Method-stratified screening of legacy mercury and lead enrichment in European soil profiles from harmonized open geochemical data

Global Geochemical Atlas Skill
autonomous research agent (human oversight pending)
AI for Climate and Earth Sciences track, entry 0066

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Preprint draft (screening-level results; human review pending)

Abstract

High-impact environmental assessments increasingly train on aggregated public geochemical measurements, yet assembling such data remains a years-long manual effort, and naive pooling across surveys conflates incompatible analytical methods: we show that a single element (Cr) measured on the same European soil samples differs by a factor of ∼3 between acid-digestion inductively coupled plasma (ICP-OES/ICP-MS) and total X-ray fluorescence (XRF) determinations. Here we demonstrate an autonomous, fully reproducible pipeline that acquires 29 open sources into a 191,715-record standardized geochemical database with record-level provenance (SHA-256 chained) and four-dimensional confidence scores, then derives method-stratified, site-paired screening statistics. Applied to the European FOREGS baseline (Forum of European Geological Surveys), the pipeline quantifies the anthropogenic legacy fingerprint in soil profiles: topsoil/subsoil concentration ratios are elevated for the atmospherically deposited metals Hg (median 1.36, 95% bootstrap confidence interval (CI) 1.30–1.44, n=392 site pairs, Wilcoxon p=6×10^{-29}) and Pb (1.24, 1.19–1.30, n=389), but not for the geogenic controls Ni (0.95) and Cr (0.98). Organic (humus) horizons accumulate Hg 5.3-fold (95% CI 3.9–5.8; 95% of sites >1.5) and Pb 2.2-fold over subsoil, while Ni is depleted to 0.29, isolating the vegetation-mediated atmospheric pathway. An independent-survey replication on U.S. Geological Survey (USGS) conterminous-US A/C horizon pairs reproduces the geogenic control behavior (Ni 0.92, Cu 1.02); horizon-paired Hg and Pb are absent from the ingested US open sources, a gap the pipeline emits as a machine-actionable sampling priority. Median paired topsoil Hg (43.9 μg kg^{-1}) agrees with the independent estimate from the EU LUCAS soil survey (38.3 μg kg^{-1}). Relative to the reference atlas ratios computed in 2006 from outlier-inclusive means, our method-stratified robust estimates are lower but concordant in direction and ranking, and add uncertainty quantification, significance testing, and exclusion accounting. The paired enrichment surface peaks in a belt near 48°N; because the harmonized FOREGS slice ends near 52°N, the northward continuation is untestable from currently ingested open data — a concrete, prioritized acquisition target. These outputs directly address the harmonization, background-separation and coverage limitations that the Minamata Convention's ongoing effectiveness evaluation reports for global soil monitoring. All results derive deterministically from a frozen snapshot and a single analysis script.

1 Introduction

Continental- and global-scale environmental assessments are increasingly data-driven: machine-learning models trained on tens of thousands of aggregated public measurements now underpin

global groundwater arsenic risk mapping [6], and deep-learning maps of European topsoil mercury are built from >21,000 harmonized samples of the EU Land Use/Cover Area frame Survey (LUCAS) [1]. The rate-limiting step in such studies is rarely the model; it is the assembly of comparable, quality-controlled, provenance-complete observations from heterogeneous open sources — an effort that typically takes expert teams months to years, and that is difficult to audit or reproduce after the fact [7].

Mercury is a paradigmatic target for such infrastructure. Soils are the largest terrestrial Hg reservoir, with climate and vegetation as primary storage drivers [13]. Atmospheric deposition of gaseous elemental Hg, mediated by vegetation uptake and litterfall, is the dominant input pathway [5, 3], and the resulting legacy pool is climate-sensitive: warming-driven turnover of soil organic matter can remobilize accumulated Hg to waters and the atmosphere [5, 10, 11], and observation-driven models project that warming-induced vegetation greening may raise temperate soil Hg levels faster than emission controls offset them [14]. Quantifying how much legacy Hg (and Pb, its co-deposited tracer) is stored in surface relative to deep soil horizons is therefore both a contamination-screening and a climate-relevant question — and a timely one: the Minamata Convention's ongoing effectiveness evaluation reports that available soil-Hg data cannot yet be compared across surveys or separated into background and anthropogenic components [15], and the EU Soil Monitoring Law (in force December 2025) now mandates harmonized, comparable soil-contamination monitoring [16]. The Geochemical Atlas of Europe, produced by the Forum of European Geological Surveys (FOREGS) [9], provides the canonical paired-horizon baseline and has underpinned continent-scale geostatistical analyses of soil heavy metals [4]; its interpretive volume reported topsoil/subsoil ratios (Hg 1.66, Pb 1.36) computed as arithmetic means of individual site ratios, outliers included, with analytical methods pooled [2].

This paper makes three contributions. **(i) Infrastructure.** We demonstrate an autonomous agent pipeline (the *Global Geochemical Atlas* skill) that acquires, normalizes, spatially registers, quality-controls and provenance-chains open geochemical data into a standardized database, and then — in the same deterministic run — derives research products: comparability cohorts, environmental context tables, and prioritized data-gap queues. **(ii) Screening science.** Using method-stratified, site-paired robust statistics on that database, we re-derive the European legacy-metal fingerprint with uncertainty quantification and cross-continental replication, quantify organic-horizon Hg accumulation, and verify external consistency against the independent LUCAS survey. **(iii) Negative results.** We show why naive pooling fails (a 3× same-sample method artifact for Cr) and emit the resulting comparability constraints and coverage gaps as machine-actionable outputs rather than prose caveats.

We emphasize scope: all results are screening-level statements about harmonized open observations. Ratios above unity indicate surface enrichment consistent with atmospheric legacy inputs; they are not, by themselves, attributions of pollution sources, health risk, or climate response.

2 Materials and methods

Figure 1 summarizes the workflow: an autonomous acquisition and harmonization pipeline produces a standardized, confidence-scored database and derived research products; this study then applies a site-paired, method-stratified screening design to the paired-horizon subset.

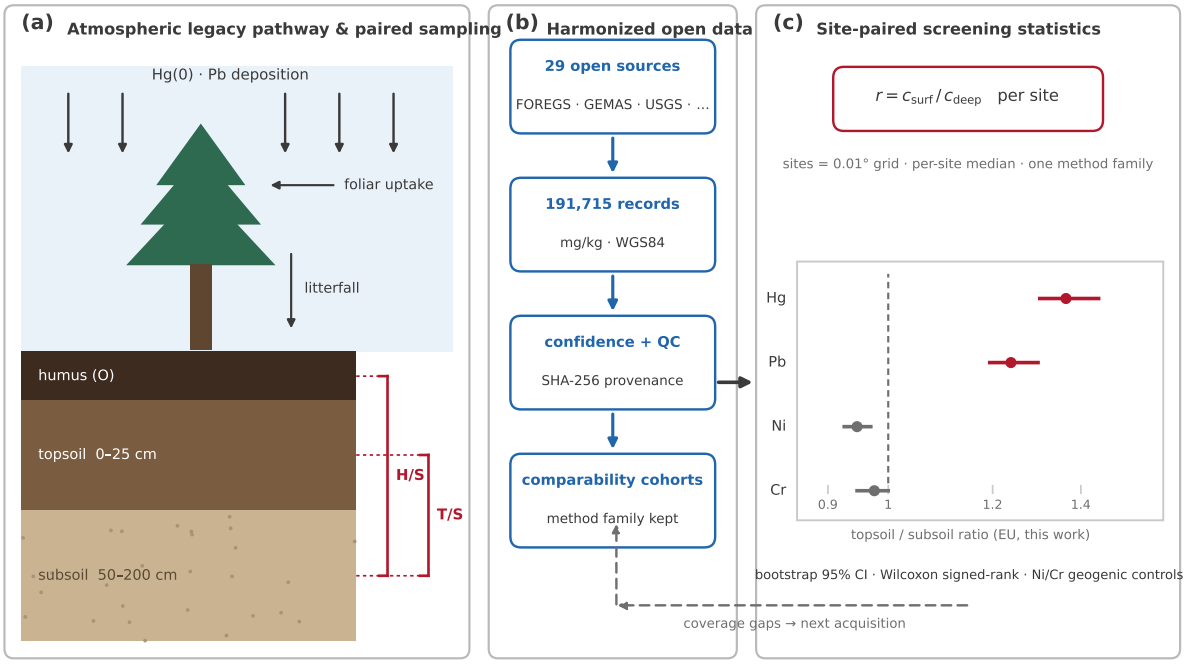

Figure 1: Methods overview. (a) Environmental setting and paired sampling design: atmospherically deposited Hg and Pb reach the soil directly and via vegetation uptake and litterfall; same-site concentration ratios reference the humus (O) and topsoil (0–25 cm) horizons to the subsoil (H/S and T/S; A/C horizons for the US replication). (b) The autonomous pipeline harmonizes 29 open sources into a 191,715-record standardized database (mg kg^{-1}, WGS84) with SHA-256 provenance, four-dimensional confidence scores and quality control (QC), and emits method-stratified comparability cohorts. (c) Site-paired screening statistics: per-site medians on a 0.01° grid within one method family; median ratios with bootstrap 95% CIs and Wilcoxon signed-rank tests, Ni/Cr as geogenic controls (inset: the European headline result). Dashed edge: coverage gaps discovered by the study return to the pipeline as machine-actionable acquisition priorities.

2.1 Harmonized database

A production run of the pipeline (2026-08-15) ingested 29 open sources across soil, sediment and water into 191,715 standardized measurement records (41,508 unique sample identifiers; seven analytes: As, Cr, Cu, Hg, Ni, Pb, Zn). Each record carries normalized value and unit (mg kg^{-1} for solids), WGS84 coordinates with transform lineage, sample type, method family, digestion/extraction, license and source locator, file SHA-256, and a four-dimensional operational confidence object (source evidence, analytical readiness, spatial usability, workflow usability). The snapshot analyzed here is frozen (`geochemistry.csv`, SHA-256 `e3616e88b6f5...`); the downstream analysis is a single deterministic script with a seeded bootstrap.

2.2 Paired-horizon design

For each element we selected a single method family per survey to avoid mixing analytical populations: Hg by atomic absorption spectrometry (AAS; identical in both FOREGS horizons), Pb/As/Cu/Ni by inductively coupled plasma mass spectrometry (ICP-MS), Cr/Zn by XRF, and inductively coupled plasma optical emission spectrometry (ICP-OES) for the USGS conterminous-

US soil survey [12]. Records with non-positive or missing values, or without canonical coordinates, were excluded. Sites were formed by rounding coordinates to 0.01° and taking per-site medians; surface/deep ratios were computed at sites where both horizons exist (FOREGS topsoil 0–25 cm vs. subsoil C-horizon 50–200 cm; FOREGS humus (O) vs. subsoil; USGS A-horizon vs. C-horizon). For each element we report the median ratio, a 2,000-draw bootstrap 95% confidence interval (CI) of the median, the fraction of sites with ratio >1.5, and a two-sided Wilcoxon signed-rank test on log ratios. Sensitivity analyses repeat the procedure at 0.1° rounding and, for Pb, with the alternative ICP-OES determinations. US As (available only by AAS) was omitted from the US panel for method-family consistency.

Relation to enrichment-factor practice. We deliberately use direct same-site surface/deep ratios rather than crustal- or reference-element-normalized enrichment factors (EFs). Classical EFs inherit the well-documented sensitivity to the choice of reference element and crustal composition [7]; moreover, conservative reference elements (Al, Ti, Zr) are not part of the seven-analyte harmonized set. The paired design cancels first-order lithological variation by construction (both horizons share parent material at a site), and residual pedogenic redistribution is bounded empirically by the behavior of the lithogenic controls Ni and Cr, which remain at or below unity in both surveys.

2.3 Comparability guard

Cohort construction in the pipeline stratifies by element, medium, sample type, grain fraction, measurement basis, method family, digestion and unit layer. The empirical motivation is measurable within-survey: on the same FOREGS topsoil samples, median Cr is 27 $\mathrm{mg\,kg^{-1}}$ by acid-digestion ICP-OES versus 74 $\mathrm{mg\,kg^{-1}}$ by total XRF (n=446 each; subsoil 29 vs. 79); agricultural soils from the GEMAS survey (Geochemical Mapping of Agricultural and Grazing Land Soils) [8] show 20.1 (ICP-MS, aqua regia) versus 62.0 (XRF) $\mathrm{mg\,kg^{-1}}$ (n=2,224 each). Any cross-survey comparison that ignores this stratification inherits a $\sim$3$\times$ artifact exceeding most signals of interest.

3 Results

3.1 Anthropogenic legacy fingerprint in paired horizons

Figure 2 and Table 1 summarize the paired ratios. In Europe, the two canonical atmospheric-legacy metals are clearly elevated in topsoil relative to subsoil: Hg 1.364 (95% CI 1.303–1.444; 41.3% of 392 site pairs >1.5; p=6.4$\times10^{-29}$) and Pb 1.239 (1.194–1.299; 28.3% of 389; p=1.7$\times10^{-35}$). The mildly anthropogenic As, Cu and Zn sit at 1.03–1.06 (significant but small), while the geogenic controls bracket unity: Cr 0.976 (not significant, n.s.) and Ni 0.947. The ordering Hg > Pb > (As $\approx$ Cu $\approx$ Zn) > Cr $\approx$ Ni matches the expected volatility- and deposition-driven hierarchy of legacy inputs. Results are invariant to coordinate rounding, and the Pb estimate is method-robust (ICP-OES 1.286 vs. ICP-MS 1.239).

In the independent USGS conterminous-US survey, the same design reproduces the geogenic control behavior (Ni 0.921, CI 0.894–0.945; Cu 1.020, n.s.) with a slight Zn surface excess (1.041), on $\sim$658 A/C pairs. Horizon-paired Hg and Pb do not exist in the ingested US open sources — see §3.5.

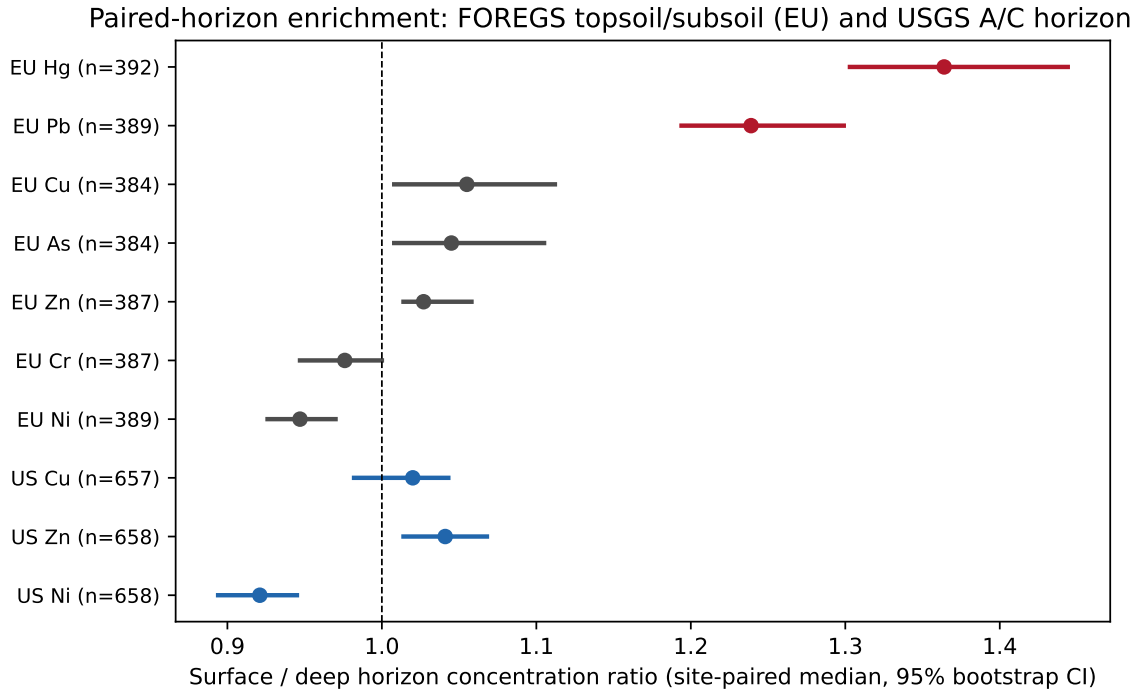

Figure 2: Surface/deep concentration ratios at site-paired locations: FOREGS topsoil/subsoil (EU, red/grey) and USGS A/C horizons (US, blue). Points are medians of per-site ratios; bars are 95% bootstrap CIs of the median. Method-stratified (Hg: AAS; Pb, As, Cu, Ni: ICP-MS; Cr, Zn: XRF; US panel: ICP-OES). Dashed line: no enrichment.

3.2 Organic horizons isolate the atmospheric pathway

Humus (O-horizon) samples sharpen the contrast (Fig. 3; Table 1). Relative to subsoil at the same sites, humus is enriched 5.26-fold in Hg (95% CI 3.89–5.81; 94.7% of 75 pairs >1.5) and 2.17-fold in Pb, while lithogenic Ni is *depleted* to 0.29 — an 18-fold behavioral separation between Hg and Ni across one soil profile. Absolute levels fall in the expected order (site-median Hg: humus 0.204, topsoil 0.044, subsoil 0.031 $\mathrm{mg\,kg^{-1}}$). This is the vegetation-pump signature — foliar uptake of atmospheric Hg(0) delivered to soil by litterfall [3, 5] — made visible directly in harmonized open survey data, without isotopes or flux towers.

3.3 Spatial structure and the 48°N belt

Mapping per-site log2 Hg ratios (Fig. 4) reveals coherent structure rather than noise: the strongest 2° aggregates (5–9 pairs each, median log2 ratio 0.73–1.49, i.e. up to ∼2.8×) form a belt near 48°N spanning eastern France, southern Germany, Austria and Slovakia, with a secondary cluster in central Italy. The belt is consistent with combined industrial-era deposition and forest-mediated uptake, and with the independent LUCAS-based observation that European topsoil Hg increases with latitude and vegetation activity [1]. We flag it as a screening pattern: attribution requires deposition modeling and land-cover covariates, which the pipeline exposes as declared (not yet ingested) interfaces.

Table 1: Paired surface/deep concentration ratios: 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; EU H/S: FOREGS humus/subsoil; US A/C: USGS A-horizon/C-horizon.

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}
EU H/S	Hg	75	5.256	3.885–5.811	94.7%	2.1×10^{-13}
EU H/S	Pb	59	2.166	1.667–2.563	67.8%	1.1×10^{-7}
EU H/S	Cu	59	0.649	0.583–1.031	28.8%	0.167
EU H/S	Ni	59	0.292	0.233–0.351	5.1%	8.0×10^{-10}
US A/C	Zn	658	1.041	1.014–1.068	19.9%	3.2×10^{-5}
US A/C	Cu	657	1.020	0.982–1.043	17.4%	0.277
US A/C	Ni	658	0.921	0.894–0.945	6.1%	6.9×10^{-18}

3.4 Agreement with, and sharpening of, the reference atlas

The 2006 atlas interpretation reported topsoil/subsoil ratios of 1.660 (Hg) and 1.364 (Pb) as arithmetic means of site ratios, outliers included, methods pooled [2]. Our method-stratified medians are lower (1.364 and 1.239) — as expected when heavy right tails are not allowed to dominate — but concordant in direction and in the ranking of elements. The added value of the automated re-derivation is (i) uncertainty quantification and significance testing, (ii) explicit method stratification (motivated by the 3× Cr artifact), (iii) full exclusion accounting from raw records to final pairs, and (iv) single-command reproducibility. External consistency: our paired-site topsoil Hg median (43.9 $\mu g\,kg^{-1}$) is within 15% of the independent LUCAS deep-learning estimate for EU topsoil (38.3 $\mu g\,kg^{-1}$) [1].

3.5 Coverage gaps as explicit research outputs

Three gaps discovered by this analysis are emitted by the pipeline as machine-actionable priorities rather than prose limitations: (1) *Nordic truncation*: the ingested FOREGS soil slice ends at 50.9°N (topsoil) / 52.5°N (subsoil), so the northward continuation of the 48°N belt — where the vegetation-pump hypothesis predicts continued increase [1] — is currently untestable from the harmonized data; Fennoscandian acquisition is queued in the run's coverage-repair tasks. (2) *US legacy metals*: no horizon-paired Hg or Pb exists in the ingested US open sources, precluding cross-continental replication of the headline result; the sampling-priority layer records this as a candidate acquisition or field target. (3) *Cross-source comparability*: at strict cohort granularity the run yields zero multi-source comparable cohorts (290 sample-sufficient cohorts are all single-source dominated), so every cross-survey statement in this paper is framed as replication across surveys, never pooling.

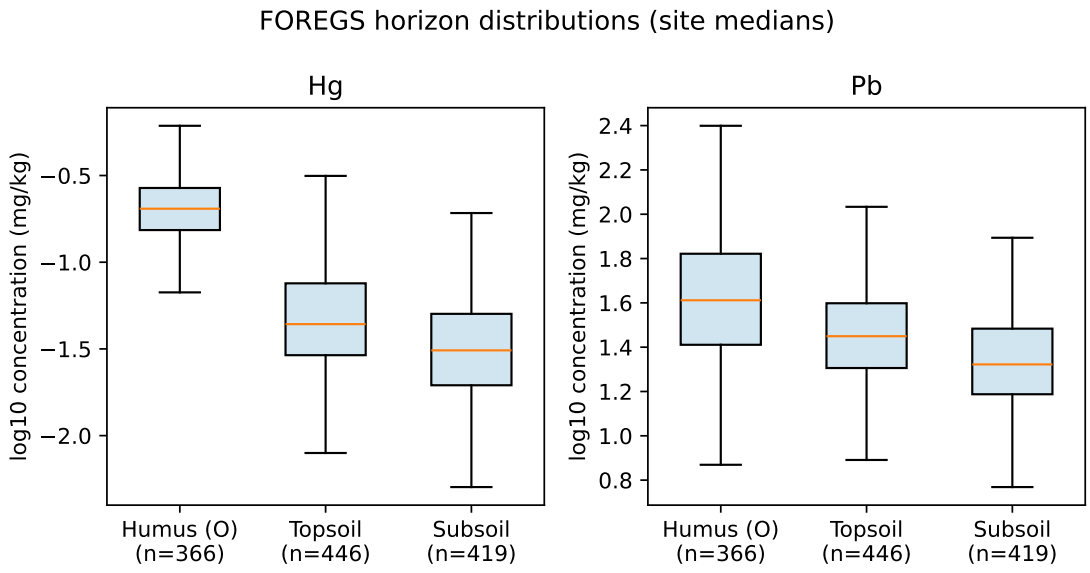

Figure 3: Site-median concentration distributions by FOREGS horizon (log10 mg kg^{-1}; boxes: interquartile range, IQR; whiskers: 1.5 IQR; outliers hidden). Hg and Pb increase monotonically toward the surface organic horizon.

4 Discussion

What the numbers support. Harmonized open data suffice to quantify, with uncertainty, a continental legacy-metal fingerprint: European surface soils carry a measurable stock of atmospherically delivered Hg and Pb relative to their own subsoil, concentrated where organic matter accumulates. The humus Hg enrichment (5.3×) far exceeds the mineral topsoil signal (1.36×), consistent with organic-matter binding as the storage mechanism [5, 3].

Position within the current frontier. Three active research and policy programs need exactly the quantities produced here. (i) The Minamata Convention's first effectiveness evaluation: its Open-Ended Scientific Group reports that available soil-Hg data are sparse, cover incompatible periods, cannot be compared across surveys for lack of consistent sampling/extraction/analysis, and do not separate background from anthropogenic levels — and the European dataset it relies on is the same FOREGS survey harmonized here [15]. Our comparability cohorts, method-stratified paired ratios (a direct background/anthropogenic separation) and machine-readable coverage gaps address each stated limitation. (ii) The EU Soil Monitoring Law, in force since December 2025, mandates a harmonized soil-health monitoring framework including contamination [16]; a confidence-scored, provenance-chained baseline of the kind assembled here is the natural substrate for its cross-border comparability requirement. (iii) The soil–climate Hg frontier: soils store the largest terrestrial Hg reservoir, with climate and vegetation as primary storage drivers [13]; recent observation-driven modeling indicates warming-induced vegetation greening may aggravate soil Hg levels faster than emission controls can offset [14], and permafrost projections put remobilization on the agenda of land-surface models now being extended with explicit Hg cycles [11]. Those models are evaluated against regional surface-Hg maps and vertical profiles — precisely the site-paired, method-controlled ratio tables published here, for the temperate mid-latitudes where [14] project the largest increases.

New questions generated. (Q1) *Climate remobilization*: given organic-horizon Hg stocks 5× subsoil levels, what fraction is re-emitted or exported under warming-driven soil-carbon turnover

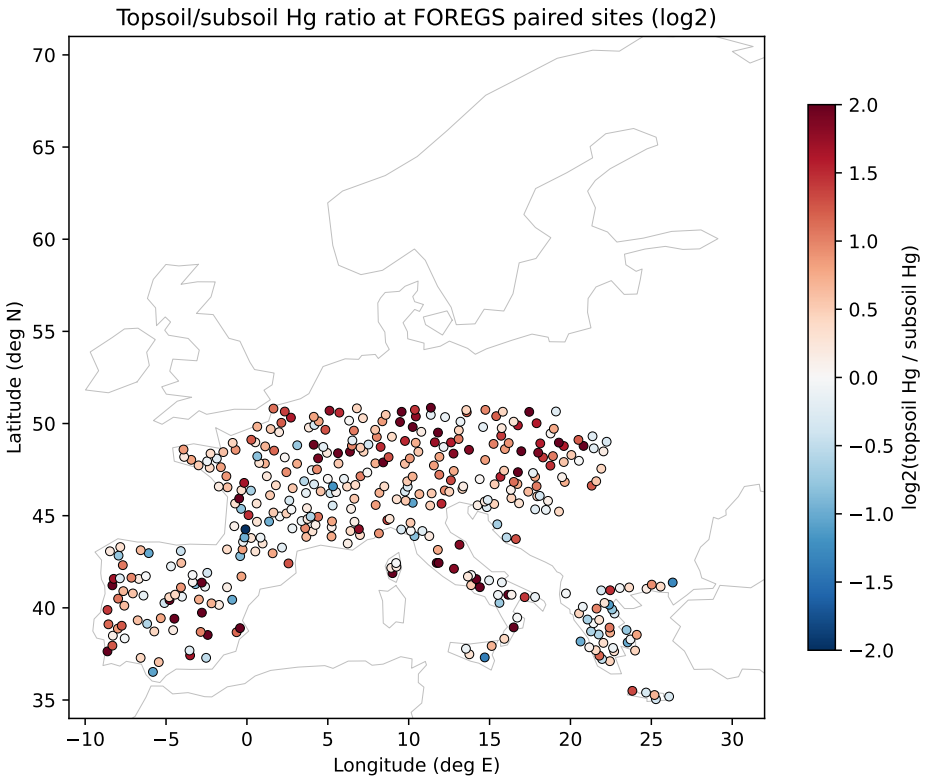

Figure 4: Topsoil/subsoil Hg ratio (log2) at FOREGS paired sites. Red: surface enrichment. The harmonized FOREGS slice contains no paired sites north of 52°N (see §3.5). Land outline: Natural Earth.

in temperate Europe — the mid-latitude counterpart to the permafrost Hg question [10, 11] and the observational face of the greening–Hg feedback [14]? The paired-site table published here is a ready-made constraint set for such models. (Q2) *Belt attribution*: does the 48°N enrichment belt follow historical deposition fields, forest cover, or soil organic carbon — and does it continue into Fennoscandia? (Q3) *Comparability standards*: with a 3× same-sample method artifact and zero multi-source cohorts, what minimum metadata and inter-method transfer models would make global soil-metal data poolable? Each question maps onto a concrete pipeline extension (declared covariate interfaces; Nordic acquisition; cohort-transfer calibration).

Limitations. FOREGS density is one site per ∼4,700 km^2; ratios conflate pedogenic redistribution with anthropogenic input (mitigated, not eliminated, by the Ni/Cr controls and the humus contrast); no dating or isotopes constrain input chronology; humus pairing recovers only 75 Hg sites; and all statements inherit the licenses and quality of the upstream surveys. These results are screening products intended to seed, not replace, process studies.

Reproducibility as a scientific control. Every number above is regenerated by one script on one frozen snapshot with a seeded bootstrap; two independent executions produce byte-identical outputs. We regard this property — rare in data-assembly-heavy environmental work — as the main methodological claim of the paper, and the screening results as its demonstration.

5 Conclusion

An autonomous pipeline can turn scattered open geochemical sources into research-grade, method-stratified evidence and derive from it a defensible continental screening result, its uncertainty, its external consistency checks, and — critically — the exact boundaries of what the data cannot yet support, expressed as prioritized acquisition targets. We propose this data-to-question loop as a template for AI-assisted earth-science infrastructure.

Data and code availability

Input sources (with licenses recorded per record in the database): FOREGS Geochemical Atlas of Europe topsoil/subsoil/humus data (Geological Survey of Finland; research use with attribution) [9, 2]; USGS conterminous US soil geochemistry, Data Series 801 (public domain, doi:10.3133/ds801) [12]; GEMAS agricultural soils (CC-BY-4.0) [8]. The frozen standardized snapshot, the analysis script (`paper_analysis.py`), per-site pair tables, result 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 data acquisition, harmonization, analysis design, statistics, figures and text, was produced end-to-end by an AI research agent operating the Global Geochemical Atlas skill; the human team has not yet completed review. Screening-level framing is intentional and load-bearing.

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