



Reframing Anthropocene metal pollution: The overlooked role of colonisation in the Southern Hemisphere

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ARTICLE INFO

Keywords:

Pollution

Colonisation

Anthropocene

Landscape-change

Paleolimnology

Atmospheric-transport

ABSTRACT

Toxic metals and metalloids, including lead, cadmium, and arsenic, have been increasingly mobilised by anthropogenic disturbance. However, interpretations of metal enrichment in the Anthropocene have largely prioritised industrialisation, often overlooking the foundational role of colonisation and its associated socio-environmental transformations. This omission is particularly pronounced in the Southern Hemisphere, where research remains comparatively limited despite the profound and often more direct legacy of colonial extractive activities. As a result, global narratives of environmental change risk underrepresenting the historical drivers shaping contamination patterns outside the Global North. We analysed lake sediment records from a latitudinal transect spanning southeastern Australia, Tasmania, and sub-Antarctic Macquarie Island, regions that experienced relatively recent but intense colonial disturbance, to quantify metal enrichment, accumulation rates, and transport pathways from pre-1850 Common Era (CE) background conditions to the present. Palaeogeochemical records were integrated with an atmospheric transport and deposition model to distinguish atmospheric inputs from catchment-derived processes. Lead exhibits the most well-defined regional signal, with post-1850 CE concentrations up to 4.7 fold higher than background and 1950s enrichment factors up to 13.5 fold greater. All lakes received episodic atmospheric metal inputs, with the Tasmanian site receiving the greatest inputs, however, expression varied with background geochemistry and catchment disturbance. Remote, low-background sites exhibited the greatest relative enrichment. These findings provide robust evidence of long-range atmospheric metal transport linked to colonial-era resource extraction and highlight the need to integrate colonial histories and social context into global assessments of environmental change in the Anthropocene.

1. Introduction

Toxic metals and metalloids (henceforth referred to as metals) including lead (Pb), cadmium (Cd), and arsenic (As) occur naturally in the environment and are released and emitted from energy production and industrial activities (Schneider et al., 2020). Over the last 200 years, colonisation and industrialisation have increased exposure to these

metals (Hawkins et al., 2017; Pérez-Rodríguez et al., 2019). However, the role of colonisation as a driver of environmental contamination is rarely explicitly considered, particularly in the Southern Hemisphere, where research remains disproportionately limited despite the often more direct and measurable legacy of colonial extractive activities (Beck et al., 2020; Schneider et al., 2022; Fisher et al., 2023).

In Australia, metal transport and deposition over the past several

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<https://doi.org/10.1016/j.gecadv.2026.100041>

Received 12 April 2026; Received in revised form 6 August 2026; Accepted 8 September 2026

Available online 18 September 2026

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centuries reflect three major socio-environmental phases, shaped by the country's distinctive historical trajectory. Firstly, European colonisation from the late 18th century rapidly transformed landscapes through land clearing, agriculture, and resource extraction, substantially increasing soil erosion and catchment disturbance (Bradshaw, 2012). Secondly, industrialisation during the early to mid-20th century, occurring later and under different historical circumstances than in Europe, introduced mining, smelting, and fossil fuel combustion, leading to increased anthropogenic metal emissions (Kristensen et al., 2017; Shanahan, 2019). Unique to Australia (and its sub-Antarctic territories, including Macquarie Island), the delayed onset of both colonisation and industrialisation, coupled with the importation of European machinery, technologies, and land-use practices, represents a major driver of environmental change that extends beyond industrialisation alone (Cook, 2019). Thirdly, from the mid-20th century, the Great Acceleration, a period of rapid growth, further intensified industrial activity, energy consumption, and long-range atmospheric metal transport (Waters et al., 2014), collectively driving substantial environmental change across Australia and its territories.

Atmospheric dust transport is a major pathway for metal dispersal in Australasia, with contaminants readily adsorbing to or incorporated within mineral dust particles (Marx et al., 2008). Lead and Cd

originating from Australian sources have been identified in dust samples in New Zealand and Antarctica, indicating long-range atmospheric transport (Marx et al., 2008; De Deckker et al., 2010). Australia hosts numerous mines, smelters, and power stations (Mudd, 2007), many of which operated with limited regulation during the 19th and early 20th centuries, generating substantial metal emissions whose transport pathways and ultimate environmental fate remain poorly quantified (Newell and Watts, 1996; Sun et al., 2022; Schneider et al., 2025).

The dispersion and deposition of metal emissions in Australia are strongly influenced by the Southern Hemisphere westerly winds, which dominate regional atmospheric circulation and facilitate transport from inland and coastal industrial centres to remote environments (Marx et al., 2008; Goyal et al., 2021). Several major legacy smelting operations including Mount Lyell (Tasmania), Port Pirie (South Australia), Broken Hill and Port Kembla (New South Wales), are located in settings conducive to the wide dispersal of emissions and have been significant sources of airborne metal contaminants (Zierer, 1940; Van Alphen, 1999; Martley et al., 2004; Mudd, 2007; Schneider et al., 2025).

We investigate the transport of the toxic metals Pb, Cd, and As from Australian sources to a sub-Antarctic Island. We use lake sediment records collected along a latitudinal transect from New South Wales in continental Australia to sub-Antarctic Macquarie Island in the Southern

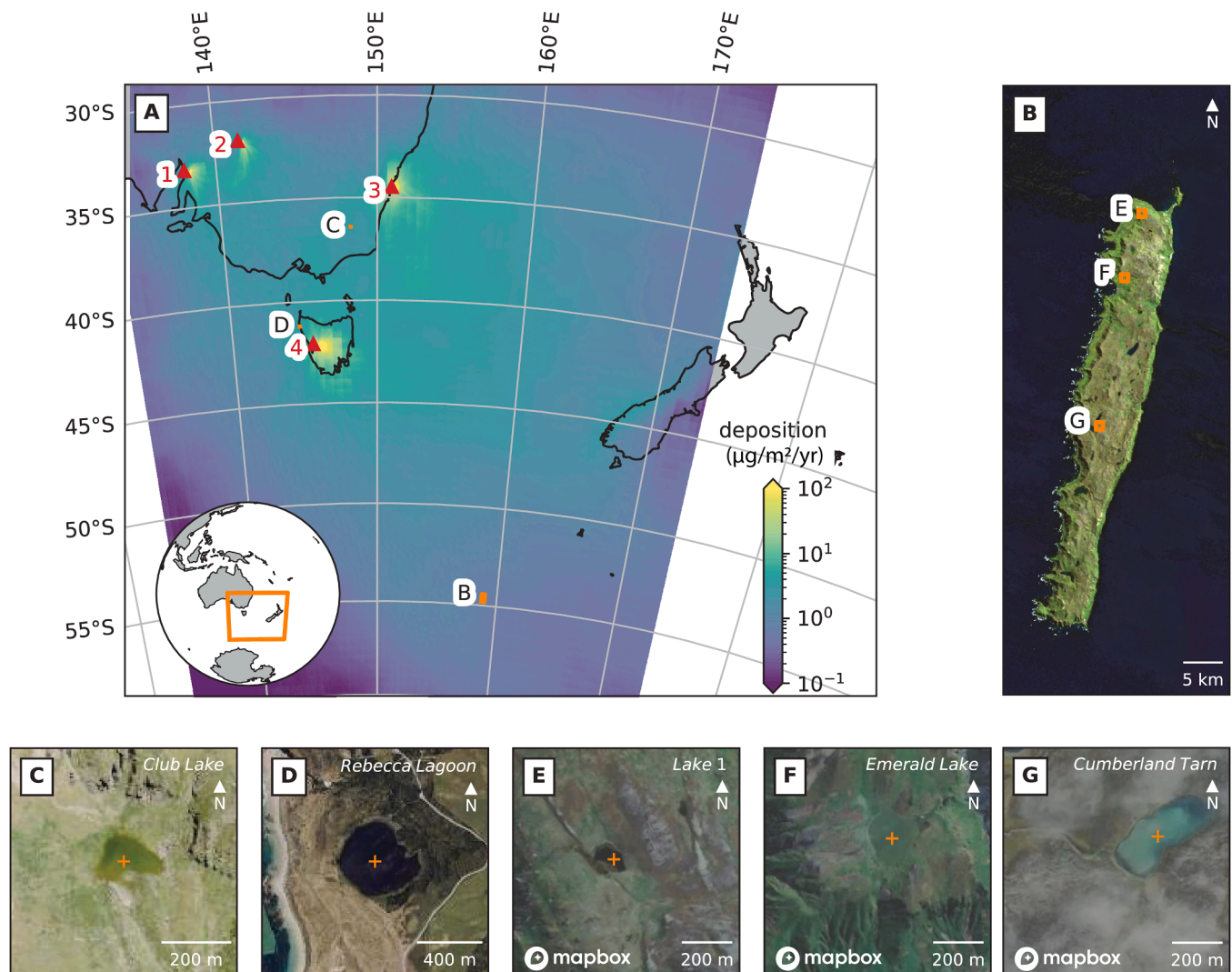


Fig. 1. A Major smelters (1 Port Pirie, 2 Broken Hill, 3 Port Kembla, 4 Mount Lyell) and modelled one-year total deposition ($\mu\text{g}/\text{m}^2/\text{yr}$) of atmospheric lead assuming that each smelter's emissions are constant at 1 kg /hr; B Macquarie Island (imagery acquired by the Operational Land Imager-2 on Landsat 9, February 8, 2022); C–G lake sediment sites (imagery in C is "NSW Imagery" from NSW Department of Customer Service - Spatial Services, CC BY; imagery in D is the "State aerial photo" dataset from the LIST © State of Tasmania, CC BY; imagery in E,F,G is © Mapbox, © OpenStreetMap).

Ocean (Fig. 1) to assess the extent to which atmospheric metal contamination emitted from mainland Australia and Tasmania has reached remote sub-Antarctic islands. We examine changes from pre-1850 CE (Common Era) background conditions through major socio-environmental phases, including colonisation (late-18th to early-19th century), industrialisation (early-20th century), and the Great Acceleration (mid-20th century), to evaluate anthropogenic influences. An additional 14 elements were analysed to characterise sediment dynamics and distinguish atmospheric from catchment-derived inputs. Finally, we integrated our geochemical records with an atmospheric transport and deposition model to assess source contributions and long-range transport pathways beyond what sediment records alone can resolve.

2. Study sites

Study sites were selected along a transect from southeastern continental Australia, Tasmania, and Macquarie Island in the Sub-Antarctic (Fig. 1). The lakes vary in proximity to pollution sources, yet none have received direct input. Club Lake, the northernmost lake located in southeastern Australia (36.4130° S, 148.2910° E) in Australia's alpine region, is 1955 m above sea level. Rebecca Lagoon lies in northwest Tasmania (41.1911° S, 144.6950° E), at 8 m above sea level. Lake 1 (54.5042° S, 158.9003° E), Emerald Lake (54.5465° S, 158.8806° E), and Cumberland Tarn (54.6438° S, 158.8523° E) on sub-Antarctic Macquarie Island (54.6208° S, 158.8556° E). Multiple lakes on Macquarie Island were studied to disentangle local and regional influences (Tibby et al., 2025; Supporting Information).

3. Methods

3.1. Sediment chronology (radiocarbon, lead-210, and plutonium-239,240 analyses)

3.1.1. Radiocarbon analyses

Club Lake was radiocarbon (^{14}C) dated by Thomas et al. (2022) using seven bulk sediment ages. The chronology for Rebecca Lagoon combines eight bulk sediment ages (published in Romano and Fletcher 2018) and five terrestrial macrofossil ages analysed in this study. Terrestrial macrofossil ages were used for Lake 1 ($n = 13$), Emerald Lake ($n = 10$), and Cumberland Tarn ($n = 11$).

Terrestrial macrofossils were selected under a stereomicroscope (Zeiss Stemi 2000), pre-treated, graphitised and measured for ^{14}C by Accelerator Mass Spectrometry (AMS) at either the Australian Nuclear Science and Technology Organisation (ANSTO) (Hua et al., 2001) or the Research School of Earth Sciences (RSES) radiocarbon facility at the Australian National University (ANU) (Wood et al., 2023; see Supporting Information).

3.1.2. Lead-210 analyses

Club Lake lead-210 (^{210}Pb) was dated by Thomas et al. (2022) using four sediment ages. Twelve samples were analysed for Rebecca Lagoon, nine for Lake 1, and 10 for Emerald Lake and Cumberland Tarn (see Supporting Information). Analyses for ^{210}Pb were conducted at ANSTO to constrain the age of recent sediment accumulation. Alpha particle spectrometry was used to determine polonium-210 (^{210}Po) and radium-226 (^{226}Ra) activities. Down-core changes in ^{210}Pb were used to calculate accumulation rate from the half-life of ^{210}Pb (22.26 ± 0.22 yr) (Appleby and Oldfield, 1992).

3.1.3. Plutonium-239,240 analyses

To develop a higher resolution chronology for the 20th century, independently validate the ^{210}Pb age-depth model, allow for comparison with atmospheric data and better understand long range transport and deposition of metals, 13 samples were analysed for plutonium-239,240 ($^{239,240}\text{Pu}$) on the Emerald Lake sediment core (Fig. 1) by Accelerator

Mass Spectrometry (AMS). The $^{239,240}\text{Pu}$ analysis was exclusively conducted on Emerald Lake due to limited sample availability across the other sediment records. For sample processing see Supporting Information and Child et al. (2008). The $^{239,240}\text{Pu}$ samples were measured using the VEGA AMS facility at ANSTO following the methods described in Hotchkis et al. (2019).

3.1.4. Age-depth models

Age depth models were produced from each entire lake sediment core, and therefore extend beyond the last 500 years that is the focus of this study. Due to distinct shifts in sedimentation in Lake 1 and Emerald Lake in particular truncating the age depth model would have resulted in unanchored bottom depths (Supporting Information). Ages are reported in common era (CE). Chronologies were constructed using a Bayesian implementation of the Constant Flux (CF) modelling approach with the R package 'rplum' v.1.0.0 (Aquino-López et al., 2018), which, firstly, assumes that the atmospheric flux of unsupported ^{210}Pb remains constant through time while allowing sediment accumulation rates to vary, and secondly, integrates information from both ^{210}Pb , ^{226}Ra . Modelling in rplum uses an autoregressive gamma process with a constant ^{210}Pb flux assumption to derive sediment chronologies. Consequently, temporal changes in sedimentation rate are explicitly accommodated within the model and do not violate the underlying CF assumption.

Integrating multiple absolute dating techniques can result in large uncertainties across different timescales. Guidelines from Sanchez-Cabeza et al. (2026) were used to produce standardised workflows for multi-proxy age depth models. The rplum model uses wider uncertainty intervals, and is better able to integrate multiple data sources using posterior probabilistic modelling, through Markov Chain Monte Carlo iterations thus allowing chronological gaps between ^{210}Pb and ^{14}C to be minimised with Bayesian frameworks without extrapolation (Sanchez-Cabeza et al., 2026). In this study, radiocarbon ages in all age-depth models were calibrated using Southern Hemisphere calibration curve (Hogg et al., 2020), and $^{239,240}\text{Pu}$ fallout data was additionally incorporated into the Emerald Lake age depth model. Emerald Lake did not have a typical Pu fallout profile associated with atmospheric nuclear weapons testing fallout. However, non-monotonic sedimentation (see Supporting Information) in the upper section of the core (0–6.5 cm) and the presence of a Pu peak at 7 cm depth coupled with a decrease to background at 11.5 cm is indicative of the 1963 peak in atmospheric weapon testing (Dharmarathna et al., 2025). The presence of a transition in the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio (Harrison et al., 2021) value from 0.22 to 0.15 at 6.5 cm further validates this assumption and independently agree with the rplum modelled age from ^{210}Pb for Emerald Lake (Supporting Information). The Pu profile was therefore incorporated as a single uncalibrated date into the age-depth model.

3.2. Total element analyses

Between 18 and 25 samples were analysed in each sediment record for 16 elements: Pb, Cd, As, Fe, aluminium (Al), gallium (Ga), cobalt (Co), magnesium (Mg), manganese (Mn), antimony (Sb), strontium (Sr), and vanadium (V), phosphorus (P), nickel (Ni), copper (Cu), and zinc (Zn). Elemental concentrations were measured following a modified protocol of USEPA Method 3051 A (USEPA US Environmental Protection Agency, 2007), at the ANU RSES with subsequent analyses by inductively coupled plasma optical emission spectrometry (ICP-OES). A total of seven of the 16 elements were analysed for higher precision inductively coupled plasma mass spectrometry (ICP-MS), P, Ni, Cu, Zn, As, Cd, and Pb (see Supporting Information). Two certified reference materials were analysed for all 16 elements, NIST 1646a and BCR320 (Supporting Information).

Total organic carbon % (TOC) was determined on each core using an elemental analyser (Thermo Fisher Scientific Flash 2000 EA) at ANSTO (see Supporting Information).

3.2.1. Accumulation rate of metal deposition

The accumulation rate (AR) ($\mu\text{g}/\text{m}^2/\text{yr}$) of Pb, Cd, and As was calculated as follows:

Eq. 1:

$$C_i \text{AR} = (\text{SR} \times \text{DBD} \times C_i) \times 10000 \quad (1)$$

SR = sedimentation rate (cm/yr)

DBD = dry bulk density (g/cm^3)

C_i = concentration of interest: Pb, Cd or As ($\mu\text{g}/\text{g}$)

3.3. Enrichment factor

Enrichment factors (EF) were calculated for each metal to quantify the anthropogenic contributions to the sediment samples (Salomons and Förstner, 2012; Stromsoe et al., 2013; Ioannides et al., 2014). The EF for Pb, Cd and As were calculated using Eq. 2 and Fe was used as the reference element (see Supporting Information).

Eq. 2:

$$EF = \left(\frac{C_i}{C_R} \right)_{\text{sediment}} / \left(\frac{C_i}{C_R} \right)_{\text{background}} \quad (2)$$

Sediment = sediment of interest

Background = is sediment dated to pre-1850 CE

C_i = concentration of element of interest ($\mu\text{g}/\text{g}$)

C_R = concentration of reference element ($\mu\text{g}/\text{g}$)

Background samples span a variable timeframe between lakes, see Supporting Information for further details.

3.4. Statistical analyses

All statistical analyses and data visualisations were undertaken in RStudio v. 4.3.0 (R Core Team, 2023). Pearson correlations were conducted for all elements. Principal component analysis (PCA) was used to evaluate multivariate patterns in sediment geochemistry among lakes and through time. Sedimentation rate and dry bulk density were excluded from the ordination. Prior to analysis, elemental concentrations were standardised and log transformed. Samples below machine lower detection limit, were assigned the lower detection limit of their respective analyses divided by 10. The PCA was performed using the rda () function in the R package 'vegan' v. 2.6–4 (Oksanen et al., 2023). In the absence of explanatory variables, is equivalent to a principal component analysis based on a centred and standardised data matrix. The proportion of variance explained by each principal component was calculated from the eigenvalues of the ordination.

3.5. Atmospheric transport and dispersion model

In this study, we use the HYSPLIT (Stein et al., 2015) model to estimate the relationship between source emission rates and lake catchment deposition rates. The modelling approach is limited in scope, employing a simplified approach to emissions, a single aerosol size, and assumes that transport patterns throughout the 19th and 20th century have not changed. For emissions, we focus on emissions from major smelters located in continental southeastern Australia and Tasmania. Across Australia, smelters were the dominant source of atmospheric metal emissions from the late 19th century through much of the 20th century (Mudd, 2007; Schneider et al., 2020). The smelters were selected based on their geographic location and potential for pollution transport to the sub-Antarctic region. Four smelters were included, Port Kembla (34.483022°S, 150.9067°E), Broken Hill (31.9698°S, 141.4731°E), Port Pirie (33.1699°S, 138.0117°E), and Mount Lyell (42.0660°S, 145.560°E) (Fig. 1; see Mudd 2007 for details). Atmospheric dispersion, specifically the relationships between the selected smelters and study lakes, were calculated using HYSPLIT (version 5.4.2). The model was run in dispersion mode and forced with MERRA2

atmospheric reanalysis fields (Gelaro et al., 2017) following the approach by McConnell et al. (2018). Lead emitted from smelters is assumed to deposit immediately onto ambient aerosol particles, meaning Pb is then transported and deposited with the aerosols. Previous research has shown significant observations of Pb containing particles ranging between 0.1 and 10 μm (Csavina et al., 2014). In the model, a single aerosol size class was simulated with a diameter of 1 μm , a density of 2.5 g/cm^3 , an emission rate of 1 kg/hr from each smelter site, and default wet and dry deposition parameters. The model was used to assess transport plausibility and was run for the year of 1991 to assist in disentangling long range from local transport of particulates.

To verify the model's representation of atmospheric transport we performed a second diagnostic run using atmospheric radon-222 as a tracer. Radon is an inert gas, with a half-life of 3.82 days, emitted continuously from ice-free land. Using known emissions from Australia (Griffiths et al., 2010), the model was run in backwards mode to predict daily mean radon measurements at Macquarie Island for the year 2021, when radon observations are available (Williams and Chambers, 2022). The modelled radon concentration (Supporting Information Figure S 14) reproduces the seasonal cycle of atmospheric transport, with stronger transport during May-August, and individual events have the correct strength, as does the annual mean.

4. Results

4.1. Chronology

Due to uncertainties in the age–depth models across lakes, cross-site alignment is approximate. Age uncertainties for each lake for key horizons discussed throughout this study can be found in Supporting Information. Sedimentation rates exhibit both inter-lake and temporal variability, with a distinct transition occurring around the 1850s (Fig. 2; Supporting Information). Prior to the 1850s, sedimentation rates were relatively low and stable (Fig. 2). Cumberland Tarn had the fastest average sedimentation rate ($0.028 \pm 0.006 \text{ cm}/\text{yr}$), followed by Emerald Lake ($0.022 \pm 0.02 \text{ cm}/\text{yr}$), Rebecca Lagoon ($0.020 \pm 0.006 \text{ cm}/\text{yr}$), Club Lake ($0.008 \pm 0.0008 \text{ cm}/\text{yr}$) and finally Lake 1 ($0.007 \pm 0.006 \text{ cm}/\text{yr}$).

After the 1850s, average sedimentation rates increased in all five lakes, with the most pronounced increase occurring from approximately 1900 CE onwards (Fig. 2). The largest increase in sedimentation rate occurred in Lake 1 (14-fold increase), which had an average sedimentation rate post-1850 of $0.1 \pm 0.08 \text{ cm}/\text{yr}$ (Supporting Information). The pre-1850 CE sedimentation rate of Lake 1 includes a long sedimentary hiatus (Supporting Information) and sedimentation rate is therefore not directly comparable to colonial impact or the other lakes in this study. Rebecca Lagoon and Emerald Lake experienced increases in sedimentation rate of 10.5- and 6-fold respectively, with post-1850 CE average sedimentation rates of $0.2 \pm 0.12 \text{ cm}/\text{yr}$ and $0.13 \pm 0.02 \text{ cm}/\text{yr}$, respectively (Supporting Information). The sedimentation rate increases in Cumberland Tarn and Club Lake were more moderate, with 2.7- and 1.3-fold increases from average post-1850s sedimentation rates of $0.076 \pm 0.05 \text{ cm}/\text{yr}$ and $0.011 \pm 0.002 \text{ cm}/\text{yr}$, respectively (Supporting Information).

4.2. Elemental concentrations, accumulation rates, and enrichment factors

Elemental concentration results align with measured certified reference materials and duplicate sediment samples (Supporting Information). Average recovery across all digestions for CRM NIST1646a (Estuarine sediment) was 95% for As, 85% for Cd, and 91% for Pb. Recovery for CRM BCR320 (Channel sediment) was lower at 91% for As, 81% for Cd, and 89% for Pb. Correlations for all elements are available in Supporting Information.

Lithogenic elements Al and Fe were strongly correlated with each

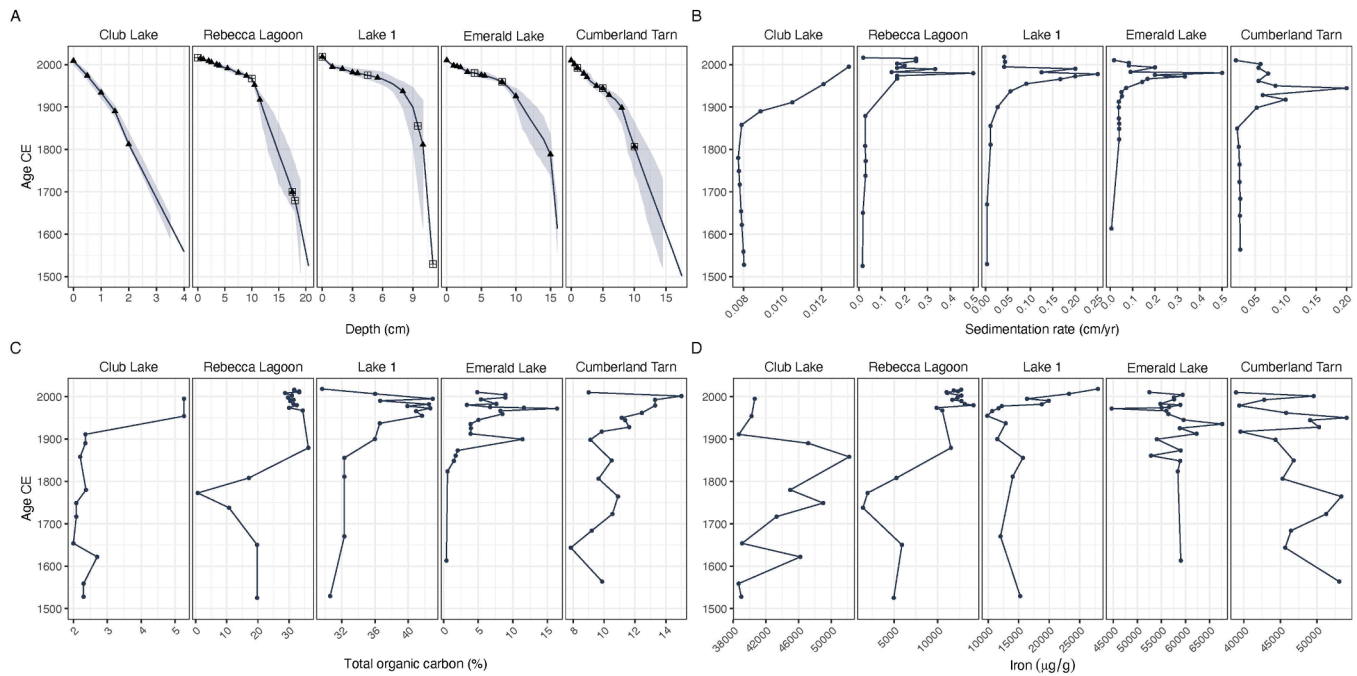


Fig. 2. A) Age-depth model for lakes in this study. Black line indicates modelled age, black triangle indicates ^{210}Pb depths, squares indicated ^{14}C depths, grey cloud indicates ± 1 standard deviation from the mean of the age depth model. B) Sedimentation rate calculation from the age depth model. Black circles indicate depths elemental analyses were conducted for this study. C) Total organic carbon (%), D) Iron (Fe). Ages are reported in Common Era (CE).

other ($r = 0.89$ across all lakes). Iron showed weaker correlations than Al to Pb (Al: $r = -0.21$; Fe: $r = 0.02$), Cd (Al: $r = 0.02$; Fe: $r = -0.08$), and a similar correlation to As (Al: $r = -0.49$; Fe: $r = -0.51$). These weak relationships suggest that variation in Pb, Cd and As concentrations cannot be explained solely by lithogenic mineral abundance. This supports the use of Fe-normalised enrichment factors to assess departures from natural geochemical backgrounds and identify potential enrichment arising from non-lithogenic processes. The Macquarie Island lakes exhibited the highest concentrations of the lithogenic elements Al and Fe followed by Club Lake and Rebecca Lagoon (Table 1).

4.2.1. Total organic carbon

Sedimentary organic matter is a well-established sorbent for many trace metals through adsorption and complexation processes and can therefore influence their accumulation in lake sediments (Adusei-Gyamfi et al., 2019). The pronounced differences in TOC observed among lakes, particularly in Lake 1 and Rebecca Lagoon, were considered in subsequent analyses through TOC normalization. This normalization was used as a standard geochemical approach to account for temporal changes in sediment organic matter when interpreting down-core elemental records, rather than to assess metal transport pathways or quantify metal-organic matter associations.

Greater between-lake variability in sedimentary TOC was observed prior to 1850 CE compared with the post-1850 CE period (Fig. 2). Before 1850 CE, TOC values from the Club Lake record remained low and stable ($\sim 2\text{--}3\%$), while Rebecca Lagoon declined from $\sim 20\%$ to near 0% by the c. 1750 CE before increasing to around 30% by 1850 CE. On Macquarie Island, Lake 1 exhibited consistently high but stable TOC ($\sim 32\%$), Emerald Lake remained low (1–2%), and Cumberland Tarn showed moderate values (8–10%).

From the 1850s to 2000s, TOC increased across all five lakes, with most systems reaching maximum values during the mid to late 1900s. Club Lake remained relatively stable until the 1900s before increasing to greater than 5%, while Rebecca Lagoon rose rapidly after the 1850s and remained elevated around 30%. On Macquarie Island, Lake 1 increased gradually to a peak of approximately 44% around the 1950s, Emerald Lake rose to more than 15% in the 1950s before declining, and

Cumberland Tarn showed a modest increase, reaching 15% at the end of the 1900s.

After 2000 CE, TOC stabilised or declined in several systems. Club Lake and Rebecca Lagoon TOC remained elevated relative to pre-industrial levels. Macquarie Island lakes decline after the 2000s, with Emerald Lake and Lake 1 decreasing to around 5% and 30%, respectively, and Cumberland Tarn returning to values comparable to pre-1900s levels of 8–10% (Fig. 2).

4.2.2. Lead (Pb)

Background Pb concentrations (Pb_C) showed an inverse relationship to lithogenic elements (Fe and Al), with higher values in Club Lake and Rebecca Lagoon compared to the Macquarie Island lakes. Club Lake recorded the highest pre-1850 Pb_C ($45.85 \pm 2.36 \mu\text{g/g}$) followed by Rebecca Lagoon ($3.61 \pm 1.55 \mu\text{g/g}$; Table 1). In contrast, pre-1850 Pb_C in the Macquarie Island lakes were substantially lower, with Lake 1 averaging $0.47 \pm 0.09 \mu\text{g/g}$, Emerald Lake, $0.64 \pm 0.14 \mu\text{g/g}$, and Cumberland Tarn, $1.85 \pm 0.16 \mu\text{g/g}$.

Post-1850 Pb_C in Club Lake declined between the 1900s and 1950s before returning to a slightly elevated Pb_C in the early 2000s, while it increased rapidly in all other lakes (Fig. 3; Table 1). Average post-1850 Pb_C increased 4.7-fold (Rebecca Lagoon), 4.2-fold (Lake 1), 2.0-fold (Emerald Lake), and 3.0-fold (Cumberland Tarn).

The Pb accumulation rates (Pb_{AR}) and enrichment factor (Pb_{EF}) results are broadly similar to Pb_C , remaining stable prior to the 1850s before increasing across all sites in the early 1900s, except Club Lake, which showed only a minor increase after the 1950s ($\text{Pb}_{EF} = 1.05$). The Pb_{AR} values vary considerably between lakes (Club Lake: $3770 \mu\text{g/m}^2/\text{yr}$ versus Lake 1: $3.67 \mu\text{g/m}^2/\text{yr}$ Table 1) potentially reducing between site comparability. Rebecca Lagoon recorded a Pb_{EF} peak in the 1960s (maximum 2.5), followed by a decline. In the Macquarie Island lakes, more pronounced mid-20th century peaks in Pb_{AR} and Pb_{EF} occurred between the 1940s and 1970s, with maximum Pb_{EF} values of 13.45 (Lake 1), 3.79 (Emerald Lake), and 4.29 (Cumberland Tarn) (Fig. 3; Table 1) the 1970s, Pb_{AR} and Pb_{EF} declined yet remained elevated in the Macquarie Island lakes (Fig. 3).

The ratio of lead concentration to total organic carbon (Pb_C/TOC)

Table 1
Summary of elements across, aluminium (Al), iron (Fe), lead (Pb), cadmium (Cd), arsenic (As), enrichment factor (EF), accumulation rate (AR), total concentration (C), Total Concentration reported in $\mu\text{g/g}$. Accumulation rate reported in $\mu\text{g/m}^2/\text{yr}$.

	Age (CE)	Club Lake					Rebecca Lagoon					Lake 1					Emerald Lake					Cumberland Tarn				
		Avg	Std	Min	Max		Avg	Std	Min	Max		Avg	Std	Min	Max		Avg	Std	Min	Max		Avg	Std	Min	Max	
Al _C	Pre-1850	44,360	2107	42,063	49,198		7954	3325	1504	10,897		46,681	8323	30,589	60,240		67,800	5814	62,977	76,211		78,869	3128	73,695	85,825	
Al _C	Post-1850	48,086	4788	42,425	53,692		22,511	3361	14,658	26,388		37,066	8967	25,907	54,332		66,413	4125	60,061	76,055		74,508	4050	69,971	80,563	
Fe _C	Pre-1850	41,611	3434	37,693	48,987		4316	1487	1447	5908		27,298	8818	12,030	36,528		63,834	10,152	58,391	79,057		47,925	3068	43,509	53,328	
Fe _C	Post-1850	43,777	5710	38,681	52,176		11,957	1115	9868	14,099		15,910	5632	9866	27,992		56,870	4688	44,660	67,618		45,367	5274	38,925	54,056	
Pb _C	Pre-1850	45,85	236	42.4	51.04		3.61	1.55	0.73	4.93		0.47	0.09	0.31	0.67		0.64	0.14	0.53	0.83		1.85	0.16	1.57	2.21	
Pb _C	Post-1850	43.11	4.3	38.31	47.16		17.04	3.99	4.73	20.43		1.98	1.26	0.59	4.18		1.27	0.39	0.66	1.96		5.72	1.26	2.97	7.08	
Cd _C	Pre-1850	0.23	0.09	0.17	0.53		0.17	0.1	0	0.27		3.32	3.62	0.74	12.72		0.69	0.15	0.53	0.89		0.6	0.03	0.55	0.67	
Cd _C	Post-1850	0.18	0.05	0.15	0.27		0.35	0.11	0	0.45		2.77	0.67	1.62	3.79		1.2	0.32	0.75	2.19		0.6	0.07	0.43	0.68	
As _C	Pre-1850	2.68	0.12	2.51	3		2.81	1.68	0	4.8		1.09	0.39	0.49	1.78		0.4	0.24	0.06	0.61		1.5	0.14	1.23	1.74	
As _C	Post-1850	2.53	0.55	1.97	3.36		4.31	0.83	3.17	5.26		1.83	0.5	1.01	2.55		0.73	0.16	0.46	1.05		2.84	1.23	1.61	5.39	
Pb _{EF}	Pre-1850	1	0.09	0.82	1.16		1	0.28	0.62	1.44		1	0.35	0.62	1.84		1	0.1	0.91	1.11		1	0.12	0.76	1.22	
Pb _{EF}	Post-1850	0.89	0.09	0.8	1.05		1.77	0.43	0.51	2.47		6.31	3.07	2.04	13.45		2.26	0.69	1.12	3.79		3.28	0.77	1.73	4.29	
Cd _{EF}	Pre-1850	1	0.46	0.66	2.4		1	0.56	0	1.5		1	0.93	0.17	3.33		1	0.29	0.75	1.39		1	0.09	0.85	1.18	
Cd _{EF}	Post-1850	0.75	0.25	0.57	1.18		0.83	0.27	0	1.2		1.59	0.64	0.47	2.43		1.96	0.72	1.29	4.44		1.07	0.15	0.88	1.38	
As _{EF}	Pre-1850	1	0.07	0.82	1.08		1	0.52	0	1.59		1	0.44	0.39	1.7		1	0.63	0.16	1.65		1	0.12	0.78	1.18	
As _{EF}	Post-1850	0.9	0.22	0.74	1.28		0.63	0.13	0.44	0.88		2.8	0.95	1.36	4.32		2.07	0.6	1.28	3.73		2.04	1.02	1.16	4.39	
Pb _{AR}	Pre-1850	3770.82	307.89	3202.67	4260.02		188.9	156.3	89.13	594.47		3.67	3.46	0.21	11.02		119.21	110.55	25.23	246.83		107.99	49.43	47.7	208.7	
Pb _{AR}	Post-1850	3561.94	554.48	3119.87	4531.32		8979.66	5972.63	458.41	25,172.88		288.1	239	7.52	730.81		1049.49	1056.56	141.08	4300.49		841.09	672.78	222.93	2640.39	
Cd _{AR}	Pre-1850	19.23	8.33	13.92	44.45		7.27	8.97	0	31.15		48.17	81.66	0.32	275.59		154.04	158.12	22.37	342.51		35.01	15.98	15.44	65.71	
Cd _{AR}	Post-1850	15.45	6.06	11.46	26.12		178.58	138.5	0	553.35		444.63	385.56	21.65	1139.55		1087.25	1176.28	149.13	4141.09		87.8	58.3	16.58	240.93	
As _{AR}	Pre-1850	220.7	21.58	191.01	262.42		117.6	101.18	0	373.86		10.95	10.97	0.25	32.49		101.4	111.27	2.7	233.79		86.86	39.1	42.48	164.32	
As _{AR}	Post-1850	210.88	63.44	170.41	322.79		2211.83	1520.22	149.06	6363.55		298.44	272.96	12.54	819.27		649.12	675.57	93.68	2612.98		363.48	201.08	184.95	821.9	

increased through the mid-1900s in Rebecca Lagoon, Lake 1, and Cumberland Tarn. Lake 1 Pb_C/TOC continued to increase at the top of the record, while in Rebecca Lagoon and Cumberland Tarn the Pb_C/TOC stabilises in the 1990s and 1950s, respectively, before declining and remaining elevated above background levels. Club Lake and Pb_C/TOC decreased from the 1850s onwards. The overall Pb_C/TOC in Emerald Lake declines, with a slight increase between the 1900s and 1950s (Fig. 3).

4.2.3. Cadmium (Cd)

Cadmium concentrations (Cd_C) and Cd accumulation rates (Cd_{AR}) are broadly similar to Pb trends. Prior to the 1850s, Cd_C was lower across most lakes (0.17–0.69 $\mu\text{g/g}$; Table 1), except Lake 1 on Macquarie Island ($3.32 \pm 3.62 \mu\text{g/g}$). Pre-1850 CE Cd_C are highly variable as a result of variable and high background concentrations of Cd (Supporting Information). Post-1850, Cd_C trends varied. Club Lake increased 1.8-fold by the early 2000s, though the average remained similar to pre-1850 values (Table 1; Fig. 3). Rebecca Lagoon Cd_C increased by two-fold, Lake 1 declined by roughly 20%, Emerald Lake experienced a two-fold increase, with no change in Cumberland Tarn (Table 1; Fig. 3). In Club Lake, Cd_{AR} increases slightly at the top of the record, the Cd_{AR} for all other lakes peak in the mid-20th century before declining to near background levels (Fig. 3).

Cadmium enrichment factors (Cd_{EF}) displayed patterns similar to those of Pb_{EF}, with a slight temporal delay. Club Lake Cd_{EF} results are similar to Pb_{EF}, with only minor enrichment (EF = 1.18) observed in the surface sample (Table 1; Fig. 3). Rebecca Lagoon, Cd_{EF} increased from the 1950s onward, though enrichment remained minimal (Maximum EF = 1.2). On Macquarie Island, Cd_{EF} rose from the 1950s, reaching maximum values in the 1970s in Lake 1 and Emerald Lake (EF = 2–5). In Cumberland Tarn Cd_{EF} peaks in the 1950s with minor enrichment (maximum EF = 1.38). Lake 1 and Cumberland Tarn decline toward pre-1850 CE levels after their peak, while Emerald Lake Cd_{EF} remains elevated (Table 1; Fig. 3).

Cadmium concentration to TOC ratios (Cd_C/TOC) reflected Cd_{EF} in Lake 1, peaking in the mid-1950s, then returning to near background. Other lakes showed declines from the early 1900s (Emerald Lake, Cumberland Tarn) or minimal variability (Club Lake and Rebecca Lagoon) (Fig. 3).

4.2.4. Arsenic (As)

Arsenic concentrations (As_C) broadly followed trends observed for Pb_C and Cd_C. Prior to the 1850s, background As_C was highest in Rebecca Lagoon ($2.81 \pm 1.68 \mu\text{g/g}$) and Club Lake ($2.68 \pm 0.12 \mu\text{g/g}$), followed by Cumberland Tarn ($1.50 \pm 0.14 \mu\text{g/g}$) and Lake 1 ($1.09 \pm 0.39 \mu\text{g/g}$), and lowest in Emerald Lake ($0.40 \pm 0.24 \mu\text{g/g}$). After 1850 CE, As_C increased by 1.5–2-fold in all lakes except Club Lake, which declined slightly (Table 1). Arsenic accumulation rates (As_{AR}) peaked in the mid-1900s before returning to near-background levels in all lakes except Club Lake which showed a minor increase at the top of the record.

The As enrichment factor (As_{EF}) was minor in Club Lake, peaking at 1.28 in the early 2000s (Table 1; Fig. 3). Rebecca Lagoon showed no increase in As_{EF} compared to pre-1850 CE. On Macquarie Island, Lake 1, Emerald Lake, and Cumberland Tarn exhibited rising As_{EF} from the 1920s, peaking after the 1970s (maximum As_{EF} = 3–5; Table 1; Fig. 3). Lake 1 declined to near-background levels post-1970s, whereas Emerald Lake stabilised but remained elevated above background. Cumberland Tarn showed peak As_C and As_{EF} at the top of the core with no measured decline. This upper-core enrichment is considered further in relation to potential post-depositional processes in the Discussion.

Arsenic concentration to TOC ratios (As_C/TOC) declined in Club Lake from the 1900s to the 1950s, with a slight increase in the early 2000s. In Rebecca Lagoon, As_C/TOC remained stable until the 1950s (excluding a ~1760 CE outlier), increasing slightly in the late 1900s, returning to background levels by the early 2000s. On Macquarie Island, Lake 1 As_C increased in the 1970s before declining slightly, while As_C in Emerald

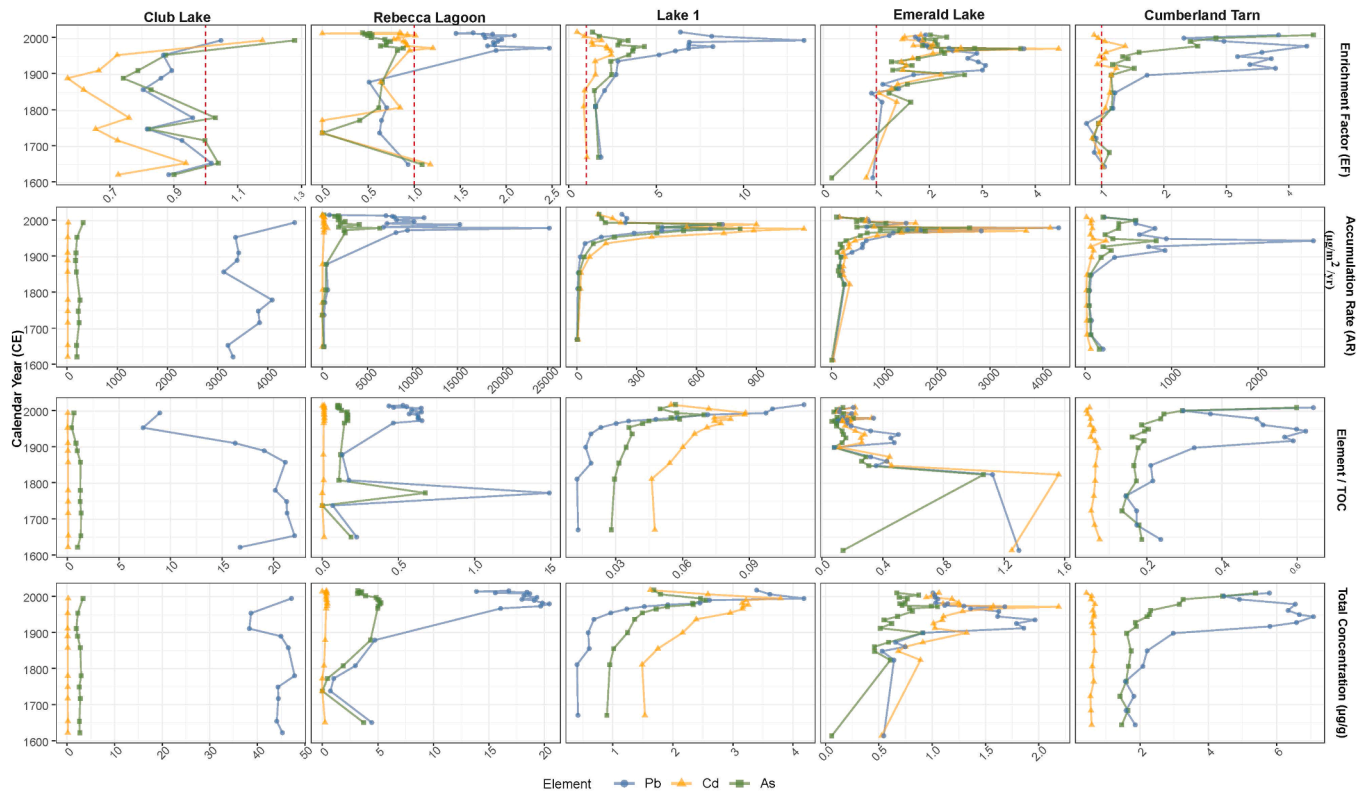


Fig. 3. Stratigraphic diagrams of lead (Pb) as blue circles, cadmium (Cd) as yellow triangles, and arsenic (As) as green squares measured by: Enrichment Factor (EF), Accumulation Rate (AR), Element divided by Total Organic Carbon (Element/C), and total concentration for all five lakes, Club Lake, Rebecca Lagoon, Lake 1, Emerald Lake and Cumberland Tarn. Red dashed lines indicate EF of 1.

Lake (excluding an ~1800 CE outlier) declining from the 1900s onward. In Cumberland Tarn, As_c/TOC increased to the top of the core into the early 2000s (Fig. 3).

4.3. Lake compositional variability

To identify compositional differences among the five lakes, all 17 analysed elements (Al, Fe, Co, Ga, Mg, Mn, Sb, Sr, V, P, Ni, Cu, Zn, As, Cd, Pb, and TOC), were included in a PCA (Fig. 4). The PCA explains 61.5% of the total variance, with PC1 accounting for 36.5% and PC2 for

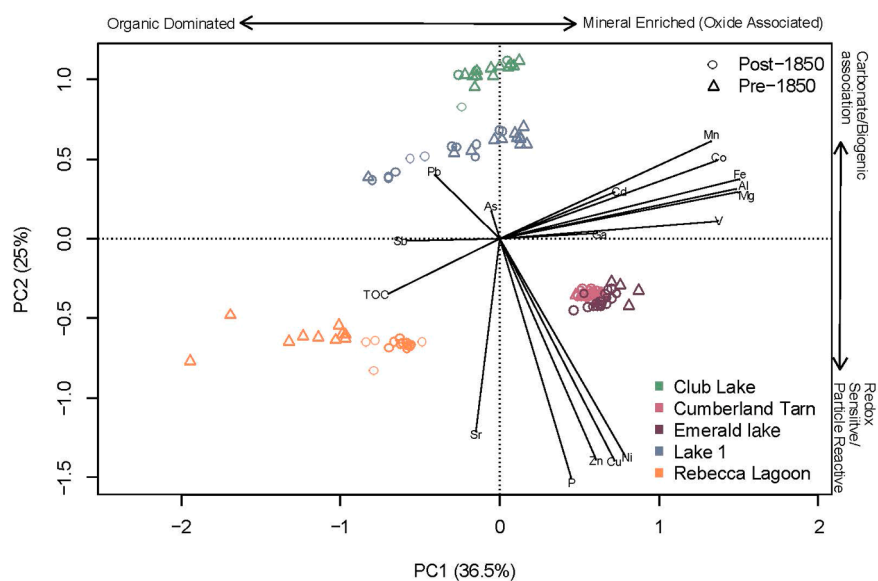


Fig. 4. Principal component analysis (PCA) biplot of sediment geochemistry across the five study lakes. PC1 (36.5%) reflects a gradient from Mineral-Enriched (Oxide Associated) compositions (positive values) to Organic Dominated, compositions (negative values), while PC2 (25%) separates carbonate/Biogenic associations (positive values) from redox-sensitive and reactive particle-associated elements (negative values). Arrows indicate elemental loadings, colours represent individual sediment samples from each lake. Symbols represent time periods, Pre-1850 CE (triangle) and Post-1850 CE (circles).

25% (Fig. 4). Separated along PC1 is a gradient from organic-dominated compositions to mineral-enriched, oxide-associated assemblages. Emerald Lake and Cumberland Tarn are plotted positively on PC1 and characterised by Zn, Cd, Ni, Fe, Mn, Co, Cu, and V (with Fe, Mg, Va, Co and Ni primarily reflecting lithogenic inputs and detrital sediment supply). Rebecca Lagoon and to a lesser extent Lake 1 and Club Lake are negatively plotted on PC1 associated primarily with TOC and, to a lesser extent, Sb. Along the PC2 axis Rebecca Lagoon, Emerald Lake and Cumberland Tarn are associated with positive loadings dominated by redox-sensitive, particle-reactive elements such as Pb and As. Alongside Fe and Mn, are Lake 1 and Club Lake with negative loadings associated with carbonate- and biogenic-linked elements, including Sr and P, as

well as TOC. Temporal variability in the lake records is most evident in Rebecca Lagoon, with pre-1850 CE sediment more organic in composition. The other four lakes do not display consistent temporal trends. Lake 1 is also separated, yet less distinctly, with greater pre-1850 CE influence from mineral-enriched assemblages, likely due to high background Cd concentrations (Table 1). Together, these axes distinguish sediment compositions controlled by mineral-associated metal enrichment from those influenced by organic matter and carbonate or biogenic components.

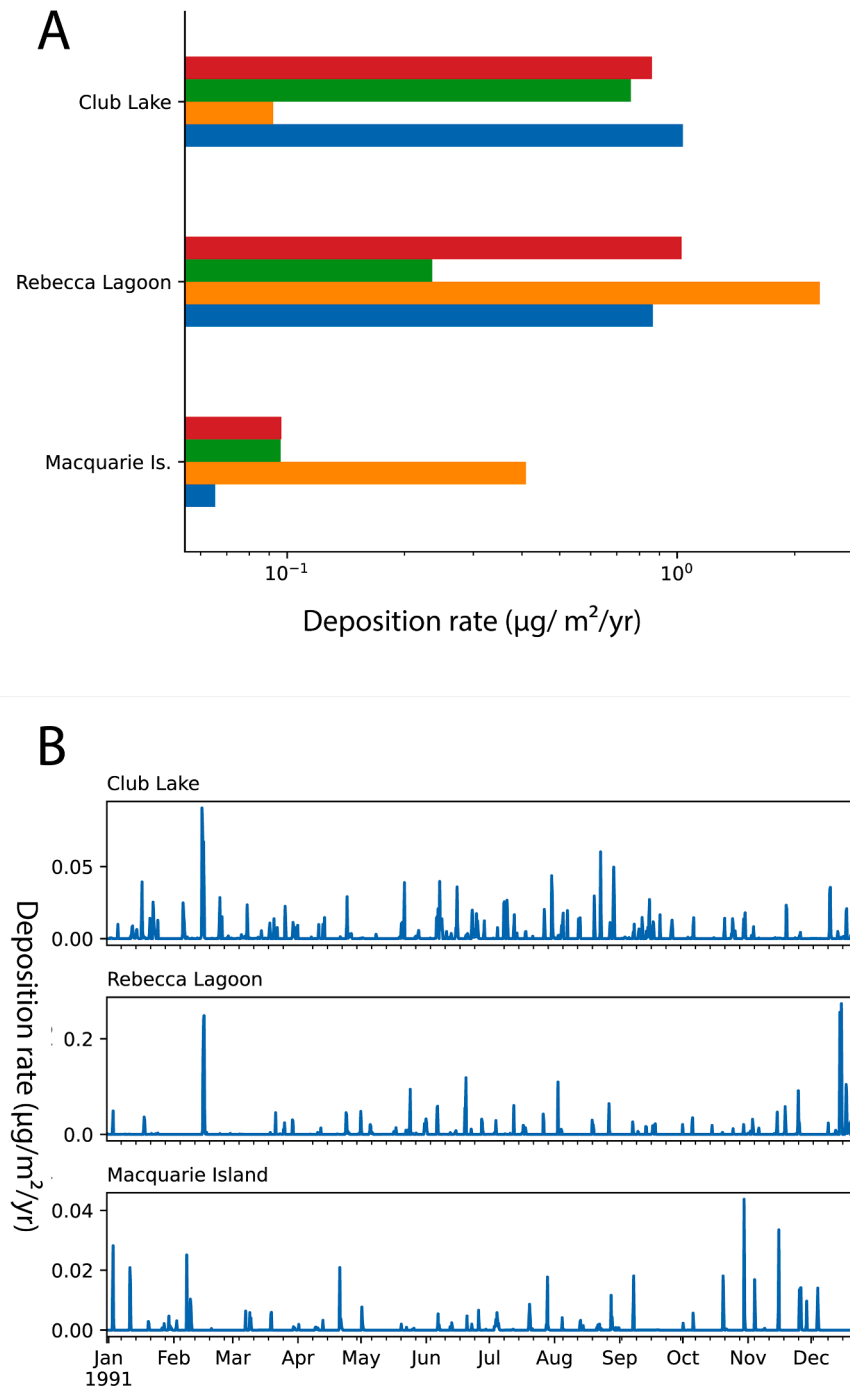


Fig. 5. Deposition rate ($\mu\text{g}/\text{m}^2/\text{yr}$) for each receptor site. A) indicates deposition of source smelter to each receptor site: blue = Broken Hill, orange = Mount Lyell, red = Port Pirie, green = Port Kembla. B) Deposition rate ($\mu\text{g}/\text{m}^2/\text{yr}$) for each receptor site from combined source by time.

4.4. Atmospheric transport

All three lakes on Macquarie Island fell into the same grid cell resolution and therefore are analysed as one receptor site in the atmospheric model. The deposition flux at the receptor sites (Club Lake, Rebecca Lagoon, and Macquarie Island, Fig. 1) relative to emissions from each of the four smelters, differs by a factor of roughly 43. The ordering of sensitivity to emissions is broadly in line with the downwind distance between source (smelter) and receptor (Fig. 1, Fig. 5A). The most efficient transport pathway is from the Mount Lyell smelter to Rebecca Lagoon, $2.32 \mu\text{g}/\text{m}^2/\text{yr}$ and the least efficient pathway is from Broken Hill to Macquarie Island, $0.065 \mu\text{g}/\text{m}^2/\text{yr}$ (Fig. 5A).

Strong deposition to Rebecca Lagoon happens only during a few anomalous events (Fig. 5B). At Rebecca Lagoon, the eight days with the highest deposition rate contribute to 50% of the deposition for the year. Deposition to Macquarie Island is also an anomalous event, with the most intense 10 days adding 50% of the year's deposition. Deposition into Club Lake although much more regular, is also episodic and 22 days contribute 50% of the deposited mass.

5. Discussion

Paleolimnological records provide valuable archives of environmental conditions prior to European colonisation (Muñoz-López and Rivera-Rondón, 2025) but interpretations are constrained by chronological uncertainty and site-specific depositional processes. These limitations are particularly relevant for the early 19th century, the primary period of interest in this study. Chronological uncertainty is further compounded by the multi-lake framework, as each record carries independent age–depth model uncertainty (Supporting Information; Thomas et al., 2022). Variable sedimentation rates and site-specific constraints mean that direct temporal alignment among lakes is necessarily approximate.

Exceptionally low accumulation rates at sites such as Club Lake and the Macquarie Island lakes reduce temporal resolution and in some cases limit interpretability of the record. For example, the period post 1850 CE to present in Club Lake is restricted to the top 1.75 cm of the sediment record. Intermittent drying or non-continuous sediment accumulation in Lake 1 and Emerald Lake (Supporting Information) compress extended time intervals into thin sediment sections. Lake 1 for example, spans approximately 7532 cal BP in 3.5 cm from 11 to 14.5 cm (Supporting Information). Because of this, background calculations span both inconsistent age ranges and sediment depths, potentially exaggerating apparent rates of change relative to more rapidly accumulating anthropogenic-era sediments (Tibby et al., 2025).

Despite these constraints, the stratigraphy provides an integrated record of long-term geochemical change, allowing robust identification of broad temporal patterns and regional coherence. Although post-depositional processes such as sediment focusing, diffuse transport, and, in organic-rich sediments, the redistribution of unsupported ^{210}Pb during early diagenesis can influence ^{210}Pb chronologies (Baskaran et al., 2014), all cores exhibit a consistent increase in age with depth (see Supporting Information). While a monotonic age–depth relationship alone does not preclude all post-depositional mobility, it provides no evidence for substantial diffuse transport. Together with the Bayesian age–depth modelling framework and the propagated chronological uncertainties, these observations support the interpretation that the chronologies are robust within their stated uncertainties. When interpreted within these constraints, the multi-lake records remain valuable for distinguishing local from regional processes and identifying coherent signals of environmental change associated with colonisation, industrialisation, and the Great Acceleration.

5.1. Site geology and background geochemistry

The five lakes span diverse geological, geomorphological, and

climatic settings, which strongly influence their background geochemistry and are reflected in the PCA structure (Fig. 4). Samples plotting towards the mineral-enriched, oxide-associated end of PC1 are dominated by lakes with strong lithogenic inputs, particularly Club Lake and the Macquarie Island lakes. Club Lake, was formed by cirque glaciation (Stromsoe et al., 2013), is underlain by Fe and Mg-rich Ordovician metamorphic schists, quartzites, gneiss, and granulites (Kolbe and Taylor, 1966; Thomas et al., 2022), explaining its elevated Fe and Al concentrations and strong association with lithogenic element loadings in the PCA (Fig. 4). Club lake is separated by Pb, due to an elevated concentration compared to the other lakes in this study, while still within expected background levels, Macquarie Island's unique geology, uplifted oceanic crust, and Rebecca Lagoon sitting on sandy soils are below continental levels (Varne et al., 2000; Saunders et al., 2012; Stromsoe et al., 2013).

Macquarie Island lakes exhibit high Fe and Al concentrations consistent with weathering inputs from the island's mafic–ultramafic bedrock (Griffin and Varne, 1980; Varne et al., 2000), and clustering towards the mineral-enriched end of PC1. Lake 1, however, does not follow the same trend. The difference between Lake 1 and other Macquarie Island lakes likely reflects a greater influence of overlying organic-rich catchment soils that dilute lithogenic inputs and modify sediment geochemistry (Tasmanian Geological Survey, 1998), shifting its position in the PCA relative to the other Macquarie Island lakes.

In contrast, Rebecca Lagoon plots towards the organic-dominated end of PC1 and is characterised by the lowest Fe and Al concentrations (Table 1). This reflects its coastal setting behind a dune–foredune system (Saunders et al., 2012), where catchment inputs are dominated by chemically inert, nutrient-poor quartz sands (Sleep and Hessler, 2006; James et al., 2008). Together, these patterns demonstrate first-order geological controls on sediment geochemistry, enabling anthropogenically driven changes to be interpreted in the context of natural background variability and facilitating the separation of catchment-derived inputs from atmospheric deposition.

5.2. Land-use change

The regions considered in this study have different indigenous and colonial use histories. Club Lake, located in Australia's Alpine region, is on the Country the Wiradjuri, Wolgalu, Ngannawal and Monaro Ngarigo people, and is the traditional gathering place for many Aboriginal groups that travelled to the area for trade and ceremony over thousands of years (Kamminga, 1995). The limited paleo-evidence from the region suggest very low/no impact of indigenous populations on the sedimentation record in this environment during the period of focus of this study (Thomas et al., 2022). Rebecca Lagoon is on the country of the Palawa people of northwest Lutruwita/Tasmania, where paleo-evidence supports the suggestions of low/no disturbance to sedimentation during Palawa land management in comparison to European arrival and the presence of people and use of fire in this landscape (Romano and Fletcher, 2018). In contrast to mainland Australia and Tasmania, Macquarie Island, which is 1500 km to the south of Tasmania, shows no evidence for indigenous occupation prior to 1850 CE. We therefore consider these regions prior to the 1850s, to have experienced minimal anthropogenic disturbance. Prior to the 1850s, sediments are characterised by geochemical signatures dominated by biogenic or/and lithogenic inputs from local catchment geology (Fig. 4). This is supported by stable sedimentation rates prior to 1850 CE, indicative of minimally disturbed systems and natural weathering processes (Fig. 2; (Cook, 2019)).

Environmental change associated with European colonisation involved widespread land clearing, grazing, agriculture, mining, urbanisation, introduction of invasive species, and industrialisation. Together, this altered vegetation cover, soil stability, and hydrological regimes (Bradshaw, 2012; Cook, 2019), leading to increased catchment erosion, modified sediment transport, and changes in lake geochemistry

(Jenny et al., 2019).

All five lakes in this study are known to have experienced colonial land-use disturbance, reflected by increases in sedimentation rate and TOC from the mid-19th century onward (Fig. 2; (Saunders et al., 2012; Saunders et al., 2013; Thomas et al., 2022)). However, the history and drivers of disturbance differ between mainland Australia and Tasmania and that of Macquarie Island. In Club Lake, sedimentation rate increases in the mid-1850s and TOC increases in the 1900s (Fig. 2), reflecting catchment modification from grazing, resulting in increased erosion and sediment delivery to the lake. While grazing in Club Lakes catchment ended in the 1950s (Lembit, 2002), there is no clear decline in sedimentation rate or TOC in our record, which likely reflects the severity of the grazing (Pulsford et al., 2004) in combination with continued presence of feral animals and altered fire regimes, limiting recovery (Dodson et al., 1994; Green, 2016; Thomas et al., 2022).

In Rebecca Lagoon, marked increases in sedimentation rate, TOC, and Fe since the 1850s (Fig. 2) reflect enhanced terrestrial inputs following catchment modification. Early European activities, including grazing and forestry-driven clearing (Saunders et al., 2012) transformed the landscape from forest to grassland, promoting erosion and nutrient mobilisation. Elevated productivity indicators including nitrogen and total chlorin, reported from the late 19th century onward support this interpretation (Saunders et al., 2012; Mackenzie and Moss, 2017). Although sedimentation and elemental concentrations decline slightly in the mid-20th century, they stabilise at levels well above pre-colonial conditions, indicating incomplete recovery.

The three lakes on Macquarie Island show increased TOC and sedimentation rates from the early 20th century (Fig. 2), reflecting extensive erosional disturbance. This disturbance was likely driven by the introduction and rapid population growth of rabbits (Cumpston, 1968). Introduced in the late-19th century, rabbit numbers peaked at over 150,000 by the mid-to-late 20th century (Soucy, 2017). The TOC trends in these lakes broadly track rabbit population dynamics, peaking in the 1970s before eradication efforts began, which culminated in eradication by 2014 (Springer, 2018). Although post-eradication sediments (after 2014) are unavailable in this study, declines in TOC at the top of all three records suggest a reduction in landscape disturbance.

We found that consistently, even across diverse ecological settings spanning over 2000 km from continental southeastern Australia to Tasmania to the sub-Antarctic, colonial-era disturbances beginning in the early-19th century produced consistent lake responses, characterised by increased sedimentation rates and organic matter delivery, likely due to enhanced erosion.

5.3. Disentangling geochemical and atmospheric controls on lake sediments

The onset of European colonisation in southeastern Australia initiated parallel processes which shaped lake sediment geochemistry. From the mid-19th century onward, lake sediments potentially record both enhanced inputs from disturbed catchments and atmospheric deposition of metals derived from industrial emissions. All study sites are positioned to receive atmospheric metal deposition, with Club Lake, Rebecca Lagoon, and the Macquarie Island lakes located along long-range atmospheric transport pathways influenced by emissions from major legacy smelting centres, including Mount Lyell (Tasmania), Port Pirie (South Australia), and Broken Hill and Port Kembla (New South Wales), although none are located directly adjacent to a smelter (Fig. 1). Mining and smelting activity expanded markedly during the late 19th and early 20th centuries, coincident with intensified land-use change across southeastern (Mudd, 2007; Schneider et al., 2022).

While mining typically produces coarse particulates that settle close to source areas, resulting in localised contamination (Dudka and Adriano, 1997). Smelting emits metals primarily as fine particulates and metal-bearing gases associated with sulphates or oxides (Pacyna and Pacyna, 2001). These particles can remain airborne for weeks, enabling

long-range transport, making smelters a dominant source of regional-scale metal contamination (Reimann et al., 2000; Csavina et al., 2014). Importantly, many smelters were commissioned pre-environmental regulation, operating with limited emission controls, the later introduction of taller stacks reduced local ground-level pollution but enhanced regional-scale dispersal. (Thomas et al., 1963).

Coal-fired power stations, established in Australia from the 1930s, generally operated at smaller scales due to low population density and under emerging regulatory frameworks for metal emissions (Schneider et al., 2021). In contrast, smelting operations were largely pre-regulatory and inefficient by modern standards. Smelting operations likely dominated atmospheric metal inputs during the early industrial period. The interpretation of metal input in lake sediments requires explicit consideration of concurrent catchment disturbance. Colonial land-use change increased erosion and dust mobilisation by four-fold from the mid 19th century across southeastern Australia (Hooper and Marx, 2018).

Increased metals may reflect both direct industrial emissions and secondary remobilisation of contaminated soils. We therefore use an atmospheric model to separate long range transport to remote ecosystems from local catchment disturbance. However, colonial-induced erosion could also contribute to secondary remobilisation of contaminated material, including fine particles originally derived from smelter emissions and coarse particulates initially deposited close to mining and processing sites. Some of the metal enrichment observed in lake sediments may reflect the redistribution of legacy contamination rather than direct atmospheric deposition alone, a pathway that is not explicitly represented in the atmospheric transport model used here.

Club Lake was modelled to receive relatively consistent deposition year round yet still receives half of its yearly deposition in 22 days (Fig. 5). However, the Club Lake sediment record showed only minimal post-1850 CE EF and AR of Pb, Cd, and As. With an overall decline in Pb_C/TOC , Cd_C/TOC , As_C/TOC and Fe from the 1900s. The decline in Pb_C Club Lake between the early 1900s and 1950s, despite increasing Australian industrial activity, may reflect dilution associated with enhanced sediment delivery rather than declining atmospheric Pb inputs. Increased catchment disturbance and erosion following the introduction and expansion of non-native grazing animals, such as feral horses (Thomas et al., 2022), could have increased the influx of relatively Pb-low lithogenic material to the lake. Consequently, the Pb concentration record reflects changes in both Pb deposition and bulk sediment accumulation, and declining concentrations do not necessarily indicate a reduction in Pb loading. Long-range metal transport from southeastern Australia has previously been documented as far as New Zealand and Antarctica (Marx et al., 2008; McConnell et al., 2024), including at Club Lake (Stromsoe et al., 2013). However, the elevated natural background Pb_C (Table 1) in combination with grazing-induced erosion (increase in TOC; Fig. 2), low sample resolution (Supporting Information), and hydrological export via a lake outflow (Stromsoe et al., 2013) likely mask low-level atmospheric inputs (Fig. 5), resulting in sediment records dominated by lithogenic and organic inputs.

Modelled atmospheric Pb input to Rebecca Lagoon is the highest of all lakes, with Mount Lyell representing the dominant source (Fig. 5). Deposition however is episodic, south-easterly winds are rare on the Tasmanian west coast (Bureau of Meteorology, 2025; Fig. 1). Half of the annual Pb deposition is modelled to occur across just eight days (Fig. 5B). Rebecca Lagoons proximity to Mount Lyell (~120 km) likely allows intermittent inputs. Our sediment record aligns with this model output, with an increase in metal concentration, EF ($Pb_{EF}=2.5$), AR, and Pb_C/TOC from the 1900s onwards. The Pb peaks in the late 1950s before slowly declining and stabilising well above background levels, aligning with late 20th century emission controls (Schneider et al., 2022), further evidencing the likelihood of atmospheric deposition at Rebecca Lagoon. While TOC and Fe also increase, likely reflecting enhanced catchment inputs associated with land-use change, Pb_C/TOC and Pb_{EF} continue to increase. As Pb may also be transported to lakes bound to or associated

with terrestrially derived organic matter we therefore interpret Pb_C/TOC in combination with Pb_{EF} and regional emission histories. This indicates that Pb enrichment exceeded the increase in organic matter accumulation, suggesting an additional Pb source beyond catchment-derived material. Together with the elevated Pb_{EF} values and regional emission history, this is consistent with a contribution from atmospheric Pb deposition, likely originating from the Mount Lyell smelter.

On Macquarie Island, the atmospheric Pb model showed episodic depositional events primarily from Mount Lyell (Fig. 5B). The modelled deposition quantities for Macquarie Island are significantly lower than Rebecca Lagoon and Club Lake (Fig. 5), a product of Macquarie Island's remote location. However, all three lakes on Macquarie Island show greater relative metal enrichment than both Club Lake and Rebecca Lagoon. The temporal correspondence between metal enrichment and modelled atmospheric transport from southeastern Australian smelters is consistent with an industrial contribution to Macquarie Island. However, the absence of Pb isotope measurements prevents definitive source attribution, and alternative sources or combinations of sources cannot be excluded. Consequently, the atmospheric model should be interpreted as identifying plausible source regions and transport pathways rather than providing conclusive evidence of source identity. The Pb_C , Pb_{EF} and Pb_{AR} all increase from the early 20th century, peaking mid-century before declining (Fig. 3), consistent with the timing of industrial emissions from south-eastern Australia (Schneider et al., 2022), yet also overlaps with rabbit population trends (Saunders et al., 2013). Yet, the continued increase in Pb/C and Pb_{EF} in Lake 1 and Cumberland Tarn (Fig. 3) indicates that Pb increased disproportionately relative to organic matter accumulation. While this pattern is consistent with an atmospheric contribution, catchment-derived Pb inputs cannot be excluded.

The magnitude of Pb enrichment observed on Macquarie Island exceeds that expected from its remote location and modelled atmospheric deposition estimates (Figs. 3 and 5). Additionally metal accumulation rates, in particular Pb_{AR} should be interpreted cautiously when comparing lakes, varying by three orders of magnitude in this study (Table 1), a product of potentially atmospheric inputs and site-specific sedimentological processes. Differences in Pb_{AR} among lakes likely reflect variations in sediment accumulation, bulk density, catchment characteristics, and metal availability, in addition to differences in atmospheric deposition. Therefore, Pb_{AR} trends within individual lakes provide the strongest evidence for temporal changes, while cross-lake comparisons are interpreted alongside concentration-based proxies. The low background metal concentrations, a result of the island being uplifted oceanic crust, and remote setting of Macquarie Island make the system highly sensitive to external input, even small absolute increases can produce large relative changes, particularly when interpreting EF. Cumberland Tarn has a background Pb_C of 1.85 $\mu\text{g/g}$ and post-1850 CE average of 5.72 $\mu\text{g/g}$, representing a three-fold increase of 3.87 $\mu\text{g/g}$ from background levels. For comparison, Club Lake would require a Pb_C increase of 91.72 $\mu\text{g/g}$ to achieve the same threefold increase. Modelled deposition, is 4.5 times greater at Club Lake than Macquarie Island (Fig. 5), yet background Pb_C is almost 25 times greater in Club Lake than Cumberland Tarn on Macquarie Island (Table 1). These low geogenic background concentrations likely amplify the apparent degree of enrichment on Macquarie Island and may explain the discrepancy between relative enrichment and modelled deposition quantities. Additionally, a 35% increase in annual rainfall on Macquarie Island over the last 100 years (Bergstrom et al., 2015) could be contributing to increased wet deposition (Zhang et al., 2025). However, quantifying the relative contribution of changes in precipitation versus atmospheric metal loading is beyond the scope of this study. The Fe_C and TOC in Lake 1, and to a lesser extent in Cumberland Tarn, increases in the mid-1900s, with catchment inputs and other climatic factors likely compounding atmospheric signals from Australian smelters due to these overlapping timelines (Cumpston, 1968; Saunders et al., 2013; Soucy, 2017).

In Emerald Lake on Macquarie Island, Pb_{EF} remains elevated while

Pb_C/TOC declines from the early 1900s, indicating that Pb inputs are increasingly dominated by catchment derived organic matter rather than direct atmospheric deposition. In contrast to Pb, Cd and As exhibit less consistent behaviour across all lake records (Fig. 3). At Club Lake, Cd_{EF} and As_{EF} are higher than Pb_{EF} , likely reflecting elevated background Pb concentrations that dampen apparent Pb enrichment. In Rebecca Lagoon, Pb_{EF} increases consistently through time, whereas Cd_{EF} and As_{EF} remain low, with smaller accumulation rate increases relative to Pb. Although Cd_{EF} followed a broadly similar temporal pattern to Pb_{EF} , its maximum enrichment was substantially lower. The magnitude of EF values for different elements is not directly comparable because each is expressed relative to its own natural background concentration. The lower Cd_{EF} may therefore reflect differences in baseline abundance, source emission ratios, atmospheric transport and lake-specific retention. Moreover, the similar timing of Pb and Cd enrichment does not require an identical source history. Both metals are emitted during non-ferrous metal production, but their relative emissions depend on ore composition and industrial processes, while twentieth-century Pb deposition may also have included substantial contributions from leaded fuel combustion. The records are therefore interpreted as reflecting broadly concurrent increases in industrial atmospheric deposition rather than equivalent inputs from a single source.

Across Macquarie Island, Cd and As trends in Lake 1 broadly mirror those of Pb, suggesting a shared atmospheric influence, with Cd_C and As_C higher than the other Macquarie Island lakes. In contrast, Cd_C and Cd_C/TOC ratios in Emerald Lake and Cumberland Tarn ($Cd_C < 1 \mu\text{g/g}$) have declined since the 1900s, indicating dominance of catchment-derived inputs (Gao et al., 2025). Arsenic in Cumberland Tarn shows a distinct pattern, continuing to increase at the top of the record, not aligning with known industrialisation or landscape disturbance (Fig. 3). This likely reflects diagenetic remobilisation or post-depositional redistribution rather than external loading, as it is only present in one lake (Leclerc et al., 2021; Gao et al., 2025). However, speciation data or porewater profiles would be required to definitely identify diagenetic remobilisation as the mechanism of increased As in Cumberland Tarn, a limitation to this interpretation. The relatively muted variability in As_C/TOC compared to accumulation rates and As_{EF} supports this interpretation, indicating mineral in-wash rather than organic matter-associated transport.

All lakes were modelled to receive atmospheric Pb inputs; however, the expression of this signal varies with metal, background concentrations, catchment processes, and geographical position. Mid-20th century peaks in Pb, and to a lesser extent Cd and As, are consistent with enhanced atmospheric deposition during industrial expansion, followed by declines associated with emission controls (Schneider et al., 2022). Club Lake shows limited post-1850 CE enrichment, with the atmospheric signal largely masked by high Pb background. In contrast, Rebecca Lagoon records a clearer atmospheric input, supported by modelled deposition, reflected by pronounced mid-20th century Pb enrichment. The Macquarie Island lakes exhibit the largest relative increases in Pb despite their isolation, likely a product of very low background concentrations that amplify the magnitude of enrichment. Among these, Lake 1 and Emerald Lake show evidence of atmospheric deposition variably masked by catchment disturbance and organic matter inputs, while Cumberland Tarn preserves the clearest atmospheric signal. Together, these records indicate widespread long-range atmospheric metal transport from Australia, with site-specific sensitivity governed by background conditions and local disturbance.

6. Conclusions

Overall, our findings establish pre-disturbance metal contamination baselines for southeastern Australia (Club Lake), Tasmania (Rebecca Lagoon), and Macquarie Island (Lake 1, Emerald Lake, and Cumberland Tarn), separating natural variability from anthropogenic enrichment, placing modern contamination within its historical (colonial) context,

and providing evidence to support informed environmental policy. The synchronous metal enrichment observed across southeastern Australia, Tasmania, and the sub-Antarctic further demonstrates that lake sediments are robust long-term archives of Southern Hemisphere Anthropocene change. Persistent metal enrichment in these remote ecosystems reveals the enduring legacy of atmospheric pollution and landscape disturbance, even far from industrial centres, highlighting the importance of integrating palaeoenvironmental records into environmental monitoring programs to track change, assess ecosystem recovery, and guide future management. Collectively, these results provide compelling evidence for long-range atmospheric metal transport associated with colonial-era resource extraction and emphasise the need to incorporate colonial histories and their social context into global assessments of environmental change in the Anthropocene.

CRedit authorship contribution statement

Margot Aurel Schneider: Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Larissa Schneider:** Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Conceptualization. **Krystyna M. Saunders:** Writing – original draft, Methodology, Investigation, Funding acquisition, Conceptualization. **James Oliver Latimer:** Writing – original draft, Investigation. **Stephen J. Roberts:** Writing – original draft, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **Brett Knowles:** Writing – original draft, Methodology, Formal analysis, Data curation. **Stewart J. Fallon:** Methodology, Formal analysis. **David Child:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Simon Graeme Haberle:** Writing – review & editing, Funding acquisition, Conceptualization. **Michael-Shawn Fletcher:** Formal analysis. **Alan D. Griffiths:** Writing – original draft, Methodology, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the author(s) used ChatGPT to assist with language editing and improving clarity of the text. After using this tool, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the published article.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgment

The authors thank Michael Hotchkis for his role in AMS measurement and data calculation for plutonium analyses. The authors acknowledge the support for the Centre for Accelerator Science at ANSTO, through the Australian National Collaborative Research Infrastructure Strategy (NCRIS). The authors thank Sabika Maizma at ANSTO for her role in low-level radiochemical analyses for ^{210}Pb analyses, and her support throughout the project. The authors thank Narelle Hegarty and her team at ANSTO for her role as a Stable Isotope Analyst for the analyses of the TOC samples and review of portions of the manuscript. The Authors thank Gavin Mudd and Patrice de Caritat for their valuable initial discussions of the manuscript. This work forms part of Margot Aurel Schneider's PhD thesis and is supported by an Australian Institute for Nuclear Science and Engineering Post Graduate Research Award (AINSE PGRA). This research was funded by an Australian Research Council (ARC) Discovery project DP220100828. The TOC, ^{210}Pb , $^{239,240}\text{Pu}$ and some ^{14}C dates were undertaken at ANSTO funded by

ANSTO grants AP16422, AP18040 and AP18122 and the ANU Radio-carbon Laboratory. The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model used in this publication and the ANSTO HPC facility for supplying the computational resources and support for running the model. We thank the Tasmanian Parks and Wildlife Service and Australian Antarctic Division (AAS 4156, awarded to Krystyna M. Saunders) for field support and access to Macquarie Island. The authors acknowledge the data providers: <https://earthexplorer.usgs.gov/>, <http://data.nsw.gov.au>, <https://services.thelist.tas.gov.au>, <https://www.mapbox.com/>, for imagery used in Fig. 1.

Applications, Solutions and Forecasts

- **Colonisation:** Metal increases are not driven by industrialisation alone, colonial land use and resource extraction initiated widespread environmental change, especially in the Southern Hemisphere.
- **Remote systems are sensitive:** Low-background environments show large relative enrichment from small metal inputs, making them highly vulnerable to disturbance.
- **Local and global drivers:** Metal accumulation reflects both atmospheric transport and catchment disturbance, and both must be considered in environmental assessments.
- **Long-term records are essential:** Sediment archives provide critical baselines for identifying human impact and improving predictions under future environmental change.

Supporting Information

For additional experimental details, materials, methods, and data see [Supplementary File 1](#). Hysplit atmospheric model results will be made available on zenodo.

Appendix A. Supporting Information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.gecadv.2026.100041](https://doi.org/10.1016/j.gecadv.2026.100041).

Data availability

Data is available in [Supporting Information](#). Atmospheric model is available on Zenodo: <https://doi.org/10.5281/zenodo.22701450>

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