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Colonisation-driven mercury cycling overrode industrial pollution in sub-Antarctic lakes

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Abstract

The rise of mercury since the Industrial Revolution is regarded as a global signature of industrialisation. We use lake sediment records from sub-Antarctic Macquarie and Campbell Islands spanning the period before and after 1850 Common Era (CE) to investigate how local colonial disturbance influences mercury sources and cycling. Although mercury accumulation increased 1.1- to 74-fold across all lakes following European arrival (1810 CE), isotope responses were lake specific. Our findings identify colonisation as an unrecognised driver of mercury cycling that can override the global industrialisation signature in the Southern Hemisphere. Colonisation-driven ecological changes, including the introduction of invasive

rabbits altered mercury cycling through two connected processes. Firstly, vegetation disturbance and erosion increased catchment-derived mercury. Secondly, marine biological mercury cycling shifted to terrestrial biological cycling through food web reorganisation. These responses reveal that colonisation, primarily through the introduction of invasive species, was a significant driver of Anthropocene mercury cycling, alongside industrialisation.

Introduction

Mercury (Hg) enrichment over the past two centuries is widely interpreted as a consequence of industrialisation, with increased atmospheric emissions from mining, fossil fuel combustion and manufacturing considered the primary drivers of rising Hg deposition in remote environments (Driscoll et al. 2013; Fisher et al. 2023). This interpretation is founded largely on records from the Northern Hemisphere (Fitzgerald et al. 2005; Biester et al. 2007). This is despite growing evidence that Hg predominantly cycles hemispherically rather than globally (Lamborg et al. 2002; Obrist et al. 2018; Schneider et al. 2023). Whether similar processes underpin Anthropocene Hg accumulation in the Southern Hemisphere remains unresolved (Horowitz et al. 2017; Saiz-Lopez et al. 2020; Shah et al. 2021; Zhang and Zhang 2022).

Mercury is a naturally occurring element that cycles continuously through the atmosphere, lithosphere, hydrosphere and biosphere (Fitzgerald and Lamborg 2005). Although anthropogenic emissions have substantially perturbed this cycle, Hg transport, transformation and accumulation remain fundamentally controlled by Earth system processes. Evidence of long-range atmospheric transport and global cycling established Hg as a pollutant of international concern and provided the foundation for international action (Selin 2009). Understanding cycling processes remains central to implementing the Minamata Convention; however, understanding and knowledge of global Hg cycling and historical contamination remains heavily biased towards the Northern Hemisphere.

In the Southern Hemisphere, where ocean–atmosphere exchange plays a proportionally greater role than in the Northern Hemisphere (Obrist et al. 2018; Schneider et al. 2023), European colonisation over the past ~500 years fundamentally transformed ecosystems (Bradshaw 2012; Lightfoot et al. 2013). These distinct environmental histories suggest that the controls on Hg accumulation may differ from those inferred from predominantly Northern Hemisphere records. Yet historical Hg trends are still largely interpreted as signals of industrial emissions,

with the potential influence of localised colonisation-driven ecological change remaining poorly understood.

This imbalance is particularly important in the Southern Hemisphere where environments have experienced distinct histories of human disturbance. In Australasia, European colonisation began in the late 18th century, followed by major industrialisation and associated Hg emissions during the 19th and 20th centuries. This history contrasts with many Northern Hemisphere regions, where earlier and more prolonged industrial activity produced fundamentally different Hg emission trajectories (Schneider et al. 2022; Fisher et al. 2023). Limited interhemispheric exchange further constrains the transferability of Northern Hemisphere-derived models to Southern Hemisphere environments (Fisher et al. 2023; Schneider et al. 2023). Consequently, models developed from these records may not adequately represent global Hg cycling. This bias also shapes interpretation of lake sediment archives, where increases in Hg accumulation are commonly attributed to industrial emissions. This potentially overlooks changes in ecological baselines and biologically mediated Hg inputs associated with more local human-driven ecosystem transformations as a consequence of exploitative practices associated with European colonisation.

Sub-Antarctic lake sediments and peat profiles provide unique archives for testing the environmental imprint of industrialisation in the Southern Hemisphere. Located at ~50–60°S, these records capture environmental change and atmospheric Hg deposition (Li et al. 2020) in a region where terrestrial archives such as tree rings and speleothems are scarce (Neukom and Gergis 2012; Turney et al. 2016; Thomas et al. 2018). Importantly, these records do not reflect atmospheric inputs alone, seabirds are important vectors of elemental transport to Sub-Antarctic island environments and ecosystems (Otero et al. 2018; Li et al. 2026). Seabirds in the Sub-Antarctic depend on the surrounding ocean as a major food source, consuming fish and other marine mammals (Treasure et al. 2015). Sub-Antarctic lakes and their sediments therefore integrate both marine and terrestrial influences, both from seabird-mediated Hg and nutrient cycling (Otero et al. 2018; Li et al. 2026), and landscape disturbance, including the introduction of invasive species, following human arrival. These interacting processes provide a natural experiment for disentangling industrial emissions from colonisation-driven changes in Southern Hemisphere Hg cycling.

101 In this study, we combine Hg, carbon (C) and nitrogen (N) isotope analyses of lake sediments
102 from five lakes on sub-Antarctic Macquarie and Campbell Islands (Figure 1, Supporting
103 Information Note 1), using Hg as a tracer to investigate how colonisation-driven ecological
104 change reshaped elemental cycling in remote Southern Hemisphere ecosystems during the
105 Anthropocene. We assess: (1) how elemental sources and cycling differed among lakes with
106 contrasting ecological baselines; (2) how ecological disturbance following European colonial
107 practices altered nutrient and Hg pathways cycling and accumulation; and (3) whether common
108 environmental drivers produced consistent or divergent isotope responses across sub-Antarctic
109 lake ecosystems.

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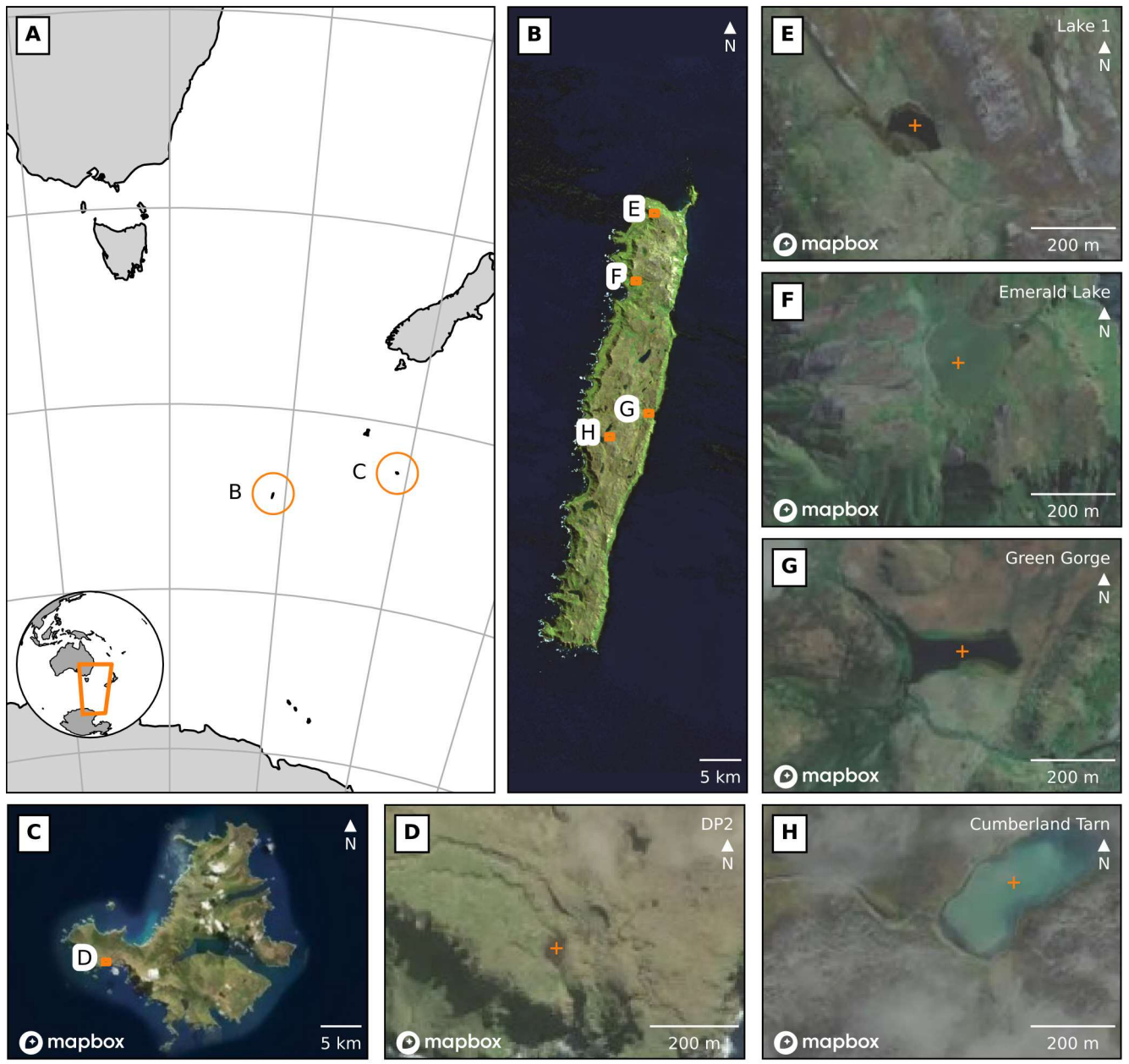


Figure 1 A) Location of Macquarie and Campbell Islands; B Macquarie Island (imagery acquired by the Operational Land Imager-2 on Landsat 9, February 8, 2022 <https://earthexplorer.usgs.gov/>); C) Campbell Island; D–H) lake sediment sampling sites on Macquarie and Campbell Islands; imagery in C, E,F,G, H is © Mapbox, © OpenStreetMap: <https://www.mapbox.com/>

Results

The 1850s marked the transition to widespread prospecting and unregulated industrial smelting emissions across the Australian continent (Mudd 2023). We therefore establish 1850 Common Era (CE) as a baseline delineator between background (pre-1850 CE) and active disturbance (post-1850 CE) in the Southern Ocean region. This date effectively consolidates the delayed

regional signatures of early unrecorded extraction with the immediate, acute impacts of global and regional industrialisation.

All lake sediment records encompass the past 500 years (Supporting Information Note 3; (Schneider et al. in press). Sedimentation rates remained relatively low and stable before approximately 1850 CE (<0.1 cm/yr). After c. 1850 CE, all lakes showed increased sediment accumulation rates, greater than any period of time pre-1850 CE (Figure 2). The magnitude of change after 1850 CE differed among lakes, with sedimentation increasing approximately twelve-fold in Lake 1, five-fold in Emerald Lake, four-fold in Cumberland Tarn, 1.5-fold in Green Gorge and 1.4-fold in DP2, although the increase in DP2 was not statistically significant (Supporting Information Note 3).

Collectively, TOC, $\delta^{13}\text{C}$, TN, $\delta^{15}\text{N}$, and TOC/TN indicate that ecological responses following the mid-19th century differed substantially among lakes (Figures 2, 3). Background conditions before 1850 CE were characterised by pronounced inter-lake variability in TOC, TN, TOC/TN, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ (Figures 2 and 3), reflecting contrasting ecological baselines. Pre-1850s TOC ranged from $0.8 \pm 0.5\%$ in Emerald Lake to $31 \pm 1.2\%$ in Lake 1, while TN ranged from $0.1 \pm 0.02\%$ in Emerald Lake to $2.5 \pm 0.2\%$ in Lake 1. The $\delta^{13}\text{C}$ ranged from $-27.5 \pm 0.27\text{‰}$ in DP2 to $-23.5 \pm 0.26\text{‰}$ in Cumberland Tarn. The $\delta^{15}\text{N}$ ranged from $-0.47 \pm 0.18\text{‰}$ in Cumberland Tarn to $11.3 \pm 0.58\text{‰}$ in DP2.

Post ~1850 CE, TOC increased in all Macquarie Island lakes, most significantly in Emerald Lake (8.8-fold) followed by Lake 1 (1.2-fold). Cumberland Tarn, and Green Gorge showed non-significant increases, DP2 declined slightly (Figure 2). In contrast, TN showed no consistent response, increasing significantly only in Emerald Lake (1.7-fold) and Green Gorge (1.4-fold), while remaining stable or declining slightly in the other lakes. Consequently, TOC/TN increased significantly in Emerald Lake, moderately in Lake 1 and Cumberland Tarn, remained unchanged in Green Gorge, and declined in DP2 (Figure 2).

Stable isotope ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) records similarly revealed divergent ecological trajectories post 1850 CE (Figures 2 and 3). The $\delta^{13}\text{C}$ became significantly more depleted in Lake 1, Emerald Lake and Green Gorge, whereas in Cumberland Tarn $\delta^{13}\text{C}$ became significantly enriched, and DP2 remained relatively unchanged (Figure 2). In contrast, $\delta^{15}\text{N}$ increased significantly in Lake 1 and Emerald Lake (2.03‰ and 1.26‰ increase respectively), but showed little change in

Cumberland Tarn, Green Gorge and DP2. Together, these proxies demonstrate that ecological change following ~1850 CE was highly site-specific despite occurring over a broadly synchronous time interval.

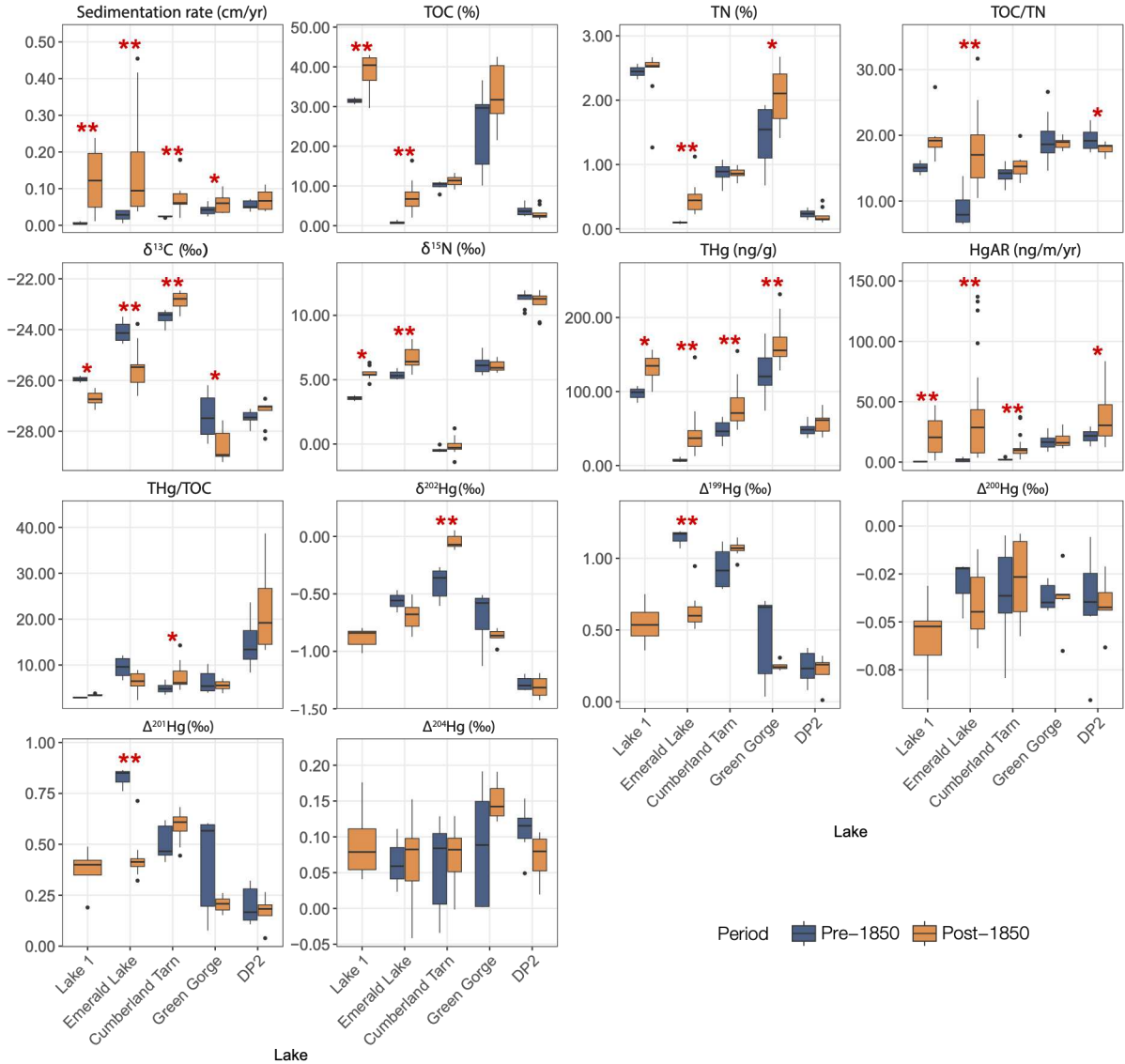


Figure 2 Boxplots of proxies by lake. Welch's *t*-test for pre and post 1850 CE significance in difference are presented with red asterisks: *p* < 0.01 "***", *p* < 0.05 "*". Blue indicates pre-1850s and orange indicates post 1850s values for each lake. Note: Pre-1850 CE Hg isotope data for Lake 1 are unavailable

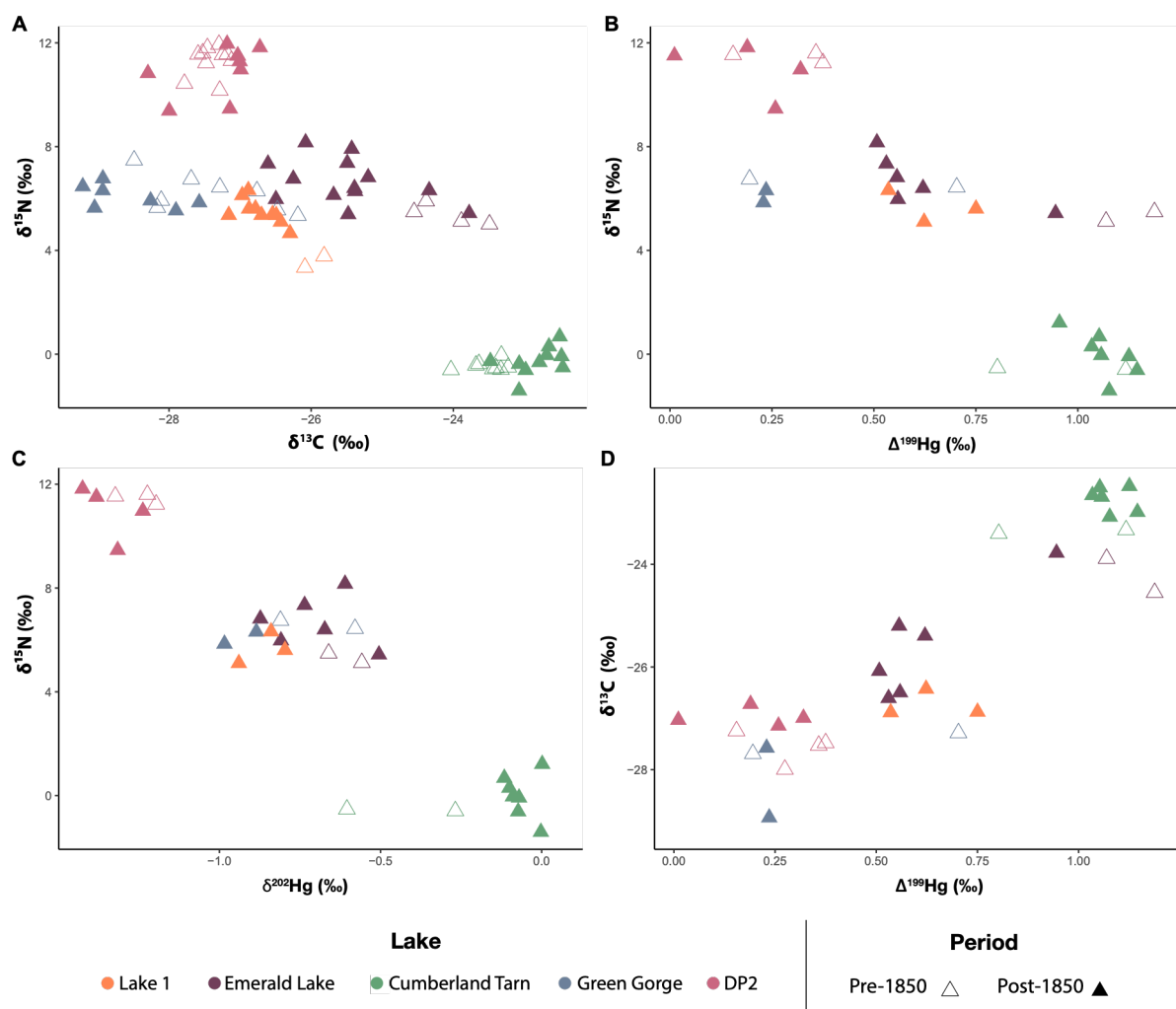


Figure 3 Relationship between $\delta^{202}\text{Hg}$, $\Delta^{199}\text{Hg}$, $\delta^{15}\text{N}$, $\delta^{13}\text{C}$ by lake. A) $\delta^{15}\text{N}$ vs $\delta^{13}\text{C}$ B) $\delta^{15}\text{N}$ vs $\Delta^{199}\text{Hg}$, C) $\delta^{15}\text{N}$ vs $\delta^{202}\text{Hg}$, and D) $\delta^{13}\text{C}$ vs $\Delta^{199}\text{Hg}$. Cumberland Tarn (green), DP2 (pink), Green Gorge (grey), Emerald Lake (purple), and Lake 1 (Orange). Note: Pre-1850 CE Hg isotope data for Lake 1 are unavailable

Total mercury concentrations (THg) remained relatively stable before ~1850 CE but increased in all lakes thereafter (Figure 2, Supporting Information Figure S 8). Background THg concentrations differed substantially among lakes, ranging from an average of 7 ± 3 ng/g in Emerald Lake to 97 ± 11 ng/g in Lake 1. Following ~1850 CE, THg increased significantly in all Macquarie Island lakes, with the largest increase observed in Emerald Lake (5.5-fold), followed by Cumberland Tarn (1.7-fold), Lake 1 (1.4-fold) and Green Gorge (1.3-fold). DP2 on Campbell Island showed only a modest non-significant increase (1.2-fold; Figure 2).

Mercury accumulation rates (HgAR) showed an even stronger response than concentrations because of increased sedimentation rates. The HgAR increased significantly in Lake 1, Emerald Lake, Cumberland Tarn and DP2, whereas Green Gorge showed only a minor increase

(Figure 2). Maximum HgAR generally occurred during the mid- to late 20th century before declining, although values remained well above background in all Macquarie Island lakes (Supporting Information Figure S 8).

Normalising Hg to TOC revealed contrasting responses among lakes. The THg/TOC increased significantly in Cumberland Tarn and non-significantly in Lake 1 and DP2 yet decreased in Emerald Lake and Green Gorge (Figure 2), indicating that post-1850 CE Hg enrichment cannot be explained solely by changes in organic matter accumulation.

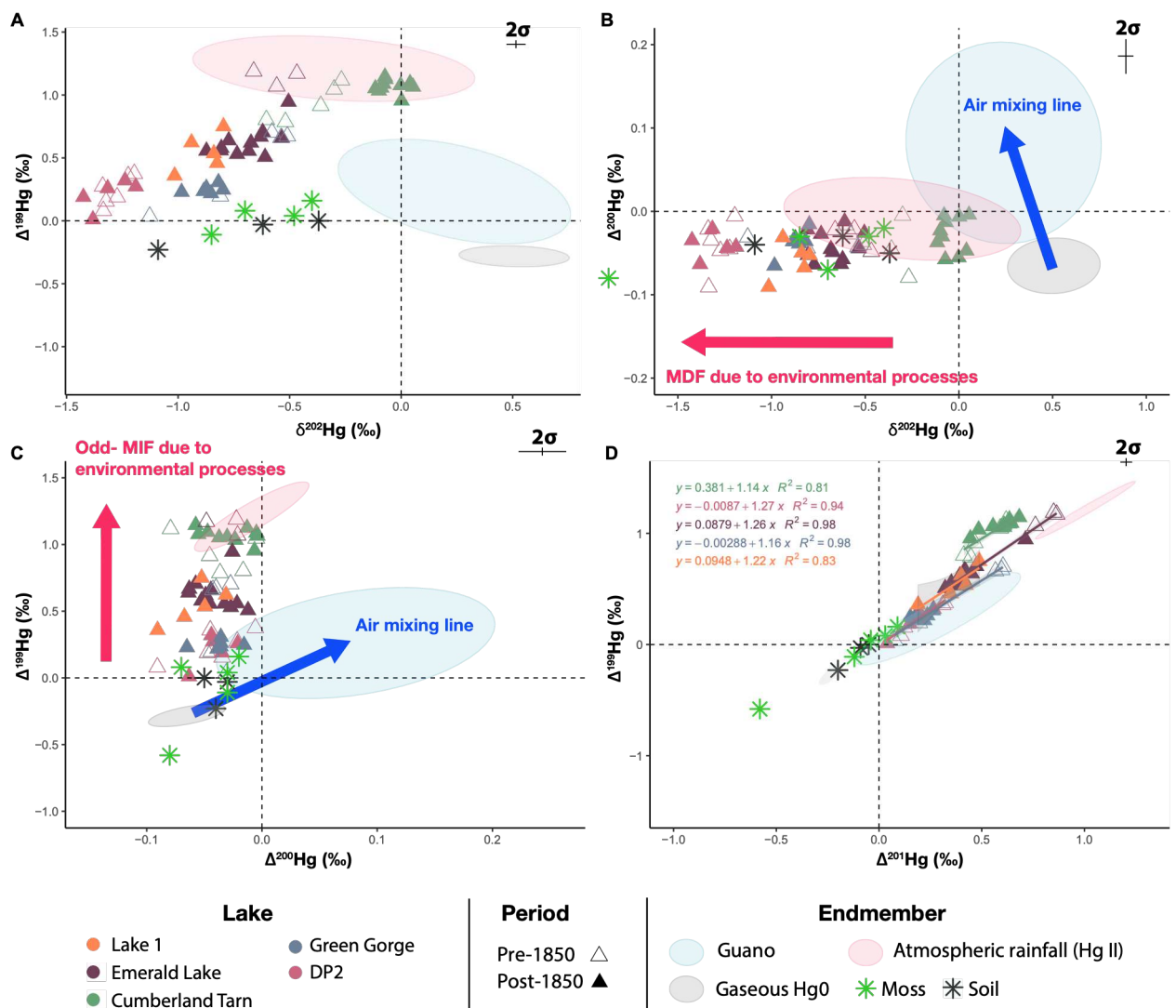


Figure 4 Comparison of Hg isotope signatures for source and process tracing. (A) $\Delta^{199}\text{Hg}$ vs $\delta^{202}\text{Hg}$ (B) $\Delta^{200}\text{Hg}$ vs $\delta^{202}\text{Hg}$, (C) $\Delta^{199}\text{Hg}$ vs $\Delta^{200}\text{Hg}$, and (D) $\Delta^{199}\text{Hg}$ vs $\Delta^{201}\text{Hg}$. Triangles indicate lake sediment. Moss (bright green) and Soil (Black) were measured as part of this study. Ellipses (1 SD) represent the endmembers, including guano from seal, penguin and albatross (pink)(Zheng et al. 2015; Li et al. 2026), atmospheric rainfall Hg^{II} (blue), and atmospheric gaseous Hg⁰ (grey) (Li et al. 2023). The blue arrow indicates the mixing of atmospheric Hg sources. The red arrows indicate the deviations of $\delta^{202}\text{Hg}$ (B)

and $\Delta^{199}\text{Hg}$ (C) values from those predicted by mixing lines based on $\Delta^{200}\text{Hg}$. Note: Pre-1850 CE Hg isotope data for Lake 1 are unavailable. MDF = mass dependant fractionation, MIF = Mass independent fractionation.

Mercury isotope compositions differed markedly among lakes despite the broadly synchronous increase in HgAR after ~1850 CE (Figures 2–4). Mass-dependent fractionation ($\delta^{202}\text{Hg}$) showed contrasting temporal trajectories. $\delta^{202}\text{Hg}$ values in lake sediments were mostly negative, falling between 0.05‰ and -1.42‰ (Figure 2, Supporting Information Table S 6). After the ~1850s, $\delta^{202}\text{Hg}$ became more negative in Emerald Lake, Green Gorge and DP2, but less negative in Cumberland Tarn (Figures 2 and 4).

Odd-mass independent fractionation ($\Delta^{199}\text{Hg}$ and $\Delta^{201}\text{Hg}$) also varied among lakes. $\Delta^{199}\text{Hg}$ for all lake sediment samples are positive, ranging between 0.01‰ and 1.19‰ (Figure 2, Table 1). Emerald Lake showed the largest decline from $1.14 \pm 0.06\text{‰}$ to $0.63 \pm 0.12\text{‰}$ following ~1850 CE, while Green Gorge and DP2 exhibited smaller decreases, and Cumberland Tarn increased slightly from $0.93 \pm 0.15\text{‰}$ to $1.07 \pm 0.05\text{‰}$ (Figures 2, 4A, Table 1). The $\Delta^{201}\text{Hg}$ was strongly correlated with $\Delta^{199}\text{Hg}$ across all lakes, with regression slopes ranging from 1.14 to 1.27 (Figures 3D and 4A). Even-mass independent fractionation ($\Delta^{200}\text{Hg}$ and $\Delta^{204}\text{Hg}$) varied within small ranges. All values for $\Delta^{200}\text{Hg}$ range between 0‰ and -0.08‰ (Figure 2, Table 1). The $\Delta^{204}\text{Hg}$ values fall between -0.03 and 0.19‰ (Figure 2). There is no significant change in average $\Delta^{200}\text{Hg}$ or $\Delta^{204}\text{Hg}$ values pre- and post-1850 (Figure 2).

For soil and moss, $\delta^{202}\text{Hg}$, $\Delta^{199}\text{Hg}$, and $\Delta^{200}\text{Hg}$ values ranged from -1.87‰ to -0.37‰, -0.58‰ to 0.16‰, and -0.08‰ to -0.03‰, respectively (Figure 4, Table 1, Supporting Information Table S 7).

Table 1 Hg isotope compositions and the estimated source contribution of a three endmember isotope mixing model on $\Delta^{199}\text{Hg}$ and $\Delta^{200}\text{Hg}$. Hg isotope compositions are reported in ‰ ± standard deviation. Isotope mixing model values are median estimated source contributions (%) of endmembers: guano (Zheng et al. 2015; Li et al. 2026); atmospheric rainfall Hg^{II} (Li et al. 2023); and catchment Hg (soil/moss, Supporting Information Table S 7). 2.5-97.5th percentile is indicated in parenthesis. Values are normalised to 100%.

Island	Lake	Period (CE)	Hg isotope composition (‰)		Estimated source contribution (%)		
			$\Delta^{199}\text{Hg}$	$\Delta^{200}\text{Hg}$	Guano	Rainfall Hg^{II}	Catchment (soil/moss)
Macquarie Island	Emerald Lake	Pre-1850	NA	NA	NA	NA	NA
		Post-1850	0.55 ± 0.15	-0.06 ± 0.02	49 (4–66)	16 (1–74)	35 (4–60)
		Pre-1850	1.14 ± 0.06	-0.03 ± 0.02	88 (75–97)	6 (0–22)	6 (0–17)
		Post-1850	0.63 ± 0.12	-0.04 ± 0.02	55 (4–71)	15 (1–79)	30 (3–54)

Campbell Island	Cumberland Tarn	Pre-1850	0.93 ± 0.15	-0.04 ± 0.03	80 (43–90)	9 (1–53)	11 (1–33)
		Post-1850	1.07 ± 0.05	-0.03 ± 0.02	85 (64–93)	7 (0–31)	8 (0–22)
	Green Gorge	Pre-1850	0.45 ± 0.31	-0.04 ± 0.01	60 (5–75)	17 (1–91)	23 (2–48)
		Post-1850	0.25 ± 0.03	-0.04 ± 0.02	21 (5–42)	21 (1–74)	58 (4–83)
	DP2	Pre-1850	0.24 ± 0.12	-0.04 ± 0.03	20 (4–42)	21 (1–74)	59 (4–87)
		Post-1850	0.21 ± 0.12	-0.04 ± 0.02	22 (6–39)	23 (1–73)	55 (4–86)
Endmember							
Macquarie Island	Catchment (soil/moss)	Post-1850	-0.08 ± 0.02	-0.04 ± 0.02	—	—	—
Amsterdam Island	Gaseous Hg ⁰	Post-1850	-0.22 ± 0.32	-0.06 ± 0.03	—	—	—
	Rainfall Hg ^{II}	Post-1850	0.25 ± 0.24	0.08 ± 0.09	—	—	—
Antarctica & Bird Island	Guano	Post-1850	1.13 ± 0.29	0.004 ± 0.05	—	—	—

*Note: Pre-1850 CE Hg isotope data for Lake 1 are unavailable

Discussion

Colonisation reshaped elemental cycling during the Anthropocene

The prevailing interpretation of post-1850 CE Hg increases in remote sedimentary archives is that they primarily reflect industrialisation associated emissions (Amos et al. 2015; Li et al. 2020; Lee et al. 2021). Our study demonstrates that industrialisation alone cannot explain Hg changes in remote Southern Hemisphere ecosystems. Although Hg_{AR} increased after the mid 19th century in all five sub-Antarctic lakes in both Macquarie and Campbell Island, Hg isotope values, $\delta^{15}\text{N}$, and $\delta^{13}\text{C}$, diverged among lakes (Figures 2 - 4). These differences indicate that each ecosystem responded differently to anthropogenic perturbations rather than industrialisation alone.

The timing of geochemical change across the records coincided with European arrival on Macquarie and Campbell islands. Following their discovery in the early 19th century, both islands experienced seal and penguin hunting and the introduction of invasive species (Saunders et al. 2013; Brown et al. 2022; Bird et al. 2025; Supporting Information Note 1). However, the intensity and type of ecological disturbance differed between the islands. Macquarie Island was significantly impacted by invasive rabbits, which caused widespread vegetation loss, soil destabilisation, erosion and food web re-organisation (Bergstrom et al. 2009; Saunders et al. 2013; Travers et al. 2021; Brown et al. 2022). Campbell Island experienced less wide-spread geomorphic disturbance than Macquarie Island, as livestock did not reach all parts of the island, in the way that rabbits did on Macquarie Island. This contrast helps separate the effects due to local colonisation from those of global industrialisation. Both

islands experience broadly similar atmospheric forcing (Saunders et al. 2018), and thus were exposed to increasing atmospheric Hg from industrial emissions, but their lake records responded differently (Figure 1, Table 1). In Macquarie Island, the post-1850 CE records show stronger changes in sedimentation rate, organic matter compositions, $\delta^{202}\text{Hg}$, $\Delta^{199}\text{Hg}$, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, while DP2 on Campbell Island shows comparatively little change (Figure 2). This suggests that the small increases in THg and HgAR reflect limited global industrial Hg inputs, while the different geochemical responses between islands reflect the type and intensity of ecological disturbance following European arrival and exploitative practices.

Interpreting post- colonial records in the Southern Hemisphere requires consideration of both ecological change and atmospheric forcing. The global atmospheric burden of Hg during the 19th and 20th centuries has increased as a result of industrialisation globally (Amos et al. 2015; Li et al. 2020), which has increased the amount of anthropogenic Hg available for deposition to remote ecosystems, including sub-Antarctic islands. However, Hg reaching lake sediments does not necessarily represent direct atmospheric deposition, because atmospheric Hg can first be incorporated into vegetation, soils and food webs before being transferred to aquatic systems and incorporated into sedimentary records (Meili et al. 2003; Driscoll et al. 2013; Zhang et al. 2018; Zhou et al. 2021).

Hg isotopes allow these atmospheric and ecosystem pathways to be distinguished. Even-MIF ($\Delta^{200}\text{Hg}$) is considered to originate primarily from upper-tropospheric and/or stratospheric photochemical reactions and is largely conserved during Hg transformations at the Earth's surface (Blum et al. 2014). $\Delta^{200}\text{Hg}$ can therefore provide a relatively conservative tracer of atmospheric Hg deposition pathways. In the Southern Hemisphere, atmospheric Hg(0) is characterised by negative $\Delta^{200}\text{Hg}$ values ($-0.06 \pm 0.02\text{‰}$), whereas Hg(II) in rainfall has positive values ($0.08 \pm 0.08\text{‰}$; Li et al. 2023). Using these atmospheric endmembers, our $\Delta^{200}\text{Hg}$ mixing model can be used to estimate the relative contribution of Hg derived from atmospheric Hg(0) as well as rainfall Hg(II). The consistently negative $\Delta^{200}\text{Hg}$ values measured in the lake sediments plot closer to the atmospheric Hg(0) endmember, with the model indicating that Hg(0)-derived Hg to the lake systems accounts for $75.4\% \pm 6.8\%$.

While $\Delta^{200}\text{Hg}$ constrains atmospheric deposition pathways, $\delta^{202}\text{Hg}$ and $\Delta^{199}\text{Hg}$ provide information on subsequent Hg cycling. The significant deviations of $\delta^{202}\text{Hg}$ and $\Delta^{199}\text{Hg}$ values among lakes relative to the mixing lines indicate that atmospherically derived Hg was

subsequently modified by terrestrial, biological and photochemical processes (Figure 4). The $\Delta^{199}\text{Hg}/\Delta^{201}\text{Hg}$ relationships provide further evidence for photochemical processing, with slopes within the ranges associated with Hg(II) photoreduction ($\sim 1.0\text{--}1.2$) and MeHg photodemethylation ($\sim 1.3\text{--}1.4$; (Bergquist and Blum 2007; Chandan et al. 2015; Zheng et al. 2015). However, the importance of these processes differs among lakes, reflecting differences in the pathways through which Hg was transferred and preserved.

Ecological baselines shape ecosystem responses to anthropogenic disturbance

While the five lakes in this study experienced broadly similar regional climate and atmospheric Hg forcing, the mixing models and multi-isotope records demonstrate that Hg was processed and delivered to the sediments through different pathways at each site (Figures 2-4; Table 1). Stable Hg isotope mixing models were constructed to assess the relative contribution of three endmembers (marine guano, rainfall Hg^{II} and catchment Hg; Figure 4) into each lake catchment. The relative contributions of the endmembers varied between the lakes and through time (Table 1). Before the 1850s, marine guano contributed greater than 75% of Hg to Emerald Lake and Cumberland Tarn, and almost 60% to Green Gorge, whereas its contribution to DP2 was only 22%, with catchment Hg making a 59% contribution. After the 1850s marine guano input decreased by more than 30% in Emerald Lake and Green Gorge, accompanied by increases in terrestrial Hg by more than 20%. In contrast, marine guano input in Cumberland Tarn increased by 5%, while DP2 remained largely unchanged with greater than 50% of Hg attributed to terrestrial catchment sources.

The separation of the lakes in $\delta^{15}\text{N}$, $\delta^{13}\text{C}$, and Hg isotopes before 1850 CE and changes post-1850 CE (Figure 3, Table 1) indicate that site-specific ecological baselines exert a strong control on how shared anthropogenic forcing (in this study, global industrialisation) is expressed in sedimentary Hg records (Figures 2-4). The lake records do not converge towards a common post-industrial signature in isotope composition despite collectively increasing HgAR. Instead, they maintain distinct isotopic relationships (Table 1), indicating different contributions and transportation pathways of atmospheric, terrestrial and biological derived Hg.

On Macquarie Island, the clearest ecological pathway is the biological transfer of marine-derived Hg from the Southern Ocean to terrestrial ecosystems. Emerald Lake, Lake 1, and to a lesser extent Green Gorge, have relatively positive $\Delta^{199}\text{Hg}$ and $\delta^{15}\text{N}$ values (greater than 0.5‰

and 3-5‰, respectively), consistent with marine Hg transported through seabird food webs (Table 1; (Bergstrom et al. 2002; Zheng et al. 2015; Li et al. 2026)). Elevated $\delta^{15}\text{N}$ indicates trophic enrichment and marine nutrient inputs to island ecosystems (Cocks et al. 1998; Bergstrom et al. 2002; Harding et al. 2004). The very positive $\Delta^{199}\text{Hg}$ is inherited from photochemical processing of marine MeHg before biological uptake (Bergquist and Blum 2007; Zheng et al. 2015; Sun et al. 2020). Their association therefore provides independent isotopic evidence for biological connectivity between the Southern Ocean and terrestrial sub-Antarctic lake ecosystems.

The strength and expression of this marine influence, however, depended on catchment characteristics. Emerald Lake shifted after 1850 CE from predominantly autochthonous organic matter towards substantially greater terrestrial input (Devesa-Rey and Barral 2012), as indicated by an increase in TOC/TN and decrease in $\delta^{13}\text{C}$ (Figure 2). This change coincided with shifts in Emerald Lake's Hg isotope composition (decrease in $\Delta^{199}\text{Hg}$), demonstrating that increased terrestrial material altered how biologically and atmospherically supplied Hg was processed and preserved in the lake (Table 1). Green Gorge provided a contrasting example: despite its coastal location, the record remained strongly influenced by terrestrial inputs throughout the sequence (Figure 2). Organic matter proxies, TOC, TOC/TN and $\delta^{13}\text{C}$ in combination with a comparatively lower $\Delta^{199}\text{Hg}$ (Figure 3) indicate that Green Gorge is terrestrially influenced by its large catchment (Supporting Information, Note 1 and 3). This has overwhelmed the marine-MeHg signature at Green Gorge. Whereas pre-1850 CE Emerald Lake maintains this marine-MeHg signature (Table 1). Thus, geographic proximity to the ocean alone does not determine marine MeHg from seabird influence. Instead, the size, ecological and hydrological characteristics of individual catchments (Meredith et al. 2022) regulate how marine, terrestrial, and atmospheric signals are integrated into sediments.

Cumberland Tarn and DP2 further demonstrate that marine Hg and nutrient signals do not necessarily co-vary together. Cumberland Tarn, with high $\Delta^{199}\text{Hg}$, $\delta^{13}\text{C}$, and relatively positive $\delta^{202}\text{Hg}$, both pre- and post-1850 (Figures 2-4), suggests strong marine MeHg contribution and less terrestrial input (Table 1). The low $\delta^{15}\text{N}$ (-0.5‰) in Cumberland Tarn and relatively high $\delta^{13}\text{C}$ (-23‰; suggesting marine contribution) in comparison with the other Macquarie Island lakes present further evidence of distinct ecological systems, even within a small geographic area. The relatively low $\delta^{15}\text{N}$ values at Cumberland Tarn may reflect limited guano inputs. Low pre-1850 CE Hg concentrations (47 ng/g) suggest that small guano inputs were sufficient to

influence the lake's Hg isotope composition, while the corresponding N inputs may have been insufficient to override the negative $\delta^{15}\text{N}$ signature of the surrounding upper-plateau catchment soils (Wilson et al. 2019). This is consistent with Cumberland Tarn and its associated catchment not being a major breeding or nesting site (Australian Antarctic Data Centre 2021), suggesting that limited biological inputs could have altered Hg isotope composition without substantially affecting the bulk sediment $\delta^{15}\text{N}$ signal. Cumberland Tarn consequently shows that $\Delta^{199}\text{Hg}$ and $\delta^{15}\text{N}$ can trace different components of biological transport and should not be expected to respond proportionally across ecological settings.

DP2 on Campbell Island represents a fundamentally different biological pathway. DP2 is strongly enriched in $\delta^{15}\text{N}$ ($>11\text{‰}$), indicating substantial trophic or biological influence (Bergstrom et al. 2002), yet $\Delta^{199}\text{Hg}$ remains below 0.5‰ , and $\delta^{202}\text{Hg}$ is negative (Figures 3 and 4). This combination is inconsistent with the marine-MeHg pathway evident in the Macquarie Island lakes and instead indicates a greater contribution from atmospheric and terrestrially cycled Hg (Table 1; Zheng et al. 2015; Li et al. 2026). The high $\delta^{15}\text{N}$ therefore records biological transport, but the Hg carried through that pathway has a predominantly terrestrial rather than marine origin. We suggest that this decoupling reflects biological transport through insectivorous (terrestrial) food webs rather than marine. Native species such as pipits (*Anthus novaeseelandia*), snipe (*Coenocorypha aucklandica perseverance*) and the Campbell teal (*Anas nesioti*) consume terrestrial invertebrates (Miskelly and Fraser 2006; Armitage 2007), providing a pathway through which terrestrially and atmospherically derived Hg can be biologically concentrated and redistributed. Isotopic studies of insectivorous birds demonstrate that Hg accumulated through terrestrial food webs can retain signatures closely linked to atmospheric Hg(0) rather than the positive $\Delta^{199}\text{Hg}$ characteristic of marine MeHg (Luo et al. 2024; Luo et al. 2025). This provides a potential mechanism for the combination of high $\delta^{15}\text{N}$ and relatively low $\Delta^{199}\text{Hg}$ observed at DP2.

Colonisation altered biological mercury transfer from sea to land

A defining characteristic of sub-Antarctic ecosystems is the strong ecological connectivity between the Southern Ocean and the terrestrial environments of its sub-Antarctic islands. Seabirds are recognised as major vectors transporting marine-derived nutrients to remote islands through feeding and guano deposition, linking marine food webs with terrestrial nutrient cycling (Otero et al. 2018). Our isotope results demonstrate that this biological

connectivity also extends to Hg. Before European disturbance, the relationships between $\Delta^{199}\text{Hg}$ and $\delta^{15}\text{N}$ indicate that biological transport was an important pathway transferring marine-derived Hg into several Macquarie Island lake catchments. This established an ecological pathway through which changes to seabird populations and food webs could directly alter Hg cycling.

European colonisation fundamentally altered biological pathways on Macquarie Island by reorganising wildlife communities and trophic interactions. The intensive exploitation of seals and penguins for furs and oil, in conjunction with the impacts of invasive species, including rabbits, cats, and rats changed the abundance, distribution and diets of native fauna (Bergstrom et al. 2009; Saunders et al. 2013; Travers et al. 2021; Brown et al. 2022). These changes altered not only nutrient transfer between ocean and land, but also the biological transport of Hg. Emerald Lake provides the clearest evidence of this disruption. After the mid-19th century, $\Delta^{199}\text{Hg}$ declined while $\delta^{15}\text{N}$ increased, indicating that the relationship between marine-derived Hg and trophic nutrient inputs had changed (Figures 2-4, Table 1). This apparently contradictory response can be explained by food web re-organisation. On Macquarie Island, Brown Skuas (*Stercorarius antarcticus lonnbergi*), shifted from feeding on burrowing petrels, as their population declined, to introduced rabbits, as their population increased (Travers et al. 2021). The Hg isotope mixing model indicates a reduction of greater than 30% of marine guano input (Table 1). This shift from a marine to a terrestrial diet explains both the reduction in the transfer of photochemically processed marine MeHg while maintaining an enriched $\delta^{15}\text{N}$ signature (Figure 2-4). The simultaneous increase in TOC and TOC/TN and depletion in $\delta^{13}\text{C}$ further indicate increasing terrestrial material, and a reduction in marine guano entering Emerald Lake (Figure 2 and Table 1). Human arrival and the introduction of invasive species therefore altered Hg cycling through two connected processes: 1) food web reorganisation reduced marine biological Hg transport; while 2) vegetation disturbance and erosion increased terrestrial input.

The contrasting response of Cumberland Tarn demonstrates that this disruption was not uniform across Macquarie Island (Figure 2 and Table 1). Cumberland Tarn retained a post-1850 CE elevated $\Delta^{199}\text{Hg}$ and enrichment in $\delta^{13}\text{C}$ (0.67‰ increase), indicating that marine influence was maintained despite widespread ecosystem changes due to the introduction of invasive species. The relative stability of TOC and TOC/TN (Figure 2) further suggests less catchment disturbance in Cumberland Tarn than at Emerald Lake. Thus, where ecological conditions

preserved marine-terrestrial biological connectivity, marine Hg transfer continued despite the broader transformations occurring on the island.

DP2 on Campbell Island, which experienced different ecological disturbances than Macquarie Island, including the absence of rabbits, provides an important lower-geomorphic disturbance comparison. Its consistently low $\Delta^{199}\text{Hg}$, negative $\delta^{202}\text{Hg}$ and enriched $\delta^{15}\text{N}$ values (Figures 3 and 4), with no significant change post-colonisation, indicates that biological transport can remain important without transfer of marine MeHg. This decoupling is consistent with a different ecological pathway involving terrestrial insectivorous food webs rather than the seabird-mediated marine pathway evident on Macquarie Island (Table 1).

Together, these records demonstrate that colonisation and the introduction of invasive species changed not only the amount of Hg reaching sub-Antarctic lakes, but also the biological pathways through which Hg moved across ecosystem boundaries. The magnitude and direction of these changes depended on local food webs, wildlife communities and the severity of catchment disturbance. This provides a mechanistic basis for understanding why broadly similar increases in Hg accumulation rates were accompanied by divergent Hg, $\delta^{13}\text{C}$, and $\delta^{15}\text{N}$ isotope trajectories among the five lakes.

We therefore propose a conceptual model in which industrialisation and colonisation acted through complementary yet distinct mechanisms to reshape elemental Hg cycling in remote World Heritage sub-Antarctic island lakes (Figure 5). Industrialisation increased the atmospheric availability of Hg, while colonial practices, primarily through the introduction of invasive species such as rabbits, altered how this excess Hg was intercepted, transported, and stored within island ecosystems. The $\Delta^{200}\text{Hg}$ mass balance model indicates that a substantial proportion of the Hg preserved in the sediments ultimately derives from atmospheric $\text{Hg}(0)$ deposition ($75.4\% \pm 6.8\%$), but the $\Delta^{199}\text{Hg}$ – $\Delta^{200}\text{Hg}$ source model demonstrates that this Hg reached individual lakes through differing combinations of atmospheric/rainfall deposition, catchment transport and biological transfer (Table 1). Thus, the atmospheric origin of Hg and the pathway by which it enters the sedimentary archive are not equivalent.

The exploitation of wildlife, and the introduction of invasive species, subsequently modified nutrient cycling and food-webs through landscape degradation (Bergstrom and Selkirk 2007;

Saunders et al. 2013; Travers et al. 2021). Colonisation therefore not only shifted the quantity of Hg entering the lake, it also changed the relative importance of terrestrial and biological pathways through which atmospherically derived Hg was transferred and preserved (Table 1). This provides a mechanistic explanation for why broadly similar increases in HgAR are accompanied by strongly divergent Hg, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ isotope trajectories among the five lakes (Figure 3, Table 1). The relative importance of these different pathways is not uniform between lakes (Figure 5). This highlights a key limitation of interpreting HgAR alone. Although HgAR increased broadly across all records (Figure 2), the accompanying changes in THg/TOC, $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ and Hg isotope compositions were lake-specific (Figures 2-4, Table 1), demonstrating that similar HgAR trends can result from different ecological and geomorphic processes.

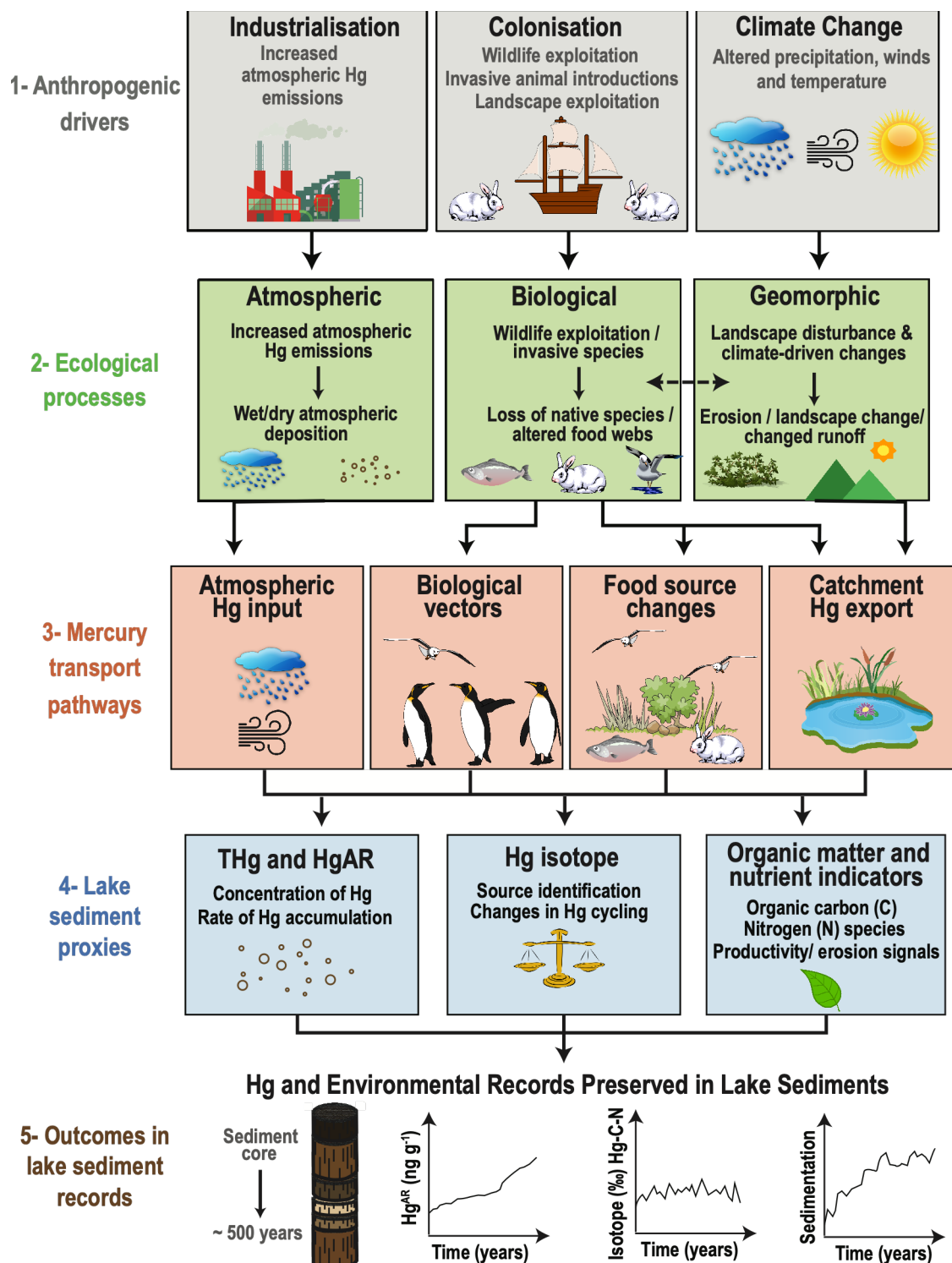


Figure 5 Conceptual diagram of possible pathways to altered Hg cycling and deposition in sub-Antarctic island systems.

Together, $\delta^{13}C$, $\delta^{15}N$ and Hg isotopes provide a more complete mechanistic framework for understanding Hg sources and cycling, thereby allowing biological and geomorphic consequences of local colonisation to be separated from increasing global industrial Hg emissions. The conceptual model therefore links a common atmospheric Hg forcing to

divergent sedimentary responses mediated by local ecological baselines, food web structure and catchment disturbance.

Implications for interpreting Anthropocene Hg records

Our findings demonstrate that ecological transformation is a fundamental, yet largely overlooked, component of Anthropocene Hg records in the Southern Hemisphere. For more than three decades, increases in HgAR preserved in remote lake sediments have largely been interpreted as evidence of increasing industrial emissions and long-range atmospheric transport (Amos et al. 2015; Li et al. 2020; Lee et al. 2021). While industrialisation undoubtedly increased global Hg emissions, our results demonstrate that ecological responses to local colonisation substantially modified how this Hg was transferred, transformed, and ultimately preserved within sub-Antarctic ecosystems. This distinction is important because conceptual models of Anthropocene Hg enrichment have been developed largely based on Northern Hemisphere records, where industrialisation occurred earlier and ecosystems differ from the strongly ocean-influenced environments of the Southern Hemisphere. Current conceptual models may therefore not fully capture the diverse ecological processes that influence Hg cycling across different environments (Otero et al. 2018; Schneider et al. 2023).

Our results caution against interpreting HgAR as a direct proxy for atmospheric Hg deposition. Remote lake ecosystems are not passive archives of atmospheric Hg deposition. Instead, the sediments integrate atmospheric inputs with ecosystem-specific processes including biological transfer, catchment transport, and biogeochemical cycling. Similar increases in HgAR can consequently arise from different combinations of these processes operating against contrasting ecological baselines. Integrating $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ and Hg isotope signatures provides a means of resolving this complexity by distinguishing organic matter and nutrient pathways from Hg sources and transfer processes. In this framework, Hg isotopes can serve not only as tracers of changing Hg availability, but also of the ecological pathways through which Hg moves across landscapes (Figure 5).

Most significantly, our study demonstrates that colonisation-driven ecological transformation can fundamentally alter elemental cycling and, thereby modify how Anthropocene Hg signals are generated and preserved. Reconstructing past Hg pollution therefore requires consideration not only of changes in atmospheric emissions, but also of the ecosystems through which that Hg was processed. Incorporating ecological context into interpretations of sedimentary Hg

records will provide a more robust basis for reconstructing Southern Hemisphere Hg histories and, ultimately, a more globally representative understanding of the Anthropocene Hg cycle.

Methods

Radiocarbon dating: Lake 1, Emerald Lake and Cumberland Tarn were radiocarbon (^{14}C) dated by Schneider et al. (in press) using 13, 10, and 11 macrofossil ages, respectively. Macrofossil ages were used for Green Gorge ($n = 14$) and DP2 ($n = 10$) as part of the present study. Macrofossils were selected under a microscope (Zeiss Stemi 2000 stereomicroscope), and processed with a modified acid-base-acid protocol (Norris et al. 2020). Due to the small sample size and fragility of the macrofossils, the standard method was modified with a lower base concentration (0.1M NaOH) and multiple base extractions. All samples were then combusted and graphitised and measured for ^{14}C by Accelerator Mass Spectrometry (AMS) following standard methods 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). To ensure consistency between dates measured at Australian Nuclear Science and Technology Organisation (ANSTO) and The Research School of Earth Science (RSES) at the Australian National University (ANU), duplicate samples were run in both labs, which fell within standard error for the two facilities (Supporting Information Table S 3)

Lead-210 dating: Lake 1, Emerald Lake and Cumberland Tarn were lead-210 (^{210}Pb) dated by Schneider et al. (in press) using nine sediment ages for Lake 1, and 10 for both Emerald Lake and Cumberland Tarn. Nine sediment ages were analysed for Green Gorge and four sediment ages for DP2 (Supporting Information Table S 4). 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 change in unsupported ^{210}Pb were used to calculate accumulation rates from the half-life of ^{210}Pb (22.26 ± 0.22 yr) (Appleby and Oldfield 1992). Unsupported ^{210}Pb activities were insufficient to construct a reliable chronology; therefore the DP2 chronology relied solely on radiocarbon dating (Supporting Information Note 3).

Plutonium-239,240 analysis: Thirteen samples from Emerald Lake were analysed for plutonium-239,240 by Schneider et al. (in press) to develop a higher resolution chronology for the 20th century (Supporting Information Note 3).

Age-depth modelling: Age-depth models established in Schneider et al. (in press) and this present study are reported in common era (CE). Details of the Age-depth models for Lake 1, Emerald lake, and Cumberland Tarn are found in Schneider et al. (in press) and in Supporting Information Note 3. The chronology for Green Gorge, Lake 1, Emerald Lake, and Cumberland Tarn was constructed using the Bayesian modelling approach using the R package ‘rplum’ v.1.0.0 (Aquino-López et al. 2018). The package can integrate both ^{210}Pb , ^{226}Ra and the model produced applies an autoregressive gamma process, in addition to a constant ^{210}Pb flux assumption, to derive sediment chronologies. Radiocarbon ages were incorporated into the age-depth model and calibrated using the Southern Hemisphere calibration curve (Hogg et al. 2020). Due to the lack of ^{210}Pb data for DP2, a chronology for DP2 was constructed using ‘rbacon’ 4.3.3 (Blaauw and Christen 2011), which uses the same Bayesian modelling approach as rplum, in combination with Markov Chain Monte Carlo techniques (Blaauw and Christen 2011; Hogg et al. 2020; Supporting Information Note 3).

Total and stable isotope carbon and nitrogen analyses: Total organic carbon (TOC) and total nitrogen (TN) were analysed in conjunction with their stable isotope ratios $^{13}\text{C}/^{12}\text{C}$ and $^{15}\text{N}/^{14}\text{N}$, henceforth referred to as $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, respectively. Analyses were conducted at the Australian Nuclear and Scientific Organisation (ANSTO) using an elemental analyser (Thermo Fisher Flash 2000 EA). A two point calibration is employed to normalise the data, utilising standards that bracket the samples being analysed (Ohlsson and Wallmark 1999). Two quality control references were also included in each run. Samples were calibrated using a methionine standard (B2045, Elemental MicroAnalyses). Results are accurate to 1% for both N % and C % and ± 0.3 ‰ for $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ (Supporting Information Table S 12).

Isotope ratios are calculated as follows:

$$\delta^{13}\text{C} = \delta(^{13}\text{C}/^{12}\text{C})_{\text{sample/reference}} = \frac{R(^{13}\text{C}/^{12}\text{C})_{\text{sample}} - R(^{13}\text{C}/^{12}\text{C})_{\text{reference}}}{R(^{13}\text{C}/^{12}\text{C})_{\text{reference}}}$$

$$\delta^{15}\text{N} = \delta(^{15}\text{N}/^{14}\text{N})_{\text{sample/reference}} = \frac{R(^{15}\text{N}/^{14}\text{N})_{\text{sample}} - R(^{15}\text{N}/^{14}\text{N})_{\text{reference}}}{R(^{15}\text{N}/^{14}\text{N})_{\text{reference}}}$$

The ratio between TOC and TN (TOC/TN) was calibrated for their respective atomic weight and was therefore multiplied by 1.167 and is reported as the atomic TOC/TN ratio to better determine the source of organic matter.

Total mercury concentration: Total Hg (THg) analyses were conducted at the Australian National University (ANU) School of Culture History and Language (CHL) using a Milestone Direct Mercury Analyser (DMA-80 Tri-cell; Milestone, Bergamo, Italy) according to the USEPA method 7473 (USEPA (US Environmental Protection Agency) 1998). THg was determined for 23 samples from Lake 1, 33 from Emerald Lake, 36 from Cumberland Tarn, 43 from Green Gorge and 28 from DP2. Sampling resolution was between 0.5 – 1 cm, depending on the sediment record (Supporting Information Note 2).

Mercury Accumulation Rate: THg accumulation rate (THgAR, ng/m²/yr) was calculated as follows:

$$\text{THgAR} = (\text{SR} \times \text{DBD} \times \text{Hg}) \times 10$$

where SR = sedimentation rate (cm/yr); DBD = dry bulk density (g/cm³); Hg = Hg concentration (ng/g). The factor of 10 is applied for unit conversion purposes.

Mercury pre-concentration: A total of five samples were analysed for Lake 1, 18 for Emerald Lake, 17 for Cumberland Tarn, 11 for Green Gorge and DP2 for stable Hg isotopes. Additionally, five moss and three soil samples from Macquarie Island were analysed. We selected samples using THg and THg/TOC values, capturing down-core variability and background concentration throughout the sediment records. The sediment records from Emerald and Cumberland Tarn were analysed at a higher resolution. Between 0.06 and 2.7 g of each sample was preconcentrated and purified for Hg using the combustion-trapping protocol, via a drying/decomposition-catalyst system of the DMA (Sun et al. 2025a). The volatilized Hg was trapped in an impinger containing an oxidized solution where Hg(0) is oxidized to Hg(II) (Sun et al. 2025b). Procedural blanks, and certified reference material (GSD-11), run every five samples were processed with the samples in the same manner. The procedural blanks of double-stage tube furnaces and drying/decomposition-catalyst systems accounted for <1% of Hg mass in sample trapping solutions. The Hg preconcentration recoveries were 91 ± 5.8% for procedural CRMs and 85.4 ± 17.5% for samples. Before Hg isotope measurement, all the trapping solutions were diluted at least two times by Milli-Q 18.2 Ω deionized water.

Stable mercury isotope measurement: Stable Hg isotope ratios were measured using a custom-made cold vapor generation (CVG) system coupled to a multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS, Nu Plasma 3D) at Tianjin University, China, according to established protocols (Shi et al. 2023; Cao et al. 2025; Li et al. 2025). Faraday cups were configured to simultaneously monitor six Hg isotopes (^{198}Hg , ^{199}Hg , ^{200}Hg , ^{201}Hg , ^{202}Hg , ^{204}Hg) and two thallium (Tl) isotopes (^{203}Tl and ^{205}Tl). The acquisition time was 7 min per sample (3 blocks $\times$ 33 cycles, 4.2 s integration time per cycle). Sample trapping solutions were diluted with ultra-pure water to yield Hg concentrations of 0.5-1 ng/mL. The solutions were introduced into the CVG system via a peristaltic pump and continuously mixed online with a 0.03 g/mL SnCl_2 solution to reduce Hg^{2+} to Hg^0 vapor. A typical ^{202}Hg signal intensity was ~ 1.8 V per ng mL^{-1} Hg at a sample uptake rate of 0.6 mL/min. Following each sample or standard measurement, the system was rinsed with the sample matrix solution for five minutes to prevent carryover, ensuring that blank signals were $<0.5\%$ of the preceding measurement. Instrumental mass bias was corrected using both the NIST 997 Tl internal standard and the NIST 3133 Hg standard-sample bracketing method. The Tl standard solution (~ 15 $\mu\text{g/mL}$ in 3% HNO_3) was introduced via an Aridus II desolvation nebulizer, yielding a ^{205}Tl signal intensity 3-5 times greater than that of ^{202}Hg . The bracketed NIST 3133 standard solutions were matrix- and concentration-matched to sample solutions within $\pm 10\%$.

The Hg isotope ratio is expressed as $\delta^{\text{xxx}}\text{Hg}$ (‰, xxx = 199, 200, 201, 202, and 204) by normalizing to the NIST 3133:

$$\delta^{\text{xxx}}\text{Hg}(\text{‰}) = \left(\frac{(^{\text{xxx}}\text{Hg}/^{198}\text{Hg})_{\text{sample}}}{(^{\text{xxx}}\text{Hg}/^{198}\text{Hg})_{\text{NIST SRM-3133}}} - 1 \right) \times 1000 \quad (1)$$

Mass-independent fractionation (MIF) value is expressed as $\Delta^{\text{xxx}}\text{Hg}$ (‰, xxx = 199, 200, 201, and 204), representing the difference between the measured $\delta^{\text{xxx}}\text{Hg}$ value and that predicted from $\delta^{202}\text{Hg}$ using a kinetic MDF law:

$$\Delta^{\text{xxx}}\text{Hg}(\text{‰}) = \delta^{\text{xxx}}\text{Hg} - ^{\text{xxx}}\beta \times \delta^{202}\text{Hg} \quad (2)$$

The mass-dependent scaling factor $^{\text{xxx}}\beta$ is 0.2520 for ^{199}Hg , 0.5024 for ^{200}Hg , 0.7520 for ^{201}Hg , and 1.4930 for ^{204}Hg .

Internal analytical precision of Hg isotope ratios (e.g., $^{202}\text{Hg}/^{198}\text{Hg}$) was typically $\leq 0.04\%$ (1SE) per analysis. The instrument's long-term uncertainty was monitored using secondary standard NIST 8610, analyzed every five samples. The measured THg isotope values of procedural CRMs (GSD-11) analyzed during different analytical sessions were consistent with previously reported values (Supporting Information Table S 11). Typical 2s analytical uncertainties for samples were reported as larger 2SD uncertainties between secondary standard NIST 8610 and procedural CRMs (Blum and Bergquist 2007).

Statistical Analysis: All statistical analyses and data visualisations were undertaken in Rstudio v.3.0 (R Core Team 2023) using packages listed in Supporting Information Table S 2. Differences between pre- and post-1850 CE samples were assessed using Welch's-t-test which accommodates unequal variances and unequal sample sizes. The statistical significance was set at $P < 0.05$.

Stable isotope mixing model for quantifying atmospheric Hg deposition pathways: A two-endmember isotope mass balance model was used to estimate the relative contributions of atmospheric gaseous Hg^0 and atmospheric rainfall Hg^{II} of Hg deposited in the lake sediments. We use $\Delta^{200}\text{Hg}$ as a conservative tracer, as no significant MIF has been measured at the Earth's surface during Hg transformations (Blum et al. 2014; Li et al. 2023). This serves as an initial contribution of Hg^0 and atmospheric rainfall Hg^{II} , assuming environmental endmembers (soil/moss and guano, see stable isotope mixing models) and lake sediments predominantly receive their Hg from atmospheric deposition. Endmember distributions were defined from the measured $\Delta^{200}\text{Hg}$ values of samples classified as gaseous Hg^0 ($n = 23$) and rainfall Hg^{II} ($n = 17$), respectively, measured by Li et al. (2023).

To propagate variability in the endmember compositions into the source estimates, a Monte Carlo approach was implemented. For each sample, 100,000 iterations were performed. In each iteration, one $\Delta^{200}\text{Hg}$ value was randomly sampled with replacement from the observed gaseous Hg^0 and rainfall Hg^{II} dataset (Supporting Information Note 2).

Stable isotope mixing models for quantifying source contributions: A three endmember Monte Carlo isotope mixing model was developed to estimate the relative contributions of potential Hg sources for each lake, 1) guano ($n = 9$): seal and penguin guano, Ross Sea Antarctica (Zheng

et al. 2015) and Albatross guano, Bird Island (Li et al. 2026), 2) atmospheric rainfall Hg^{II} (n =17; Li et al. 2023), and 3) catchment soil/moss (n = 8).

For each sediment sample, 100,000 random source mixtures were generated. Source proportions were sampled by generating exponentially distributed random variables and normalising them so that the three source fractions summed to one. During each iteration, $\Delta^{199}\text{Hg}$ and $\Delta^{200}\text{Hg}$ values for each source were randomly sampled with replacement from the measured endmember datasets to incorporate the observed variability of each endmember into the model (Supporting Information Note 2).

To evaluate temporal changes in Hg sources, sediment samples were classified as pre-1850 CE or post-1850 CE. Median source contributions from individual samples were averaged within each period to compare changes in the relative contributions of atmospheric, ornithogenic and catchment-derived Hg associated with European impacts.

Data Availability

The authors declare that all data supporting the findings of this study are available within the paper and the Supporting Information.

Code Availability

Isotope mixing model code available: <https://github.com/margotschneider16/Mixing-models>

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Additional Information

Supplementary information is available in the Supporting Information document.

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928 **Competing Interests**

929 The authors declare no competing interests.

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