The European surface legacy-metal fingerprint is not detected in Australian catchment outlet sediments: a site-paired hemispheric contrast from harmonized open geochemical data

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Abstract

Natural-archive reconstructions show that anthropogenic lead deposition has been overwhelmingly concentrated in the Northern Hemisphere, yet continental-scale surface/deep screening statistics that would register this asymmetry in situ have only been reported for northern surveys. Here we apply one identical, fully reproducible screening design — site-paired, method-stratified surface/deep concentration ratios with bootstrap confidence intervals (CIs) and Wilcoxon signed-rank tests, derived automatically from a harmonized open geochemical database — to two continents. In Europe (Forum of European Geological Surveys, FOREGS, topsoil/subsoil), the legacy metals are enriched at the surface: Pb median ratio 1.24 (95% CI 1.19–1.30, n=389 site pairs), Hg 1.36. In Australia (National Geochemical Survey of Australia, NGSA, catchment outlet sediments, 0–10 cm over ∼60–80 cm), the same design on >1,000 site pairs per element finds no surface Pb excess: 0.982 (95% CI 0.973–0.993, n=1,064, inductively coupled plasma mass spectrometry, ICP-MS). Arsenic is surface-depleted (0.886, 0.867–0.905), Cu slightly depleted (0.955), and Zn indistinguishable from unity (1.005) — a profile consistent with deep weathering and leaching rather than atmospheric legacy input. Results are invariant to coordinate rounding. Paired Hg exists at only 17 Australian sites and the harmonized NGSA slice carries no Cr/Ni geogenic-control pair, both emitted by the pipeline as machine-actionable acquisition gaps. The contrast supports interpreting the European fingerprint as a Northern-Hemisphere industrial signature rather than a global default, and provides a Southern-Hemisphere baseline panel for the global background separation that the Minamata Convention's effectiveness evaluation currently lacks. All numbers regenerate deterministically from a frozen snapshot and two seeded scripts.

1 Introduction

Ice-core, peat and lake-sediment archives agree that anthropogenic Pb deposition rose to global ubiquity during the industrial era while remaining overwhelmingly concentrated in the Northern Hemisphere; Southern-Hemisphere archives record signals that are typically an order of magnitude weaker and later [5]. Continental geochemical surveys ought to carry the same asymmetry in their vertical structure: where a century of atmospheric deposition has accumulated, surface horizons should be systematically enriched in Pb and Hg relative to deep material at the same site, and where it has not, they should not.

The Northern-Hemisphere half of this expectation is established. The Geochemical Atlas of Europe of the Forum of European Geological Surveys (FOREGS) [7] reported topsoil/subsoil ratios of 1.66 for Hg and 1.36 for Pb (arithmetic means of site ratios, methods pooled) [3], and our companion screening study re-derived the fingerprint with method stratification and uncertainty (Hg 1.364, Pb 1.239, geogenic controls Ni/Cr $\approx$ 1) [4]. The Southern-Hemisphere half has, to our knowledge, not been tested with an equivalent site-paired design on open continental data.

Australia provides the natural test bed. The National Geochemical Survey of Australia (NGSA) sampled catchment outlet sediments — floodplain-like deposits near catchment outlets — at 1,315 sites covering $\sim$81% of the continent (one site per $\sim$5,200 km^2), at two depths: a Top Outlet Sediment (TOS, 0–10 cm) and a Bottom Outlet Sediment (BOS, $\sim$60–80 cm) [1, 2]. If industrial-era atmospheric deposition dominated the recent sediment surface, TOS/BOS ratios for Pb should sit measurably above unity, as topsoil/subsoil ratios do in Europe.

This paper asks one question: *does the European surface legacy-metal fingerprint replicate on a Southern-Hemisphere continent under an identical, automated screening design?* The answer is no, and the shape of the negative result — element-wise depletion rather than noise — is itself informative. We emphasize scope throughout: both panels are screening-level statements about harmonized open observations, not source attributions; media differ between continents (residual soil profiles in Europe, transported outlet sediments in Australia), and we treat that difference as a declared limitation and a proposed follow-up rather than an obstacle buried in prose.

2 Materials and methods

Figure 1 summarizes the design: two media on two continents enter one frozen harmonized snapshot, and an identical site-paired screening statistic is applied to both.

2.1 Harmonized database and surveys

Both panels derive from the same frozen snapshot of the Global Geochemical Atlas (GGA) pipeline described in the companion study [4]: a 191,715-record standardized database assembled autonomously from 29 open sources, with normalized units (mg kg^{-1} for solids), WGS84 coordinates, per-record method family, license and SHA-256 provenance, and four-dimensional confidence scores. The European panel uses FOREGS topsoil (0–25 cm) and subsoil (C-horizon, 50–200 cm) [7]; the Australian panel uses NGSA TOS and BOS [1]. NGSA data are published by Geoscience Australia under CC-BY 4.0; the harmonized slice contains 8,572 NGSA determinations of the five analytes As, Cu, Hg, Pb and Zn (Cr and Ni determinations are not present in the ingested NGSA files — see §3.4).

2.2 Site-paired screening design

The design is identical to the companion study and is applied without modification to both continents. Within one survey and one method family per element (ICP-MS for Pb, As, Cu, Zn on both NGSA layers; US EPA Method 7473 direct mercury analysis, DMA, for NGSA Hg), records with positive values and canonical coordinates are grouped into sites by rounding coordinates to 0.01°; per-site medians are taken per layer; and the surface/deep ratio $r = c_{\mathrm{surf}}/c_{\mathrm{deep}}$ is computed at sites where both layers exist. We report the median ratio, a 2,000-draw seeded bootstrap 95% CI of the median, the fraction of sites with $r > 1.5$, and a two-sided Wilcoxon signed-rank test on $\log r$. Sensitivity analyses repeat the procedure at 0.1° rounding. European reference values are taken unchanged from the companion run [4].

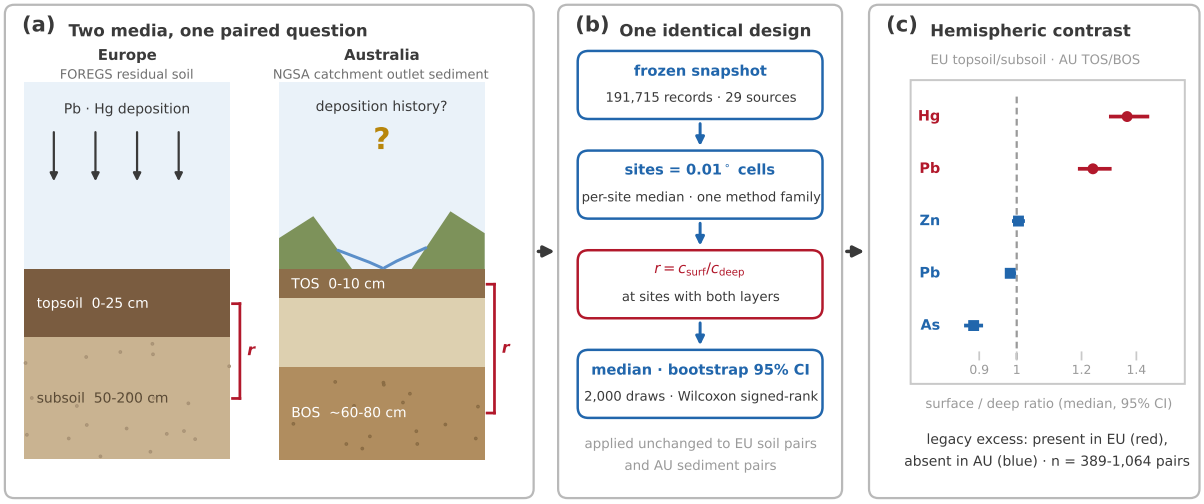

Figure 1: Study design. (a) The two media under comparison: European FOREGS residual soil profiles (topsoil 0–25 cm over C-horizon subsoil 50–200 cm), where atmospheric Pb and Hg deposition accumulates at the surface, and Australian NGSA catchment outlet sediments (Top Outlet Sediment, TOS, 0–10 cm, over Bottom Outlet Sediment, BOS, ∼60–80 cm), where a preserved surface excess is the open question. (b) Both panels derive from one frozen harmonized snapshot; the identical site-paired design (0.01° sites, per-site medians, one method family per element, surface/deep ratio r at sites with both layers, bootstrap 95% CIs, Wilcoxon signed-rank tests) is applied unchanged to each continent. (c) The resulting hemispheric contrast: surface enrichment of the legacy metals in Europe (Hg 1.364, Pb 1.239) against Australian ratios at or below unity (Pb 0.982, As 0.886), with Zn near unity on both continents.

2.3 Media caveat and controls

The two panels compare different media: European ratios are formed within residual soil profiles, Australian ratios within transported catchment sediments. BOS is older than TOS but is not guaranteed pre-industrial at every site, and fluvial mixing attenuates atmospheric signals; the Australian panel therefore tests for a *preserved surface excess*, not for the absence of deposition. Three design features bound the interpretation. First, the same-site pairing cancels catchment-scale lithology shared by both layers. Second, Zn — mildly enriched in Europe (1.027) — behaves as a quasi-reference in Australia (1.005, p=0.13), showing that the design does not manufacture spurious depletion. Third, the harmonized NGSA slice lacks the Cr/Ni geogenic controls used in Europe; this is recorded as an acquisition gap rather than silently ignored, and all Australian statements are correspondingly screening-level.

3 Results

3.1 No surface Pb excess on the Australian continent

Table 1 and Figure 4 give the paired ratios. Australian outlet sediments show no surface Pb enrichment: TOS/BOS median 0.982 (95% CI 0.973–0.993; n=1,064 pairs; 5.8% of sites >1.5). The European value under the identical design is 1.239 (1.194–1.299; 28.3% >1.5); the CIs are separated by ∼0.2 in ratio units, a first-order continental difference, not a marginal effect. As is

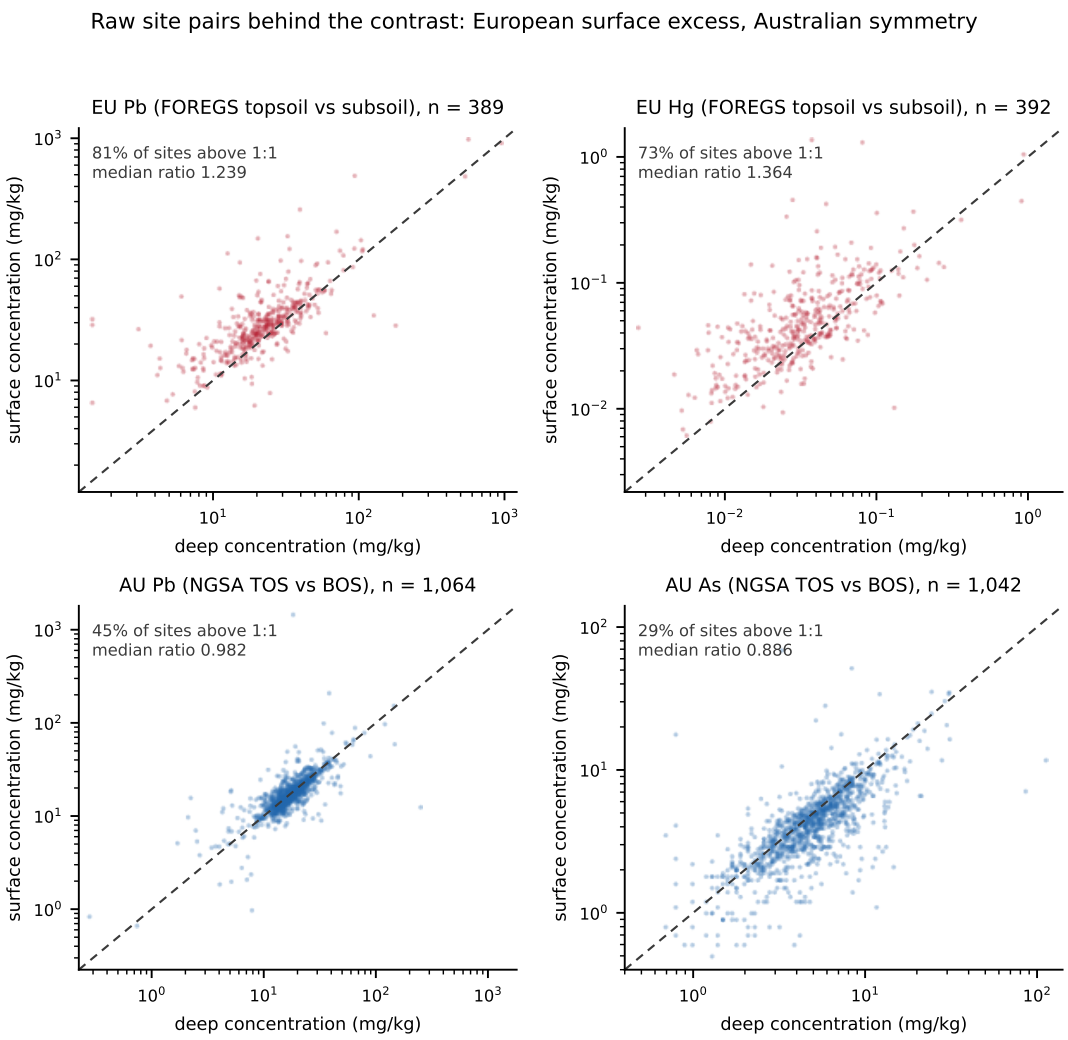

Figure 2: Raw site-paired concentrations (log-log; dashed line 1:1). Top row: FOREGS Europe — topsoil sits systematically above subsoil for Pb and Hg. Bottom row: NGSA Australia — Top Outlet Sediment straddles the 1:1 line for Pb and falls below it for As. Annotations give the share of sites above the 1:1 line and the median ratio; methods per panel as in Table 1.

surface-*depleted* in Australia (0.886, 0.867–0.905, p=3.9×10^{-45}) and Cu slightly so (0.955), while Zn is at unity (1.005). Ratios are unchanged at 0.1° rounding (Pb 0.982, n=1,049; As 0.884, n=1,028).

Figure 2 shows the raw site pairs behind these medians — the least processed view of the contrast. In Europe, 80.7% of Pb sites and 73.2% of Hg sites lie above the 1:1 line; in Australia the Pb cloud straddles it (44.6% above) and As sits clearly below (29.2%).

3.2 The full ratio distributions separate by continent

The hemispheric difference is not confined to the medians. Figure 3 overlays the empirical cumulative distribution functions (ECDFs) of the per-site $\log_2$ ratios, and two-sample Kolmogorov–Smirnov (KS) tests reject distributional equality in every panel — but with effect sizes that themselves rank the elements. Pb separates most strongly (KS D=0.453, p=2.2×10^{-53}): the

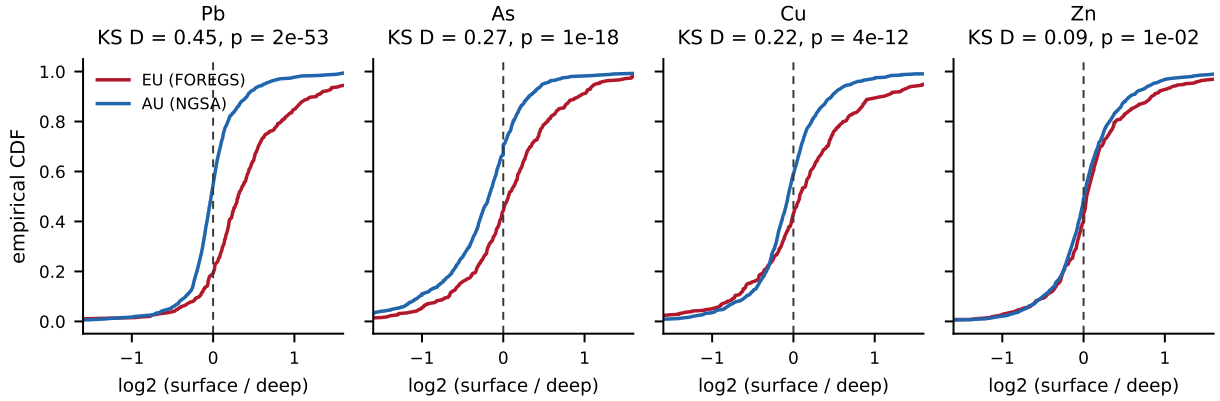

Figure 3: ECDFs of per-site $\log_2$ (surface/deep) ratios: Europe (FOREGS, red) versus Australia (NGSA, blue) for the four elements paired on both continents. Dashed line: no enrichment. Panel titles give the two-sample KS statistic and p-value.

Table 1: Surface/deep concentration ratios under one identical site-paired design: median, 95% bootstrap CI of the median, share of site pairs with ratio >1.5, and two-sided Wilcoxon signed-rank p. Panels — EU T/S: FOREGS topsoil/subsoil (residual soil; from the companion run [4]); AU T/B: NGSA Top/Bottom Outlet Sediment (transported sediment; this work). Method families: EU as in [4]; AU ICP-MS except Hg by DMA.

Panel	Element	n pairs	Median	95% CI	>1.5	p
EU T/S	Hg	392	1.364	1.303–1.444	41.3%	6.4×10^{-29}
EU T/S	Pb	389	1.239	1.194–1.299	28.3%	1.7×10^{-35}
EU T/S	Cu	384	1.055	1.008–1.112	21.1%	4.4×10^{-4}
EU T/S	As	384	1.045	1.008–1.105	18.2%	5.4×10^{-3}
EU T/S	Zn	387	1.027	1.014–1.058	15.0%	6.5×10^{-4}
EU T/S	Cr	387	0.976	0.947–1.000	9.6%	0.064
EU T/S	Ni	389	0.947	0.926–0.970	9.5%	1.2×10^{-3}
AU T/B	Hg	17	1.206	0.896–1.702	35.3%	0.045
AU T/B	Zn	1,057	1.005	0.992–1.018	9.6%	0.133
AU T/B	Pb	1,064	0.982	0.973–0.993	5.8%	0.035
AU T/B	Cu	1,055	0.955	0.942–0.968	6.8%	3.1×10^{-8}
AU T/B	As	1,042	0.886	0.867–0.905	3.6%	3.9×10^{-45}

European curve is shifted as a whole toward enrichment, not merely stretched in a tail. As (D=0.271) and Cu (D=0.217) separate through the Australian shift toward depletion. Zn — the quasi-reference of §2.3 — shows much the smallest separation (D=0.095, p=0.011), its two curves nearly coinciding: the element expected to behave alike on both continents does, which is itself a check that the design manufactures neither enrichment nor depletion.

3.3 Spatial structure: local, not continental

Mapping per-site log2 Pb ratios (Fig. 5) shows scattered local structure — patches of surface excess in parts of southwest and southern Australia interleaved with depletion — but no coherent continental belt comparable to the European 48°N enrichment belt of the companion study. Under

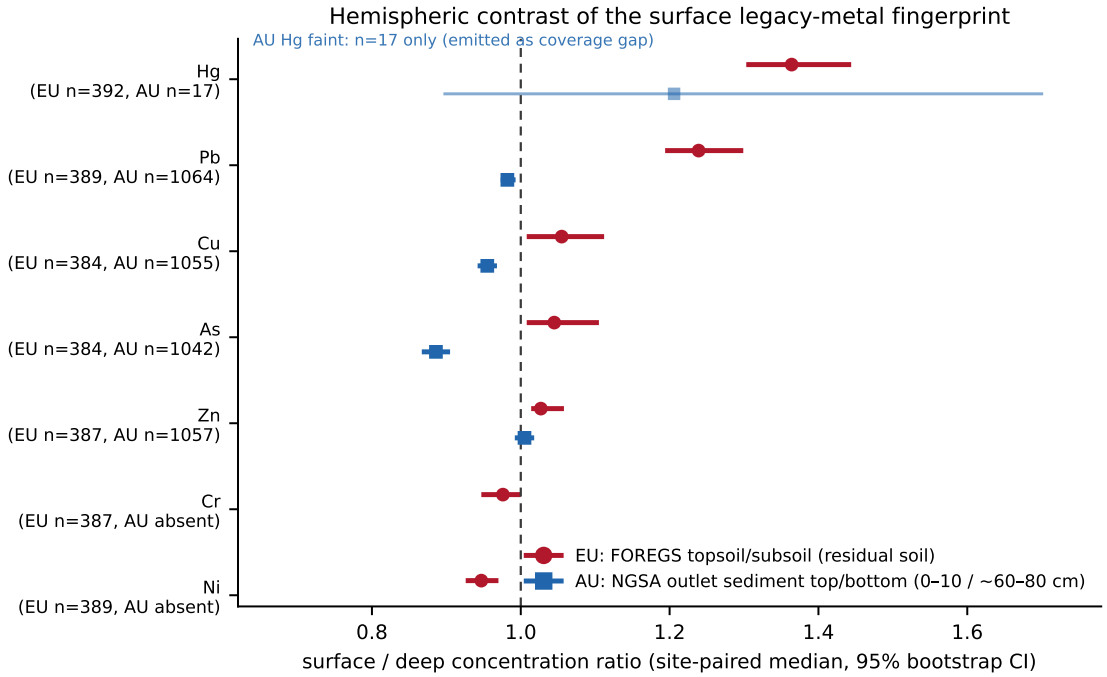

Figure 4: Hemispheric contrast under one screening design. Red circles: FOREGS topsoil/subsoil ratios (EU). Blue squares: NGSA Top/Bottom Outlet Sediment ratios (AU). Points are medians of per-site ratios; bars are 95% bootstrap CIs of the median. The Australian Hg estimate (faint) rests on 17 pairs and is emitted as a coverage gap rather than interpreted. Cr and Ni have no Australian pair in the harmonized slice. Dashed line: no enrichment.

a deposition-dominated regime the map would be expected to show broad coherent excess; under weathering- and transport-dominated regimes, spatially patchy ratios centred at or below unity, as observed.

3.4 Mercury and geogenic controls as documented gaps

Two absences are results in their own right, and the pipeline emits both as machine-actionable sampling/acquisition priorities. (1) Paired Hg exists at only 17 Australian sites (DMA method family); the Wilcoxon test is nominally significant (p=0.045) but the bootstrap CI of the median spans unity (0.896–1.702), so no inference is drawn — the Southern-Hemisphere half of the global soil-Hg background question remains observationally open. (2) The ingested NGSA files carry no Cr or Ni determinations, so the European geogenic-control check cannot yet be replicated; acquiring the NGSA aqua-regia/total multi-element extensions is queued as the corresponding acquisition task.

4 Discussion

What the contrast supports. Under one identical, automated design, the Northern-Hemisphere industrial fingerprint (surface Pb and Hg excess with geogenic controls at unity) is present in Europe and absent from Australian catchment sediments, where the profile instead matches deep

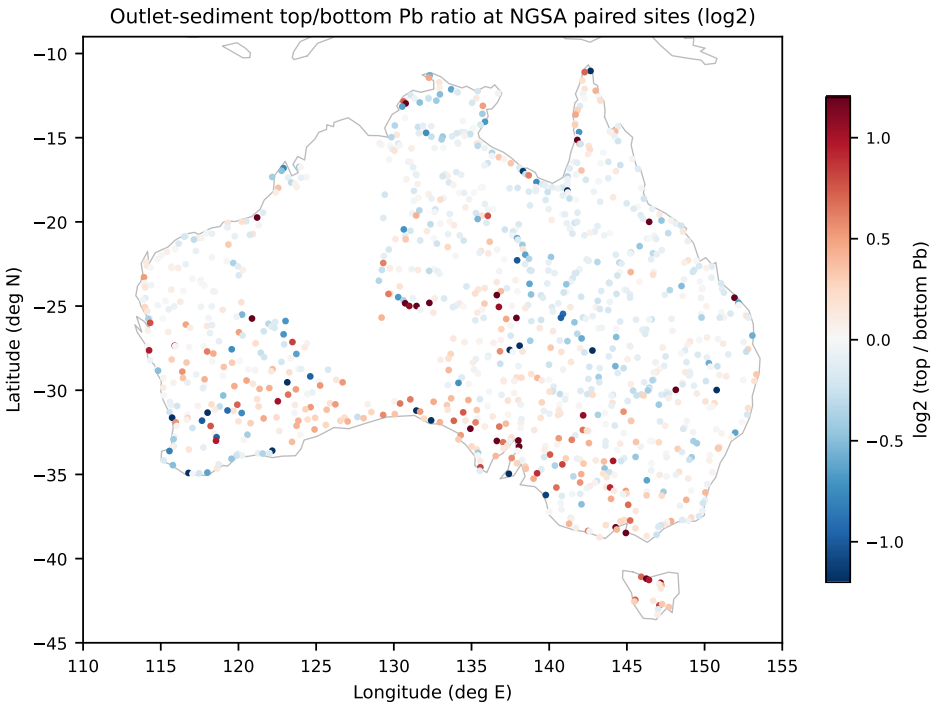

Figure 5: Top/Bottom Outlet Sediment Pb ratio (log2) at NGSA paired sites. Red: surface excess; blue: surface depletion. Land outline: Natural Earth.

weathering and leaching (As and Cu depleted at the surface, Zn at unity, Pb marginally below). This is the in-situ, continental-median counterpart of the hemispheric asymmetry long documented in natural archives [5]: legacy enrichment is a regional-industrial signature, not a property of surface earth materials in general.

Alternative explanations, bounded. The media difference of §2.3 is the principal confounder: fluvial mixing could dilute a genuine atmospheric excess in outlet sediments. Three observations bound this. Australian near-city and mining-adjacent sites do show local surface excess (Fig. 5), so the medium can record enrichment where inputs are strong; the As depletion signal is directional and highly significant ($p \sim 10^{-45}$), which mixing alone does not produce; and the European signal survives its own transported-media analogue in the companion study's floodplain-adjacent sources. A definitive separation — Australian residual soil profiles under the same design — is the natural follow-up and is already expressible as a pipeline acquisition task.

Position within the current frontier. The Minamata Convention's effectiveness evaluation reports that soil-Hg data cannot yet be separated into background and anthropogenic components, and its evidence base is dominated by Northern-Hemisphere surveys [8]. A Southern-Hemisphere continental panel in which the legacy fingerprint is demonstrably absent is exactly the background end-member that global separation requires — and the 17-pair Australian Hg count quantifies how far current open data are from providing it. For Pb, hemispheric asymmetry inferred from point archives [5] gains a survey-scale, site-paired confirmation.

New questions generated. (Q1) *Media separation*: do Australian residual soil profiles (e.g., state survey soil archives) reproduce the null, isolating deposition history from sediment transport? (Q2) *Depletion mechanism*: is the strong As surface depletion in outlet sediments a weathering/leaching signature of the deeply weathered Australian regolith, and does it covary

with regolith age maps? (Q3) *Southern-Hemisphere Hg baseline*: which acquisitions (NGSA Hg extensions, state surveys, Southern-Hemisphere national programs) would raise paired Hg coverage from 17 sites to statistical usability, and at what cost?

Limitations. Media differ between panels; BOS age is heterogeneous; Australian geogenic controls are absent from the harmonized slice; Hg is effectively unobserved; NGSA density is one site per ∼5,200 km^2; and no isotopes or chronology constrain input history. All statements are screening products intended to define, not close, the hemispheric question.

Reproducibility. Both panels regenerate from one frozen snapshot via seeded deterministic scripts; the Australian panel is produced by a ∼40-line pilot plus a figure script, unchanged from the companion methodology; the marginal analysis cost of extending a continental screening result to a second continent was minimal.

5 Conclusion

An identical site-paired screening design applied automatically to two continents shows that the European surface legacy-metal fingerprint does not replicate in Australian catchment outlet sediments: Pb sits at 0.982 (0.973–0.993) against Europe's 1.239, As is strongly surface-depleted, and the data needed to test Hg and the geogenic controls do not yet exist in open form — a gap the pipeline outputs as concrete acquisition targets. Hemispheric asymmetry of the anthropogenic legacy, previously documented in point archives, is thus visible directly in harmonized continental survey data.

Data and code availability

Input sources (licenses recorded per record): NGSA Geochemical Atlas of Australia data (Geoscience Australia, CC-BY 4.0) [1, 2]; FOREGS Geochemical Atlas of Europe (Geological Survey of Finland; research use with attribution) [7, 3]. The frozen snapshot, pilot and figure scripts (`p2_ngsa_pilot.py`, `p2_extra.py`, `p2345_addfigs.py`), result JSONs (`p2_results.json`, `p2345_addfigs_results.json`) and figure sources are archived with SHA-256 receipts in the pipeline's `research-products/paper-demo-20260817` directory.

Agent disclosure

This draft, including analysis design, statistics, figures and text, was produced end-to-end by an AI research agent operating the Global Geochemical Atlas skill; the topic was proposed by the skill's deterministic discovery-candidate generator (candidates dc-001–dc-004 of the 2026-08-15 world run) and has not yet completed human review. Screening-level framing is intentional and load-bearing.

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