Government of **Western Australia**
Department of **Water and Environmental Regulation**

Murujuga Rock Art Monitoring Program: Monitoring studies data collection and analysis plan

Department of Water and Environmental Regulation
and Murujuga Aboriginal Corporation
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Acknowledgements

The Department of Water and Environmental Regulation recognises the Traditional Owners and Custodians of Murujuga; the past, present and future generations of Ngarda-Ngarli, and their ongoing connection to this sacred country. All aspects of the program will be conducted with respect for, and be guided by, the cultural law, knowledge and practices of the Circle of Elders, Traditional Owners and Custodians of Murujuga.

This document has been jointly approved by the Murujuga Aboriginal Corporation and the Department of Water and Environmental Regulation, who jointly oversee the implementation of the Murujuga Rock Art Strategy. Cultural approval has been granted by the Murujuga Circle of Elders to undertake research on Murujuga country and all scientific elements are subject to independent peer reviews.

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Cover photograph: Fish engraving, Murujuga Aboriginal Corporation

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Murujuga Rock Art Monitoring Program: Monitoring studies data collection and analysis plan

Final Report

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Calibre Rev.	Curtin Rev.	Date	Revision Reason
-	1	30 July 2020	Initial Release – Part A
A	2	25 August 2021	Draft Re-release to Calibre with − study design updates following peer review by the DWER-appointed scientific reviewers and the Stakeholder Reference Group. − site locations and sample size determination following statistical modelling.
B	2D	06 September 2021	Re-Release to DWER post internal revisions.
C	2E	18 November 2021	Update post field sample selection and review comments
0	3	27 January 2022	Final version issued for use following review process

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Executive summary

The Murujuga region is home to what is believed to be the largest collection of petroglyphs in the world, as well as a number of large industrial facilities and supporting infrastructure. A significant body of research and environmental monitoring has been conducted – especially since 2004, with the aim of determining if anthropogenic emissions are accelerating natural weathering processes occurring on the petroglyphs and surrounding rock surface.

However, some of the previous research has been criticised for a range of issues including sub-optimal site selection, insufficient sample size and methodological flaws. Monitoring, modelling and rock art weathering research has also been conducted by relevant industries, government and academia; however, systematic integration and meta-analysis of datasets have not been conducted. As such, because of the above issues, there remains uncertainty over the levels of anthropogenic emissions which would cause an acceleration of weathering of the petroglyphs.

For these reasons, the Murujuga Rock Art Monitoring Program (Monitoring Program) has been started by the Government of Western Australian (in partnership with the Murujuga Aboriginal Corporation) to:

- develop and implement a world's best practice monitoring campaign
- ensure the rock art can be protected
- support the World Heritage listing application process.

Field measurements on the petroglyph surfaces are inherently challenging, because of the range of parameters which cannot be controlled. These include:

- significant spatial variation in colour and texture of the rock patina – even on very small scales
- the need to ensure measurements are non-detrimental
- vastly changing climatic conditions
- rugged and varied terrain
- uncertainties in petroglyph dating etc.

As such, there remains some uncertainty on whether colourimetric or spectral analysis of the patina in the field will produce significant results unless an extremely large sample size and/or very long-term monitoring programs are deployed.

This study therefore details a dual approach of non-invasive field monitoring coupled with highly resolved laboratory measurements on rock patina, in order to allow a complete understanding of the bio/geo/physico-chemical processes which can occur on the petroglyph and non-petroglyph patina, and the sensitivity of these processes to various gaseous or particulate air pollutants which they could feasibly be exposed to. Both arms of the study have undergone appropriate statistical design with respect to sample size and spatial location (see Appendix I), which will be revisited for studies where preliminary data is yet to be obtained.

The above dual-armed approach increases the potential for success of the Monitoring Program, as the laboratory-based arm would be sufficient to establish pollutant thresholds for Environmental Quality Management Frameworks (EQMF), even if the findings of further field studies (and meta-analysis) of field datasets remain inconclusive. The field measurements of air quality near the petroglyph surfaces can be combined with the 'dose-response' information from the laboratory studies.

The main goals of this study are to:

1. undertake studies to measure air quality parameters which may foreseeably impact rock art
2. study rock art petroglyphs and adjacent rocks and soil to determine if measurable impacts on colour or bio-physico-chemistry can be found associated with air pollutant exposure
3. link the evidence in the studies above to provide a rigorous evidence base for the Conceptual Model, Environmental Quality Criteria (EQC) and the EQMF.

The specific components of the proposed study program are:

1. non-invasive monitoring:
    (a) spectral analysis of petroglyphs
    (b) prescribed air quality monitoring – nominally at locations proximal to (a)
    (c) high-resolution laser scanning of monitoring sites and DGPS location mapping for all measurement points used in (a)
    (d) regional-scale air quality modelling
    (e) sub-scale air quality modelling using data from (c)
2. Laboratory experiments on non-petroglyph samples:
    (a) detailed characterisation of the rock microbiome (abundance, diversity, and physiological properties/gene expression) and response to environmental exposures
    (b) detailed characterisation of the inorganic geochemistry of the rock patina and response to environmental exposures
    (c) detailed characterisation of the organic geochemistry of the rock and interaction of organic pollutants with (a) and (b).
    (d) isotope analysis of key emissions to enable source apportionment of measurements in (1b).

The following sections detail the proposed analyses for each of the above. The above components inform the development of the Conceptual Model, which has been previously detailed in a separate document that should be read in conjunction with this document. Following peer review and approval of this and the Conceptual Model document, it is planned to conduct preliminary method development/validation for the work packages in 2(a-d) and modelling (1d), which will be combined with plume dispersion modelling commissioned by the Department of Water and Environmental Regulation (DWER) and industry monitoring datasets, to allow optimal statistical design for selection of locations of study sites (1a-c,e) and selection of locations for sample collection (2a-c). Following peer review of this second stage, the site locations would then proceed to consultation with the Murujuga Aboriginal Corporation (MAC) and the Murujuga Elders to obtain the approval needed to commence.

Figure E-1 shows the coarse-scale site selection. In this figure:

- Magenta 'EX': denotes existing Air Quality (AQ) monitoring stations included in the design
- Blue 'AQ': denotes proposed new AQ monitoring sites
- Green 'RS': denotes representative sample sites
- Orange 'AS': denotes additional sites to capture known dolerite and granite rock art.

Blue and magenta sites will be used for all of the studies outlined above. Green sites will be used for 1(a) and all of 2.

A full list of the inclusions in each of the course study sites is included in Section 5, and Appendix I and II contain further detail on the selection process for these sites.

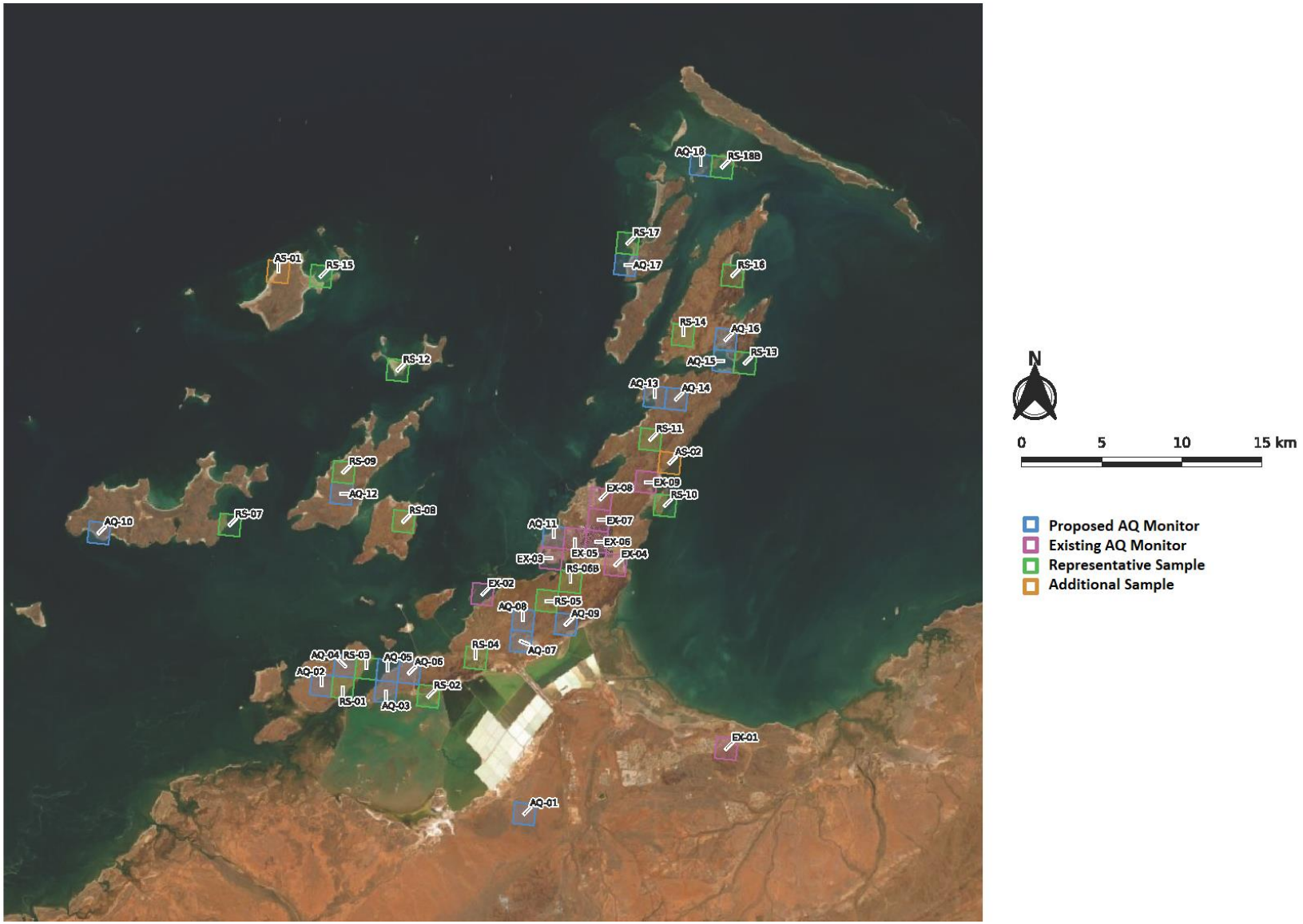


Figure E-1 Optimised site selection.

1 Study objectives

The objectives of the Monitoring Program, including data quality objectives, are described in the following sections.

It is important to note that both the Monitoring Program contract and the design/development stages, required by the research team to develop a 'world's best practice' scientific study, necessitated that the project be split into a number of parts, namely:

1. Conceptual Model(s) (DWER, 2021)
2. Monitoring Studies Data Collection and Analysis Plan (Current Document)
3. *Study of the Cumulative Impacts of Air Emissions in the Murujuga Airshed* (Ramboll, July 2021 – link in references).

These documents should be considered in concert in order to capture the complete study design, rationale, hypotheses and inter-relationships, as far as can be established based on currently published data. Document 2 was previously issued for review as a part A document (without finalised site locations or sample size information). The above has now been completed, as well as incorporating revisions from all reviewers commissioned by DWER.

The previous Part A document has been revised to include:

1. a finalised plan for the studies for the first year of monitoring (subject to peer review and guided by the reviewer comments on Part A).
2. statistically optimised study site locations (subject to MAC approval)
3. optimised number of study sites and number of replicate samples/analyses per site
4. improvements to the design incorporating reviewer suggestions (increased real-time air quality monitoring (number of sites and duration), soil monitoring, expanded pH monitoring).

This work also includes hypotheses to be tested (where appropriate) and a timeline for the Initial Studies (IS) which form part of the project.

In addition to revisions because of further peer review, the study design has been additionally revised based on preliminary fieldworks to select study sites. Specifically, two additional rock types (granite and basalt) were included as an abundance of rock art was found on these two rock types. The site selection was also amended to include two additional sites with known rock art on dolerite and granite.

1.1 Overarching principles of the Monitoring Program

The authors have taken on this Monitoring Program to assist the Murujuga people in preserving their rock art. Before commencement of the project, DWER made it clear that we are to consider MAC and their Circle of Elders equal partners in awarding and leading the project. As such, we wish to emphasise that we consider that we are working for MAC and the Murujuga Elders, and we undertake that no part of this project or the proposed sub-studies will adversely impact any rock art, either intentionally or unintentionally. Every effort will be taken to work with MAC and the relevant State and Commonwealth authorities to ensure all appropriate approvals and agreements are in place before collection of any samples or undertaking fieldwork. Collection of samples for destructive analysis or installation of any monitoring equipment is only proposed for areas where every effort

has been made to verify no rock art is present and we have obtained full agreement from MAC and the Circle of Elders.

1.2 Ultimate goal and guiding principles

This Monitoring Program is designed to provide the best possible scientific evidence base for the assessment, approval, management and compliance responsibilities of governments, industry and MAC under the *Environment Protection and Biodiversity Conservation Act 1999* (Commonwealth) and the *Environmental Protection Act 1986* (WA). The ultimate goal of the Monitoring Program is to obtain rigorous scientific findings about the condition of the Murujuga petroglyphs, about the chemical, biological and physical processes which affect their condition, and about the impact and potential future impact of industrial output on their condition, all of which provides an evidence base for the Conceptual Model and the EQMF. It is also envisaged that the program of monitoring and EQMF will form a fundamental part of the Murujuga Rock Art Strategy, which in turn has a critical role in the development of a nomination for World Heritage listing of the Murujuga Cultural Landscape.

An EQMF provides a risk-based framework that aims to protect the Murujuga petroglyphs from accelerated weathering because of anthropogenic emissions. It will also form the basis of the ongoing monitoring and assessment program at Murujuga. Environmental monitoring data will be compared with EQC that provide an early warning indicator and a threshold of unacceptable change in rock art condition. Each criterion will be linked to appropriate management responses to control the level of risk associated with anthropogenic emissions.

The following are important considerations for the Monitoring Program:

- MAC and the State Government have jointly commissioned the research and will jointly approve the details of the work to be carried out.
- Key principles of the research carried out by the Program are:
    - respect for the cultural law of Murujuga
    - commitment to world's best scientific practice in all elements of the research.
- The monitoring program will ultimately be jointly run by the Murujuga Aboriginal Corporation and DWER (ongoing monitoring). The Program will strive to transfer skills and technology to the Murujuga rangers and will be designed to be carried out with portable equipment, phone apps and other convenient technology wherever possible.

The Monitoring Program broadly consists of:

1. development of a conceptual model (completed subject to revisions)
2. design of the Study Phase of the program – current document
   (undertake initial studies – 12-18 months)
3. design of Monitoring Phase of the program
   (transition from initial studies to ongoing monitoring)
4. develop EQC(s) and EQMF in parallel to 2 & 3.

The Studies Phase of the program will involve measuring and studying many variables which will not be part of the final Monitoring Phase of the program, but which we need to understand to demonstrate links between the environmental variables and the impacts on the petroglyphs. For example, the microbiome study must identify which species of bacteria, fungi and endolithic lichens

are present on and in the rock surface; this is necessary to understand the process of change, and to predict how the rock will be affected by different environmental conditions. The monitoring program will necessarily reduce works to only those which have been found to demonstrate a causal link and also offer minimally invasive and/or sample intensive study designs.

1.3 Research objectives

The main research goals of the study are to:

1. accurately measure the colour and surface texture of rocks across the Murujuga region, establish baseline values for a long-term monitoring program, identify important differences in colour or surface texture which may be associated with accelerated (anthropogenic) weathering, and develop criteria for assessing weathering rates
2. identify and characterise the minerals, inorganic and organic chemicals, and microbes present on the rock surface and in the sub-layers, which are relevant to the appearance and integrity of the petroglyphs or may be involved in accelerating or preventing the degradation of the petroglyphs
3. determine which atmospheric pollutants are present in industrial (e.g. port, shipping and local industry) and natural emissions (e.g. marine aerosols, emissions from bushfires) on Murujuga, and which are capable of causing degradation of, or change in, the petroglyphs
4. under controlled laboratory conditions with small samples of rock, measure the effect of a known amount of pollutant chemical on the constituents of the rock surface and sub-layers
5. measure the concentration of atmospheric pollutants to which rocks are exposed, across the Murujuga region
6. identify the weather conditions, environmental conditions and industrial output conditions which are likely to pose the greatest risk of degradation to petroglyphs
7. calculate the timescales over which changes are predicted to occur, and thresholds of pollutants giving rise to accelerated change
8. identify sentinel variables (‘canary in the coal mine’) which can be observed in the field, and which can serve as indicators (EQC) of increased risk of degradation
9. determine if soil monitoring will inform some processes or measurements on the rock surface
10. utilise world-leading statistical design, methodology, and analysis techniques
11. link all of the evidence in the studies above to reach rigorous scientific conclusions and provide a rigorous evidence base for the Conceptual Model and the EQMF and the Monitoring Program as a whole.

These broad objectives are further expanded below into specific objectives for each of the component studies.

1.4 Organic geochemistry

The objectives of the Murujuga organic geochemistry study are to:

1. measure the molecular and stable isotopic composition of organics from all sources, including natural emissions (terrestrial vegetation, fire), anthropogenic emissions (shipping, traffic, industry) and organics present in the host rocks (including rock dust from iron ore transport),

to produce a detailed characterisation of organic emissions in the Murujuga region, including determination of the occurrence and magnitude of seasonal variations in emissions

2. measure the molecular and stable isotopic composition of organics deposited on rock surfaces and in soils, to link organics on rock surfaces to the various emission sources, and determine which sources contribute the highest quantity of organics to the surface deposition
3. identify any organics that are likely to affect weathering of rock art, e.g. by affecting growth of microorganisms (in conjunction with microbiome expert(s)) or interaction with specific minerals (in conjunction with inorganic geochemist(s))
4. understand confounding factors introduced by (reported) changed fire management regimes approximately coinciding with the commencement of industrial activity on the Burrup.

1.5 Inorganic geochemistry

The objectives of the inorganic geochemistry investigations are to:

1. characterise the mineralogy and mineral compositions of the fresh rock, weathered rind, and patina for the granophyre, gabbro, dolerite, granite and basalt rock types from a range of settings including high and low exposure to industry emissions and differences in exposure to rain and wind
2. determine the mineralogy for these systems at thermodynamic equilibrium using thermodynamic calculations and use best practice approaches to estimate the timescales of change, based measured reaction rates, reaction mechanisms, and the results of other component studies (organic, microbiome, field observations, weathering experiments)
3. compare the expected and observed mineralogy for sites with different exposure to anthropogenic emissions to:
   (a) obtain proof-of-concept for the thermodynamic calculations
   (b) investigate the presence of statistically significant differences among the sites and relate these to industry emissions
4. use the observations and calculations to identify components of the mineralogy that are most sensitive to industrial emissions and that can act as early indicators of change (EQC) and devise a monitoring strategy that will form part of the EQMF and ongoing monitoring.

1.6 Microbiome

The objectives of the Murujuga rock patina microbiome study are to:

1. use advanced molecular biological approaches to characterise:
   (a) microbial community composition associated with rock patina and the underlying weathered rind. *This answers* '*Who is there?'*
   (b) diversity and relative abundance of functional genes involved in microbial processes and pathways that may contribute to stabilising or bio-deteriorating the rock patina are present: *This answers 'What are they potentially doing?*'
   (c) characterise which of these functional genes are expressed to functional gene transcripts that can potentially be translated into functional proteins/enzymes that carry out these processes

2. Use advanced bioinformatics tools to:
   (a) build metagenome assembled genomes (MAGs) from the shotgun metagenome dataset of objective 1b and map the functional gene transcripts from objective 1c to the MAGs. This will reveal all sequenced genes belonging to the same species that were expressed at the time of sampling
   (b) determine the number of mutations that occurred post-mortem in the assembled metagenomic bins (from objective 2a) to distinguish between microbial taxa that have been preserved in the patina as ancient DNA versus those that represent modern taxa. Since this DNA damage increases with time it may be possible to determine the order in which these taxa died
   (c) estimate the growth rate of the patina associated microbial communities by measuring genome replication rates from shotgun metagenomic bins without the need for cultivation
3. perform cultivation experiments to follow seasonal changes in the composition and number of viable bacteria and fungi isolated from the rock (sub)surfaces and to compare the community composition and metabolic potential with the microbiomes from the pristine field samples.

   The above objectives involve the analysis of patina and weathering rind samples from the five main rock types (granophyre, gabbro, dolerite, basalt and granite), which will be obtained directly during wet and dry seasons of years one and two of the initial study)
4. expose fresh rock cubes from the three main rock types with the patina removed to field environmental conditions to monitor the rate in which a patina-building microbiome re-colonises the weathering rind and which taxa and processes are involved. These datasets will be compared with the established rock patina microbiomes.
5. perform cloud/exposure chamber time series exposure experiments to determine the threshold concentrations of key pollutants in the region that result in a shift in microbial communities with bioweathering properties.

The datasets of the five overarching aims will be integrated with changes in mineral and lipid biomarker compositions and in the context of changes in the environmental parameters, which will be analysed in parallel as a team.

1.7 Surface condition

Monitoring of the rock surface condition will assess three main aspects: colour, texture, and elemental composition.

The objectives of the surface condition study are to:

1. accurately measure the colour, texture and elemental composition of selected rock surfaces across Murujuga
2. study the secondary reflectance characteristics to monitor chemical change, anticipating future colour change
3. capture the broad surface elemental distribution to understand colour change across whole surfaces to correlate with and inform spot measurements
4. establish baseline values for a long-term monitoring program

5. identify important differences or changes in colour, surface texture and elemental composition which may be associated with degradation of appearance or condition
6. develop deeper understanding of variation in elemental composition across the rock surface and its relationship to colour and texture
7. develop criteria for assessing 'degradation' of appearance or condition
8. ensure that the study data provide information required by other component studies (such as the precise spatial location of each observation).

1.7.1 Perceptual and spectral colour

Change in the visible colour of a surface is a reasonable indicator of change in its composition and condition. Colour change was adopted as the main indicator of degradation in several earlier studies of the Murujuga petroglyphs, but will be only one of several measures of change in this study. We will integrate results from different techniques to determine the causal relationships between external factors that affect the rock art, the physical characteristics of the rock art (e.g., colour, mineralogy, indigenous microbiome), and the processes that affect change. It is considered likely that, within the life of this Monitoring Program, colour change will not be of sufficient magnitude to allow meaningful interpretation in its own right.

To better observe and characterise changes, photospectrometric analysis will not only describe the perceptual colour but will study the entire reflectance spectrum in the range of the nominated instrument, namely wavelengths from 250–1,000 nanometres (the visual spectrum ranges from about 380–740 nanometres). Spectral change will be studied across the entire spectrum to identify trends indicating mineralogical change not yet manifesting as colour change. This spectral analysis beyond perceived colour will provide more informative data analysis, greater opportunity to detect change, and improve the ability to predict change as required for the development of the EQMF.

Undisturbed rock surfaces undergo dynamic change through natural processes, both biological and mineralogical, which continually alter the appearance of the rock. In a natural system the colour of the disturbed surface progressively approaches that of the undisturbed background. The rate of this convergence has been estimated to take millennia.

In an industrially influenced scenario, the process of change is less well understood, but has been anticipated in recent studies to be an erosion of the manganese/iron mineralisation crust. While the granophyres are not iron- or manganese-rich, these elements become concentrated in the residue left after the rocks weather and more soluble components are lost. Erosion of the patina would result in a lighter surface as the dark mineralisation gives way to the lighter weathered rind beneath. The colour trend in a natural system would be darkening with an anticipated trend towards red and yellow values. In a dissolution model the colour trend would be towards lighter and less red and yellow values.

Data from previous colorimetric studies, conducted in the period 2000-16, can be used to establish approximate baseline values, and approximate estimates of the variability, for colorimetric values. These pilot estimates, together with the above-mentioned considerations of the potential directions of colour change, will inform the study design and data analysis.

1.7.2 Elemental distribution

Coupled to spectral analysis, elemental distribution will enable better detection of seasonal variations as well as progressive long-term trends in the interaction between the rock system and the

atmosphere. In situ (portable) X-Ray Fluorescence (pXRF) measurements will plot fluctuating elemental composition on the surface to support evidence of spectral changes and further build a case for stability or change. The benefit of elemental mapping is that it better links change to deposited elements, such as sulphur, and more precisely observes the ratios of manganese and iron in relation to reference elements. The XRF studies will also explore spatial elemental distribution of visual variability across a surface to give deeper interpretation of the composition of the variously coloured surfaces.

Element ratios can be used to constrain the possible minerals present, but may not provide a unique identification. For this reason, the pXRF work is complemented by other techniques, such as XRD that can identify the minerals uniquely. Preparation and analysis of thin sections (section 3.3.2ii) will also enable us to determine the changes occurring within the substrate.

1.7.3 Texture

One aspect of change that requires deeper consideration is that of surface morphological change. Preliminary site visits have highlighted the issue of subsurface mineral formations that have the ability to alter the appearance of the surface, including its colour, through the formation or loss of minerals at and below the surface. Mineral stability depends on the thermodynamics of the mineral systems and environmental parameters such as pH, redox, temperature, and the concentrations of a range of elements, such as silicon (Si). Minerals can form by precipitation from solution or be lost by dissolution. Delamination has been identified in previous studies (Pillans and Fifield, 2013) as the most threatening mechanism for the engraved surfaces. Not only can this be directly linked to mineral formations beneath the surface but a quantum loss of surface through delamination can vastly alter the spectrometric measurement of a surface. The morphological studies, centred on microscope-based photogrammetry but also applying Reflectance Transformation Imaging techniques will study select engraved surfaces to measure dimensional change. The study will provide comparison of the same surface through time to identify dimensional change in the surface at 10 micrometre resolution. Ten micrometres is the resolution available for the field techniques, but higher spatial resolution is available using the laboratory-based techniques. The results will be integrated with those using other laboratory-based techniques to link textural observations to inferred mechanisms of mineralogical change.

1.7.4 Surface Eh-pH measurements

Eh-pH is a measure of the stability and/or reactivity of mineral and chemical systems (refer glossary and Section 2.1.5). The objectives of the surface Eh–pH measurements are to:

1. add to, and corroborate with, existing Eh–pH measurement datasets (Macleod, 2005; Black et al., 2017; Macleod and Fish, 2021)
2. place Eh–pH measurements in the context of the air quality measurements and weather records
3. interpret Eh–pH measurements in the context of the processes observed in the weathering experiments.
4. link Eh–pH measurements to ex-situ inorganic and organic geochemical observations and microbiome analyses to constrain the reactions that stabilise and destabilise Fe- and Mn-bearing minerals
5. use Eh–pH measurements to augment the in-situ field-based colour monitoring and elemental distribution results, to reduce uncertainty and provide constraints on the processes occurring on the rock surfaces.

pH measurements can and will be conducted adjacent to petroglyphs being studied, on non-engraved rocks included in the study and on prepared rock cubes at AQ monitoring stations.

1.8 Air quality

The objectives of the air quality modelling and monitoring are to:

1. provide long-term monitoring of air quality at a sufficient number of locations to enable us to determine the degree of exposure of the petroglyphs to pertinent air pollutants
2. conduct fine-scale air quality and fluid dynamics modelling on existing data, both to inform the choice of monitoring location, and to determine fine-scale airflow and pollutant transport in study regions – especially those with highly variable topography (gorges etc.)
3. conduct air quality measurements to validate the modelling above and that done by others and collect large mass samples to establish isotope signatures for source apportionment.

1.9 Climate

Peer reviewers have strongly suggested that climate change be considered as an additional anthropogenic impact. In addition to global anthropogenic climate change, local effects such as changed burning practices – likely cessation of cultural burning in 1868, changes in bushfire management in the modern era and gradual reintroduction of cultural burning currently – also need to be considered. We aim to consider all such effects as far as possible. Bushfire impacts can be accounted for in chamber studies and comparison of samples proximal to recent fires. Climate effects (temperature, humidity, rainfall, extreme weather events) based on the best available models for the region, will be explored in conjunction with geochemical and microbiome models – once established – in order to permit long-term climate impacts to be explored. Therefore, although not explicitly stated throughout the document, climate change impacts will be implicitly considered in all studies and analyses.

2 Evaluation of previous studies and data

Previous primary studies have included air pollution monitoring (Gillett, 2010), modelling of the atmospheric transport of air pollution (Sinclair *et al.*, 2009), laboratory experiments on the effect of airborne chemicals on rock surfaces (Lau *et al.* 2007), microbial diversity (O'Hara 2008), measurement of colour change and mineralogy (Markley *et al.*, 2014) and measurement of pH and its possible association with measured colour change (Macleod, 2005; Black et al., 2017; Macleod and Fish, 2021).

2.1 Scientific value, limitations, usefulness for future work

2.1.1 Spectrophotometry

The data files associated with the CSIRO colour change study (Markley *et al.*, 2014) were made available to us (Table 2-1).

Table 2-1 Data files associated with the CSIRO colour change study.

File	Description	Originator
allLabFromASD2004-2016.xlsx	ASD portable spectrometer data	CSIRO
Burrup Rock Art Monitoring Program_Metadata Information_23Dec2019.docx	Metadata for Burrup Rock Art Monitoring Program	CSIRO
dwer4_colour_spectral_data_2004_2016.zip	ASD portable spectrometer data	CSIRO
BYKall.csv	BYK Data	CSIRO

Rock surfaces at Murujuga were measured spectrophotometrically at seven sites in the period 2004-2016, with a further three sites added in a study for Yara Pilbara from 2014-2016. The early studies employed a combination of three colorimeters and spectrometers, with the later study introducing stereo imaging to generate three-dimensional relief profiles. The colorimetric and spectrometric data from 2004 has been provided to us, but without precise location data. The stereo imaging work has been described in public reports but certain details of the study protocols are missing from those reports, making further analysis difficult.

Scientific commentary on the colour change studies and on their general context has been published by Bednarik (2002, 2007), Black *et al*. (2017;2017b), Smith *et al.* (2020), and reported by Data Analysis Australia (2016).

i. Site selection

Sites for spectrophotometry were selected by a process described by Markley *et al.* (2014). In brief, sites were 'determined' by the Rock Art Monitoring Management Committee. One 'representative' petroglyph at each site was selected in consultation between the committee's technical advisor and the indigenous community. Several 'spots' on each petroglyph were identified and are shown in photographs in Markley *et al* (2014). A 'spot' typically includes both engraved and non-engraved

areas. It is unclear to what level of accuracy the researchers were able to reposition the spectrophotometer on the same spot if the measurement had to be repeated.

These selected sites are not representative in the statistical sense, that is, the observations at these sites cannot be extrapolated to other petroglyphs or other rocks using the principles of sampling theory (Thompson, 2012). In order to draw conclusions about the entire population of rocks, the sample of sites would need to have been chosen at random, in such a way that every rock in the target population had a chance of being selected in the sample, and the probability of selection is known. Of course, this may not be possible, for practical and cultural reasons. This is a weakness of the previous colour study data, regardless of the instrumentation and techniques used. Overcoming this weakness should be a priority for this Monitoring Program. Otherwise, the study will need to deploy alternative, less-satisfactory methods of extrapolation based on model assumptions.

ii. Instrumentation

Colour measurement was performed using three different instruments:

- The *BYK-Gardner* hand-held spectrophotometer reports colour values in the CIE L*a*b* coordinate system (see below).
- The *Konica Minolta (KM)* spectrophotometer measures reflectance at 30 wavelength bands, and reports colour values in CIE L*a*b* coordinates, with capability to report reflectance spectra.
- The *Analytical Spectral Devices, Inc (ASD) FieldSpecPro* spectrometer measures the reflectance spectrum between wavelengths 350–2,500 nm in 1 nm steps with a wavelength resolution of 3–10 nm.

Reproducibility of colour measurements is essential, especially for detecting gradual trends. Inadequate reproducibility of the data from these instruments has apparently been an important issue in the previous studies, and may have motivated the choice of different instruments over time. The report by Data Analysis Australia (2016) specifically identifies the BYK instrument data as suffering from problems of calibration, and we concur with this assessment, as discussed below. Data Analysis Australia (2016) recommends that the ASD instrument should be adopted.

Some commentary has noted that the instrument manufacturers' disclaimers discourage the use of these instruments in field work. This is a concern because regardless of the intrinsic value of the data or the scientific validity of the disclaimer, it presents the risk that scientific findings that are partially based on these instruments may not be accepted by the scientific community. In response, the proposed instrument, the Jaz Spectrometer (Ocean Insight, Dunedin, FL, USA) is rated to 50 °C and will only be operated below that value (Peak ambient temperatures rarely exceed 40 °C in the region; however, radiant heating from rocks may exceed this and cooling or moderation of time of work will be undertaken as needed). Generally, thermochromatic variations, that is the variation in target temperature, is far more significant and has been addressed in the measurement protocols.

iii. Data curation

The data files provided to us would need additional work by the original providers in order to meet the latest standards of reproducible research (NHMRC and Universities Australia, 2018). These standards include openly accessible documentation on the conduct of the study, definitions of all the variables, the units in which they are expressed, and the encoding of missing values. Data files should use a consistent format, consistent naming conventions for files and variables, and have an openly documented format. There should be a documented chain of 'provenance' for the data, including all

adjustments, recalibrations and transformations of the original data, preferably documented using reproducible computer code (e.g. in the languages R or Python) and preferably supplying the original raw data files for complete reproducibility. Adherence to these standards is an important safeguard when research is likely to be subjected to close scrutiny, is strongly encouraged by top scientific journals, and increases the likelihood that research findings will be accepted. Many of the files provided lack such information and contain the datasets themselves with limited metadata (method, calibration data, etc.).

We have been able to glean some of the missing information from the publications of the original investigators, and from the Data Analysis Australia (2016) report which includes information from the original investigators. Data Analysis Australia (2016) recommends that the data files should be thoroughly reorganised and documented. We concur with this recommendation.

A complete chain of provenance for the data is not available to us. Most concerning is the possibility (also noted in the Data Analysis Australia, 2016 report) that some of the data may not be consistently calibrated. See our discussion of the BYK data in Section 2.1.1v.

iv. Colour change detection

The previous colorimetric analyses were performed and reported in CIELAB coordinates (also known as CIELab or *L*a*b** coordinates). The CIELAB system encodes visible colour using three coordinates *L* (representing lightness), *a* (representing hue between green and red) and *b* (representing hue between yellow and blue) so that two colours with the same *L,a,b* coordinates are perceived as identical by human eyes, and so that uniform changes in the coordinate space correspond to roughly uniform changes in perceived colour (Schanda, 2007, chapter 3). CIELAB values depend on the white point chosen for reference; for reflected colour the white point should correspond to the illumination source. The ranges of *a* and *b* values depend on the specific implementation; typically *a* and *b* are scaled to the range of 8-bit signed integers (-128 to +127) or signed percentages (-100 to +100). We have not yet been able to identify the nominal ranges of *a* and *b* used in previous analyses, and this affects some of the conclusions of the data analysis.

Colour change was assessed in previous studies using the Euclidean distance in the CIELAB space:

$$\Delta E = \sqrt{\Delta L^2 \Delta a^2 \Delta b^2}$$

where *ΔL* is the difference in *L* value between two colours being compared, and similarly *Δa* and *Δb* are the differences in the other coordinates. This reduces a colour difference to a numerical value; a threshold of two units is often taken as the smallest perceptible colour difference ('Just Noticeable Difference'). Again, this calculation depends on the range of *a* and *b* values. Some studies appear to have used the '1976 definition':

$$\Delta E = \sqrt{\Delta a^2 + \Delta b^2}$$

which ignores the discrepancy in lightness, and corresponds to Euclidean distance in the two-dimensional *(a,b)* space of hues. Shades of grey are indistinguishable (*ΔE*=0) according to the latter definition, but not according to the former definition. Colours which are noticeably different according to the latter definition must also be noticeably different according to the former definition, but not conversely.

In the absence of other benchmarks, a provisional definition of 'accelerated degradation' could be based on exceeding the threshold for just-noticeable difference in colour ($\Delta E > 2$) within a relatively short time scale.

Some previous studies appear to have used *ΔE* to infer *trends* in colour (not shown), by comparing the colour values at the beginning and end of a time period. From a statistical viewpoint this is unsatisfactory because it does not make the most efficient use of the available data (and therefore may fail to detect a trend when it is present). A trend should be measured by fitting a trend line (a linear regression line, or a more complicated and flexible version of regression) using all of the available data. This also enables the analyst to assess the strength of evidence for the trend, and to identify anomalous data. This approach has been advocated in the report by Data Analysis Australia (2016).

v. Issues with BYK data

Initial inspection of the data from the BYK instrument shows some anomalies that are not consistent with progressive/gradual change and are possibly attributable to change of technique or to inconsistent calibration. This has also been noted by Data Analysis Australia (2016). Figure 2-1 shows boxplots of the L coordinate against the year of observation. An abrupt change around the year 2010 is evident.

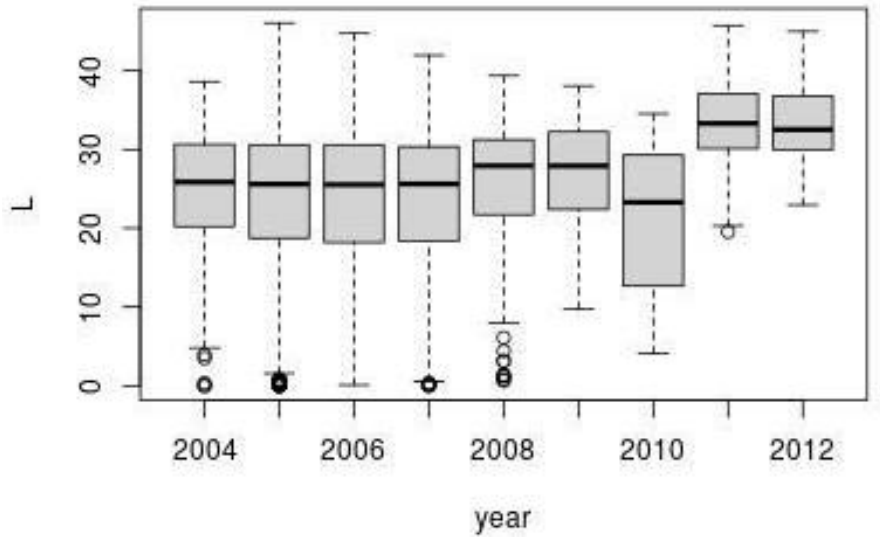


Figure 2-1 L coordinates by years.

Tabulation of the data indicates that in 2010, data were recorded for only one site. Restricting the analysis to data collected only at this site throughout the study, gives the boxplot below (Figure 2-2), which reinforces the impression of an abrupt change in 2010. A similar impression is given by the boxplots of the *a* and *b* variables, Figure 2-3.

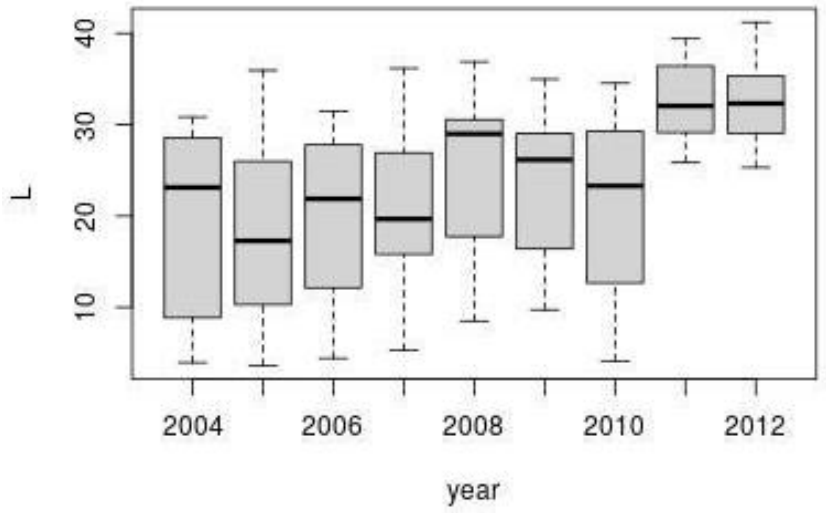


Figure 2-2 L coordinates by year, restricted by site.

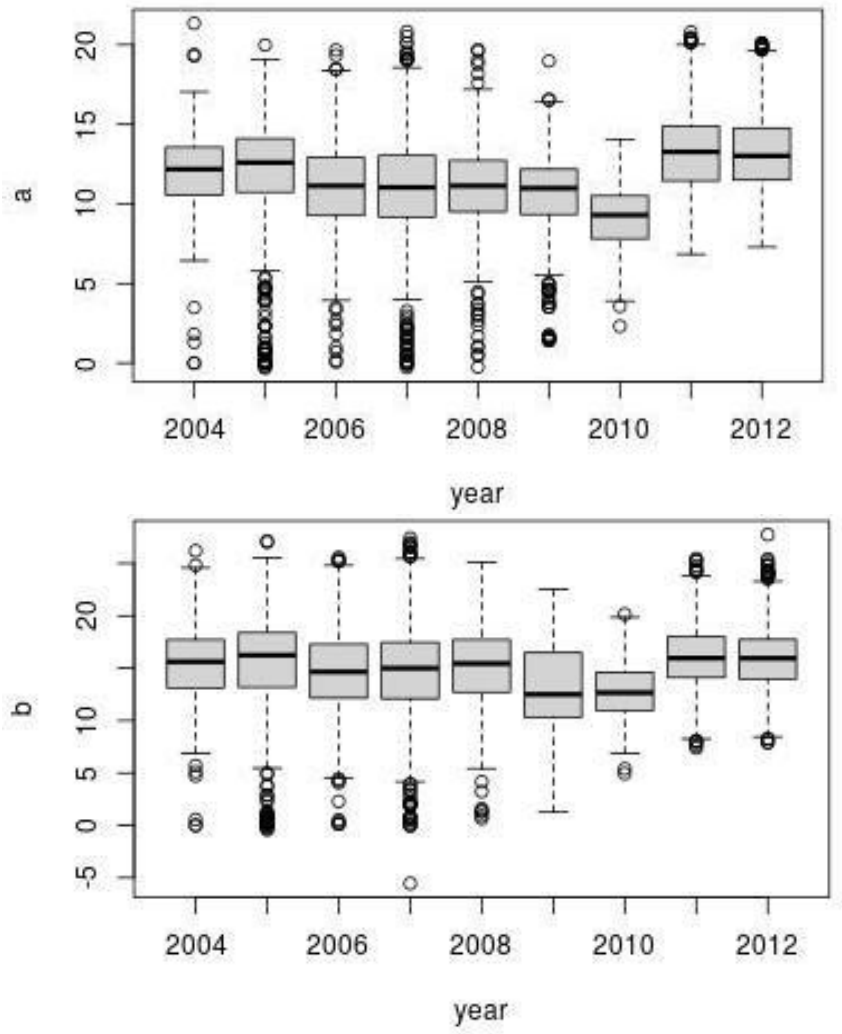


Figure 2-3 a and b variables by year.

Simple linear regression of each CIE coordinate against calendar year shows a statistically significant positive slope. The estimated trends for *L, a, b* are respectively 1.45, 0.25, 0.13 units per year, and are all judged highly significant according to the regression t-test. (Similar results are obtained from analysis of covariance taking the study design into account.) A linear trend of this magnitude would exceed the threshold for just noticeable difference in colour after two years (according to the definition of *ΔE* first given) or seven years (according to the 1976 definition which ignores lightness). Positive trends in *a* and *b* imply more red and more yellow colours, consistent with the direction expected for natural mineralisation. However, when the regression is performed only on the data from 2004-2009, the regression slopes for *a* and *b* are significant and *negative*, respectively -0.36 and -0.42 units per year suggesting a trend in the reverse direction. A broken-stick linear regression of *a* and *b* against calendar year gives a negative trend before 2010 and a positive trend after 2010. The inconsistency between these trend estimates is expected, in the light of Figure 2-1 and Figure 2-2.

These anomalies mitigate against a straightforward interpretation of colour differences or colour trends over the period 2004-2012 and suggest that the BYK instrument data should not be used, or should be used with extreme caution, in any future analysis.

The previous colorimetric data do have substantial value for this Monitoring Program in that they provide approximate benchmarks of the colorimetric values. More importantly, the variability in colorimetric values has helped shape the current studies through the refinement of a range of measurement procedures, including instrument operating temperature, thermochromaticity, probe contact angle and several other controllable parameters.

Markley *et al* (2014) state that there was high variability between successive repeated measurements in the BYK data; that the original intention was to take seven (7) replicate measurements at each spot; this was later revised to 21 replicates. This is consistent with the data available to us. A simple analysis of covariance for the individual coordinates gives approximate standard deviations of 3.0 units for *a* and 3.8 for *b.* Taking the average of 21 replicate observations would give standard errors of 0.7 and 0.8 units respectively. These standard errors are about double the yearly change in the *a* and *b* coordinates according to the (probably overestimated) linear trends fitted above. Deeper analysis of the results of previous studies has been undertaken where possible and prudent, in order to estimate different components of variance, to inform the final study design (refer Appendix I).

2.1.2 Inorganic geochemistry

Large-scale geological maps of the area are available, but smaller scale maps (1:50,000 or lower) are not publicly available.

Ideally, it would be possible to compare rocks exposed to industrial emissions to ‘control’ rocks, which are not exposed to industrial emissions, but are exposed to environments that are comparable apart from the emissions. However, this is not possible, and attempts by previous workers to set up ‘control’ sites have been shown to be flawed.

A small number of studies have described the mineralogy and geochemistry of the Murujuga rocks (e.g. Donaldson, 2011; Ramanaidou *et al.*, 2017; Ramanaidou and Fonteneau, 2019, Fairweather, 2019). However, the amount of data and spatial coverage are insufficient to assess the extent and length scales of variability in the composition and mineralogy of the fresh rock, the weathered clay layer, and patina. Selected studies are discussed below.

An experimental weathering program (Ramanaidou *et al.*, 2017) characterised minerals and textural relationships by optical microscopy and scanning electron microscopy (SEM), performed colour measurements using a Konica Minolta CM-700d (KM) instrument, reflectance spectroscopy (400–2500 nm) with a spectral resolution of 10 nm at 700 nm and equipped with three detectors: a 512 element Si photodiode array for the 400–1,000 nm range and two separate TE cooled, graded index InGaAs photodiodes for the 1,000–2,500 nm range. This instrument was used to identify minerals at the mineral group level of identification (e.g., chlorite, iron oxide), but did not identify the minerals at the level of detail needed to monitor small changes in the mineralogy, which might include changes in the composition of chlorite or changes in the degree of iron oxide hydration. Statistical tests (t-tests) were used to compare the artificially weathered and unweathered samples, but these used some assumptions (e.g. equal variance) that are difficult to justify, based on the small sample sizes.

Ramanaidou and Fonteneau (2019) obtained samples from 10 sites close to the monitoring locations used for the study of Ramanaidou *et al.* (2017). A suite of techniques including X-ray diffraction (XRD), SEM, micro-X-ray fluorescence mapping, and reflectance spectroscopy were used to characterise the composition and mineralogy of the rock surfaces. Electron probe microanalysis (EPMA) was used to measure the composition of the minerals. Whole-rock chemical analyses were performed on eight samples, and the data were combined with three analyses of the quench gabbro, two analyses of the basal gabbro and eight analyses of the granophyre taken from the databases of the Geological Survey of Western Australia. The whole-rock compositions of the fresh rock and weathered rind were compared for two sites. The chemical composition of apatite ($CaPO_4$) was measured, because difference in the distribution and characteristics of phosphorous-bearing minerals was noted during the chemical mapping, and apatite in the weathered rind and fresh rock showed significantly different compositions and shapes. The spectrometric measurements were used to derive hematite: goethite ratios, that were tested as a proxy for the age of the engravings, estimated from the colorimetric measurements. The quality of these data, excepting possibly the colorimetric measurements are likely to be good, based on the use of appropriate analytical equipment and application of mature analytical techniques, but the sample sizes are relatively small and further work is needed to elucidate the spatial variability of the different parameters that were measured. Furthermore, the work did not include any techniques with resolution at the sub-mm scale, and these are required to detect any early signs of mineralogical change that might be associated with anthropogenic emissions.

An experimental approach has been used to compare the consequences of washing rocks collected before the current industrial activity (and hence emissions exposure), and those collected by Black *et al.* (2017). The results of these experiments are valuable, in that they indicate differences in the pH of solutions that interact with the surfaces of the rocks, but further work is necessary to determine the statistical significance, implications, and geochemical processes that underlie these results.

Field monitoring studies have measured the colour and spectral mineralogy of the rocks over a period of years (e.g. Lau *et al.*, 2011). The colorimetric work is discussed in detail above. For example, spectral mineralogy was measured by an analytical spectroscopy device (ASD) in the field at seven sites (Lau *et al.*, 2011). These results are valuable but do not provide the spatial resolution, details of the mineralogy, or information on reaction relationships that are necessary to determine whether early and invisible consequences of exposure to anthropogenic emissions are modifying the appearance of the Murujuga Rock Art.

2.1.3 Microbiome

The petroglyphs of the Burrup Peninsula have predominantly been carved into the patina-covered weathering rind of the parent granophyre and gabbro igneous rock types. This rind can reach a thickness of up to 10 mm, mainly consists of relatively soft kaolinite clays, and is covered with a 1–200 um-thin patina. The carved petroglyphs owe their visibility from the contrast generated between the exposed lighter coloured weathering rind and the dark patina that covers the entire rock surface areas where carvings are absent. While there is spatial heterogeneity in mineral compositions, the patina is mainly composed of clay minerals (~70%; mainly montmorillonite and kaolite), and 25% manganese (Mn) and iron (Fe) oxides. The concentration of Mn is 50-300x higher than in the source dust material, while the ratio of Mn to Fe in surrounding soil is 1:40–1:60 as opposed to 1:1 in the patina (e.g., Dorn, 2009). In arid deserts with low rainfall, patina is formed at a slow rate of 1–10 um per thousand years (Liu and Broecker, 2000). Five theories have been postulated to explain the origin and mechanisms of patina formation:

(a) Natural pH fluctuations and abiotic oxidation are the main factors contributing to the enrichment of Mn oxides (Hook et al., 1969).

(b) Aerosol, dust particles, and organic compounds are cemented by amorphous hydrated silica and silica minerals (Perry and Kolb, 2004).

(c) Bacterially mediated accumulation of Mn and Fe oxides and subsequent cementation of bacterial remains to the other patina components (Allen et al., 2004).

(d) Both biotic and abiotic factors are at play with microorganisms accumulating Fe and Mn oxides, and precipitation of dust collating the patina to the rock surface (Dorn, 2007).

(e) Microbial activities in endolithic biofilms transform the initial mineral matrix and creates an organo-mineral film that comprises the initial microhorizon of the endolithic pedogenesis process. According to the concept, the patina is this microhorizon that is exposed to the surface because of chemical or mechanical weathering.

While abiotic factors cannot be excluded, there is accumulating evidence for a biological contribution to desert rock patina formation. A snapshot of significant findings from microbiome studies on patina covered rocks from desert and non-desert regions, located overseas, is presented below.

i. Microbiome studies performed on desert rock varnish outside of Australia

Various studies have used imaging techniques (e.g. SEM) to describe morphological features indicative of the presence of bacteria and/or fungi and to determine signs of interactions with the mineral surface. For example, SEM revealed the presence of deposited oxides on the outer membrane of bacterial and fungal morphotypes in patina samples as well as on the cell membranes of bacteria and/or fungi that were enriched in selective growth media (e.g. Esposito et al., 2015; Kuhlman et al., 2005; Lang-Yona et al., 2018; Northup et al., 2010; Parchert et al., 2012).

Previous SEM also revealed a distinct presence of bacterial coatings involved in the formation of biofilms such as extracellular polymeric substances (EPS), capsules, and other membrane structures such as pili and microfilaments on rock patina from Matsch Valley (South Tirol, Italy) (Esposito et al., 2015). Confocal microscope imaging of coloured patina from Ethiopian petroglyphs also identified the presence of patina that appeared to be covered in a monolayer of EPS (Wu et al., 2020). Being associated with a microbial biofilm offers a broad range of advantages to its members especially in hostile conditions such as adhesion, including providing a source of nutrients, cellular communication (e.g. metabolite exchange, chemotaxis and quorum sensing) as well as resistance to antibiotics, and protection from environmental stressors such as dehydration and harmful UV irradiation (dos Santos

et al., 2018 and references therein). In addition, a preferred colonisation of microorganisms within microscopic cracks and fractures was observed and microbial cells seemed to be growing inside as well as outside the rock surface layer (Esposito et al., 2015). Cracks and fissures are considered to be protective microhabitats for the endolithic microbiome through the maintenance of humidity and protection against harmful UV radiation (e.g. Wierzchos et al., 2012).

Parallel 16S rRNA gene profiling showed that cyanobacteria capable of oxygenic photosynthesis were low in abundance in patina of the Matsch Valley rocks (Esposito et al., 2015). Instead, they reported a significantly higher abundance of *Rhodopila* (Acetobacteraceae). Species in this genus contain acidophiles, anaerobic, or microaerophilic species, including anoxygenic photoautotrophs, which use Fe^{2+} as electron donor. *Rhodopila* are known to produce a type-c cytochrome oxidase with a very high redox potential (Benning et al., 1996), which could provide advantageous conditions in the highly oxidative varnish environment. Furthermore, varnish differed from non-varnish layers in the abundant presence of the extreme acidophilic genus *Acidocella* (Acetobacteraceae), indicative of the presence of acidic microenvironments. The authors postulated that under such conditions, stable divalent Fe^{2+} cations would be formed, creating the possibility of microbially mediated Fe oxidation (Esposito et al., 2015).

Despite the well-known role that fungi play in (in)organic matter transformations, elemental cycling, bioweathering (e.g. of building materials and structures), rock and mineral transformations, and interactions with clay minerals (Gadd et al., 2017 and references therein), fewer studies have focused on fungal associations with desert rock patina. For example, lichenised fungi (i.e., heterotrophic fungi forming a symbiotic relationship with microalgae) can grow within rock substrate, colonise internal cracks or pores, and deposit acidic metabolites (organic acids) that lower the surface pH causing dissolution of minerals (Favero-Longo and Viles, 2020 and references therein). More frequently reported is the presence of dark pigmented microcolonial fungi (MCF) that form compact colonies of specialised microorganisms, which inhabit pittings that they have produced themselves through the production of organic acids (e.g. Sterflinger, 2010).

However, MCF have also been reported to deposit Mn oxides directly on rock surfaces as well as in cultured isolates and may therefore also play a role in patina formation. For example, using a combination of SEM, cultivation, and 18S rRNA gene profiling, MCF were found to be associated with Mn oxidisers in patches of maximum 60-year-old incipient varnish at the Black Canyon, located in the semi-arid Chihuahuan desert of New Mexico, US (Parchert et al., 2012). The identified fungi were also found to be distantly related to known Mn-oxidising genera, and 17 isolates were able to oxidise Mn in culture (Parchert et al., 2012). The fact that MCF at more developed varnish sites were not associated with metal deposits, led to the assumption that MCF act as pioneer organisms in the harsh environment of exposed rock and that the early formation of dark Mn oxide coatings by MCF offers protection against UV irradiation to allow secondary colonisers to inhabit the same space (Parchert et al., 2012). In an earlier study initially aimed to study bacterial communities on the Black Canyon rock patina, MCF were observed on the patina, but not directly associated with Mn deposits. It was postulated that changes in metal oxidation capacity could be a result of maturation of MCF colonies and the production of stable biofilms, to allow energy being utilised for colonial growth instead of metal oxidation, which is then no longer required for survival (Northup et al., 2010).

The purpose of this incomplete overview was to show that the role that microorganisms play in the formation and/or bioweathering of patina is complicated and still not very well understood in general. This is in part because of the limitations associated with the main approaches that have been used to study the microbiology of rock patina:

> **Cultivation approaches**: Because of not well understood growth requirements of microbial species that inhabit hostile extreme environments, the success rate to bring them in culture

is very low. For example, using several targeted media to grow chemolithotrophic as well as heterotrophic bacteria from deep subsurface marlstone and granitic rocks, only yielded six enrichment cultures out of 240 (e.g. Cockell et al., 2021). Therefore, many organisms have likely been overlooked from cultivation of microorganisms from patina samples.

Microbial physiology inferred from environmental barcoding genes: Sequencing of environmental taxonomic marker genes such as 16S rRNA (bacteria) and ITS (fungi) have proven to be very useful to identify microbes associated with patina, including those that cannot be brought into culture (e.g. Kuhlman et al., 2005; Lang-Yona et al., 2018; Northup et al., 2010; Parchert et al., 2012; Esposito et al., 2015). However, many of these studies have made claims about the physiological properties of environmental bacterial 16S rRNA, eukaryotic 18S rRNA or fungal ITS sequences based on their 'close' affiliation with database sequences from 'related' well-studied cultivated species that are known to, for example, capable of Mn oxidation. This is an impossible task since these markers cannot reliably identify taxa at species level, while even closely related species can have very different physiological properties.

Viability of rock patina microbes: Since even extracellular DNA can be preserved for thousands of years when adsorbed to clay minerals, there is no way of telling if the microbial sources of the sequenced environmental DNA were alive at the time of sampling or whether they have died a long time ago, and preserved as ancient DNA (see Capo et al., 2021 for a review).

ii. Microbiology of rock patina at Murujuga

The Burrup Peninsula is not only home of the world's largest collection of Holocene rock art, the region also harbors a large petrochemical industrial complex as well as fertiliser plants, which produce ammonia, urea, and ammonium nitrate. These industrial emissions can form acid rain, which potentially increases the acidity of the Burrup rocks, whereas the deposition of ammonium nitrate PM10 dust particles is expected to accelerate the growth of opportunistic microorganisms including bacteria, yeast, fungi, and lichens on the rock patina in the region (MacLeod, 2005; Giesen et al., 2014). Many of these microorganisms are expected to deposit metabolites in the form of organic acids that will contribute to the dissolution of Mn and Fe compounds from the patina (MacLeod, 2005). In cases where the rate of patina erosion from an increased acid load will outcompete the rate of patina formation, the mineral composition and colour of the patina will change, ultimately causing a reduction in the colour contrast between the carved petroglyphs and the patina background, which then will reduce the visibility of the precious rock art collection (Gordon and Dorn, 2005). Newly formed fissures and crevices in the bioweathering patina will be preferably colonised as shown elsewhere (Esposito et al., 2002) which will enable fungal hyphae to invade the softer weathering rind below the patina and erode the edges of the petroglyphs (Black et al., 2017). Based on the rate that the ferruginous surface crust had disappeared because of increasing surface acidity from pre-industrial times until 2002, it was predicted that the petroglyphs would disappear by mid-century based on the then current levels of acid emissions.

The study of MacLeod (2005) has perhaps contributed to the majority of what we currently know about the patina microbiome in Murujuga. They performed 750 surface pH measurements on rocks throughout Murujuga and reported a mean pH of 5.84 ± 0.85, which was significantly more acidic compared with reference museum samples (pH 6.8 ± 0.2), which were collected before the major industry development. The lowest pH of 3.04 was measured during the winter months on rocks located downwind of the nearby LNG plant. In addition, swab samples from the rock surfaces (collected during winter, spring, and summer) were plated on solid microbial growth media. This

study revealed a linear relationship between the number of bacterial/yeast/fungal colonies on the plates and acidity of the rocks, indicating that microbial metabolites were a likely source of the rock surface acidity. Furthermore, the number of plated bacteria increased during the wet season and with the amount of nitrate present. During the dry season the pH of the rock surface was higher and bacterial counts were extremely low while viable yeasts and fungi remained relatively abundant during both wet and dry seasons. This implied that yeast and fungi had better coping mechanisms and the ability to survive the harsh environmental conditions during summer, whereas bacteria seem to be capable of rapidly re-establishing or re-colonising the rock surfaces when more favourable growth conditions during the winter season returned (MacLeod. 2005).

However, the same bias related to the difficulty to bring extremophilic microorganisms into culture as outlined above, most likely also influenced the results of the MacLeod study. Moreover, a comprehensive overview of the diversity and composition of microbial communities associated with rock patina at Murujuga is lacking. In addition, nothing is known about the metabolic properties of the various members of the community and which functional genes are actively expressed that inform about the processes and pathways that are relevant for patina formation/bioweathering as well as for long-term survival. In addition, it also remains to be determined which members of the community are ancient versus recent as well as their growth rates, which we will determine through cultivation-independent molecular and bioinformatics approaches. In addition, the microbiome part of the project aims to generate a full understanding of to what extent various environmental parameters, which will be analysed in parallel as a team effort, contribute to a potential shift from a patina forming microbiome (if present) to a patina weathering microbiome at Murujuga. We will address these knowledge gaps using our state-of-the-art multidisciplinary approach that are described in detail in section 3.3.3. of this study design.

2.1.4 Spatial datasets

It has been found that systematic errors or offsets exist in some spatial location data for the region, which has led to issues such as emission source locations being incorrect. For example, the Gorgon Gas Development Air Quality Assessment report (prepared by SKM, Appendix B, p49-50) shows a location mismatch of ~120 m for about half of Karratha Gas Plant (KGP) stacks, and ~200 m for the Yara stack. For the dispersion modelling this is smaller than the grid size, so unlikely to make a difference. However, it is important to correct this for the higher resolution CFD (computational fluid dynamics) modelling we are proposing, as it would be a significant source of error.

2.1.5 Eh-pH-chloride measurements

A number of studies (Bednarik, 2002; 2007; Black et al. 2017; Macleod 2005; Smith et al. 2020) have pH monitoring data, and in some cases, redox potential (Eh), of the rock surfaces at Murujuga and inferred the consequences of pH variations for the mineralogy of the patina that is removed to form the rock art. The patina takes its colour from iron (Fe) and manganese (Mn) compounds which are combined with clay minerals and cemented by organic material in places.

The chemical stability of these compounds is sensitive to pH and Eh of the local environment, which measure the acidity (proton de-availability) and electron availability, respectively. Transfer of protons and electrons amongst Fe and Mn bearing minerals and aqueous solutions in contact with the rocks has the potential to destabilise Fe and Mn bearing minerals. If the Fe and Mn minerals are dissolved, then the patina loses its colour and the rock art loses definition. Conversely, if Fe and Mn bearing minerals precipitate rapidly, then the rock art is obscured by mineral formation and the rock art is lost.

It has been proposed (Macleod, 2005) that deposition of compounds related to the oxides of nitrogen (NOx) and oxides of sulfur (SOx) produced by industrial and shipping emissions at Murujuga, or the production of organic acids might decrease the pH of rock surfaces. Macleod (2005) carried out measurements of the surface pH in a three-year study that also included the concentrations of soluble ions on the rock surfaces including chloride (Cl^-), nitrate (NO_3^-), sulphate (SO_4^{2-}) and oxalate ($C_2O_4^{2-}$), and the microbiological activity at a range of field sites at Murujuga. Of relevance here, the pH and Fe and Mn concentrations of double-distilled water used to wash the surfaces of rocks were measured for field sites at Murujuga and compared with rocks from Murujuga held by the Museum of Western Australia. The results show that rocks from field sites returned lower pH values (4.3–5.3) than the museum-held rocks (6.8 ± 0.2), and that the concentration of Fe and Mn increased with decreasing pH. These findings were combined with Eh–pH measurements and diagrams and interpreted by Macleod (2005) and Black et al. (2017) as evidence that anthropogenic emissions have decreased the pH of the rocks and increased the rate of removal of Fe and Mn, thereby increasing the rate of rock art degradation. Macleod and Fish (2021) carried out similar measurements between 2019 and 2021 and concluded that the rock art surfaces did not record industrial emissions other than by an ammonia leak in 2018, and that the surfaces are highly dynamic with alternation between periods of low pH and Fe and Mn loss with periods of higher pH and growth of Fe and Mn bearing minerals. Growth and dissolution of Fe and Mn bearing minerals are attributed to a complex interplay among rainfall, nitrate production by microbial activity, an ammonia leak in 2018, and changes in speciation induced by chloride deposition.

These works provide useful insights and pose challenges for future work. For example, Macleod and Fish (2021) inferred a highly dynamic environment for Fe and Mn oxide precipitation from the year-on-year variation in patina colour recorded by Lau (2008) as part of the CSIRO monitoring. It is difficult to reconcile this finding with the very slow growth rates of desert varnish type patinas, which are of the order of microns (µm) per thousand years in arid environments, and somewhat faster in wetter settings, that are described in the review of rock varnish formation by Dorn (2019). Possible explanations are that the desert varnish paradigm is not appropriate for Murujuga, that desert varnish is more dynamic than previously thought, that the dynamic nature of Fe and Mn mineral formation and dissolution at Murujuga reflects the impacts of industrial emissions, or that the pH measurements reflect rapid processes that affect only parts of the mineral assemblage.

This latter point requires further investigation. Macleod and Fish (2021) state that the slopes of Eh–pH data are consistent with the equilibrium conditions for reactions involving addition and removal of Mn-bearing minerals at various times, but the data are not provided or plotted, and other reactions that would show a similar Eh–pH slopes are not discussed. While this is understandable given the limited space available in conference presentations, it is not possible to assess the validity of the conclusions with confidence, particularly given the highly unreliable nature of field Eh measurements, which seldom reach equilibrium in most environmental media because of the low concentrations of multiple redox-sensitive species and the sluggish nature of electron transfer reactions (Stumm and Morgan, 1981). The meaning of the field pH measurements is also unclear. Aho and Weaver (2006) show that rock pH measurements equilibrate on the timescale of hours, which is much longer than the minute used in the field measurements of Macleod (2005) and Macleod and Fish (2021). Additionally, the existence of reaction relationships such as those proposed by Macleod and Fish (2021) does not unambiguously identify the direction of the reaction, so any of these reactions could have proceeded from right to left, or left to right, at the time of measurement, before measurement, or subsequently.

Exploration of these issues requires further work utilising mass-balance approaches and rigorous consideration of the consequences of equilibrium and kinetically controlled processes and the effects of microbiota on these processes.

Monitoring for previous studies was relatively infrequent, with yearly or unreported measurement frequency. This frequency makes it difficult to deconvolute the effects of rainfall, nitrate production by microbial activity, and chloride accumulation, which are linked, but occur at different rates. Macleod and Fish (2021) refer to a 'normal' build-up of acidity, but it is not clear what processes are regarded as normal, or the rate and absolute amounts of acidity build-up. More regular monitoring would prove beneficial. Further, it would be beneficial to develop a repeatable technique using a consistent amount of water; Macleod and Fish (2021) refer to the use of 1–2 drops, and variable dilution of acidity might have provided a source of variability in the results. It will also be necessary to derive a reliable and defensible value for pH in the absence of pollution. Macleod and Fish (2021) use a value of 5.5 ± 0.2 but it is not clear how this value is derived, given that pollution is ubiquitous at Murujuga. Black et al. (2017) use a value of 6.8, derived from measurements of rocks collected before the production of industrial emissions and held by the West Australian Museum, but these rocks would have been dry for many years, so the effects of microbiological activity, which is inhibited by dry conditions, would be less than at the field sites, preventing meaningful comparison among the measurements.

A robust interpretation of pH measurements requires a detailed characterisation of the minerals that form the surface patina, as proposed in the inorganic geochemistry program of work. Macleod and Fish (2021) state that the purple-black patina at Deep Gorge is trimanganese tetraoxide (Mn_3O_4), but without description of the methods or results used as a basis for this interpretation. The description is consistent with Mn_3O_4 but other minerals, including metastable precursors to the thermodynamically stable oxides, must also be considered.

The role of chloride ions as a control on pH and mineral stability remains an open question. Macleod and Fish (2021) data show that a decrease in pH is associated with a decrease in the concentration of chloride ions for six of the seven monitored sites. This relationship between pH and Cl^- is interpreted as an indication that acidity produced by microbiological processes is buffered by the presence of Cl^- ions. However, the speciation of the pH buffer is not stated; bicarbonate-related buffers are plausible, and Na (sodium) would play a role in these through the sodium bicarbonate ($NaHCO_3^-$) ion, but the precise causal relationships are not identified and alternative reasons for the relationship were not discussed. For example, Macleod (2005) proposes that Cl^- is removed from the rock surface by integration into iron oxyhydroxides such as lepidocrocite (γFeO.OH), which can hold up to 18 wt.% Cl within the crystal lattice, and the formation of iron oxyhydroxides is intimately linked to pH variations.

It is difficult to reconcile the proposed causal relationship between Cl^- and pH with the proposal that acidity produced by microbial activity is rinsed away by rainfall events, increasing the pH. Under these circumstances, Cl^- would be expected to increase as a result of sea spray between rainfall events and an increase in Cl^- would be associated with a decrease in pH, but the data show the opposite trend. Of course, it is likely that there is an interplay between a range of processes including salt deposition, microbial activity, and industrial deposition, but the frequency of pH measurements is insufficient to deconvolute the processes.

There are also knowledge gaps around the relationship between NOx compounds in the air and nitrate on rock surfaces. Macleod and Fish (2021) state that pH decreases with increasing nitrate concentration at four sites, but the method of nitrate measurement is not supplied in the methods, the data are not shown, and the reason for exclusion of three of the seven monitored sites is not given. While these omissions are understandable in a short conference publication, it is difficult to determine the validity of the conclusions. It is also stated that a decrease in the NOx concentration associated with low NOx burners installed by Woodside has led to a decrease in soluble nitrate between 2003-2004 and 2017-2019, stabilisation of iron-rich minerals within the rock patinas, and a

reduction in surface acidity, but the data to support these statements are not provided. However, the statement is difficult to reconcile with the similarities in recorded pH. The pH range recorded by Macleod (2005) and Black et al. (2017) is 4.3–5.3, similar to that recorded by Macleod and Fish (2021) for 2019. Higher and lower values are recorded for 2017 and 2018 and attributed to an ammonia leak and cyclonic rainfall.

2.2 Data and information gaps

2.2.1 Organic geochemistry

No previous organic geochemical studies of Murujuga have ever been reported to our knowledge. Data and information gaps which can be filled by organic geochemistry (see section 3.3.4) include:

- molecular and stable isotopic composition of organics from the various emission sources (natural and anthropogenic) including solid, liquid and gas mixtures
- possible transport mechanism/s of organics (solid, liquid and gas mixtures) to rock surfaces, and changes in molecular/stable isotopic composition of organics during transport
- interactions of organics with inorganics and microbiome on rock surfaces; changes in molecular/stable isotopic composition, influence on microbial growth including the role of organics in the production of metabolites
- effect of organics (and their interactions with inorganics and microbiome) on rock art degradation.

2.2.2 Inorganic geochemistry

Mineralogical characterisation of the surface/patina/varnish, weathered rind, and fresh rock present at Murujuga have been limited. Such information along with mechanical information (e.g. elastic modulus, patina/rind adhesion), is vital to understanding the thermodynamic and geochemical process which are occurring in response to natural and anthropogenic environmental challenges. Such work should form an integral part of experiments used to investigate the consequences of interactions between deposition, precipitation, and the patina. Minerals, morphologies, and textural relationships must be characterised at the micrometre and nanometre, as appropriate, to detect any early signs of modification by anthropogenic emissions.

Additionally, there is scope to examine the potential to utilise soil as an indicator of processes which may occur on the rock. One useful and novel indicator of weathering may be plutonium isotopes, which have only existed in the environment during the Anthropocene.

2.2.3 Microbiome

As discussed above, microbiome studies at Murujuga have been limited. Not only is there a very limited understanding of the microbial community compositions associated with rock patina throughout Murujuga, but it also remains to be investigated which individual members of the microbiome possess the genetic machinery and actively express genes to carry out processes that would lead to the formation and/or weathering of varnish.

With our proposed multidisciplinary approach (section 3.3.3), which combines the more traditional cultivation and SEM approaches with advanced molecular biological and bioinformatics/biostatistical toolsets and in the context of the minerology, organic geochemistry, spectral, and air quality

metadata (which will be generated by the various experts in their fields), we will be able to answer additional more specific but important outstanding questions not limited to:

1. what are the mechanisms behind the long-term survival of the extremophiles that colonise desert rock patina?
2. do soils and dust deposits represent background sources of the patina microbiome?
3. which species are predicted to be ancient versus modern constituents of the rock patina microbiome based on the level of post-mortem mutations in their DNA?
4. which microbes carry the genetic machinery that can potentially form or deteriorate rock patina and which of these genes are actively expressed and potentially translated into enzymes that carry out these processes?
5. how do the members of these complex microbial ecosystems interact at individual to whole community levels?
6. what rate do patina forming bacteria and fungi colonise the patina-free rock slab surfaces, and in what order?
7. how does the patina microbiomes vary during wet and dry seasons, and which natural versus anthropogenic organic and inorganic pollutants cause significant changes in these microbial communities and in the composition of functional genes that are predicted to be involved in patina formation/bioweathering, and what are the mechanisms behind their long-term survival?

The unprecedented level of information about desert rock patina microbiomes in the context of environmental changes that can be achieved with the novel interdisciplinary approaches is a first of its kind. Therefore, this project builds on previous work involving desert rock patina in general.

2.2.4 Air quality and source apportionment

The previous air quality studies have mostly been well conducted and received little criticism over the quality of the measurements or the methods used (Black *et al.* 2017). However, the integration between the various monitoring campaign datasets (e.g. Industry and Government/CSIRO) has been less than ideal and the rationale for site selection not fully detailed and likely opportunistic (CSIRO, 2010). Furthermore, air quality modelling studies have also been limited in scope (sources and physico-chemistry) and spatial resolution (Physick *et al.* 2002). Finally, the complete characterisation of natural and anthropogenic emissions sources and exploration of likely interactions with the rock patina has also been limited. The capture of transient events and peaks, as well as mist/dew deposition events have been limited. Such events may serve to concentrate air pollutants from the air column (Andronache *et al.*, 2006; Ladino *et al.*, 2011; Lu *et al.*, 2010; Macleod, 2005; Black *et al.* (2017).

Previous air quality monitoring and modelling data can be used to inform both the optimal selection of additional monitors and overall/meta-analyses to answer the fundamental research questions. Most importantly, however, relevant threshold values need to be developed for pollutants that are relevant to ongoing preservation of rock art, rather than the human health values applied previously.

2.2.5 Eh-pH measurements

More frequent Eh–pH and chloride ion measurements are required around major rainfall events, because it is currently difficult to deconvolute the roles of rainfall, nitrate build-up, chloride, microbial activity, and industrial emissions in the formation and removal of Fe- and Mn-bearing

compounds in the patina of the Murujuga rocks. Integration of the results with the outcomes of microbial activity monitoring, inorganic geochemistry, thermodynamic calculations, and air quality monitoring, will enable us to answer a number of questions around the relationships between key parameters that include:

- acidity production by microbial activity
- acidity production by industrial deposition
- the role of rainfall
- the role of chloride ions
- the extent of rock–fluid equilibration during Eh–pH–chloride ion measurements
- the Fe- and Mn-bearing minerals that form
- the relative roles of equilibrium- and kinetically controlled mineral formation and dissolution.

2.2.6 Chamber studies

Despite the prior studies, it remains possible that the current level of anthropogenic emissions at Murujuga may not have a measurable impact on the rock art – or an effect below the level of measurement accuracy and precision inherent in field work. If that is the case, no field study of any size will result in a statistically significant outcome. For this reason, chamber studies (beyond the limited inorganic studies mentioned above) are vital to give the project certainty in its ability to establish EQC levels.

2.2.7 Petroglyph Age

Since the condition of a petroglyph declines gradually (naturally) over time after it is created (e.g. as the engraved surface reverts to the original patina), it would be useful to know the true age (time since creation) of each petroglyph to assess the natural rate of weathering and other questions.

However, dating of petroglyphs is beyond the scope of this study, for several reasons, including the terms of contract. Dating is a challenging task, as explained in Section 3.1 below. The most reliable method of dating a petroglyph is based on the motif. Different techniques for creating petroglyphs (scoring, pecking etc) were used at different times, and these differences could also affect the rate of weathering over time, so that age is confounded with technique. Another possible confounding factor is whether petroglyphs were culturally retouched to preserve their condition.

A separate Australian Research Council (ARC)-Linkage study based at The University of Western Australia is currently addressing this gap; research arising from that study will be incorporated if pertinent and available.

The study design includes a statistically representative sample of petroglyphs, which could retrospectively be dated if a suitable technique were established. This leaves open the possibility of correlating petroglyph weathering with age, using the data collected in this study.

3 Proposed study design

3.1 Rationale for the proposed design

This section lays out the principles and strategy adopted for the design of the study leading to decisions on such matters as criteria for selecting sampling locations, sampling frequency and replication.

3.1.1 Overarching principles

Overarching principles in the design of the study include:

- respect for the cultural law of the Traditional Owners and for their custodianship of Murujuga
- insistence on rigorous methodology
- validity for the entire population of culturally altered rock
- predictive power (the ability to predict the effects of different possible levels and patterns of industrial output).

3.1.2 Variations in Murujuga rock art

The Dampier Archipelago, (Murujuga) of which the Burrup Peninsula is the principal land mass, contains the world's largest petroglyph site with an estimated one million images. It is also a place of outstanding natural and cultural values. The place continues to be of living significance to the Yaburara, Mardudhunera, Ngarluma, Yindjibarndi and Wong-Goo-Tt-Oo people whose interests on Murujuga are represented by the Murujuga Aboriginal Corporation.

On Murujuga, the rock art occurs primarily on rock of fine-grained gabbro, granophyre or to a lesser extent on dolerite, basalt and granite. Some of the nearby islands, like Legendre, comprise limestone, while on others, like Rosemary and Enderby, volcaniclastic sedimentary rocks and basalt are also present. Mulvaney (2015) recognised eight petroglyph production techniques each of which, for this study, present different scenarios for motif preservation (Table 3-1).

Shallow scratching and abrading are more suited to, and more visible on, the finer grain and shallower weathered crusts of granophyre and dolerite. These techniques are also more likely to be among the more recent art as, being shallow, they are more prone to deterioration. The relative dominance of a technique is also reflective of the rock support (type) on which the petroglyphs are produced; in areas where granophyre and gabbro are not dominant, the prevalence of scratched and abraded images reduces pecked images to about 60%.

A sample of 5,650 images found the motif types are dominated by non-figurative geometrics (40%), similar numbers of Zoomorphs (fauna: 27%) and Anthropomorphs (25%), and lesser numbers of animal tracks (8%). On the basis of variation in surface weathering and superimpositioning, Mulvaney developed a system of five variations in 'contrast-state', with the most weathered (contrast-state 1) being the oldest (illustrated in Figure 3-1). Tying this to animal species and the rising sea level around the archipelago over the past 35,000 years provided some idea of the chronology of the petroglyphs, ranging from c.20,000 years ago to the early contact period of the early to mid-nineteenth century.

Mulvaney's five contrast-states each tended to contain idiosyncratic motif types, particularly a break between land and marine types that is equated with the rising of the sea level, and the forming of the archipelago from a previously inland mountain range, beginning about 10,000 years ago.

Table 3-1 Technique percentages by lithology (From Mulvaney, 2015).

TECHNIQUE	LITHOLOGY		
	Gabbro (%)	Granophyre (%)	Dolerite (%)
Pecked (Pk)	96.4	82.6	79.3
Pounded	2.2	5.9	0
Scratched (Sc)	0.3	3.9	7.6
Abraded (Ab)	0.1	1.4	5.4
Bruised	0	2.8	0
Pk + Ab	0.9	2.2	1.1
Pk + Sc	0.1	0.9	2.2
Sc + Ab	0	0.3	4.3

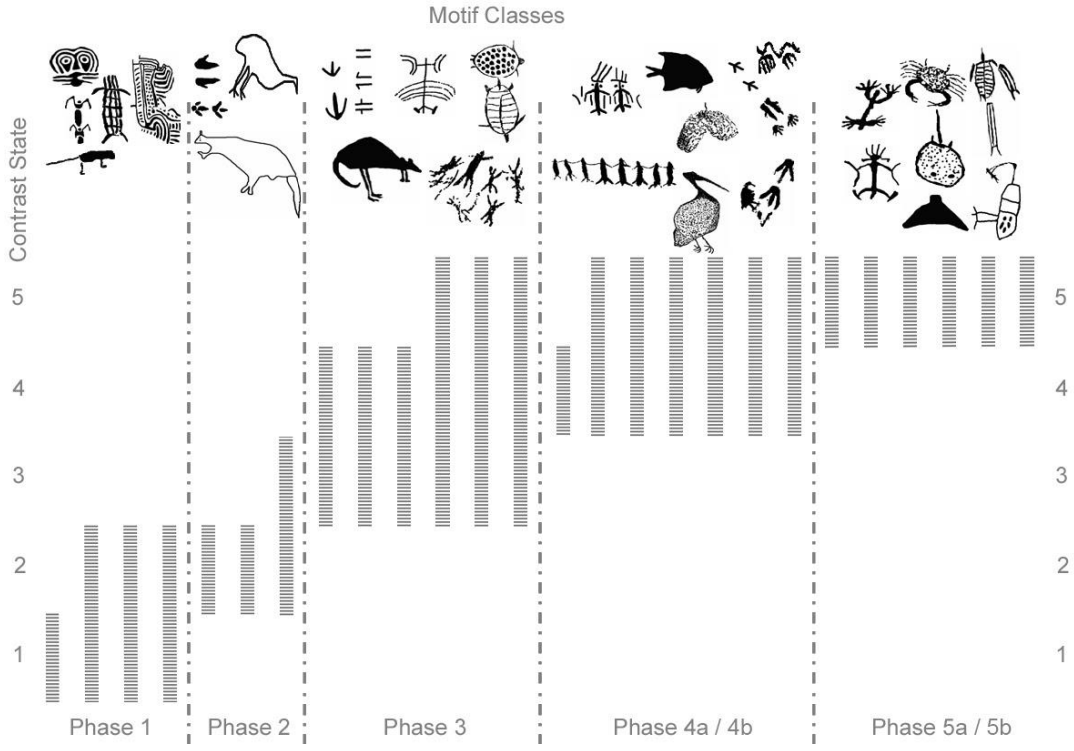


Figure 3-1 Motif classes showing the sequence of motif types, from older to younger (From Mulvaney, 2015).

3.1.3 Key considerations and constraints

The study methodology must confront substantial challenges. Those which affect the overall study design are outlined here.

- *Representative sampling under constraints*

  The selection of sites for observations must reconcile many competing requirements. In order to obtain rigorous conclusions, observations must be adequately representative of the petroglyphs, of the entire rock population and/or of the entire Murujuga region. Analysis should focus greatest attention on culturally important areas. All observation and sampling activities require approval for cultural and safety reasons. Approval will not be granted for sampling or observation at some sites – especially those sites which are the most culturally important. Practical considerations will rule out some other sites.

- *Complex inter-relationships*

  Impacts of industrial output on petroglyphs are likely to involve complex interactions between industrial output, weather, environment, and rock/patina constituents (minerals, molecular compounds, microbiological populations and physiology).

- *Need for closely located observations in the component studies*

  Atmospheric observations, destructive samples of rock, and non-destructive observations of rock surfaces, should be taken physically close together, wherever possible, in order to establish connections between them. For example, if the study is required to be capable of attributing accelerated weathering (degradation) of rock to industrial output, then we need to know how much industrial output each rock was exposed to. Pollutants from traditional activities on country (such as the use of fire for land management) and rock weathering/degradation must be measured at the same sites, or accurately interpolated from other sites nearby.

  Evidence connecting the three aspects of rock and patina composition (inorganic, organic and microbiome), requires that rock samples for the three studies should be taken very close together.

  Not all observations have to be linked in this way, but at least a substantial fraction of the observations must be, and some aspects of the statistical analysis will rest heavily on the subset of observations that are linked.

- *Detection limits*

  Laboratory techniques will be required to detect very low concentrations of chemical compounds. These low concentrations may fall below the limit of detection, unless the laboratory technique is carefully designed. Information on the chemical composition of the industrial output (e.g. the flare stacks) is needed in order to design laboratory technique so that compounds are detectable. Some of this information is available from the atmospheric modelling work commissioned by DWER (Ramboll, 2021).

- *Effect of pollutants on rock*

  An important goal of the research is to determine the effect of industrial pollutants on the rock material. One of the proposed studies is a controlled laboratory experiment in which samples of rock material will be exposed to high levels of industrial pollutants, in an effort to

artificially age and degrade the rock and patina. It is currently unknown which pollutants, which rock materials, and which chemical, biochemical or biological processes might be involved. Identifying these key elements is the goal of another part of the study.

- *Rock strength*

  The rock's rheological properties will affect preparation methods. Petrographic technique involves cutting a very thin slice of rock, polishing the cut surface, and observing it under an optical microscope (in transmitted light). Typical section thickness is 30 µm (0.03 millimetres), and if the rock is too fine-grained and breakable then it might need to be impregnated with epoxy. The alternative would be to use a 'billet' (a chip of rock about 25 x 15 x 15 mm, about the size of the top joint of a person's thumb). These considerations affect the study design.

- *Multi-component study*

  The study has several components, which are qualitatively different scientific studies, including observational monitoring, field sampling, and controlled laboratory experiments. Information from these components must be combined and integrated to obtain the final results. This is a complex task which will require advanced statistical methodology and data analysis techniques. The component studies must be designed so that they provide the data necessary for cross-referencing and integrating information between the different components.

- *Prediction objectives*

  A major objective of the study is to produce a 'dose-response' model which predicts the effects of any given level of industrial output on the petroglyphs. An evidence base for these predictions requires that we observe the effects of different levels of industrial output, in laboratory experiments and in the field.

- *Previous study data*

  Data are available from previous colour studies of the Murujuga petroglyphs, and from ongoing atmospheric monitoring of pollutant levels. These legacy data are valuable input for the design of our study. However, because of concerns expressed in Section 2, it is not a simple matter to combine the colour data from previous studies with the data from our study, to create a longer time series of colour observations. If the legacy data are included at all in the formal statistical analysis, this will need to be analogous to a 'meta-analysis' (Borenstein *et al.*, 2009) in which information is weighted by its reliability.

3.1.4 Sources of variability

'Sources of variability' are factors which have an influence on the observations. A careful consideration of sources of variability is important for study design, to determine sample sizes and allocation of sampling effort, and to avoid methodological errors such as confounding and bias (Andersen, 1990).

i. Variability across the rock surface

Photographs (Figure 3-2) show that there can be high variability in colour and texture across the surface of an individual rock. The condition of the patina may vary widely from solid, to partially degraded, to absent across the rock surface (Figure 3-2c shows regions where the surface patina

appears to have detached.) These variations may be associated with variation in mineral composition.

Other potentially important sources of variability across the surface of an individual rock include the angle of the rock surface to the sun, angle to the vertical, and angle to the prevailing wind directions.

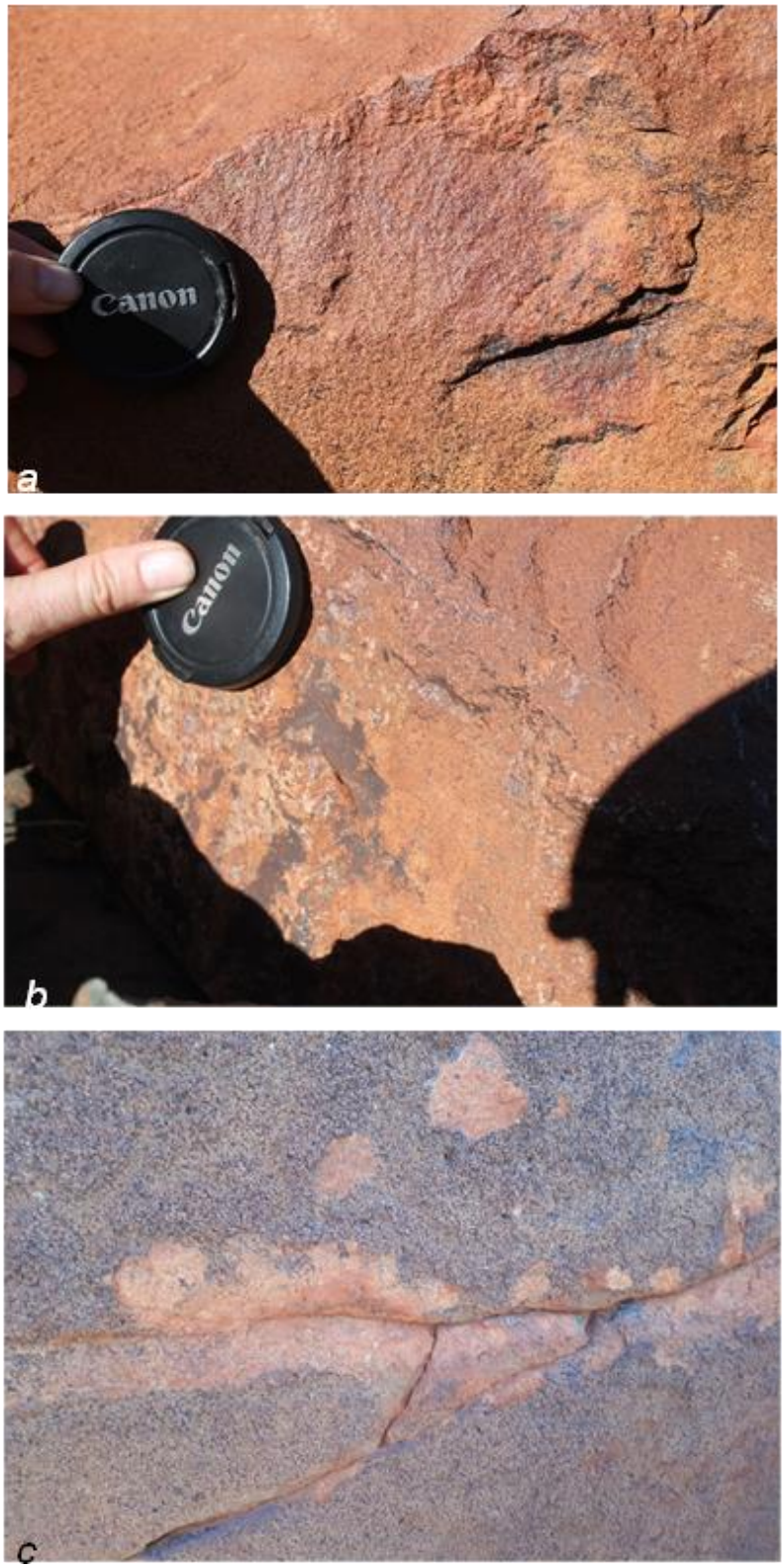


Figure 3-2 (a, b) Variation of colour, texture, surface mineral (c) patchy patina.

ii. Differences between rocks

Potentially important sources of variability or difference *between* rocks include spatial variation in rock type and rock and patina composition across the Murujuga Region, terrain slope, proximity to the coast, weathering history, physical damage and level of exposure to atmospheric pollutants.

Rock type is considered to be an important explanatory variable, but rock type classification in the field can be challenging ('cryptic') for some rocks.

Rock surfaces which have been culturally altered may be physically different from the overall population of rock surfaces, for example because the engraving process may require the rock surface to be strong and homogeneous.

Rock surfaces selected for colourimetry in previous studies may have been selected for their homogeneity, colour, texture, angle to the vertical, accessibility and other factors.

iii. Impact on colour measurement

The impact of these sources of variability on colour measurement are considered in Table 3-2.

iv. Impact on inorganic chemistry study

Table 3-3 summarises important sources of variability for the inorganic study.

Table 3-2 Sources of variability on colour measurements.

	Source of variability	Impact	Magnitude	Likelihood of impact
1	Target area variation	Measurements of colour depend on hitting the target precisely every time	Significant variation in colour readings because of inaccurate relocation	False assumptions of colour change. Errors will be minimised through the number of readings taken and the relocation method.
2	Operating temperature, instrument	Readings affected by operating temperature	Minor variations within 5 °C, but increasingly large above 10 °C	To minimise thermal error in the instrument all measurements at a given spot or target will be taken within a 5 °C window. The temperature will be measured at the first reading on each target and this will set the parameters for all subsequent readings. This thermal stabilisation is for the temperature of the rock surface to minimise the influence of thermochromism, the colour variation because of the temperature of the measured surface. The instrument itself will be maintained in a Peltier cooled container that will provide a stable temperature for the instrument.
3	Rock surface temperature, thermochromism	Minimal within 5 °C range but surface colour is temperature dependant	Below 5 °C colour variation is negligible	Record surface colour within prescribed surface temperature range. Surface temperature will be measured using infrared thermometer. This will define the recording temperature range that will be adhered to for all future readings. This can be likely be achieved by adhering to seasonal and diurnal recording times.
4	Humidity variations	Condensed water in rock pores can affect the colour measurement. Pores begin to fill at relative humidity (RH) as low as 16%, and progressively through higher RH.	The impact of RH variation has been studied by the researcher but not yet quantified in CIELAB units	Impact will only be significant where RH at recording time varies substantially. RH of recordings will be documented and an operating range established. It is anticipated that RH will follow temperature range settings provided wind direction is constant. Air off the ocean at a set temperature will be close to a constant RH.
5	Undocumented change to the surface, bird droppings, tourist impact.	A bird excrescence on a recording target will be visible for some time. It will reach a point where it is no longer perceptible but contributory to spectrometric values. Such incidents will be	Substantial colour variation will occur if a surface has been compromised by unknown material application.	Any such impacts should be readily recognised in the expanded spectral study and reinforced by dramatic increases in specific elements in the XRF studies.

	Source of variability	Impact	Magnitude	Likelihood of impact
		seen in the laboratory values but more clearly in the complete spectra and XRF element numbers. This transitory incidence may have contributed to some of the previous variability.		
6	Micro-spalling	The loss of a microscopic surface fragment in the target area will result in dramatic change to spectral results	Potentially large change in spectral data.	Macro-photogrammetry of all targets, combined with micro-photogrammetry of colour targets will correlate any spectral shift to surface morphological change.
7	Mineralisation	Growth of discrete mineral particles in the target area can change spectral data dramatically. This may be in the form of transitory soluble salt phases or permanent insoluble phases.	Potentially large change in spectral data.	Macro-photogrammetry of all targets, combined with micro-photogrammetry of colour targets will correlate any spectral shift to surface morphological change.
8	Probe contact angle	Conventional geometry for Photospectrometry is 0 or 45 ° to the vertical. Preparatory research has shown that a 20 ° orientation is optimal for a rough surface	Where a surface is not flat the returning beam may have recorded off surfaces with more than 10 ° of orientation variability. Preparatory studies show a greater error between 40 and 50 ° than between 10 and 30 °.	It is proposed that measurements be taken at 20 °rather than 45 °

Table 3-3 Sources of impacts for the inorganic chemistry study.

	Source of variability	Impact	Magnitude	Likelihood of impact
1	Concentrations of chemical components in fresh rock (SiO_2, Al_2O_3, FeO, Fe_2O_3)	The concentrations of these elements control the alteration products that form.	Concentrations may vary by ±5%	Images suggest that rocks of a single rock type (e.g., gabbro, granophyre, dolerite, etc.) have similar weathering products, but the authors are unaware of any studies which confirm this. Preliminary fieldworks suggest this is not the case. Fairweather (2019) shows that the trace elements vary by a factor of two or three, and the major elements are different by ~10% relative (based on two dolerite samples), however, using a measuring technique that has high uncertainty. The Ramanaidou and Fonteneau (2019) paper indicates lower variability.
2	Different mineral assemblages in the weathered rind. Might occur in response to differences in the rock bulk composition, the time scales of weathering, and the removal and addition of elements by open system processes	Different minerals react differently to form the patina	A range of different clay minerals and iron oxides might form.	The authors are unaware of a systematic study of the mineralogy of multiple examples of the weathered rind
3	Different minerals in the patina	Different minerals react differently with chemicals in the atmosphere or in dust	Potentially large; some iron oxides and manganese oxides are very stable, some much less so	The authors are unaware of a systematic study of the mineralogy of multiple examples of the patina. However, variations in the surface mineralogy on the scale of 10s of mm have been noted by the authors during preliminary field visits. An initial approach is to split the surface into four types: 1. dull orange (potentially iron hydroxides; 2. shiny dark orange/brown (potentially iron oxides such as hematite) 3. same as (1) with manganese oxides 4. same as (2) with manganese oxides. The thickness also varies from almost undetectable to opaque. Opacity indicates thicknesses above 100 µm .

	Source of variability	Impact	Magnitude	Likelihood of impact
4	Exposure to weathering – some rock surfaces are more protected than others, this would also affect the build-up and flushing of soluble salts.	Rock surfaces protected from surface deposition or flushing might undergo less or different reactions with chemicals in the atmosphere or dust	This is likely to be a question of degree rather than a major difference, as in the mineralogical sources above. However, some rocks could see, for example, only a few percent of the atmospheric deposition if they are sheltered.	Impact may be proportional to aspect or degree of sheltering; however, the authors could not find studies which have confirmed or refuted this hypothesis.
5	Exposure to chemicals as a consequence of proximity	Impacts as for 4.	Magnitude as for 4.	See 4.
6	Positioned where water flow is focused	High flow rates at times could cause fundamental differences in the evolution of the weathering rinds and patina	Different minerals and sequences of minerals could form where the reacting substrates are eroded and interact with large volumes of water	The authors are unaware of studies of the effects of this factor
7	Exposure to marine aerosols	Marine aerosols probably transport inorganic and organic components to the rock surfaces, including marine pollutants.	Wet and dry salt deposition and subsequent 'refluxing' through wetting and drying cycles might physically destabilise rock surfaces. The evaporation pans at the salt works are in close proximity to petroglyph sites and are a specific potential source of marine salts. Cl, SO4 and Br as ingredients in the surface films provide the potential for formation of additional secondary chemical and mineralogical phases, and effects on pH.	Unknown high. Preliminary field visits have allowed us to observe pronounced differences in the patina as a function of proximity to the ocean.

v. Impact on organic chemistry

The preceding factors will also result in variability in the organics on the rock surfaces. Molecular and isotopic composition of surface organics are likely to be highly variable on a fine scale, dependent on such factors as the surface mineral composition, exposure to emissions and the surface microbiome.

vi. Impact on microbiome

Parameters that influence the microbial communities include the following (in order of estimated importance). Quantitative parameters are:

1. moisture
2. nutrients/dust
   (a) natural versus industrial organic pollutants/dust
   (b) mineral composition of the rocks
3. salt exposure
4. temperature
5. surface pH
6. light/UV exposure.

Categorical parameters include:

1. summer/winter (would capture major differences in precipitation; it is not feasible to sample a contrasting gradient along the pollutant gradients)
2. rock type (gabbro, granophyre, dolerite, basalt, granite).

vii Impacts on Eh, pH, and chloride ion concentrations

Historic and current moisture levels influence the Eh and pH via their effects on microbial activity. Additionally, the period since cyclonic or other heavy rainfall influences the build-up of acidity, seawater-derived chloride, and industrial emission-derived N- and S-bearing compounds. All of the parameters (3.1.4 vi above) that affect the microbiome also affect Eh and pH, via their impacts on microbiome activity.

3.1.5 Principles of study design

i. There are no ‘control’ sites

The technical term ‘control’ refers to samples in an experiment which have not been subjected to any ‘treatment’. This applies to a controlled experiment, where the experimenter has the freedom to assign each individual sample or unit to be either subjected to a ‘treatment’ (such as exposure to pollution) or to be an untreated ‘control’. Any observed differences between ‘control’ and ‘treatment’ samples can then be attributed to the effect of the treatment (all other things being equal).

In previous studies of the Murujuga petroglyphs, some sites have been designated as ‘control’ sites where (it is asserted) industrial pollution exposure has been negligible. This terminology is strictly incorrect, because the researcher did not have the power to assign an arbitrary level of pollution exposure to each site. Observation at sites which experienced different levels of pollution exposure

(whether this is just asserted or was actually measured) is not a controlled experiment. Comparison of rock condition at sites which experienced high and low levels of pollution exposure, does not have the same statistical validity as a controlled experiment. Differences between rocks that experienced high and low pollution levels could be attributable to any of the other sources of variability listed above. It has been argued in some commentaries (e.g. Black *et al*., 2017) that a comparison between 'treatment' and 'control' sites is technically invalid, oversimplified, unnecessarily restrictive, and would be less likely to detect an effect.

In epidemiology and public health research, the term *'case-control'* (Breslow and Day, 1980; Borgan *et al*, 2018) refers to studies which compare human subjects who have a disease ('cases') with others who do not (controls). Statistical analysis is possible if the cases and controls do not differ in other important respects, for example, the proportion of females should be roughly the same among cases and among controls. Conduct of a case-control study, and statistical analysis of case-control data, are much more complicated and tendentious than a controlled experiment.

It could be argued that previous studies of the Murujuga petroglyphs used a case-control approach to estimate the effect of industrial output. However, it is unclear whether the cases (petroglyphs exposed to industrial output) and 'controls' (petroglyphs allegedly unexposed to industrial output) were adequately matched for other characteristics, which is the crucial requirement (Breslow and Day, 1980). Accordingly, these studies do not have a sufficiently solid foundation for inference about the effect of industrial output; however, they may be useful in the preliminary design of the study since they can provide approximate benchmark estimates of the effect size (the magnitude of the effect of industrial output).

A more important point is that, for the Murujuga petroglyphs, exposure to industrial output is a matter of degree, rather than a binary distinction between exposed and unexposed sites. Previous studies seem to have implicitly assumed that a statistical analysis would require sites to be labelled as either exposed or unexposed and would assess differences between these sites using a 'two-group' comparison between the data observed at the exposed and unexposed sites. There is no such requirement, and in this context, it would make much more sense to perform an analysis like linear regression (or a more sophisticated and flexible version of regression) in which the degree of weathering/degradation is related to the degree of exposure by a linear relationship or another functional relationship. This approach would be more statistically efficient and statistically powerful (i.e., would obtain more accurate estimates of the effect, and would have a higher probability of detecting an effect). Similar comments were made in the report by Data Analysis Australia (2016).

ii. Spatial interpolation of atmospheric monitoring observations

Data are available from previous and ongoing atmospheric monitoring of pollutant levels. Our study will add atmospheric monitoring stations at other locations

The existing atmospheric monitoring sites were selected for practical reasons, as is quite common for weather-monitoring data. Observations from these sites can be 'interpolated' or 'extrapolated' to other locations within the Murujuga region, using well-established statistical techniques such as space-time kriging (Cressie and Wikle, 2011; Wikle *et al*., 2019).

Figures 3-2 to 3-4 illustrate the concept of spatial interpolation using kriging. Figure 3-3 shows the maximum temperatures for the month of January 2009, recorded at 769 sites across Australia. Figure 3-4 shows the interpolated monthly maximum temperature at every location in Australia, computed from the 769 observations using kriging prediction. Figure 3-5 shows the estimated standard error of this prediction (the kriging error); the standard error is quite small at locations close to the original

observations, and increases as we move farther away, indicating that prediction becomes less reliable.

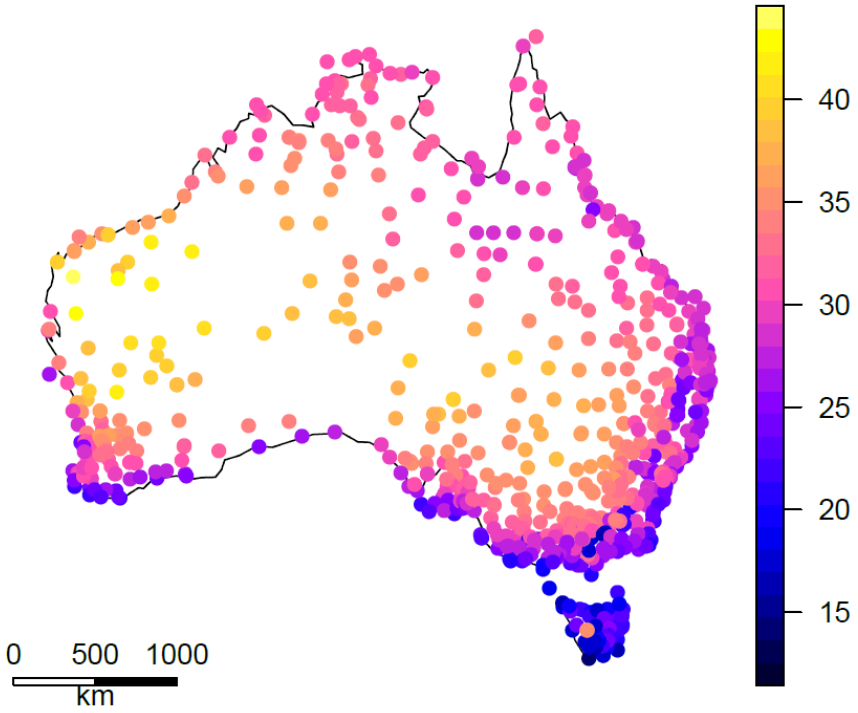


Figure 3-3 *Monthly maximum temperature for January 2009, recorded at 769 sites across Australia. Colours represent temperatures in °C .*

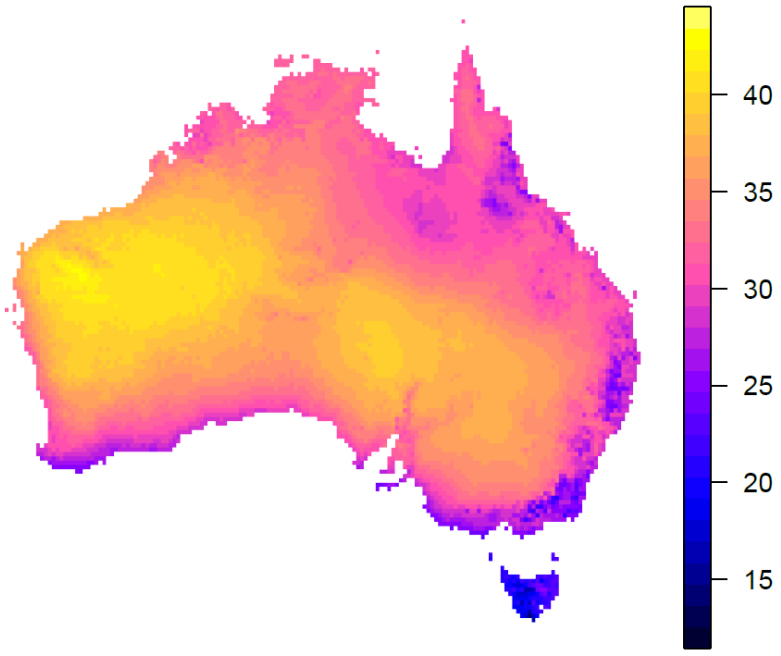


Figure 3-4 *Spatial interpolation of the monthly maxima. Kriging predictor of monthly maximum temperature at every location in Australia, computed from the 769 values in Figure 3-3. Colour scale in °C .*

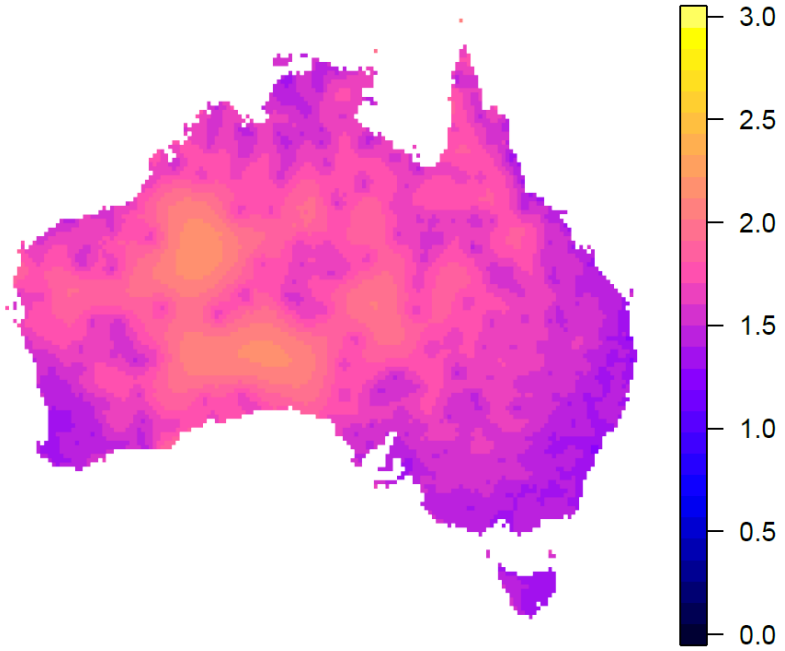


Figure 3-5 Estimated interpolation error. Standard error of the kriging predictor in Figure 3-4, for every location in Australia. Colour scale in °C .

These techniques assume, quite reasonably, that atmospheric pollution (for example NO_2, SO_2, ammonia) concentrations are spatially correlated. The accuracy and reliability of the interpolation depends on the strength of space-time correlation at different spatial distances and time lags.

Apart from the Ramboll (2021) report, there is insufficient information about space-time correlation to confidently predict accuracy, or to determine the optimal locations for additional monitoring stations. The current location/selection assumes that the Ramboll modelling is accurate; however it has been sufficiently 'overdesigned' to allow for some error and/or future/changed emissions sources.

iii. Trend estimation from a designed survey

The main statistical objective of the study design is to ensure that the study will be capable of detecting degradation over time, detecting accelerated degradation, and attributing accelerated degradation to specific causes or processes.

Urquhart *et al* (1998), Herlihy *et al* (2000) discuss the design of spatial monitoring sites for the detection of 'policy-relevant trends over time'. Crucial input information for such studies includes the components of variance associated with different sources of variation.

Urquhart *et al* (1998) give power curves for detection of anthropogenic increase in trend. Power depends on the magnitude of the anthropogenic effect. For design purposes, a back-of-the-envelope estimate of the magnitude of the anthropogenic effect could be obtained from the previous CSIRO study data. This implicitly treats the sites that were nominated as 'control' and 'exposed' sites as matched case-control data and is valid only provided they are indeed matched (i.e. are not distinguished by characteristics other than their location relative to industrial sources).

iv. Model-assisted survey inference and hybrid inference

In previous studies of the Murujuga petroglyphs, the study sites were selected without following a statistical sampling design. Consequently, it is not possible to use statistical design-based sampling inference to draw conclusions from those studies about the entire population of Murujuga petroglyphs or unaltered rocks. The selected sites are not guaranteed to be representative of the population of interest. Indeed, several potential sources of bias were identified in Section 3.1.3.

However, the information from the previous studies, or from any other arbitrarily chosen sites, can be partially utilised for model-based statistical inference. This can be regarded as a kind of interpolation or extrapolation, in which the data observed at the arbitrarily chosen sites is used to predict the values at other spatial locations. The weakness of model-based inference is that it assumes a model (for example, linear interpolation assumes a straight-line relationship) which may be wrong. Scientific conclusions derived using model-based inference always remain open to critique.

Our study aims to combine design-based inference and model-based inference. The sampling design for our study will follow recognised principles of randomised design-based survey sampling (Thompson, 2012) which guarantee valid estimates of target quantities for the target populations of petroglyphs and unaltered rocks. Sample sizes and relative survey effort will be determined using model-based estimates obtained from the previous studies (Särndal *et al*, 1992); these calculations can markedly improve the efficiency and accuracy of the study, while its basic validity rests on the design-based principles. The final data analysis will utilise data from previous studies as well as the newly gathered data, and will combine design-based inference, spatial model-based inference and statistical modelling of variance components in order to reach the objectives.

3.2 Non-Invasive monitoring of rock art

3.2.1 Selection of sites

i. Requirement for new sites

The selection of sites where rock surfaces will be monitored is important to the validity of the study. In our proposed study, the selected rocks need to be representative of the entire population of rocks (to allow statistical inference about the entire population), and their spatial locations should be spread across the Murujuga region (to allow spatial interpolation from the study sites to other locations).

Ideally it would be desirable to continue monitoring the same sites which have been visited in previous studies, because this would produce longer time-series of colour observations, making it possible to detect more gradual changes in colour. The study by Markley *et al* (2014) involved 11 ‘sites’ which are nominated rocks which bear a petroglyph; for each site, several ‘spots’ which are nominated positions on the rock surface close to the engraved part; and for each spot, a pair of colorimetric measurements, one taken on the engraved part and one taken on the unaltered part of the rock surface. The main concerns we have raised about this design are (1) the accuracy with which the colorimeters can be repositioned at the same location on the rock surface each time, and (2) that the selection of sites was not representative. Based on analysis of prior data and discussion with the key researchers who undertook the previous work, we have little to no confidence that we could accurately relocate the instrument(s) on sites used previously.

The reliability and reproducibility of the positioning of the colorimeters can be increased by improvements in technique, which are discussed in Section 3.2.3 below.

The sites used for previous studies are not sufficient, because they are not representative. We propose that, while incorporating previous sites where possible, a new set of observation sites should be selected according to a sampling procedure which guarantees a representative sample. This combined set of sites will be the basis for statistical inference about the entire population of rocks, as well as allowing spatial interpolation.

ii. Sampling protocol for survey sampling sites

Our proposed sampling protocol for sites uses techniques and principles of randomised design-based survey sampling (Thompson, 2012). This approach gives us enough freedom to make a valid design that is practically feasible and acceptable.

A map of the entire Murujuga region (including islands) has been partitioned into discrete subsets ('areas') in consultation with the Traditional Owners (Circle of Elders) and MAC, utilising the 1.3 km grid cells used in the Ramboll (2021) modelling. As explained by Thompson (2012), the partition can be drawn in any arbitrary fashion, and can be based on any relevant information. Areas may be delimited by cultural boundaries, by physical markers such as the crest of a hill, by a road or fence, or any other kind of boundary or criterion. As explained by Thompson (2012), use of this information affects the efficiency but not the fundamental validity of the method. The partition may be 'nested' (each area may be divided into sub-areas) or 'stratified' (areas may be organised into groups which have the same cultural permissions, for example).

Grid cells which are deemed unsafe for any reason, or for which permission will not be granted under any circumstances, will be designated as such, and removed from consideration.

Nested cluster sampling (Thompson, 2012) will then be used to select a random sample of areas using systematic random sampling at each stage. The use of systematic random sampling ensures that we obtain a predetermined sampling fraction. If the areas have been stratified, then each stratum may be sampled with a different sampling fraction, to increase statistical efficiency.

Each grid cell that is randomly selected for potential study would be reviewed by the study team under the guidance of the Circle of Elders to determine which individual rocks are potentially available to be studied. Considerations include cultural requirements, physical safety, accessibility, and technical constraints. Within each selected area, individual rocks will be selected according to a randomised sampling design subject to these constraints.

The resulting sampling design has both a known discrete sampling fraction (for use in estimating population totals via survey sampling inference) and a known spatial probability density conditional on the selected areas (for geostatistical purposes).

Although this design has a fixed sampling fraction rather than a fixed sample size, the sampling fraction p will be determined by the desired sample size n using $p = n/N$ where N is the number of areas delineated in the partitioning.

Owing to COVID/project restrictions, the partitioning step has not yet taken place as it requires close collaboration of key personnel, Murujuga Elders and MAC rangers on-site. Accordingly, N is not yet determined, but this is not a major consideration.

iii. Sample size

See Appendix I for sample size determination.

3.2.2 Spatial mapping of study sites and surrounds

The purpose of the spatial mapping is to capture the surrounding area (shown in Figure 3-6) in order to model the surrounding landscapes, location and orientation to be used for sub-scale CFD simulations (see below) to better capture airflow and pollutant transport in and around petroglyphs (particularly important in rapidly changing topography – gorges etc.). It can also be used to provide contextual information and spatial documentation for general use in the Monitoring Program. The primary method of data capture will be 3D terrestrial laser scanning. However, some alternative methods have been proposed to adapt to some potential issues or challenges that may be faced in the field.

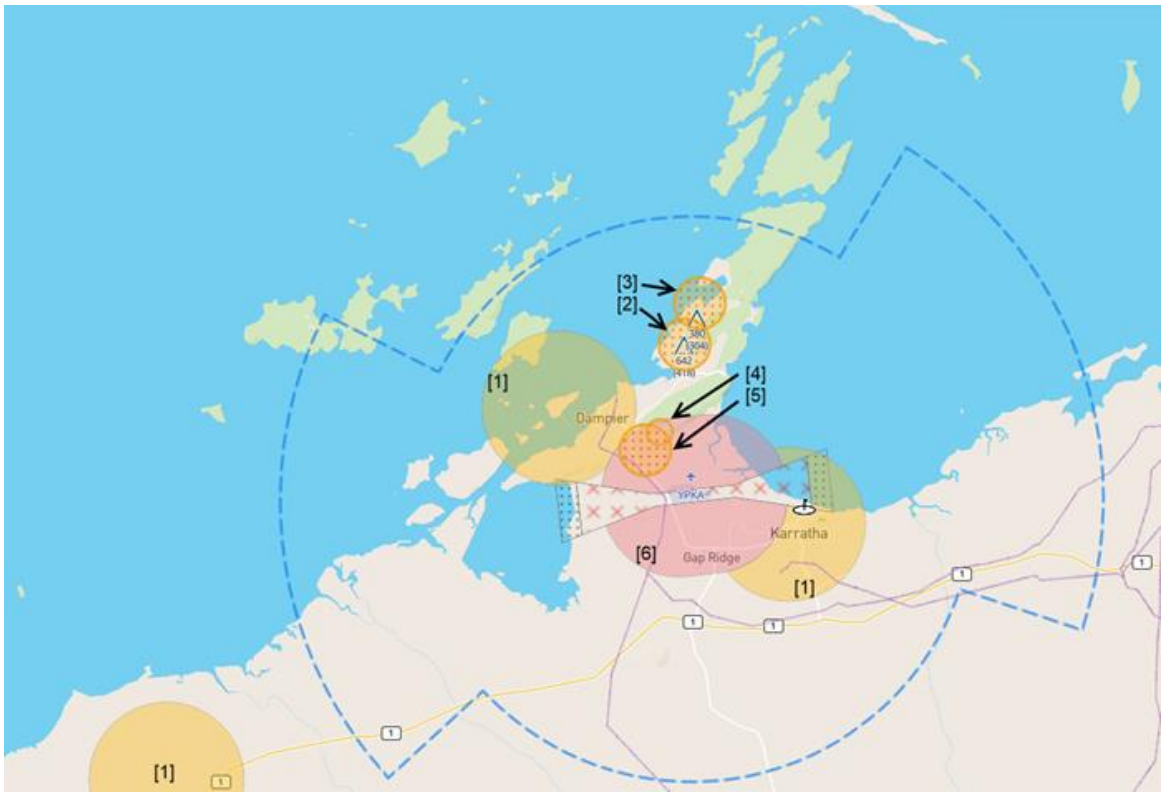


Figure 3-6 Map of the Burrup Peninsula and nearby islands.

Figure notes show to following information overlaid from the ok2fly and CASA websites:

[1] Nearby helipad, do not fly in ARR/Dep paths and over runways and taxiways without permission.

[2] D149 Danger Area, exhaust plume (24hr operation), contact Woodside Withnell Bay, check to fly.

[3] D150 Danger Area, exhaust plume (24hr operation), contact Woodside Withnell Bay, check to fly.

[4] D154 Danger Area, rifle range (NOTAM), contact Army DOTAM WA check to fly.

[5] D172 Danger Area, blasting (Monday to Friday, sunrise to sunset), contact Karratha AD OPR, check to fly.

[6] Controlled Aerodrome, permission by airport required to fly, contact AD OPR or ATC.

[7] Karratha Control Zone (Class D airspace).

i. Aim

The site capture will be done as an overview and general record, and also to provide context of the site for other parts of the Monitoring Program. The capture method will depend on the availability, accessibility, and time. As such it may be completed by a combination of laser scanning, terrestrial photogrammetry and/or UAV (unmanned aerial vehicle) photogrammetry. Capturing the surrounding area is to provide context of the surrounding landscape to the site (such as rock formations, elevation changes, vegetation, etc.). This will be more where the topography of the area is required (for air flow or water run off analysis). There is some existing data (DEM from Geoscience Australia) which can be used, but the resolution is often not sufficient for detailed modelling. It is also insufficient where accurate position and orientation is required.

Within each study site, the region between the air quality (AQ) monitor (if present) and the selected petroglyph and (non-engraved) sample rocks will be mapped at the finest detail, with coarser mapping for the remaining area within each study site. This is to enable the CFD model to accurately apportion pollutants between measured (or grid average model) values and local exposure/deposition at specific study sites. One or more survey markers will be established at each AQ site to aid accurate location of the monitor and study sites for other studies relative to the AQ monitor.

ii. Laser scanning

Laser scanning will be the primary method used to capture 3D data to model the surrounding environment of the site. It allows for areas between 10–100 m to be captured quickly and efficiently at resolutions of 10–100 mm (which can be manually adjusted to highlight or remove small details). The main advantage with laser scanning is that it captures 3D point representations of the surrounding area, which can then be modelled and classified into the component elements. Because it is a laser-based point sampling method, it also has the advantage of being able to penetrate vegetation to capture both the vegetation and the surface being occluded by the vegetation. This makes it ideal for modelling surfaces that would otherwise be occluded. However, since it is a point-based system, it does not necessarily have the resolution or speed of other methods.

For laser scanning, a Leica C10 or RTC360 scanner will be utilised. It has the ability to capture points with ± 6 mm accuracy or better, up to 100 m with typical resolution of a point very 5–10 mm at a distance of 10–20 m depending on instrument settings and time of scan. A scan from the Leica 10 will take about 10–20 minutes, while a scan from the RTC60 will take about 4–10 minutes. The trade-off between instruments is that the C10 has a longer range, while the RTC is faster to complete the same resolution scan.

As a line-of-sight instrument, the scanner is required to be set up at multiple locations around the object or scene of interest to ensure that the whole area is captured because of occlusion. For precise location and orientation of the scans, each of the scans are registered to each other, and to global coordinates through the use of survey targets (minimum of 3). These targets can be used to align the point cloud through the common targets, provide a check based on how well the geometry of the targets matches between overlapping point clouds, and to allow the points to be positioned and orientated absolutely in a global coordinate system. In order to register the 3D point information globally, the targets must be surveyed in. This requires the targets to be surveyed precisely using Raw GPS, Differential GPS/GNSS (DGPS), Real Time Kinematic GPS/GNSS (RTK), or a post processing service such as Auspos (Table 3-4).

If marks with known coordinates are available, two receivers will be used for RTK (one as a rover for the target, one as a base station on the known coordinate) to provide good accuracy of less than 20 mm. If no known marks are available, then RTK will still be used, but the base station will be left running for about four hours to be post-processed later (and survey markers established as above). This way corrected coordinates can be calculated to within 10–100 mm. If precise positioning is deemed not required, a hand-held GPS providing raw data will be used to get a rough position.

Table 3-4 Positioning system performance.

Method	Accuracy	Time	Requirements
Raw obs	+ 1 metre	Instant	Receiver
DGPS	~1-10 cm	Seconds	Subscription to Ominstar
RTK	~10-20 mm	Seconds	Another receiver set up over a known point or base station
Auspos	~10-100 mm	2-8 hours	External processing service.

iii. Drone/UAV photogrammetry

UAV photogrammetry (imagery taken from drones) is included here as an alternative method for mapping and capturing the surrounding landscape. It is proposed here as it allows for faster capture of large areas than the laser scanning (when areas are greater than 100 m). It does suffer from occlusions caused by vegetation and overhanging structures, requires permissions and approvals to be obtained, land owners and custodians (e.g. DPAW in the case of national parks) to be notified before operations commence, and the adherence to strict CASA regulations (flying heights and safety). However, it is useful for broad area mapping.

For the UAV drone, a DJI Phantom 4 Pro or a Mavic Air will be primarily used for mapping as it offers a good trade off in terms of form factor, endurance and image quality (20 mp camera). Before the flight, the ground sampling distance, flight height, overlap, and oblique camera angle need to be planned to ensure sufficient resolution and model accuracy. A flight planner software for drones (such as Drone Deploy or Pix4D mapper) will be used in this respect to plan the UAV flight path and image capture points. Normally a sampling resolution of 20 mm is achievable.

In the normal case, aerial targets will be employed (as per the targets described in the laser scanning section). These are used and surveyed to provide absolute geo-referencing and control, as well as to provide checkpoints to use in verification of the data. Targets will be placed around the edge of the area of interest (e.g. one in each corner) and at least one target placed in the middle of the area, as well as any significant elevation points (top of outcrops or bottoms of depressions). This is to ensure control to position the data, to ensure that the 3D model does not flex (bow upwards or downwards across the site) because of poor calibration of the camera, and that any significant elevation changes are captured and accounted for in the output. The targets can be surveyed by the same methods described in the laser scanning section. If precise positioning is deemed not required, the raw GPS/GNSS from the drone (delivered as Geotagged images) can be used to deliver +1 m level positional accuracy.

Optimal conditions for the flight are low winds, with overcast or cloudy conditions to eliminate shadow effects. If the sun is not occluded, then the flights should be performed near midday (to reduce the presence of shadows), and all flights for an area should be concluded in the shortest

period of time so that changing shadows do not interfere with image matching and generation of 3D points. Otherwise, the site may need to be broken down into smaller sub-regions. Also, if a point of interest is present (rock art or geology structure), or there is a lot of elevation change in the area, then consider flying an orbit around the point of interest or flying the site with an oblique camera angle (70 ° tilt for example) in a cross hatch or grid formation to improve the quality of the 3D data and redundancy.

iv. Advantages and benefits

The benefit of the laser scanning method is its ability to capture 3D geometric and position data (in the form of a 3D point cloud), being less affected by vegetation than imaging methods, and requiring less post-processing time. However, it does suffer from the lack of accurate colour information and potentially less resolution than imaging techniques at closer range (accuracy is fixed based on the instrument, whereas photogrammetry depends more on object distance). In addition, while the 3D point information can be utilised directly, it normally requires post-processing to create useful models and be used for data analysis. While it is easy to use, it is more labour intensive in the field compared with other laser method. The advantages for the UAV data collection (though imaging) is that it can cover and capture images over large areas quickly and efficiently. This enables a variety of products such as ortho-mosaics, meshed surfaces, point clouds and elevation models to be generated for a site. While the field capture is quick, some forethought and experience are needed in the planning stage to get the most out of the data (in terms of camera angles, lighting, flying height, required ground sampling distance, etc.), as these factors can affect the data density and accuracy. It benefits the most when ground control points are used to help strengthen the final solution and check the quality of the final product. It also does not penetrate vegetation when capturing data, so this can be a source of occlusion (or missing data), or erroneous modelling of the area. Processing methods and filtering procedures help to mitigate these factors. Also note that while a larger number of images may increase the area of coverage and the detail captured, it can significantly increase the processing overhead when calculating the final solution.

Terrestrial photogrammetry (close range and macro) also has similar properties to the UAV data. It has the ability to capture high resolution and accurate 3D data on the mm (and sub-mm) level, depending on the camera properties, focal length, object distance, and lighting and surface conditions. The lighting and texture of the surface is important in this regard as the image matching is reliant on the detection of feature points, and improper lighting or low surface texture will lead to a greatly reduced number of feature points, hence a coarser or poorer quality of 3D model will be generated. This can be mitigated by targets and the use of proper targets or increasing the detail on the surface (through external dusting and speckling, or filtering of the colour channels). Here scale is an important aspect to solve when creating a 3D model (to ensure measurements taken from the end product are accurate), but this can be defined through the use of a scale bar or known coordinates/distances of targets or surface features.

Table 3-5 Site capturing equipment summary.

Method	Essential	Optional
Laser scanning	Laser Scanner 1x Heavy Duty Survey Tripod	3+ Survey Tripods 3+ Targets 2x GNSS Receiver
UAV	Drone (P4 Pro or other) Mavic Air	5+ Aerial Targets 2x GNSS Receiver (for accurate

Method	Essential	Optional
		positioning)
Terrestrial Photogrammetry (macro and site)	Cameras (configuration dependant on the job site) 1x Tripod	5+ Targets (to be surveyed) 1x Scale bar (highly recommended) 1x Colour calibration chart 1+ Lighting source or shade
Micro-photogrammetry	DSLR camera 4x Zeiss microscope objective Automated scanning frame Agisoft Metashape software Blender software Photmodeller software Meshlab software CloudCompare software	Shade canopy
Colorimetry	Jaz spectrophotometer Pico tristimulus colorimeter Spectrasuite software Excel spreadsheet Voxler 3D data modelling	
Elemental surface distribution	Skyray pXRF Microsoft Excel Blender surface mapping	

3.2.3 Surface change analysis

i. Colour change

Instrumentation

Colorimetric measurement will be conducted using a Jaz Spectrometer as the primary research-grade instrument and a Pico tristimulus colorimeter as a comparative tool to establish its usefulness for ongoing MAC ranger monitoring.

The ASD Field SpecPro used in previous studies has been deemed by the current team and reviewers to be the superior instrument. The ASD has slightly lower specifications than the Jaz as a photospectrometer, but the ASD has additional Infrared spectroscopy capabilities, being effectively two instruments in one case. There is an argument that using the ASD would allow us to incorporate data from previous studies which used the ASD. However, our assessment is that the previous data cannot be satisfactorily included in the analysis.

The Jaz has been chosen both for its portability and the spectral analysis unique to a xenon light source (Thorn 2021a). It is further noted that several characterising features of the minerals forming iron/manganese films have distinguishing peaks in the UV range 250-350 nm, a function unique to xenon light sources but not the ASD FieldSpecPro.

Preparatory studies of the Pico (by A. Thorn) have shown it to provide surprisingly greater repeatability precision than the Jaz and hence its suitability to be included as a comparative tool and one that can be carried by MAC rangers on a daily basis. Colour measurement undertaken in previous studies has been criticised with regard to the selection of sites, instrumental calibration, and the high variability and low reproducibility of the colour measurements. In the present study, a rigorous process of site selection has been proposed in Section 3.2.1 above. Concerns about the instrumentation and technique are addressed here, including target relocation, surface temperature, and instrument operating parameters.

The Jaz spectrometer collects colour data via a coaxial fibre optic cable transmitting xenon light to the surface and transmitting the spectral data back to the instrument. This alone is a major improvement on target relocation and instrument operating conditions, compared with the fixed geometry of the KM and BYK instruments. The Jaz is rated to operate effectively at temperatures below 50 °C and the fibre optic ensures that the instrument is not subjected to rock heat radiation. The instrument will also be housed in a Peltier cooled insulating case to maintain a constant temperature close to the collection conditions. By dividing the day into morning and afternoon shifts, as indicated in Figure 3-7, any site can be measured at its designated ambient temperature. With shading, the surface can be stabilised to a consistent temperature within 5 °C. A 5 °C thermal variation is considered reliable for instrument stability and for thermochromatic effects on the rock surface itself. It is not yet known whether this narrow range will be achievable for all sites at all times of year, given some more remote sites have small windows of tidal accessibility and work after sunset is not permitted for cultural reasons.

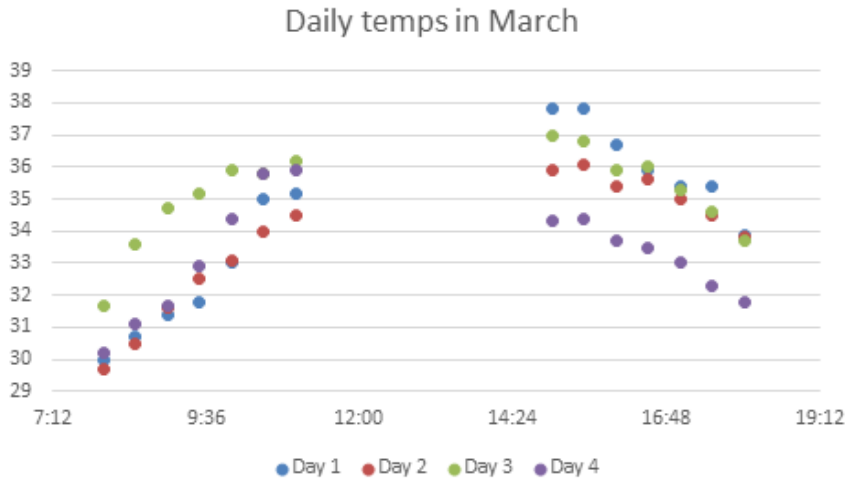


Figure 3-7 Ambient temperature against hour of day, for four days in March, in the proposed operating periods 8:00-11:00 and 15:00-18:00.

The Jaz collects spectral data in the wavelength range 220–1,100 nm, but with useful data confined to the range 250–900 nm. The data is presented as raw xenon spectral data points, from which CIELAB values can be derived within the software. The spectra will be processed both in the raw xenon spectral mode and as derived laboratory units. Xenon is specified here as it is a unique light source that emits in very distinct wavebands, the reflected results of which contain diagnostic features not seen in a daylight corrected spectrum generated by halogen or other more uniform illuminants.

Colour measurement of rock surfaces

The reproducibility of colour measurements of a spot on a rock surface is vitally important and has attracted close scrutiny in previous research and commentaries. Sources of variability, discussed in Section 3.1.4, include variation in the ambient light; error in physically relocating the sensor at the same position; spatial inhomogeneity of the rock surface itself; operator differences; and instrument variability and calibration.

This study introduces techniques that eliminate the effect of ambient light, and techniques that reduce errors in physically relocating the sensor.

The colour measurements will be conducted both on a precise point and over an area of 10 mm x 10 mm. The areal acquisition will be achieved by mounting the fibre optic head on a microscope stage micrometer, attached to the photogrammetry scanning frame. The micrometer will be progressed in 1 or 2 mm increments in a grid, centred on the precise point, to provide 25 or 81 readings over the grid.

The scanning approach will be conducted for the monitoring studies and extended to all readings if proven more reliable than single point acquisition. Much has been discussed about the unreliability of target relocation, which is a real issue using large spectrometer heads such as the Konica Minolta used in previous studies.

A remaining challenge is that spatial inhomogeneity of the rock surface affects the reproducibility, and even the feasibility, of colour measurements.

One possible strategy would be to specifically select rock surface patches (1 cm across) which appear homogeneous in colour, since these are likely to provide the most reproducible colour measurements and permit the most sensitive analysis of trends in colour. This is the convention in art museum analysis of paintings. However, this preferential selection could introduce bias in the study design. Indeed, recent findings suggest that poor surface condition is encountered more frequently with older petroglyphs; this would imply that the selection of homogeneous surfaces would favour younger petroglyphs. It seems likely that this approach has been followed in previous research studies, and this may be a previously unrecognised weakness in those studies.

Another possible strategy would be to select surface patches by a randomised sampling procedure, and to perform the colorimetry procedure on each selected surface patch, including those where a measurement is ultimately found to be unobtainable. This would be a valid study design but could be statistically inefficient (i.e. low information content obtained per unit effort) because of the expected high variability and low repeatability of colour measurements from some surfaces.

Our proposed approach combines these two strategies:

- Before any fieldwork, a comprehensive cross-validation of the ASD, Jaz and Pico instruments will be undertaken, to determine the optimum instrument for use on the project. The relocation method will also undergo a ‘blind’ validation using rock art from the region which is held at the Western Australian Museum.
- At the ‘Air Quality’ sites, rock surface patches will be selected specifically for their apparent chromatic homogeneity. This maximises the statistical power to detect trends in the colour of an individual petroglyph over time, and to cross-reference such trends to the environmental air quality which is measured only a short distance away.

- At the 'Representative' sites, a formal, randomised sampling procedure will be used to select rock surface patches. The colorimetry procedure will be applied to each selected patch unless it is impossible or unsafe.
- A practical, objective index of 'chromatic homogeneity' will be developed during the first year of the monitoring study. This index will be included in the list of attributes that are to be recorded about each rock that is monitored (other attributes include spatial aspect and rock type).
- At the 'Air Quality' sites, the study will also visit a random sample of rock surfaces, and record only the rock attributes (spatial aspect, rock type, chromatic homogeneity index, etc).

This approach will allow the study to incorporate information from correctly 'representative' samples that is expected to include some observations which have very high variability. The chromatic homogeneity index and other rock attributes will be used to determine statistical weights (inversely proportional to the variability). It will also allow us to reconcile the results from the two types of sites, and will increase the power of the final analysis,

Analysis of colour change

Statistical analysis will be applied both to the raw reflectance spectra, and to the derived laboratory colour values.

The raw xenon spectra provide a great deal of information about alteration processes that do not necessarily manifest as changes in visible colour. (This principle is well understood in other fields such as remote sensing, where modern `multispectral' satellites capture reflectance spectra at hundreds or thousands of wavelengths, because this greatly increases the ability to detect change.) The derived L*a*b colour values represent only the colour as perceived by the human eye. Visible colour may not change appreciably during some kinds of alterations of the reflectance spectrum and in other situations those alterations will lie outside the visible waveband. As an example, almost all white pigments in western art can be characterised by their reflectance characteristics below the visible at about 400 nm. Similar relationships exist for minerals.

The final, definitive statistical analysis of the Program data will combine the colorimetric and spectrometric data with pollution exposure data and other explanatory variables to rigorously assess evidence of degradation and evidence of anthropogenic impact.

During the monitoring studies and while the Monitoring Program is ongoing, Program members will conduct simple exploratory data analysis to detect emerging evidence of trends in colour over time, and other effects on colour. Since it is important to distinguish between natural change and anthropogenic impact, an exploratory analysis will look for simple trends, where it is provisionally assumed that natural mineralisation corresponds to a trend towards darker, redder, yellower values and towards convergence between the colours of engraved and background surfaces, whereas erosion of the manganese and iron mineral films will trend in the opposite directions. This provisional analysis will be performed using the CIELAB values, using various statistical models and including plotting the L, a, b, year, surface data values in Voxler software. Voxler displays three primary values (L*a*b) in rotatable 3D space and can attach labels and shading for two other critical attributes. Other attributes such as distance to source can be substituted into the five variables. Visualisation of some of the CSIRO data sets has already revealed trends not commented upon in their own reports, which only presented averaged colour data and only between adjacent years rather than trends throughout the study period. When studying the trends, there are many instances where the changes are numerically significant but not trending.

ii. Surface elemental distribution

Portable X-ray fluorescence (pXRF) is a non-destructive technique that reports on elemental composition, either by spatial distribution maps or numerical element counts at set locations. It is considered that within the current Monitoring Program numerical element counts meet the statistical integrity requirement best. The proposed pXRF records 45 elements simultaneously, of which no more than 15 will play a part in the analysis, both as elements of interest and known stable reference elements within this geological context.

The XRF study will examine the same surfaces as those from which colour measurements are being taken. As both techniques and all photogrammetric techniques in this study design are non-contact, there is no opportunity for contamination or surface alteration that might influence results across systems. Element distribution will confirm the findings of the spectral studies, particularly the examination of the secondary peaks within the Jaz generated xenon spectra. The combination of colour change with elemental distribution data will better clarify change mechanisms. For example, a natural darkening should be matched to an increase in manganese or iron in relation to stable ions such as zirconium (Zr) and niobium (Nb). A colour shift that is matched to an increase in sulphur might be more confidently interpreted as a result of that elemental shift. In addition to precise location measurements to compound surface change at the nominated targets, the XRF will be used to build an element distribution of the various phases of the rocks, from the core to the depletion crust to the outer enriched film that provides the all-important contrast to make the engravings more visible. Spatial mapping of whole surfaces of select engraved surfaces will also be undertaken to more clearly associate colour change with composition. Typical Murujuga surfaces consist of a range of coloured areas, some of which have formed since the surface was broken through. This variability will not be understood from pre-set target analysis and only fully understood with a more free-ranging exploration of the surface, across all of its chromatic range. The data will be presented as distribution maps either in 2D or 3D, wherever the latter more fully describes the pattern. Typically, a surface will be analysed point-by-point where each sample location is plotted onto a labelled image of the surface. Each element of relevance will be mapped, where the findings indicate relevance to change mechanisms. The data will either be represented as real values (ppm) or as ratios of stable elements (manganese/zircon), whichever provides the most efficient picture of change.

The XRF data will also better indicate seasonal fluctuations and adventitious events such as bird or human impacts not visually discernible. A colour change corresponding to a spike in phosphorous not matched on neighbouring rocks can be interpreted as a bird rather than industrial derivation. The colour change in isolation has no ability to understand this impact. XRF, while being less precise than the laboratory techniques described elsewhere in this document, will rely on its non-contact portability to connect the detailed analysis carried out on adjacent surfaces with engraved surfaces approved for non-contact analysis. The usefulness of this can only be discovered during early phases of the research but does not add to the outlined sampling regime above.

iii. Morphological change

The primary study of colour change, enhanced through elemental distribution change, does not provide an understanding of all forms of alteration of the surface. The deposition of a pollutant element such as sulphur, has been described until now in terms of acid dissolution, resulting in thinning and consequent lightening of the surface layer. This study acknowledges that surface change is a dynamic process not requiring anthropogenic input. While sulphur, the one element focused on for this discussion, certainly has the ability to form sulphuric acid, it has also been identified as a significant contributor to mineral formation such as gypsum crusts in granite and

sandstones. Gypsum formation on a surface can be white or black depending on the hydration state in its formation, but equally may form below the surface in capillaries such as micro-delaminations and erosion voids. The growth of gypsum below the surface is not a colour issue but purely mechanical. This has been described by Pillans and Fifield (2013) as the most significant destabilising mechanism at Murujuga. The statements are not disputed but this study has an obligation to confirm the extent of such instability. Crypto-fluorescence is not confined to gypsum but any pollutant element may combine with any surface or atmospheric element to deposit in and on the engraved rock surface. Crypto-fluorescence will not appear as a colour or element change but as a micro-spall, as described by the cited authors. Spalling is a progressive mechanism characterised in its early phase as an expansion of the surface followed by loss of surface material. Crypto-fluorescence may also manifest simply as surface growth, both seasonal and permanent.

Adapting existing technology and capabilities, selected surfaces, embracing the full width of an engraved channel and its surrounding undisturbed rock, will be scanned to develop a detailed 3D model from which various metrics can be determined. The primary imaging will be conducted through a scanning microscope over an area nominally 100x100 mm. The scanning frame can fully automate the system to complete the scan area in about 120 minutes. The scanning frame is positioned over the scan area on adjusted rubberised feet, that will be further isolated from the rock with non-abrasive Kevlar protection fabric. Once levelled to the general conformation of the study surface the scanning frame operates continuously in three axes. The Z axis runs from the upper focal plane to lower focal plane in a continuous cycle to provide 20 frames on each up or down cycle. Once a Z-cycle is completed the frame advances in the X-axis by 65% of the frame width, which will be about 15 mm. After seven steps the frame advances the same distance in the Y-axis until completion. Depending on settings the scanner will have established either 49 or 100 locations each containing a stack of eight images at predefined focal distances. Each stack has all completely out-of-focus images removed, and the remaining useful images processed through Helicon focus stacking software to produce one fully focused image from each stack. The 49–100 resultant sharp images retain sufficient exchangeable image file format data (exif format data) to be recognised in Agisoft Metashape photogrammetry software, through which a 3D image is constructed. The scan area has 3D targets placed around its perimeter for future measurement in the model. A 36 Mp image will provide 490 pixels per linear mm of image (2 µm per pixel) and from this a resolution of at least 10 µm will be clearly readable in the model.

The model will be queried in two fundamental ways. The first is a time-based comparison between models of different years using Cloud Compare software. This provides a topometric road map of dimensional difference between two versions of the same 3D form, highlighting these in heat map format. Once the heat maps are developed the original models can be compared to identify the alteration in visual terms. Distinct landmarks in the scan area will be used as alignment references. These might be protruding sand grains or other permanent features that are unlikely to disappear over the study period. Natural features can also be used as landmarks, such as indentations or ridges.

The model will also be queried to measure the profile through the engraved channel. This is achieved by direct measurement using Cloud Compare, Photomodeller or Blender, all of which will be deployed in the quest for analytical resolution, where the dimensions of any one feature can be recorded with great precision, but also by slicing the model into a thin ribbon that can then be oriented to provide a 10 µm resolved cross section of the slice. Depth, width and area of the profile, or any other dimension can be accurately measured with great precision. This provides a numerical record of incremental erosion as opposed to the quantum loss from a micro-delamination. All such losses are also highly visible in all of this examination. For reference a sand grain will be in the range 100-600 µm and hence its departure will be highly visible in the Cloud Compare study and in the profile study.

A further morphological study will be achieved using Reflectance Transformation Imaging (RTI) techniques. Like photogrammetry this is an imaging technique but differs in that the camera remains fixed while the artificial light source is moved in an orbit around the object camera common hemisphere. Two glossy spherical targets are positioned in the scene to provide lighting position data. Relying on the stereo light spots on the spheres, the RTI software is able to rebuild the surface viewed from every possible lighting position within the hemisphere. This is also the case with standard raking light but the benefit of RTI is that the imaging is comprehensive in coverage and that any lighting position can be reproduced off-site within the software. An adaptation relying on the movement of the sun and time lapse imaging has been trialled with some useful results and will be assessed during the monitoring studies.

The benefits of RTI over photogrammetry are that finer variations in a flat surface can be detected. It is not suited to highly undulating surfaces, but Murujuga provides an abundance of suitably flat cultural surfaces. The other significant benefit is that RTI images a complete surface rather than the reduced area of the micro-photogrammetric study. A cultural surface of a meter wide will show all surface undulations, and here the software can provide lighting for a particular orientation within an undulating surface. The other benefit of RTI is that it is low-technology, beyond installing the software and can be carried out by MAC rangers at any future time. MAC rangers will be fully experienced in RTI, Photogrammetry and Spectrometry using the Pico during the Monitoring Program relationship. This will be augmented by condition surveying that feeds valuable data into all of these studies.

The equipment needed for surface change study is shown in Table 3-6.

iv. Description of the quality assurance/quality control procedures

The processing pipeline for the laser scanning data will use the Leica Cyclone software to import the raw data and register it together into a single geo-referenced dataset. The first step is to import the scanner files and a list of control points into the software. If any control targets are used, these are labelled in the scan file to help with geo-referencing. A registration is created to merge all the data together using common targets and by matching common points together in overlapping regions/scans to perform a cloud to cloud (or Iterative Closest Point) matching. A check is then undertaken based on the results to ensure: a) for overlapping point clouds that there is a substantial number of detected points and the overlapping region and the RMS value between the points are within the noise levels of the point clouds; b) that the errors between registered point clouds and targets are within instrument specification; and c) cut planes and surfaces in the registered point clouds are check to ensure that the overlapping point clouds are aligned (and there are no offsets between them). Then the data can be cleaned (of vegetation or obstacles) and checked to ensure no occlusions are present. The data can then be exported for further analysis or to derive further products such as meshed surfaces, and terrain models. This last step will depend on the results required to support the other parts of the Monitoring Program and will use a combination of CAD and Modelling software (Reshaper, Descartes, Revit) or custom scripts and programs. It should be noted that some of this can be completed in the field; however, the steps should still be followed to check and validate the data as this is harder to achieve in the field.

The imagery from the close-range photogrammetry and UAV flights will be processed using photogrammetric software to combine, adjust and produce the 3D information. A combination of software is available and include Agisoft Metashape, Bentley Context Capture, and Adams Technology 3DAnalyst, and they all follow similar processes. Images (preferably raw or uncompressed) are imported. Feature points are automatically identified and matched between images to approximate the orientation and camera calibration parameters. If ground control is

present, then the targets are labelled in the images and their geo-referenced coordinates are then imported to orientate and position the camera locations and the final 3D data. If control is not available, then GPS image metadata (geotagging from the images) are used to constrain the position and final solution. For terrestrial data, this may have to be taken either from another data source (laser scanned data for example) or added in manually by other means. Regardless, scale should be fixed through either a scale bar or know distance between two points (features or targets). A bundle adjustment is then performed to produce the final solution including the camera locations, camera calibration parameters, and 3D coordinates of the matched feature points. At this step, the output will be interrogated to ensure quality of the product.

If ground control has been used, then the errors associated in their position will be checked to see if they are acceptable. If redundant control is present, then the bundle adjustment can be run independent of then, and then their calculated coordinate values from the adjustment can be compared against their true locations. The re-projection errors (or the errors between the matched points) will be verified to see if it is reasonable considering the surface sampling resolution (that is, that it is not significantly larger than the assumed point accuracy, and that there are no significant trends in the final data). The camera calibration parameters will also be checked to make sure that there are no significant variations between the assumed values. For example, the focal length is close to the camera and lens specifications, the principal point is close to the centre of the images, the radial and other distortion parameters are not significantly large, and their uncertainties are small, and there are no large correlations between parameters which would indicate poor geometry between images. Finally, the ground sampling distances, point uncertainties, and image overlap and geometry are all within specifications (and small for the uncertainties). Note that multiple image captures for a site can be run independently to see if they give similar solutions and captures across multiple sites can be compared to see if the parameters between processing runs are stable, and whether comparable results are produced. A dense image reconstruction can then be performed, and the final product (whether it is an ortho-mosaic, DEM, 3D point cloud or surface mesh) can be produced, and any filtering (such as removing vegetation) can be executed. Similar software to the laser scanning can be used to produce further digital models.

Table 3-6 Surface change analysis equipment summary.

Method	Essential	Optional
Spectrophotometry	Jaz spectrophotometer Pico tristimulus colorimeter Spectrasuite software Excel spreadsheet Voxler 3D data modelling	Scanning frame for spatial colour measurement
Micro-photogrammetry	DSLR camera 4x Zeiss microscope objective Automated scanning frame Agisoft Metashape software Blender software	Shade canopy

Method	Essential	Optional
	Photmodeller software Meshlab software CloudCompare software	
RTI	DSLR Ryobi portable LED light	
Elemental surface distribution	Skyray pXRF Microsoft Excel Blender surface mapping Surfer software	
Condition surveying	Xiaomi smartphone Various software for GPS, database and general site data management.	

While photogrammetric software follows the same general procedures and methodologies, their use will be determined based on the application. For example, Context Capture has the advantage for UAV data when using ground control points and for when the data can be combined and adjusted together with the point cloud, as well as producing detailed error analytics. 3DAnalyst has advantages when analysing rock structures and performing close range surface reconstruction and is highly tuneable in terms of processing parameters. Metashape has the advantage of being simple to use and reliable results for visualisation and providing site context.

v. Spectrophotometry

All data generated by the Jaz and Pico instruments will be stored in three formats:

1. Jaz spectral reflectance data presented in numerical format at 0.5 nm intervals. This is the generated data used to develop reflectance spectra and ultimately the CIELAB values. This data can be used by any future analyst for further querying.
2. AZ spectra files. These files contain the graphical representations of spectra that are used to study secondary peak transformations and shifts, indicating mineral alteration.
3. Spectra images in .jpg format for future study and illustration in reports. All spectra that have interpretive value will be included in reports and provided in a dedicated folder. These are static views of the spectra and have limited use for diagnostic purposes. Where a spectrum shows more than one feature of relevance an image will be saved of each view.

vi. Elemental distribution

1. Numerical data on the 45 elements will be presented for the record.

2. The subset of relevant elements will be copied out into a duplicate folder to provide easy access to the relevant material for future analysis.
3. All calculated data, that is x/y ratios and other calculations will be provided in a separate folder.
4. All visual mapping of elemental distribution will be provided in the native format of Blender, Surfer, and other software used to construct interpretative visuals. Where 3D representations are used, both the Blender file and the Metashape file used to build the form will be provided. Metashape files will be in native format and converted to industry standards such as .obj and .stl where used for downstream processing.

vii. Morphological studies

1. The morphological studies will be presented in the original native formats generated by the instrument.
2. All photogrammetric models will be further saved to industry transferrable formats such as .obj and .stl.
3. All RTI models will be made available as created, with links to the freeware used to view them.
4. All RTI key views will be saved to .jpg for interpretative use in reports and stored in labelled folders.

viii. Condition surveying

Condition surveying will add additional context to the surface morphological studies and feed into various other surface impact studies such as solar impact. It serves a broader role in the Monitoring Program in enabling the MAC rangers to develop and maintain a system of condition reporting that will become increasingly significant in monitoring the surfaces they are charged with the care of. This section has been included to document the resources required and data delivered.

1. Condition surveys will be prepared using a mobile phone or tablet. This will be maintained and stored both on-site and at the central research location (Cloudstor). All database files will be converted to standard files readable in all commercial database formats, spreadsheets, and document readers.
2. Summary data generated from the database will be presented in reports and in folders for storage.
3. GPS reference points of all sites will be stored in digital format and as a kmz file for viewing in Google Earth and other general navigation software products.
4. All digital images of sites and features will be presented in their generated format and as jpgs, filed by site and year of creation.

ix. Number of samples for spectrophotometric analysis

Refer Appendix I and II. Some of the broad site selections cover petroglyphs currently being studied by Macleod and Fish (2021). These will be included in the design if possible.

x. Departures from best practice principles of sampling program

Colour measurements and elemental data will follow conventional protocols. Analysis of the raw xenon generated spectrum is a new development that studies secondary features within the

spectrum that do not contribute to colour but act as early indicators of change. This technique has been developed by the researcher and published (Thorn, 2021), indicating peer acceptance of the technique. The xenon spectral analysis does not replace more conventional daylight corrected spectra but does provide a far more probing analysis of changes within the spectrum, especially changes within the regions outside the visible band.

Photogrammetric study of detailed forms is not a new science. However, the current study proposes to take this to the microscopic level. This again is not a new technology as 3D SEM images at very high resolution have been used for many years. The use of microscopic images to construct 3D models of small-scale surfaces will be a departure in terms of in-situ 3D imaging.

3.2.4 Eh-pH-chloride ion measurements

Eh–pH–chloride ion measurements will be performed at the same time as the colour monitoring measurements and will use similar equipment to that described by Macleod and Fish (2021). The methods will be developed on advice from the instrument manufacturers and tested extensively on the preliminary sample rocks that have been collected from Murujuga.

The starting point for method development is the method of McLeod and Fish (2021). In this method, pH measurements are made with a flat surface pH electrode (VWR model no. W7567287) connected to a TPS Aqua pH/ORP/°C meter after the addition of a fixed number of drops of distilled water between the rock surface and the electrode, and after waiting 30–60 seconds for stable readings. The electrode is calibrated at the start of each day's measurements.

We note that the 30–60 seconds proposed for the reading is very different to the timescale of hours proposed by Aho and Weaver (2006) as necessary for rock–fluid equilibration. This means that Eh–pH measurements reflect only a subset of the equilibria or reactions in progress on the rock surfaces, and might reduce the utility of the conclusions based on systems at thermodynamic equilibrium, such as those represented by Eh–pH diagrams, particularly with reference to the stability of minerals that are slow to equilibrate or have metastable precursors, such as hematite, magnetite, and some of the manganese oxides. However, the more time-consuming methods proposed by Aho and Weaver (2006) are impractical as in-situ monitoring techniques, and it is useful to derive an indication of type of rapidly occurring reactions that might occur on the Murujuga rocks. Our strategy is to begin with the methods of Macleod (2005) and Macleod and Fish (2021) and to investigate the extent to which limited thermodynamic equilibrium affects the results and interpretation by measurement of pH on the sample rocks over varying timescales, along with other variables such as the number of drops of water and temperature of the rock surface. Based on this information, we will determine an optimal measurement strategy.

A similar approach will be taken for chloride ion activity. The starting point is to measure with a calibrated Orion Thermo 1609-186881 chloride ion-specific electrode coupled to a TPS WP-90 ion-pH-mV-°C meter (or comparable instrument), and wet the rocks with two drops of 0.05 M sodium nitrate solution to stabilise the liquid junction between the sensing head and the rock surface, and continue measurements until stable readings are obtained, which in the work of McLeod and Fish (2021), occurred after about one minute.

Redox measurements suffer the same potential issues as pH measurements, in that only a subset of the possible minerals will equilibrate with the solutions on the short timescales available for in-situ monitoring. We will take a similar strategy as for the pH monitoring. We will begin with the method of McLeod and Fish (2021), using a 2 mm o.d. platinum wire electrode pushed through a sponge rubber mat, wet with local tap water, and an Ag/AgCl (3M KCl) reference electrode held near the

platinum electrode (or comparable instrument). The reference electrode will be calibrated using a saturated quinhydrone solution at pH 4.0. Tests of measurements over different lengths of time, with different wetting solutions and different amounts of wetting solutions, and comparison of the results with those predicted by equilibrium thermodynamic models, will be used to optimise the method and refine the interpretations.

Once the methods are finalised, spots for analysis will be noted and adjacent but non-identical spots will be used each time to reduce the possibility of contamination from the addition of nitrate. Preliminary testing will be used to optimise the amount of water to be added before pH measurements.

3.3 Destructive analysis of non-petroglyph rock samples

3.3.1 Overview of general principles for destructive sampling and site selection

All samples have been selected under guidance from Murujuga Elders, rangers and archaeologists. We have also obtained preliminary (calibration/validation) samples. We will endeavour (using these preliminary samples) to design all studies that require rock samples to minimise sample mass and number of samples required. It should be noted, however, that the microbiome, inorganic and organic methods proposed all require different sampling methods. It is, however, envisaged that the samples for these methods can be co-located in a small region of a rock. As there is significant evidence that the bio/geo/physico-chemical processes which occur on the rock patina are comparable on both culturally altered (petroglyphs) and unaltered rocks of all types (Dorn, 2020) and are consistent across the whole of Murujuga (subject to environmental gradients), we are able to select rocks for sampling (across the appropriate environmental and air quality gradient) which are distant from petroglyphs and perhaps in less culturally significant areas.

Insufficient information is available on the expected variability between replicate samples for some of the laboratory-based experiments to be fully designed at present. It is proposed that once we have obtained peer review and approval for the work contained in this section, a detailed validation of methods and determination of replicate variability and detection limits be undertaken. This would require approval from MAC and the Circle of Elders for the collection of a small number of preliminary rock samples. Until such time as this preliminary work can be completed, indicative sample numbers have been proposed below.

For the organic and inorganic analyses, a minimum sample size is 25 g of rock material (in triplicate at minimum) from selected sites covering the five different rock types present on Murujuga will be required. The minimum sample size for the gabbro, which is coarse-grained, is larger, because larger samples are required to ensure a representative sample at any locality. The samples identified during the preliminary fieldwork are a minimum of 500 g. Samples will be taken as cores or whole rocks. Rocks showing variation in surface colouration and mineral composition will be investigated for organic composition.

3.3.2 Inorganic geochemistry

i. Overview

Sampling will be undertaken at the sites identified in Section 3.2.1, but on rocks with no petroglyphs. All five rock types will not be present at each site, because, for example the dolerite and granite rock types are rare, and most petroglyphs are on the granophyre and gabbro.

After geological mapping of an area of ~100 × 100 m around the sample sites, aided by pXRF where necessary, a number of samples determined by the statistical team, and representing the five rock types that host rock art (gabbro, granophyre, dolerite, basalt and granite), where available, will be collected and analysed. The fresh rock (F), weathered rind (W), and patina (P) will be considered separately. The workflow is summarised in Figure 3-8. After sample site selection, samples will be collected using a diamond coring bit, 25–40 mm diameter, attached to a battery-powered angle grinder or drill. This enables precise sampling of the selected site and removes the need for hammering. Site selection will be guided by the MAC rangers and will be as unobtrusive as possible. Sample sites might include disturbed land, where sampling protocols may be less restrictive and larger sample sizes are possible.

The samples will be split into a polished thin section plus billets and any other material (Section 3.3.2ii). All material will be retained for return at the end of the current Monitoring Program. The billet will be used as a source of material for X-ray diffraction (XRD, Section 3.3.2ii). The other proposed techniques will be applied to the polished thin section (Section 3.3.2iv-vii). Analysis will start at the macro-scale and move to progressively higher spatial resolution as areas of interest are recognised and selected for further work. The results will be compared with the results of equilibrium thermodynamic models and literature estimates of weathering rates (Section 3.3.2ix), and then combined with those of other modules to establish the types of reactions that produce the weathered rind and patina, to test for any differences among sites exposed to low and high levels of anthropogenic emissions and establish proxies for the different reactions that can be measured in the field (Section 3.3.2x).

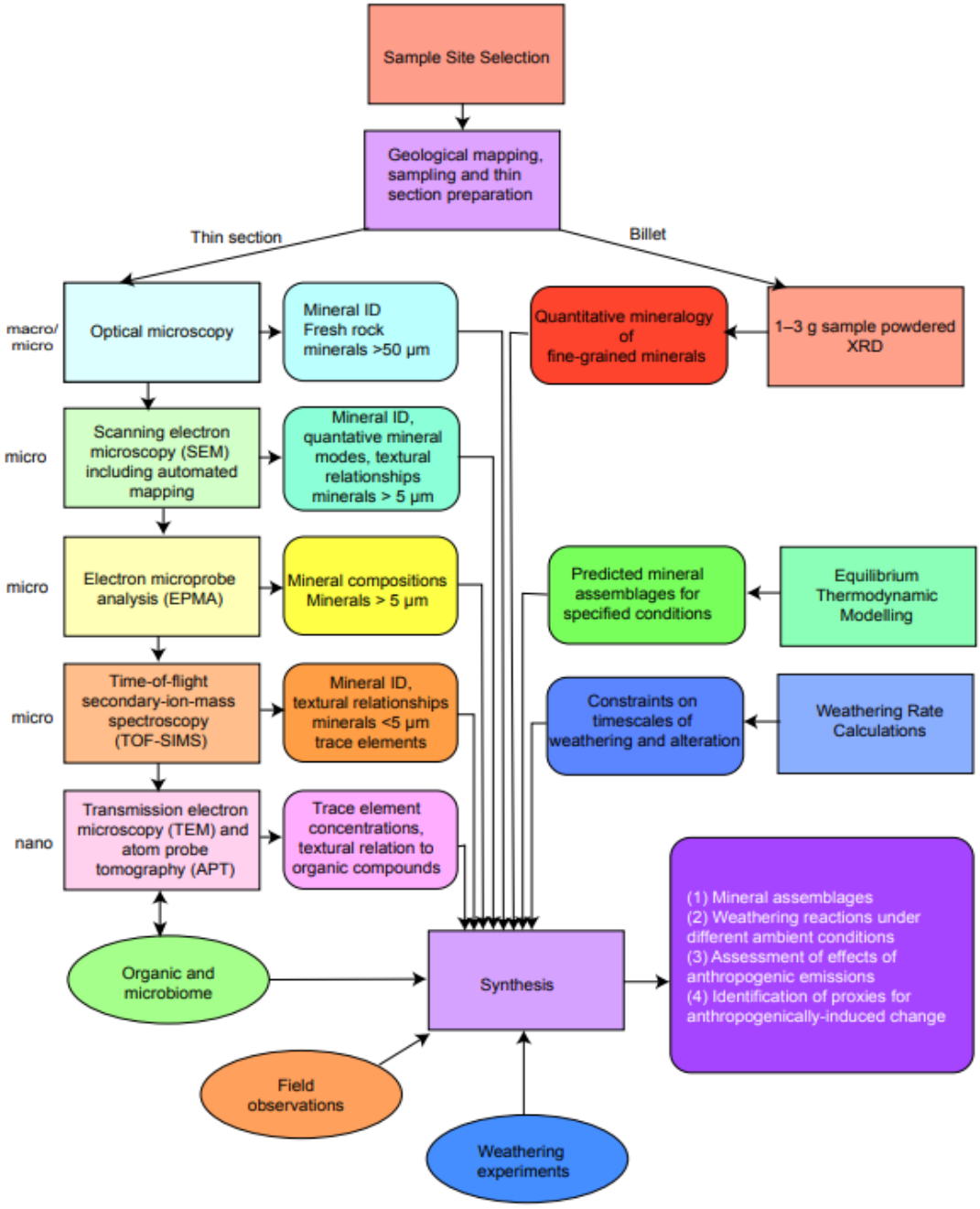


Figure 3-8 Overview of the inorganic geochemistry workflow.

ii. Production of polished thin sections and billets (F, W, P)

Samples, about 40 × 20 × 10 mm in size, that provide a representative cross-section through the patina, weathered rind, and fresh rock, and taken as described above (Section 3.3.2i), will be used as the basis of polished thin sections (Figure 3-9). The sections are glued onto a glass slide, cut using a diamond saw, and polished using a range of grit sizes and diamond paste to a thickness of 30 micrometres. Thin section preparation will be performed using water-free liquids such as kerosene to minimise dissolution of water-soluble minerals. The billets, which form the remainder of the sample after the thin section has been prepared, will be retained for further analysis.

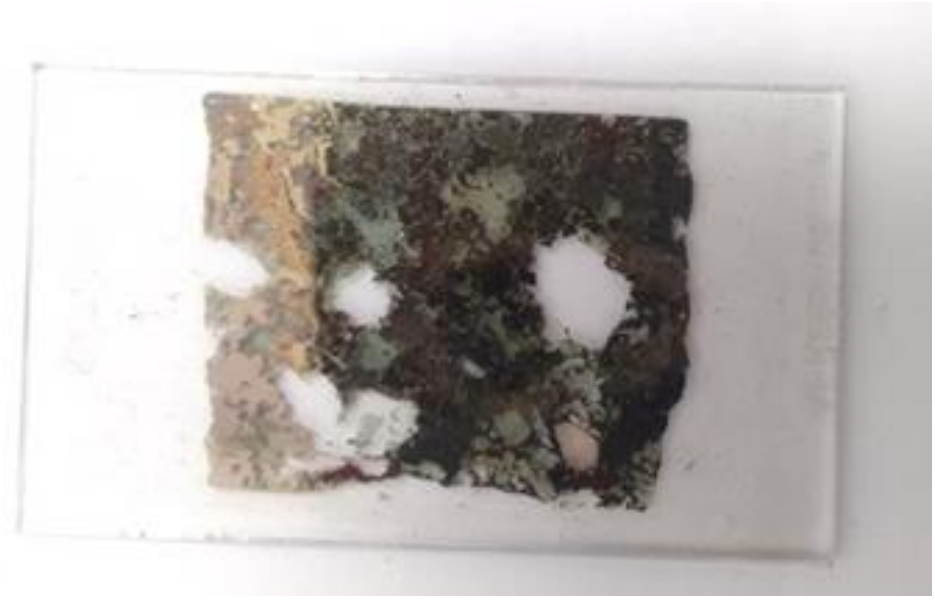


Figure 3-9 40 × 20 mm thin section of rock used for optical microscopy, SEM, EPMA, and as a source of needles and foils for TEM and APT.

iii. Identification of clay minerals by X-ray diffraction and Raman micro-analysis (W, P)

Clay minerals within the weathered rind are important because they provide a substrate for the patina, and the identity of the minerals records the rock's weathering history.. An effective way to identify fine-grained clay minerals with a grain size <5 µm , at detailed family level, is by XRD complemented by Laser Raman micro-analysis, so these techniques will be applied to the weathered rind and patina separately. X-ray diffraction measures the spacing between layers of atoms within crystals, and this spacing can be used to identify the mineral and mineral groups present. This technique can be performed on a small amount of powdered sample (~1–3 g) taken from the billet. The material will be crushed using a high-purity ring-mill to minimise contamination, micronised, and spiked with a known amount of a reference mineral such as quartz, or performed in-situ on the thin section, using the Bruker D8A XRD instrument hosted by the John de Laeter Centre at Curtin University. This instrument uses a Cu K-α source. Iron-rich samples will be analysed using the D8D instrument, also hosted by the John de Laeter Centre, which uses a Co K-α source. Issues with preferred orientation are common when clay minerals are analysed, and these will be minimised by best practice sample preparation, and post-acquisition data processing. The XRD patterns will be analysed by the Reitveld method using Bruker Topas software to quantify the amounts of the different minerals present. The uncertainties on the amounts of these minerals varies but is typically of the order of ~5% relative for minerals with modes >10%, and ~10% relative for minor minerals. Quality assurance and quality control for this technique are provided by spiking with reference materials, duplicate and replicate analyses of the unknown materials, and by monitoring of the RWP (R-factor weighted profile) and quality of fit parameters which are provided as fit diagnostics.

Laser Raman microscopy is a useful complement to XRD because: (1) it measures the vibration frequency of bonds within the minerals analysed, which differs from the crystallographic spacing information derived from XRD; (2) the spatial resolution is excellent (<1 micrometres), whereas XRD is applied to bulk mixed samples so that spatial context is lost; and (3) because Laser Raman is well suited to mapping of the spatial distribution of the minerals. Laser Raman microscopy will be performed on the polished thin sections using the Horiba® 229 LabRAM HR Evolution instrument located at the Australian Research and Resources Centre (ARRC) hosted by CSIRO, using a 600 gr/mm

grating and a 230 Synapse Visible detector. Analysis will use 532 nm incident radiation produced by a 100 mW Laser 7 231 Quantum Torus consisting of a continuous wave single frequency diode laser. The spectra will be processed (background removal, peak identification), using the MagicPlot software, and the peaks will be assigned to minerals or mineral groups using reference spectra from online databases (e.g., RRUFF database). This technique is qualitative, rather than quantitative, so quality assurance/quality control will consist of analysis of duplicate and replicate samples, and multiple processing of the same dataset.

iv. Optical Microscopy (F, W, P)

Polished thin sections will be investigated using a Nikon Eclipse LV100N optical microscope with transmitted and reflected light (Figure 3-10) to determine the identity of all minerals sufficiently large for unambiguous identification (>20 micrometre grain size), using a range of criteria such as colour, relief, cleavage, birefringence, and other crystallographic characteristics. This technique is likely to be adequate to determine the mineralogy of the fresh rock, but the weathered rind and fine-grained minerals are likely to be too fine-grained for comprehensive mineralogical identification by optical microscopy.

Figure 3-10 Nikon Eclipse LV100N optical microscope for identification of minerals in the fresh rock and preliminary investigations of the weathered rind.

v. Imaging by scanning electron microscopy (F, W, P)

The sections are usually coated with a very thin layer of carbon or platinum, a few nanometres thick. The use of carbon or platinum depends on whether further analysis for organic-related parameters is required; carbon will not be used in this case, because it can be difficult to differentiate between organic carbon that is part of the original sample and the carbon that forms part of the coating.

The samples will then be imaged using one of the scanning electron microscopes (SEMs) at Curtin University (e.g., Hitachi Desktop instrument hosted by the School of Earth and Planetary Sciences, the TeSCAN MIRA field-emission-gun (FEG) SEM, or the TeSCAN TIMA instrument, which is equipped with energy-dispersive analysis system to allow rapid compositional mapping. The MIRA has the additional capacity for electron back scatter diffraction (EBSD), which identifies minerals using their crystallographic characteristics, and is a valuable technique if polymorphs with different structures but similar compositions are present. However, the EBSD technique has limited application to very fine-grained or close-to-amorphous clay minerals and iron oxides, so these will be analysed by XRD (see below). Large area mapping will be performed on the new TESCAN CLARA. The TESCAN instruments are hosted by the Microscopy and Microanalysis Facility within the John de Laeter Centre. The Hitachi will be used for preliminary work, which includes chemical element analysis using energy-dispersive spectrometry (EDS), which enables many minerals to be recognised, based on the elements present. The MIRA instrument, which has better spatial resolution and more comprehensive mapping capabilities, will be used for more detailed work on selected areas. The detection limit for most elements on this instrument is 0.1 wt.% and the spatial resolution is 500 nm. The TIMA is set-up for rapid compositional mapping of entire thin sections, and all thin sections will be mapped using this instrument. Post-acquisition processing can be used to identify the minerals present, so long as their composition is relatively consistent. This technique should also reveal the presence of some components of the microbiome (Section 3.3.3), and the results will be integrated with the work of Marco Coolen and his team so that functional and causative relationships among the minerals and micro-organisms can be identified.

The spot size for this analysis is of the order of a few microns (µm), so very fine-grained intergrowths of clay minerals, where the individual mineral grains are less than ~5 µm will be difficult to image by this technique. However, it should be sufficient to recognise the main components of the different layers, and to image transitions between the fresh rock, weathered rind, and patina. The EDS analysis is not fully quantitative, especially for the more volatile elements such as potassium and sodium, so the element analyses produced by this technique are considered semi-quantitative.

vi. Mineral compositions by electron probe micro-analysis (F, W, P)

Quantitative mineral composition analysis will be performed on the JEOL 8530-F electron microprobe at the Centre for Microscopy, Characterisation and Analysis at the University of Western Australia. This instrument is equipped with five detectors to perform wavelength-dispersive spectrometry (WDS) and a full range of appropriate standards are available to enable the mineral compositions to be quantified with relative uncertainties of ~1% for most chemical elements. Elements and oxides of interest include SiO_2, TiO_2, Al_2O_3, FeO, MgO, CaO, Na_2O, K_2O, MnO, Cl, P, and V. Detection limits are about 100 ppm for most elements, and elements from boron to uranium can be analysed. The spatial resolution of this technique is a few micrometres, so the unique compositions of minerals with grain sizes <5 micrometres are difficult to determine by this technique. However, it is possible to combine the results of multiple analyses of mixed phases with the end-member compositions of likely candidate minerals to extract the probable end members of two- or three-component mixing.

Quality assurance and quality control will be achieved using suitable matrix-matched standards for initial calibration and analysis of secondary standards after every 10 analyses to monitor instrument drift, accuracy and precision.

vii. Time-of-flight secondary ion mass spectroscopy (W, P)

A new capability at Curtin University is the Australian Research Council-funded IONTOF M6 Time-of-flight secondary ion mass spectroscopy instrument (TOF-SIMS). This instrument has the unique capability of allowing analysis of organic and inorganic species on the same sample in the same session, so it provides new opportunities to relate the composition and location of elements hosted by inorganic materials (i.e. minerals) to the composition and location of organic species related to the presence of microorganisms. These data will be invaluable to derive causal relationships among the different minerals and microorganisms and will provide a sensitive monitor of any differences in the composition of minerals, and composition and type of microorganisms, between sites exposed to high and low anthropogenic emissions.

This instrument is purpose-built for elemental and molecular imaging and analysis. The sensitivity (detection limits and precision) are at the ppm to ppb level and the mass resolution is <30,000. The spatial resolution is excellent, with a lateral resolution of ~50 nm and a depth resolution of <1 nm, which will prove extremely useful when low volumes of sample present in the thin patina are analysed. The main purpose of this instrument is to determine spatial relationships among the different minerals, microorganisms, and organic material, so quality assurance and quality control will be checked by multiple analyses of the same material and benchmarking against lower resolution techniques.

viii. Observation of nano-scale features by transmission electron microscopy and atom probe tomography (W, P)

The techniques described above should be sufficient to determine the mineralogy, mineral modes, and, in most cases, the compositions of the minerals within the samples. However, the fine-grained nature of the minerals, the narrow width of the patina, and the expected complexity of the relationships among the minerals mean that higher resolution imaging is necessary to observe features such as growth relationships, mineral degradation, twinning, mineral inter-layering, mineral zoning, including oscillatory zoning, and element clusters that might provide an early indication of mineral breakdown or formation of new minerals. To observe these features, observation at the nanometre scale is required, and this can be accomplished by transmission electron microscopy (TEM) and atom probe tomography (APT). Atom probe tomography is a relatively new technique, and the Curtin University instrument is the only one, worldwide, that is dedicated to earth science applications (Reddy *et al.*, 2020).

For both techniques the focused-ion-beam-equipped TESCAN LYRA SEM will be used to excise selected areas, 5–10 micrometres in size, from the thin sections and used to make needles, 1–2 micrometres in length (Figure 3-12, Figure 3-10) or TEM lamellae, ~5 x 5 µm in size, and ~100 nanometres (nm) thick (Figure 3-11).

The TEM will be performed using the FEI Talos FS200X G2 FEG TEM hosted by the John de Laeter Centre at Curtin University. This TEM is equipped with highly sensitive Super-X EDS detectors for elemental analysis, and can perform 3D tomography, enabling a 3D image of the sample to be constructed. Using the TEM, which is equipped with an EDS facility, and the capacity for scanning-TEM, we can map the distribution of elements within the lamellae, at a spatial resolution of 0.14 nm.

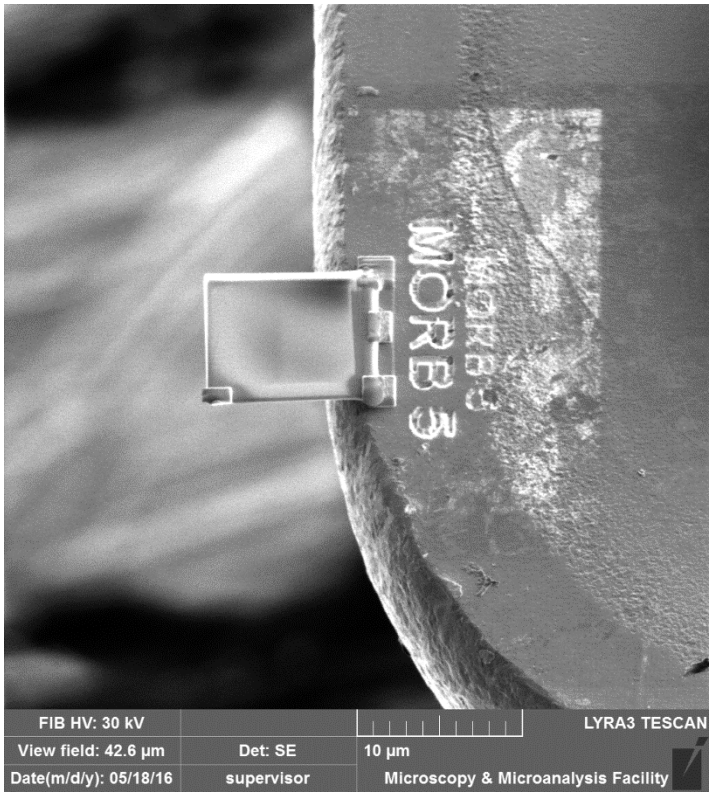


Figure 3-11 Typical TEM lamellae cut using the focused ion beam (FIB) facility at the Microscopy and Microanalysis Facility at the John de Laeter Centre, Curtin University. The lamella is ~5 micrometres in size, and a few 10s of nanometres thick.

Atom probe tomography will be performed on the LEAP 400X HR Atom Probe located within the Geoscience Atom Probe Facility, John de Laeter Centre, Curtin University. The spatial resolution of the atom probe is better than 1 nm, and the field of view is about 500 x 200 nm. The mass resolution is better than 1,000 (M/DM_{FWHM}), and the detection limit is ~100 ppm. Using APT, which allows the positions of individual atoms to be determined, we can map the variations in the concentrations of elements on the sub-micrometre scale and quantify their proximity to interfaces between the patina and weathered rind, and the weathered rind and fresh rock. In this way, nanoscale diffusion profiles can be recognised, and these might provide a highly sensitive indicator of the effects of anthropogenic emissions.

This method will also reveal the relationships between organic material and the minerals, so the results of this work will be integrated with the organic and microbiome analysis (Sections 3.3.3 and 3.3.4) to understand the relationships between the microbiome and the minerals. This method will provide the most sensitive indicator of changes to the minerals that characterise a geologically stable patina and will identify small morphological or geochemical changes a long time before those changes manifest on the microscopic or macroscopic scale.

Quantification of absolute timescales of change does not fall within the scope of methods described here. However, a major objective is to predict the factors that enhance or retard alteration, so that we can recognise natural rates of change and distinguish these from rapid rates of change, even where the processes are identical.

Quality assurance and quality control for these instruments will be provided by repeat analyses of minerals with well-known compositions analysed by the larger scale techniques. However, the

precision is typically less than that of techniques such as EPMA, because the amount of material analysed is much less, so the counting statistics are associated with larger errors. For this reason, we will use the nanometre-scale techniques mainly to determine the distributions of the minerals and use techniques such as EPMA to determine mineral compositions.

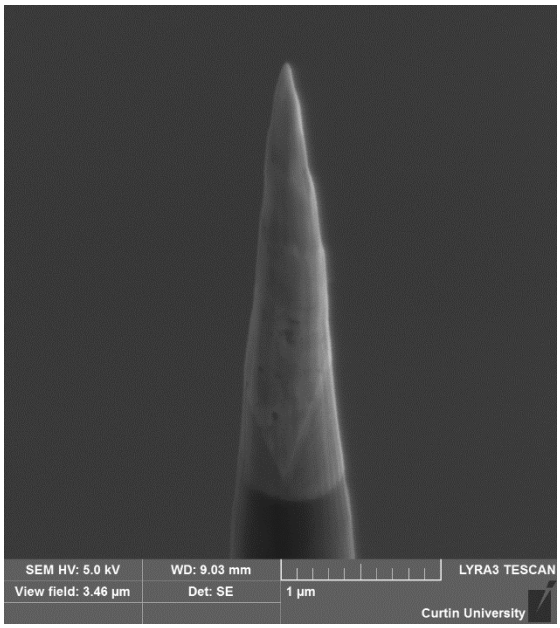


Figure 3-12 Typical atom probe needle cut using the focused ion beam (FIB) facility at the Microscopy and Microanalysis Facility at the John de Laeter Centre, Curtin University. The needle is <2 µm long.

ix. Thermodynamic calculations

Thermodynamic calculations performed using the Phreequality control or PhreeqE software will be used to determine the equilibrium mineral assemblage for the ambient conditions at Murujuga, and to investigate how these vary with changes in the composition of the atmosphere and rain related to industrial emissions. Equilibrium mineral assemblages form in environments where the timescales are sufficiently long for new minerals to form. This process also requires there to be no kinetic barriers to the minimisation of free energy, and that conditions are sufficiently constant that the timescale of reaction is small compared with the timescale of mineralogical changes. These conditions are rarely achieved in surface weathering settings, so equilibrium thermodynamic calculations provide guidelines, rather than absolute predictions, of the minerals expected in various conditions. Fortunately, much research has been devoted to the types of reactions that occur as rocks and their weathering products approach equilibrium, and the rates of those reactions, so these literature data will be combined with our observations to identify the processes and reactions that occur as the rocks react with their environment to approach thermodynamic equilibrium. Furthermore, the results of the smoke cell accelerated weathering experiments will provide additional constraints on the reactions that occur in the field.

Microorganisms, such as bacteria, play a key role in the approach to equilibrium. They can enhance reaction rates by catalysing reactions and create micro-environments that favour one reaction over another. The roles of microorganisms and the reactions that they facilitate, will be constrained by integrating the results of the thermodynamic calculations and mineralogical characterisation with those of the organic geochemistry and microbiome characterisation (Sections 3.3.3 and 3.3.4).

x. Isotope monitoring in soil and rock samples

The onset of nuclear testing in the 1940s produced 137Cs (caesium) and 239Pu (plutonium) isotopes that circulated within the atmosphere and were deposited worldwide. These isotopes have been proposed as a global marker for the beginning of the Anthropocene period. It may be possible to use 239Pu as a sensitive test of the effects of anthropogenic processes such as industrial emissions on the Murujuga Rock Art, and a pilot study to investigate this possibility is proposed here.

This study will test the hypotheses that (1) 239Pu was incorporated into soil at Murujuga; (2) 239Pu was incorporated into the patina that the petroglyphs are engraved into; and (3) there are differences in 239Pu levels among patina exposed to high and low amounts of industrial emissions.

To test these hypotheses, soil and rock samples will be taken from the sampling sites identified for the Murujuga Rock Art project where soil and rock occur in close proximity. Trial analyses of the material will be performed at the TRACE (Trace Research Advanced Clean Environment) Facility at the John de Laeter Centre at Curtin University. If the detection limits, blank measurements, and precision of the XR-Thermo Fisher Thermal Ionisation Mass Spectroscopy (TIMS) instrument hosted within this laboratory are insufficiently precise, then external laboratories with a proven track record in this technique will be sought (e.g., ANU: see Tims et al. (2010)).

The results will be used to test the hypotheses outlined above. If there are consistent and statistically robust differences in 239Pu levels among patina exposed to high and low amounts of industrial emissions, such that those exposed to high amounts of emissions, have less 239Pu, then it will be possible to quantitatively link industrial emissions to patina destruction. Preservation of the rock art relies on preservation of the patina, because it is the contrast between the patina and the underlying clay that delineates the petroglyph. Similarly, if the data show a consistent and statistically robust increase in 239Pu levels in patina exposed to high amounts of industrial emissions, relative to those exposed to low amounts, then it might be possible to conclude that industrial emissions are increasing the rate of patina production, which increases the rate at which the rock art is lost and is considered a negative impact.

Additional soil testing will also be performed in a pilot study to determine the validity of using soil as a less-invasive sampling medium for processes or deposition occurring on the rocks. Noting, however, that soil is scarce in many regions containing rock art.

xi. Synthesis

Our investigations will collect data on a range of length scales, from nanometres (using transmission electron microscopy) to hundreds of metres (using geological mapping). We anticipate that the nano and micro-scale observations will inform us of the processes and identities of the minerals: this information cannot be obtained from the macro-scale observations. Macro-scale observations will be used to quantify the heterogeneity of the processes and mineral distributions recognised from the micro and nano-scale studies. This information cannot be obtained from the smaller-scaled studies.

By integrating across the length scales, we will derive an understanding of the minerals present, the processes occurring, and the heterogeneity and, by relating the macro-scale observations to parameters such as aspect or proximity to the ocean, we will constrain the larger scale controls on those processes.

The available information will be used to construct a conceptual model based on well-constrained data sets:

1. Observed mineralogy of the fresh rock.
2. Observed mineralogy of the weathered rind.
3. Observed mineralogy of the patina.
4. Results of organic geochemistry studies.
5. Results of microbiome studies.
6. Results of smoke-cell weathering experiments – the accelerated weathering within these experiments will place constraints on the sensitivity of the system to different parameters (pH, CO_2, NO_x, SO_x concentrations, temperature).

xii. Predicted equilibrium mineral assemblage for the different ambient conditions

By comparing the observed mineral assemblages with the predicted equilibrium assemblage, precursor assemblages, and roles of the microorganism communities, we will determine the reactions that are in progress, and their likely end points. Based on these inferences, we will answer questions related to the conceptual model for weathering of the Murujuga rocks, which include:

- what are the minerals and proportions of minerals in the fresh rock, weathered rind, patina and re-formed patina?
- how do they relate to the composition of the atmosphere and water that formed them?
- are the patina and weathered rind affected by air pollutant levels?
- how are these reactions and relationships affected by anthropogenic emissions?
- how do the minerals relate to the microbiome?
- how are the minerals related to the colour and surface roughness?
- what is the nature/state of each process under natural weathering?
- what are the interactions that occur among the biogenic and abiogenic processes?
- what are the impacts of the predicted reactions on the condition of the petroglyphs?
- what minerals that provide the best proxies for the extent of the weathering reactions?

Our conclusions will be tested against the in-situ surface studies and tested for consistency against those from other modules within the Monitoring Program, such as the organic module, microbiome module, and smoke-cell weathering experiments.

Once the best mineral proxies are identified, we will develop a field monitoring program using in-situ techniques that:

- target the presence or absence of sensitive minerals, such as in-situ Laser Raman or Infra-Red Spectroscopy
- incorporate surface characterisation techniques that monitor reactions, such as high-resolution reflectance transformation imaging techniques.

This will enable field monitoring to be integrated with the monitoring described in Section 3.2.3 and the results will be combined to provide a holistic record of the evolving condition of the rock art.

3.3.3 Microbiome

i. Detailed description of the sampling procedures

Patina, weathering rind, background source material (soils and dust deposits)

DNA+RNA analysis: Using a battery-operated Dremel tool (subject to preliminary validation work) and sterile grinding bits, we propose to obtain 0.25 g of ground varnish samples overgrowing the main rock types (Granophyre and Gabbro) and from dolomite rock surfaces if present. Before sampling the surface will be sprayed with sterile molecular grade water to wash off dust to ensure that the pulverised patina surface can be sampled as a paste. Occasionally, we will also collect and analyse the deposited dust to elucidate if this comprises microbes that have colonised the underlying patina. Traces of varnish paste will be removed from the sampled spots using wipes drenched in 70% ethanol before the weathering (subsurface) rind immediately below the patina will be subsampled in parallel in a similar fashion. Sterile grinding bits will be exchanged between sampling to prevent cross contamination. We successfully tested this approach on a varnish covered rock at Curtin University. These samples will be transferred to sterile tubes in the field and immediately flash frozen inside a dry shipper. A dry shipper is a cryogenic storage dewar with a double wall that contains porous material to trap liquid nitrogen. This way liquid nitrogen is only present in a 'solid state' for safe handling, while the temperature inside will be maintained at a temperature of -196 °C for the duration of the sampling trip. This will be required to secure the integrity of the nucleic acids, notable the extremely labile functional gene transcripts (messenger RNA). Nearby soil samples will also be collected and analysed as a possible background source of patina microbes.

Cultivation: Using sterile swabs, small amounts of ground varnish and weathered rind material will be resuspended in 2 mL sterile peptone broth and kept on ice until returned to the lab where we will monitor seasonal changes in the number and diversity of bacteria and fungi following the plate count approach by MacLeod (2005).

SEM: In addition, small chips of rock surface and underlying weathered rock will be sampled in parallel for scanning electron microscopic (SEM) analysis.

These field samples will be obtained during the wet and dry seasons of sampling in year one of the initial study. The locations will be chosen in agreement with MAC, the Circle of Elders, and the rest of the research team, and to ensure that they differ significantly in the level of petrochemical and fertiliser pollutant exposure, which will be analysed by the team in parallel. All samples can and will be obtained from representative varnished surfaces on rocks that do not contain petroglyphs. If present, visible lichens overgrowing the rock varnish will be sampled as well as the rock surfaces below (scraping with a sterile scalpel versus Dremel tool).

Microbial populations on rock surfaces are likely to be variable at specific investigation sites because of differences in the rock fabric, and on the orientation and slope of the rock surface. To address this inter-site variability, we will augment the in-situ microbiome studies with data from experimental rock slabs that will be mounted at each site. With permission from MAC and the Circle of Elders, these slabs will be cut from one specimen of rock (per rock type) to eliminate rock fabric variability issues. A flat surface that is mounted in the same way at each site will also reduce orientation effects. The slabs can be sterilised before mounting to allow new populations of microbes to populate their surfaces. This would enable spatial and temporal population differences to be determined in areas of different air quality. These samples will be placed inside the air quality monitoring stations at deployment and subsampled annually during the monitoring program (and subsequently if they form

part of ongoing monitoring in the EQMF). Change in pH over time and the microbial deposition of organic acids on these slabs will also be measured.

ii. Experimental design

The field samples will be brought to the clean laboratory at Curtin University for subsequent DNA and RNA extraction. Library preparation and amplicon sequencing (MiSeq) of environmental barcoding genes encoding for 16S ribosomal RNA (bacteria + archaea, e.g. Orsi et al., 2017), 18S rRNA (fungi and other micro eukaryotes, e.g. More et al., 2018) and ITS (best for fungi, Usyk et al., 2017) will provide a holistic overview of the microbial communities and their relative abundances that are associated with patina as well as with the underlying (subsurface) weathering rind. The taxonomic analysis of the genomic markers (DNA) will inform about living, dormant, and dead taxa, whereas the parallel sequencing analysis of 16S, 18S, and ITS transcripts (as cDNA) will reveal taxa that were alive (metabolically active or dormant) at the time of sampling since the transcripts of these non-coding structural housekeeping genes degrade within hours to days after cell death whereas environmental DNA can survive for decades and even tens of thousands of years (e.g. Capo et al., 2021 and references therein).

The other component studies will provide data on air quality indicators measured near and on rock surfaces, (in)organic deposits of natural versus anthropogenic organics, surface mineralogy, and colour and texture of rock. We will use ordination techniques and multivariate biostatistical methods (e.g., Coolen et al., 2013; Campbell et al., 2020) to identify to what extent the measured variables (e.g. moisture, mineral composition, UV exposure, pH, exposure to light/UV, sea salt and pollutant exposure) contributed to variations in the microbial community composition.

In addition, we will plate out resuspended bacterial and fungal cells on selective solid growth media to determine seasonal changes in viable bacteria and fungi (colony counts) after MacLeod (2005). The obtained colonies will be sequenced (16S and ITS) and compared with the pristine prokaryotic and fungal communities and those that colonised the rock slabs over time. The presence of lichenised and/or microcolonial fungi in Fe- and Mn-crusted patina will furthermore be confirmed through cryoSEM at the John de Laeter Centre at Curtin University.

Moreover, small pieces of varnish-overgrown rocks will be incubated in a cloud chamber and exposed to aerosols with elevated concentrations of selected pollutants to determine the threshold concentrations of these pollutants that cause microbial overgrowth and/or bioweathering of rock varnish and to follow shifts in the microbiome and functions during the time series cloud chamber exposure experiment.

At the end of year one of the study, samples that harbour unique microbial profiles and which show a significant response to environmental conditions will be selected for more in-depth testing. These samples will undergo shotgun sequencing of environmental functional metagenomes (e.g. Kose et al., 2018; More et al., 2019; Orsi et al., 2017;) and subsequent mapping of functional metatranscriptomes to metagenome assembled genomes (MAGs) (e.g. Wong et al., 2018). This will identify the physiological potential of the most important active microbial members of the rock associated microbiomes.

iii. Number of samples

Table 3-7 shows the estimate of sample size for the microbiome study, based on the sample selection process recently performed.

Table 3-7 Sample Number estimates for each study.

Study Type	Number of Sites	Number of Rock types	Sampling Season	Number of samples
Rock Surface Microbiome	43	5	Wet + Dry	256*
Electron Microscopy	43	5	Wet + Dry	128
Rock Cubes (AQ Monitoring sites)	27	5	Annual (Wet)	135
* 64 rock samples selected. Patina and top underlying weathered interval will be sampled. The powdered samples will be divided in two fractions for the cultivation and molecular analysis.				

iv. Departures from best practice principles of sampling program design

In the detailed description below we describe the best practice in cultivation- and cultivation-independent molecular biological techniques and the use of environmental DNA and RNA for the study of microbial diversity and function in complex environmental samples. We do not deviate from the best practice principles.

v. Detailed description of the proposed methodologies, instruments, equipment, and procedures

In the clean laboratory at the WA Organic and Isotope Geochemistry Centre (WA-OIGC) of Curtin University, DNA and RNA will be simultaneously extracted from ~0.25 gram of patina and rind material as well as background soil and dust samples and from ~0.25 mL of culture materials after Coolen and Orsi (2015). The DNA pool will serve as a template for the PCR amplification of bacterial and archaeal 16S rRNA genes (Caporaso et al., 2012), eukaryotic 18S rRNA genes (More et al., 2018), and fungal ITS (Usyk et al., 2017). Barcoded libraries will then be sequenced (2x 300 bp paired end) using the MiSeq Illumina sequencer at the Australian Institute for Genomic Research (AGRF) in Perth (e.g. More et al., 2018). Abundance matrices of bacterial + archaeal and fungal amplified sequence variants (ASV) will be generated using the Quantitative Insights into Microbial Ecology pipeline (QIIME2, version June 2020) (Bolyen et al., 2019). These datasets will therefore reveal the identity of the varnish associated microbial community members down to species level as well as their relative abundances. The comparison with the microbial communities identified through the parallel sequencing of reverse transcribed 16S, 18S, and ITS transcripts will reveal which of these communities were likely to be alive at the time of sampling.

These ASV abundance matrices will form the template for subsequent ordination and biostatistical analysis using a variety of R packages (https://cran.r-project.org) to visualise and explain to what extent these microbial communities were selected by the chemistry of the rocks and/or by anthropogenic and natural gradients of organic and inorganic pollutants, which will be measured in parallel by the other members of the research team.

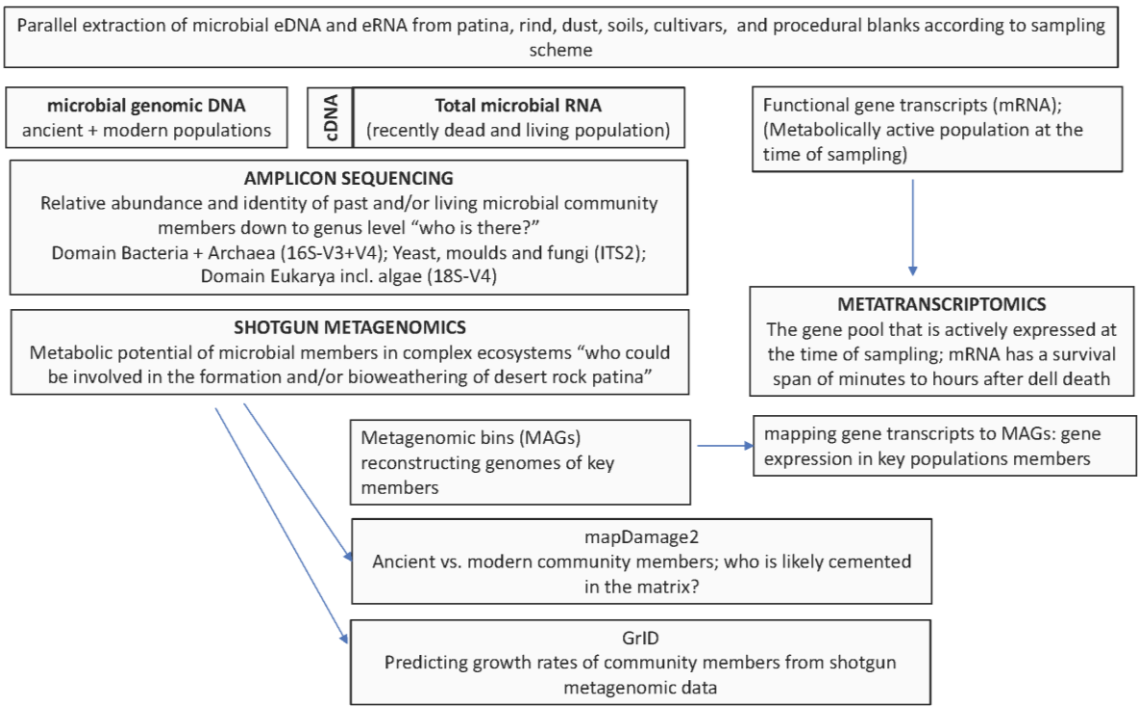


Figure 3-13 Amplicon sequencing and data analysis.

Within the cultivation laboratory of WA-OIGC at Curtin University we will plate out the inoculated peptone broth on peptone agar plates versus potato dextrose agar amended with the antibiotic chloramphenicol to selectively grow bacterial versus fungal colonies (modified after MacLeod, 2005). The plates will be incubated aerobically for 24 hours or until bacterial and fungal colonies have formed. The number of colonies will be counted, and the (entire) microbial biomass will be collected from the plates for DNA extraction and subsequent 16S and ITS profiling as described above. The viable, cultivated bacterial and fungal communities will be compared with those that were associated with the rock varnish and underlying weathered rind at the time of sampling as well as with the bacteria and/or fungi that have colonised the weathering rind of the rock slabs during the two-year time series field exposure experiments.

After the key pollutants at current ambient concentrations have been identified that may shape the rock (sub)surface microbiomes, we will place small pieces of varnish covered rock (~10 cm3; triplicates) inside an aerosol chamber (see 3.4). The rock surface microbiome will be exposed to an aerosol spray containing elevated levels of the pollutant. The threshold concentration that will lead to significant changes to the rock microbiome will be tested on the three most essential organic and/or inorganic pollutants. The same molecular approaches as outlined above will be used to follow the taxonomic and functional changes of the microbiome during the time series experiments. We anticipate following the changes for up to one year with 10 sampling time points. At t=0 and t=end, samples will also be obtained for plate counts and subsequent profiling of the viable and culturable bacterial and fungal communities as described above.

By the end of the initial study of year 1, the DNA and RNA extracts from a small number of selected samples that harbor unique microbial profiles and that showed a significant response to environmental conditions will be used as template for the preparation of shotgun metagenome and metatranscriptome libraries (e.g. Orsi et al., 2017; Kose et al., 2018; More et al., 2019; Campbell et al., 2020). Metagenome and metatranscriptome libraries will be prepared using the DNA Prep Tagmentation kit (Illumina) and the Ovation RNAseq System V2 kit combined with the Ovation

Ultralow Library System V2 kits (Nugen), respectively. Both library prep kits are designed for handling ultralow environmental DNA and RNA templates. These libraries will be sequenced (2x150 bp) on an Illumina NovaSeq 6000 system at AGRF in Melbourne, which generates up to 2.5 billion reads using the S4 flow cell and can accommodate ~100 uniquely 'barcoded' samples per run to obtain a desired >40 million sequence reads per sample. The obtained metagenomes and metatranscriptomes will be analysed using the fully automated pipeline SqueezeMeta, which offers multi-metagenome support and the co-assembly of related metagenomes and the retrieval of individual genomes via binning procedures (Tamames and Puente-Sanchez, 2019). The entire SqueezeMeta output will be uploaded into the R package SQMtools (Puente=Sanchez et al., 2020) for data visualisation. With SQMtools it is, for example, possible to generate a heatmap of the top 50 microbial taxa and genes predicted to be involved in the degradation of aromatic compounds and to produce graphs of entire pathways involved in the various processes. SQMtools will also be used to generate excel tables with reads per sample (taxonomy and functions) for subsequent multivariate analysis using third-party R packages such as Vegan (Oksanen et al., 2020) and MixOmics (Rohart et al., 2017) to study associations with the in parallel measured environmental, (in)organic parameters, and spectral colour data. Furthermore, we will map the recovered functional gene transcripts to metagenome assembled genomes (MAGs) (e.g., Wong et al., 2018; Salazar et al., 2019). The latter approach will link the actively transcribed functional gene transcripts to the key microbial players associated with rock patina and subsurface weathered rind. We will focus on genes and transcripts that are relevant for varnish formation and/or bioweathering as well as long-term survival strategies (e.g. energy metabolisms; defence mechanisms such as the production of biofilms, antibiotics, pigments; communication between cells such as quorum sensing and chemotaxis; processes involved in iron and manganese cycling, degradation of hydrocarbons/xenobiotics/aromatics; production of organic acids; dormancy such as DNA repair).

The metagenomic bins will furthermore be compared with reference genomes of closely related modern taxa in MapDamage2 (Jónsson et al, 2013) to distinguish between past versus modern taxa based on the level of post-mortem diagenetic alterations of the environmental DNA. Moreover, we will use the package Growth Rate index (GRiD) to estimate the growth rate of the rock associated microbial communities by measuring genome replication rates from shotgun metagenomic bins. The sensitivity of the approach allows to infer growth rates from rare taxa with ultralow 0.2x sequence coverage (Emiola and Oh, 2019).

These metagenomic, metatranscriptomic, and bioinformatics studies will then be continued for all field samples, slabs, and cloud chamber experiments that revealed unique microbial communities from the initial 16S, 18S, and ITS profiling. All experiments will be repeated from samples collected during the second sampling expedition.

vi. Description of the quality assurance/quality control procedures

Flash-freezing samples in the dry shipper during sampling at -196 °C and subsequent transport on dry ice is the best means to fully preserve the integrity of both DNA and the much more labile ribosomal and messenger RNA molecules. Vinyl gloves and masks will be worn at all times to prevent cross contamination with skin bacteria, but we will not attempt to sterilise the rock surfaces as this will destroy the patina associated microbiome. DNA and RNA will be extracted simultaneously to make sure that both biomolecules are derived from the same chemically and mechanically lysed microbial cells to allow a direct comparison between total versus active microbiome members. Triplicate extractions will be performed to compensate for differences in microbial compositions at the same sampling site. All PCR reactions will also be performed in triplicate to monitor differences in the amplification efficiency. Procedural blanks will be amplified and sequenced in parallel to monitor

contamination introduced during DNA extraction and the preparation of enzymatic reaction mixtures. Sequence reads that occur in both samples and blanks will be removed from the datasets.

The QIIME2 pipeline offers DADA2, a revolutionary approach to denoise sequence data including the removal of chimera's (Boylen et al., 2019).

Rarefraction curves will be generated in QIIME2 (Boylen et al., 2019) to confirm if deeper sequencing would have resulted in an increased diversity.

3.3.4 Organic geochemistry

i. Rationale

Organic geochemistry is the study of organic compounds in the environment, their sources (both natural and anthropogenic) and the alteration processes they undergo (Peters *et al.*, 2005; Volkman, 2018). Through molecular and stable isotopic characterisation of organics that have been deposited on rock surfaces, compounds may be linked back to their original sources (e.g. Dawson *et al.*, 2007; Vizthum von Eckstaedt *et al.*, 2012; Grice & Eiserbeck, 2014). The aim of this study is to identify organic compounds present on rock surfaces with the potential to cause accelerated degradation of petroglyphs, either through interactions with specific minerals (see section 3.3.2) or the microbiome (section 3.3.3). Organic geochemical analyses will be performed at the Western Australia Organic and Isotope Geochemistry Centre (WA-OIGC), Curtin University.

ii. Extraction of organics from rock, soil and dust samples

Rock samples will be separated where possible into layers; patina, weathered rind, and fresh rock. Rock samples will be ground to a fine powder using Rocklabs ring mills. Dust and soil samples will be extracted as collected. To analyse organics from rock, soil and dust samples, the powdered sample is extracted with solvents (typically a mixture of 9:1 dichloromethane and methanol by volume). The extracted mixture of organics contains molecules of biological origin (biomarkers and modern natural products) which give information on the source of the organics and post-depositional alterations (Brocks & Grice, 2011; Walters *et al.*, 2018). A range of extraction techniques are available at WA-OIGC. The most commonly used are Soxhlet apparatus (e.g. Sinninghe Damsté *et al.*, 1989; Plet *et al.*, 2016) or microwave extraction (e.g. Birgel *et al.*, 2014; Cesar & Grice, 2017). The Soxhlet apparatus is potentially more efficient, as extraction can be extended effectively indefinitely to ensure all organics are extracted (e.g. Holman *et al.*, 2012), though time requirements can be prohibitive for large numbers of samples. Microwave extraction takes much less time, but if repeated extractions are necessary then solvent requirements can be higher than for Soxhlet. Samples where only small amounts of material are available for extraction (less than 5 g) will be extracted in an ultrasonic bath (Wakeham *et al.*, 1995), as this can be done on a smaller scale than the larger Soxhlet apparatus and microwave vessels. Dust samples will be extracted using the ultrasonic bath, as these are likely to be of much lower mass than rock and soil samples.

iii. Wet laboratory processing

Organic extracts are complex mixtures, typically containing hundreds of different compounds (ranging from mass 15 up to 650 Daltons; these encompass isoprenoids, *n*-alkanes, steroids, hopanoids, fatty acids, sterols, hopanols, alkyl benzenes, alkyl naphthalenes, and polycyclic aromatic hydrocarbons), including classes that are not amenable to the most common analytical techniques (see 3.3.4 iii). It is therefore standard practice to separate organic extracts into sub-fractions before analysis. All organic extracts will be initially separated by polarity using column chromatography with silica gel (Bastow *et al.*, 2007), which is a rapid separation sufficient for most analyses. For very

complex mixtures or for analyses with more strict requirements for separation (e.g. compound-specific isotope analysis), additional separation methods will be employed such as molecular sieving (West *et al.*, 1990; Grice *et al.*, 2008) for saturated fractions and alumina chromatography (Jiang *et al.*, 2013) for aromatic compounds. The polar fraction is not amenable to analytical techniques based on gas chromatography and must be modified through derivatisation procedures before analysis. The specific technique used will depend on the type of compound to be analysed. The most common include methylation of fatty acids (Pagès *et al.*, 2014) and silylation of alcohols (Volkman *et al.*, 1992).

iv. Gas chromatography – mass spectrometry

Organic extracts will be analysed using gas chromatography – mass spectrometry (GC-MS), which is the most commonly-used technique for analysis of complex organic mixtures (Grice & Eiserbeck, 2014; Gruber *et al.*, 2020). GC-MS allows for the identification and quantification of compounds in complex mixtures of organics extracted from sediments and petroleum (e.g. Williford *et al.*, 2011; Plet *et al.*, 2016). A suite of Agilent GC-MS instruments are available at WA-OIGC, with a range of GC column types and sample inlets.

In cases where organic extracts are too complex to be analysed using conventional GC-MS, the technique of comprehensive two-dimensional gas chromatography (GCxGC-TOFMS) will be employed. GCxGC-TOFMS employs two GC columns of different polarities, greatly expanding peak resolution compared with GC-MS and allowing much more detailed characterisation of highly complex mixtures (e.g. Eiserbeck *et al.*, 2015; Nelson *et al.*, 2016). The LECO Pegasus 4D GCxGC-TOFMS at WA-OIGC will be used for these analyses.

v. Isotope ratio mass spectrometry

Isotope ratio mass spectrometry (irMS) will be employed to measure the ratios of stable isotopes of carbon ($\delta^{13}C$) and hydrogen (δD). These stable isotope ratios can give information on the source of organic molecules, biosynthetic pathways of biomolecule formation, and post-depositional alteration of organics (e.g. Grice & Brocks, 2011; ; Hayes, 2004; Holman & Grice, 2018; ; Sessions, 2016). WA-OIGC hosts two Thermo Delta V irMS instruments, which can perform two types of irMS analyses. The simplest is elemental analysis (EA-irMS), which combusts or pyrolyses a solid sample into a single peak of gas, thereby measuring $\delta^{13}C$ or δD of the entire sample as a single bulk measurement (Skrzypek & Paul, 2006).

Much more information can be obtained from compound-specific isotope analysis (CSIA), in which a gas chromatograph is connected to the irMS instrument through a specialised interface (the technique is also known as GC-irMS). This technique allows the isotope ratios of individual compounds in complex mixtures to be measured; however, to achieve reliable results strict analytical requirements must be met in terms of amount of sample and separation of chromatographic peaks (Sessions, 2006). CSIA will therefore only be done on select samples where these requirements are able to be met. For both EA-irMS and GC-irMS, standard reference materials with verified isotopic composition are run alongside samples, so that results are correctly normalised to the international isotopic scales for each element (Paul *et al.*, 2007; Meier-Augenstein & Schimmelmann, 2019).

vi. Pyrolysis techniques

After extraction, residual rock and dust samples will be subjected to various pyrolysis techniques to investigate the chemical composition of any high-molecular-weight material that cannot be analysed by conventional GC techniques. The simplest method of pyrolysis is flash pyrolysis connected to GC-MS (Py-GC-MS), which is done at WA-OIGC using a CDS Analytical Pyroprobe 5250. This technique breaks high molecular weight organics into smaller fragments by rapid heating to high temperatures

(500 °C and above) in an inert atmosphere of pure helium gas. This is a quick and straightforward analysis which requires only small amounts of powdered rock (~5 mg), so it is suitable as a rapid screening technique. Disadvantages are that the pyrolysate cannot be purified before analysis, and re-arrangement of organics can occur because of the high temperatures used (Larter & Horsfield, 1993).

Hydropyrolysis (HyPy) is a closed-system method whereby rock powder is heated under high hydrogen pressure in the presence of a catalyst. The resulting fragments are rapidly hydrogenated and cryogenically trapped to prevent rearrangement, hence they more closely reflect the composition of the original organic matter (Love *et al.*, 1995). The Strata Technology HyPy system at WA-OIGC has been shown to be effective in analysis of high-maturity organics (Meredith *et al.*, 2015; Robert *et al.*, 2016). As HyPy is much more time consuming than the simpler Py-GC-MS technique, it will be done only on selected samples of particular interest.

vii. TOF-SIMS analyses

The new IONTOF M6 Time of Flight – Secondary Ion Mass Spectrometer (TOF-SIMS), hosted by the John de Laeter Centre, Curtin University, will be employed to investigate the relationships between organics and inorganics on mineral surfaces. TOF-SIMS employs an energetic ion beam to ionise small areas from sample surfaces, allowing organics and inorganics to be identified and their spatial relationships to be mapped (e.g. Thiel & Sjövall, 2011; Siljeström *et al.*, 2017). Selected thin sections will be provided by the inorganic geochemistry team (Section 3.3.2) for TOF-SIMS analysis.

viii. Potential deviations from best practice

The amount of organics in rock samples from Murujuga is expected to be low, hence sample sizes for extractions would ideally be at least 25 g to ensure sufficient material is recovered for GC-MS and GC-irMS analyses. However, restrictions on sample collection mean that many samples are likely to be much smaller, especially those of thin patina and weathered rind layers. For these samples, quality of data produced from analysis may be impacted, e.g. low peak heights resulting in higher signal-to-noise ratios, and insufficient abundance for reliable isotope ratio measurement. Results will not be reported for measurements if quality controls cannot be met.

3.4 Chamber experiments – accelerated patina weathering

To ensure that a dose-response relationship for (elevated) air pollutant concentrations can be established, controlled chamber experiments on patinated rock core samples are planned. These experiments will adapt validated exposure methods developed to accurately expose cell cultures to a range of gases, particles and droplets (Tollstadius *et al*, 2019, Landwehr *et al.*, 2019) – See Figure 3-14 and Figure 3-15.

Both systems (Figure 3-14 and Figure 3-15) are designed to use 23 mm diameter cell culture dishes (albeit 6 and 36 simultaneous samples respectively in each device), which will be substituted to hold 23 mm (or smaller) diameter rock cores with intact patina. The systems are both equipped with incubators to hold the test substrate (rock core) at the desired temperature. In this case mean rock surface temperatures obtained from field measurements using an infra-red thermometer will be used.

The systems permit exposure to uniform, controlled deposition of any desired droplet, particle, or gas. Samples can be exposed either directly from a combustion source (with cooling of exhaust) or reconstituted from field samples or synthesised mixtures based on field measurements or air quality modelling.

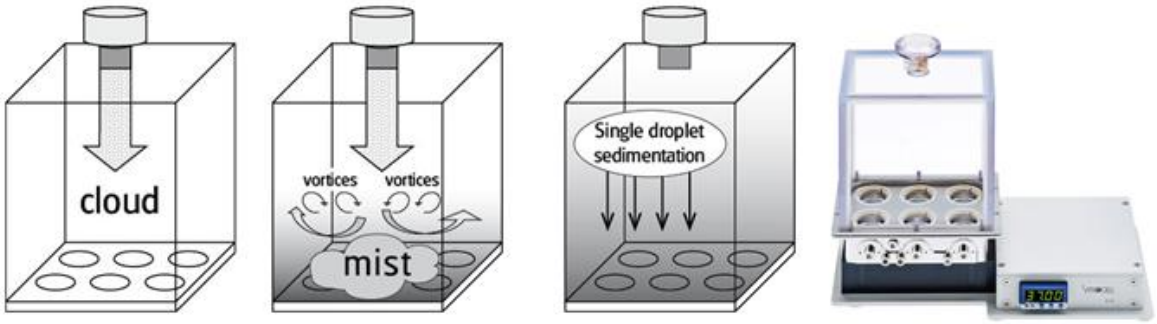


Figure 3-14 Cloud chamber – showing the condensation/coagulation of a cloud of aerosol.

The cloud chamber replicates mist deposition on the rocks, which as mentioned above is thought to be a mechanism capable of concentrating air pollutants from the air column. This hypothesis will be tested.

The apparatus in Figure 3-15 can be used to replicate dry exposure conditions. Post exposure (and subsequent incubation to permit microbiome and geochemical responses to the exposure), the full suite of characterisation detailed in Section 3.3.2–3.3.4 can be applied.

Rock samples with intact patina will be co-exposed or sequentially exposed to typical emissions (natural gas/condensate combustion, ship diesel/fuel oil combustion, vegetation combustion and biogenic VOCs). The sample size determination and full program of experiments for this component of the work will be completed in conjunction with the statisticians, once preliminary 'range-finding' studies have been undertaken, as is common in toxicological type testing. Negotiations to obtain these preliminary samples from the Murujuga region are still ongoing.

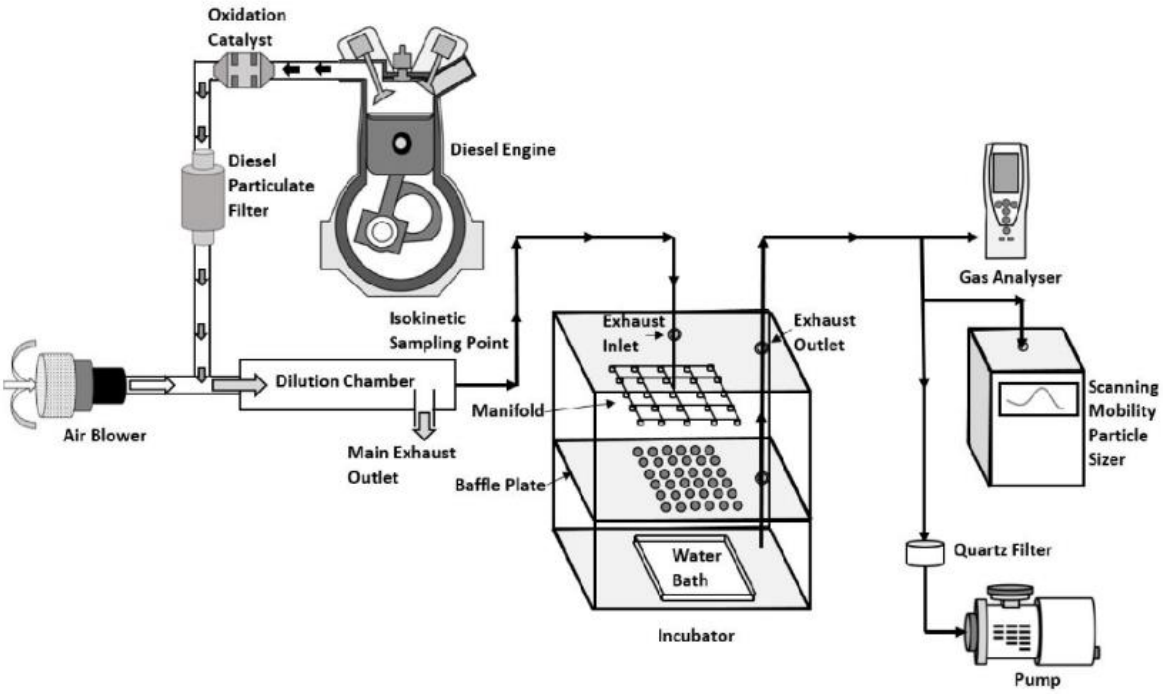


Figure 3-15 Exposure system for gaseous and particle deposition. Can be operated with a range of air pollutants in addition to combustion engine exhaust (Landwehr et al., 2019).

3.5 Air quality modelling, monitoring at receptors and source apportionment

The two main elements in our conceptual model which impact any processes occurring on the rock surface are the atmospheric environment and air quality. Ultimately any deposition at the rock art sites will be dependent on what is present in the atmosphere (either from natural or anthropogenic sources) and what is transported to the individual sites, whether gaseous or particulate. In this part of the Monitoring Program we aim to use a combination of measurement and modelling to capture information on these air pollutants.

This work will be comprised of air quality monitoring at a number of sites within the broader Murujuga area. The selection of these sites has been described in Appendix II and is based on plume/chemical transport models based on known emissions sources across the Burrup Peninsula and other more distant sources. Through this process we have identified a total of 21 sites (including four with existing electrical power) where we can establish monitoring stations, some (or all) of which are expected to form part of the longer term monitoring program.

In addition, we intend to use a suite of real-time measurement technologies to provide a more detailed picture of transient emissions, and additional analysis over that which can be obtained from the more traditional air-quality monitoring stations. This will include isotopic analysis, as well as real-time monitoring to permit validation of plume and computational fluid dynamics models and peak detection and tracking. We will also undertake laboratory-based combustion of key shipping fuels and other hydrocarbon samples to determine isotope signatures. It remains an ongoing question if long-term average emissions or transient concentrations or peak emissions are more critical.

Using the above, we can then analyse selected isotopes in the deposition samples proximal to the petroglyphs to determine the relative contribution of natural and anthropogenic sources (see Section 3.3.3).

3.5.1 Prescribed monitoring

The specification issued by DWER was most prescriptive with respect to air sampling and specified the use of deposition sampling to measure a range of ionic species which could have impacts on both the rock surface and the rock art itself. These impacts may be geological, mineralogical, microbial, etc. and may be complex in nature. There may also be impacts which are synergistic in nature, requiring multiple pollutants to be present, and may be dependent on atmospheric conditions. Any impacts therefore may not be purely because of a single species/pollutant. To allow us every opportunity to uncover any impacts, we are proposing, in line with the original tender, a comprehensive monitoring program to measure not only deposition, but also ambient air, and atmospheric condition/quality.

i. Deposition sampling

In order to collect both deposition samples, a network of wet and dry (bucket) samplers will be situated at each of the monitoring sites. These samplers will allow us to assess the variability in deposition across the larger Rock Art Area and to provide a better estimation of deposition rates throughout the year. We are proposing to leave the monitoring equipment in place for the duration of the monitoring program with samples to be collected at regular intervals. This methodology is consistent with that used in the literature with the same wet and dry samplers used by Morales-Baquero *et al.* (2013), and similar samplers used by Castillo *et al.* (2017) and Inomata *et al.* (2009).

The difference here is that we add a third bucket to cover dew events (where the humidity may be high, but rain does not or has not yet occurred).

The samplers utilise humidity and/or precipitation sensors which are linked to a mechanism which will rotate the cover to open the appropriate collection bucket. In a rain event, the wet deposition collection bucket will be opened, and the others covered. When the humidity drops, the cover will again rotate and the dry deposition bucket will open, and the other buckets will remain covered. When the humidity is high, but the weather station is not reporting rain (sampler and weather stations to be co-located) the third bucket will be uncovered. Thereby allowing us to collect both wet and dry deposition samples, and a third set of samples for mist events, without needing to continuously be onsite.

ii. Gas sampling and analysis

As a number of species which may contribute to wet deposition may be in the gas phase, we will sample for acid gases, at the same locations as the wet and dry samplers. Sampling will be conducted using Gradko International Acid Gases diffusion tubes (subject to availability and preliminary validation). These tubes may be deployed for up to four weeks, with collected samples remaining stable for 12 weeks. This will provide sufficient time for the samples to be transported to the laboratory. The analysis will be conducted by Gradko International in their UK laboratories. Analysis of acid gases will be via ion chromatography. The method allows for detection of hydrofluoric, hydrochloric, nitric, hydrobromic, phosphoric and sulphuric acids (HF, HCl, HNO_3, HBr, H_3PO_4, H_2SO_4) and has been previously employed by Alves *et al.* (2018).

Given the number of surrounding industries, we also propose sampling for a suite of volatile organic compounds (VOCs). This will allow us to identify the presence of VOCs which will provide an indication of the atmospheric organic matter present at the monitoring sites. Gradko International VOC diffusion samplers will be used here. As with the acid gas sampling this will be done passively, allowing us to collect a sample over four weeks. Analysis will be conducted by Gradko International in their UK laboratories, via thermal desorption in accordance with ISO 16017-2. The tubes are configured to sample VOCs and semivolatile organic compounds (SVOCs) in the C2-C28 range. Given previous air quality monitoring in the region has measured BTEX (benzene, toluene, ethylbenzene and xylene) these will be included in the sample suite.

Similarly, analysis of ozone, ammonia, sulphur dioxide and nitrogen dioxide (O^3, NH^3, SO_2 and NO_2) will be performed using Gradko International sampling tubes. This means that each sampling location will be fitted with a set of sampling tubes, mounted on a pole with a rain cover. This will result in a suite of sampling tubes being used in each location, with a pair of tubes for; VOCs, acid gases, ozone, ammonia, sulphur dioxide and nitrogen dioxide. The full set of 12 tubes will be exchanged every four weeks and immediately transported for analysis.

Given the remoteness of a number of the monitoring sites identified and the lack of available power in most sites, passive sampling techniques will be favoured over active sampling in our default monitoring station configuration. These also have the advantage of being able to be deployed for longer time periods, and avoid issues associated with the calibration and re-calibration of sampling pumps in the field. There are some cases where passive sampling is not possible or not effective.

It is acknowledged that the use of such tubes provides averaged data, which may not be sensitive enough to detect short duration peaks. To counter this, where power is available, we will use direct reading instruments to capture transient data. At these (powered) locations we will deploy a range of direct reading instruments to measure species of interest. One such species is nitric oxide (NO) which

readily oxidises to nitrogen dioxide (NO_2) in the atmosphere, so must be directly measured rather than collected for later analysis. For NO monitoring we will utilise a nitrogen oxides analyser, allowing us to use chemiluminescence to monitor and log NO and NO_2 levels. This is something that can be deployed at each of the powered sites identified.

iii. Weather measurements

To allow us to collect accurate weather data throughout the Monitoring Program, a network of weather stations will be used. These will be present at each of the air quality monitoring sites and will record temperature, relative humidity, pressure, precipitation, solar radiation, and wind speed and direction.

iv. Sample sizes

The exact number of monitoring sites was determined statistically using the approach described in Appendix I and II. In addition to the monitoring sites set up by the research team, we will also have access to monitoring data from the Woodside and Yara air quality monitoring stations. We have identified four powered sites where we will deploy a full complement of monitoring equipment, consisting of our air quality monitoring station (comprising a weather station, deposition sampler and passive sampling tubes) and a suite of direct reading instruments (as described under Equipment and Procedures). To help ensure reliability of the results and to account for any failure or damage of the passive sampling tubes, there will be duplicates of each tube at each location (i.e. they will be deployed in pairs).

In addition to the four powered sites there are 17 other sites that have been identified for monitoring. These sites do not have power and in some cases are relatively remote. At each of these sites we will deploy an air quality monitoring station. This means that there will be a total of 21 air quality monitoring stations, four or which will have continuous monitoring. Nine of the sites are industry sites; however, it is planned to co-locate powered/continuous monitors at two of these.

This will provide three deposition samples (dry, wet and mist) from each site, each month; two passive samples for each of acid gases, VOCs, NO_2, SO_2, ozone and ammonia at each site, each month; and continuously logged temperature, humidity, rainfall and wind speed and direction data.

Additionally, at each of the powered sites we will have continuous monitoring of NO, NO_2, sulphur dioxide (SO_2) ozone and hydrocarbons, allowing us to observe any short terms peaks, which will not be captured by the passive samplers. By co-locating passive and active samplers at the four powered sites we will also be able to cross-validate our measurements.

v. Departures from best practice

Given the mix of equipment used at each site, and any unique characteristics of the site, it may also be necessary to alter the way the equipment is distributed at each site, meaning that the distance between and relative orientation of the weather station, sampling tube and deposition sampler, may vary between sites.

For the measurement of wind speed, we will adhere to the standard 10 m measurement height, where possible. Noting that the existing industry monitoring sites use either a 5 or 10 m measurement height. There may be cases, for particular sites where this is not possible, and if so, the measurement of wind speed will be conducted at the height that is practical/permitted. Ultimately, we want to be able to estimate the quantities of species contacting and depositing on the rocks and

rock art creating a link between ambient air measurements and observed processes taking place on the rocks.

In terms of specific locations of the monitoring stations at the identified sites, these will be determined based on:

- AS/NZS 3580.1.1:2016 Siting Air Monitoring Equipment Guide
- AS/NZS 3580.14:2014 Methods for sampling and analysis of ambient air- Part 14: Meteorological monitoring for ambient air quality monitoring applications
- any cultural considerations
- practicality.

Stations will be sited at least 10 m from any roadway, which satisfies AS guidelines for all pollutants apart from SOx and Particulate Matter (PM) (which require 50 m). However, it is important to note that most roads in the region do not have sufficient traffic volume to require this separation.

vi. Air quality monitoring station inclusions

Each of the 21 air quality monitoring stations we deploy will include a weather station, deposition sampler and a suite of passive sampling tubes, as described in the sections below.

Weather station

Each monitoring station will contain a weather station to measure atmospheric conditions throughout the study, these will need to be procured. We are proposing the Campbell Scientific GRWS100 Weather Station, which comprises a wind monitor, an air temperature and relative humidity probe, a pyranometer, barometric pressure sensor, rain gauge, data logger and power supply. Options are available for each measurement device to ensure that the requirements outlined in AS/NZS 3580.14:2014 are met. These options also allow us to exchange sensors with alternatives as appropriate. For example, the standard anemometer requires a start threshold of 0.5 m/s, this can be replaced with an ultrasonic anemometer which will avoid this issue. The weather stations allow the measurement of wind speed and direction, air temperature, relative humidity, barometric pressure, solar radiation and precipitation. These stations have both on board data-logging and a range of communication options.

The proposed fit out of the weather station is with the following instruments/sensors:

- CS300-L Pyranometer
- EE181-L Air Temperature and Relative Humidity probe
- CS100 Barometric Pressure sensor
- TE525MM-L Metric rain gauge
- WINDSONIC1-L Ultrasonic Anemometer.

Deposition sampler

This will be developed by the research team in house. The operating principal is relatively simple, with a movable lid that will keep one bucket open and the remaining two closed. This will allow us to collect samples for dry deposition, wet deposition and mist-type deposition. It is believed that mist

type events may result in significant deposition, and therefore we consider it important to capture such events separately. A schematic of the proposed device is shown in Figure 3-16.

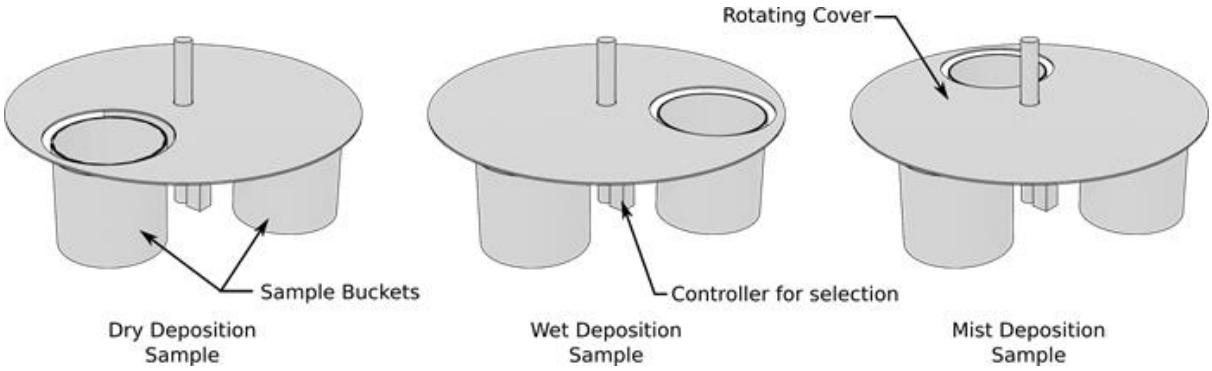


Figure 3-16 The '3 Bucket' deposition sampler.

In addition to the dry and wet deposition samplers we plan to add a third bucket for mist/fog events. This will allow us to assess the relative contribution of mist/fog driven deposition which we hypothesise may be significant. This will also provide us with 'real world' data to extrapolate the results of our lab-based rock exposure studies.

The deposition sampler will be fitted with a rotating lid, which allows one bucket to be open at a time, this will be controlled by a stepper motor, which will open/close the buckets based on weather information provided by the weather station.

The default position will be with the dry bucket open. There will be a daily test of the function of the lid, with a single complete rotation performed at the same time each day.

The dry deposition bucket will be closed, and the wet deposition bucket opened based on readings from the rain gauge. The mist/fog bucket will be opened, based on the dewpoint (T_{dp}), which will be determined based on the measured dry bulb temperature and relative humidity obtained from the weather station. The controller for the 3-bucket sampler will calculate the dew point using the following expression (Lawrence, 2005), with constants based on the recommendations of Alduchov and Eskridge (1996).

$$T_{dp} = \frac{243.04\left(ln\frac{RH}{100} + \frac{17.625T}{243.04+T}\right)}{17.625 - \left(ln\frac{RH}{100} + \frac{17.625T}{243.04+T}\right)}$$

Where *T* is the dry bulb temperature and *RH* is the relative humidity.

The following parameter will be used to determine which bucket to open:

- If the value of $T_{dp} + 2.5 \geq T$ and the reading in the rain gauge is 0 mm then the mist/fog deposition bucket will open (the other buckets will be closed, by virtue of the single opening in the rotating lid).
- If the value of $T_{dp} + 2.5 < T$ and the reading in the rain gauge is 0 mm then the dry deposition bucket will open.
- If the reading from the rain gauge is not 0 mm (i.e. a detectable amount of rain is collected) the wet deposition bucket will open.
- If the rain gauge reading does not change for 20 minutes and $T_{dp} + 2.5 < T$ then the dry deposition bucket will open.

- If the rain gauge reading does not change for 20 minutes and $T_{dp} + 2.5 \geq T$ then the mist/fog deposition bucket will open.

Samples will be collected every four weeks and transported to the laboratory for analysis. These will be transported from the monitoring site to the ChemCentre. For the dry deposition samples, the samples will be washed out of the sampler with ultra-pure distilled water and will be measured for Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-}. A similar approach will be taken for the wet deposition samples, with pH and conductivity measurements taken before analysis for the same ions listed above. These samples can then be analysed for carbon and iron ore dust and sea salt.

Prepared 'fresh' rock sampler/test section

In addition to the deposition samplers described above we plan to deploy a collection surface or test section of rock in each of the air-quality monitoring stations. These rock surfaces would be subject to the same dry, wet, and mist deposition processes as the rock art in the area, as well as exposure to the same gas and particulate species. These surfaces would also allow assessment of the microbiome that develops as a proxy to the rocks containing the rock art, which we will not disturb. A section of each to the three predominant rock types present in Murujuga will be prepared such that a layer of 'fresh' rock (i.e. unexposed rock with no patina, no weathered rind etc.) is newly exposed at each site. These can then be analysed, providing a link between the measured air-quality and what is happening on the rocks.

Passive sampling tubes

Given the remoteness of the monitoring sites, passive sampling tubes with long deployment times offer a favourable approach for monitoring a number of species. The passive sampling tubes will be prepared and analysed by the supplier. These tubes will be shipped to Perth and then transported to and deployed at the monitoring sites. These tubes are stable for 12 weeks, allowing sufficient time for transport, deployment, sampling, and analysis. Tubes will be ordered in batches to ensure that there is not more than 12 weeks between preparation and analysis.

All tubes will be mounted on a post which forms part of the sampling station, this will allow the tubes to be protected by rain cover, and a solar shield, preventing excessive heating of the tubes and keeping water out of the tubes. The detection limits for the tubes are provided in Table 3-8.

Table 3-8 Detection limits for the passive sampling tubes, provided by the supplier.

Tube	Detection Limit (µg/m^3)
Acid Gases	3.5 (HCl), 0.5 (HF), 2 (HBr), 2.5 (H_3PO_4), 2 (HNO_3), 2 (H_2SO_4)
Ammonia	1.5
Ozone	10
Nitrogen dioxide	1.5
Sulphur Dioxide	2
VOCs	Low ppb range*
* There is some variance depending on species.	

The 12-week stability time, the ability to deploy the sampling tubes for four weeks, the detection limits available and the cost of the tubes make a compelling case for their use; however, we must also consider potential limitations. More frequent sampling would give more measurements, though would require more frequent site visits, and would be more resource intensive. We also need to consider any potential interferences with the sampling tubes. The NO_2 and SO_2 tubes have known interferences with nitrate and sulphate particles, respectively. As we are also measuring deposition, we will be obtaining measurements of deposited nitrates and sulphates, which we can use to judge if any such interferences are occurring. There is also the possibility of wind speed impacting the sampling measurements. For most of the tubes this can be offset by the use of a filter. For the NO_2 tubes, wind speeds between 1-4.5 m/s can lead to up to a variance up to 10%. Here we will also collect wind speed data for the sampling period so we can determine to what extent this has been impacted and calculate the appropriate error.

Passive sampling tubes: cross validation study

Before deployment of the monitoring stations, we will field trial the passive sampling tubes to confirm supplier provided specifications and compare performance of the different tubes available. This will consist of deploying triplicate tubes for different monitoring periods to assess performance over time. This will also allow us to confirm the 'real world' detection limits of the sampling tubes.

Several different passive sampling tubes have been used in the Murujuga area as part of previous studies or ongoing monitoring work conducted by Industry. These have used Radiello tubes, Ferm/IVL tubes (Ferm and Rodhe, 1992). For many species the detection limits of the tubes are comparable, but the length of deployment variable. We are proposing a new tube, supplied by Gradko International, owing to the ability to deploy for up to 30 days.

Given some differences in quoted detection limits and the vagueness around values quoted as 'low ppb range', particularly for VOCs, we will perform a comparison of tubes, Before deployment of the ambient air monitoring stations. This will allow us to choose the most appropriate tube for each species. If for example the Gradko tube cannot give satisfactory performance for VOCs we would use a tube with better performance.

Complete sets of tubes will be deployed for seven days, 14 days and 30 days. This will allow both the variability between tubes to be assessed as well as comparative performance. Given the remoteness of several monitoring sites identified, the deployment period for the tubes needs to be sufficiently long to make returning to site to swap out tubes efficient. It may not be practical to visit each site more frequently than monthly, so longer deployment times are preferred. If scheduling or environmental constraints delay the monthly sample collection, averaging intervals for the results will be altered accordingly.

As with any sampling there is also the possibility that pollutant levels may be present, but at a concentration that falls below the detection limit. Should there be repeated measurements that are below the detection limits, we propose revising the sampling protocol. The initial monitoring period provides an opportunity to explore this, particularly for species that haven't been monitored in the area before, and therefore there is no indicative value on which to base the monitoring program design.

Real-time monitoring

Reviewers of the previous release of this document noted a preference for active or real-time monitoring. We agree that expanded real-time monitoring would be beneficial, given concerns expressed above and in the literature regarding the importance of peak and transient air pollution. At each of the four powered sites we plan to deploy a range of direct reading instruments to log analyte levels continuously. These sites will be fitted with instruments for NO_x (NO and NO_2), SO_2, NH_3, O_3, hydrocarbons and particulates, in an air-conditioned enclosure to ensure the equipment stays within the manufacturers quoted operating ranges (in most cases this is below 40 °C). The enclosure will provide sufficient room for cylinders of calibration gas and data handling and communications systems. We are considering different options for these monitors based on performance and availability.

The use of such instruments will allow us to detect short-term and long-term peaks in contaminants of interest. Through the continuous monitoring of these species, we can also consider any trends as well as the relative ratios of species, e.g. the variation of NO:NO_2 ratio during day versus night.

- *Nitrogen oxide and nitrogen dioxide*

  Each powered monitoring site will be equipped with a nitrogen oxide sensor. The sensors under consideration are the Sernius® 40T which is designed for trace analysis and can detect concentrations as low as 50 ppt or the ThermoFisher 42iQ, or the TAPI T200U which are equipped for trace levels. This is considered important as NO levels will likely be low, owing to the ready oxidation of NO to NO_2 under normal atmospheric conditions. These instruments can be deployed in such a way that the requirements of AS 3580.5.1-2011 are met, which will require the provision of cooling and calibration gas. These sensors also measure NO_2, which we can compare to the results from the passive sampling tubes to cross-check our measurements.

- *Sulphur dioxide*

  An SO2 sensor will be deployed at each powered site. The analysers under consideration are the ThermosFisher 43iQ, the TAPI T100U and the Sernius 50T.

- *Ammonia*

  Each of the four powered sites will be equipped with an ammonia analyser to allow the real-time measurement The instruments under consideration are the Thermo Scientific Model17i

Ammonia analyser, which can measure down to 1 ppb, and the Sernius 44 Ammonia analyser, which has similar performance, though with a slightly lower detection limit.

- *Ozone*

  An ozone sensor will be deployed at each powered site. The analysers under consideration are the Sernius 10, the ThermoFisher 49i and the TAPI T400.

- *Hydrocarbons*

  Each powered site will have a hydrocarbon analyser to measure total hydrocarbons present. Given the nature of the industries present around the Murujuga area and potential likely emissions, we will deploy an analyser which reports both methane and non-methane hydrocarbons. The analysers currently under consideration are the ThermoFisher Model 55i and the EcoTech VOC1000.

- *Particulates*

  In addition to the gaseous species, we will also directly monitor particulates at each powered site. Here we plan to deploy the PALAS FIDAS 200E aerosol spectrometer, which will allow us to measure $PM_{2.5}$ and PM_{10} simultaneously, as well as PM_1, PM_4 and particle number concentration. This instrument is approved and certified to the requirements of EN15267.

- *Carbon*

  While we will assess carbon deposition at each of our monitoring sites, this will only give monthly average. Through the use of an appropriate real-tie instrument we can also measure the black carbon (soot) content of the particulate matter present. This would serve to complement the real-time particulate measurements being conducted at the powered monitoring sites. We propose deploying a single Magee Aethalometer Model AE43 to measure black carbon. The instrument would be rotated through each of the four powered sites based on a schedule determined based on weather data and the particulate matter readings from the FIDAS instruments.

- *Open Path FTIR*

  To provide additional characterisation of emissions present we plan to deploy an Open-Path Fourier Transfer Infrared Spectrometer (FTIR) in a series of monitoring campaigns across the four powered sites. The timing of the campaigns would be subject to instrument availability and weather patterns. It would also be contingent of suitable locations for the instrument's mirrors being found. Through the use of the instrument, we would be able to simultaneously measure a broad suite of relevant chemical species present (Weber et al., 2004).

- *LIDAR*

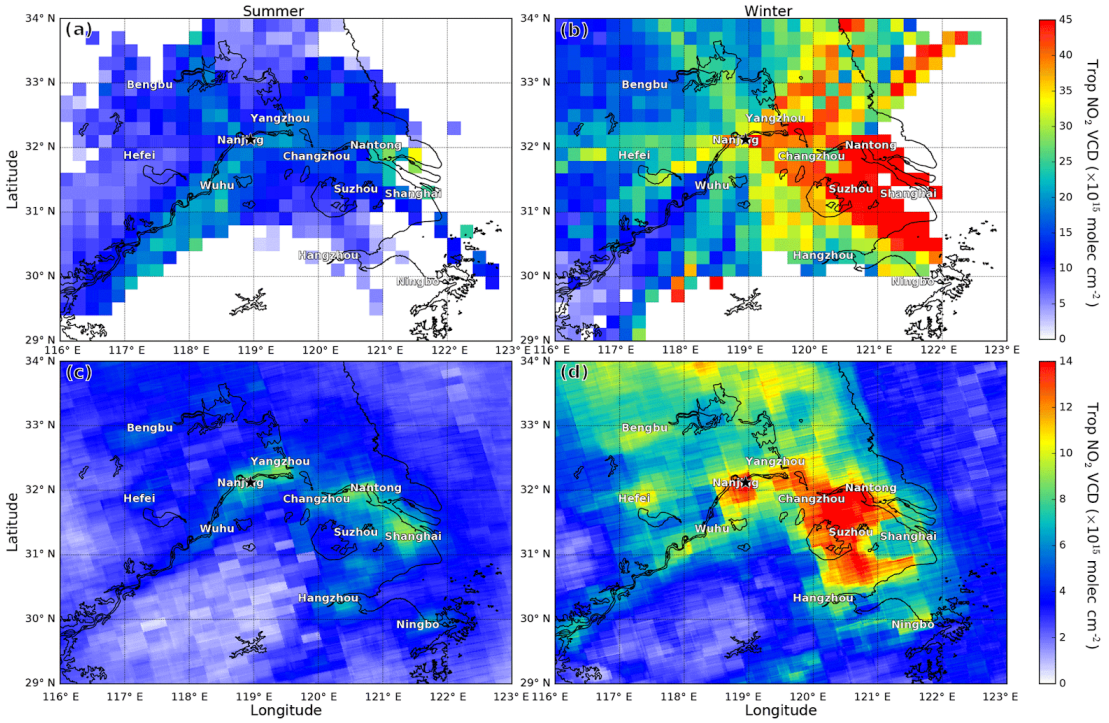
  DWER has a Windcube 200S LIDAR, which we can borrow for use. This is considered a low-cost add-on to the project, which can provide us with additional useful information that would not be otherwise captured. The instrument has been previously used by DWER to map dust plumes (DWER 2019) and would be employed here for the same purpose. The modelling conducted by RAMBOLL for example, does not feature locally specific dust input parameters, so there is some question over the accuracy of the data, particularly for fine dust.

While dust may not be the most important pollutant with respect to the weathering of the rock art it may contribute via mechanical abrasion, or act as a carrier for specific pollutants. It is therefore useful to validate the dust plume data. This will be done using the LIDAR alternately at each of the four powered sites, with the monitoring schedule determined based on weather parameters.

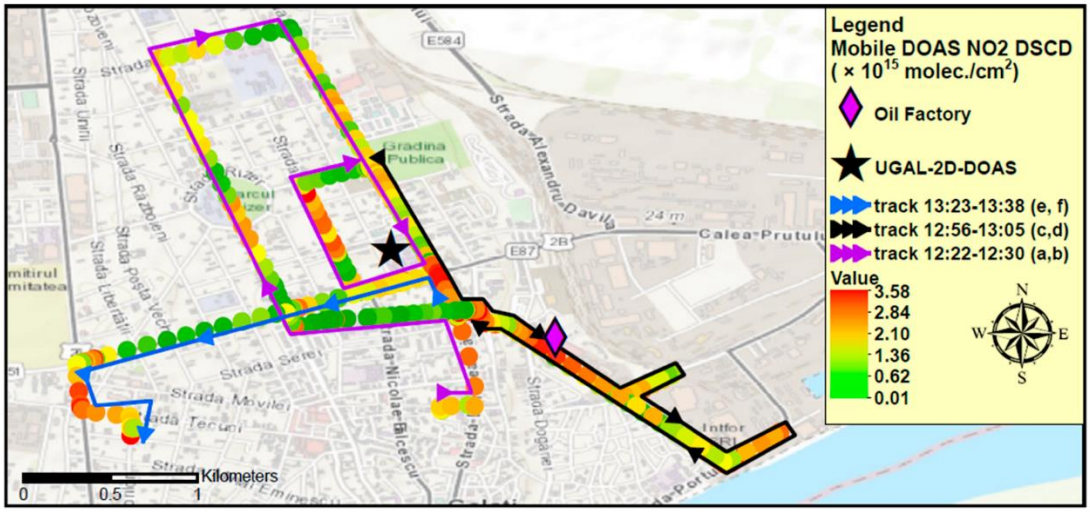
- *MAX-DOAS*

  A single multi-axis differential optical absorption spectrometer (MAX-DOAS) will be employed at site EX09. This site is on the top of a hill (Mt Wongama) and will provide good coverage of the surrounding area. This will allow us to perform NO_2 mapping of the surrounding areas and provide information which can be used to assist in the analysis of the NO_2 measurements obtained using the gas analysers.

  Figure 3-17 shows examples of the types of measurements which can be obtained using the MAX-DOAS. It is possible to get both full column data over a larger area (a), or to get ground level data (b). The MAX-DOAS can be set to the appropriate protocol to measure the column height of interest. This will give us the ability to map pollutant (NO_2, SO_2) levels over about a 4 km radius.



(*a*)



(*b*)

Figure 3-17 Examples of the spectrum of MAX-DOAS measurement capability for NO_2 sensing. (a) shows the full air column measurements (Chan et al. 2019) and (b) ground level (0 Azimuth) measurements from a smaller mobile unit (Rosu et al. 2020).

- *Isotopic analyser*

  A single Picarro G2201-I Isotopic Analyser (Perez et al., 2017) will be deployed initially at site EX09. This will allow us to measure methane (CH_4) and carbon dioxide (CO_2) levels in real-time while simultaneously measuring Carbon-13. The instrument is designed to be field deployable and can be moved between powered sites should it be required. Furthermore, it can measure ethane/methane ratios, to differentiate between natural and anthropogenic sources.

- *'Low-cost' sensors*

  The experimental program necessarily requires high accuracy instruments which meet appropriate standards and QA/QC protocols. However, the remoteness of the sites and lack of power limit the potential to deploy full-scale real time monitoring for all pollutants of concern. Therefore, in addition to the highly accurate passive and active monitors proposed for the unpowered/powered sites respectively, low-cost solar powered, sensor-type instruments will be additionally deployed to provide qualitative or semi-quantitative trends in pollutants. These sensors will measure PM, CO and NO_2, using components (SDS011 or Plantower PM sensors, Alphasense CO and NO_2 sensors) which have been proven to be more accurate than most sensors on the market (Jayaratne et al., 2020). The combination of PM and two key combustion gases, when combined with weather station data, will permit the tracking of pollutant plumes/peaks and basic source identification (e.g. geogenic/ore/dust/salt versus combustion sources).

Data-logging

The weather station is equipped with a dedicated control unit and data logger. These allow data to be stored directly to removable media on each device. This storage space is more than sufficient to record all data gathered over the four-week period between sample recovery visits. The weather stations can also be equipped with a range of data communications equipment. We plan to utilise the local 4G mobile network to regularly transmit data back to the project Cloudstor repository. Initial inspection of network coverage maps indicate that this will be achievable for most of the Murujuga area. Should a site fall outside network coverage we will find an alternative means to transmit the data (likely RF / LoRa). This will allow us to both monitor weather conditions and instrument performance more regularly and provide us with a redundancy should the on-board data storage become corrupted. This will also allow us to monitor the performance of the sensors, to allow early detection of sensor drift (or failure).

The direct reading (continuous/real-time) instruments have a number of data interfaces so we will develop an external data logger to record and transmit data from each of the instruments. The measurement data will also be stored locally, such that there is an in-built redundancy measure for the direct reading instruments. This will be developed in house by members of the research team, using similar technology to that used in an ARC Linkage project on sensor networks, in which members of the research team are involved. All continuous monitoring stations are within the 4G coverage network.

Power supply

Each of the four powered sites (see Table 3-9) has sufficient power infrastructure to allow the deployment of the proposed instruments. The weather stations and deposition samplers will be powered by on board solar panels, with battery back-up.

vii. Equipment siting

As described earlier our site selection process has identified sites based on the minimum grid sizes employed in the modelling conducted by Ramboll (2021). The exact siting of the equipment at these locations will be determined in collaboration with the Murujuga Aboriginal Corporation and the Circle of Elders (except for powered sites, and where the location is already used for industry monitoring). This is to ensure a culturally appropriate location within the identified grid square is found, without disturbing any rock art or impacting on cultural practices.

In addition to any cultural considerations the exact siting of the equipment will, where practical, follow the requirements of AS/NZS 3580.1.1.2016. This will require the monitoring stations to be set a certain distance from any roads (50 m where possible). The distances from the road for background monitoring are chosen, as while the emissions of any passing vehicle do contribute to the local ambient air, we want to ensure they don't disproportionately impact the measurements. The aim here is to use the results of our network of monitoring stations to predict levels at any location in Murujuga, so we need to avoid disproportionately weighting individual contributing sources.

Table 3-9 Equipment to be installed at each monitoring location.

Site designation	Passive/solar monitoring station	Weather station	Rock cubes	Powered/ Realtime monitoring station
EX01[(h)]	* (Woodside)	*	x	
EX02	x (DWER decomm.)	x	x	x
EX03	* (Woodside)	*	x	
EX04	* (Yara)	*	x	
EX05	* (Yara)	*	x	
EX06	* (Yara)	*	x	
EX07	* (Woodside)	*	x	x
EX08	* (Woodside)	*	x	
EX09	* (Woodside)	*	x	x+
AQ01	x	x	x	x
AQ02	x	x	x	
AQ03	x	x	x	
AQ04	x	x	x	
AQ05	x	x	x	

Site designation	Passive/solar monitoring station	Weather station	Rock cubes	Powered/ Realtime monitoring station
AQ06	x	x	x	
AQ07	x	x	x	
AQ08	x	x	x	
AQ09	x	x	x	
AQ10	x	x	x	
AQ11	x	x	x	
AQ12	x	x	x	
AQ13	x	x	x	
AQ14	x	x	x	
AQ15	x	x	x	
AQ16	x	x	x	
AQ17	x	x	x	
AQ18	x	x	x	

(h) denotes (human/urban) health monitoring station which has different measurement parameters. Passive samplers and deposition monitor will be added to this site.

* denotes existing monitoring stations with owner shown in brackets

\+ EX09 is centrally located on the peninsula at a slightly elevated location and deemed the most appropriate site for scanning-type instruments (Lidar, MAX-DOAS, etc.).

viii. Chemical analyses (ChemCentre)

The ChemCentre will provide services for the analysis of atmospheric deposition samples. These samples will include dry depositions, aqueous solutions collected in the field and solutions derived from rinsates or extractions. Robust methods suited to analysis of low ionic strength solutions and capable of achieving very low detection limits will be employed.

- *Iron ore dust and sea salt*

  Iron ore dust will be digested with strong acid (high-purity nitric and hydrochloric acids) with a microwave digestion system such as Milestone Ethos Easy.

  Sodium from sea salt will be analysed in the same acid digest or from a separate extraction with deionised water as appropriate based on levels and amount of material.

Analysis for iron and sodium (from salt) will be by inductively coupled–plasma – optical emission spectrometry (ICP-OES) on instruments including an Agilent 5110 with protocols based on APHA 3120 and US EPA 200.7. This technique offers excellent detection limits and selectivity for the metals. Where appropriate, confirmation using inductively coupled plasma mass spectrometry (ICP-MS) on an Agilent 7900 can be employed.

Analysis for chloride (from salt) – where required – will be by ion chromatography (IC) with conductivity detection on a Dionex ICS-5000 with protocols based on APHA 4110 B. and US EPA 300.1 and will require a deionised water rinsing of the particulates.

- *Cations (including Mg^{2+}, N^{+}, K^{+}, Ca^{2+})*

  Aqueous solutions presented to the laboratory or other materials extracted with deionised water or other appropriate extractant will be analysed as follows.

  Inductively coupled–plasma – optical emission spectrometry (ICP-OES) on instruments including an Agilent 5110 with protocols based on APHA 3120 and US EPA 200.7. This technique offers excellent detection limits and selectivity for the metals. Where appropriate, confirmation using inductively coupled plasma mass spectrometry (ICP-MS) on an Agilent 7900 can be employed.

- *Ammonium (NH_4^+)*

  Aqueous solutions presented to the laboratory or other materials extracted with deionised water or other appropriate extractant will be analysed as follows.

  Continuous flow analysis will achieve excellent detection limits. Instruments employed will be flow injection analysis (FIA) on a Lachat QUALITY CONTROL8500 Series 2 following protocols based on APHA 4500-NH_3 H, or segmented flow analysis (SFA) on a Seal Analytical Quattro following protocols based on APHA 4500-NH_3 G.

- *pH*

  pH of solutions will employ the standard electrochemical (pH electrode) technique following the protocol of APHA 4500-H^+ B.

- *Electrical conductivity*

  The conductivity of the solutions will employ the standard electrode and meter technique and will follow the protocol of APHA 2510 B.

- *Carbon*

  Analysis of aqueous solutions for total carbon (TC), total inorganic carbon (TIC) and total organic carbon (TOC) will use combustion – infrared detection following protocols based on APHA 5310 B using instruments including a Shimadzu TOC-V CSH. As an alternative, persulfate oxidation – infrared detection based on APHA 5310C using instruments including an OI Analytical Aurora model 1030.

  Analysis of solids for total carbon (TC), total inorganic carbon (TIC) and total organic carbon (TOC) will use combustion – infrared detection following in-house protocols using instruments including an Elementar vario MAX cube.

ix. Quality assurance and quality control

Deployment of monitoring stations

Each of the components of the monitoring stations will be tested before deployment to verify they are in working order. They will again be tested when the monitoring stations are assembled on site. Data logging (and communications where sufficient network coverage) will be verified on site. Each sensor on the weather station meets the requirements outlined in AS/NZS3580.14:2014.

On each visit to collect samples the complete monitoring station will be inspected for any signs of damage or deterioration. This will include inspection of the sensors themselves and any wiring and cables. Damaged or faulty sensors will be replaced, with repairs and recalibration of any faulty sensors to be completed by the supplier.

A set of standard operating procedures will be developed for the deployment of the sensors and these will be prepared during the testing of the systems before deployment.

Sample collection and transport

The monitoring sites will be visited every four weeks for sample recovery. This will involve swapping out the passive sampling tubes and recovering the three deposition samples from each station. All samples will be labelled with a unique label, identifying the sample location, time, type of sample and who collected it. Passive sampling tubes will be immediately sealed and removed in the order they were deployed. These will be inspected for any visible damage, with any observed being recorded. These samples will then be placed in a secure insulated container to protect them from heat and sunlight. They will be shipped to the supplier's laboratory for analysis. Local options were explored; however, even with shipping costs this remains the most cost-effective option by far. As mentioned above, a validation study will be undertaken to confirm accuracy and sampling interval can be achieved.

Deposition samples will be recovered in labelled sample jars and transported by air back to Perth. These will then be delivered directly to the ChemCentre for analysis by the research team and using the ChemCentre's chain-of-custody documentation.

Field blanks

In addition to the sampling tube supplier's internal QA/QC (Quality Assurance/Control) procedures we will also make use of field blanks for each batch of tubes deployed. One of each tube batch will be transported to site and remain unopened. We will therefore have one field blank for each tube type for each 4-week sampling period.

Calibration

All sensors on the weather station will be manufacturer calibrated before use. These will be tested before deployment, with the results reported by the sensor to be monitored for any unusual readings. During site visits for sample collection the function of sensors will be verified against hand-held instruments to ensure consistency, faulty sensors will be replaced and returned to the supplier for repair and/or recalibration. A calibration record will be developed for each sensor, such that calibration schedules provided in AS/NZS 3580.14:2014 can, where possible, be maintained.

External laboratories

The passive sampling tubes will be prepared, supplied and analysed by Gradko International in their UK-based laboratories, using their in-house Quality Assurance/Quality Control (QA/QC) procedures. This laboratory uses a variety of United Kingdom Accreditation System (UKAS) certified methods and ISO standards to conduct the work.

ChemCentre

The Chemcentre will conduct all deposition sample analysis apart from isotopes. ChemCentre's Quality Management System reflects a history of accreditation dating back to 1954, as one of the first laboratories in Australia to achieve National Association of Testing Authorities (NATA) accreditation. They currently maintain an extensive scope of accreditation within the fields of Chemical Testing and Forensic Science. Therefore, their Quality Management System provides validated and accredited methodology that complies fully with all relevant clauses of ISO/IEC 17025 and ISO 9001. Their systems are actively reviewed and managed and are subject to regular external scrutiny by accreditation bodies. New methods are constantly being considered for NATA accreditation or peer review including those relevant to this tender.

Routine analytical work entails replicate analysis of at least every 10^{th} sample (sample volume permitting), an ongoing external Quality Control (QC) every 20^{th} sample, the use of matrix matched certified reference (CRM's) with every analytical batch (where available), fortified sample matrix spikes with every analytical batch and method blanks. Where practical, results of analysis are verified by chemists using confirmation with alternate methodologies (e.g. ICPAES and ICPMS for metals analysis), use of chemical relationships (e.g. ionic balance within 5%) and results of previous analysis (e.g. long-term monitoring of the same site).

x. Urea monitoring

We have been specifically requested to consider and comment on monitoring of urea emissions, because of the likely addition of urea manufacturing to the airshed. Urea ($CO(NH_2)_2$) is a basic solid, with a melting point of ~135°C; however, in atmospheric, aquatic and terrestrial environments, it is generally accepted that it breaks down relatively rapidly to form ammonia (NH_3) gas (a weak base) and or ammonium (NH_4^+) ions (a weak acid). Depending on humidity, weather and other air contaminants, complex partitioning between these latter compounds and between solid/liquid and gaseous states can occur (Ge *et al.,* 2011). Highly specialised instruments are needed to fully speciate particulate emissions in real time – e.g. monitoring of urea particulate emissions directly. Thus it is proposed that urea releases may be more appropriately measured via measurement of ammonia (NH_3) gas and or ammonium (NH_4^+) ions. The ratio of these two substances, combined with appropriate models, and sufficient spatio-temporal resolution of data, could possibly be used to differentiate between direct release of ammonia versus release of urea.

The sections above already propose the measurement of ammonium in wet/dry/dew deposition bucket samplers at monthly intervals. However, several additional instruments will be added in order to provide better spatio-temporal resolution of ammonia and urea emissions within the airshed.

Furthermore, CO_2 is an integral feedstock in the production of urea from ammonia, with more CO_2 being required than is emitted in the production of ammonia from natural gas. Therefore, localised CO_2 emissions and any changes associated with urea production may also be useful to monitor.

Previously proposed monitoring:

1. NH3 passive sampler, monthly samples at each location.
2. NH4+ analysis in wet/dry/dew deposition sampler 15 sites *3 monthly samples at each location.

Additional monitoring proposed:

1. ThermoFisher 17i real-time NH3 monitor (or equivalent) – one per each powered monitoring station
2. Low cost (Alphasense) NH3 sensors added to each low cost monitor
3. Picarro G2210-i CO2 monitor

The low-cost sensors will permit spatio-temporal trends to be extracted from the monthly average samples, thereby providing much richer information. Additionally, the OP-FTIR campaign will provide a useful baseline dataset, which could be repeated once the urea plant is operational.

3.5.2 Source apportionment (including isotopes)

i. Isotope signatures from combustion emissions

In order to better detect trace levels of organics and inorganics collected from the deposition monitors proximal to the petroglyphs and undertake source apportionment based on isotope signatures of anthropogenic and natural sources, it is important to undertake source characterisation studies. These will be conducted via (a) controlled combustion using validated methods/apparatus existing at Curtin University, and the isotope fingerprinting method developed by members of the research team (Vitzthum von Eckstaedt *et al.*, 2012).

The method will use the diesel engine apparatus shown in Figure 3-15 and validated methods used previously (Landwehr *et al.* 2019) to combust a representative range of shipping (diesel engine) fuels obtained from industry, expected to consist of heavy fuel oil, marine diesel and possibly natural gas. A validated tube furnace method for biomass combustion (Ordou and Agranovski, 2019) will be used to combust the most abundant floral species from Murujuga to obtain isotope signatures corresponding to bushfire smoke and biogenic volatile organic compound (bVOC) emissions. In all studies the method developed by members of the research team (Vitzthum von Eckstaedt *et al.*, 2012) will be used in conjunction with particulate sampling in order to develop isotope 'fingerprints' for key carbonaceous emissions. Such carbonaceous emissions have been shown to impregnate the patina (Dorn, 2020) and may act as a carbon source for the microbiome. Inorganic isotopes will also be determined from filter samples using the methods described above. Isotopic analysis of soil samples and surplus deposition samples will also be undertaken.

3.6 Computational fluid dynamics modelling

3.6.1 Modelling and site characterisation

In order to assess the impact of atmospheric pollution on the Murujuga petroglyphs, it is essential to know the amount and kind of pollution to which they were exposed. In this study, atmospheric pollution will be measured at several sites across the Murujuga region, and the pollution levels experienced by the petroglyphs will be inferred by spatially interpolating the monitoring data.

It is vital that the monitoring network should be capable of providing a representative picture of the pollution pattern, for any combination of pollutant sources, across all expected meteorological conditions.

The importance of correct site selection is illustrated in Figure 3-18. The top panel shows a (simulated) pollution field as a colour image. Red and white dots show the locations of two possible networks of monitoring stations. The lower panel shows the result of observing the pollution levels at the red network sites and interpolating these values to other locations within the region bounded by the network. The centre panel shows the result if the white network is used. The red network can capture a reasonably accurate picture of the pollution pattern, whereas the white network records almost zero pollution everywhere and infers that the pollution is very low across the entire space.

While Figure 3-18 is an artificial illustration, our preliminary analysis has shown that during some periods of the year, with very stable atmospheric conditions prevailing over Murujuga, industrial output from a point source such as a flare stack could be transported as a relatively thin plume across the peninsula. The scenario sketched in Figure 3-18(b), where a poorly placed monitoring network fails to detect any emissions, is entirely possible.

Pollution modelling is included as an integral component of this Monitoring Program as it allows the effectiveness of a proposed monitoring network to be assessed in this manner up front, but more importantly it also allows optimisation of the network such that the network is known to be effective before deployment (cf. the red network in FigI 3-18(c)) and to what level. Furthermore, statistical performance of the network can be assessed, including scenarios where one or more monitoring stations are unavailable.

While design of the new monitoring network is the main goal, modelling and site characterisation makes it possible to assess previous monitoring campaigns objectively by assessing the effectiveness using the same approach, for example by assessing the air quality monitoring program carried out by CSIRO between 2004 and 2009 (Gillet, 2010).

Computational fluid dynamics (CFD) will aid in the final site selection as additional smaller scale models can be used to characterise local effects at a particular site, such as those because of trees, rocks or structures.

It should be noted that the aim of this component of the overall project is to design a monitoring network that is able to accurately measure and record the emissions that occur at Murujuga, and to allow reconstruction and interpolation of these measurements to rock art sites with low and quantifiable error. As such, in the modelling that underpins the design it is not necessary to perfectly capture all emission sources, or to predict the actual exposure levels, but only for the modelling to be a good representation of the conditions at Murujuga. Any modelling is strongly dependent on the quality of the assumptions that underpin it, and it is unlikely that these could be known with sufficient accuracy and certainty to predict future exposure with known error. It is also highly likely that these assumptions will change over the monitoring program. The final network needs to be robust against uncertain modelling assumptions, as well as any changes to them during the monitoring program (such as the construction or decommissioning of an industrial site). Hence, the design philosophy is that the air-quality monitoring network must allow conclusions to be drawn from measured data only and be unaffected by the initial modelling. The modelling is, however, an essential part as it provides detailed data in advance of any physical measurements, and therefore allows an optimal network to be designed up front.

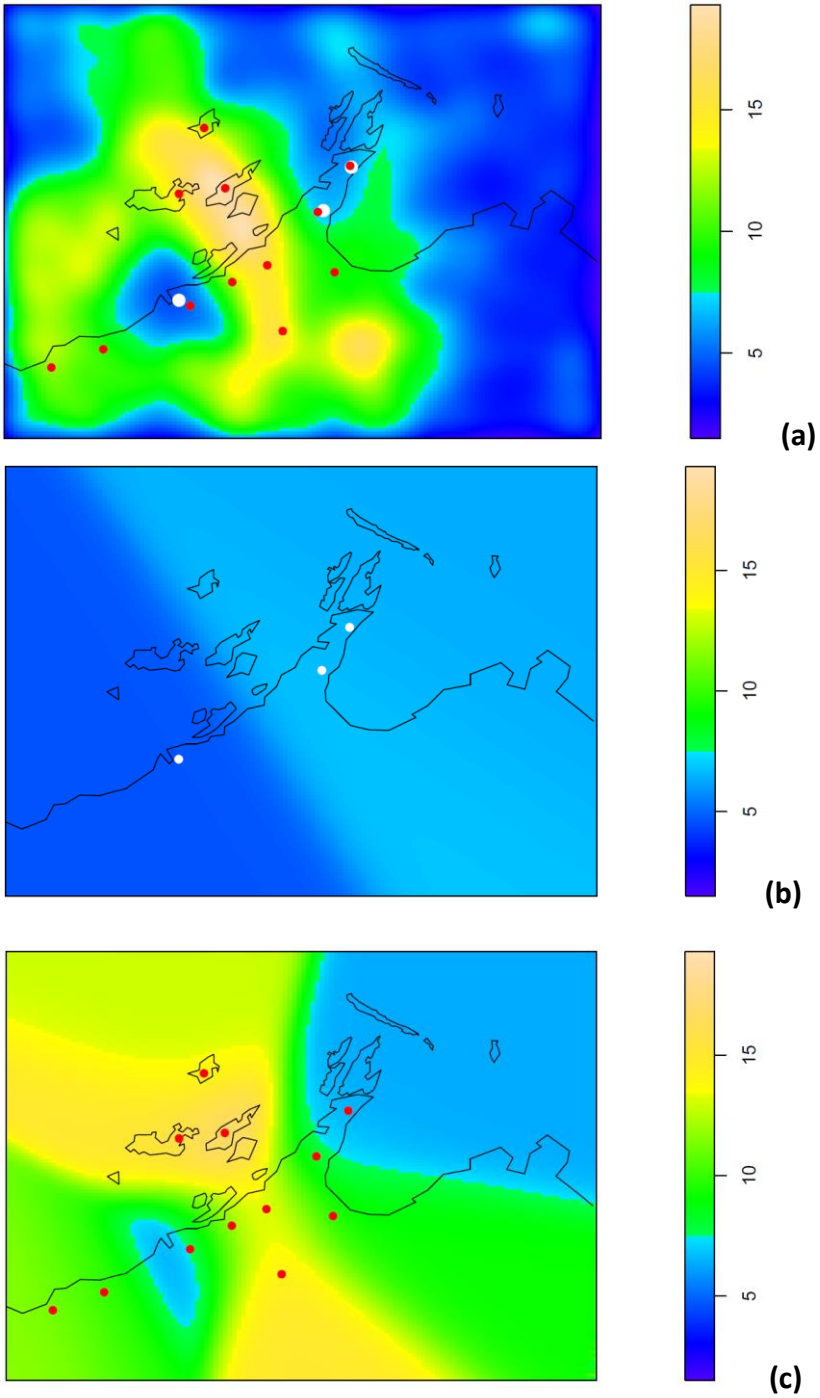


Figure 3-18 *Example monitoring network. (a) shows actual gradient with white and red points two potential monitoring networks. (b-c) show the fields calculated from the sampled data for the respective networks.*

3.7 Detailed description of the proposed data collection plan

Air-quality modelling for environmental impact assessment allows airflow and pollutant transport to be estimated under a range of emission and weather conditions. For environmental approvals dispersion modelling may be used to estimate pollutant levels over a specified time period to

establish average and maximum pollutant levels under expected (future) operating scenarios. The resolution of dispersion modelling is necessarily relatively coarse because of the timescales of the studies and complexity of the chemistry, with grid cell sizes in the order of 500 m–5 km. It is also likely that for environmental assessment the physical monitoring of air-quality is separate to the environmental impact modelling. As a result, comparisons are usually qualitative only. Because of the reduced resolution of dispersion modelling it is not possible to use it precisely locate monitoring instruments beyond the resolution of the mo–el grid (500 m–5 km).

Computational fluid dynamics (CFD) also predicts fluid flows numerically for a given set of flow inputs; however, it is conducted at a much finer resolution than dispersion modelling, with cells sizes in the order of 1–2 m. This allows any adverse effects of local features (ie. buildings and structures) to be avoided and ensure that air quality monitoring stations are reading representative values. The finer resolution though means that CFD has a higher computational cost than dispersion modelling and requires increased fidelity of boundary conditions and emissions data. As such it not possible to conduct long-term temporal studies over larger regional areas (i.e. in the order of tens of kilometres).

This project will use both these approaches, in combination with advanced optimisation techniques to establish an air-quality monitoring network which is able to determine the level of pollutant exposure of the rock art at Murujuga with confidence. At a broad scale, dispersion modelling will be used to establish an optimal network at low resolution (about 1.5 km). CFD modelling will be used to fine-tune placement of monitoring station within these regions, accounting for local features (such as roads, buildings, and terrain). These two approaches together will allow the air-quality monitoring network to be designed, characterised, and optimised before deployment. Designing the network in this way will make it possible to estimate the exposure (magnitude and estimated error) at any particular rock art site at Murujuga through mathematically rigorous interpolation of the field measurements.

i. Detailed description of the proposed methodologies

This part of the study comprises three parts, each of which are described in more detail below. For the dispersion modelling the steps in the modelling process are as follows:

1. Collect meteorological data for the regional area containing the Burrup Peninsula and use this to determine the simulation input parameters, supported by a large-scale meteorological model.
2. Establish an emissions inventory for the Burrup Peninsula, identifying emission source locations and rates.
3. Find and review other required modelling inputs (i.e. ground condition/roughness, surface elevation, etc).
4. Prepare and run the model for the meteorological conditions identified in 1, including the emission sources in 2.
5. Post-process and collate the model results, including sensibility and QA/QC checks

The steps above establish a baseline emissions ‘year’ (at an hourly sampling rate), which records the pollution levels across the whole region, albeit at low spatial resolution (in the order of 1.5 km).

Using the dispersion modelling data as a proxy for real-world measurements, it is possible to design (and test) an air-quality monitoring network virtually. The steps that will be taken to assess a potential monitoring network consisting of ‘n’ air-quality monitoring stations are:

6. Identify and sample the pollutant levels at sites where it is necessary to accurately capture pollutant exposure levels (i.e. the rock art sites). This creates virtual exposure records for the rock art sites.
7. Sample the pollutant levels from the modelling results for each of the 'n' virtual AQ monitoring stations. This creates virtual measurement records for the AQ monitoring network.
8. Using spatial analysis and statistics, interpolate from the virtual AQ station measurements to the rock art sites to predict the level of exposure at each site (locations identified in step 6).
9. Determine a measurement error for the rock art sites by comparing the interpolated pollutant measurements (from step 8), and the sampled pollutant levels for each rock art site in step 6.
10. Determine an overall measure of fitness for the network by combining the errors calculated in step 9.

Steps 6 to 10 allow a proposed AQ monitoring network to be objectively assessed. By repeating the process for a large number of candidate AQ monitoring networks, it is possible to find the best among them. This requires repeating these steps for each network and finding the one with the lowest error. Taking this further, advanced optimisation techniques allow an optimal AQ monitoring network to be identified from an exceedingly large number of potential air quality monitoring networks.

For the network design a genetic algorithm will be used to find the optimal network for a given number of AQ monitoring stations. Furthermore, by varying the number of AQ monitoring stations and repeating the optimisation it will not only be possible to find an optimal network for a specific number of monitors, it will also be possible to see how the monitoring program performance is affected as the number of AQ monitoring stations is increased. Using a genetic algorithm will also allow complex constraints to be included, such as accounting for regions where it would not be possible to site a monitoring station, and allow for measurements from existing monitoring stations to be included in the analysis.

The results from the optimisation will assist the project statisticians in determining the required number of AQ monitoring stations after considering other factors which may affect the statistical rigour (e.g. if there is a need to locate two AQ monitoring stations in proximity). After this assessment the final locations of the AQ monitoring stations will be known to within about 1.5 km.

These 1.5 km regions will allow sample selections to be made for co-located samples in each of the complementary study programs. For these regions, CFD simulations will be made of the sites and will assist in fine tuning the location of each AQ monitoring station, accounting for any adverse effects because of local geometry or emissions (i.e. industrial stacks, gorges, rocks, roads, etc) that may not have been resolved in the broad scale dispersion modelling.

The steps that will be taken in the CFD modelling are listed below, (these are similar to steps 1 to 5, albeit at a finer scale):

11. Collect and analyse meteorological data for each site to establish the variability in conditions and determine the simulation input parameters required to adequately capture the conditions at each AQ monitoring site (or group of sites).
12. Revise, and review the broad scale emissions inventory for the Burrup Peninsula to refine the locations and rates, accounting for the increased resolution of the model.

13. Create a 3D topographical model of the simulation domain.
14. Prepare and run the model for the conditions identified in step 11, including the emission sources identified in step 12.
15. Post process the results for each site to identify any potential issues at this finer scale (< 5m).

Note that the aim of the CFD is to ensure that there are no local features or flow patterns that would adversely affect the measurement at a given site and make them unrepresentative. The CFD modelling is not intended to be a tool to predict the actual exposure levels at a given location.

Further details on the proposed approach are given below.

ii. Existing data – dispersion modelling

Broad scale dispersion modelling (steps 1 to 5) was separately commissioned by DWER and was undertaken Ramboll, with the assumptions and results summarised in their report (Ramboll, 2021). Through DWER, the project team has access to the raw input and result data from the model runs made by Ramboll. This data has been checked and assessed and will be the source data for the broad scale AQ monitoring network optimisation.

Figure 3-19 shows data for SOx and NOx from the Ramboll model runs. The dispersion was modelled on a 1.3 km grid, at hourly temporal resolution to generate one year of data. Meteorological inputs were calculated using WRF (weather research and forecasting model), and cross checked against measured meteorological data for a 2014 weather year. Ramboll modelled many emissions scenarios, with the 2030 scenario best matching the expected emissions inventory over the duration of the Murujuga Rock Art Project. The 2014 weather year was used for all scenarios modelled by Ramboll.

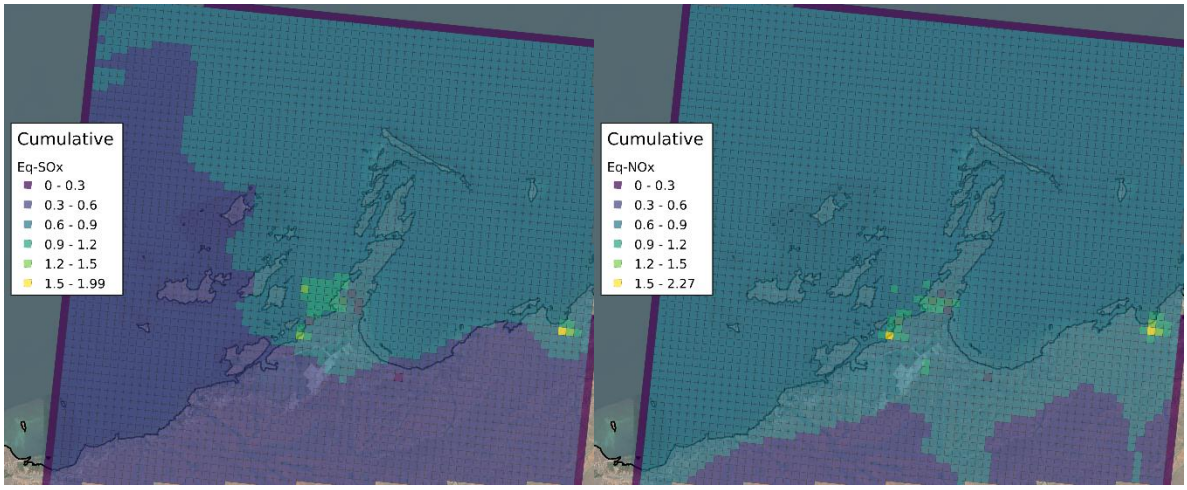


Figure 3-19 Ramboll modelling data at 1.3km grid resolution (annual cumulative NOx, SOx, 2030 model year).

iii. Existing data – rock art sites

The aim of the AQ monitoring network is to establish the exposure to emissions of the rock art at Murujuga. To account for this in the AQ monitoring network design the sampling error of the network is weighted by the prevalence of rock art. The WA Department of Planning, Lands and Heritage (DPLH) curates the Register of Aboriginal Sites or Heritage Places and this data will be used to determine the weighting. The full dataset kept by DPLH will be used for the analysis as it is more detailed than the publicly available register. Figure 3-20 shows the raw database after filtering to

exclude entries that are not engravings or artworks and the resulting weightings after resampling on the 1.33 km grid.

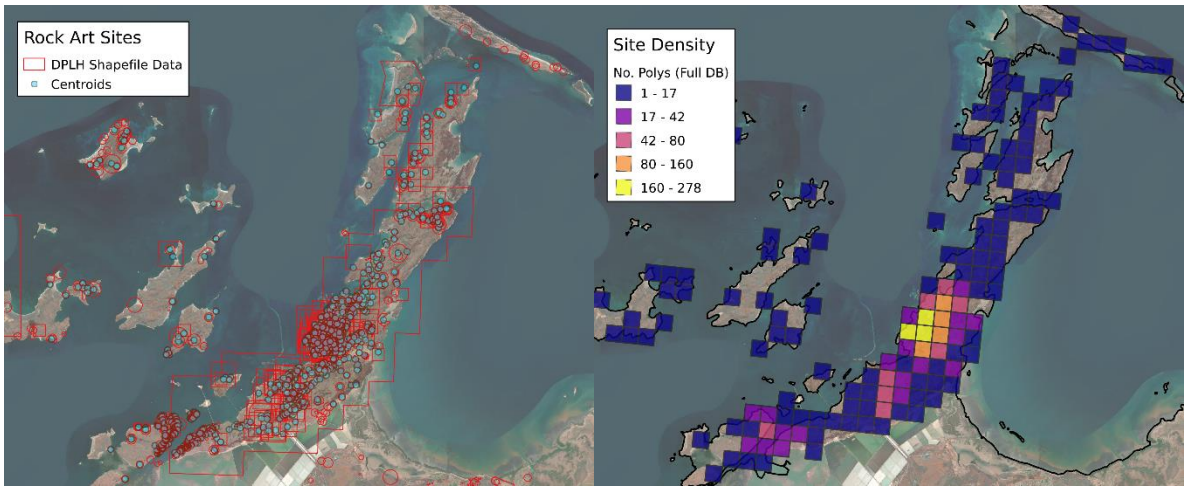


Figure 3-20 DPLH registered site data, original data and resampled for weighting.

iv. Existing data – allowable monitoring station locations

As stated above, the 1.33 km grid used in the Ramboll modelling will form the basis for broad scale siting of the AQ monitoring stations. Allowable locations for siting new stations have been identified after accounting for water bodies and the regional extent of the study. The possible locations are shown in Figure 3-21. There are nine existing monitoring stations which are expected to be available for the study or will be recommissioned in this project (shown in red), and there are two monitoring stations which are remote from the study region. These may optionally be included in the network if selected during the optimisation (shown in orange).

'Virtual' samples will be taken directly from Ramboll model data in the matching grid cells. Optimisation will be based on 'Eq-NOx' and 'Eq-SOx', as these occur across the Archipelago. Ammonia compounds were excluded because they were expected to bias the optimisation as they are concentrated around Yara Pilbara's facilities only, while fine particulate was excluded on two grounds: firstly there is an abundance of iron across the Pilbara, so any chemical weathering processes were assumed to not be limited by a low level of iron compounds; and secondly, there were some issues raised concerning the modelling inputs for fine particulates in the data that the project has access to (corrected results will not be available before site selection needs to be finalised).

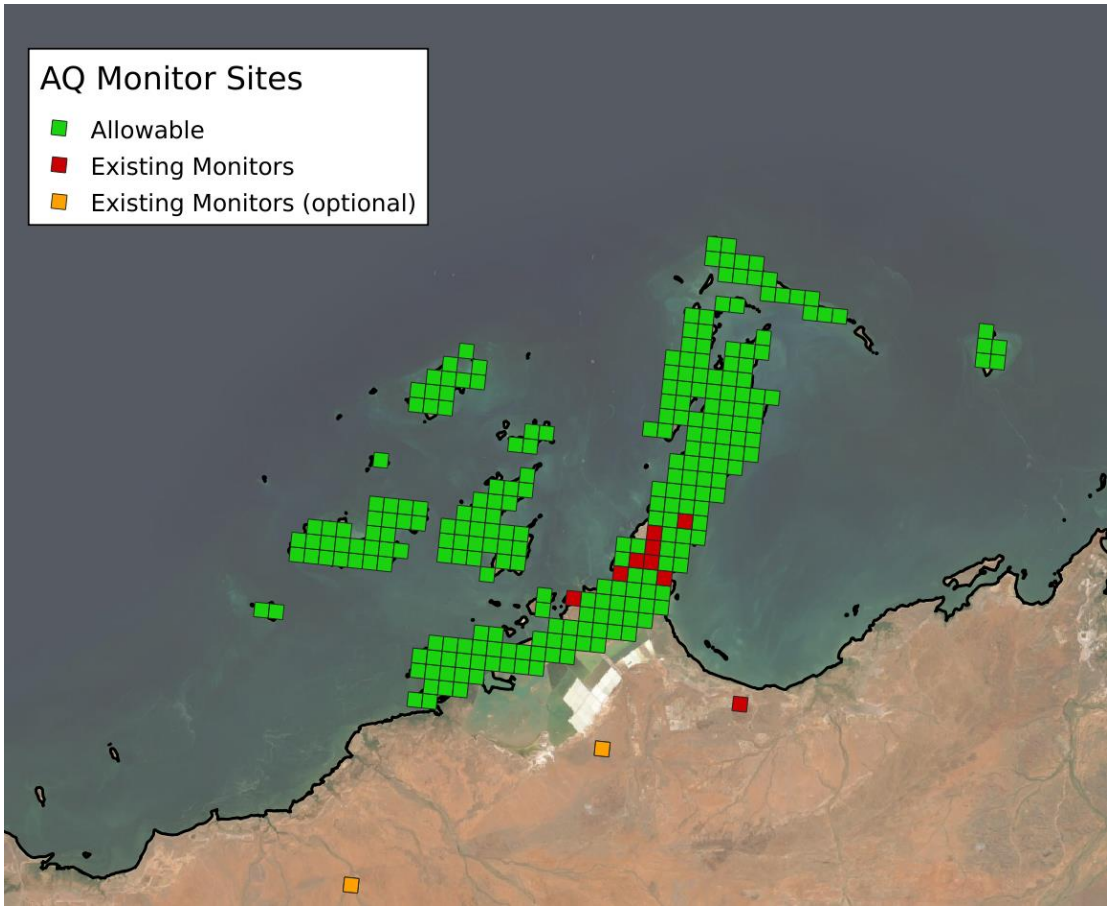


Figure 3-21 Allowable locations for additional monitors forming part of the AQ monitoring network.

v. Spatial Analysis and Interpolation

Interpolation of the measured air quality at the monitoring sites is required to predict the species concentrations at locations that are not coincident with a monitoring station. For spatial data interpolation a kriging process will be used. This process makes use of the fact that nearby monitors are expected to record similar readings. Analysis of the Ramboll data will be made in collaboration with the project statisticians to tune the kriging process. This will then feed into the optimisation process.

vi. Optimisation

There is an exceptionally large number of combinations of air quality monitoring sites that could form a network for measuring the air quality at Murujuga. Figure 3-21 shows 258 potential monitoring sites, and even when only considering these there are between 10^{13} and 10^{30} possible combinations (for an additional seven and 20 monitoring stations, respectively) which could form a monitoring network. For this reason, numerical optimisation is desirable, as it can efficiently search potential combinations of monitoring networks to find those which give the best performance (lowest error), while also accounting for other constraints.

As stated above, a genetic algorithm (GA) will be used to perform the optimisation of the AQ monitoring network. A GA works by creating several (random) networks from the possible combinations (the population). These are then iterated through generations, where in each generation each candidate network (an individual) is assessed against how well it meets the

optimisation objective (its fitness). After the fitness of each individual is assessed, the population is ranked, and a number of processes follow to improve the overall fitness of the population. These are (typically): dropping the worst performing individuals and replacing them with new individuals, swapping characteristics between individuals in the population (breeding), and introducing random changes to an individual (mutation). This allows better exploration of the problem space. The end result is that after a sufficient number of successive generations (i.e. through repeating the process), the population should find a globally optimal solution, noting that each run may not necessarily converge on the same solution. This approach will be used to identify an optimal air quality monitoring network for Murujuga.

After the optimisation has been conducted, the project statisticians will review the results, and incorporate them when making the final site selections.

vii. CFD simulations

Following the selection of the monitoring station locations, simulation domains for CFD modelling will be created and simulations made to investigate any local, fine scale flow and pollution transport issues under the prevailing conditions identified from meteorological data. Particular attention will be paid to point and line source emissions in the modelling as these have strongly localised effects and are likely not precisely captured in the dispersion modelling.

Species transport will be used to model each of the pollutant species, with source terms matching those identified in the emissions inventory. For particulate species (which have non-negligible settling), settling will be accounted for using a representative settling velocity, after Wu *et al. (2000)*, which has been used successfully for other particulate studies.

OpenFOAM will be used for the modelling as it exhibits exceptional scalability on high-performance computing (HPC) systems and has well validated and fully auditable source code. As it is open-source it is also possible to develop and use custom tools for solving as well as pre- and post-processing.

These simulations are relatively large, so access to HPC resources is essential. This requirement will be met by the Pawsey Supercomputing Centre, with a commercial HPC provider also available if required.

Representative domains and a sample result are given in Figure 3-22. Depending on the prevailing wind direction and the location of the monitoring stations, domains will be adjusted to encompass more than one monitoring station where possible.

Validation of the models to be used in the CFD simulations has been undertaken to ensure that the modelling will be able to accurately predict the flow and species transport from the emissions sources identified in the emissions inventory.

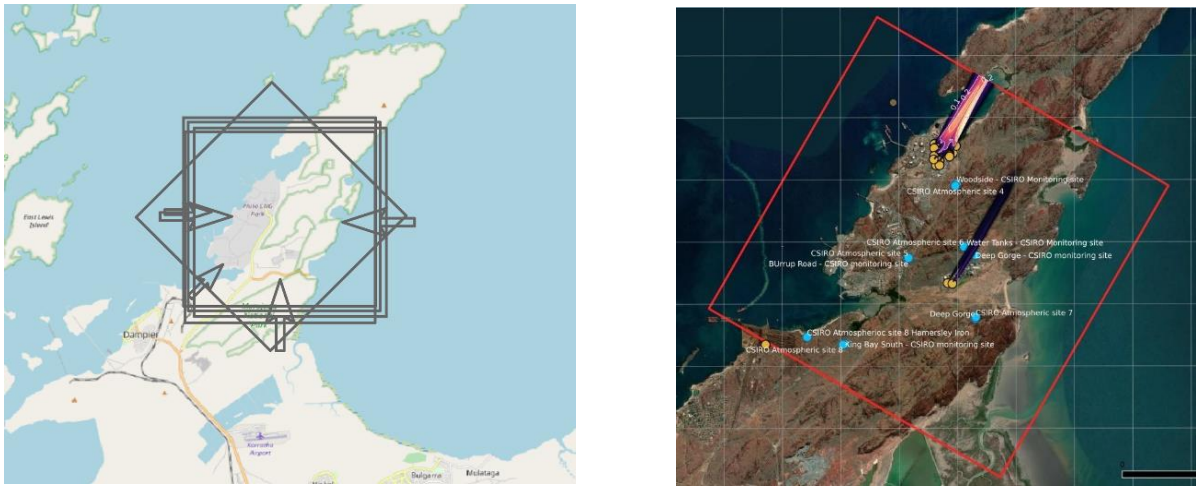


Figure 3-22 Sample domains for CFD analysis (left) and sample simulation result (right).

viii. Data sources

The following section gives further detail on the inputs that will be used in the CFD modelling. Assumptions, Inputs, and results of the dispersion modelling are detailed in the report by Ramboll (Ramboll 2021).

Meteorological data

Meteorological data has been sourced from the Bureau of Meteorology for Karratha Aero (Station 4083) from 2015-2019 (the most recent five years at project commencement). This data was subsequently grouped based on wind speed and direction. Wind directions were classified based on eight compass directions – N, NE, E, SE, S, SW, W, NW, while four wind speed bands were selected as in Table 3-10.

From the analysis the 14 most common conditions account for 90% of observations and are shown in Table 3-11 with cumulative occurrences (a more expansive table is given in Appendix III). These conditions also capture the extremes of wind speed – a calm wind condition (from the south), and a high wind condition (from the west).

Table 3-10 Windspeed band and range at Karratha Aero.

Windspeed band	Windspeed range	% Occurrence
Calm	0 – 2 m/s	6.6
Low	2 – 5 m/s	42.4
Medium	5 – 10 m/s	47.4
High	> 10 m/s	3.6

Table 3-11 Conditions accounting for 90% of observations, with cumulative occurrences.

Rank	Wind speed	Wind direction	Average wind speed in band (m/s)	% Occurrence	Cumulative % occurrence
1	Medium	W	7.2	19.5	19.5
2	Low	SW	3.5	8.5	28.0
3	Medium	NE	6.9	8.5	36.5
4	Low	W	3.9	8.0	44.4
5	Medium	NW	6.4	6.7	51.2
6	Low	S	3.2	6.3	57.4
7	Medium	E	7.4	5.3	62.7
8	Low	NW	3.8	4.9	67.6
9	Low	NE	3.7	4.8	72.4
10	Low	E	3.6	4.0	76.3
11	Low	SE	3.3	3.5	79.9
12	Medium	SW	6.4	3.2	83.0
13	Low	N	1.3	2.8	85.9
14	High	W	10.4	2.1	88.0

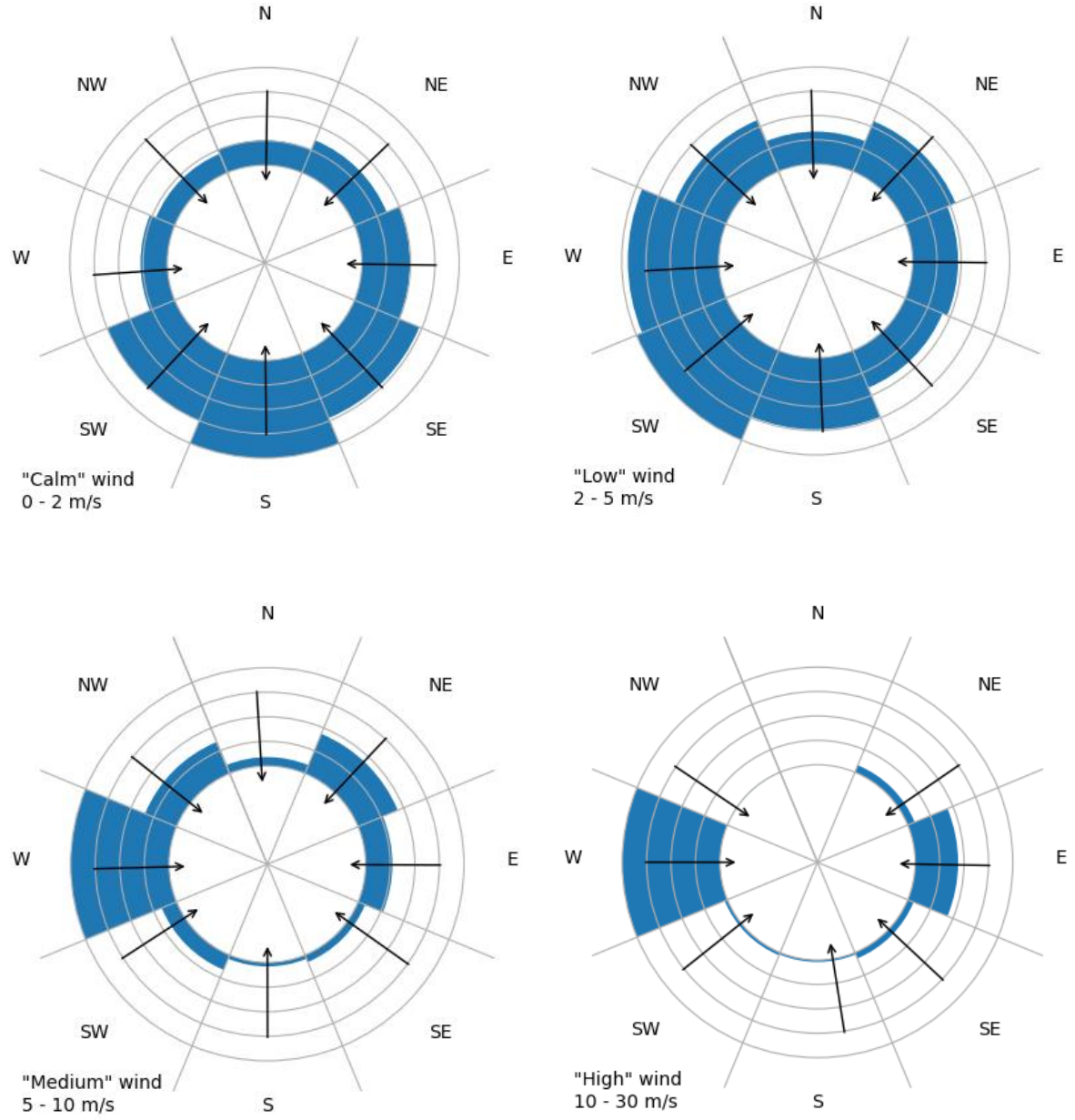


Figure 3-23 Wind data for Karratha, grouped by wind speed and direction.

This data will be the primary source for the wind conditions simulated with CFD. The purpose of these simulations is to identify any potential problems with final site locations (in the order of 10s of metres) and not to perform a statistically rigorous analysis, and it is likely that the angular resolution of wind data as classified above is not resolved finely enough for complete characterisation. Where results indicate that there may potentially be an issue at wind directions not covered above, intermediate directions may be simulated as well.

Emissions inventory

The emission inventory will be primarily drawn from the Ramboll input data. Where necessary (i.e. in the case where resolution is insufficient) recent data may be found in past environmental impact studies and/or directly from the industries operating on the Burrup Peninsula. For example, the most

recently available Air Quality Impact Assessment was finalised in March 2020 for the Perdaman Urea Project and includes relatively detailed emissions data.

The emission sources will be kept the same for all simulations.

Modelled species

The species that will be included in the modelling are:

- NOx
- SOx
- NH_3
- PM_{10}
- $PM_{2.5}$

Site topography

The high resolution of the proposed CFD models means that high resolution topography of the Peninsula and relevant Islands is required. Initial simulations made use of the 1-arc second resolution Digital Elevation Model (DEM) bare earth dataset published by Geoscience Australia, captured by the Shuttle Radar Topography Mission (SRTM), subsequently filtered, and smoothed to remove buildings and vegetation (among other features). In June 2020 Landgate released a 1 m resolution DEM of the Burrup Peninsula, created from aerial imagery taken in March 2020. This DEM has been received from Landgate as well as the point cloud used to create it. This dataset will be used for the base topographic data. Buildings/structures will be recovered from the point cloud data where possible. Beyond the extents of the Landgate data the DEM will be supplemented with the 1-sec SRTM mission data as required. Further local refinement (obtained by laser scanning) of proposed sites may supplement this data to provide a still higher resolution for the geometry.

Post-processing and quality assurance

The results from each simulation will be post-processed to extract the pollutant species concentration at ground level, and other data necessary to sanity and QA check the simulations (such as convergence data and residuals). The resulting suite of simulation results will cover the prevailing conditions at Murujuga and will assist in final siting of the monitoring stations within the 1.3 km regions identified in the initial site selection.

4 Data Management

4.1 Standard data management practice

The Monitoring Program team will follow standard data management practices, following the Data Management Plan and Data Access Agreements developed as part of the project.

All source data will be stored in CloudStor as it becomes available; this includes raw and processed data, and model and analysis output data. The data will be made available to each Program affiliate according to Program role and will be partitioned by each specialist research group. Data request forms are available in the Data Access Agreement and Management Plan. Requesters are required to complete the forms provided by the Data Manager (at Curtin Univerity) via Calibre. The forms will also be reviewed and sent to DWER for approval. Secure download links can also be made available for one-off access to specific data if needed.

Activity-specific data management approaches are outlined below.

4.2 Spectral imaging

All data captured will be stored in the following:

- Raw scan format (both proprietary format from the scanner and exported as an open standards format).
- The registered and geo-referenced data into a unified point cloud, including any transformations, and validation/error checking results.
- Processed model in the format desired for further processing with respect to other parts of the Monitoring Program (e.g. ortho images, mesh surfaces, digital terrain models).

The storage of the raw data allows for reprocessing of the data, if required, by third parties, or to check and validate the output and analysis using different and emerging software. For laser scanning, this will likely be in the Leica format, and the open format of LAS or e57. For the photogrammetry it will be in the raw image format (uncompressed jpeg or tiff files depending on the camera specifications). In the past, different software versions (let alone different parameters or software) have caused different results. For example, the photogrammetric software Agisoft Metashape produces slightly different results each time an adjustment is performed, even when using the same images and program settings. By storing it in an open format, it ensures the ability to reuse the data with different software and improved techniques in the future, without worrying about legacy or proprietary formats.

The aligned, registered, and geo-referenced data is also stored, along with all the input and output parameters used and produced. This will depend on the specific software solution, but in most cases it will be similar. For photogrammetry, this will include the images, ground control points (with their survey method and accuracy), number of tie points to use (and accuracy or image resolution to base these on), camera properties, and calibration parameters if known (including geometric and spectral calibration properties). As outputs will be the matched feature (tie) points, re-projection errors, control and check point errors, camera calibration values (if a self-calibration is performed, with the estimated parameter errors), camera locations and orientations (and error estimates), overlap and dense image reconstruction. For the laser scanner, the input will be the individual setups and the control points (with their survey method and accuracy). The output is the combined point cloud,

error alignment and overlap between point clouds and control points (the uncertainties are harder to derive but are calculated based on the RMS of the overlapping regions). For all output, the report will also be stored to enable reprocessing and validation as required.

For the outputs, these will be dependent on the needs of the sites. These can include ortho-images, digital elevation and surface models, meshed surfaces, or analysis of these and the raw point files (e.g. vegetation density, surface orientations and normal, roughness index). The outputs of these will be stored in relation to the other parts of the Monitoring Program, but generation will be documented and stored. This documentation at a minimum will include software, methods and parameters used (and any reports generated), as well as any scripts or codes used in their generation to ensure that their production can be repeated and justified as needed.

4.3 Microbiome

The data obtained from the initial screening of microbial communities through 16S/18S and ITS profiling will form the basis for the selection of the smaller sample set for more in-depth metagenome and metatranscriptome profiling to study the physiological activities of the key members of the varnish microbiome. Each step in this process generates additional data and the bioinformatics and biostatistics approaches have been integrated after each practical step.

In collaboration with other members of the research team, the microbiome researchers will use advanced ordination and statistical analysis to determine the environmental parameters that play a significant role in stimulating an accelerated growth of varnish-building bacteria/fungi/lichens versus the development of varnish-dissolving acidic microbial biofilms.

Sequence data will be processed using various highly specific third-party, command-line based bioinformatics packages that are installed at the Pawsey Supercomputer (Nimbus server). Both the raw and processed sequence data as well as the results stemming from the analysis will be stored on Cloudstor.

4.4 Air quality control

Data from the weather stations and real-time instruments, where possible, will be live streamed to the Cloudstor repository. Where this is not possible the data will be recovered on the sample collection trips to the monitoring stations and uploaded to Cloudstor manually.

Data from the laboratories (ChemCentre and Gradko International) will be uploaded to the Cloudstor in the form of reports and extracted from the reports for analysis as required.

4.5 Computational fluid dynamics modelling

Simulations will be prepared on the workstation machines before transferring to supercomputing resources at the Pawsey Supercomputing Centre. Completed simulation cases will be transferred to the Monitoring Program repository as soon as practicable.

Post-processing is likely to occur on both personal workstations and supercomputing resources; in each case completed analyses will be transferred to Cloudstor as soon as practicable.

Where possible, analysis will be scripted to ensure consistency between analyses, and minimise the likelihood of transcription or process errors.

Standard CFD methods will be applied for validation, verification, and quality assessment. These include residual monitoring, grid independence and checking compliance with turbulent model constraints. Where necessary, additional validation simulations will be carried out to ensure that models are appropriate. This data will be stored in the Monitoring Program's CloudStor repository.

5 Study locations and indicative activity timing

Figure 5-1 shows the coarse-scale site selection, conducted using a combination of Kriging-Variance (blue and magenta sites) and randomised sites (green). Figure 5-2 shows the major activities planned for each type of site.

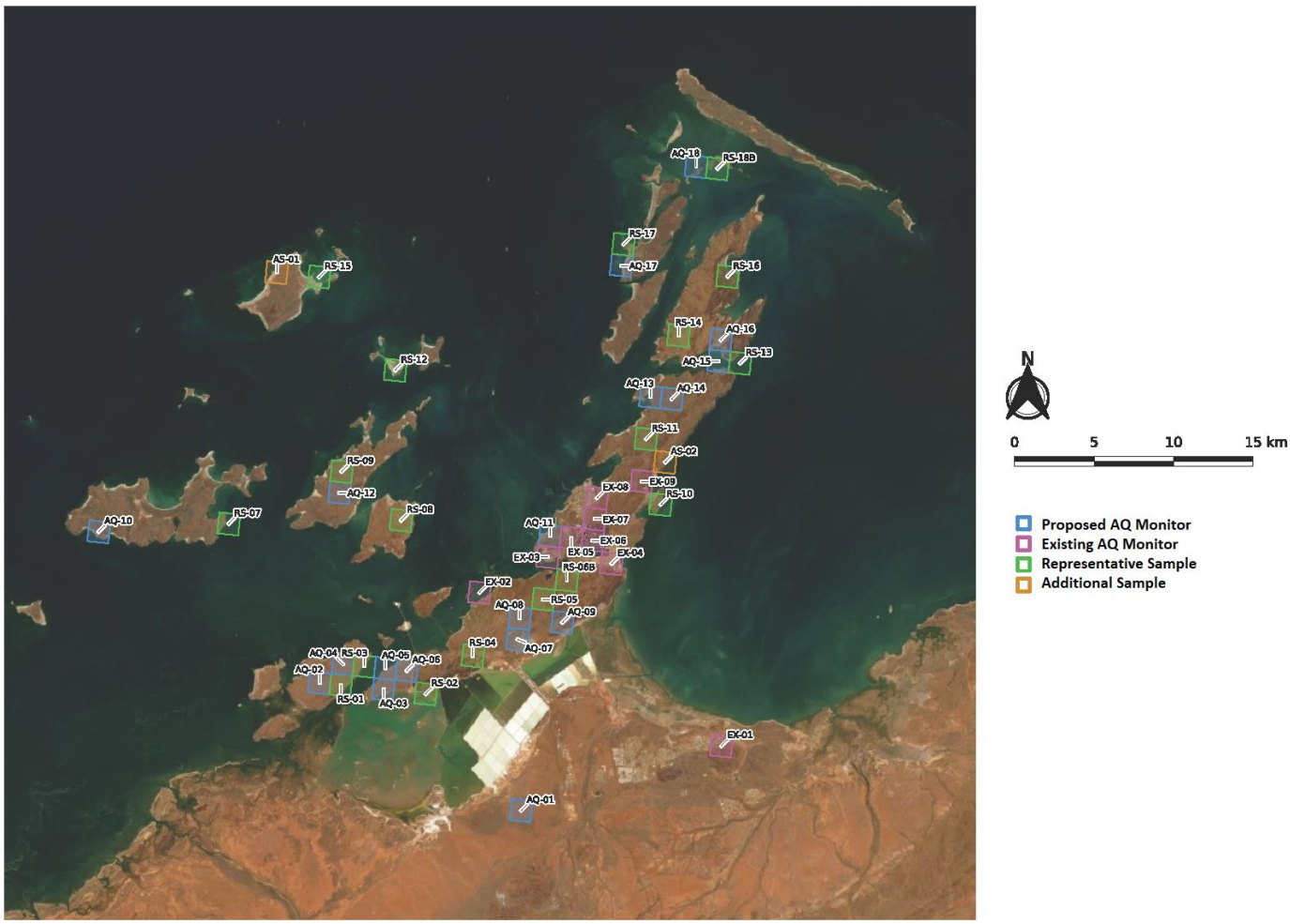


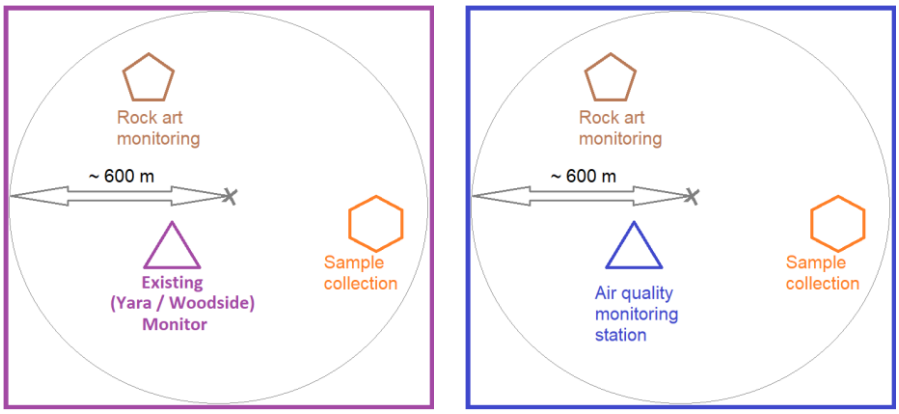
Figure 5-1 Optimised site selection.

For the study squares shown in Figure 5-1:

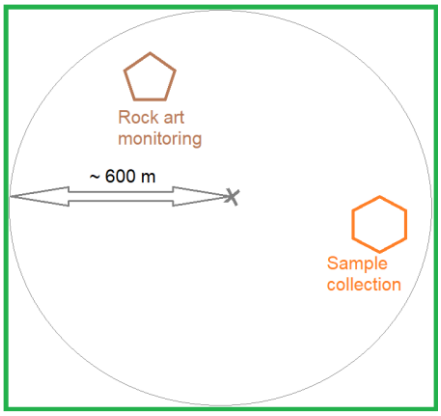
- magenta ‘EX’ denotes existing Air Quality (AQ) monitoring stations included in the design
- nlue ‘AQ’ denotes proposed AQ monitoring sites.
- green ‘RS’ denotes sample survey sites.
- orange ‘AS’ denotes additional sites to capture known dolerite and granite rock art.

Blue ‘AQ’ and magenta ‘EX’ sites are identical, apart from a pre-existing AQ monitor at the former. The study design has been adapted to include additional (orange ‘AS’) sites where known rock art exists on either dolerite or granite.

Green 'RS' sites have been randomly selected from an appropriate sample of rock types of interest (see Appendix I). These sites will only be used for sample collection and rock art monitoring (if present).



(a) Existing AQ Monitor sites *(b) New AQ Monitor Sites*



(c) Representative Sample Sites

Figure 5-2 Key activities at each site type.

5.1 Selected study samples

Since the original version of this document, preliminary fieldworks to select study sites has been undertaken. This has allowed us to:

(a) finalise the sample numbers for the first phase of the study

(b) adapt the study design to incorporate the abundance or rock art found on basalt and granite as well as adjust the site selection criteria to include known rock art sites with petroglyphs on dolerite and granite.

Tables 5-1 to 5-5 summarise the selections for each study site/square.

Table 5-1 Summary statistics for all sample squares.

Site	Granophyre		Gabbro		Dolerite		Granite		Basalt		Soil sample*
	S	A	S	A	S	A	S	A	S	A	
Cumulative totals	31	30	12	13	8	5	6	2	7	4	30
* Indicates soil present proximal to at least one sample rock in square											

Table 5-2 Samples selected in AQ (blue) squares.

Site ID	Granophyre		Gabbro		Dolerite		Granite		Basalt		Soil sample	Notes
	S	A	S	A	S	A	S	A	S	A		
AQ01	0	0	0	0	0	0	0	0	0	0	Y	Only sand and pisoliths present in square. Powered AQM
AQ02	1	1	0	0	0	0	0	0	0	0	Y	Dolerite present in square but no rock art found
AQ03	1	1	1	1	*	1	0	0	0	0	Y	* Dolerite sample matched with AQ5
AQ04	1	1	0	0	0	0	0	0	0	0	Y	Dolerite present in square but no rock art found
AQ05	1	1	0	0	1	*	0	0	0	0	Y	* Dolerite art matched with AQ3
AQ06	1	1	0	1	0	0	0	0	0	0	*	Only gabbro deposit in square highly significant site so no sample selected. Soil sample possible at granophyre.
AQ07	1	1	0	0	0	0	1	1	0	0	Y	AQ7 and AQ8 AQ monitors located ~25m apart
AQ08	1	1	0	0	0	0	0	0	0	0	Y	AQ7 and AQ8 AQ monitors located ~25m apart
AQ09	1	1	1	1	0	0	0	0	0	0	N	Monitor and granophyre samples slightly outside square boundary
AQ10	1	1	0	0	0	0	0	0	0	0	Y	
AQ11	1	1	0	0	0	0	0	0	0	0	N	
AQ12	0	0	0	0	0	0	0	0	1	1	Y	
AQ13	1	1	0	0	0	0	0	0	0	0	N	

Site ID	Granophyre		Gabbro		Dolerite		Granite		Basalt		Soil sample	Notes
	S	A	S	A	S	A	S	A	S	A		
AQ14	1	1	0	0	0	0	0	0	0	0	Y	
AQ15	0	0	1	1	0	0	0	0	0	0	N	
AQ16	0	0	1	1	0	0	0	0	0	0	Y	
AQ17	1	1	0	0	0	0	0	0	0	0	Y	
AQ18	1	1	0	0	0	0	0	0	0	0	Y	
Totals	**14**	**14**	**4**	**5**	**1**	**1**	**1**	**1**	**1**	**1**	**14**	

Table 5-3 Samples selected in EX (Magenta) squares.

Site ID	Granophyre		Gabbro		Dolerite		Granite		Basalt		Soil sample	Notes
	S	A	S	A	S	A	S	A	S	A		
EX01	0	0	0	0	*	*	0	0	0	0	*	Requires discussion with Ngarluma Aboriginal Corporation regarding rock samples and monitoring. Woodside Powered Health AQM. Plan to add deposition monitor and rock tiles.
EX02	1	1	0	0	0	0	0	0	0	0	Y	Powered AQM to be installed (Ex DWER site). Rock art and rock sample slightly beyond square; however, only ~800m from AQM.
EX03	1	1	0	0	0	0	0	0	0	0	N	Heavily disturbed area, little/no undisturbed soil areas.
EX04	0	0	1	1	0	0	0	0	0	0	N	Rock art on granite present however excluded on cultural grounds.
EX05	1	1	0	0	0	0	0	0	0	0	N	
EX06	1	1	0	0	0	0	0	0	0	0	N	
EX07	1	1	0	0	0	0	0	0	0	0	Y	Existing health monitor. Additional active and passive monitors to be added if possible or neighbouring station to be built.
EX08	1	1	0	0	0	0	0	0	0	0	N	
EX09	1	1	1	1	0	0	0	0	0	0	Y	Powered AQM site on Mt Wongama. Undisturbed soil samples possible near Gabbro

Site ID	Granophyre		Gabbro		Dolerite		Granite		Basalt		Soil sample	Notes
	S	A	S	A	S	A	S	A	S	A		
												and Granophyre.
Totals	**7**	**7**	**2**	**2**	**0**	**0**	**0**	**0**	**0**	**0**	**3**	

Table 5-4 Samples selected in RS (green) squares.

Site ID	Granophyre		Gabbro		Dolerite		Granite		Basalt		Soil sample	Notes
	S	A	S	A	S	A	S	A	S	A		
RS01	1	1	0	0	0	0	0	0	0	0	Y	
RS02	0	0	1	1	0	0	1	0	0	0	Y	Much of site excluded by leaseholder on safety ground (steep terrain). Soil near granite.
RS03	1	1	0	0	0	0	1	0	0	0	Y	
RS04	1	1	1	1	1	0	0	0	0	0	*	Soil sample near Granophyre possible. Location selected via random selection from visible rock outcrops, then random x, y position within outcrop. Granophyre rock art selected as only culturally appropriate panel found.
RS05	2	1	0	0	1	0	0	0	0	0	N	2nd sample rock selected in case of difficulty approving initial selection. Selection via random division
RS06B	1	1	0	0	0	0	0	0	0	0	N	Random selection of divisions and random selection between first five rock art panels found in division
RS07	0	0	0	0	0	0	0	0	1	1	N	Intertidal – no/limited soil.
RS08	0	0	0	0	1	1	0	0	1	1	Y	RND 1 lowest at/near (Basalt) outcrop. Dolerite samples found near RND 98.
RS09	0	0	0	0	1	1	0	0	2	1	Y	Rock art and 2nd sample rock on Basalt is atypical basalt in transition region beside Dolerite dyke
RS10	0	0	1	2	0	0	1	0	0	0	N	RND 4 lowest in Gabbro region. RND 20 lowest in Granite region. Rock art re-selected as closer panel found to RND 4 (<25m). Original retained as unusual

Site ID	Granophyre		Gabbro		Dolerite		Granite		Basalt		Soil sample	Notes
	S	A	S	A	S	A	S	A	S	A		
												plagioclase feldspar in patina.
RS11	1	1	0	0	0	0	0	0	0	0	Y	
RS12	0	0	0	0	0	0	0	0	1	0	Y	No rock art found in square.
RS13	0	0	1	1	0	0	1	0	0	0	N	
RS14	1	1	0	0	1	0	0	0	0	0	Y	
RS15	0	0	0	0	0	0	0	0	1	0	N	Very weathered Basalt. No rock art found.
RS16	0	0	2	1	0	0	0	0	0	0	Y	2nd sample rock unusual patina – likely biofilm because of animal activity
RS17	1	1	0	0	0	0	0	0	0	0	Y	Selected via x/y coord generated in field.
RS18B	1	1	0	0	0	0	0	0	0	0	Y	Rock art sample selected by randomly selecting from visible rock outcrops, then random x,y position within outcrop. Rock sample selected via random coordinates relative to rock art panel.
Totals	**10**	**9**	**6**	**6**	**5**	**2**	**4**	**0**	**6**	**3**	**11**	

Table 5-5 Additional samples selected in AS (orange) squares.

Site ID	Granophyre		Gabbro		Dolerite		Granite		Basalt		Soil Sample	Notes
	S	A	S	A	S	A	S	A	S	A		
AS01	0	0	0	0	1	1	0	0	0	0	*	Opportunistic dolerite sample on Rosemary. Soil sample possible but in drainage channel.
AS02	0	0	0	0	1	1	1	1	0	0	*	Region slightly outside EX09. Soil sample possible near granite.
Totals	**0**	**0**	**0**	**0**	**2**	**2**	**1**	**1**	**0**	**0**	**2**	

5.2 Approximate schedule

Figure 5-3 shows the approximate schedule for the first 12-18 months of field and laboratory studies, including additional validation studies, commencement of petroglyph monitoring, deployment of air quality monitoring and sampling of non-engraved rocks. Additional studies such as topographic scanning will be conducted after detailed site selection and once appropriate approvals are in place

to confirm AQ monitor location. Discussions are ongoing with the Murujuga Circle of Elders as to whether the rocks will remain in-situ or be relocated to the MAC offices for secure storage post sample collection.

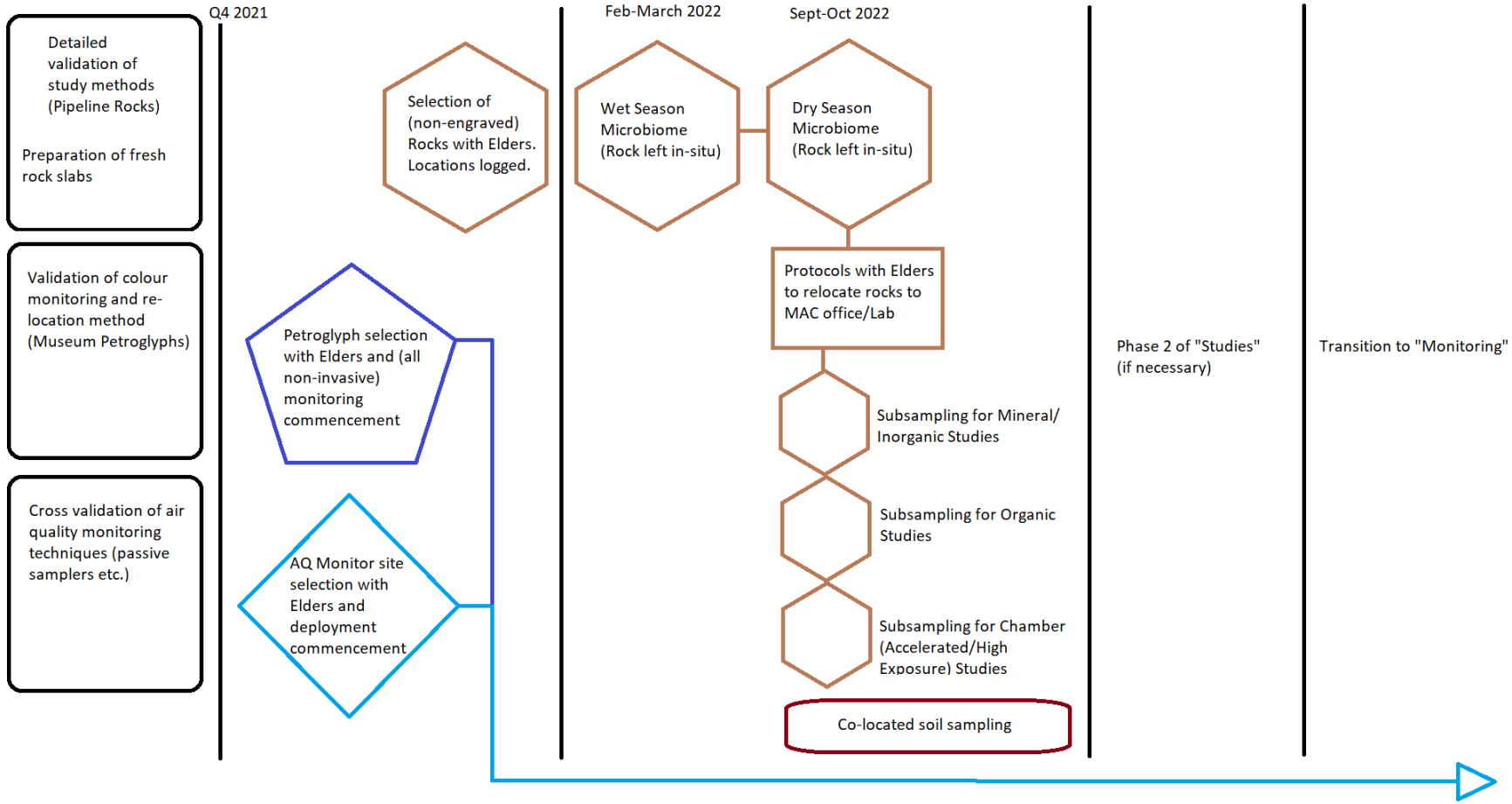


Figure 5-3 Outline schedule for the first 12-18 months of studies.

6 Summary

This document summarises the limitations and future utility of previous studies, which will be incorporated into meta-analyses where possible.

The study design for further work proposes a dual-armed approach of field and laboratory measurements. Both sets of studies aim to characterise all relevant bio-physico-chemical relationships which are occurring on the Murujuga rocks, or which may occur at foreseeable environmental conditions. The authors believe the works proposed for the first 12 months of monitoring studies will provide a comprehensive, robust, and resilient means of determining the response of the rock patina to air pollutants. However, the study design permits additional detailed studies to be undertaken, if warranted, before transition to an ongoing monitoring framework.

Preliminary fieldworks have been conducted, visiting all proposed study sites with Murujuga Elders and rangers, as well as geology and archaeology teams. This has permitted scientifically and culturally appropriate sites, rock art panels and sample rocks to be selected. This work also permitted us to observe the complexity and heterogeneity of the patina, as well as to commence the compilation of a pXRF dataset for the study sites. Through this work, an abundance of rock art on basalt was found, as well as several significant sites with rock art on granite. This discovery has led to the inclusion of these two additional rock types in the study design.

We are confident that the approach presented in these documents will provide sufficient depth and breadth of data, as well as statistical power, to permit the development of EQC for specific air quality parameters, and ultimately lead to the development of an overarching EQMF.

Refer to Appendix I and II for further detail on the site selection and sample size determination and Section 5 for the samples selected.

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9 Glossary and Acronyms

9.1 Glossary

Anthropogenic	From human activity. In the context of this research anthropogenic includes human impact, including industrial, transport, tourism, site management, and all other impact that can be attributed to human activity. It can also be considered to include distal or global human activity which may impact the natural environment through changes in climate.
Barcoding gene	A gene that is shared by a group of organisms but differs in the genetic code between species because of evolutionary changes. Sequencing analysis of these genes in environmental samples can reveal evolutionary differences between the organisms within the group. This makes it possible to classify bacteria that cannot be distinguished based on morphological features.
Biomarkers	Organic compounds produced from natural degradation of biochemicals produced by living organisms. The structure of a biomarker can sometimes be linked to a biochemical produced by a specific organism or group of organisms, while others are more general. They are known as 'molecular fossils' as they can be used to infer the presence of certain organisms in ancient environments.
Biofilm	A biofilm growing on a surface comprises a syntrophic (feeding together) consortium of microbial cells that are embedded in a slimy extracellular matrix that is composed of extracellular polymeric substances (EPSs). The bacteria that live close together can 'communicate' with each other (share nutrients, exchange genes to make them immune to antibiotics etc.)
Chemoautotrophic	Chemoautotrophs are organisms that are able to synthesise their own organic molecules from the fixation of carbon dioxide. They are able to produce their own source of food, or energy. The energy required for this process comes from the oxidation of inorganic molecules such as iron, sulfur or magnesium. Chemoautotrophs are able to thrive in very harsh environments because of their lack of dependence on outside sources of carbon other than carbon dioxide.
Chemolithotrophic	the energy metabolism of bacteria that can, in the absence of light, use the oxidation of inorganic substances as a source of energy for cell biosynthesis and maintenance
Culturally Important Place	A Place, area of land/sea, in the landscape nominated by Traditional Owners

Culturally Important Site	A specific location, such as a rock waterhole etc., within the landscape. Such sites may or may not lie within a Culturally Important Place.
Dispersion	The spreading out of emissions from a localised source (e.g. industry stack, wildfire) over a wide area because of the effect of wind.
Eh-pH	A parameter which indicates the stability of mineral or chemical systems based on the activity of hydrogen ions (pH) and electrons (Eh). These are often compared using an Eh-pH diagram (Pourbaix diagram).
End Member	A pure chemical compound (/mineral) component entering into solid solution with other pure chemical compounds to form a series of minerals.
Endolithic lichens	Lichens (fungi that live together in a colony with photosynthetic algae) that grow in rocks
Environmental Quality Criteria (EQC)	Scientifically based limits of 'acceptable' change within an EQMF.
Environmental Quality Management Framework (EQMF)	A framework to guide the assessment and management of activities related to a particular environmental value.
Environmental Quality Objective (EQO)	A specific management goal for a designated part of the environment that signals the level of environmental quality needed to protect the environmental value of an EQMF.
Environmental Value (EV)	A beneficial use or an ecosystem health condition which requires protection from the effects of emissions or environmental harm.
Eukarya	Members of the domain Eukarya—called eukaryotes—have membrane-bound organelles (including a nucleus containing genetic material) and are represented by five kingdoms: Plantae, Protista, Animalia, Chromista, and Fungi.
Extracellular polymeric substances (EPS)	EPS consists of a variety of macromolecules that are secreted by bacteria in the environment to make biofilms.
Fermentation	The microbial enzymatic degradation of organic matter (e.g. carbohydrates) into short-chain fatty acids (notably acetic, propionic and butyric acids) that can lower the pH of the patina.
Gene transcription	The active expression of a gene that produces an RNA copy. This can be a structural component such as ribosomal RNA that make up the ribosomes. Functional gene products are messenger RNA, which is

	translated into proteins within the ribosomes. Transcripts are indicative that the cell was alive and possibly active at the time of sampling since RNA is much more insatiable and prone to degradation than DNA. DNA survives much longer in the environment.
Heterotrophs	Heterotrophic bacteria and fungi that derive energy from organic compounds.
Hypolithic	Microbial communities that colonise and grow below the rock surface where they are protected against UV damage and desiccation.
MAGs	Metagenome Assembled Genomes. This technique combines short environmental DNA sequences (metagenomes) to build genomes of key species present in the microbiome. Expressed genes (metatranscriptome) that are sequenced in parallel can be mapped against the assembled genomes to link these processes with the key species involved in patina formation and/or degradation.
Metabolic pathways	A metabolic pathway is a linked series of chemical reactions occurring within a cell. The reactants, products, and intermediates of an enzymatic reaction are known as metabolites, which are modified by a sequence of chemical reactions catalysed by enzymes. The genes involved in the individual enzymatic processes as part of the metabolic pathways and the active expression of these genes will be identified through respectively metagenomics and metatranscriptomics as defined below.
Metagenomics	The study of sequenced genetic material in environmental samples. Depending on sequence depth, this can reveal a holistic overview of the relative abundance of all microbial genes present. In our study this refers to microbial genes that encode for enzymes involved in processes leading to patina formation and/or degradation.
Metatranscriptomics	The study of microbial gene expression in environmental samples. Depending on sequence depth, this can reveal a holistic overview of the relative abundance of all actively expressed microbial genes present. In our study this refers to actively expressed microbial genes that encode for enzymes involved in processes leading to patina formation and/or degradation. Because of the short-lived survival of gene transcripts in the environment, their presence is indicative of an active role in these processes
Microbiome	An integrated community of micro-organisms (bacteria, archaea, unicellular eukaryotes and fungi) occupying a particular habitat.

Microbial metabolites	Breakdown products of larger complex organic compounds that the microbial communities use as energy sources and for growth.
Microcolonial fungi	Colonies of fungi growing on rock surfaces, notably in extreme environments (desert). They are highly resistant against desiccation and ultraviolet damage.
Mineral Assemblages	Presence and abundance of mineral species in a given spatial region (either across the rock surface or from the rock surface to the 'fresh' rock below the outer weathered rind.
Murujuga	Traditional name for Burrup Peninsula and surrounding islands of the Dampier Archipelago.
Murujuga Rock Art Monitoring Program (MRAMP)	Overall program of work to be conducted to2026. Includes Initial studies to inform the design of the ongoing monitoring framework, as well as the development of EQCs and the EQMF.
Ongoing monitoring	The ongoing monitoring refers to the longer-term monitoring program to be jointly run by MAC and DWER once this program of works is complete (expected to commence from 2026). Also referred to as 'long-term monitoring'.
Ordination	This is a group of statistical approaches to visualise how much microbial communities differ between samples by projecting the distances in a multidimensional space. The closer the dots in the graph, the more related they are and vice versa. This can also show if communities are significantly different between sample categories such as between rock types.
Organic geochemistry	The study of organic compounds in the environment, including in rocks, sediments, soils, petroleum, aquatic environments and the atmosphere. Organic geochemistry studies the origin of organic compounds, their transportation processes, and the alteration they undergo in the environment, over time scales ranging from the present day to hundreds of millions of years ago.
Petroglyph	Literally 'rock mark', the term describes any cultural marking into a rock surface. The marks can be produced by a range of techniques, including pecking, pounding, incising, scratching or abrading, or a combination of two or more techniques. Techniques such as scratching can be very shallow (<1 mm), while pecking can be from 1 mm to more than 100 mm deep. All petroglyphs at Murujuga are Culturally Important.
Patina	In the Murujuga context the texture and colour of the rock surface is referred to as a patina. This is a deliberately broad definition, which encompasses other characterisation such as rock varnish or desert

	varnish including any biota which may be present on the rock surface. The patina has been shown to form over a depletion zone, referred to as the crust, which generally has a lighter appearance than both the patina and the underlying rock. An engraving is formed by breaking through the naturally formed patina to expose the lighter crust beneath. There may be cases where the engraving has exposed the underlying rock, which may result in a darker engraved channel.
Photoautotrophs	Photoautotrophs are organisms that can make their own energy using light and carbon dioxide via the process of photosynthesis. Examples are cyanobacteria and green algae, known to colonise rock surfaces. Photoautotrophs are considered primary producers since their biomass can be consumed by heterotrophs (defined above) as a source of carbon and energy.
Photospectrometry	An instrumental technique for measuring the chromatic reflectance of a surface by scanning at frequencies that cover the visible spectrum and beyond. Photospectrometry provides both a single value colour designation and spectral data that can indicate non-chromatic change. The techniques has been applied to both the measurement of change and the characterisation of unknown minerals and compounds.
Photolysis	The process by which molecules are broken into small fragments by exposure to sunlight (typically UV radiation).
Primary producers	See definition of 'photoautotrophs'.
Secondary Organic Aerosol (SOA)	Solid organic molecules produced in the atmosphere by the reaction of smaller, gaseous organic molecules with oxygen (and often ozone and UV/OH radicals).
Stable isotopes	Atoms of the same element which contain a different number of neutrons in the nucleus. Most elements consist of more than one stable isotope, the ratios of which can be measured using specialised instrumentation. Stable isotope ratios in organic compounds are affected by the processes of formation and alteration, hence their measurement gives information on source and alteration history.
Weathered rind	The outer portion of the rock that is sufficiently close to the surface to have interacted with oxygen or other environmental conditions. This layer is significantly thicker than the layer forming the patina and has different colouration to the underlying fresh rock (core).

Weathering	In the Murujuga context the concepts of weathering are differentiated as natural weathering and accelerated (/anthropogenic) weathering, which MAC would prefer is referred to as degradation. However, these effects may be difficult to decouple. Natural Weathering: the alteration of a rock surface through natural agents such as the impacts of temperature cycles and interactions with water and aerosols/gases released by the surrounding terrestrial and marine environments. Weathering can be subtractive (erosion) or additive (mineralisation or accretion). Accelerated weathering: degradation because of anthropogenic activity and not be considered as natural weathering.

9.2 Acronyms

Acronym	Definition
AQ	Air Quality
ASD	Analytical Spectral Devices (spectrophotometer)
BYK	BYK-Gardner spectrophotometer
CIELAB	Commission Internationale de l'Éclairage (International Commission on Illumination) L*a*b colour space. (Also CIELab or CIE L*a*b).
CM	Conceptual Model
CoA	Commonwealth of Australia
CSIRO	Commonwealth Scientific and Industrial Research Organisation
DWER	Department of Water and Environmental Regulation (Western Australia)
EPMA	Electron Probe Micro-Analysis
EPS	Extracellular Polymeric Substances
EQC	Environmental Quality Criteria
EQMF	Environmental Quality Management Framework
EQO	Environmental Quality Objective
(OP)FTIR	(Open Path) Fourier Transform Infra Red
ITS	Internal Transcribed Spacer (genomic marker)
KM	Konica Minolta
LIDAR	Light Detection And Ranging
MAC	Murujuga Aboriginal Corporation
MAG	Metagenome Assembled Genome
MAX-DOAS	Multi Axis Differential Absorption and Spectroscopy
NOx	Oxides of Nitrogen ($NO + NO_2$)
$PM_{2.5, 10}$	Particulate Matter < 2.5 and <10 microns (µm) aerodynamic diameter, respectively

Acronym	Definition
SEM	Scanning Electron Microscope (/Microscopy)
SOA	Secondary Organic Aerosol
SOx	Oxides of Sulphur (SO_2 + SO_3)
TLS	Terrestrial Laser Scanner
WA	Western Australia
XRD	X-Ray Diffraction
(p)XRF	(portable) X-Ray Fluorescence

Appendices

Appendix I — Statistical study design

APPENDIX I
Study design methodology

Statistical Team

Abstract

This Appendix summarises the study design, focusing on its statistical validity and power. It reports on the selection of the monitoring sites and calculations of sample size.

Contents

Introduction

This is an Appendix to

> Murujuga Rock Art Monitoring Program:
> Monitoring Studies Data Collection and Analysis Plan

(referred to as the "Study Design document").
This Appendix summarises the study design, focusing on its statistical validity, efficiency and power. Statistical challenges are discussed, and appropriate strategies are developed. The Appendix also reports on the selection of the monitoring sites and calculations of sample size.
Should there be any discrepancies between this Appendix and the Study Design document, the Appendix prevails on matters of study design and methodology, while the Study Design document prevails on details of experimental procedure and operational matters.
Source code for the calculations presented in this Appendix is available to technical reviewers on request.

1 Overview of study

1.1 Study objectives

The ultimate objectives of the study are

1. to obtain a scientific *evidence base* about the processes that cause deterioration of the petroglyph surfaces;
2. to develop a *monitoring framework*, supported by the evidence base, consisting of practical measurement techniques and benchmark values that have actionable consequences ("traffic lights").

The first objective (evidence base) requires a very complex study design. In order to obtain an evidence base of high standard, — that is, one which can withstand a high level of scientific scrutiny and will be accepted by the international scientific community, — it will be necessary to undertake a thorough and wide-ranging investigation of the many natural and anthropogenic processes that are involved or could be involved in the degradation of the rock surface condition. This will be necessary in order to exclude potential alternative explanations and to establish unequivocal evidence. Great care is required in the design and execution of the study, and a wide range of advanced technical instrumentation and technique will need to be deployed.
The second objective (monitoring framework) requires that the evidence base clearly identifies a single process or a small number of processes which are chiefly responsible for degradation of rock surface condition, if it occurs. Indicators of these processes must then be identified, and practical methods for measuring these indicators must be developed. Ideally these indicators would have been amongst the observable data recorded during the monitoring study.

1.2 Study components

The proposed study consists of several components:

1. **rock colour and condition monitoring:** in order to detect changes in the colour and condition of rock surfaces, a number of individual rocks will be selected; these rocks will be repeatedly re-visited in the field during the five year study; and the researchers and indigenous rangers will carefully measure the spectral colour and surface texture of these rocks under controlled conditions.

2. **air quality monitoring:** in order to determine the likely exposure of rock surfaces to chemicals which might cause degradation of the appearance of the rock, a network of automatic monitoring stations, placed at carefully-chosen locations across the Murujuga region, will measure the presence and amount of different gases in the atmosphere, at different times throughout the five-year study.

3. **microbiome sampling in the field:** in order to understand what biological processes might be likely to cause the colour and appearance of rocks to degrade over time, researchers will scrape a small amount of material from the outer surface of selected rocks (rocks which are not culturally altered), and study the population of micro-organisms that are present.

4. **microbiome on prepared slabs:** in order to understand what biological processes might be likely to occur in the *engraved* parts of a *culturally-altered* rock surface, the researchers will collect pieces of un-altered rock from a disturbed site, cut them to make small slabs with a fresh surface, and place these prepared slabs in the field. In subsequent years these prepared slabs will be studied by the same techniques as for the Microbiome Sampling study.

5. **inorganic chemistry and mineralogy:** in order to understand what physical/chemical processes might be likely to cause the colour and appearance of rock surfaces to degrade over time, the researchers will obtain core samples of rock, and study their inorganic chemical composition and mineral structure, at different microscopic depths below the rock surface.

6. **organic chemistry:** in order to understand what biochemical processes might be likely to cause the colour and appearance of rock surfaces to degrade over time, the researchers will obtain core samples of rock, and study their organic chemical composition, including comparison between the surface and the deep interior of the rock.

7. **chamber experiments:** in order to predict how much degradation in colour and texture would occur if the rocks were exposed to higher concentrations of chemicals in the atmosphere, researchers will take core samples of rock to the laboratory in Perth, place the samples in a closed container, add progressively higher concentrations of chemicals into the air in the container, and observe changes over time, particularly changes in the microbiome.

8. **soil sampling:** where possible, a small jar of soil will be scooped from the ground close to a rock that has been physically sampled. This is part of the initial study. It will make it possible to assess whether the organic chemistry of the soil is a good predictor of the organic chemistry of the rock. It may also indicate the occurrence of chemical processes such as leaching.

1.3 Locations to be visited

1.3.1 Objectives

Each of the component studies requires the researchers to visit sites in the Murujuga region. Several of the component studies are directly linked, in the sense that we will need to *spatially cross-reference* at least some of the observations made in different component studies. For example, it must be possible for air quality to be monitored at locations close to some of the rocks whose colour will be measured.
There are several competing goals for the selection of sampling locations for each kind of observation:

- accurate extrapolation of air quality from the monitoring sites to other locations in the Murujuga region
- accurate measurement of the effects of different levels of exposure to air pollutants
- validity of extrapolating from a sample of rocks to the entire population of rocks
- ability to measure different sources of variability so that the uncertainty in the final results can be quantified

1.3.2 Proposed locations

To reconcile these competing requirements, the study design proposes two sets of sampling sites on the Murujuga region: the **Air Quality** Sites and the **Sample Survey** Sites.

Air Quality Sites are placed at strategic locations across the Murujuga region so that observations of air quality at these locations will be sufficient to predict air quality at any other location in the Murujuga region.

Sample Survey Sites are nominated "at random" according to principles of survey sampling (design-based sampling).

Each Site (of either kind) is a circle of radius 600 metres which is designed to contain an air quality monitoring station (for the air quality sites only), a rock for colour monitoring (study 1), a rock for microbiome sampling (study 3), a rock providing core samples (for studies 5, 6, 7), several nearby locations for soil sampling (study 8) where possible, and a location for a prepared slab (study 4).
In addition to these sites there is also an area of very disturbed rocks on the **Interconnector Pipeline Site** which could provide large quantities of rock material for study purposes. For example the Prepared Slabs (component study 4) could be made from these rocks. It is unclear whether all three main rock types are present on this site; dolerite appears to be absent.

1.3.3 Initial permission to access the proposed locations

Access to some sites may be denied, restricted or inappropriate, due to cultural safety concerns, inaccessibility, or other reasons.
A large scale map of the *proposed* site locations across the Murujuga region, together with higher-resolution satellite images of each proposed site location, will be presented to MAC for

assessment. The proposed site locations will also be submitted for cultural heritage approvals and other government approvals.
If access to any proposed site is denied in its entirety, we will propose an alternative site. For the Air Quality Sites, we would revisit the algorithm that was used to propose the air quality monitoring network, add new conditions that reflect the permissions obtained or denied, and compute a new network of proposed sites subject to these conditions. For the Sample Survey sites, if access to a site is forbidden entirely, we will seek permission to visit a nearby site (identified by attaching serial numbers to the sites and proposing the "next" site in the sequence).
Within a large site, access to particular areas or particular petroglyphs may be denied for cultural or other reasons. In the Air Quality Sites, no particular adjustment will be made for this. In the Sample Survey Sites, techniques from model-assisted survey inference will be used to adjust for the missing information [1, 12, 11].

1.4 Physical sampling

The initial part of the study involves physical sampling of rocks, conducted as follows, and summarised in Figure 1.

- At each Air Quality Site and each Sample Survey Site, up to three rocks will be chosen (ideally, one of each of the three main rock types if present).
- Each rock will be visited twice: first in the wet season, and then in the dry season.
- At the first visit (wet season), a small amount of material from the outer surface of the rock will be scratched off, using a small battery powered tool ("Dremel" or similar) on a small patch of surface. Three small patches will be scratched on each rock.

  The scratched material will be analysed using genetic methods to understand the population of microbes that are present ("microbiome").

- At the second visit (dry season),
  - The surface will be scratched again, on three small patches close to the previous patches;

    The microbiome of this sample will be analysed.
  - A small jar of soil will be removed from the ground close to the rock, where possible;
  - The *pH* (acidity) of the rock surface will be measured by wetting it with a small volume of distilled water, and touching a sensor to the rock surface;
  - Under supervision of MAC, core samples will be drilled from the rock. Two alternative procedures are:
    * Core samples are extracted from the rock on site, without moving the rock; *or*
    * A cultural ceremony is performed; the rock is removed from country and taken to the MAC precinct; cores are drilled at the MAC precinct; and the remaining rock is kept in the custody of MAC.
  - The core samples will be taken to Curtin University in Perth, where they will be divided into sub-samples for
    * studying organic chemistry

    * studying inorganic chemistry and mineralogy
    * conducting gas chamber experiments
  - Prepared slabs of rock (prepared from material at the pipeline site) will be placed near the centre of the site.

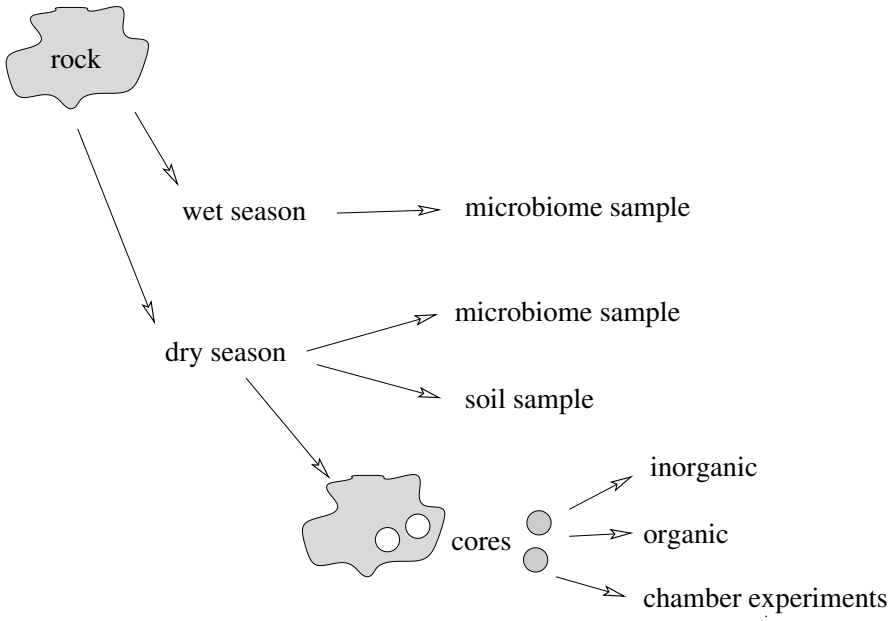


Figure 1: Schematic diagram of proposed visits for physical sampling of individual rocks.

1.5 Elaboration of the study design

The design is constructed as follows:

- First the locations for the air quality monitoring stations are selected so that they enable accurate prediction of the air quality at other locations in the Murujuga region.

  This is determined by generating many "weather scenarios" which are computer maps of the concentration of pollutants across the Murujuga region, under different weather conditions and industrial emission patterns. Then we consider different possible arrangements of the monitoring stations. For each possible arrangement, we assume that air quality is measured only at these locations and must be extrapolated to the entire Murujuga region using a prediction technique; we compare this extrapolated concentration map with the actual concentration map, and measure accuracy of extrapolation. The arrangement of monitoring stations with the highest accuracy is chosen. See Section 3.1 below for further details.

- At each site selected for an air quality monitoring station (study 2), within the 600-metre radius circle we select rocks for colour monitoring (study 1), microbiome sampling (study

3), organic and inorganic chemistry (studies 5–7), and locations for prepared slabs (study 4) and soil sampling (study 8) where possible.

- Another set of locations is selected using survey sampling principles (randomised sampling design using stratification and nested cluster sampling). These locations can be regarded as (statistically) representative of the entire Murujuga region. In consultation with MAC it will be determined which of these locations are permissible and safe to visit under appropriate conditions.

    Ideally we would like to perform colour and condition monitoring (study 1), microbiome sampling (study 3) and chemistry (studies 5–7) at each location, but for the locations where this is not possible, we will seek to obtain covariate (predictor variable) information such as the frequency of different rock types, the typical compass directions of the rock faces, etc.

At the Air Quality Sites, we collect spatially cross-referenced observations of air quality, rock colour, mineralogy/inorganic composition, microbiome, and prepared slab colour. The main methodological principle for these data is **regression** of colour against air quality, in the presence of other explanatory variables.
The Sample Survey Sites sample is a randomised design-based sample that is effectively post-stratified into safe and unsafe strata. (Strata could be culturally unsafe for the researchers, or physically unsafe to visit, or physically difficult to access, or could be disallowed by the field-work safety policy, etc.) We shall apply **survey sampling** principles to both strata: unbiased design-based estimation to the "safe" stratum and regression sample estimation to the "unsafe" stratum using model-assisted survey inference [11, 13].
It is necessary to record some details of the immediate environment of each sample rock. Such details include the spatial aspect of the rock surface (the compass direction it is facing, and the angle to the sky), and the presence of other rocks which shadow the surface or protect the surface from wind erosion. A suitable protocol for recording these details needs to be developed in consultation with MAC.

2 Approach to sample size determination

Sample size is determined by stipulating a minimum standard of statistical performance that must be achieved — for example, that the study must have an 80% probability of detecting a rate of change in rock colour of 0.2 colour units per year or greater, if that change is actually present. The minimum number of samples required to achieve this performance is then determined using statistical theory.
The calculations depend on the details of the study design and on the statistical analysis procedure (including the specific statistical tests to be used). They also depend on knowledge of the characteristics of the data.
Therefore, rigorous determination of sample size would require that the entire experimental protocol and analysis procedure had been specified in advance, and that there is already some information about the characteristics of the data. This is typical of research in well-established fields such as clinical trials.
The present study design is novel, complex, involves many exploratory steps whose outcome is unknown in advance, and requires the development of new statistical methodology. Con-

sequently, in the design phase where measurements have yet to be taken and concomitant information is imprecise, sample size determination will be approximate.
To determine the sample sizes, we shall adopt a precautionary approach in which each of the component studies is considered separately, and designed to achieve a simple and robust standard of minimum performance. This will ensure that each study is capable of providing adequate results and achieving its objectives, regardless of the outcome of the other studies.

3 Air quality monitoring locations

The network of Air Quality Monitoring Sites is designed so that the air quality at any site across the Murujuga region can be inferred accurately from air quality observations at the monitoring sites alone.

3.1 Air quality network design

The design of the air quality network is a problem of strategic optimisation rather than representative random sampling. The procedure used to design the network has been described in Section 3.7 of the Study Design document:

1. Collect and analyse meteorological data for the Murujuga region to establish the variability in weather conditions, and construct simulation procedures which replicate these conditions.

2. Identify the locations of emission sources in the Murujuga region and their typical emission rates.

3. Create a 3D topographic model of the Murujuga region

4. Run computational fluid dynamics (CFD) calculations using simulated meteorological conditions (step 1) using the existing emission sources (step 2).

5. Identify up to 100 potential sites for air quality monitoring in the Murujuga region. Proposed networks will be formed by choosing up to 20 of the 100 sites. Identify any constraints on the selection of sites.

6. Develop a fitness criterion which measures the performance of a proposed network when the air quality field is assumed to be known exactly. Criteria include kriging standard error and sample maximum error.

7. Select the optimal network by applying the fitness criterion to all possible networks, subject to the constraints.

Section 3.7 of the Study Design document reports that fitness reached its optimal level with a network of 18 monitoring stations. The selected Air Quality station sites are depicted in Figure 2.

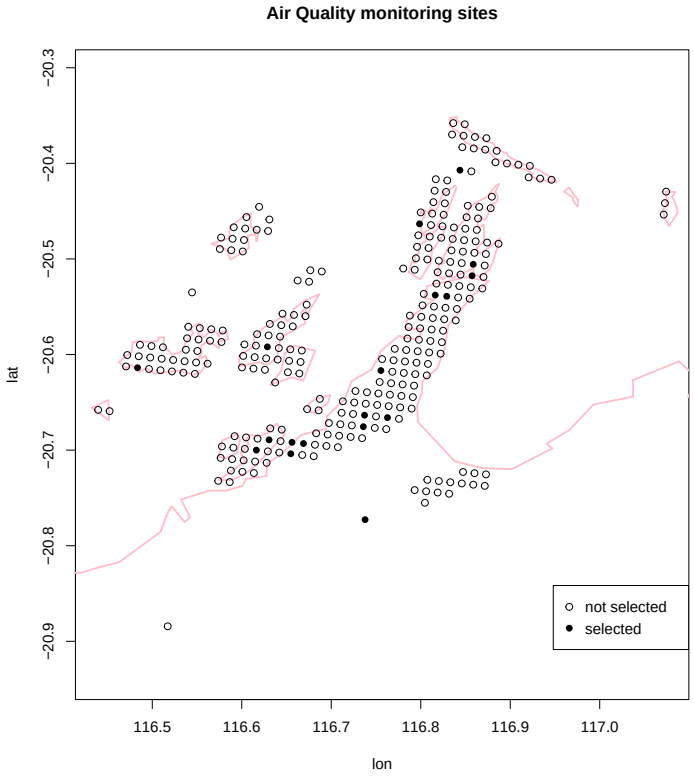


Figure 2: Selected Air Quality station sites.

4 Sample Survey locations

4.1 Methodology

Only a few dozen petroglyphs will be visited during the study, whereas it is believed that there are at least a million petroglyphs in the Murujuga region. The small sample fraction does not undermine the validity of the study, provided the selection of petroglyphs visited during the study is statistically "representative" of the entire population of petroglyphs. This can be ensured by following the principles of randomised design-based sampling [2, 14, 15].
The primary mechanism for sample selection of the Sample Survey Sites will be *nested cluster sampling*, using *systematic sampling* at each level:

1. The entire area of the Murujuga region is notionally divided into cells. (In this case the cells are the same as the 1.3 km square grid cells delineated when selecting the Air Quality network).

2. The cells are notionally arranged in some order. A random number x between 1 and 10 is selected. Starting from cell number x, we count off every 10th cell $(x, x + 10, x + 20, \ldots)$ and these cells are selected. This is a *systematic sample* with period 10, and can be regarded as a sample of one-tenth of the population.

3. Within each selected cell, locations of petroglyphs (for colour monitoring) and sample

rocks (for destructive sampling) are chosen according to a random sampling design (see Section 4.3).

This procedure ensures that the two basic criteria for design-based sampling inference are satisfied:

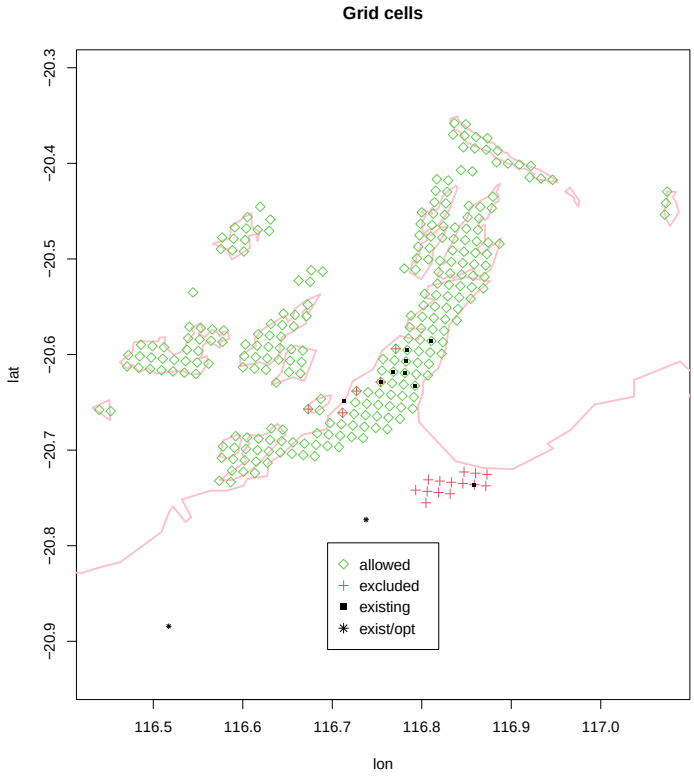
1. each rock or petroglyph which would have been permissible to sample, had a nonzero probability of being sampled; and
2. for each rock and petroglyph that *was* sampled, the sampling probability is known.

To increase statistical efficiency (and at the suggestion of a reviewer) this procedure will be modified by stratifying the population according to rock type. That is, the Murujuga region will initially be divided into a few regions ("strata") according to the predominant rock type that is shown on a geological survey map. The nested cluster sampling procedure will be applied separately to each stratum (each major rock type). We note that this strategy does not alter the fundamental validity of the sampling technique, and does not rely on the accuracy of the geological survey map classification, as the strategy is designed only to increase statistical efficiency.
It could happen that a grid cell (1.3 km square grid cell) selected by this randomised procedure could be culturally unsafe for the researchers, or physically unsafe to visit, or physically difficult to access, or could be disallowed by the fieldwork safety policy, etc. In such cases we would propose visiting the next cell in the sequence.

4.2 Selection of grid cells for sampling

Figure 3 depicts the 1.3 km square cells delineated across the Murujuga region. It shows cells which are available for sampling, those which are excluded for some reason, and those where there is an existing air quality monitoring station.

Figure 3: The Murujuga region divided into cells, classified as available or unavailable for sampling.



Figures 4 and 5 show cells where the main rock types, Granophyre, Gabbro and Dolerite are present according to an available geological survey. "Present" means that a simple point-counting estimate of area fraction indicated that at least 1/16 of the cell area was occupied by the rock type in question; or in the case of Dolerite, that a dolerite dyke was mapped within the cell.

A stratification of the sites was developed using the following definitions:

STRATUM	DEFINITION	NUMBER OF CELLS
D	Dolerite present	89
A	Gabbro present, Dolerite absent	45
R	Granophyre present, Gabbro and Dolerite absent	53
S	other locations with petroglyphs	9
N	none of the above (this stratum will not be sampled)	77

The strata are mapped in Figure 6.

The cells containing air quality monitoring sites were then excluded from consideration. Systematic random samples of period 11 were taken from each of the strata D, A, R, and a systematic random sample of period 9 from stratum S. This produced a sample of total size 18, containing 8 cells in stratum D, 4 in A, 5 in R and 1 in S. The cells selected to contain the Sample Survey Sites are shown in Figure 7.

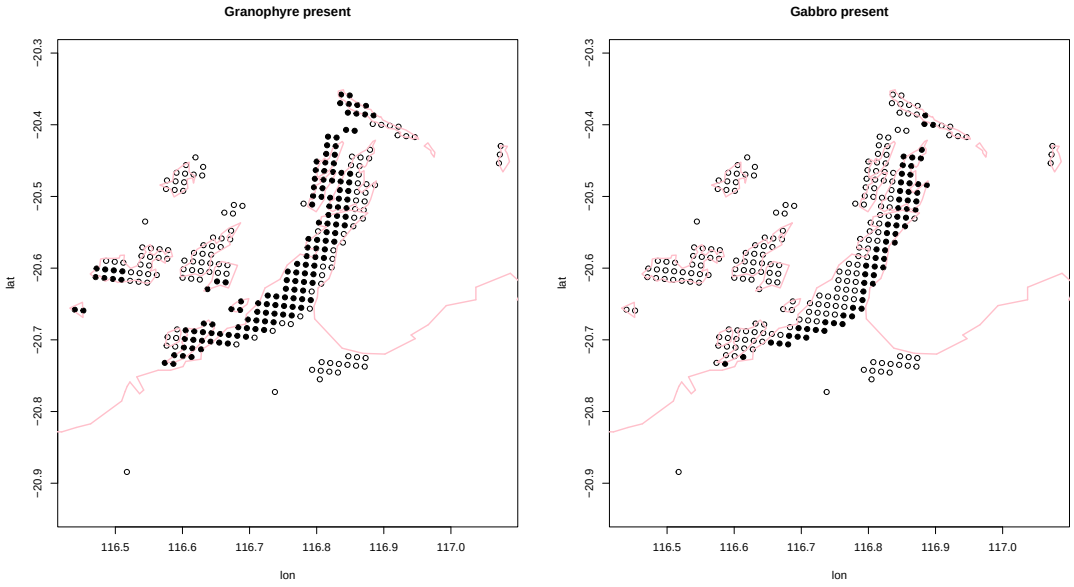


Figure 4: Sites where Granophyre and Gabbro are present (in the sense defined in the text).

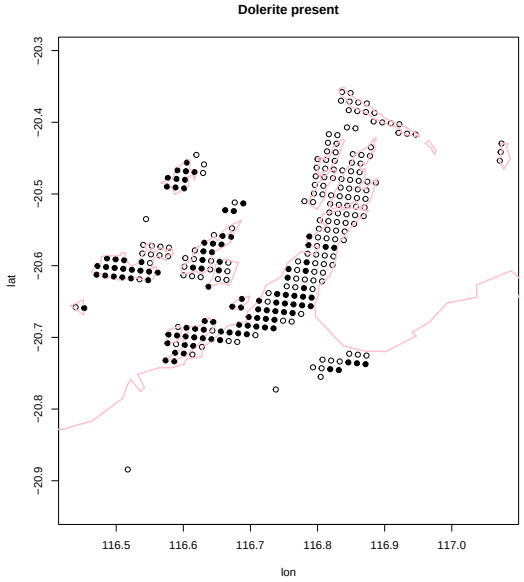


Figure 5: Sites where Dolerite is present (in the sense defined in the text).

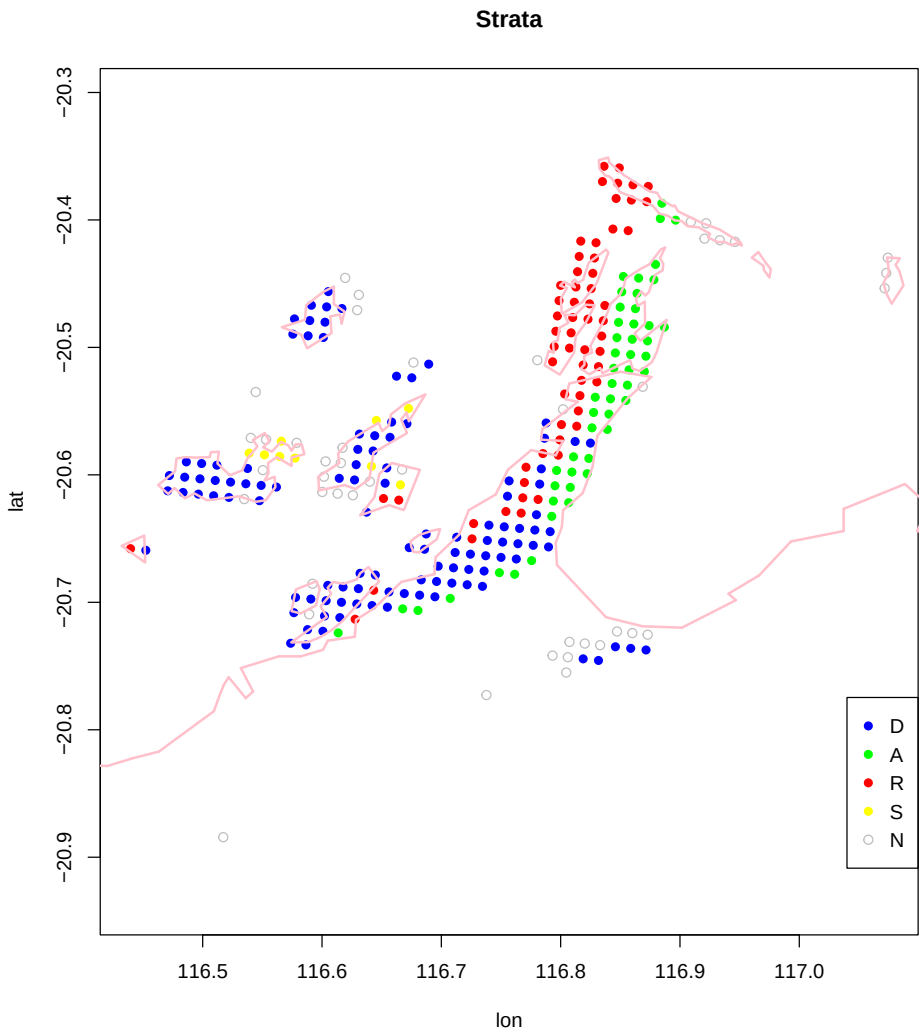


Figure 6: Stratification of the population of cells available for sampling.

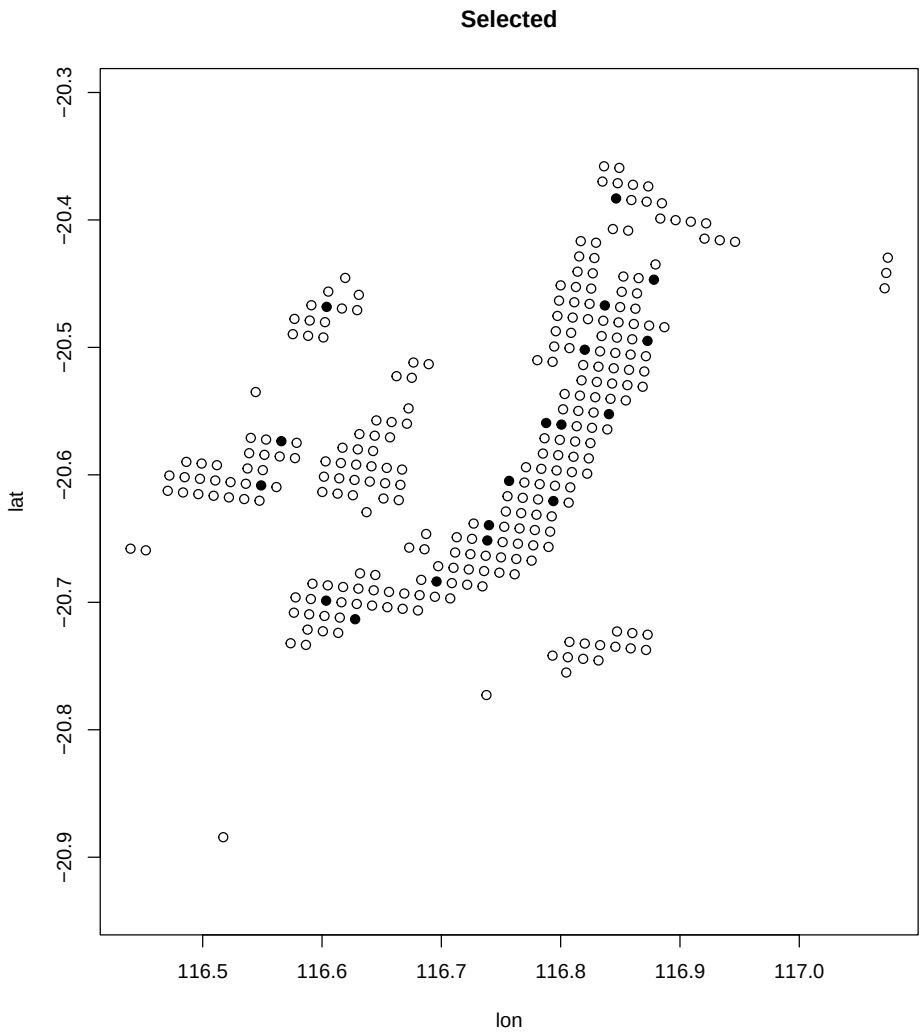


Figure 7: Cells selected to contain sample survey sites.

4.3 Fieldwork sampling protocol

This section describes the sampling protocol followed in the field, within each selected 1.3 km square grid cell.
A tentative protocol for sampling was developed before fieldwork began. However, in keeping with best practice, the plan allowed for the strong possibility that additional constraints on the sampling procedure would be discovered during the early phase of fieldwork. It was expected that these constraints could have been due to cultural factors, practical exigencies, legal considerations, spatial characteristics of the rock population, geological considerations and so on. It was also decided to collect detailed records of the fieldwork operations so that the sampling procedure could be carefully examined and so that ample covariate information would be available for later use.
The most important practical consideration identified during the early phase of fieldwork was that the cost (in time and resources) of visiting a single spatial location was much higher than expected. Reasons included physical inaccessibility; cultural safety concerns; the legal requirement to survey at least a 25-metre radius around each destructive sample rock to ensure that there are no petroglyphs within 25 metres; the difficulty of finding suitable sample rocks in some locations (due to geology or due to the high density of petroglyphs); the unexpectedly complex geology and the consequent need for more careful examination of the local geology (for example to ensure the rocks selected for destructive sampling originated in the selected locality rather than having fallen or slid down a slope from another locality); health and safety risks; and time constraints determined by boat access and cultural concerns.
For this reason, the fieldwork sampling design was altered so that, for each of the three main rock types, the team typically selected only one spatial location for a petroglyph and one spatial location for a destructively sampled rock. The sampling principle applied within each cell is uniform (i.e. equal-probability) random sampling of a fixed number of sample points.
Below is a summary of the fieldwork sampling protocol for each sampled cell.

1. obtain a satellite image of the sampled cell and superimpose any available data on the locations of registered petroglyphs.

2. supply this image to MAC rangers and elders, and in consultation with them, divide the image into broad regions which are either (A) 'unsafe' (culturally unsafe, physically unsafe for access, etc.), or (P) 'safe, but known to contain petroglyphs' (with a 25 metre margin), or (S) 'safe, and not known to contain any petroglyphs'.

3. generate a list of at least 1000 random spatial locations, independently and uniformly distributed in the selected cell. rejecting any locations which lie in the 'unsafe' region. Split this list into two lists of locations in the (P) and (S) regions.

4. visit the first location in the (S) list. If this is found to be inaccessible or unsafe, continue down the list until an acceptable location is found.

   (a) identify the rock nearest to this sample location that is suitable for destructive sampling. Suitability includes physically safe access, correct geological type, and non-anomalous geology (for example, not a rock which has rolled down a slope from another location).

   (b) survey a 25 metre radius of this candidate rock to ensure there are no petroglyphs.

i. if no petroglyphs are found, accept this candidate rock as the sampled rock.
ii. if petroglyphs are found, but subtend an angle of less than 180 degrees relative to the candidate rock, move away from the candidate rock in the opposite direction until a new candidate rock is identified. Restart step 4b above.
iii. if petroglyphs are found, and they subtend an angle of more than 180 degrees relative to the candidate rock, abandon this location, and restart step 4 above.

5. Visit the first location in the (P) list. If this is found to be inaccessible or unsafe, continue down the list until an acceptable and safe location is found. From this location, find the nearest petroglyph that is accessible and safe.

6. Each location selected for colour monitoring or destructive sampling, and their environment, are carefully documented (with photographs of the selected rocks, field labelling markers, GNSS/GPS location records, the GNSS/GPS and time tracks of the research team, photographs of the neighbouring individual rocks, the surrounding terrain and the geological context, geological survey notes, portable X-Ray Fluorescence assays and other records).

The initial fieldwork is now complete. The results of this initial phase determine the spatial locations that will be visited throughout the project. However, additional sampling is possible at these selected spatial locations, to increase the replication at the shortest spatial scale, if this is judged to be necessary.

5 Colour monitoring

5.1 Sketch of study

The colour monitoring study will record spectral colour (absorbance/reflectance values at each wavelength for a finely-spaced sequence of light wavelengths). In previous studies, such observations were immediately reduced to three-dimensional colour coordinates (CIE-LAB or similar). Our analysis will not do this; our full analysis will retain the full reflectance spectrum, which potentially contains much more information.
Nevertheless, the CIE-LAB coordinate system is an important reference, because it is designed to replicate the response of the human eye to visual stimuli. The CIE-LAB coordinates are scaled so that the ability of the human eye to distinguish different colours depends on the Euclidean distance between the colour points in the three-dimensional CIE-LAB space (conventionally denoted ΔE). Conventionally the threshold of ability to distinguish colours is set at $\Delta E = 2$.
The full analysis will therefore include both a high-dimensional analysis of the full reflectance spectra (using repeated-measures methods), and a three-dimensional analysis of the CIE-LAB colour values (using vector-valued regression models), and of their dependence on the explanatory variables in the study (time, place, exposure to pollutants, etc). All analyses will consider the direction of change as well as its magnitude.
The study design has chosen 18 Air Quality Sites and 18 Sample Survey Sites. In order to capture seasonal effects, each site will be visited at 4 different dates in the first calendar year, and thereafter on 2 different dates per year, in the peaks of the wet and dry seasons, respectively.
At each site, up to three rocks will be selected for colorimetry. On each rock surface, a number of "spots" will be selected as targets for the colorimeter sensor.

5.2 Sample size calculation

Various analyses of the previous study data give the following, very approximate, ranges of estimated effect size for the CIE coordinates a and b.

SOURCE	COORDINATE a	COORDINATE b
Site	± 3	± 3
Spot within site	± 2	± 3
Year-on-year change	0.25	0.15
Engraved vs unaltered	0.2	3

The estimated effect sizes are potentially biased, as discussed previously, but may be used for designing sample size.

Likewise we have the following approximate estimates of standard deviation

SOURCE	COORDINATE a	COORDINATE b
ASD	0.5	0.3
BYK	3	3

The CIE-LAB lightness coordinate L is not considered because estimates of L from previous studies are unreliable, and because the intention is to detect changes in hue, which is represented by the a and b coordinates.

5.2.1 Power for trend detection at a single spot

We first present sample size and power calculations for the detection of a trend in colour (hue) at a single spot.

The test of nonzero trend over time will be performed using the t test for nonzero slope in simple linear regression, or the F test of nonzero slope in parallel regressions allowing for a seasonal difference (wet vs dry season).

There are $m = 4$ years of observation and we perform simple linear regression of the response against calendar year. Assume that the trend is large enough to be detectable after m years according to the threshold of detectability $\Delta E > 2$. This implies that at least one of the CIE coordinates must have a slope of at least $2/(m\sqrt{3}) = 0.29$. Slope estimates of this size were indeed obtained from some prior data, as tabulated above. We assume the slope is $\beta = 0.2$ units per year. Using the table above we assume the error standard deviation (i.e. for replicates under identical conditions) is $\sigma = 0.5$ units.

Assume that each spot colour measurement is replicated n times per visit so that there are mn observations altogether. The test for a trend over time will be the t-test of nonzero slope in linear regression against calendar year. Assume a simple linear regression of mean response against calendar year, with slope β and error standard deviation σ. The least squares estimate of β has standard error $B = \sigma/\sqrt{S_{xx}}$ where

$$S_{xx} = S_{xx}(m,n) = \sum_{i=1}^{mn}(x_i - \bar{x})^2 = n\sum_{j=1}^{m}(j - (m+1)/2)^2, \tag{1}$$

where x_i is the calendar year in which observation i is made. For $m = 4$ years we have $S_{xx}(4,n) = 5n$. The one-side t-test of the null hypothesis $H_0 : \beta = 0$ against $H_1 : \beta > 0$ rejects H_0 if $|T| > t_{\text{crit}}$ where t_{crit} is the 95th percentile of the t distribution with $m - 2 = 2$ degrees of freedom (d.f.). The true distribution of T is the noncentral t distribution with 3 d.f. and noncentrality parameter ncp $= \beta/B$. From this we may calculate the power (probability

of correctly detecting a nonzero trend) for any number n of replicates. Using the approximate estimates above, we obtain the following power values.
First considering previous studies, for the BYK instrument (standard error $\sigma = 3$) assuming $m = 5$ years of observation and $n = 8$ replicate observations per year, the power to detect a slope of 0.2 colour units per year at a single site is 0.0949. The minimum number of replicates required to detect this change over 5 years with 80% power is 182. These calculations suggest that the previous studies did not have adequate power.
For the ASD instrument, CIELAB channel a (standard error $\sigma = 0.5$) assuming 4 years of observation and 8 replicates per year, the corresponding power is 0.529, and 18 replicates would be sufficient to achieve 80% power.
For the ASD instrument, CIELAB channel b ($\sigma = 0.3$) the same calculations give a power of 0.865, and 7 replicates would be sufficient.
In summary, 80% power could be achieved with as few as 7 replicates (for a trend of at least 0.2 units per year in the a channel) and 18 replicates (for a trend of at least 0.2 units per year in the b channel) assuming the ASD instrument is used over 4 years of observation.
Note that the difference in power between a 4-year and 5-year study is quite large, because of the high leverage of the observations at each end of the time sequence. In equation (1) if the duration m is increased from 4 years to 5 years, the value of S_{xx} is doubled from $5n$ to $10n$, and the noncentrality parameter ncp is increased by 40%. The power will be substantially increased, from 80% to more than 99%.

5.2.2 Power for trend detection using a scanning grid

An alternative observation technique proposed in Section 3.2.3 of the Study Design document is to mount the colorimeter sensor on a motorized stepping stage. Then colorimetry measurements would be performed on a grid of positions, either $7 \times 7 = 49$ or $9 \times 9 = 81$ positions centred on the target spot. The grid would be spaced so that it covers a rectangle approximately 1 centimetre across.
The use of a scanning grid would enable some statistical analysis that is impossible or difficult using other designs. In previous studies, the variability between replicate observations at a single spot included "spatial" and "non-spatial" contributions that could not be separated. The "spatial" contribution is due to spatial inhomogeneity of the rock surface itself, which has an effect because of errors in relocating the target for each replicate observation. The "non-spatial" contribution comprises all other sources of variability between replicates, such as intrinsic variability of the instrument, temperature-dependent effects, ambient light variation, and operator differences. If a scanning grid is used, the short-range spatial inhomogeneity of the rock surface colour can be observed and modelled, which enables the two variance contributions to be separated (assuming the scanning grid technique is performed at least twice).
In the proposed study design, on each rock surface selected for colorimetry, a rock surface patch approximately 1-3 cm across will be selected. In the Air Quality Sites, the surface patch will be selected for its apparent chromatic homogeneity, following standard practice (both in museum practice and in previous field studies of Murujuga). In the Sample Survey Sites, both the rocks themselves and the patches on the rocks will be selected using randomised design-based sampling procedures. This design maximises the power to detect colour changes at the arbitrarily-chosen Air Quality Sites, while also guaranteeing the validity of statistical inference about the overall population of rocks and of petroglyphs.

For sample size calculations, existing data does not provide information on the relative sizes of the "spatial" and "non-spatial" components of variation just described. The major concern is that the non-spatial variation may include random effects due to lurking variables such as temperature or operator differences. Using a precautionary approach we allocate effort in proportion to the variance components. The large variance component for spot–within–site (reported in the tables above) then implies that the number of spots on a given site should be commensurate with the number of sites.
The study design has chosen 18 Air Quality Sites and 18 Sample Survey Sites. Accordingly each site should be examined at about 10 fixed spots.

5.2.3 Conclusion for Colour Monitoring

The proposed schedule of visits is sufficient. Each selected rock should be examined at about 10 fixed spots. If individual spots are measured, the measurements should be replicated 20 times, at each spot, on each visit. If a scanning grid is used, the scan should be performed at least twice at each spot, on each visit.

6 Microbiome sampling

6.1 Sketch of study

The microbiome study is described in the Study Design document section 3.3.3. A simplified sketch is as follows:

- rocks will be selected according to the protocols described above, including at least one rock from each of the three major rock types where available.
- rocks will be visited at two times of year (wet and dry season) in years 1 and 3 of the study
- from each selected rock at each selected time, three samples will be taken, using a Dremel tool or similar, to extract powdered rock patina
- from the powdered patina, DNA and RNA will be extracted
- marker genes relevant to eukaryotes, prokaryotes and fungi will be extracted and amplified;
- genetic markers will be matched to established databases to identify the taxa involved;
- the relative frequencies of different taxa in the microbiome (from each sample) will be inferred;
- the active metabolic pathways represented in the transcriptome (from each sample) will be inferred;
- correlations will be estimated between the presence/abundance of taxa and presence/abundance of various minerals (and other explanatory variables).

The corresponding, notional sample size is $n \times 3 \times 2 \times 2 \times 3 = 36n$ where n is the number of sites visited. In practice it may be more accurate to assume that dolerite is not found in half the sites, so that the sample size is $n \times 2.5 \times 2 \times 2 \times 3 = 30n$.
There are also several validation experiments and exploratory experiments that we will not consider here.
There is some concern that a cultural smoking ceremony might affect the microbiome in the sample. This requires further investigation, once the procedure has been clarified.

6.2 Sample size calculations

Proportions of different taxa

The proportions of different taxa present in the microbiome are estimated in each powdered rock sample using a complex pipeline of laboratory techniques and data analysis (described in the Study Design document). The key question is how many times this entire procedure should be replicated to ensure adequate statistical performance.
Following common practice, it is proposed to take "triplicate" samples (scraping three adjacent patches of the same rock surface). The estimated proportions of the most abundant taxa are typically observed to vary by about ± 0.1 from one replicate to another. This suggests that an individual estimate has a standard error of about $0.1/\sqrt{2} = 0.07$. (If we interpret "abundant" to mean that the true proportion of the taxon is 20% or greater, then the "effective sample size" of a single estimate is at least $0.2(1-0.2)/0.07^2 = 32.7$, that is, the reported accuracy is equivalent to that obtained from a binomial experiment with 32 trials.)
The study should be capable of detecting changes in the proportion of a particular taxon. Preliminary advice was that a change of more than 10% would be scientifically informative, so we fix this as the minimum discrepancy that must be detectable, for abundant taxa.
The usual statistical theory for proportions based on the binomial distribution is not applicable. One approach to microbiome data uses the Dirichlet-multinomial distribution to model both overdispersion and multivariate dependence between relative abundances [8, 9]. Power calculations based on this approach require estimates of the true relative abundances, which are not available for this study.
Instead we treat the individual estimated proportions as continuous, approximately normally distributed random variables, with standard deviation 0.07. (It may be appropriate to apply Fisher's variance stabilising transformation $\varphi(x) = \arcsin\sqrt{x}$ before modelling).
The main experiment described above is a full factorial design with n sample sites, 3 rock types, 2 study years, 2 seasons and k replicates, where commonly $k = 3$. The main design objective is to be able to detect differences between the microbiomes at different sites which are attributable to differences in environmental exposure after allowing for other factors. The simplest model is a linear model with additive effects for each factor. Let γ_i be the effect for sample site i ($i = 1, \ldots, n$) with the constraint $\sum_{i=1}^{n} \gamma_i = 0$. The null hypothesis is $H_0 : \gamma_i = 0$ for all i. The interesting alternative hypotheses are those in which γ_i is related to an explanatory variable, as discussed below.
In the ANOVA F-test of a sample site effect, the test statistic is the ratio of the sample-site mean square to the residual mean square, and the test refers this statistic to the F distribution with d_1, d_2 degrees of freedom, where $d_1 = n - 1$ and $d_2 = (n-1)(3-1)(2-1)(2-1)(k-1) = 2(n-1)(k-1) = 4(n-1)$.

Under any simple alternative hypothesis (i.e. where the values of γ_i are specified), the test statistic has a noncentral F distribution with d_1, d_2 degrees of freedom and noncentrality parameter

$$\lambda = \frac{12k}{\sigma^2} \sum_{i=1}^{n} \gamma_i^2 \tag{2}$$

where $\sigma = 0.07$ is the error variance. We need to consider specific, simple, alternative hypotheses and calculate the corresponding value of the noncentrality parameter λ.

1. **Uniform spatial gradient**: Consider a scenario where the proportion of a particular taxon at each site decreases linearly across the spatial domain so that the values of γ_i can be rearranged as a sequence of equally-spaced values between $-A/2$ and $+A/2$. Thus A is the *most extreme* discrepancy between the sample sites, equal to the minimum discrepancy that we require the study to be able to detect. Then $\sum_i \gamma_i^2 \approx (n/12)A^2$ so that the noncentrality parameter (2) is approximately

$$\lambda \approx 12k \frac{n}{12} \frac{A^2}{\sigma^2} = knA^2/\sigma^2.$$

Taking $\sigma^2 = 0.1^2/2$ as above gives the power curves shown in Figure 8

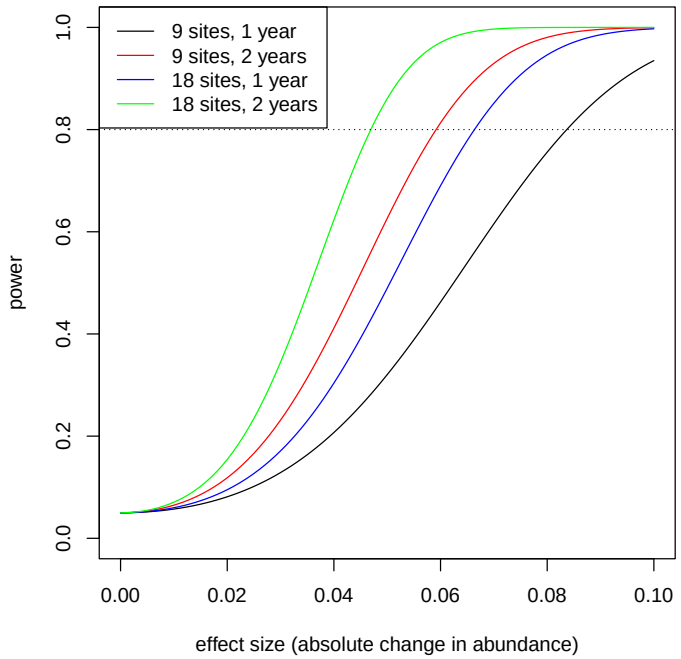


Figure 8: Power curves for detecting a given effect size.

A difference in abundance of 9% can be detected with at least 80% power using 9 sites, visited only once, and can be detected with 99% power using 18 sites visited only once or 9 sites visited twice. A difference in abundance of 5% can be detected with at least 80% power using 18 sites visited twice.

2. **Point source model:** a common model for environmental pollution is a point source (or several point sources) of emissions.The pollutant concentration, or its associated increased risk, is assumed to drop sharply as a function of distance from the nearest source [4]. If sample sites were approximately uniformly located in space, the corresponding values of γ_i would include only a few large values. For the model estimated in [4], for example, elevated risk was predicted to occur only within a radius of 2 km of the source, representing only 1 percent of the survey area.

   Simplifying further, assume that an effect of size $A = 0.1$ or greater (that is, a change of at least 10 percentage points in the relative abundance of a specified taxon) is present at only m of the possible study locations (namely those with the highest exposure) while all other locations show negligible effect. Then $\sum_i \gamma^2 = m(1 - m/n)A^2$.

Figure 9 shows the power curves assuming that 10% of sites have elevated values.

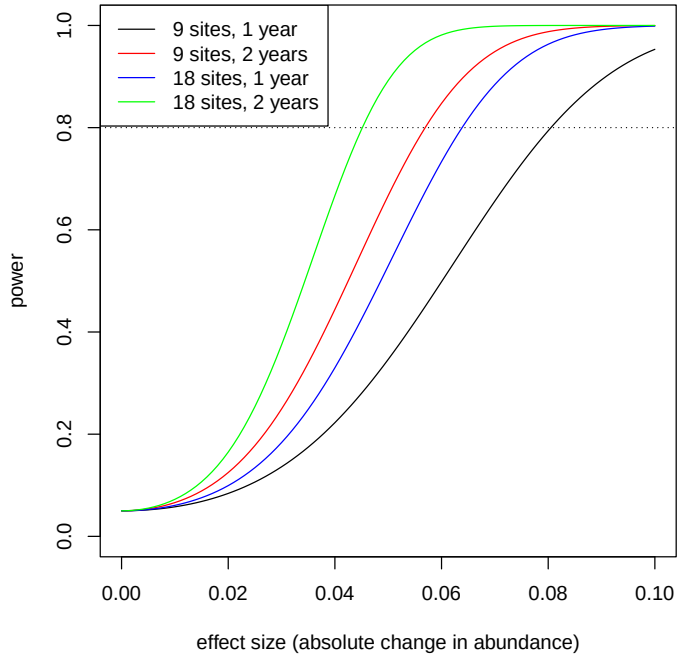


Figure 9: Power curves for detecting a given difference in abundance, assuming 10% of sites have elevated values.

Relative gene expression levels

For relative gene expression levels we shall use a similar approach to that given above.

Correlations

Assume n bivariate normal observations with population correlation ρ. Let R be the sample Pearson correlation. The variance-stabilising transformation is [5]

$$Z = z(R) = \frac{1}{2}\log\frac{1+R}{1-R}$$

with inverse

$$R = z^{-1}(Z) = \frac{e^{2Z}-1}{e^{2Z}+1}.$$

The stabilised variable Z is approximately Normal with variance $1/(n-3)$. The usual $100(1-\alpha)\%$ confidence interval for $z(\rho)$ is $Z \pm z_{1-\alpha/2}/\sqrt{n-3}$ where $z_{1-\alpha/2}$ is the upper $\alpha/2$ quantile of the standard normal distribution. Since $dz/dr = 1/(1-r^2)$, the back-transformed confidence interval for ρ has width approximately equal to $2(1-\rho^2)z_{1-\alpha/2}/\sqrt{n-3}$. If we specify that the confidence interval should have width at most w, then

$$w \geq 2(1-\rho^2)z_{1-\alpha/2}/\sqrt{n-3}$$

implying

$$n \geq 3 + \left(\frac{2(1-\rho^2)z_{1-\alpha/2}}{w}\right)^2.$$

Figure 10 shows the sample size required to achieve confidence intervals of width 10%.

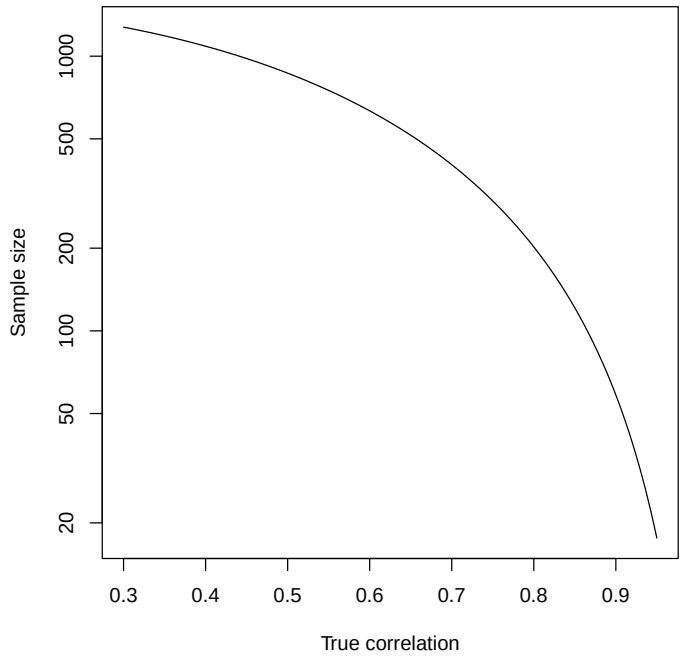


Figure 10: Sample size required to achieve confidence intervals of width 10%.

The notional sample size calculated in section 6.1 is $30n$ where n is the number of sites visited. The study envisages $n = 18 + 18 = 36$ so that $30n = 1080$. The design achieves confidence

intervals of expected width less than 0.1 whenever the true correlation exceeds 0.5. If the two types of sites are analysed separately, then the notional sample size is $30 \times 18 = 540$, and confidence intervals of width 0.1 are achieved whenever the true correlation exceeds 0.65.

7 Inorganic geochemistry

The inorganic geochemistry study is described in detail in Section 3.3.2 of the Study Design document. Its main goals are to

- gain an understanding of the minerals present in the rocks;
- gain an understanding of the inorganic chemical processes occurring;
- establish large-scale controls on these processes (such as proximity to the ocean, spatial aspect, air quality).

The detailed processes are unclear; multiple microscales are involved; a key goal is to integrate information from different length scales.
Physical rock samples for the inorganic chemistry studies will be taken on only one occasion, whereas the other component studies will make repeated visits to the Murujuga region (see Section 1.4 above). Consequently, of all the component studies, it is the inorganic chemistry studies which have the greatest influence on the sample size considerations.

7.1 Sketch of study

At each of the sampling sites, a number of rock samples will be taken.
From each rock, samples of fresh rock, weathered rind and patina will be extracted. Each sample will be split into a polished thin section and a solid billet. The solid billet will be analysed to determine the composition of the weathered rind and patina (using X-ray diffraction and Raman micro-analysis), while mineralogy and microanalysis are applied to the polished thin section (using optical microscopy, electron probe, secondary ion mass spectroscopy, transmission electron microscopy, and atom probe tomography).
Objectives for the inorganic geochemistry are listed in Section 3.3.2 *x* of the Study Design document. Three main targets are

- accurate estimation of mineral composition (the relative proportions of different minerals)
- accurate estimation of the thickness of the patina
- ability to detect differences in mineral composition and patina thickness depending on an explanatory variable such as air quality.

Weathering is faster at damage sites on the rock surface (e.g. where the rock has split). Weathering includes physical processes (such as sandblasting) and chemical processes (principally where water interacts with mineral grains to form clay minerals).

7.2 Sample size calculation for mineralogy

As the main performance criterion we nominate the power of the significance test of a difference in the mineralogy (relative composition) as a function of an explanatory variable. The theory described in Section 6.2 above can be applied with some modification.
Different rock types have different mineralogy, so that it is essential to have each of the three main rock types (gabbro, granophyre and dolerite) represented in the sample overall. There will be sites where dolerite is absent.
The simplest design requires a fixed number of replicates of each rock type. That is, at each site, for each of the three main rock types which is present, a fixed number m of rocks of that type would be chosen. The number of sample rocks at each site would be either $3m$ or $2m$. Then we would require $m \geq 2$ to ensure that it is possible to estimate variability within each rock type. Then there would be either 4 or 6 sample rocks at each site. This simple design could be modified by permitting only a single replicate ($m = 1$ implying 2 or 3 rocks) at some of the sites.
A rough estimate of variability (Section 3.3.2 *iii* of the Study Design document) is that the relative proportion of an abundant mineral varies by $\pm 10\%$ relative, at short scales in the same rock, and by less than 10% relative between rocks. Conservatively conflating these two sources of variation gives a standard error of 14% relative. For change detection it can be assumed that the true proportion is at most 50% so that the standard error is at most 7% absolute.

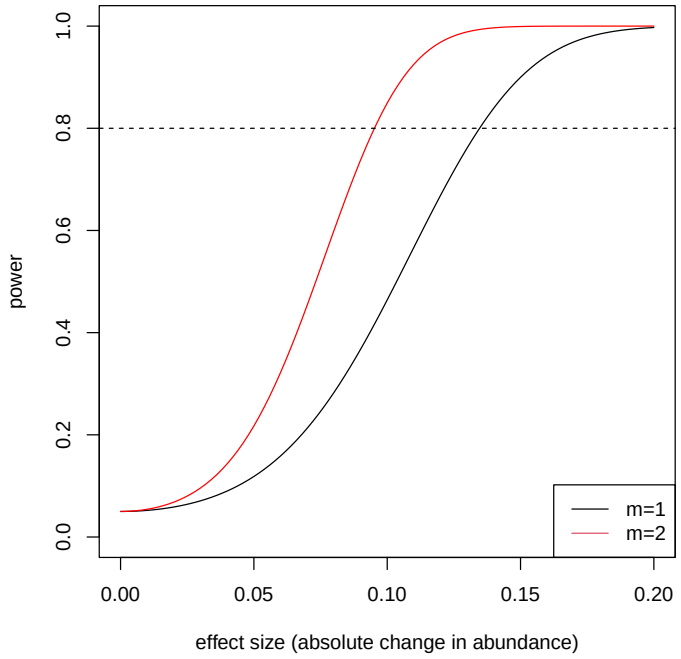


Figure 11: Power curves for detecting a change in abundance.

Figure 11 shows the power to detect a change in composition, assuming only two rock types for simplicity, with no replication ($m = 1$) or with duplication ($m = 2$). Without duplication, 80% power is achieved only with an effect size of 14% absolute. With duplication, 80% power

is achieved with an effect size of 10% absolute.

7.3 Patina depth

A rough estimate of variability in the patina is that the depth varies by $\pm 10\%$ (relative) at short scales over a rock surface where it is present. Patina is occasionally absent from the rock surface over patches of varying size.
Ignoring patina absences for simplicity, we could model the patina depth as a log-Gaussian spatial random field [3] across the surface of an individual rock. The natural logarithm of patina depth is then a Gaussian random field with pointwise standard deviation approximately equal to 0.1 (dimensionless units) across an individual rock surface.
Patina can differ markedly between different individual rocks. The mean of the Gaussian field (logarithm of patina depth) depends on the individual rock and includes random effects for the individual rock, systematic site effects, and other terms.
The power to detect a trend in patina depth as a function of an explanatory variable can be calculated by adapting the theory presented in Section 6.2 above.
Figure 12 shows the power against the alternative where the effect of the explanatory variable is a uniform gradient across the 36 sites.

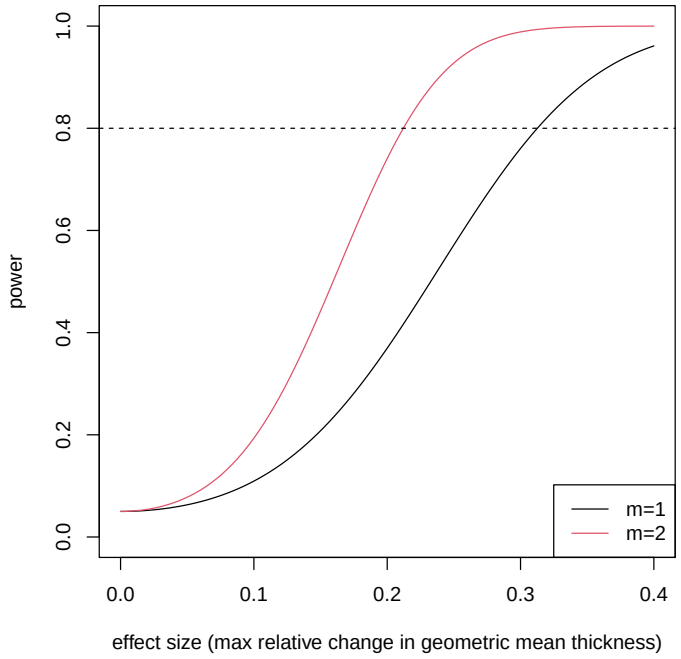


Figure 12: Power to detect a trend in patina depth, as a function of depth gradient.

An effect size of 0.1 (for example) shown on the horizontal axis indicates that the geometric mean depth of the patina is 5% (relatively) higher when the explanatory variable is at one extreme, and 5% lower when the explanatory variable is at the other extreme. Power is calculated assuming no replication ($m = 1$) or with duplication ($m = 2$). It indicates that duplicate

sampling ($m = 2$) is required to achieve 80% power of detecting an effect size of 0.2, while unreplicated sampling ($m = 1$) would suffice to detect an effect size of one-third.

7.4 Acidity (*pH*) measurement

Acidity of the rock surface has been identified as a potentially important explanatory variable by the technical reviewers and by [7].
pH will be monitored on rock art sites (up to $n = 45$), Study rocks (up to $n = 45 \times 3$) and prepared rock slabs ($n = 25 \times 3$). Monitoring will be conducted 4 times in the first year (possibly on a smaller subset), followed by annually, with additional *ad hoc* monitoring after weather events.
The sample size required for ongoing monitoring will be determined once these initial observations have been made, or using the data of [7] if these are made available.

7.5 Plutonium isotope ratio

In the initial study there will also be a pilot experiment in which the relative abundances of Plutonium radioisotopes will be assayed from the solid rock billet samples. These preliminary samples will be used to determine feasibility, detection limits, and sample sizes required if it is decided to include this measurement in the full experimental design.

8 Organic geochemistry

8.1 Outline of study

The Organic Geochemistry study is detailed in Section 3.3.4 of the Study Design document. Its main goal is to identify organic compounds present on rock surfaces with the potential to cause accelerated degradation of petroglyphs.
Organics will be extracted from powdered rock, separated into sub-fractions, and subjected to gas chromatography – mass spectrometry, isotope ratio mass spectrometry, pyrolysis techniques, and time of flight – secondary ion mass spectrometry.
Paired samples of soil and rock will also be collected where possible. Soil can be assayed very rapidly for percentage of Total Organic Carbon TOC and Total Inorganic Carbon TIC. The study will investigate the correlation between TOC in soil and rocks.

8.2 Study design aspects

For some purposes it is merely necessary to establish the presence of an organic compound — that is, any "detectable" presence is adequate — in order to form causal hypotheses about the processes involved.
The amount of organics in rock samples from Murujuga is expected to be low, so that the main concern is to ensure sufficient sample mass in each sample to permit analysis.
Major issues for such investigations are the *detection limits* and the *range of linearity* for each instrument. These must be investigated and validated for each technique and for each organic compound under investigation.

Consequently this study requires pilot (trial) experiments to establish validity and determine effect sizes, sources of variability, and controlling variables. These trials will determine the parameters for the full experiments.

9 Chamber experiments

The chamber experiments seek to establish a "dose-response" relationship between (elevated) air pollutant concentrations and (accelerated) degradation of rock surface condition. They are described briefly in Section 3.4 of the Study Design document.
The "response" in the chamber experiments is a change in the microbiome or in the organic chemistry present on the rock samples. Possible effect sizes are unknown.
There will need to be several pilot experiments to establish detectability limits, ranges of variables, and the likely effect sizes. These pilot experiments in turn depend on information from the Organic Geochemistry study (identifying organic compounds of interest) and the Microbiome study (identifying taxa of interest).
The definitive experiments should include a factorial experiment with specific molecular gases at multiple levels of concentration, and a regression experiment with a typical environmental composite pollutant (e.g. heavy fuel oil exhaust, natural gas, diesel exhaust, combusted biomass) at different concentrations.
Sample size determination for these definitive experiments depends on the results of the pilot experiments.

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Appendix II – Site selection

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Murujuga Rock Art Monitoring Program

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Table of Contents

1 Introduction

This document presents a summary of the process for selecting sites to create an air-quality monitoring network to support the Murujuga Rock Art Monitoring Project. The key inputs to the process were modelled air-quality data for Murujuga, and rock art site data from the “Register of Aboriginal Sites or Heritage Places” curated by the Western Australian Department of Planning, Lands and Heritage.

Assessment of potential air-quality monitoring networks was made by comparing the actual exposure to an exposure reconstructed from ‘virtual measurements’ on the candidate network, with dispersion modelling data acting as a proxy for field data. A Genetic Algorithm was then developed and applied to find the optimal air-quality network for a given number of monitoring stations. This allowed the dependence on the number of monitoring stations to be established. The assumptions and methodology for the optimisation process and the final selection are also presented.

The objective of the air-quality monitoring network is to provide an accurate and reliable record of any exposure of the Murujuga rock art to natural or anthropogenic airborne pollutants. The location of air-quality monitoring stations is important as it seeks to establish the level of exposure generalised across all rock art on the archipelago(/peninsula) while also providing accurate measurement of the exposure of individual rock art sites selected as part of the study. Further constraints are imposed on the selection due to operational limitations (such as accessibility), and by recognising that the final sites that are selected must be acceptable to the traditional owners of Murujuga. Additionally, to avoid re-siting air-quality monitor stations after deployment it is desirable to select the most appropriate locations at the start of the study. For this reason, advanced optimisation techniques were employed to determine the locations of the air quality monitors, based on dispersion modelling recently conducted by Ramboll for the Department of Water and Environmental Regulation (DWER). The sections below give a brief overview of the optimisation methodology and the results of the optimisations.

2 Overview of the site-selection methodology

The dispersion modelling results provide air quality data across the whole Burrup region, including Murujuga. By considering this to be a reference dataset for air quality, it is possible to assess the effectiveness of a proposed air quality monitoring network by determining how accurately the network is able to recreate the original dataset from discrete measurements. As the current project is concerned with the impact on the rock art at Murujuga, the optimisation is weighted to place more importance on the prediction accuracy at rock art sites.

The method is as follows:

1. Make (virtual) measurements of air quality at rock art sites by sampling the dispersion modelling data at each site.
2. Select 'X' locations as air quality monitoring stations.
3. Make (virtual) measurements of air quality at the 'X' chosen monitoring stations by sampling from the dispersion modelling data.
4. By interpolating the measurements from the 'X' chosen sites (i.e., through kriging) determine the predicted air quality at the rock art sites.
5. For each rock art site, calculate an exposure 'error' – i.e., the difference between (4) and (1).
6. Combine the errors to give an overall error for the air quality monitoring network.

By repeating the steps 2 to 5 for many candidate networks it is possible to optimise the location of the air quality monitors such that the error in step (6) is reduced to a minimum. Furthermore, by changing 'X' and repeating the optimisation the relationship between the error and the number of air-quality monitoring sites can be determined.

3 Background data

Optimisation of the air-quality monitoring network relies on access to a spatially complete dataset to which the air-quality predictions can be compared. In reality, such datasets rarely exist and detailed simulation data must be used instead. For the Murujuga Rock Art Monitoring Project, results from dispersion modelling were provided by DWER and served as the proxy for field data. The simulations were undertaken by Ramboll, and the information provided to the project included input source files (weather and emissions), and hourly result files. To assess the impact on the rock art this data was analysed in combination with the known locations of rock art at Murujuga.

3.1 Modelling Scenarios

The dispersion modelling considers two model years (2014, 2030) under different emissions scenarios. The aim of the modelling was to allow the relative contributions from natural, domestic and industrial sources to be characterised. The air-quality component of the Murujuga Rock Art Monitoring project is planned to commence following approval of the Monitoring Studies Data Collection and Analysis Plan and to continue beyond completion of the project term in December 2025. The current makeup of emission sources at Murujuga is closer to the 2030 scenario in the dispersion modelling, as since 2014 a large proportion of the industrial development on the Peninsula has been undertaken, and many of the emission reduction schemes identified in the Ramboll report have already been implemented in full or in part. For this reason, the optimisation was performed on results from the 2030 emissions scenario.

3.2 Specie aggregation

As part of the dispersion modelling, relatively broad chemical classifications of emission sources are allocated to individual chemical species. For optimisation, this process was reversed by combining the chemical data back into a broader classification of emissions (NOx, SOx for example).

3.3 Data resolution

The dispersion modelling provided by DWER was undertaken on two grid sizes, 4 km and 1.333 km. Site selection was made using the results from the 1.333 km grid, and the original grid cells from the Ramboll simulation were used as the basis for site selection.

3.4 Temporal resolution

The dispersion modelling resolved hourly intervals across the modelled year. Performing the optimisation on this quantity of data is not feasible, so the hourly data was summed to match the collection frequency of the passive air-quality monitors (which are to be collected approximately monthly). Taking the collection interval as 4-weeks resulted in 13 datasets across the modelled year. A test optimisation made use of an annual summation of the data, but it was determined that that the periodic data

better captured variations in the pollutant exposure across the year, at the expense of greater computational effort during the analysis.

3.5 Rock art location data

The Department of Planning, Lands and Heritage (DPLH) has provided a copy of the "Register of Aboriginal Sites or Heritage Places", which contains the locations of rock art sites at Murujuga. This dataset consists of spatial data (as polygons) outlining a site or group of sites, and other registered locations. This data was filtered to include only those sites that were noted as artwork, and the centroids of these polygons were then used as the locations of rock art sites. A rock art site density was calculated for each modelled grid cell based on the number of these locations in the cell. The DPLH makes available two datasets from the register, one is publicly available with less precise polygons, while the other is restricted and contains more detail and sites. The analysis used the more detailed restricted database, but it is unlikely that results would be significantly different using the public database. Appendix B compares the two datasets.

4 Assumptions and Constraints

The key assumptions and constraints underlying the optimisation were as follows:

- all air quality monitoring stations measure the same species and are co-located (ie. all species are measured at the same locations) and there is a single air quality monitoring network - as opposed to separate networks to measure each of the individual species (nominally NO_X, SO_X, NH_3, and particulates).
- The key species considered in the optimisation were NO_X and SO_X. The Ramboll data suggests NH_3 is concentrated around the Yara Pilbara plants and may bias the optimisation towards this region. In addition, it was deemed likely that the existing monitors near to Yara Pilbara's facility would support the new monitoring program adequately.
- Air quality data from existing air quality monitors operated by Yara Pilbara and Woodside was assumed to be available to the Murujuga Rock Art Monitoring Project over the project's duration (either directly or for co-location). Hence, virtual air quality measurements from these monitors were included in the interpolation but excluded from the list of potential new air quality monitoring sites.
- Air quality monitors were to be restricted to the region of study on the Murujuga peninsula or islands, and could only be sited on land.
- There were some potential air-quality monitoring sites distant from the Peninsula which were optionally included in the optimisation (Figure 2 shows the actual locations considered as potential sites.)
- The weights on the error used to determine the fitness of the air quality were as follows:
  - the normalised error in the NO_X and SO_X predictions were weighted equally
  - the error in each grid cell was weighted by the number of registered rock art sites in the cell.

5 Source Data, Analysis and Optimisation

5.1 Pollutant data

The key pollutant species considered for the optimisation were NO_X and SO_X. Figure 1 shows the annual aggregate pollutant levels for each species, on the 1.3 km model grid used by Ramboll.

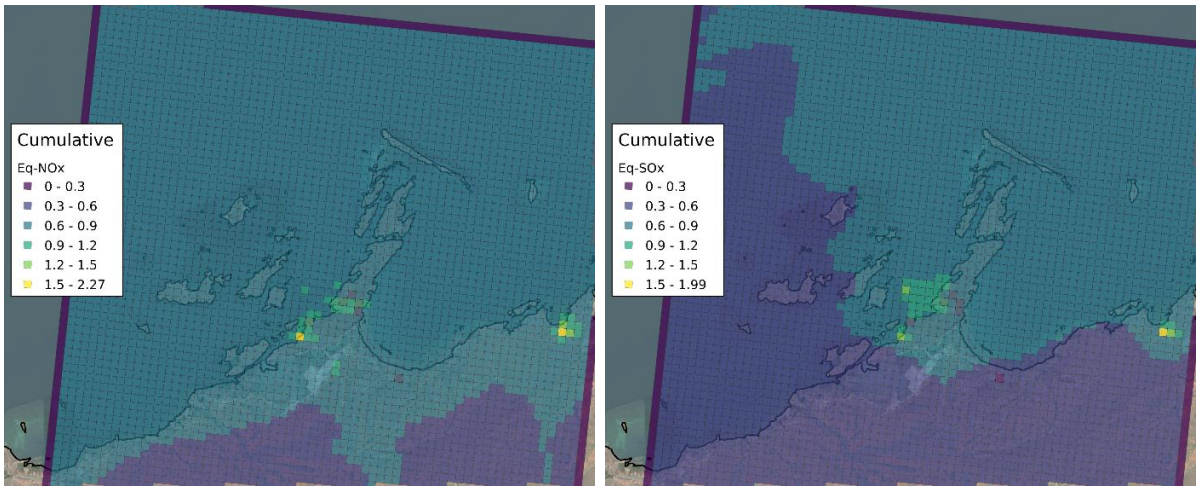

Figure 1: Cumulative annual NOx (left) and SOx (right) predictions from the dispersion modelling results, overlayed on the Burrup Peninsula (in grams per grid cell).

5.2 Allowable Monitoring Station Sites

Figure 2 shows the grid cells considered as potential sites for locating an air quality monitoring station. Sites in red are existing monitoring stations (operated by others) and the (two) sites in orange are monitoring stations that could be potentially used in the monitoring network. Due to their relative distance from the other study sites, these were not forced to be included in the network, but were only optionally included, i.e., these locations were available, but whether they were selected was an outcome of the optimisation process itself.

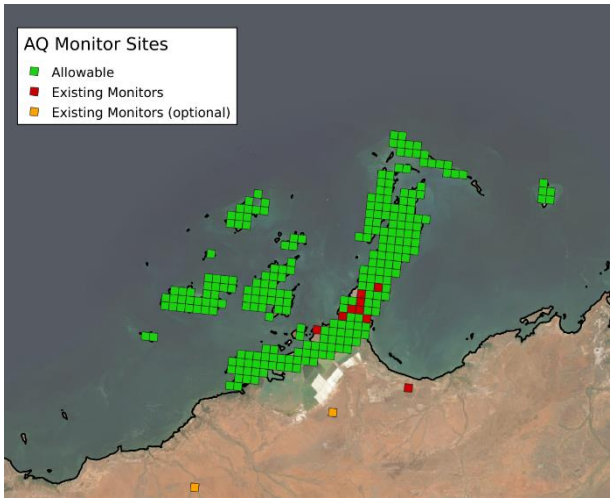

Figure 2: Grid cells considered as potential locations for air quality monitoring stations

5.3 Registered Rock Art Sites

The registered sites from the restricted DPLH register are shown in Figure 3 with the outlines and corresponding centroids. Figure 3 also includes the calculated site density used in the optimisation to weight the exposure error.

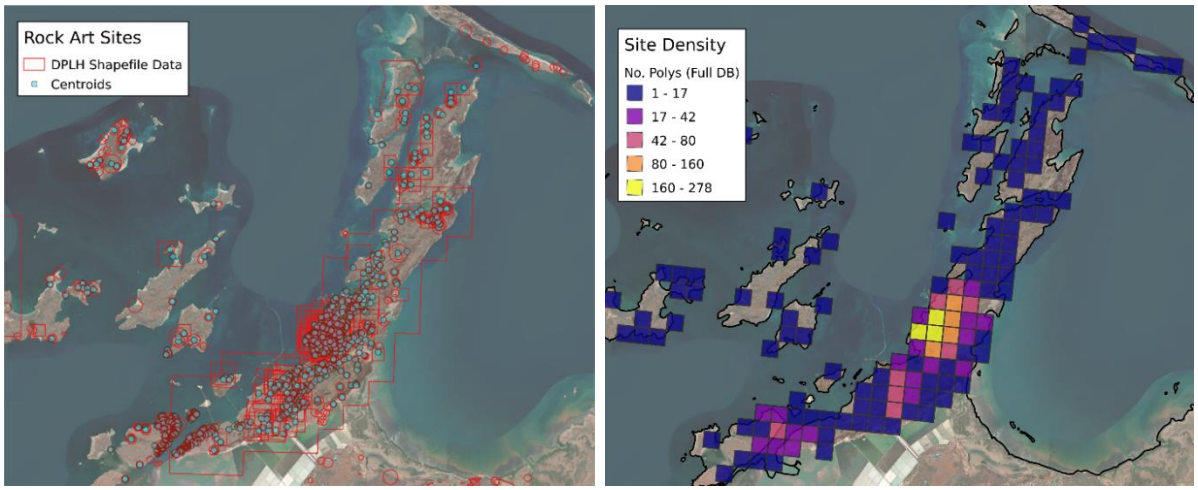

Figure 3: Restricted DPLH data for registered sites, and corresponding site density (sites/grid cell) used for weightings in the optimisation.

5.4 Data Interpolation

Interpolation of the measured air quality at the monitoring sites is required to predict the specie concentrations at locations that are not coincident with monitors. For spatial data a kriging methodology is typically used, making use of the fact that nearby monitors are expected to record similar readings. A semi-variogram is used to establish the distance-dependence of a dataset and can assist in setting the parameters for the kriging methodology 3. Figure 4 shows the semi-variograms for Eq-NOx and Eq-SOx for Murujuga using all the allowable monitoring locations as the sample locations. These demonstrate the spatial dependence of the data. A squared negative

exponential Radial Basis Function (RBF) was fitted to the data, with the length scale parameter tuned using data from the Ramboll simulations, using the “sci-kit learn” toolbox 4.

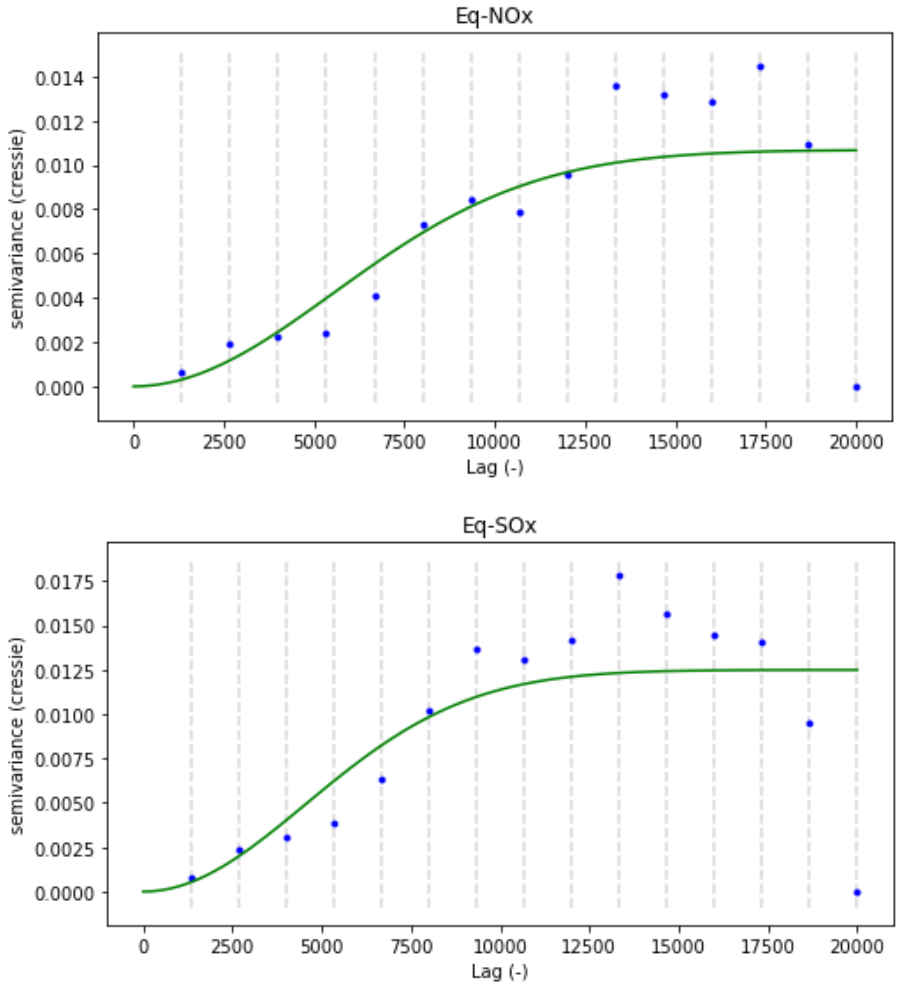

Figure 4: Variograms for NO_x and SO_x for annual data at Murujuga. A Radial-Basis-Function (RBF) has been used to fit the data.

5.5 Optimisation

There is an exceptionally large number of combinations of air quality monitoring sites that could form a network for measuring the air quality at Murujuga. For example, Figure 2 shows 258 potential monitoring sites and even when only considering these there are between 10^{13} (7 additional monitors) and 10^{30} (20 additional monitors) possible combinations which could form a monitoring network. For this reason, numerical optimisation is desirable – it allows efficient searching of the problem space to find a combination of monitoring sites that give the best performance (lowest error), while also accounting for other constraints.

For this project a Genetic Algorithm (GA) was used to perform the optimisation. When properly tuned, a GA is an efficient optimisation method able to incorporate complex constraints. A key feature of GAs is that during any iteration (or generation) there are

a number of possible networks (the population). In each generation each network (an individual) is assessed against how well it meets the optimisation objective (its fitness). After the fitness of each individual is assessed, the population is ranked and a number of processes follow in order to try and improve the overall fitness of the population. These are (typically): dropping the worst performing individuals and replacing them with new individuals, swapping characteristics between individuals in the population (breeding), and introducing random changes to an individual (mutation). This allows better exploration of the problem space. The end result is that after a sufficient number of successive generations (i.e., through repeating the process), the population should converge on a globally optimal solution, noting that each run may not necessarily converge on the same solution. This process was used to determine an optimal air quality monitoring network for Murujuga.

6 Results

The results from the optimisation using the process above are given in the following sections.

6.1 Number of air quality monitoring stations

Using the periodic data calculated from the Ramboll modelling results, a Genetic Algorithm model was created, with the constraints outlined above. The optimisation model was run multiple times, each time with a different number of air quality monitoring sites (between 5 and 20 sites in addition to the existing ones). This allowed the effect of the number of monitoring sites on the network fitness to be determined, as shown in Figure 5. This provides some insight into the trade-off between the cost of the programme (number of monitor sites) and the network performance (fitness).

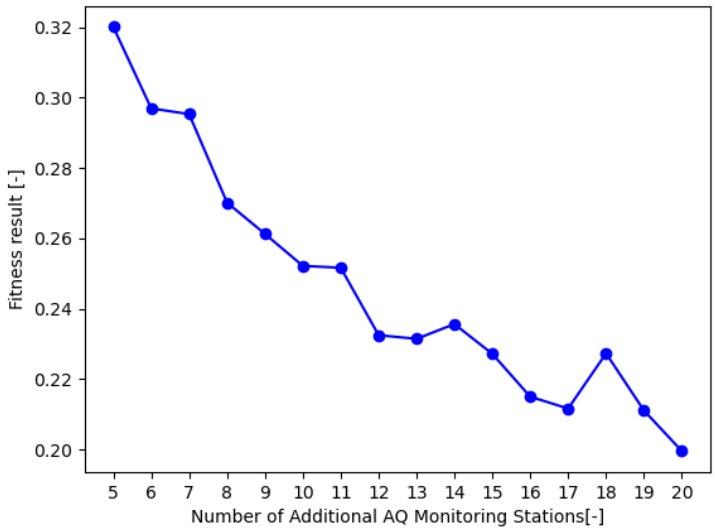

Figure 5: Final fitness of the best individual after optimisation against the number of monitoring sites

As would be expected, Figure 5 shows a decrease in the fitness (i.e., exposure error) as the number of air quality monitors is increased. There is a relatively uniform trend for the performance of the network to increase as the number of monitors increases. While the overall trend is towards a better network, there is some randomness in the results, and the addition of a monitor does not necessarily show an increase in performance. This is result of randomness inherent in the process in conjunction with the fact that as the number of monitors increases the number of potential combinations increases substantially, and the best individual found by the GA may not be the global optimum.

The final site selection for an air-quality monitoring network with 18 monitors is shown in Figure 6.

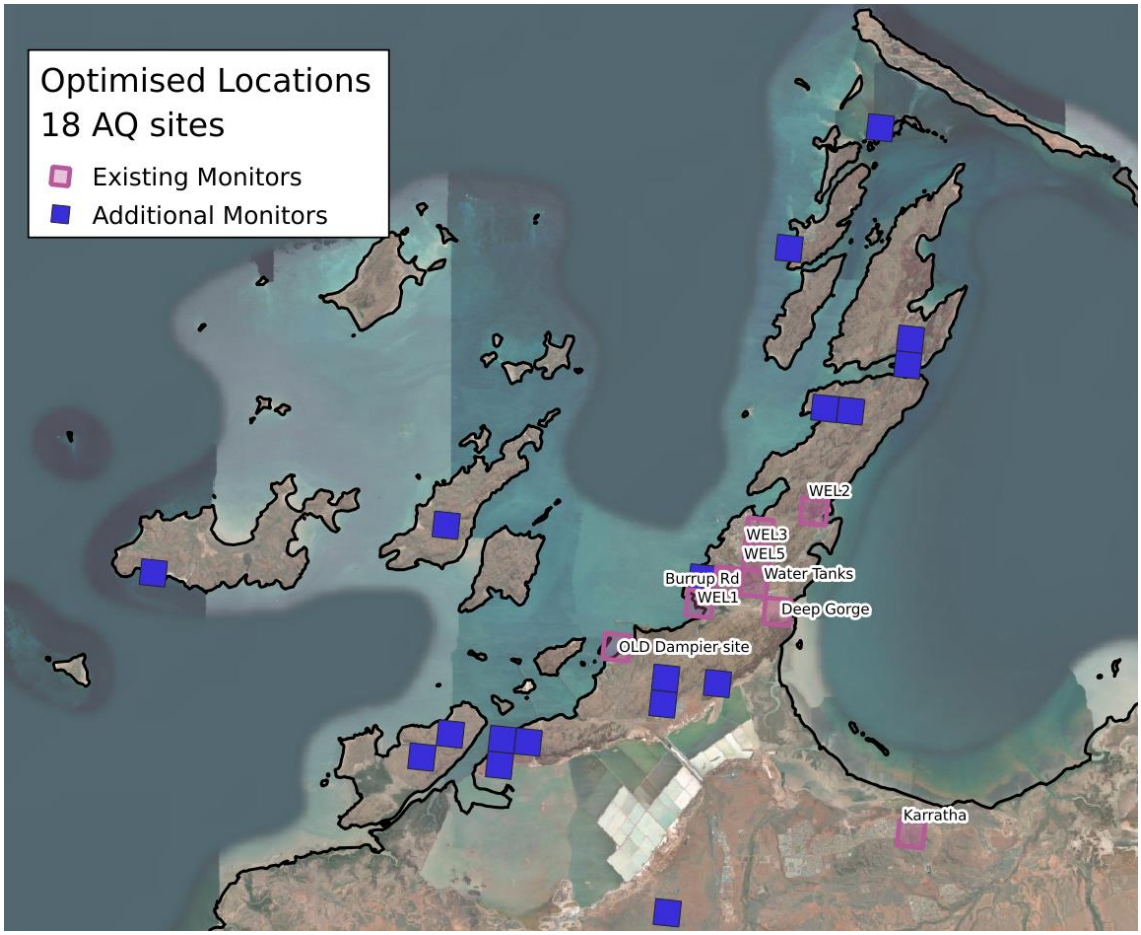

Figure 6: Optimal locations for a network of 18 air quality monitoring stations

6.2 General features of the optimal networks

There is an inherent component of randomness in the GA results, so it is useful to compare the locations that result from the optimisation as the number of monitoring stations is changed. This allows an overall understanding of what the final network may look like, and which particular regions may be more favourable for siting an air-quality monitoring station. Figure 7 shows the number of times that a potential site (from the list of allowable locations) forms part of an optimal monitoring network.

There is a large spread in the sites appearing only once or twice, but many appear in at least six (out of 16) of the optimisation results and two are selected in 15. These two locations are both southeast of Dampier and in adjacent grid cells. The next most selected areas are on the eastern part of West Intercourse Island and north of King Bay. Most of the remaining sites identified by the optimisation occur in the north of the archipelago with a relatively uniform spread, and almost all of the rest are on East and West Lewis islands. Enderby Island was selected in one of the optimisation results and the DWER owned “OLD Karratha site” was selected in two.

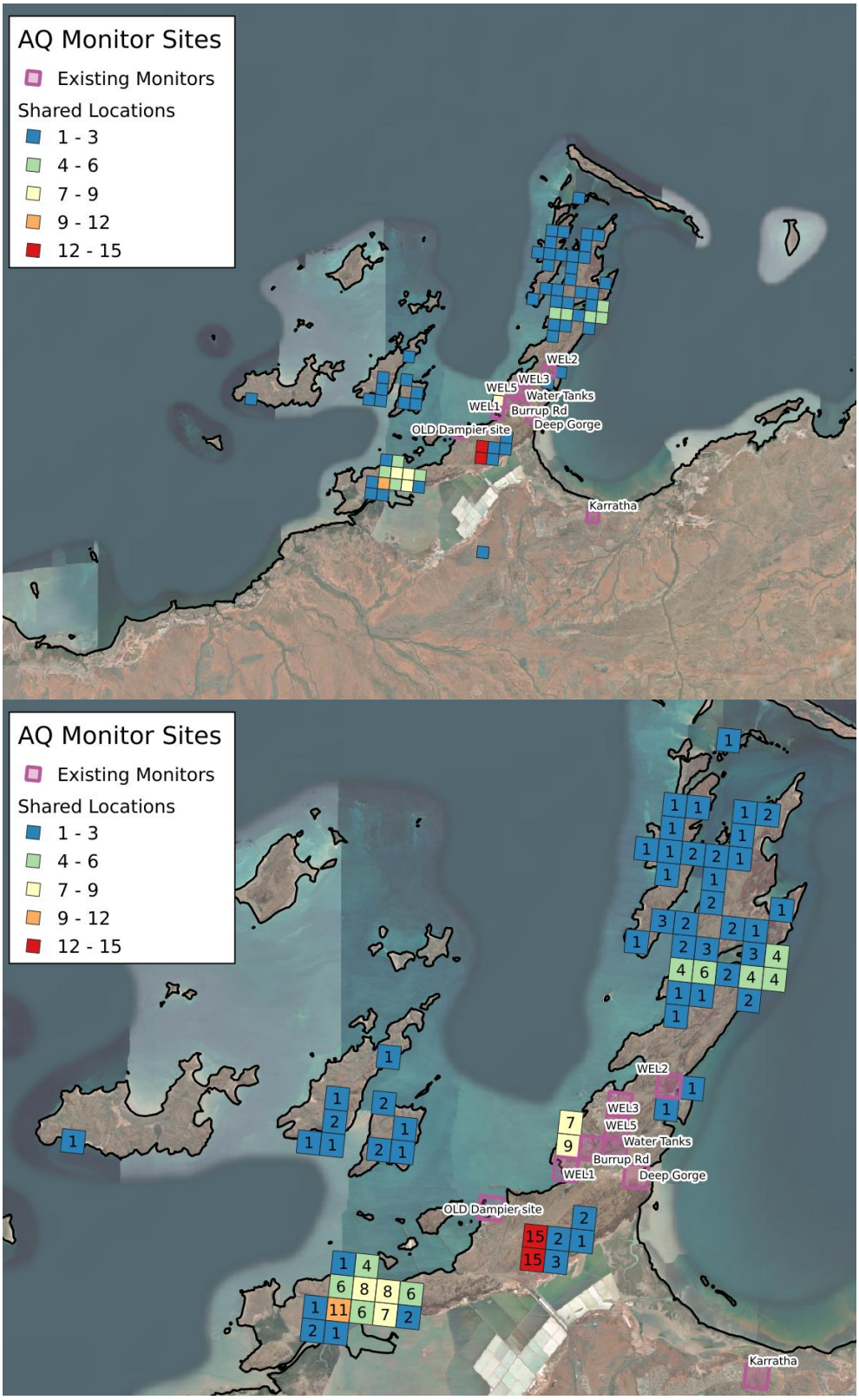

Figure 7: *Locations common between optimal networks with different numbers of monitoring stations.*

7 Conclusions and Recommendations

This report presents the outcomes of a data driven site selection process for an air-quality monitoring network at Murujuga. Hourly data on airborne pollutants for a full year (nominally 2030) was used to predict the performance of the network. The pollution data was sourced from dispersion modelling simulations commissioned by DWER and undertaken by Ramboll. The simulation data was aggregated into two datasets, cumulative exposure over 1 year, and a second dataset, containing 13 periods of cumulative exposure over 4 weeks. The objective of the optimisation was to minimise the error in predicted measurements of SOx and NOx using these datasets.

The periodic dataset was expected to be more representative of the actual monitoring network, which proposes to make use of passive sampling devices (predominantly) which will be collected on an approximate monthly basis. The annual dataset allowed for faster turnaround during testing and configuration of the optimisation while also allowing the results returned using the periodic data to be cross-checked.

The raw exposure errors from the trial networks were weighted by data obtained from the “Aboriginal Heritage Sites and Places” register, maintained by the Department of Planning, Lands and Heritage. In addition, a classification (allowed/excluded/potentially allowed) was used to filter the possible locations on which a monitor could be placed. Finally, a number of existing air-quality monitoring stations operated by others were assumed to be available to the project, either directly or as a site for co-locating new monitors. As such, the locations of these monitors were excluded from the list of allowable locations, but their data was included in the reconstruction of virtual exposure data, and fitness calculations.

Optimisations were performed for different numbers of monitoring stations, from 5 to 20 in addition to the existing monitors. By analysing the fitness as the number of monitors was increased it was established that increasing the number of monitors gives improved performance when capturing the trends in the 13, 4-week period data.

7.1 Final Selection of Air-Quality Monitoring Station Sites

The results indicate that a suitable number of additional air-quality monitoring stations is likely between 15 and 20. These, in combination with the existing air-quality monitoring stations, would result in an air-quality monitoring network that is able to reliably determine the exposure of a rock art site with a known level of error. The results presented here will be used by the project statisticians to assist them in making a final recommendation on the number of air quality monitoring stations required. The resulting locations are based on square grid cells approximately 1300m on a side. More precise siting of the monitors within these cells will be established on-site, and in conjunction with the traditional owners and the Murujuga Aboriginal Corporation. Final siting will also be informed by fine scale (<1m) resolution Computational Fluid Dynamics (CFD) models of the sites, using the prevailing conditions at Murujuga as inputs.

8 References

1. Ramboll 2021, "*Study of the Cumulative impacts of Air Emissions in the Murujuga Airshed*", Final Report prepared by Ramboll for the Department of Water and Environmental Regulation, 19 July 2021.

2. Department of Planning Lands and Heritage, 2021, "*Aboriginal Heritage Places*", url https://catalogue.data.wa.gov.au/dataset/aboriginal-heritage-places (public dataset)

3. Cressie, 1993, "*Statistics for Spatial Data, rev. edn*.", Wiley.

4. Pedregosa, F.; Varoquaux, G.; Gramfort, A.; Michel, V.; Thirion, B.; Grisel, O.; Blondel, M.; Prettenhofer, P.; Weiss, R.; Dubourg, V.; Vanderplas, J.; Passos, A.; Cournapeau, D.; Brucher, M.; Perrot, M. & Duchesnay, E., 2011. "Scikit-learn: Machine Learning in Python", Journal of Machine Learning Research, vol 12, pp 2825-2830.
url https://scikit-learn.org/stable/index.html

A Appendix A – Additional results

This appendix provides the optimised locations for different numbers of monitoring stations in the network, and the convergence history for each of the optimisations.

A.1 Monitoring station locations

The figures below show the optimised locations for the monitoring network for each number of monitors using the dispersion modelling data.

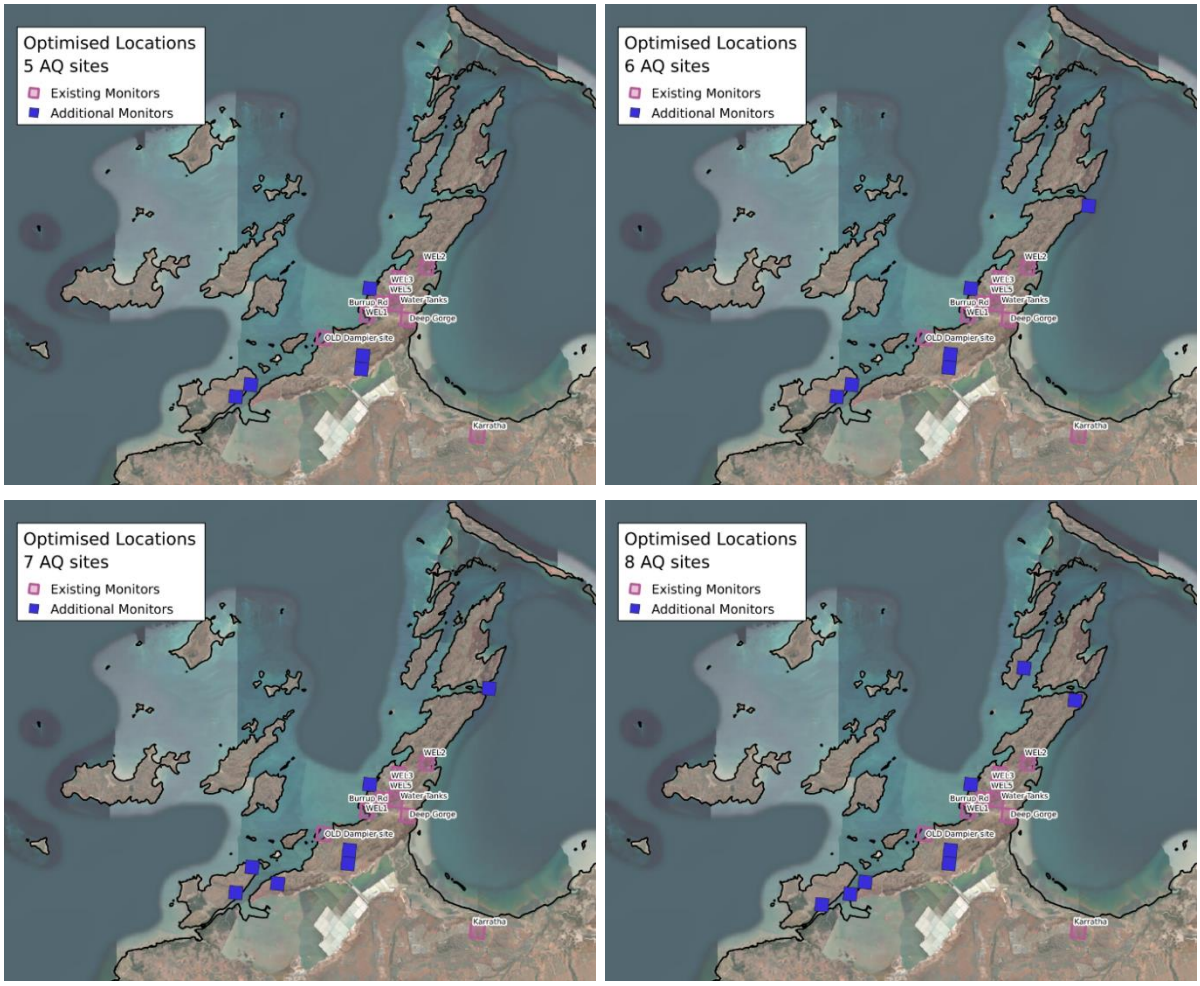

Figure 8: Final locations optimised by the GA for between 5 and 20 additional monitoring stations

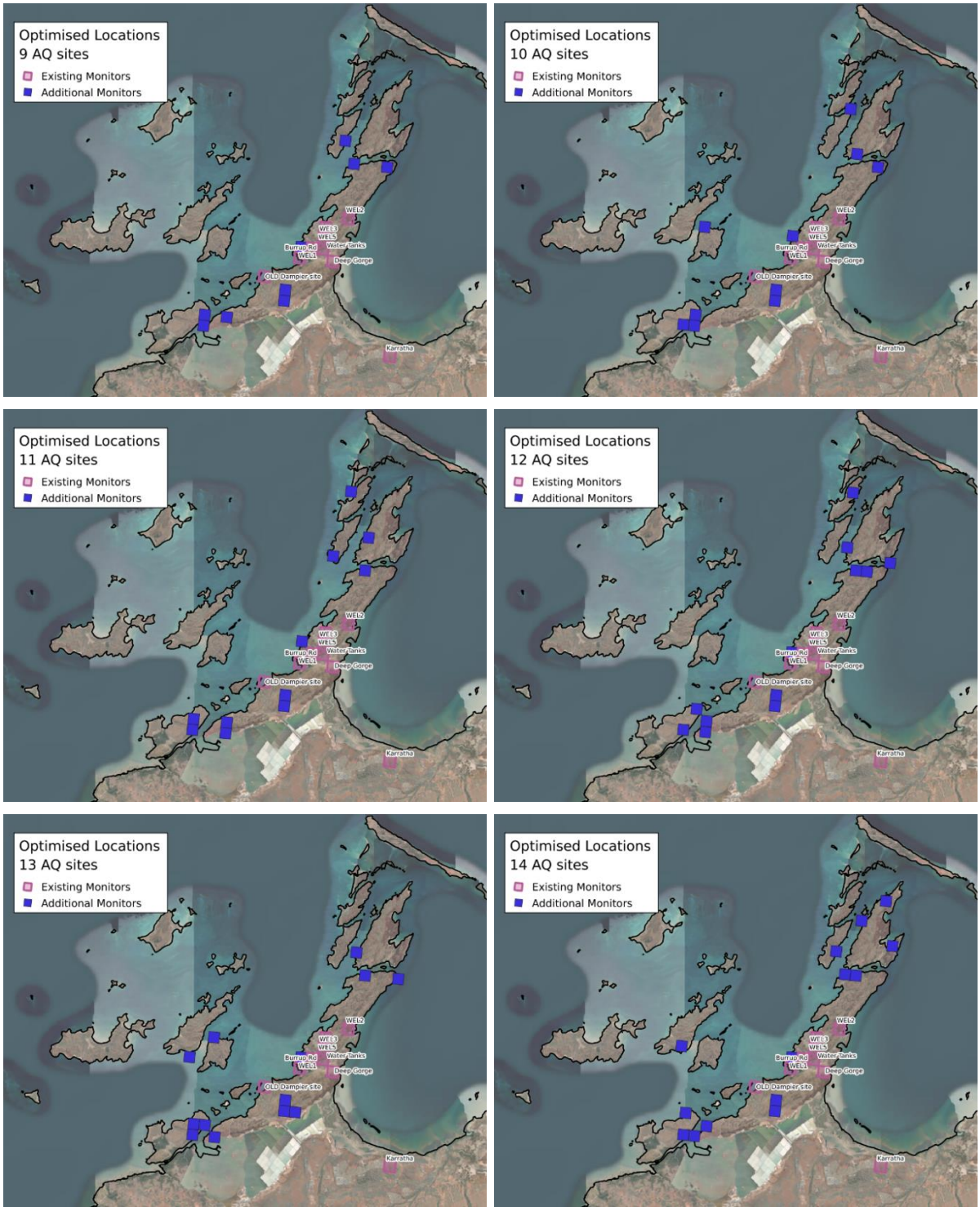

Figure 8: Final locations optimised by the GA for between 5 and 20 additional monitoring stations (continued)

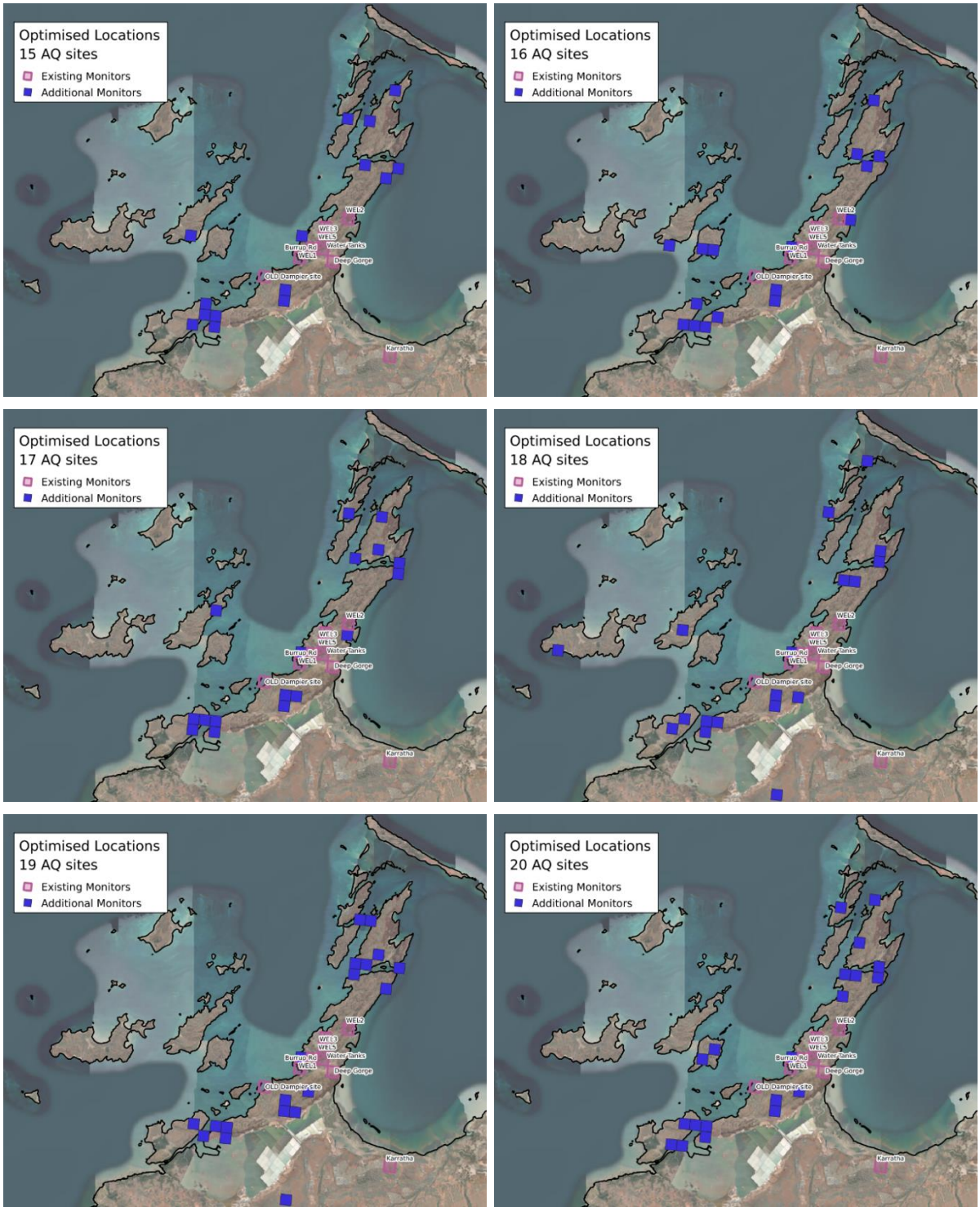

Figure 8: Final locations optimised by the GA for between 5 and 20 additional monitoring stations (continued)

A.2 GA convergence

The figures below show the GA convergence for differing numbers of monitors using the annualised dispersion modelling data.

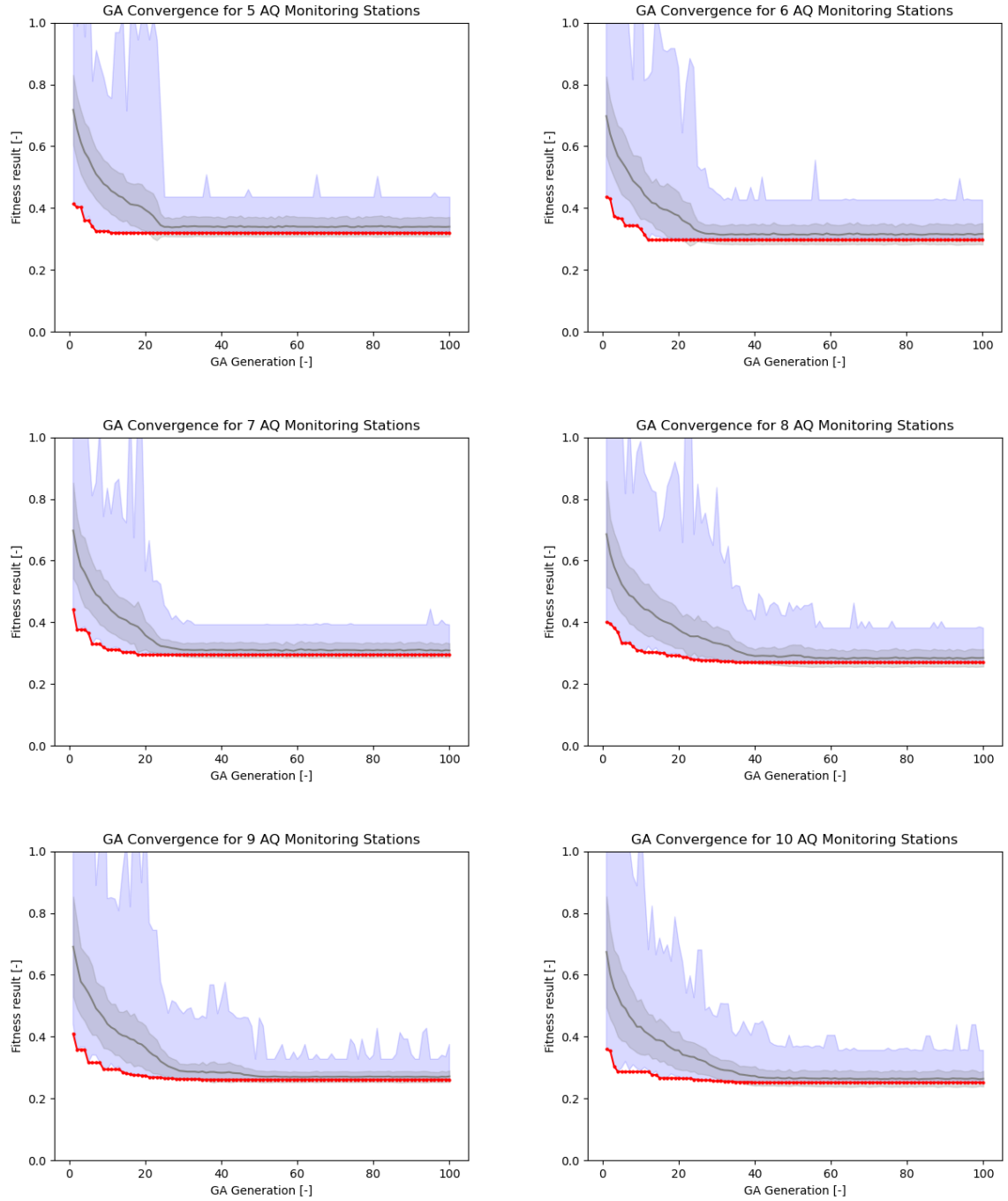

Figure 9: Convergence History for the GA for between 5 and 20 additional monitoring stations

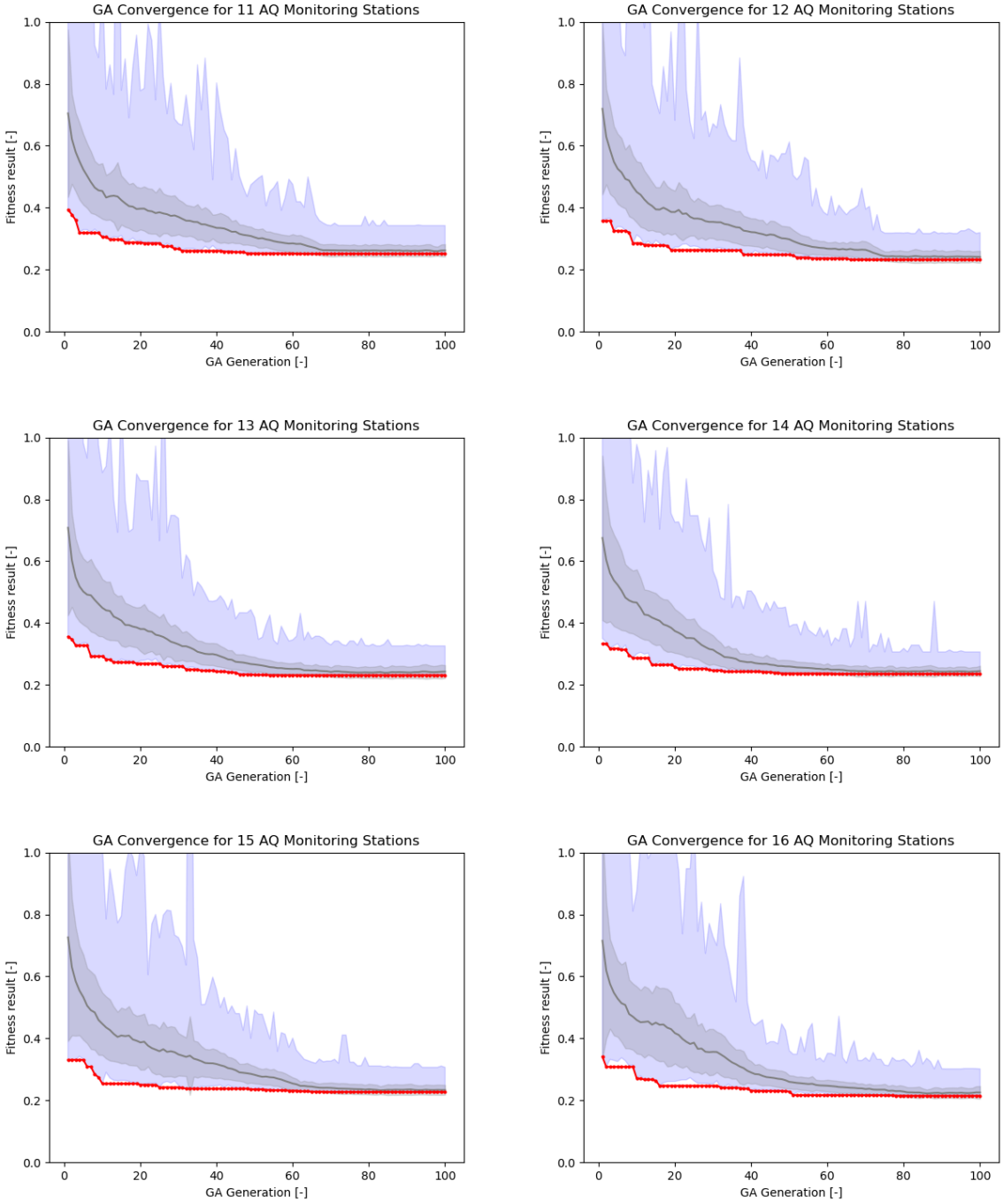

Figure 9: Convergence History for the GA for between 5 and 20 additional monitoring stations (continued)

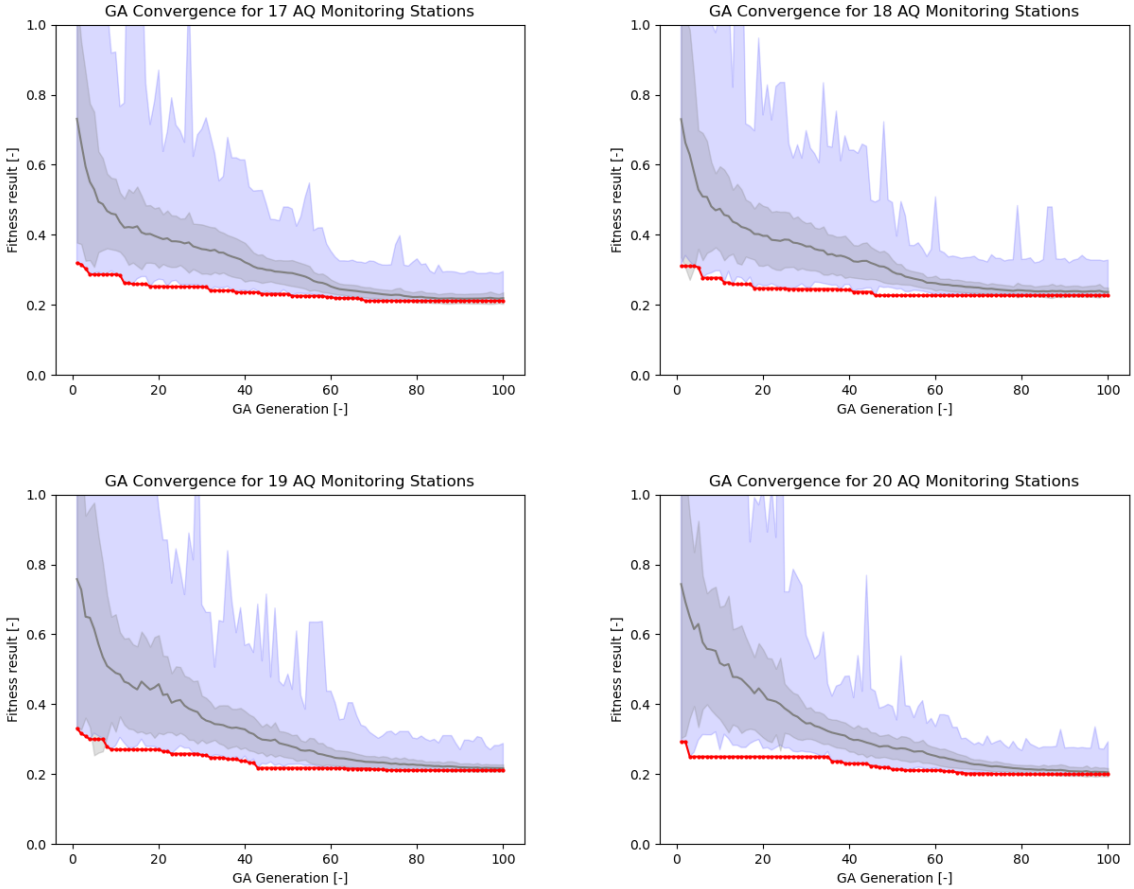

Figure 9: Convergence History for the GA for between 5 and 20 additional monitoring stations (continued)

B Comparison of Registered Site Databases

The initial trials of the optimisation process were made using the publicly available register of Aboriginal Sites from the Department of Planning, Lands and Heritage. Prior to the final optimisation run, the full (and restricted) database was made available. For completeness, a comparison was made between this dataset and the publicly available register to assess whether the choice of database was likely to affect the optimisation. Figure 10, below, shows the data from each of the databases, after aggregation onto the grid used for optimisation, while Figure 11 shows the difference in the optimisation weights, using the public database as the reference.

Apart from in a small number of grid cells the weights from both datasets are similar, and as such either dataset could be used as the input to the optimisation without a substantial impact on the result.

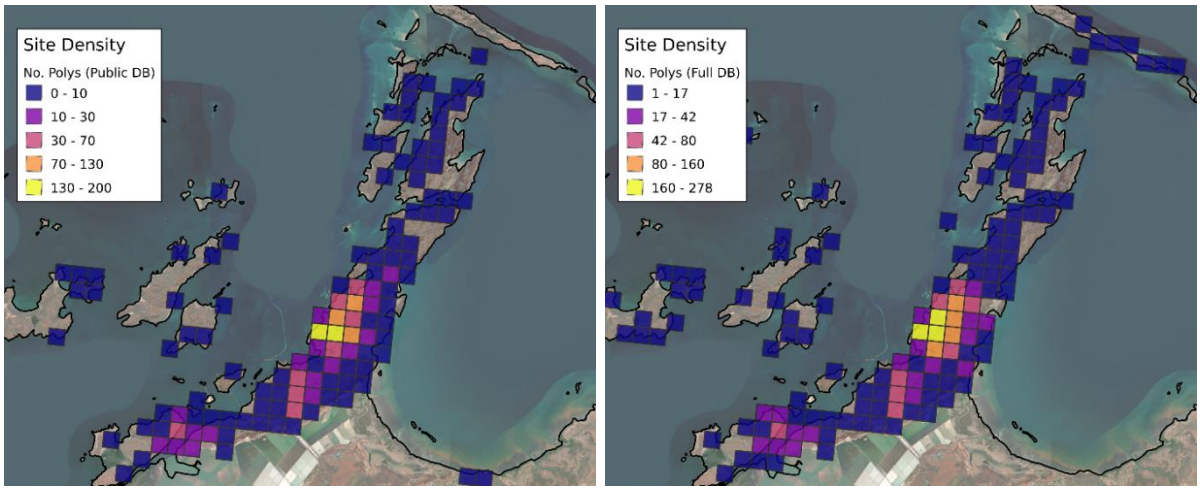

Figure 10: Comparison of the aggregated site data from the public and restricted databases.

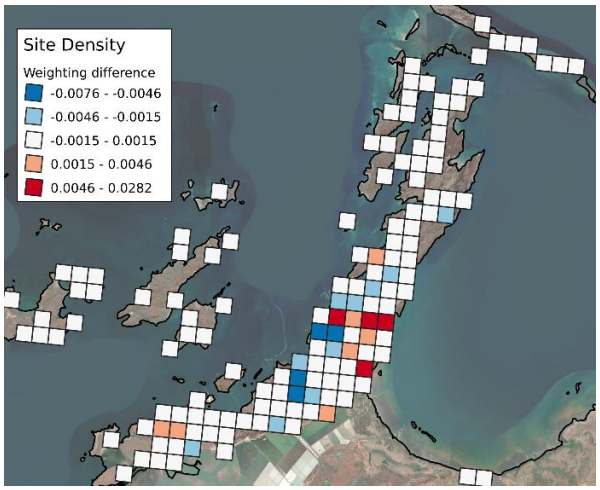

Figure 11: Changes to the weights resulting from the use of the full registered site database instead of the publicly available database.

Appendix III – Sample wind table

Full table of wind occurrence for Karratha Aero (004083):

Rank	Wind speed	Wind Direction	% Occurrence	Cumulative % Occurrence
1	Medium	W	19.5	19.5
2	Low	SW	8.5	28
3	Medium	NE	8.5	36.5
4	Low	W	8	44.4
5	Medium	NW	6.7	51.2
6	Low	S	6.3	57.4
7	Medium	E	5.3	62.7
8	Low	NW	4.9	67.6
9	Low	NE	4.8	72.4
10	Low	E	4	76.3
11	Low	SE	3.5	79.9
12	Medium	SW	3.2	83
13	Low	N	2.8	85.9
14	High	W	2.1	88
15	Medium	N	1.8	89.8
16	Calm	S	1.6	91.4
17	Medium	SE	1.5	92.9
18	Calm	SW	1.2	94.1
19	Calm	SE	1.2	95.3
20	High	E	0.9	96.2
21	Calm	E	0.8	97
22	Medium	S	0.8	97.8
23	Calm	NE	0.6	98.4
24	Calm	W	0.4	98.8
25	Calm	N	0.4	99.2
26	Calm	NW	0.3	99.6

Rank	Wind speed	Wind Direction	% Occurrence	Cumulative % Occurrence
27	High	NE	0.2	99.8
28	High	SE	0.1	99.9
29	High	SW	0.1	100
30	High	S	0	100
31	High	NW	0	100
32	High	N	0	100