Background concentrations of mercury in Australian freshwater sediments: the role of catchment's physico-chemical parameters on mercury deposition

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Abstract

Waterways in the Southern Hemisphere, including on the Australian continent, are facing increasing levels of mercury contamination due to industrialisation, agricultural intensification, energy production, urbanisation and mining. Mercury contamination undermines the use of waterways as a source of potable water and also has a deleterious effect on aquatic organisms. When developing management strategies to reduce mercury levels in waterways, it is crucial to set appropriate targets for mitigation of these contaminated waterways. These mitigation targets could be (1) trigger values or default guideline values provided by water and sediment quality guidelines or (2) background (pre-industrialisation) levels of mercury in the waterway. The aims of this study were to: (1) quantify the differences between existing environmental guideline values for mercury in aquatic systems, and background mercury concentrations, and (2) determine the key factors affecting the spatial differences in background mercury concentrations in freshwater lake systems in Australia. Mercury concentrations were measured in background sediments from 21 lakes in Australia. Organic matter and precipitation were the main factors to explain mercury concentrations in sediments of lakes. These data indicate that background mercury concentrations in lake sediments can vary significantly across the continent, and the background concentrations are up to nine times lower than current sediment quality guidelines in Australia and New Zealand. This indicates that if waterway managers are aiming to restore systems to 'pre-industrialisation' mercury levels, it is highly important to quantify the site-specific background mercury concentration. We found that the geology of the lake catchment correlates to the background mercury concentration of lake sediments, with the highest mercury background levels being identified in lakes in igneous mafic intrusive regions and the lowest in areas underlain by regolith. Taking into account these findings, we provide a preliminary map of predicted background mercury sediment concentrations across Australia that could be used by waterway managers for determining management targets.

Keywords: waterway management, sediment quality guidelines, background conditions, mercury

Introduction

Mercury is a highly toxic compound, especially in its methylated form (Ullrich et al., 2001). Anthropogenic activities such as fossil fuel combustion (Fabris et al., 1999), industrialisation (Navarro et al., 1993), mining (Davies et al., 2018) and agricultural intensification (Denton and Breck, 1981) have led to increasing levels of mercury contamination in aquatic environments. Mercury is currently listed as one of the top ten chemicals or groups of chemicals of major public health concern by the World Health Organisation (WHO 2010). Previous studies of centennial-scale changes in mercury levels in Australian freshwater systems have shown that metal concentrations in sediments had low variability over time prior to European settlement, the development of mines and industrialisation (Lintern et al., 2015). These studies indicate that it was only with European settlement and industrialisation that the concentrations in metals in aquatic sediments started to fluctuate significantly. These pre-industrialisation and pre-European concentrations of metals in aquatic sediments are often assumed to represent 'background' levels of metals in aquatic sediments.

There are many waterways throughout the world where the mercury concentrations in aquatic sediments are elevated compared to pre-industrialisation concentrations (Balogh et al., 1999; Balogh et al., 2009; Cooke et al., 2020). Australian aquatic systems such as, Reedy Creek (Churchill et al., 2004), Lerderderg River (Bycroft et al., 1982), the Gippsland Lakes (Fabris et al., 1999), Molonglo River (Stinton et al., *in press)*, the Connewarre Complex (Reeves et al., 2016) and Newell Creek (Schneider et al., 2019), all show elevated concentrations of mercury in post-industrialisation sediments. This is of concern due to the toxic impacts of elevated concentrations of mercury on humans (Castilhos et al., 2006), as well as on biodiversity and ecosystem survival (Selin, 2009). As such, efforts are currently being made to reduce mercury levels in waterways. These methods include: controlling and reducing mercury sources within catchments, increased regulations to reduce atmospheric emissions of mercury, treatment of wastewater, and stabilisation, dredging or capping of mercury-contaminated sediments (Wang et al., 2004; Hylander and Goodsite, 2006; Mathews et al., 2013).

When remediating a polluted aquatic environment using these methods, target sediment and water toxicant levels are required to calculate a pollutant's enrichment factor compared to its background levels and to establish mitigation measures to restoration programs. It is of crucial importance to identify the optimal sediment and water quality guidelines that should be used when restoring waterways affected by mercury contamination in Australia.

Water and sediment quality management frameworks in many parts of the world, including Australia, state that pollution reduction targets should be based on the background conditions of the aquatic system (Canadian Council of Ministers of the Environment, 2003; Water Quality Australia, 2018). Water quality management frameworks also often provide guideline values calculated using eco-toxicity data (the probability of a toxic effect of a certain concentration of the chemical on benthic organisms), which can be used when background levels cannot be quantified (MacDonald et al., 2000; Water Quality Australia, 2018). The current default guideline value (DGV) for mercury in Australia and New Zealand is 150 ng/g. This represents the value below which there is a low risk of toxic environmental effects due to mercury. The guidelines also provide an upper guideline value (GV-high) of 1000 ng/g for mercury, which represents the value above which we are likely to observe some toxic effects of mercury on the

environment (Water Quality Australia, 2018). When the GV-high is exceeded, there is a high probability that the level of mercury in the sediments is toxic to benthic organisms. Therefore, while GV-high threshold values provide an indication of concentrations at which toxicity-related adverse effects are expected to be observed, the DGV threshold value is used for guidance on the remediation of contaminated sediments. Our paper focuses on the lower threshold value, the DGV, and aims to provide a guideline for restoration of polluted sites.

However, we currently have very little understanding of the difference between background conditions of aquatic systems and existing guideline values (Lintern et al., 2016). This is largely because quantitative data of background conditions are unavailable due to a lack of water and sediment quality monitoring until the late 20th century (Alexander et al., 1998). Understanding the quantitative differences between background mercury levels and guideline values provided by water quality guidelines is critical in guiding waterway managers to select the most appropriate water quality and sediment quality targets for remediation projects. The water or sediment quality target selected will depend on the objectives of the project, which could range from restoring the aquatic environment to 'pre-pollution' levels, protecting key aquatic species, to protecting human health.

Therefore, the aim of this study is to fill this gap in our understanding and determine the difference between background mercury concentrations and existing sediment quality guideline values in aquatic systems. We had the additional objective of determining the spatial variability in background mercury concentrations and predicting background mercury concentrations across Australia using our understanding of the key factors driving background mercury concentrations. In this study, we do not intend to assess the health risks of mercury to aquatic environments. Rather, we use sediment cores from 21 Australian lakes as a case study, to identify background levels of mercury across the region (Förstner and Salomons, 1980; Dubois et al., 2017). Lake sediment cores have been used in previous studies to understand long-term historical trends in metal pollution of aquatic environments (Latimer et al., 2003; Lintern et al., 2015). Although a dataset larger that 21 lakes would result in more robust statistical findings, these data provide an initial insight into background mercury levels within Australia and the Southern Hemisphere. Little is known about mercury in Australia, and this lack of evidence for national levels of risk appears to be partly responsible for Australia not ratifying the Minamata Convention (Sinclair and Schneider, 2019). This study is the first step in filling our knowledge gap of background mercury levels in Australia and the Southern Hemisphere, which will provide evidence that may advance the call for ratification of this international convention.

Methodology

Site selection and sediment sub-sampling

Sediment cores with known age-depth models from previously published studies were selected for this study. This included cores from 21 freshwater lakes, the majority of which were located in South-East Australia (Figure 1). The characteristics of these sites have been summarised briefly in Table 1.

From the 21 cores listed in Table 1, one to four sediment sub-samples were obtained from sediments deposited prior to industrialisation and European settlement in Australia in 1788 AD (Powell, 1994). These sediment samples were taken from depths corresponding to approximately 2000, 4000, 6000, 8000 calibrated years BP. For the rest of this study, we use *ka* to represent thousands of calibrated years BP. It should be noted that not all cores had available sediments

corresponding to each of these depths. A detailed inventory of the sub-sampling depth of each core and the approximate ages of the sediment sub-samples are provided in the Supplementary Materials (Table S1). Seventy one sub-samples were collected in total, each having a mass greater than 4 g wet weight.

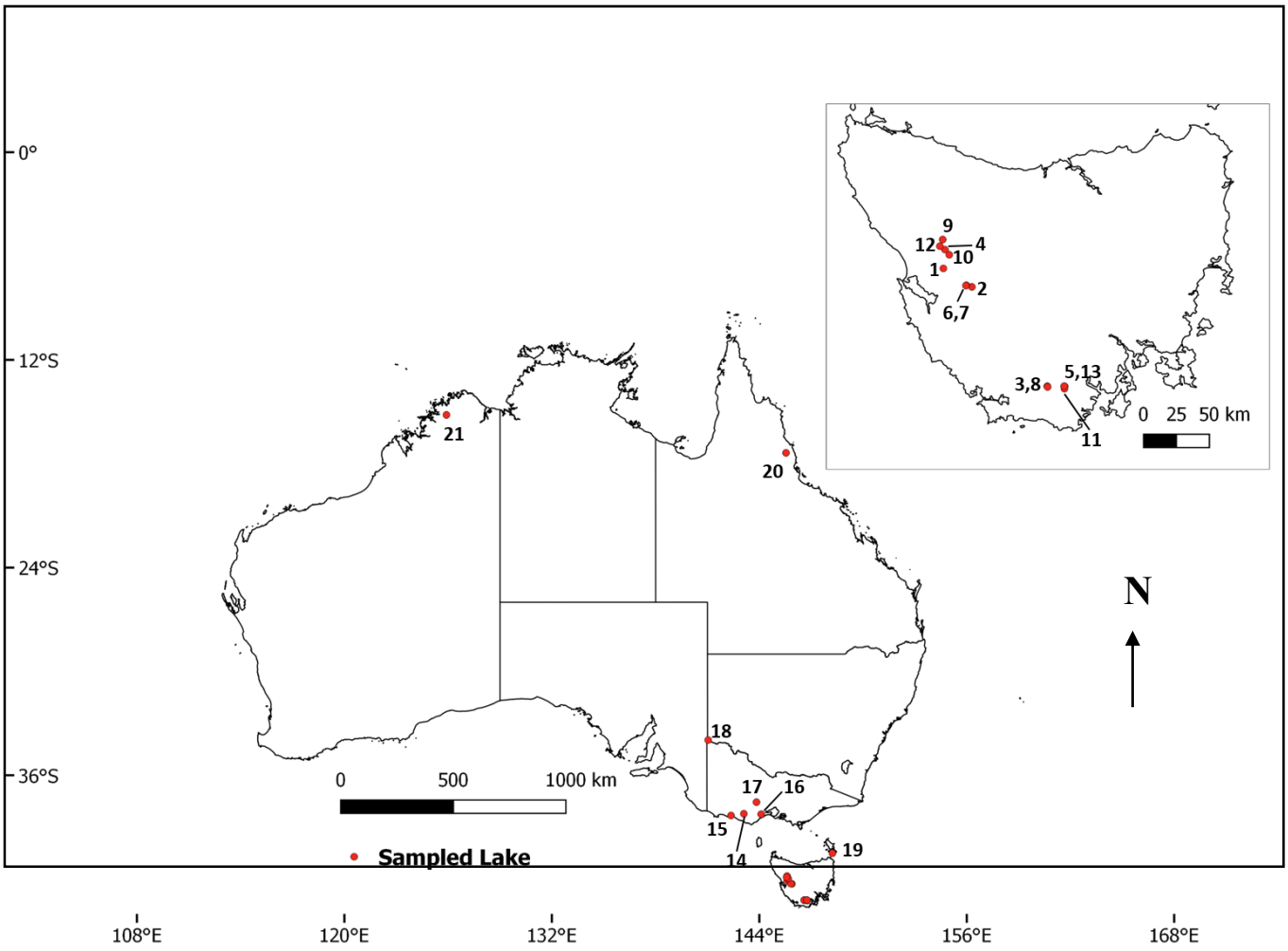

Figure 1: Locations sampled for pre-industrial reference sediments. Insert shows zoomed in map of the state of Tasmania.

Table 1: Published works showing age-depth models of sediment cores sub-sampled for pre-industrial reference sediments

Site Name (State)	Longitude	Latitude	Year cored	Catchment Area (km^2)	Lake water level at time of coring (m)	Corer used	Catchment type	Published age-depth model	Reference Number used to show location in Figure 1
Owen Tarn (Tasmania)	145.60943	-42.09961	2015	76.6	7	Gravity corer	Sub-alpine lake	(Mariani et al., 2019)	1
Vera (Tasmania)	145.87992	-42.27459	2011	5,204	48	Nesje corer Gravity Corer	Sub-alpine lake	(Beck et al., 2019) (Beck et al., 2018)	2

Square Tarn (Tasmania)	146.59422	-43.21431	2012	0.048	3.5	Gravity corer	Sub-alpine lake	(Mariani and Fletcher, 2017)	3
Rolleston (Tasmania)	145.62476	-41.92149	2015	7.59	42	Nesje corer	Sub-alpine lake	(Mariani and Fletcher, 2017)	4
Osborne (Tasmania)	146.75930	-43.21621	2011	0.21	9.5	Gravity Corer	Sub-alpine lake	(Fletcher et al., 2014; Fletcher et al., 2018)	5
Nancy (Tasmania)	145.82712	-42.25877	2011	0.12	24.1	Gravity Corer	Sub-alpine lake	(Fletcher et al., 2015)	6
Gwendolyn (Tasmania)	145.80000	-42.30000	2011	1.09	30	Gravity Corer	Sub-alpine lake	(Fletcher et al., 2015)	7
Burgess (Tasmania)	146.59713	-43.22148	2012	1.62	26.2	Gravity corer	Sub-alpine lake	(Fletcher et al., in preparation)	8
Gaye (Tasmania)	145.60333	-41.82642	2011	1.18	1.2	Gravity corer	Sub-alpine lake	(Mariani and Fletcher, 2017)	9
Isla (Tasmania)	145.66544	-41.97053	2015	1.18	14	Gravity corer	Sub-alpine lake	(Mariani and Fletcher, 2017)	10
Hartz (Tasmania)	146.75656	-43.23809	2012	0.77	40.5	Gravity corer	Sub-alpine lake	(Mariani and Fletcher, 2017)	11
Julia (Tasmania)	145.57614	-41.88923	2012	1.5	12	Gravity corer	Sub-alpine lake	(Mariani and Fletcher, 2017)	12
Perry (Tasmania)	146.75440	-43.21357	2013	0.20	74	Gravity corer	Sub-alpine lake	(Cadd et al., 2019)	13
Lake Gnotuk (Victoria)	143.10300	-38.22100	2016	2.3 (Leahy et al., 2010)	20	Gravity Corer	Volcanic plains Lake	(Fletcher et al. in preparation)	14
Tower Hill Crater (Victoria)	142.36092	-38.32195	2010	1.8 (Leahy et al., 2010)	0.1	Piston Corer	Volcanic Plains Lake	(Mills et al., 2013)	15
Lake Modewarre (Victoria)	144.10529	-38.24558	2009	5.5 (Leahy et al., 2010)	Dry	Piston Corer	Volcanic Plains Lake	(Mills et al., 2013)	16
Lake Wendouree (Victoria)	143.83369	-37.55304	2009	5.5	Dry	Russian D Corer	Volcanic Plains Lake	(Mills et al., n.d.)	17
Tareena Billabong (New South Wales)	141.03924	-33.96555	2001	12.4	1.0m	Russian D Corer	Billabong (Oxbow Lake)	(Gell et al., 2005)	18
Rexy Boy Lake (Tasmania)	148.225	-40.5074	2015	0.0155	0.6m	Russian D Corer	Lacustrine wetland	Hopf et al. in prep)	19
Bromfield Swamp (Queensland)	145.543726	-17.371719	2009	2.72	1.0 m	Piston Corer	Lacustrine wetland	(Burrows et al., 2016)	20
Mitchell River (Western Australia)	125.8997	-15.1763	2012	0.07	1.0m	Piston Corer	Billabong (Oxbow Lake)	(Connor et al. in prep)	21

Analysis of sediments

Sediment sub-samples were taken to the Palaeoworks Lab at the Australian National University and freeze-dried using a FreeZone Plus 6 freeze-drier (Labconco, Kansas City, MO) for 48 hours. Samples were then homogenised and crushed into a fine powder, and 100 mg of the sample was analysed for total mercury concentration using the USEPA method 7473 (US EPA, 1998) and a Milestone Direct Mercury Analyzer (DMA-80 Tricell; Milestone, Bergamo, Italy). The Direct Mercury Analyzer uses thermal decomposition, amalgamation and atomic absorption spectrometry to identify total mercury concentrations in solid materials. A pair of blanks and a pair of certified reference materials (WQB-1 Lake Ontario sediment from the National Water Research Institute in Canada and 2711a Montana II soil from the National Institute of Standards and Technology in the USA) were analysed for every 36 samples. A duplicate sample was run for every three samples, and results for these were within 10% of the original sample and reported as the mean between the duplicates. When the two duplicates had a difference in concentration higher than 10%, a triplicate was run.

All sediment samples were also analysed for additional metals including: aluminium (Al), arsenic (As), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni) and zinc (Zn). These analyses were conducted at ALS in Scoresby, Victoria, a commercial laboratory accredited by the National Association of Testing Authorities (NATA). Sediments were first ground into a fine powder, and then were digested using aqua regia (hydrochloric and nitric acids), and the digested sediments were analysed using inductively coupled plasma mass spectrometry (ICP-MS) following USEPA SW846 (US EPA, 2007). Limits of reporting (LORs) in dry weight are 5 mg/kg. All laboratory blanks were below the detection limit and duplicate runs were within 6.8% difference of each other for all metals. Recovery rates were between 80.7% and 116% for all metals.

Particle size distribution of all sediment samples was determined at the Coastal Lab (University of Melbourne, Parkville, Victoria) using 0.25 g wet weight of the sediment sample. The sediment samples were first digested to remove all organic matter by heating the samples and slowly adding 30% Hydrogen Peroxide until all organic matter was removed. 1 mL of tetra-sodium pyrophosphate decahydrate was then added to disperse each sample and samples were ultrasonicated for 30 minutes. The particle size distribution of sediments was then analysed in a Beckman Coulter LP 13320, ensuring an obscuration of at least 7%.

Total organic carbon of the sediment samples were determined by weight loss on ignition as described by Wang et al. (2011). After freeze-drying samples, shells and other visible organic materials were removed. Sediment was weighed to approximately 1 g per sample and then heated in a muffle furnace (LABEC, model CEMLL) at 550°C for 8 hours. After 12 hours, samples were allowed to cool to room temperature and then weighed again.

Data analysis

We calculated the median, minimum and maximum concentrations of the pre-industrial mercury for each lake. These distributions were then compared to sediment quality guideline values (default guideline values; DGVs) in Australia

(Water Quality Australia, 2018) to quantify the differences between background mercury concentrations and the DGVs.

We attempted to explain the spatial variability in background mercury concentrations by identifying relationships between background mercury concentrations and lake catchment characteristics. The lake catchment characteristics assessed included: catchment area, elevation, current average annual temperature, current average annual rainfall, lithology and age of underlying geological bedrock. These catchment characteristics were obtained from national spatial datasets (Geoscience Australia, 2011; Geoscience Australia, 2012). For all categorical catchment characteristics, we used the Kruskal-Wallis chi-squared test (α=0.05) to assess whether statistically significant differences in background mercury concentrations were explained by catchment characteristics. For numerical catchment characteristics, the Spearman Correlation Coefficient (Spearman, 2010) (α=0.05) was used to assess whether there was a statistically significant relationship between the catchment characteristic and normalised background mercury concentration. These analyses were completed in RStudio Version 3.5.2 (RStudio Team, 2015).

Finally, we developed predictions of background mercury concentrations in waterways across Australia. We used the statistical distributions of background mercury concentrations from each geological formation for which background sedimentary mercury concentrations were available. We log-transformed all background mercury concentrations and then used the Shapiro-Wilk test (α=0.05) to verify that the log-transformed data were normally distributed for each geological formation. We then determined the mean, 2.5th and 97.5th percentiles of the distributions of background mercury concentrations for each geological formation.

Mapping

A map with the expected mercury concentrations across Australia was produced using Arc Map 10.6.1. The Join Feature tool was used to combine a layer of Australian lithology (Geoscience Australia, 2012) with the 2.5th, 50th and 97.5th percentiles of mercury background distribution calculated for the four lithologies related to the lakes in this study: igneous mafic intrusive, igneous mafic extrusive, regolith and sedimentary siliciclastic. Areas where lithological data were not available were left blank.

Results and Discussion

Background mercury concentrations in sediments compared to sediment quality guidelines

Background mercury concentrations in the 21 Australian lakes ranged from 15.6 ng/g to 249 ng/g. Default guideline values (DGVs) for mercury derived using eco-toxicological data are 150 ng/g in Australia (Water Quality Australia, 2018). As such, in some Australian lakes, background mercury concentrations were considerably lower than the existing DGVs. Indeed, Figure 2 indicates that 19 out of 21 lakes had median background mercury concentrations that were less than the existing DGVs derived from eco-toxicological data. These results indicate that existing sediment quality guidelines in Australia can overestimate Australian background sediment mercury concentrations by at least nine times.

Two lakes (Hartz and Perry, all located in Tasmania) had median background mercury concentrations higher than the default mercury guideline values. This suggests that under current guideline values, these lakes would potentially be

considered contaminated even prior to industrialisation. This indicates the importance of identifying the background sediment concentrations of aquatic systems, and using these to help determine restoration targets.

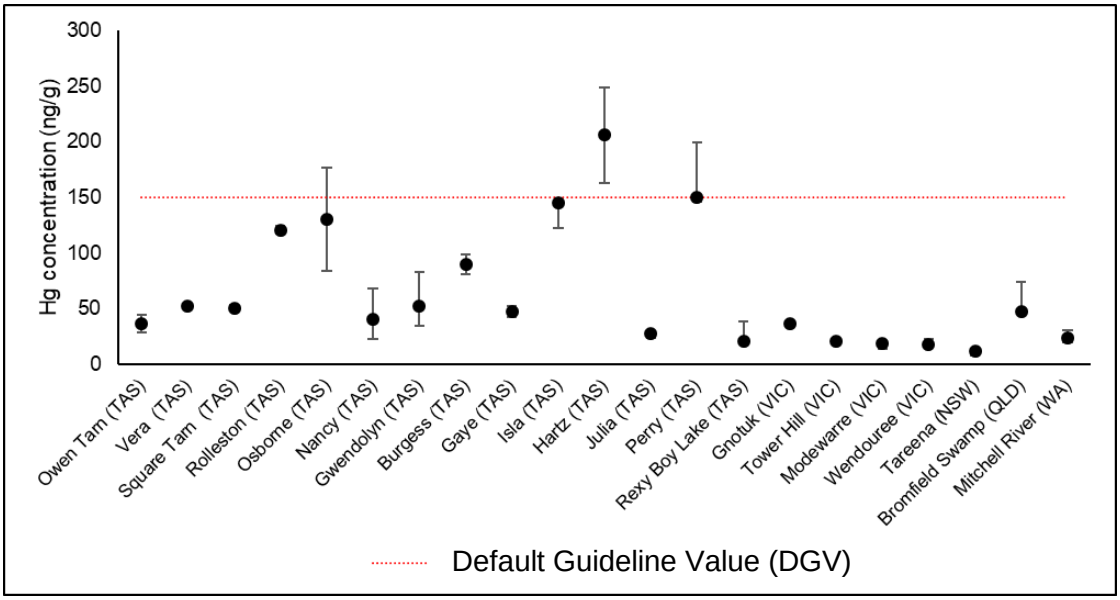

Figure 2: Mercury concentrations (ng/g) in pre-industrial sediments compared to the Australia New Zealand sediment quality guidelines (Water Quality Australia, 2018).

Background concentrations of other metals (As, Cr, Cu, Pb, Ni and Zn), were mostly below the detection limit of 5 mg/kg, so no data analyses were performed with these data (presented in Supplementary Table S1).

Variability in background sediment mercury concentrations

As indicated in Figure *3*, there is considerable spatial variability in background mercury concentrations in lakes across Australia. Background mercury concentrations ranged from 12 ng/g at Tareena Billabong in New South Wales to 206 ng/g at Hartz Lake in Tasmania.

From the 21 lakes studied, the spatial variability is considerably greater than the temporal variability in background mercury concentrations. The spatial variability in background mercury concentrations (represented by the mercury concentration range divided by the median) is 433% between the 21 lakes, but ranges from 5% to 114% for the temporal variability (also represented by the mercury concentration range divided by the median) in background concentrations. In Figure *3*3, black dots represent the background median values (calculated using the samples taken from 2 ka to 8 ka). The error bars represent the ranges (i.e., temporal variability) in background mercury concentrations between 2 ka and 8 ka for each site. This graphical representation demonstrates that, between 2 ka and 8 ka, spatial variability in background concentrations is greater than temporal variability. This agrees with previous works indicating consistent background levels of metals in lake cores prior to European settlement and industrialisation of Australia (Hollins et al., 2011; Lintern et al., 2015). Note that lakes from mainland Australia have less variability of mercury concentration than sites in Tasmanian.

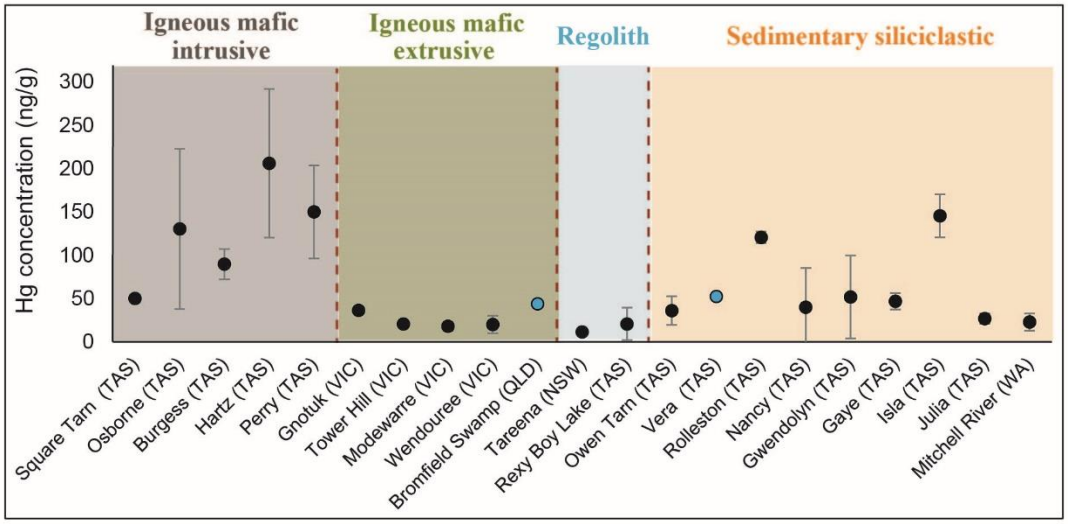

Figure 3: Comparison of spatial vs temporal variability in mercury concentrations of 21 freshwater lakes in Australia. Black dots represent the background medians (calculated using the samples taken from 2 ka to 8 ka). Blue dots represent lakes with only one background sample available, represented in the graph by the original mercury concentration instead of the median). The error bars represent the ranges in background mercury concentrations between 2 ka and 8 ka for each site. (All measured mercury concentrations provided in Supplementary Materials Figure S1). Sites are grouped by catchment's geological formation.

Relationship between background mercury concentrations and catchment characteristics

There is a strong positive correlation between mercury concentrations and organic matter (ρ=-0.90, $p<0.05$), as well as a positive correlation between mercury (ρ=-0.56, $p<0.05$), elevation and rainfall (ρ=-0.32, $p<0.05$) (Figure 4). Sediment grain size and catchment evapotranspiration and temperature are not significantly correlated to spatial differences between the background mercury concentrations (Figure 4). The high correlation of mercury with organic matter agrees with previous studies (Bengtsson and Picado, 2008; Chakraborty et al., 2015). As for other locations worldwide, mercury in these 21 freshwater catchments of Australia is highly influenced by reduction and complexation with organic matter (Chakraborty et al., 2015), which plays an important role on spatial differences in mercury concentrations. The correlation between rainfall and elevation can be explained by the process of wet deposition in which mercury aerosol and reactive gaseous forms of Hg(II) (RGM) are efficiently scavenged by precipitation events, facilitating the deposition of atmospheric mercury to the earth surface and freshwater catchments (Guentzel et al. 2001). Elevation is correlated to mercury in this study due to its correlation with rainfall. The highest lakes in this study are located in Tasmania, one of the wettest regions in Australia (BOM, 2019).

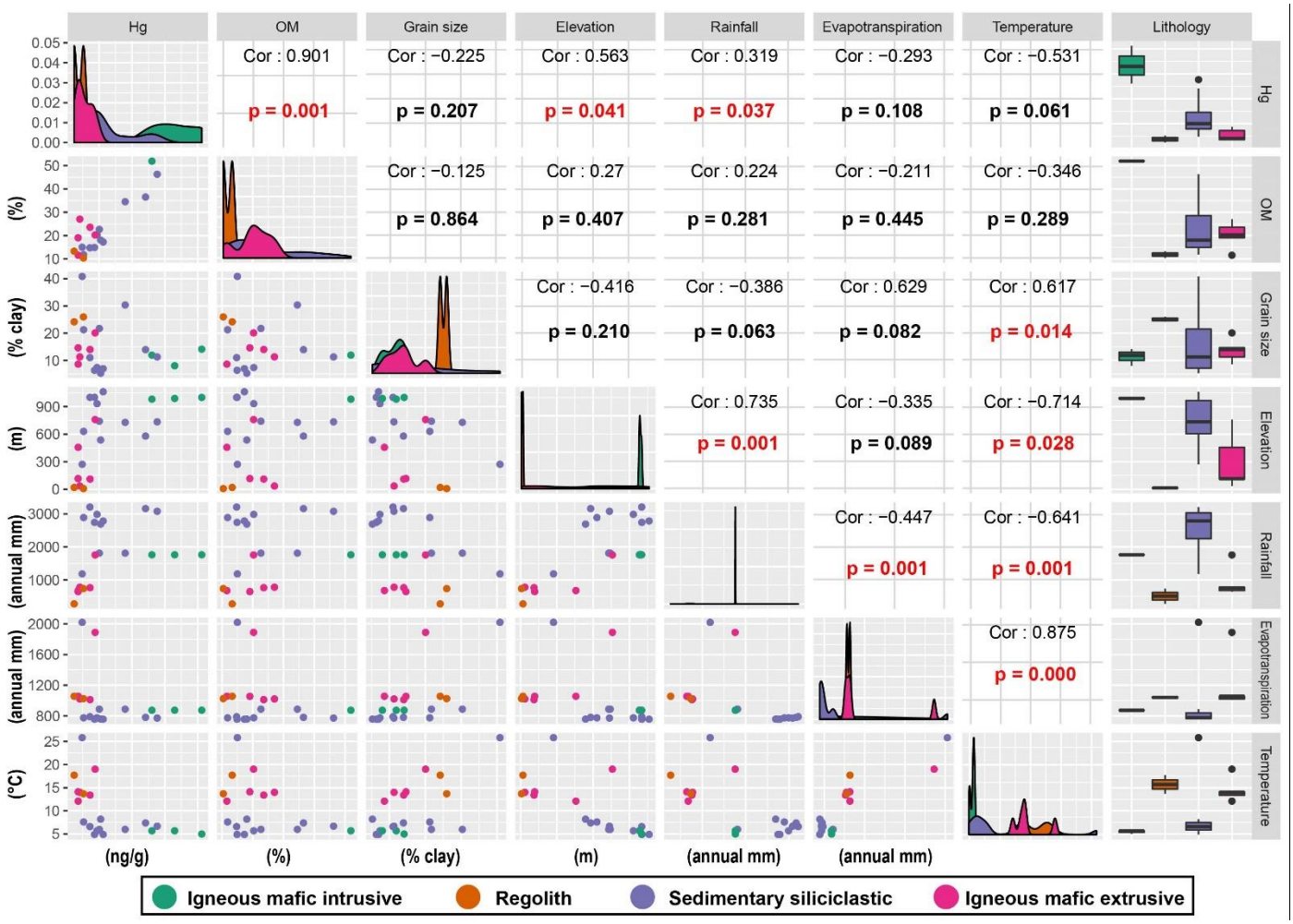

Figure4: Relationship between background mercury concentration (ng/g), organic matter (%),grain size (% clay), elevation (m), rainfall (mm), evapotranspiration (mm) and temperature (°C) in sediments prior to European settlement .

A Kruskal-Wallis H test showed mercury concentrations were significantly different depending on the catchment lithology of the lakes $\chi^2(2) = 12.818$ p = 0.014 (Figure5). Post hoc comparisons using the Bonferroni test indicated that 'igneous mafic intrusive' was the catchment rock type with highest mercury concentrations, significantly different from 'sedimentary siliciclastic, regolith' and 'igneous mafic extrusive' rock (p = 0.35, p=0.025, p = 0.13 respectively). There was no significant difference in mercury concentrations among Siliciclastic, regolith and igneous mafic extrusive rock formations (p> 0.05).

The largest mercury concentrations were measured in igneous mafic intrusive formations, and the lowest concentrations in the regolith. The higher mercury concentrations in the igneous mafic intrusive formations is a result of mercury being sourced from volcanic activities (Gustin et al 2000). The most important and efficient natural source of mercury on Earth is by far volcanic activity, which liberates mercury via quiescent degassing events that overwhelm the atmospheric budget of mercury (Higheras et al 2013). Thus, igneous mafic intrusive formations in Australian freshwater catchments play important roles on mercury concentration in freshwater lakes in the country.

We suggest the use of this link between lithology and background mercury concentrations in lake sediments as a useful way to predict background mercury concentrations in lakes where background sediment samples are not available. In addition, this assessment of catchment lithology and distribution of mercury may identify potential

hotspots for natural mercury emissions in Australia. Currently there is no published material available on natural emissions of mercury from soils in the country. The link between rock formation and mercury concentrations provided in this study is the first step to understand the natural mercury emissions in the country and provides guidance to future studies on soil emission fluxes. This is particularly the case in mercury emissions from fire as bushfires promote volatilisation of mercury in soil (Schneider et al 2019).

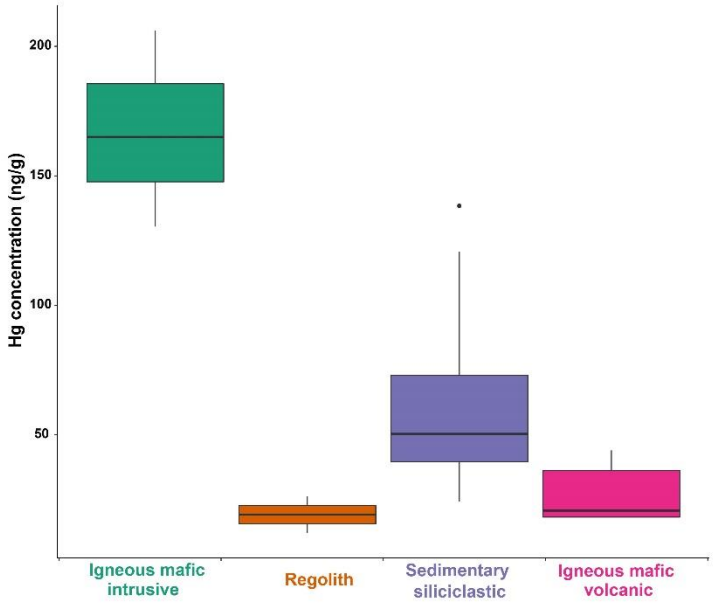

Figure5: Background mercury concentrations (ng/g) in 21 freshwater lake sediments in Australia, plotted by catchment lithology.

Predicted background mercury concentrations across Australia

Predicted background mercury concentrations (log-normalised) across Australia are presented in Figure . Regions across eastern Tasmania and north-western Australia are likely to present the highest levels of background mercury concentrations in aquatic sediments (with a mean of 4.3 ng/g), with the lowest background mercury concentrations likely in areas near the east coast (with a mean of 1.02 ng/g). This is a concern for mercury management in Tasmania considering that several current and legacy mining sites, which mobilise mercury from the earth's crust to the atmosphere, are located in this area of the country (Unger et al., 2012).

Whilst these maps could be used as a preliminary guide when determining background mercury concentration for water quality and sediment quality management purposes, further analysis is recommended to either refine and expand these maps, and validate results with fine-scale analyses. Firstly, as only 21 lakes were used in this analysis, there are several lithologies that were excluded from Figure 5, including sedimentary carbonates, organic-rich rocks, and argillaceous detrital sediments. The inclusion of cores from lakes with these geologies in the analysis could expand the coverage of our predicted background mercury concentrations across Australia. Secondly, some of the distributions of background mercury concentrations are based on as little as 12 data points (e.g., for the igneous mafic intrusive and igneous mafic extrusive formations). The inclusion of a larger number of lake core samples could further refine the statistical distributions available for background mercury concentrations for waterways with these geologies. Further, the risk of mercury contamination to society and the environment suggests a more comprehensive program of

background analyses based on pre-european sediment cores is warranted. Finally, the estimates provided in these maps should not be considered in areas in which cinnabar deposits have been recorded (McQueen, 2011).

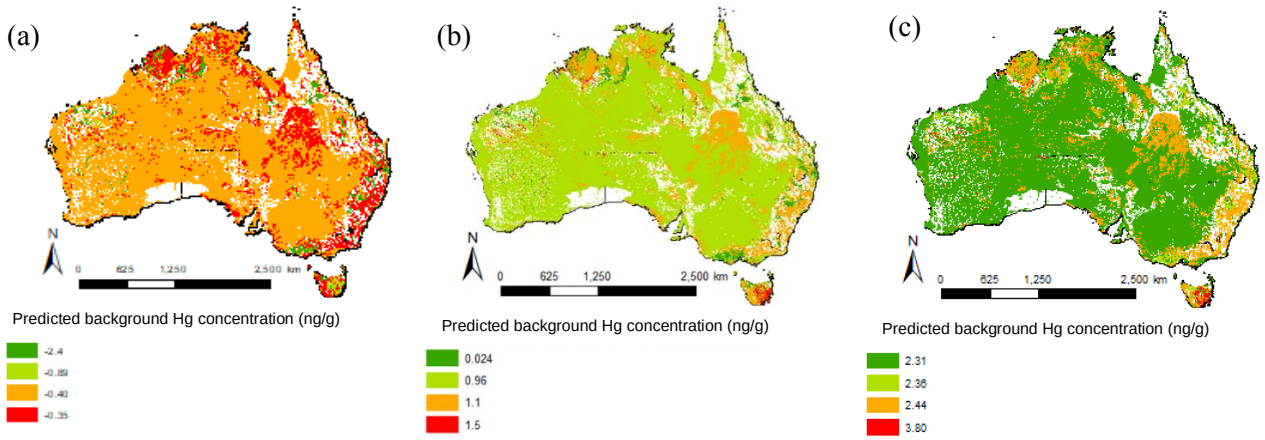

Figure 6: Maps showing the (a) 2.5th percentile, (b) mean and (c) 97.5th percentile of predicted log-normalised background mercury concentrations normalised to grain size across Australia.

Conclusions

This study aims to compare background lake sediment concentrations of mercury in Australia to existing sediment quality guidelines. We defined 'background' lake sediments as those deposited prior to industrialisation, mining and intensification of agricultural activities in Australia in the early 19th century. Mercury concentrations in background lake sediments from 21 lakes across Australia indicate that a large proportion of lake sediments have mercury concentrations that are lower than the Australian sediment quality guidelines. Sediment quality guidelines for mercury were nine times larger than background mercury sediment concentrations. This indicates that, for mitigation or waterway management projects aiming to restore waterways polluted by mercury, 'background' levels should be considered in addition to the existing Australian sediment quality guideline values. An understanding of site-specific background mercury concentrations is necessary for setting these restoration targets. Furthermore, this suggests that adoption of the existing sediment quality guidelines for mercury in Australia should not be used as an absolute value to protect some ecosystem services (e.g. benthic organisms or vegetation) native to the local region. Studies have reported that mercury toxicity tolerance is linked to exposure history. Flora and fauna that live in high mercury concentration sites are able to tolerate higher dosages of mercury than those that have no previous exposure (Chang, 1977, Singh, 2005). In this context, the DGV value should be used more carefully at sites with lower background mercury concentrations.

We recognise that determining background mercury concentrations for every waterway in Australia is not feasible. Using our understanding of the background mercury concentrations in 21 lake systems across Australia, we have identified that these background concentrations are mainly driven by organic matter, precipitation and underlying geology of the region. We have produced maps that predict background mercury concentrations, based on geology, across Australia. These maps offer an initial baseline against which current sediment mercury concentrations can be

compared. Further studies using a greater number of sites, particularly from northern and western parts of the continent, would provide more robust predictions of background mercury concentrations in waterways. We recommend that future researchers and the Australian government work together to develop a coordinated program of analysing pre-European sediments to improve the robustness of this map. Regardless, this study provides the first inventory of mercury concentrations in sediments in Australia that could be used as restoration targets in remediating Australian waterways with elevated mercury levels. Furthermore, the results of this study provide baseline guidance for future mercury studies in the freshwater lake systems in Australia.

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Contributions

Study Conception and design: AL, MM

Data acquisition: AL, LS, SH, KB, MM, MSF, PG

Data analysis and interpretation: AL, LS, KB, MM

Article drafting and revision: AL, LS, KB, MM

Approved the submitted version for publication: AL, LS, SH, KB, MM, MSF, PG

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Supplemental material

Table S1: Background mercury concentrations used to calculate median and range of background mercury concentrations, and other metals measures in 21 lake sediments in Australia.

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Data accessibility statement

All data generated in this study are available in the supplementary materials.

Figures

Figure 4: Locations sampled for pre-industrial reference sediments. Insert shows zoomed in map of the state of Tasmania.

Figure 5: Mercury concentrations (ng/g) in pre-industrial sediments compared to the Australia New Zealand sediment quality guidelines (Water Quality Australia, 2018).

Figure 6: Comparison of spatial vs temporal variability in mercury concentrations of 21 freshwater lakes in Australia. Black dots represent the background medians (calculated using the samples taken from 2 ka to 8 ka). Blue dots represent lakes with only one background sample available, represented in the graph by the original mercury concentration instead of the median). The error bars represent the ranges in background mercury concentrations between 2 ka and 8 ka for each site. (All measured mercury concentrations provided in Supplementary Materials Figure S1). Sites are grouped by catchment's geological formation

Figure4: Relationship between background mercury concentration (ng/g), organic matter (%),grain size (% clay), elevation (m), rainfall (mm), evapotranspiration (mm) and temperature (°C) in sediments prior to European settlement .

Figure5: Background mercury concentrations (ng/g) in 21 freshwater lake sediments in Australia, plotted by catchment lithology.

Figure 6: Maps showing the (a) 2.5th percentile, (b) mean and (c) 97.5th percentile of predicted log-normalised background mercury concentrations normalised to grain size across Australia.

Tables

Table 1: Published works showing age-depth models of sediment cores sub-sampled for pre-industrial reference sediments

Figure 1

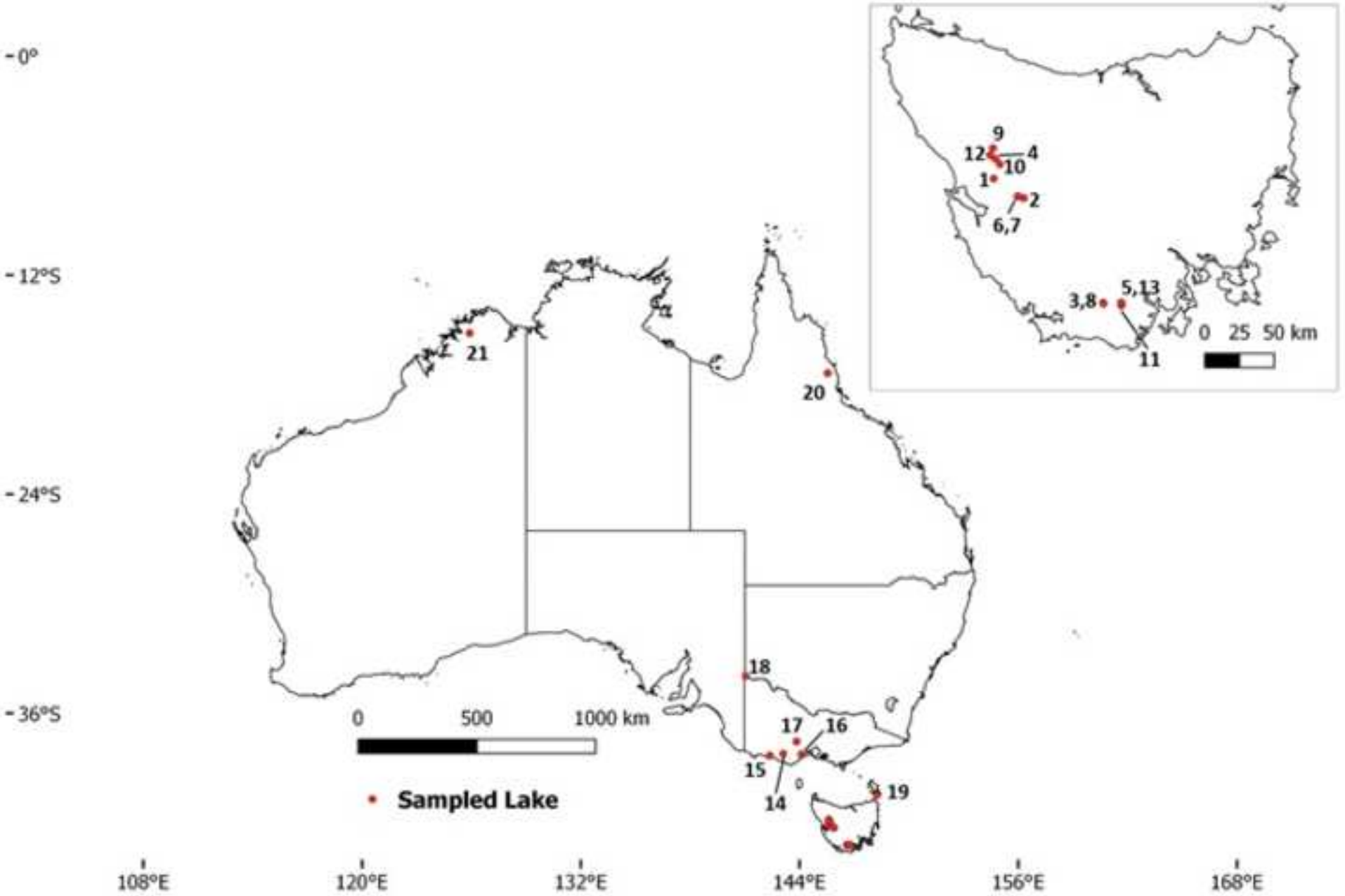

0°
12°S
24°S
36°S
108°E
120°E
132°E
144°E
156°E
168°E
0 500 1000 km
Sampled Lake
1
2
3,8
4
5,13
6,7
9
10
11
12
14
15
16
17
18
19
20
21
0 25 50 km

Figure 2

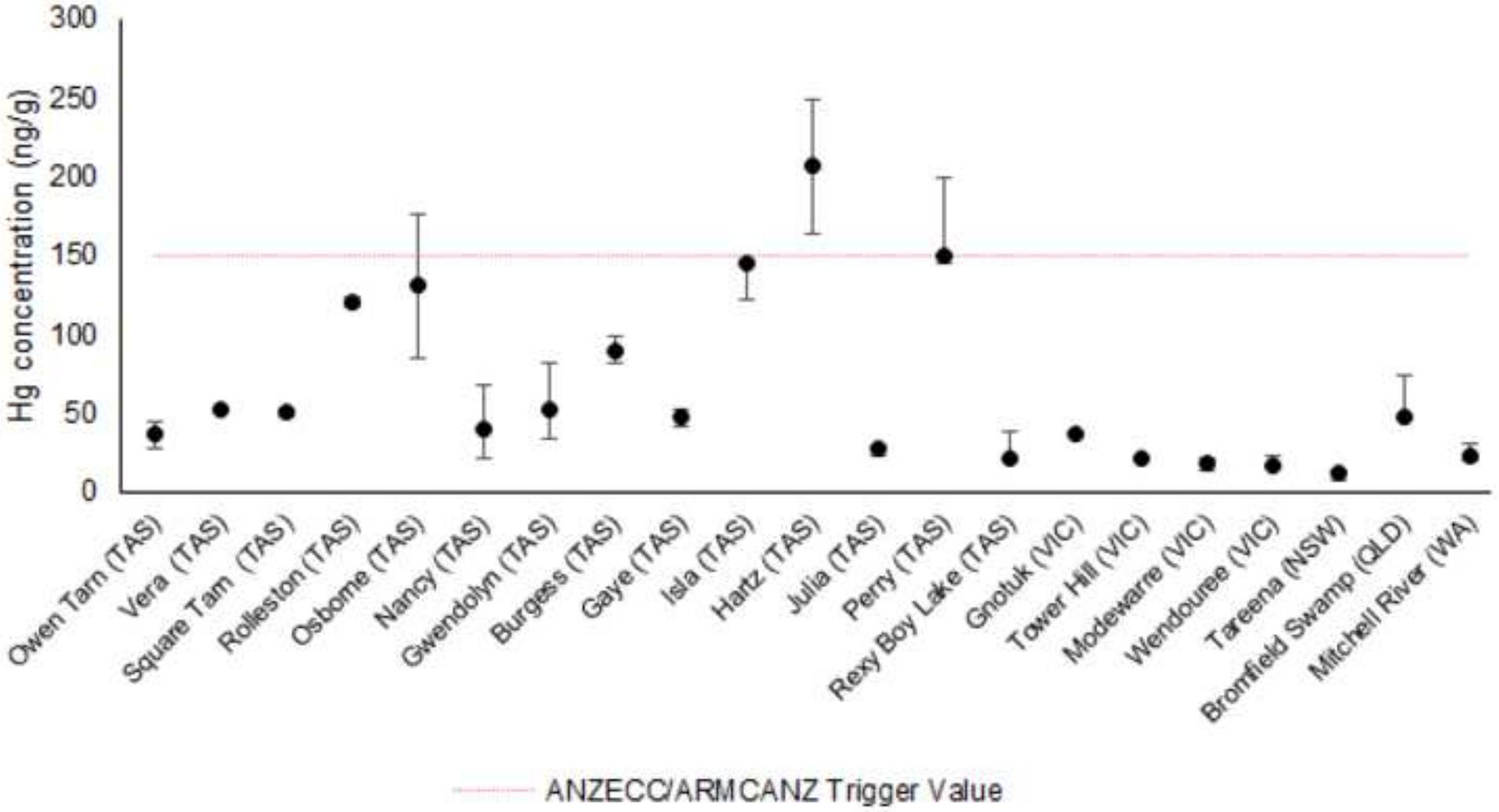

Hg concentration (ng/g)
300
250
200
150
100
50
0
Owen Tarn (TAS)
Vera (TAS)
Square Tarn (TAS)
Rolleston (TAS)
Osborne (TAS)
Nancy (TAS)
Gwendolyn (TAS)
Burgess (TAS)
Gaye (TAS)
Isla (TAS)
Hartz (TAS)
Julia (TAS)
Perry (TAS)
Rexy Boy Lake (TAS)
Gnoluk (VIC)
Tower Hill (VIC)
Modewarre (VIC)
Wendouree (VIC)
Tareena (NSW)
Bromfield Swamp (QLD)
Mitchell River (WA)
ANZECC/ARMCANZ Trigger Value

Figure 3

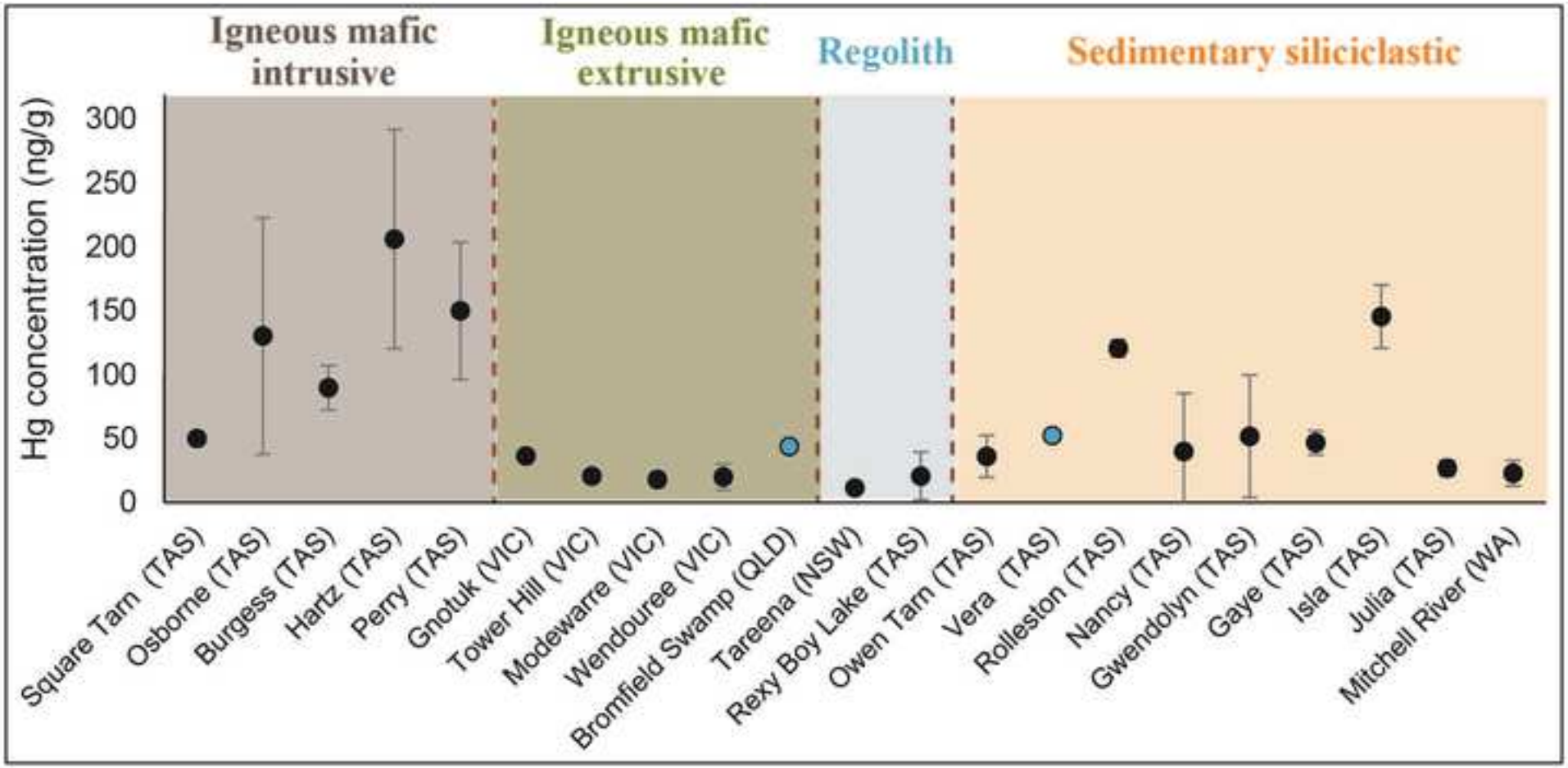

Igneous mafic intrusive
Igneous mafic extrusive
Regolith
Sedimentary siliciclastic
Hg concentration (ng/g)
300
250
200
150
100
50
0
Square Tarn (TAS)
Osborne (TAS)
Burgess (TAS)
Hartz (TAS)
Perry (TAS)
Gnoluk (VIC)
Tower Hill (VIC)
Modewarre (VIC)
Wendouree (VIC)
Bromfield Swamp (QLD)
Tareena (NSW)
Rexy Boy Lake (TAS)
Owen Tarn (TAS)
Vera (TAS)
Rolleston (TAS)
Nancy (TAS)
Gwendolyn (TAS)
Gaye (TAS)
Isla (TAS)
Julia (TAS)
Mitchell River (WA)

Figure 4

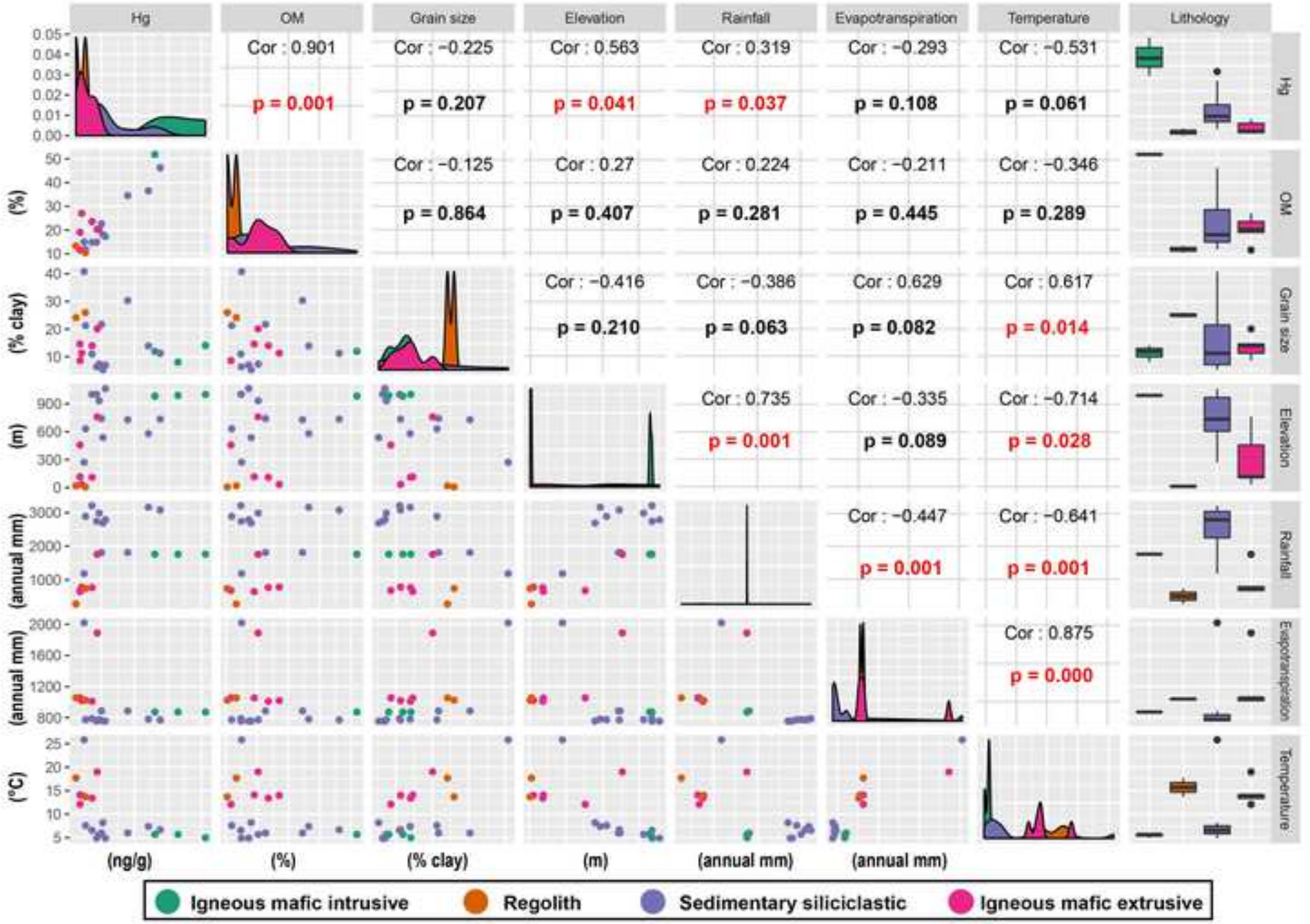

Hg
OM
Grain size
Elevation
Rainfall
Evapotranspiration
Temperature
Lithology
Cor : 0.901
p = 0.001
Cor : −0.225
p = 0.207
Cor : 0.563
p = 0.041
Cor : 0.319
p = 0.037
Cor : −0.293
p = 0.108
Cor : −0.531
p = 0.061
Cor : −0.125
p = 0.864
Cor : 0.27
p = 0.407
Cor : 0.224
p = 0.281
Cor : −0.211
p = 0.445
Cor : −0.346
p = 0.289
Cor : −0.416
p = 0.210
Cor : −0.386
p = 0.063
Cor : 0.629
p = 0.082
Cor : 0.617
p = 0.014
Cor : 0.735
p = 0.001
Cor : −0.335
p = 0.089
Cor : −0.714
p = 0.028
Cor : −0.447
p = 0.001
Cor : −0.641
p = 0.001
Cor : 0.875
p = 0.000
(ng/g)
(%)
(% clay)
(m)
(annual mm)
(annual mm)
Igneous mafic intrusive
Regolith
Sedimentary siliciclastic
Igneous mafic extrusive

Figure 5

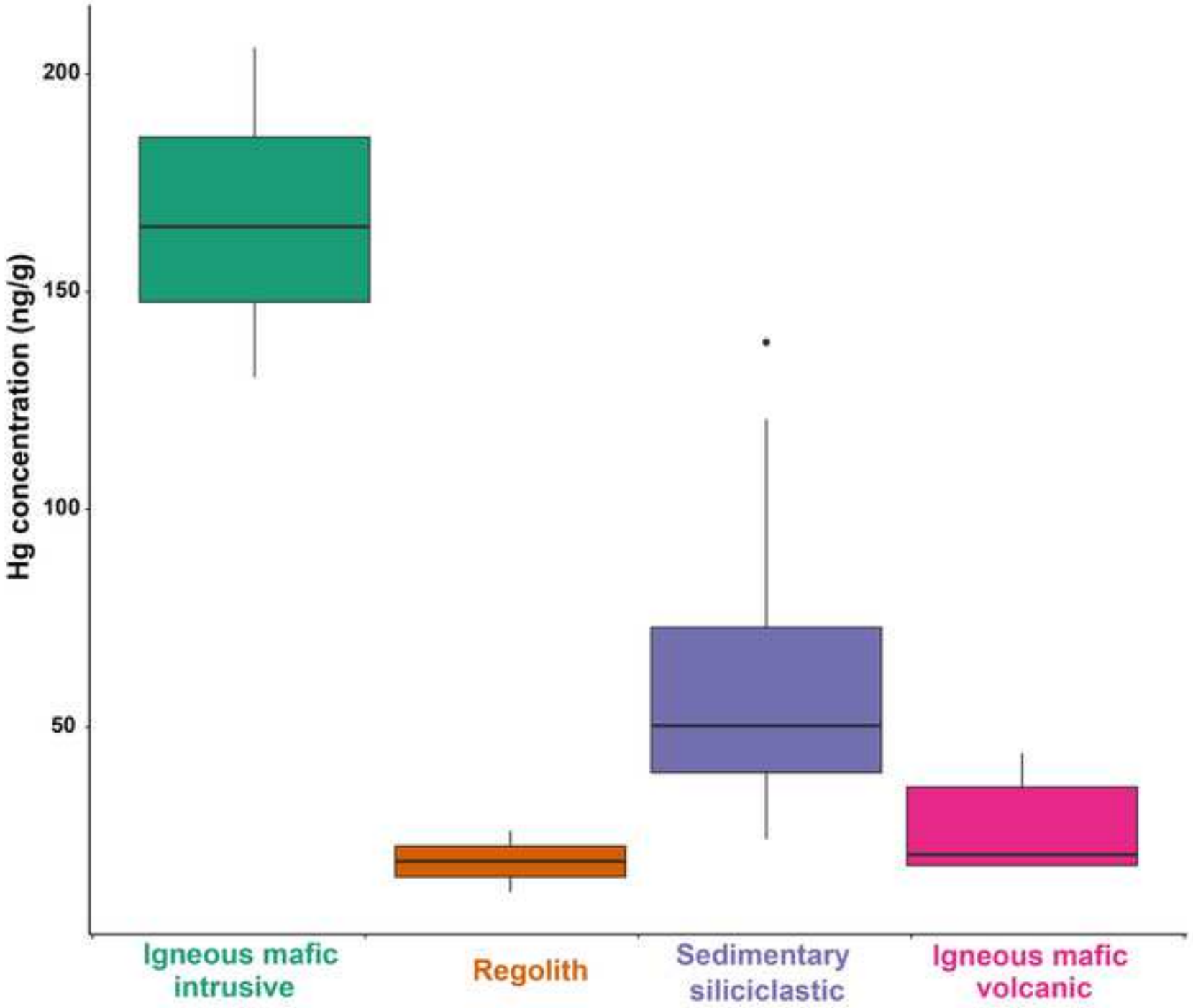
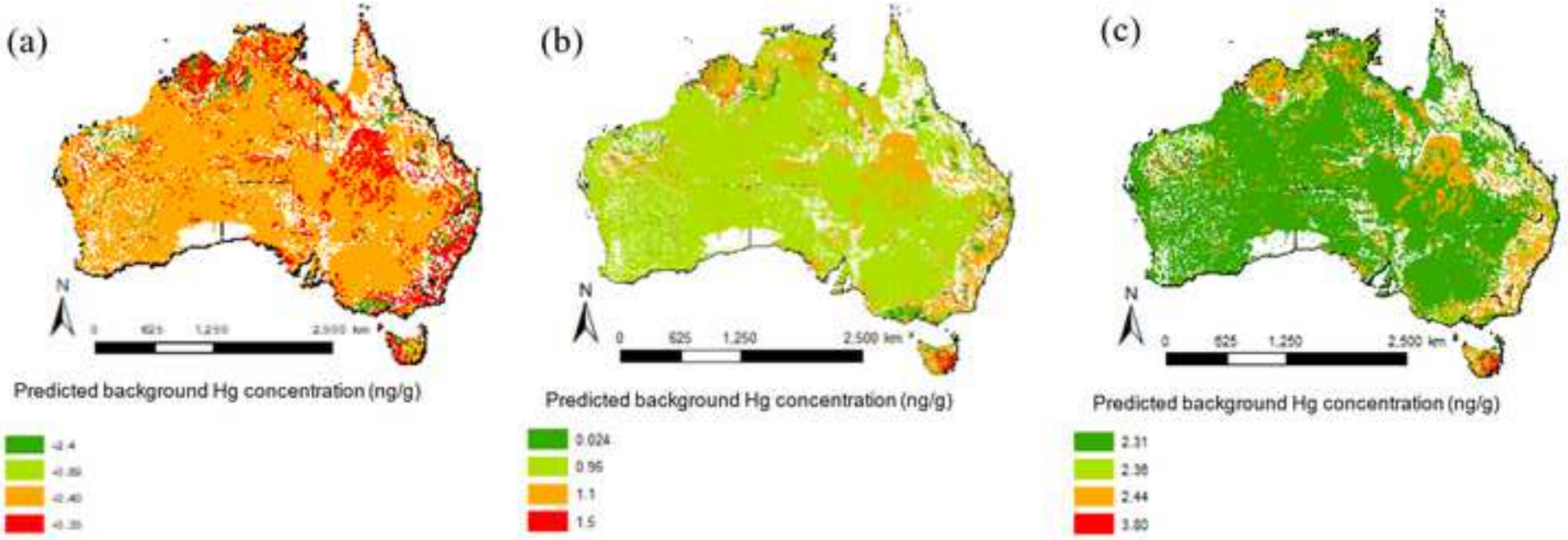

Hg concentration (ng/g)
200
150
100
50
Igneous mafic intrusive
Regolith
Sedimentary siliciclastic
Igneous mafic volcanic

Figure 6


(a)
N
0
625
1,250
2,500 km
Predicted background Hg concentration (ng/g)
(b)
N
0
625
1,250
2,500 km
Predicted background Hg concentration (ng/g)
0.024
0.95
1.1
1.5
(c)
N
0
625
1,250
2,500 km
Predicted background Hg concentration (ng/g)
2.31
2.38
2.44
3.80