On the use of lithogenic tracer measurements in aerosols to constrain dust deposition fluxes to the ocean southeast of Australia.

Claudia Hird [a*], Morgane M.G. Perron [ab*], Thomas M. Holmes [c*], Scott Meyerink [c], Christopher Nielsen [a], Ashley T. Townsend [d], Patrice de Caritat [e], Michal Strzelec [a], Andrew R. Bowie [ac]

a Institute for Marine and Antarctic Studies (IMAS), University of Tasmania, Battery Point, Tasmania, Australia.

b Université de Brest - UMR 6539 CNRS/UBO/IRD/Ifremer, Laboratoire des sciences de l'environnement marin (LEMAR) - Institut Universitaire Européen de la Mer - Rue Dumont D'Urville, 29280 Plouzané, France

c Australian Antarctic Program Partnership (AAPP), University of Tasmania, Battery Point, Tasmania, Australia.

d Central Science Laboratory, University of Tasmania, Hobart, Tasmania, Australia

e John de Laeter Centre, Curtin University, Bentley WA 6845, Australia

* These authors contributed equally to this work.

Correspondence to: Morgane M.G. Perron, morgane.perron@univ-brest.fr

Abstract

Australia contributes a significant amount of dust-borne nutrients (including iron) to the Southern Ocean, which can stimulate marine primary productivity. A quantitative assessment of the variability of dust fluxes from Australia to the surrounding ocean is therefore important for investigating the impact of atmospheric deposition on the Southern Ocean's carbon cycle. In this study, lithogenic trace metals (aluminium, iron, thorium and titanium) contained in aerosols collected between 2016 and 2021 from kunanyi/Mount Wellington in lutruwita/Tasmania (Australia) were used to estimate dust deposition fluxes. Lithogenic fluxes were calculated using each tracer individually, as well as an average using all four tracers. This latter approach enabled an assessment of the uncertainty associated with flux calculations using only individual tracers. Elemental ratios confirmed the lithogenic nature of each tracer in aerosols when compared with both Australian soil samples and the average Earth's upper continental crust. Lithogenic flux estimates showed annual dust deposition maxima during the austral summer, following the Australian dust storm season, and annual minimum deposition flux over winter. The data provided

here will help to constrain model estimates of southern hemisphere atmospheric deposition fluxes and their subsequent impact on global ocean biogeochemical cycles.

Environmental significance / Plain language summary

Dust deposition flux was investigated in lutruwita/Tasmania, Australia, between 2016 and 2021. Results show that the use of direct measurement of aluminium, iron, thorium and titanium in aerosols to estimate average dust deposition fluxes limits biases associated with using single elements. Observations of dust deposition fluxes in the Southern Hemisphere are critical to validate model outputs and better understand the seasonal and interannual impacts of dust deposition on biogeochemical cycles.

a supprimé: Determined lithogenic flux estimates were consistent with a regular dust deposition peak during the austral summer, in line with the dust storm season in the southeast of Australian, and a low atmospheric deposition in winter. ocean This study provides an insight into the seasonal and interannual variability of dust deposition fluxes from the southeast of Australia based on aerosol sample measurements. This information will enhance our understanding of nutrient-bearing dust deposition to the Australian sector of the Southern Ocean and may prove useful in refining modelling estimates of southern hemisphere atmospheric deposition fluxes and their subsequent impact on global biogeochemical cycles.¶

1. Introduction

Lithogenic mineral particles such as iron oxyhydroxides, kaolinite, illite and smectite are commonly entrained into the atmosphere (Cudahy et al., 2016) following the erosion of the Earth's Upper Continental Crust (UCC) (Crawford et al., 2021). Such dust particles are the primary source of trace metals including aluminium (Al), iron (Fe), thorium (Th) and titanium (Ti) to the atmosphere, which can therefore be used as tracers of aeolian lithogenic inputs to the ocean (Baker et al., 2020). Dust carries important nutrients, including Fe, to marine ecosystems, feeding primary producers (Mackie et al., 2008). Due to the current lack of field observations on the concentrations of aeolian trace metals and their corresponding dust deposition fluxes, large uncertainties remain regarding how and to what extent dust supply fertilises key oceanic regions such as the Southern Ocean with vital nutrients. This leads to a poor understanding of the impact of dust deposition on the biological carbon pump.

The amount of dust entrained into the atmosphere depends on soil surface roughness, vegetation and coverage, on particle size, composition, and moisture content, and on local conditions such as wind speed and rainfall, which change both regionally and seasonally (Mahowald et al., 2009). Air masses can carry dust over thousands of kilometres before particles return to land or fall onto the surface ocean (Mackie et al., 2008). Atmospheric deposition of dust to the open ocean has been demonstrated to act as a key supplier of vital macro- and micro-nutrients (such as Fe) to the marine ecosystem (Mackie et al., 2008; Weis et al., 2024). For example, during the austral summer 2019-

2020, nutrient supply from large dust-containing bushfire emissions (Perron et al., 2022; Hamilton et al., 2022) was identified as the main trigger of a large and long-lasting phytoplankton bloom in the South Pacific Ocean (Weis et al., 2022).

Field and modelling approaches to estimating dust deposition both offer various benefits and drawbacks. Field observations at sea are influenced by local environmental conditions (i.e., weather, surface ocean properties) and are not representative of the large scale or long-term atmospheric deposition trends (Anderson et al., 2016). Time-series stations on land can overcome the issue of temporal coverage but may not be representative of atmospheric loading and deposition over remote oceanic regions. To date, global models are not capable of reproducing atmospheric concentrations of trace metals transported in dust to remote areas and cannot accurately quantify particle settling rates (Anderson et al., 2016). Considering the Southern Hemisphere, model estimates tend to overestimate total dust emission at the source and underestimate soluble trace element deposition fluxes over the ocean (Anderson et al., 2016; Ito et al., 2020). To reduce uncertainty in dust deposition fluxes to the open ocean it is essential to validate model fluxes using field-based observations. Long-term atmospheric observatories, particularly near the coasts, are attracting increasing interest from the scientific community as a platform to better understand seasonal to interannual patterns of deposition events in addition to shipboard observations and satellite estimates (Perron et al., 2022; De Deckker, 2019).

In Australia, the large spatial heterogeneity of soil types and the highly episodic nature of weather events such as droughts, bushfires and dust storms make it particularly difficult to model dust deposition fluxes (Mackie et al., 2008), though modern dust emissions from South Australia have been observed as far as the snow sheet of inner Antarctica (Vecchio et al., 2024). A main source of trace metals to the Australian sector of the Southern Ocean is dust carried from kati thanda/Lake Eyre and dhungala-barka/Murray-Darling geological basins (De Deckker, 2019). The typical dust storm season in Australia spans from September to November (austral spring), with the most extreme storms occurring in September (O'Loingsigh et al., 2017). The dust season can extend through the austral summer due to bushfires (and postfire unvegetated ground) across southern Australia (Hamilton et al., 2022). In a study conducted by Perron et al. (2022), atmospheric concentrations of mineral dust and associated lithogenic tracers (Al, Fe and Ti) were reported to

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be 2.5-fold higher, on average, during fire events compared to days not impacted by bushfires in lutruwita/Tasmania, Australia.

Dust deposition fluxes reported by different models range over an order of magnitude (from 0.55 to 5.48 mg m^{-2} d^{-1}) over the Southern Ocean region southeast of Australia (Mahowald et al., 2006; Weis et al., 2024). Different methods have also been used to estimate dust deposition fluxes from field samples. While broadly used in air pollution studies (Bindu et al., 2016), total aerosol loading measurements based on gravimetry does not enable the discrimination of atmospheric sources (e.g., dust vs anthropogenic). In addition, such a method is not compatible with atmospheric trace metal studies, where strict protocol requires minimal filter handling to prevent contamination. from the analysis of a single tracer element, for example Al or Th, in aerosol samples and in seawater (Anderson et al., 2016). However, single element dust flux estimates are subject to anomalous data stemming from contamination, deviation from the mean UCC, or preferential mineralization following a particular laboratory protocol. Recently, the analysis of four lithogenic tracers (namely, Al, Fe, Th, and Ti) in marine sinking particles collected at the Southern Ocean Time-Series (SOTS) mooring station (140°E, 47°S) were used to calculate an average 'multi-tracer' estimate of dust deposition fluxes to surface waters of the subantarctic ocean south of Australia (Traill et al., 2022). The latter field-based flux estimates showed good agreement with remotely sensed proxies of dust transport and modelled deposition estimates. Elemental ratio analysis in the same sediment trap samples suggested that lithogenic material from southeastern Australia was the most likely source of Al, Fe, Th and Ti to this area of the Southern Ocean (Traill et al., 2022).

In this study, the analysis of the same four lithogenic tracers (Al, Fe, Th, and Ti) was performed in aerosol samples collected at the kunanyi/Mount Wellington time-series sampling station in southern lutruwita/Tasmania (Australia). Dust deposition fluxes were estimated from both individual tracer concentrations and using the multi-tracer approach used by Traill et al. (2022). Here we report a 5-year (2016-2021) time-series of dust deposition flux estimates downwind of the south-eastern Australian dust path, at one of the gateway to the Southern Ocean. The suitability of the four metals as lithogenic tracers was also verified by comparing elemental ratios (relative to Al) in the aerosol samples to the average topsoil composition in Australia (this study) and to the averaged UCC composition (McLennan et al., 2001).

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2. Material and methods

2.1 Aerosol collection and study site

kunanyi/Mount Wellington overlooks Hobart, the capital city of the Australian island state of lutruwita/Tasmania. The mountain is in a strategic position for sampling one of the three major atmospheric pathways in Australia (Baddock et al. 2015; Bowler 1976), where air-masses from mainland Australia are transported south-eastwards over lutruwita/Tasmania (and our sampling site, Figure 1) before reaching the Southern Ocean. This study uses aerosol filters collected on a HiVOL 3000 air particulate sampler (Ecotech, Acoem, Melbourne, Australia) positioned at 1,271 m above sea level, on the summit of kunanyi/Mount Wellington. Filter samples have been collected for Total Suspended Particulates (TSP) since September 2016, with each sample representing a period ranging from a few days to 2 weeks, depending on weather conditions and specific weather events, and allowing for sampler servicing and calibration. Samples suspected for contamination or that were significantly wet at the time of recovery were discarded and sampling was suspended in the winter time of 2017, 2018 and 2019 for operational reasons. As a result, 125 aerosol samples were selected from the kunanyi/Mount Wellington atmospheric time-series collection for this study (November 2016 - February 2022). The origin and concentration of aerosol Fe in 80 samples from this dataset was previously reported in Perron et al. (2022), however the

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present study differs in using total concentrations of Fe, Al, Th and Ti to calculate atmospheric (dust) deposition fluxes and the associated seasonal and interannual trends at the sampling station.

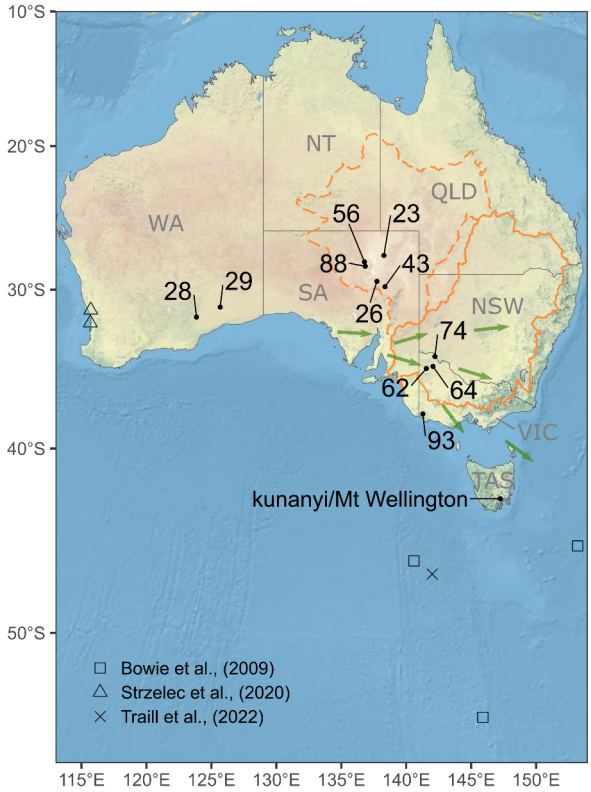

Figure 1. Location of the aerosol sampling station at kunanyi/Mount Wellington in Tasmania (TAS). Black dots display the locations of selected NGSA soil samples in the Australian states of Western Australia (WA), South Australia (SA) and Victoria (VIC) with identification numbers annotated (see Table S2). Prevailing dust pathways of southeast Australia are displayed as green arrows adapted from Sprigg (1982) and Mackie et al. (2008). The kati thanda/Lake Eyre (dashed line) and dhungala-barka/Murray-Darling (solid line) geological basins are delineated in orange. Location of previous field studies that estimated dust deposition fluxes are also indicated.

2.2 Aerosol leaching protocol

Laboratory work for aerosol and soil sample processing (sections 2.2 and 2.3) followed GEOTRACES recommended procedures for ultra-trace sampling and analysis (Cutter et al., 2017).

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All reagents were ultra-high purity (UHP) and either purchased (Baseline, SeaStar Chemicals) or distilled in-house using instrument quality reagents (IQ grade, SeaStar Chemicals). Whatman W41 (203mm x 254mm paper sheets, Sigma-Aldrich) filters were acid washed in a series of 24h hydrochloric acid (0.5 M HCl) baths and rinsed with UHP water to leach any impurities and reduce the impact of the cellulose filter on the analysis of trace elements in aerosols (Perron et al., 2020a). Perron et al. (2020a) suggested a 3-step leaching method to define trace metal concentrations and solubility in aerosols taken from land-based stations in Australia (Strzelec et al., 2020a, 2020b) and on research vessels operating around Australia and in the Southern Ocean (Perron et al., 2020b, 2021). Although samples were collected and analysed in batch over several years, the collection and analysis of each batch of samples follow the same protocol and the resulting data was quality-controlled against blanks, replicate analysis and Certified Reference Materials (Table S1).

One sub-sample of 47mm diameter was cut off each aerosol filter sheet collected at our sampling station using a sharp titanium punch cutter (Perron et al., 2020). Sub-samples were successively leached using UHP water (Milli-Q®, 18.2 MΩ) and 1.1 M ammonium acetate (10 mL, pH 4.7). The remaining filter residue was then digested using a mixture of concentrated nitric acid (HNO_3, 1 mL) and concentrated hydrofluoric acid (HF, 0.25 mL) at 120°C for 12 hours (Perron et al., 2020a). The sum of all three steps in the protocol provided the total concentration data for each lithogenic tracer in aerosols which is used in this study (Perron et al., 2020a). Satisfactory recoveries (>90%) were obtained for Al, Fe and Ti when applying the total metal digestion step of the protocol to two reference materials, the Arizona Test Dust (ATD) (Morton et al., 2013) and the GeoPT13 certified Koeln loess (International Association of Geoanalysts) (Potts et al., 2003) (supplementary Table S1). A smaller recovery of 87% (using only certified reference material) obtained for Th highlights the unique extraction and stability chemistry of the metal which our protocol is not optimised for. Thorium concentrations are therefore likely to be slightly underestimated in this study as discussed in section 3. Only total metal concentrations are discussed in the present study.

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2.3 Atmospheric deposition flux estimates

The total concentration of each lithogenic tracer in our samples was used to calculate single tracer-dust deposition flux estimates. Additional measurements on the collected aerosols (e.g.; carbon

and major ion analysis) were not available for this study, so intrinsic calculation of the total aerosol mass on each individual aerosol filter using these parameters was not possible.

In addition, due to the lack of necessary meteorological data to estimate particle deposition velocities specific to our study site, a single coarse particle deposition velocity of 0.2 cm s^{-1} was applied to trace metal-bearing dust deposition estimates based on the literature in similar study regions (Baker et al., 2017; Perron et al., 2020b; Winton et al., 2015). In this study, "F(X)" denotes the deposition flux estimate for the individual lithogenic tracer "X". F(X) (in mg m^{-2} d^{-1}) was obtained following equation (1):

$$F(X) = Cx * Vd \quad (1)$$

where X is the lithogenic tracer – Al, Fe, Th or Ti ; C_x is the total metal concentration (ng m^{-3}) in aerosols and V_d is a constant deposition velocity of 1723 m d^{-1} (0.2 cm s^{-1}). It should be mentioned that a factor of 3 uncertainty was previously attributed to the use of a set deposition velocity as it does not account for specific particle size in different aerosol samples or for specific atmospheric conditions such as humidity and wind speed at the collection time (Baker et al., 2016; Winton et al., 2016; Duce et al, 1991).

Similar to other studies reported in the literature, a single-tracer dust (lithogenic) deposition flux estimate, $F_{Lith(X)}$, was calculated by dividing F(X) by the average abundance ($[X]_{UCC}$, wt%) of the element X in the UCC as reported in McLennan (2001); Al = 8.04%, Fe = 3.5%, Th = 1.07x10^{-3}%, Ti = 0.41% following equation (2).

$$F_{Lith(X)} = \frac{F(X) \times 100}{[X]_{UCC}} \quad (2)$$

While $F_{Lith(X)}$ estimates are solely based on the analysis of a single lithogenic tracer, a multi-tracer dust deposition flux estimate, F_{LithAv}, was obtained by calculating the average of all four $F_{Lith(X)}$ (equation 3, N=4) for each individual aerosol sample.

$$FLithAv = \frac{\sum FLith(X)}{N} \quad (3)$$

Multi-tracer $F_{LithAv,}$ estimates were calculated using both the reported average UCC composition (McLennan, 2001) and Australian soil measurements (see section 2.4 in this study) as references for comparison.

2.4 Soil sampling and processing

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Eleven topsoil (0-10 cm) samples were selected from the National Geochemical Survey of Australia (NGSA) Project: Geochemical Atlas of Australia (Geoscience Australia), a continental-scale geochemical survey covering most of Australia (Caritat and Cooper, 2011; Caritat, 2022). In this study, soil samples originating from the southern part of Western Australia, from South Australia and from Victoria were selected and analysed (Figure 1) as they likely better represent particles entrained through the south-east Australian dust path towards our sampling station and the Southern Ocean (Baddock et al., 2015, Supplementary Figure S1). These sources include the geological basins of kati thanda/Lake Eyre and dhungala-barka/Murray-Darling, It should be mentioned that no sample from New South Wales was used for this study although a large part of the dhungala-barka/Murray-Darling basin is located in this state.

Ten milligrams of each soil sample was dry sieved through a 63 µm nylon screen to capture the soil fraction fine enough to be entrained into the atmosphere (Strzelec et al., 2020a). The sieved fraction was then processed through the same sequential leaching method described in section 2.2 (Perron et al., 2020a). Aerosol and soil leachates were analysed for a suite of elements, including Al, Fe, Th and Ti, by Sector Field Inductively Coupled Plasma Mass Spectrometry (HR-ICP-MS, Thermo Fisher Scientific, Element 2) at the Central Science Laboratory of the University of Tasmania. Increased spectral resolution was employed to resolve major spectral interference overlaps associated with analysis of Al, Fe and Ti. Further details on the ICP-MS analysis procedure are provided in Perron et al. (2020a).

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2.5 Atmospheric source tracking

The ratio between the total concentration of each lithogenic tracer of interest, T(X), and the total Al concentration, T(Al), in individual aerosol samples was calculated and compared to the same ratio in the average UCC reported in McLennan (2001) and in the average topsoil from southeastern Australia (Section 2.3). The so-called enrichment factor (EF, equation 3) was used to ascertain the lithogenic origin of Fe, Th and Ti in this study.

$$EF = \frac{\frac{T(X)}{T(Al)}aerosol}{\frac{T(X)}{T(Al)}UCC} \quad (3)$$

Using this approach, an EF value below 10 was considered to indicate a prevailing lithogenic source origin for the metal tracers, while an EF exceeding the threshold value of 10 is associated

with an enrichment from non-lithogenic atmospheric sources such as anthropogenic combustion (Shelley et al., 2015; Perron et al., 2022). Reimann and Caritat (2005) warned about the biases associated with using a low EF threshold to identify anthropogenic sources due to the natural variability in the Earth's crust composition, fractionation of elements during their emission to – and transport within – the atmosphere, and biogeochemical processes during and after aeolian transport. Here, a high EF threshold of 10 is adopted to account for such variability.

3. Results and discussion

3.1 Evaluating the lithogenic origin of the four tracers in aerosols

Enrichment Factors (EF) were calculated for Fe, Th and Ti measured in aerosols, and compared to the Australian soil samples selected from the NGSA (this study) and compared to averaged UCC composition from McLennan (2001) (Table 1). Calculated EF values were used to discard significant contributions of non-lithogenic sources to our lithogenic tracers in kunanyi/Mt Wellington aerosols as indicated by EF>10. Metal concentrations in individual NGSA soil samples analysed in this study are reported in the supplementary Table S2 (lithogenic tracers) and in Table S3 (other analysed trace metals not discussed in this study).

Table 1. Comparison of mean Al, Fe, Th and Ti concentrations measured (ppm) in Australian soil samples (n = 11) compared to concentrations reported in the average UCC by McLennan (2001). Enrichment factors (EFs) calculated for Fe, Th and Ti (using Al as a reference) in aerosols collected at kunanyi/Mount Wellington (n = 125) are also displayed using both crustal references

	UCC	Australian soils		kunanyi/Mount Wellington aerosols	
Al	80400	38560		/UCC	/Australian soil
Fe	35000	22616	**EF(Fe)**	1.6 ± 0.6	1.2 ± 0.5
Th	10.7	10.3	**EF(Th)**	1.3 ± 2.5	0.7 ± 1.2
Ti	4100	4313	**EF(Ti)**	1.2 ± 0.6	0.6 ± 0.6

Overall, EFs close to 1 were measured for all aerosol samples, suggesting that the lithogenic tracers used in this study are indeed of a prevailing crustal origin. Using Australian soil concentration (Table 1 and supplementary Table S2) to calculate EFs resulted in values further away from the threshold of 10. In particular, EFs calculated using Australian soil data are closer to 1 for Fe and

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Th when compared to using average UCC values (McLennan, 2001). Indeed, underestimated Th measurements due to incomplete sample digestion (section 2.2) in our study result in a similar underestimate of EF. Elemental ratio of Ti/Al in aerosol samples collected at kunanyi/Mt Wellington (Figure 2) were closer to the average ratio of the UCC, resulting in EF(Ti) closer to 1 when compared to using average Australian soil measurement as a reference.

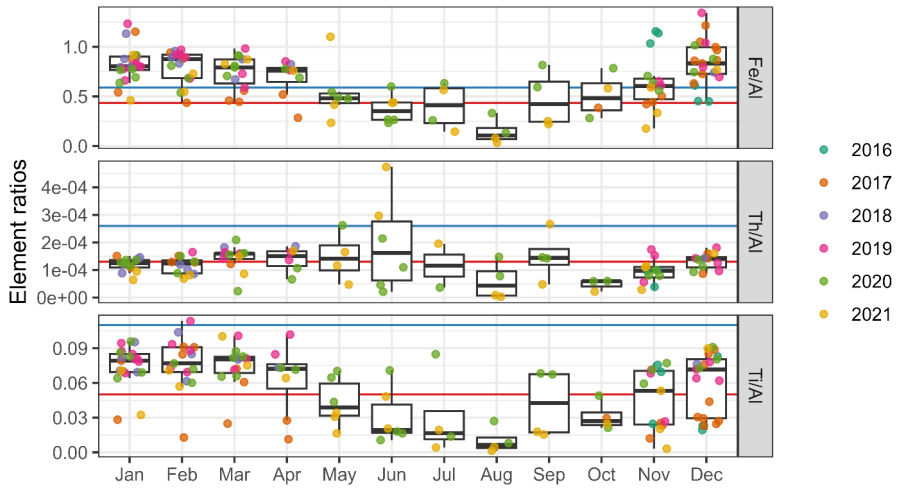

Figure 2. Boxplot of elemental ratios of Fe/Al (top), Th/Al (middle), and Ti/Al (bottom) in kunanyi/ Mt Wellington aerosol samples collected between 2016 – 2021, grouped according to month. Whiskers represent 1.5 times the interquartile range (75^{th} – 25^{th} percentile) beyond the boxes, while the upper, middle, and lower horizontal lines of the box represent the higher interquartile, median value and lower interquartile of the average monthly dataset, respectively. Colours represent each sample collection year. Horizontal red lines represent metal ratios in the average UCC (McLennan, 2001). Horizontal blue lines represent average metal ratios in the eleven selected NGSA Australian soil samples (this study). Two Th outliers (May and June 2020) were excluded from the Th dataset and subsequent calculation for clarity.

Mean Al and Fe concentrations measured in our Australian soil samples were both twice smaller than the average UCC values reported by McLennan (2001) while Ti and Th concentrations were similar within 10% (Figure 2 and supplementary Table S2). While Australian soil is known for its high Fe content (Mahowald et al., 2019, Strzelec et al., 2020a), a high soil heterogeneity across this vast country may explain such surprising observation. This resulted in calculated Th/Al and

Ti/Al ratios higher for Australian soil samples while Fe/Al ratios remained similar compared to the average UCC.

Elemental ratios calculated for individual aerosol samples are summarised in the supplementary Table S4. Both Fe/Al and Ti/Al ratios showed a clear seasonal trend, with higher ratios resembling mean ratios measured in Australian soil samples (Fe/Al=0.59 and Ti/Al=0.11, Figure 2) in the summertime (December-February) and lower Fe/Al and Ti/Al ratios closer to the average UCC ratios (Fe/Al=0.435 and Ti/Al=0.05, McLennan, 2001) in wintertime (June-August, Figure 2). Summertime Fe/Al ratios in kunanyi/Mt Wellington aerosols were slightly higher (Fe/Al =0.72) than the Australian soil measurements. This can be explained by increased contribution of local soil emission from Tasmania under drier weather conditions as the NGSA database shows higher Fe/Al ratio (on average 0.7, n=21 samples) in Tasmanian soil compared to other soil across Australia (Caritat and Cooper, 2011; Caritat, 2022). While enhanced air-masses originating from the Australian mainland cannot be observed in the summertime using HYSPLIT model (supplementary Figure S1), the Australian Bureau of Meteorology reports increasing southwards blowing winds at our sampling station from January through to March (supplementary Figure S2). Such discrepancies emphasise the complex regional wind pattern influencing our sampling station and highlight the need to consider other parameters such as seasonal changes in environmental conditions at the source region when investigating aerosol entrainment and transport. Ti/Al ratios were found to lie between our Australian soil (Ti/Al = 0.11) and UCC (Ti/Al = 0.05) references from December through to May, then falling below the UCC ratio in the cooler months of the year. This monthly variability indicates different lithogenic sources of Fe and Ti are likely to influence the atmospheric loading at our sampling station throughout the year. The onset of the dust season on the Australian mainland (October-November, Baddock et al., 2015) may explain part of the summer (dusty) season atmospheric inputs at our kunanyi /Mt Wellington aerosol sampling station, as evidenced by higher Fe/Al (and Ti/Al) ratios in aerosols. On the other hand, other atmospheric sources (locally derived from Tasmania or from long-range transport over the Southern Ocean) with a similar (lower) metal/Al signature than the UCC seem to prevail in our study region during winter. However, the small number of aerosol samples available between May - October in our study does not allow for accurate assessment of trends during the winter period. Much smaller variability was observed for the Th/Al ratio calculated in kunanyi/Mt Wellington aerosols (mean

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Th/Al = 0.00017) across the time-series, with an overall median ratio close to that of the UCC (mean Th/Al = 0.00013) across most of the year except during August and October.

Differences between elemental ratios in soil and in aerosol samples may stem from atmospheric processes occurring during transport between source regions and the sampling site including the preferential settling of denser (e.g., oxyhydroxides) minerals over lighter minerals (e.g., clay), and from the mixing of different lithogenic air-masses during atmospheric transport. Analysis of a large set of soil samples, including more locations across Australia and particularly in Tasmania, as well as high resolution information on wind speed and direction at the sampling site and for the duration of the timeseries is necessary to better assess the relative contribution of different Australian dust sources to the lithogenic particulate loading at kunanyi/Mount Wellington.

3.2 Single tracer lithogenic particle fluxes at kunanyi/Mount Wellington: characteristics and trends

Thorium and Ti are commonly used as tracers of lithogenic atmospheric deposition fluxes as they are almost exclusively derived from lithogenic material and have little reactivity or biological utility in the atmosphere (Boës et al., 2001; Ohnemus and Lam, 2015). While Al may be emitted to the atmosphere by anthropogenic sources, its prevailing source in the offshore atmosphere remains crustal material (Xu and Weber, 2021). Although Fe solubility vary following physico-chemical processes during the atmospheric transport, the soluble Fe fraction remains small compared to the total (mostly refractory) fraction of Fe delivered by dust. Hence, if all four tracers have a unique lithogenic source, the use of a multiple tracer lithogenic flux estimate can reduce the uncertainty associated with the variability of a single metal's concentration due to contamination, deviation from the UCC or secondary atmospheric inputs (Traill et al., 2022).

Table 2. Correlation coefficient (R^2) between tracer concentrations in kunanyi/Mount Wellington aerosols.

	Al	**Th**	**Fe**	**Ti**
Al	1	-	-	-
Th	0.90	1	-	-
Fe	0.87	0.82	1	-
Ti	0.74	0.84	0.83	1

A strong correlation (mostly $R^2 > 0.8$) was found between the total atmospheric concentrations of Al, Fe, Th and Ti in the individual samples (Table 2). The strongest correlation ($R^2 = 0.90$) was found between Al and Th and the weakest correlation ($R^2 = 0.74$) was found between total Ti and Al concentrations in aerosols. Such strong correlations suggest that a common prevailing source may supply all four tracers to kunanyi/Mt Wellington sampling station. Australian soil samples collected in the state of Victoria and analyzed in this study also showed a good correlation between the four lithogenic tracers, with R^2 of 0.97, 0.74 and 0.73 for Fe, Th and Ti when compared to Al (based on Table S4 data). Such correlation was not found for soil samples from South Australia (only 2 soil samples from Western Australia). However, the small number of soil sample analysed in this study (n=4 for Victoria and n=5 in South Australia)) is not sufficient to draw conclusion on the potential origin of metals in kunanyi/Mt Wellington aerosol samples. The NGSA database available from Caritat and Cooper (2011) also shows a strong correlation (>0.70) between Fe and Al measurements in soil samples from New South Wales, South Australia and Tasmania using an aqua regia mineralization and x-ray florescence analysis protocol.

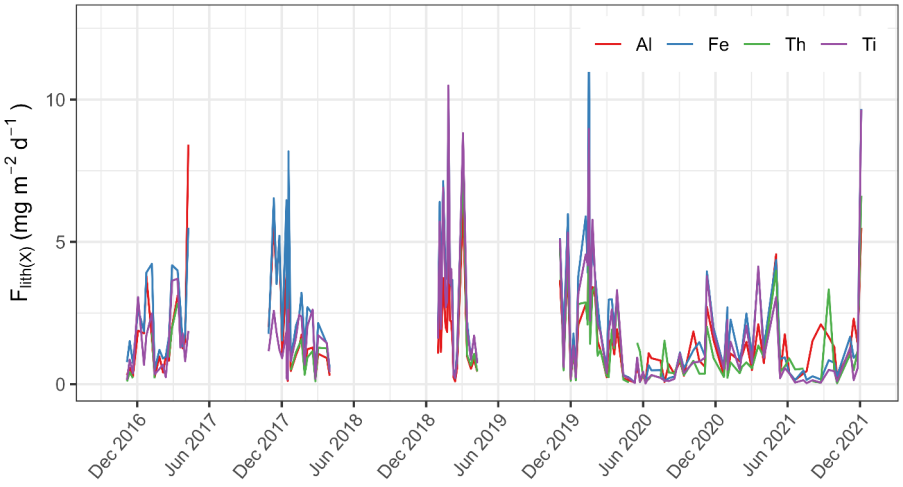

Figure 3. Individual tracer flux, $F_{Lith(X)}$ (mg m^{-2} d^{-1}), at the kunanyi/Mount Wellington aerosol sampling station from 2016 to 2021. Data points represent each aerosol mid-sampling period. Gaps in the time series are periods where samples were not collected due to logistical limitations (winters) or instrument maintenance. Here, $F_{Lith(X)}$ are calculated using the average UCC content for each metal as reported in McLennan (2001).

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Dust deposition fluxes estimated using individual tracer (Al, Fe, Th, and Ti) concentrations measured in kunanyi/Mount Wellington aerosols, called $F_{lith(X)}$, showed similar variability throughout the time-series (2016-2021, Figure 3). Overall, the smallest $F_{lith(Th)}$ flux was estimated using Th as a single lithogenic tracer, ranging between 0.03 and 7.8 mg m^{-2} d^{-1}. The largest dust flux was obtained using Fe as a lithogenic tracer and ranged between 0.05 and 12.7 mg m^{-2} d^{-1} ($F_{lith(Fe)}$, Figure 3). Lithogenic flux estimates calculated using Al and Ti concentrations in aerosols ranged from 0.06 - 8.4 mg m^{-2} d^{-1} and from 0.03 - 10.5 mg m^{-2} d^{-1}, for $F_{lith(Al)}$ and $F_{lith(Ti)}$, respectively. Despite slight differences found between $F_{lith(X)}$ estimates obtained using different lithogenic tracers, the magnitude of the difference between the highest and lowest $F_{lith(X)}$ estimates varied by only a factor of 2, which reinforces the likelihood of a common prevailing atmospheric source for all four tracers.

This finding corroborates work presented by Traill et al. (2022), where concentrations of all four lithogenic tracers showed similar variabilities in marine sinking particles collected in the subantarctic region of the Southern Ocean south of Tasmania (SOTS station). Similarly, Traill et al. (2022) estimated higher lithogenic fluxes when using Fe as a lithogenic tracer and lower lithogenic fluxes when using Th as a lithogenic tracer (Traill et al., 2022). Median $F_{lith(X)}$ estimates measured at the kunanyi/Mt Wellington sampling site (this study: 1.2, 1.7, 0.8 and 1.1 mg m^{-2} d^{-1} using Al, Fe, Th and Ti as individual lithogenic tracer, respectively) compares well with reported dust deposition fluxes of 1.4 - 5 mg m^{-2} d^{-1} estimated by models in the study region (Jickells et al., 2005; Mahowald et al., 2005; Weis et al., 2024) and other Southern Hemisphere dust fluxes <2.7 mg m^{-2} d^{-1} reported off the coasts of South Africa and South America, away from major dust sources (Menzel Barraqueta et al., 2019). Our flux estimates are smaller than mineral dust deposition estimates of 4.0 - 25.0 mg m^{-2} d^{-1} (based on Ti concentration in aerosols) reported by Strzelec et al. (2020a) in Western Australia, much closer to large Australian deserts.

Overall, maximum $F_{Lith(X)}$ estimates in our study were calculated during austral summer months (roughly December – March). Different metals are observed to dominate the summer $F_{Lith(X)}$ peak each year, with Al showing the highest $F_{Lith(X)}$ flux in summer 2016/17 (8.4 mg m^{-2} d^{-1}), Fe in 2017/18 (8.2 mg m^{-2} d^{-1}) and in 2019/20 (12.7 mg m^{-2} d^{-1}), and Ti in 2018/19 (10.5 mg m^{-2} d^{-1}) and in 2021/22 (9.6 mg m^{-2} d^{-1}). This may be due to variabilities in the nature and composition of

the dominant dust source arriving at the sampling site each year, including the impact of dust-containing fire emissions during the summer seasons 2018/19 and 2019/20 (Perron et al. 2022).

3.3 Multi tracer particle flux

All four tracers (Al, Fe, Th, and Ti) measured in kunanyi/Mount Wellington aerosols showed a strong correlation with one another and a similar variability over time (section 3.1), suggesting that they originated from a single terrestrial source. This supports the approach taken in this study whereby a multi-tracer lithogenic deposition flux, called F_{LithAv}, is estimated by averaging $F_{Lith(x)}$ fluxes obtained using each of the four tracers for each sample. The resulting F_{LithAv} estimated at our station between 2016 and 2021 is displayed in Figure 4 and provides a more robust estimate of deposition flux by smoothing variability between tracers (displayed in Figure 3). Individual and average lithogenic flux estimates ($F_{Lith(x)}$ and F_{LithAv}, respectively) calculated in this study are summarised for individual samples in the supplementary Tables S5 and S6, respectively.

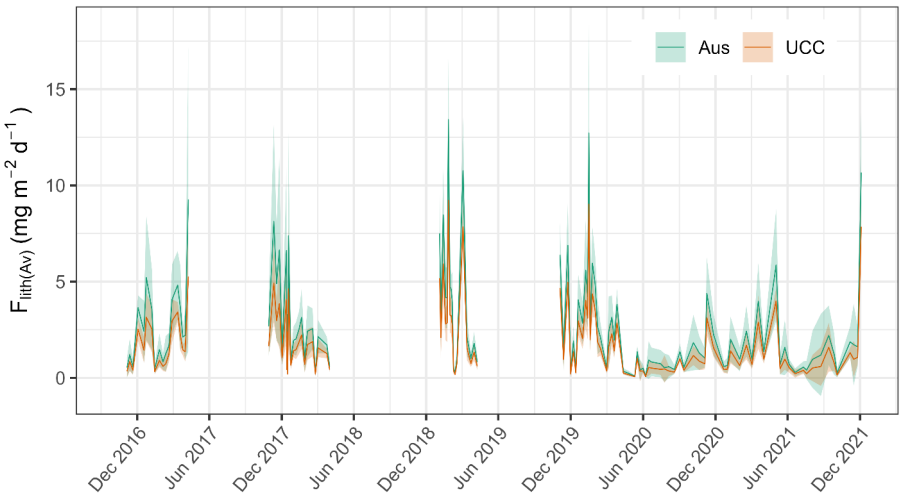

Figure 4. Multi-tracer lithogenic flux estimate, F_{lithAv}, expressed in mg m^{-2} d^{-1}, corresponding to the average of all individual tracer fluxes ($F_{lith(X)}$) calculated based on the lithogenic composition of the UCC (orange colour) and that of the eleven Australian soil samples measured in this study (green colour) . Shadings represent +/- one F_{lithAv} standard deviation of the average (solid lines).

A mean F_{LithAv} value of 1.8 ± 1.3 mg m^{-2} d^{-1} was calculated based on the analysis of aerosol samples collected between 2016 and 2021 at kunanyi/Mt Wellington (Tasmania, Figure 4 orange colour).

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Throughout our time series, the highest F_{LithAv} values were observed in January 2019 and 2020, with flux peaks reaching 9.2 in January 2019 and 9.0 mg m^{-2} d^{-1} in January 2020, respectively. Noticeable peak fluxes of 7.9 mg m^{-2} d^{-1} also occurred in early March 2019 and in mid-December 2021. Extended periods of low F_{LithAv} estimates ($\leqq$ 0.5 mg m^{-2} d^{-1}) were observed during the two austral winter periods sampled, with a minimum flux of 0.06 mg m^{-2} d^{-1} reached in May 2020 (Figure 4). There is therefore an apparent seasonal trend in dust deposited at the kunanyi/Mt Wellington site, with higher F_{LithAv} observed in warmer periods (November - March) and lower fluxes in cooler periods of the year (May - August). It should be mentioned that a mean F_{LithAv} value of 2.7 ± 1.9 mg m^{-2} d^{-1} is estimated when using the average metal content in Australian soil analyzed in this study (Figure 4 green colour). Indeed, while Th and Ti contained in our eleven Australian soil samples show similar concentrations (within 10%) as in the average UCC (McLennan, 2001), Al and Fe concentrations in these local soil samples differ by 52 and 35%, respectively. This result in higher F_{LithAv} estimated using Australian soil data (Figure 4).

The mean F_{LithAv} observed in this study, of 1.8 mg m^{-2} d^{-1} when using the average UCC and 2.7 mg m^{-2} d^{-1} when using the average Australian soil measurement (Table S5), fall within the dust deposition range of 1.1 - 5.5 mg m^{-2} d^{-1} reported by models in southeastern Australia, which account for soil erodibility, soil particle size distribution and wind friction velocity (Albani et al., 2014; Weis et al., 2024). In the Southern Ocean south of Tasmania, smaller mineral dust fluxes of 0.37 mg m^{-2} d^{-1} and 1.0 mg m^{-2} d^{-1} were reported based on aerosol Fe measurements at sea, particle size and surface wind speed (Bowie et al., 2009) and based on Al, Fe, Th and Ti measurements in marine sinking particles (Traill et al., 2022), respectively. Traill et al. (2022) reported a similar annual variability in lithogenic deposition flux at SOTS between 2011 and 2018, with minimum F_{LithAv} around 0.5 mg m^{-2} d^{-1} in July-September and an earlier dust deposition peak (compared to our study) in November-December, up to 2.5 mg m^{-2} d^{-1}. Strzelec et al. (2020a) also reported (up to 6 times) higher mineral dust fluxes in warmer months compared to cooler months based on Ti analysis in aerosols from Western Australia. In particular, the two summer seasons showing F_{LithAv} over 9.0 mg m^{-2} d^{-1} correspond to large bushfire seasons in Tasmania and in Australian mainland upwind from Tasmania, respectively (Perron et al., 2022). Indeed, fire events are known to exacerbate dust entrainment into the atmosphere both during (pyro convective updrafts) and post (burnt ground) fire event (Hamilton et al., 2022). It is worth noting that F_{LithAv} estimated using

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Australian soil measurements (this study) fall closer to the mean reported estimate found in the literature while using the average UCC value result in lower-end F_{LithAv} estimate compared to the literature. While F_{LithAv} estimated using the average UCC may present an advantage in being more comparable with other studies worldwide, F_{LithAv} estimated using Australian soil data may be more relevant for validating model outputs as it likely better represents true deposition fluxes in our study region. Overall, the choice of one or the other crustal source result in up to a factor 2 difference in the calculated $F_{LithAv.}$

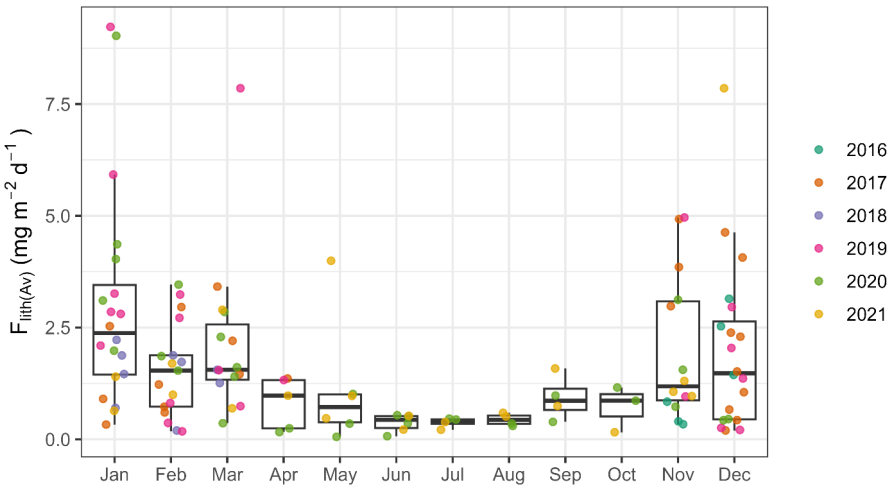

Figure 5. Monthly F_{LithAv} estimates, in mg m^{-2} d^{-1}, based on lithogenic tracer analysis in aerosol samples collected between 2016-2021 at the kunanyi/Mt Wellington site. Individual (weekly) samples are shown as dots and the colour code represents each collection year. Whiskers represent 1.5 times the interquartile range (75^{th} – 25^{th} percentile) beyond the boxes, while the upper, middle, and lower horizontal lines of the box represent the higher interquartile, median value and lower interquartile of the average monthly dataset, respectively.

Greatest F_{LithAv} fluxes are annually observed during the austral summer (December - March), with median F_{LithAv} of 2.4 mg m^{-2} d^{-1} in January and around 1.4 mg m^{-2} d^{-1} in December, February and March across all years (Figure 5). This tendency aligns with higher frequency of dust storms occurring in Australia's main geological basins during warmer months (late austral spring and summer), resulting in higher dust deposition fluxes (O'Loingsigh et al., 2017). The summers of 2017/2018 (Nov-Dec), 2018/19 (Jan-Feb) and 2019/20 (Dec-Feb) had especially high F_{LithAv} fluxes compared to other summer periods in the time-series (Figure 5). These observations are consistent with the year 2017 being identified as the third driest year since

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records have been kept in Australia (Steffen et al., 2018), and the two following summer periods being identified as strong bushfire years, across Tasmania in 2018/2019, and across southeast Australia in 2019/2020 (Perron et al., 2022). Relatively smaller peaks were observed during the summer of 2020/21 and, to a lesser extent, during the 2016/17 summer (Figure 5). This may reflect two wetter summer periods under the influence of El Niño Southern Oscillation positive phase (La Niña), where increased moisture in the topsoil restricted particles from being eroded and entrained by air masses (Bureau of Meteorology, 2022). In addition, fewer bushfire emissions during these two wetter summer periods may have resulted in less dust emissions due to increased vegetation cover on the soil (Bureau of Meteorology, 2022). Wetter summer seasons may also explain a shift in F_{lithAv} peaks towards the end of the summer seasons 2016/17 and 2020/21 (February - March) compared to the December-January $F_{lith(Av)}$ peak observed in 2017/18, 2018/19, and 2019/20 (Figure 5).

4. Conclusions

This study explores the seasonal and interannual variability of the lithogenic deposition flux using analysis of Al, Fe, Th, and Ti (as lithogenic tracers) in aerosol samples collected at kunanyi/Mt Wellington (Tasmania, Australia). Enrichment factors close to 1 and elemental ratios similar to those measured in soil samples collected in Australian dust source regions confirmed the crustal origin of all four tracers. Deposition fluxes, $F_{Llith(X)}$, calculated using each tracer individually showed small differences, of a factor 2 on average, between one another throughout the 2016-2021 time series. This study suggests the use of a multi-tracer (averaged) dust deposition flux estimate as a more robust method to account for variability of individual tracers in aerosols.

The mean F_{lithAv} of 1.8 mg m^{-2} d^{-1} calculated in this study across the 2016-2021 time-series is consistent with earlier lithogenic deposition fluxes reported in the literature. Dust flux maxima (up to 9.2 mg m^{-2} d^{-1}) were consistently observed during the austral summer, while minimum annual F_{lithAv} (down to 0.06 mg m^{-2} d^{-1}) were estimated in the winter. Overall, individual year F_{lithAv} fluxes aligned well with the occurrence of known dust and bushfire events in the summertime as well as other global meteorological events such as El Niño Southern Oscillation (ENSO) which drive the weather patterns across southeast Australia.

Dust deposition fluxes calculated in this study hold some uncertainties -- a factor 3 and a factor 2 due to the use of a set deposition velocity and the assumption of metal abundance as per the average UCC, respectively. While using the averaged UCC to calculate F_{LithAv} may present an advantage

in being more comparable with other studies worldwide, we show that F_{LithAv} estimates (0.09 – 13.4 mg m^{-2} d^{-1}) calculated using metal abundance in Australian soils are in better agreement with dust fluxes reported by global models in our study region. Therefore, understanding local soil composition is essential to estimating dust deposition fluxes in different study regions. Overall, the uncertainty in field-based dust deposition flux estimates likely remain smaller than uncertainties associated with model parametrization.

As dust deposition is now being recognized as a major source of vital micro-nutrients such as Fe to the Southern Ocean, accurately quantifying dust fluxes is vital to understanding primary production in the Southern Ocean and how this may change under future climate scenarios. Our observed dust flux maxima in austral summer may also provide a much-needed regional pulse of nutrients to phytoplankton when water column nutrients are being depleted through the growing season.

Additional observations covering a wider geographical area and greater temporal coverage (including time-series stations and winter sampling periods) are required to better constrain seasonal and interannual variability. Our study adds vital data to the relatively few field-based dust deposition flux estimates available to validate model outputs, especially for Southern hemisphere dust sources (including Australia) which greatly vary in nature and composition.

Author contributions

A.R.B. was responsible for project conceptualisation, funding acquisition, resources and supervision. M.M.G.P. was responsible for part of the sample collection, sample processing, data interpretation, processing and curation as well as for manuscript drafting. S.M was responsible for part of the sample collection and analysis, and for laboratory supervision. T.H was responsible for data curation. C.N was responsible for the analysis of soil samples. C.H was responsible for part of the sample collection, sample processing, data curation and the original draft writing. A.T. was responsible for instrumental analysis. P.dC. was responsible for part of the sample collection and data curation. M.S. was responsible for part of the sample collection and sample processing. All authors were responsible for data interpretation and validation and reviewing and editing the manuscript.

Conflicts of interest

There are no conflicts to declare.

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Acknowledgment to country

Before the white settlement of lutruwita/Tasmania, kunanyi/Mount Wellington was a prominent feature in the lives of the Moomairremener people for thousands of years and continues to be. We pay our respects to elders' past, present and emerging and are thankful to have been able to study this region.

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