



Weathering release and ecosystem retention of trace metals along a 127-yr glacial retreat soil chronosequence

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ABSTRACT

Glaciers are retreating worldwide, exposing fresh substrates that undergo primary soil and vegetation development. Glacier retreat chronosequences provide unique settings to study trace metal sources (lithogenic vs. atmospheric) and their redistribution during early ecosystem development. Along the 127-yr Hailuoguo Glacier retreat chronosequence, we analyzed the vertical metal distribution in soil to distinguish rock- and atmosphere-derived trace metals, and quantified fast- and slow-reacting mineral pools and their release rates in a laboratory weathering experiment to assess the metal availability for redistribution. We calculated total trace metal stocks in the upper 10 cm of mineral soil, organic layers, and above- and below-ground vegetation compartments (leaves, branches, bark, wood and roots). Our results indicate that Co, Cu, Ni, U and V mainly derived from the parent material, whereas Cd, Pb, and Zn originated from atmospheric inputs, with initial carbonate weathering contributing to Cd and Zn release. Weathering released trace metals from labile primary minerals abundant in younger soils, while vegetation establishment and organic layer formation enhanced trace metal accumulation. However, this accumulation did not compensate for the substantial leaching losses, which accounted for 39–49% of the initial stocks, except for Pb and Cd, whose atmospheric inputs offset losses, resulting in their long-term retention within the ecosystem. The dynamic interplay of weathering, atmospheric inputs and plant-driven redistribution in this young ecosystem reflect a rapid responsiveness of trace metals to environmental changes. The observed dynamics at decadal scale challenge the view that trace metals are generally little reactive in ecosystems.

1. Introduction

Mountain glaciers are retreating globally because of increasing temperatures (IPCC, 2023) which is expected to continue during the coming decades (Hugonnet et al., 2021; Bosson et al., 2023; Rounce et al., 2023). The latter would even be the case in the absence of further global warming (Zekollari et al., 2025). The ongoing glacier mass loss will not only contribute to sea-level rise and alter downstream water chemistry (Beard et al., 2022; Cazenave et al., 2018; Huss and Hock, 2018), but also mobilize sediments, nutrients and trace metals, thereby impacting ecosystems and global biogeochemical cycles (Velasquez Casallas et al., 2025; Wang et al., 2020a). Trace metals such as Cu, Ni, and Zn are essential micronutrients for plants but become toxic to the vegetation in high concentrations. Other trace elements such as Co and V, although not classified as essential, can also positively influence plant growth and metabolism in small amounts (Marschner,

2012), while they are essential for some microorganisms (Einsle and Reels, 2020; Helmann, 2025). In contrast, elements such as Cd, Pb and U are non-essential and toxic to plants and microorganisms (Giller et al., 1998; Fajardo et al., 2019).

The fate of trace metals in ecosystems is partly controlled by their origin. Lithogenic metals are mainly bound in primary minerals and only become available after weathering, which is a slow process occurring at centennial to millennial scales. In contrast, anthropogenically-derived metals in soils are frequently more mobile and bioavailable, because they only equilibrate slowly with the soil solid phase by aging (Kabata-Pendias, 2010). Understanding the dynamics of trace metals in newly established ecosystems is essential for assessing how climate change and anthropogenically derived metal inputs influence the ecological risks of trace metals for the ecosystem development in high mountain regions (Le Roux et al., 2020; Sundriyal et al., 2026).

Weathering can be studied in the field, e.g., via the open-system

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element budget (Brimhall and Dietrich, 1987; Chadwick and Brimhall, 1990) and by constructing hydrological catchment budgets (Likens, 2013) or in the laboratory. Because of its natural slowness, weathering is usually artificially accelerated if studied in the laboratory to allow for measurable observations within a feasible time frame. One approach to reach an acceleration of weathering is a pH_{stat} experiment (e.g., Alt et al., 2013; Hacker et al., 2018; Schöndorf et al., 2025). In such an experiment, the pH is lowered to below the soil's equilibrium value and maintained constant to promote weathering. Frequently, this kind of experiment is performed in titration processors (Süsser, 1987), where the released ions accumulate in the solution, creating a risk of secondary mineral formation, which would represent stagnant soil water. However, in the approach of Kaupenjohann and Wilcke (1995), the ions released into the solution by proton buffering are continuously removed by mixed-bed ion exchange resins which simulates metal leaching in well-drained soils and avoids exceeding solubility products of little soluble minerals. Element release during pH_{stat} experiments can frequently be described as the sum of two simultaneous first-order reactions, representing a fast- and a slow-reacting pools with their respective release rate constants (e.g., Süsser, 1987; Schwarz et al., 1999; Schöndorf et al., 2025).

The Hailuoguo Glacier, located on the eastern slope of Mount Gongga in southwestern China, has been continuously retreating since the end of the Little Ice Age (Li et al., 2010), resulting in a glacier retreat chronosequence that allows for the study of chemical weathering and trace-element mobilization during the early stages of soil formation. Trace metals occur naturally and are commonly derived from parent materials, but their deposition from the atmosphere is increasingly influenced by human activities. The latter is particularly true in China, where the rapid industrialization since the 21st century caused an increase in trace metal deposition from the atmosphere (Pratte et al., 2018). Earlier research has shown that the eastern Tibetan plateau receives Cd and Pb inputs originating from South Asia and southwest China (Bing et al., 2016a, 2014; Wu et al., 2011), leading to increased metal concentrations in the soils of the remote Hailuoguo region (Bing et al., 2016b; Wang et al., 2020b). So far, in the Hailuoguo region, weathering intensity was approached by geochemical indices (Zhou et al., 2013) and major mineral composition analysis (Zhou et al., 2016). Zhou et al. (2018) showed that a rapid decrease in soil pH together with the rapid development of vegetation accelerated P weathering along the chronosequence. Basdediós et al. (2022b) reported that total Ca and Mg stocks decreased with increasing site age, mainly because of the dissolution of carbonates and easily weatherable Mg-containing minerals. Trace metal dynamics were evaluated based on environmental enrichment factors (Bing et al., 2016c, 2016b). However, the relationships between trace metal release from parent material through weathering, inputs via deposition from the atmosphere, and their retention and ecosystem-internal redistribution among developing vegetation, organic layers, and mineral soils remain poorly explored. Measuring total metal stocks in vegetation and soil captures long-term biomass accumulation and soil development, enabling direct comparison of metal pools across successional stages (Basdediós et al., 2022b, 2025). The knowledge of the development of trace metal stocks therefore facilitates the assessment of trace metal retention, accumulation from atmospheric deposition, and ecosystem-internal redistribution. Understanding metal dynamics at early successional stages further helps predict whether metal reactivity will respond in the short term to environmental change, particularly climate change. This is important to anticipate potential risks to soil health and headwater quality.

Our overall goal was to determine the extent to which the cycling of eight selected trace metals (Cd, Co, Cu, Ni, Pb, U, V and Zn), spanning a broad range of geochemical mobilities and biological roles, responds to advancing ecosystem succession during 127 years as proxy of fast environmental change. We aimed to (a) quantify weathering-driven release rates of trace metals from mineral topsoils along the Hailuoguo glacier retreat chronosequence by calculating the sizes of the fast- and

slow-reacting mineral pools and their associated release rate constants, (b) evaluate the contribution of geogenic versus anthropogenic sources to trace metal accumulation, and (c) assess how primary succession and organic layer formation influence the retention of trace metals in the ecosystem. To achieve this, we conducted a laboratory weathering experiment (pH_{stat}) and calculated total trace metal stocks including the upper 10 cm of the mineral soil, the organic layer (Oi, Oe and Oa horizons) and above- and below-ground vegetation compartments. We hypothesized that (i) in spite of the short development time (127 yr), soil metal release potential decreases as a result of the rapid depletion of easily weatherable minerals in the mineral topsoil, (ii) increasing plant biomass enhances the uptake from the soil and retention of trace metals along the chronosequence, leading to increasing vegetation stocks, particularly of biologically active elements (e.g., Co, Cu, Ni, V, Zn), and (iii) the formation and development of a soil organic layer promotes the retention and surface accumulation of trace metals with high affinity for organic matter (e.g., Cu and Pb), derived from litterfall recycling and deposition, while total ecosystem stocks of other trace metals decline with soil age because of leaching losses at rates controlled by the weatherability of their dominant mineral hosts.

2. Materials and methods

2.1. Study area

The Hailuoguo glacier is located on the eastern slope of Mount Gongga in the transition zone of the Sichuan Basin and the Tibetan Plateau, southwest China (Fig. S1). Increasing temperatures since the late nineteenth century (IPCC, 2023) resulted in the retreat of the glacier, exposing an approximately 2 km long and 50–200 m wide glacier retreat chronosequence that spans elevations from 2800 to 2950 m above sea level (m a.s.l.). The study area has a mean annual temperature of 4.2 °C and a mean annual precipitation of 1947 mm, with the majority of rainfall occurring between May and September (Wu et al., 2013). The successive exposure to ecosystem development created a well-established soil and vegetation chronosequence along the natural succession path (Li et al., 2010). The parent material for soil formation is a granitoid glacial moraine (Searle et al., 2016). The debris consists mainly of silicates (~90%) with a minor contribution of carbonates (<6%), which are completely weathered within less than 50 years of soil development (Basdediós et al., 2022b). The short duration of pedogenesis (<130 years) resulted in the formation of initial soils without a B horizon, classified as Leptic Calcaric to Follic Dystric Regosols from youngest to oldest (IUSS Working Group WRB, 2022).

Our study comprises seven sites deglaciated between 0 (2982 m a.s.l.) and 127 yr ago (2855 m a.s.l.), which were previously described by Zhou et al. (2013). Biological soil crusts and pioneer mosses started colonizing the soils approximately five years after glacier retreat, followed by the establishment of N_2 -fixing shrub-*Frankia* symbioses (*Hippophae tibetana* Schltdl.; 37 yr-old site), a half-mature broad-leaved tree forest dominated by poplar (*Populus purdomii* Rehder; 47 yr-old site) and ending in a conifer-dominated mixed forest with Faber's fir [*Abies fabri* (Mast.) Craib; 59 and 87 yr-old sites] and Sargent spruce [*Picea brachytyla* (Franch.) E. Pritz; 127 yr-old site; Fig. S1, Table S1].

2.2. Field sampling

Soil profiles were hand-dug in August 2017 at seven ecosystem succession stages (Fig. S1). Each successional stage was sampled in triplicate (i.e., 3 replicate plots $\times$ 7 ecosystem stages = 21 sites), with a minimum distance of 20 m between soil profiles, to ensure site representativeness and statistical independence of the replicate plots. Because the valley near the glacier is narrower, the distance between soil profiles was reduced to 10 m only at the 0 and 5 yr-old sites. Five soil horizons were sampled: Oi (fresh litter), Oe (shredded litter), Oa (dark layer of decomposed humus), A (surface mineral soil with humus enrichment)

and C (weathered soil parent material; Table S1). No organic layer was present at sites ≤ 5 yr-old. Additionally, we collected 16 rock samples from the glacial debris (0 yr-old site).

Leaves, needles, bark, branches, trunk and roots of the dominant tree and shrub species were collected between August and October 2017. A minimum of three trees were randomly sampled and composited into a single sample from the area surrounding each of our replicate soil profiles. Four species were sampled along the chronosequence: *H. tibetana* (37 yr-old site), *P. purdomii* (47 yr-old site), *A. fabri* (59 and 87 yr-old sites) and *P. brachytyla* (127 yr-old site), as described in Basdediós et al. (2022b).

Mineral and organic soil samples were air-dried to constant weight. Leaf, needle, branch, bark, wood and root samples were oven-dried at 40 °C for 72 h. Mineral samples were sieved to collect two fractions: fine earth (<2 mm) and stones (>2 mm). Trunk and root samples were manually homogenized using an agate pestle and mortar. Bark and branch samples were homogenized using a blade grinder (Polymix PX-MFC 90 D) equipped with a 3.0 mm mesh size sieve. Aliquots of all samples were ground using a ball mill (PM 200, Retsch, Haan, Germany) or a vibratory disc mill (TS, Siebtechnik GmbH, Germany) prior to chemical analysis.

Bulk deposition (i.e., wet deposition and the soluble fraction of coarse particulate dry deposition which can sediment into the open collectors) was weekly sampled with four Hellmann-type collectors, placed on a vegetation-free area (i.e., sites < 5 yr old), which were combined and filtered through a 0.2- μ m filter from 14/08/2017 to 02/10/2017. Soil solution was weekly collected using suction cups installed at the 0.1-m depth in the mineral soil at the 127 year-old site at the end of the vegetation period (September 2017). Because suction cups require equilibration with the surrounding soil, the initial soil solution samples were discarded to allow sufficient time for equilibration. Stream water was weekly collected from 07/08 to 02/10/2017.

2.3. Chemical analyses

Trace metal concentrations in the soil samples (mineral and organic horizons), stones and rocks were determined after digestion of ~ 0.1 g of sample with a mixture of 4 mL of HNO_3 (69%), 1.5 mL of HF (48%) and 1 mL of H_2O_2 (30%). Plant compartments were digested using 8 mL of HNO_3 (69%) and 2 mL of H_2O_2 (30%). All digestions were performed in a microwave oven (MARS6Xpress, CEM, Kamp-Lintfort, Germany), and each microwave run of approximately 20 samples included at least one certified reference material (CRM) and one blank. Accuracy was assessed using SRM1547 (peach leaves) and SRM1515 (apple leaves) as CRMs for organic samples, and BCR2 (Columbia River Basalt 2) for soil samples. The average recoveries were $100 \pm 10\%$ ($\pm$ standard deviation). Precision, assessed through duplicate measurements, was $< 5\%$.

To determine the release kinetics of the trace metals in the mineral topsoils (10 cm) along the chronosequence, we performed a pH_{stat} experiment following the method of Schwarz et al. (1999) as described in Basdediós et al. (2022a). Briefly, we used 2 g of mixed ion-exchange resin (Amberlite MB-20, Rohm and Haas) enclosed in polyethylene bags (100 μ m mesh width) to monitor element release from soils. The resin was previously saturated with HNO_3 and rinsed with deionized water (>18 M Ω cm^{-1}) to remove free acid. Then, the resin bag was added to a 125-mL PE bottle containing 100 mL of DI water and 1 g of soil, together with the required amount of $\text{Ba}(\text{NO}_3)_2$ to decrease the pH to 3.0 by releasing H^+ from the resin. The pH value of 3.0 was chosen because it was lower than the native pH values of all studied soils to accelerate the weathering release of metals. A pH value of 3.0 is a common choice in many other pH_{stat} experiments (e.g., Kaupenjohann and Wilcke, 1995; Schwarz et al., 1999; Süsser, 1987). The bottles were shaken for ten sequential times from 10 min to 168 h to determine the kinetics of element release, resulting in 210 samples (7 sites $\times$ 3 replicates $\times$ 10 shaking times). After shaking, resin bags were removed, rinsed, and the adsorbed ions were extracted with 2 M HNO_3 in three extraction steps.

The release kinetics obtained with our pH_{stat} experiment represent potential weathering rates under strongly acidic conditions and may differ from those under field conditions, but allow for direct comparison among sites of different ages. Tests of element recovery from the resin with a standard solution (ICP multi-element standard solution IV, Certipur, Merck KGaA, Darmstadt, Germany; Cat. No. 1.04498) containing a mixture of elements yielded $105 \pm 3\%$ (Cd, $n = 5$), $105 \pm 12\%$ (Co, $n = 3$), $103 \pm 6\%$ (Cu, $n = 3$), $99 \pm 7\%$ (Ni, $n = 5$), $107 \pm 2\%$ (Pb, $n = 5$), and $97 \pm 6\%$ (Zn, $n = 5$). Uranium and V were not included in the multi-element standard. Element concentrations in the digests, extracts and water samples were determined by inductively-coupled plasma mass spectrometry (ICP-MS, 7900 Agilent, Waldbronn, Germany).

2.4. Calculations

The element release during our pH_{stat} experiment was described following a two-step first-order equation (Eq. (1)):

$$Y(t) = \text{Pool A } (1 - e^{-k_a t}) + \text{Pool B } (1 - e^{-k_b t}) \quad (1)$$

where $Y(t)$ represents the element release from soil (mg kg^{-1}) at time t . Pool A and Pool B represent two differently reactive (fast and slow; mg kg^{-1}), operatively defined pools, each comprising a range of different but unknown metal forms. The variables k_a and k_b are the corresponding release rate constants of each pool (h^{-1}). Pools and release rate constants were estimated using a nonlinear regression model with sequential quadratic optimization. The goodness of the fit between the observed data and the model was evaluated using the coefficient of determination (R^2).

Element stocks in tree compartments ($E_{\text{tree-compartment}}$) were computed as the product of compartment biomass (B_c) and the corresponding element concentration (C_E) measured in each compartment (leaf/needle, branch, trunk, bark and root; Eq. (2)). Plant biomass and proportional contribution of the different tree compartments to the total biomass have been reported by Basdediós et al. (2022b) for the same study sites based on data from Luo et al. (2004) and Zhou (2013).

$$E_{\text{tree-compartment}} (\text{g m}^{-2}) = B_c (\text{g m}^{-2}) \times C_E (\text{g g}^{-1}) \quad (2)$$

Element stock in soil organic layer and mineral horizons was calculated considering the element concentration (C_E), the bulk density (BD_i) taken from Wang et al. (2019) and the thickness (T_i) of the respective horizon measured during soil sampling (Eq. (3)). Total element stock in the organic horizon at each plot was calculated by summing up the element stocks of all individual horizons (Oa , Oe and Oi).

$$E (\text{g m}^{-2}) = C_E (\text{g g}^{-1}) \times \text{BD}_i (\text{g m}^{-3}) \times T_i (\text{m}) \quad (3)$$

The total element stock in the mineral soil was calculated by summing up the individual A and C horizon stocks to a depth of 0.1 m, considering the contribution of the stones (i.e., soil > 2 mm) in the soils along the chronosequence as described in Basdediós et al. (2022b). The thickness of the C horizon (T_C) underlying the A horizon was adjusted to account for the decrease in bulk density of the A horizon related to soil loosening during pedogenesis, using Eq. (4),

$$T_C (\text{m}) = 0.1 (\text{m}) - T_A (\text{m}) \times \frac{\text{BD}_A (\text{g m}^{-3})}{\text{BD}_C (\text{g m}^{-3})} \quad (4)$$

where T_A is the thickness of the A horizon, 0.1 m is the initial mineral soil depth and BD_A and BD_C are the field bulk densities of the local A and C horizons, respectively. With this approach we corrected for the changes in bulk density of the soils in the course of soil development to make sure that we indeed compared the same initial mineral masses among the various study sites. The total stock ($E_{\text{total stock}}$; g m^{-2}) at each plot was obtained by summing the stocks of the vegetation, and organic and mineral horizons.

2.5. Statistical analyses

Linear regression and Pearson correlation were used to evaluate the relationships between variables (i.e., fast- and slow-reacting pools, as well as their associated constant release rates k_a and k_b , Fig. S2). The regression analyses were conducted on the mean values of each site age ($n = 7$). One-way analysis of variance (ANOVA) followed by a Tukey's HSD post-hoc test were applied to detect significant differences between total stocks and pool sizes among the various study sites. Normal distribution of residuals was visually inspected using Q-Q plots. Homoscedasticity was confirmed for our data after applying Levene's test. No

data transformations were necessary. Because we observed partly different trace metal concentrations in the parent material between the three younger and the four older sites we compared the mean trace metal concentrations between these two site groups in the stone fraction (>2 mm) of the C horizons using a t -test (Fig. S3). When significant differences were found, the trace metal loss from the mineral topsoil at the 47–127 yr sites was adjusted by accounting for these differences; and both the adjusted and unadjusted results were reported. Statistical analyses were conducted using the statistical software R (R Core Team, 2020). Significance was set at $p < 0.05$.

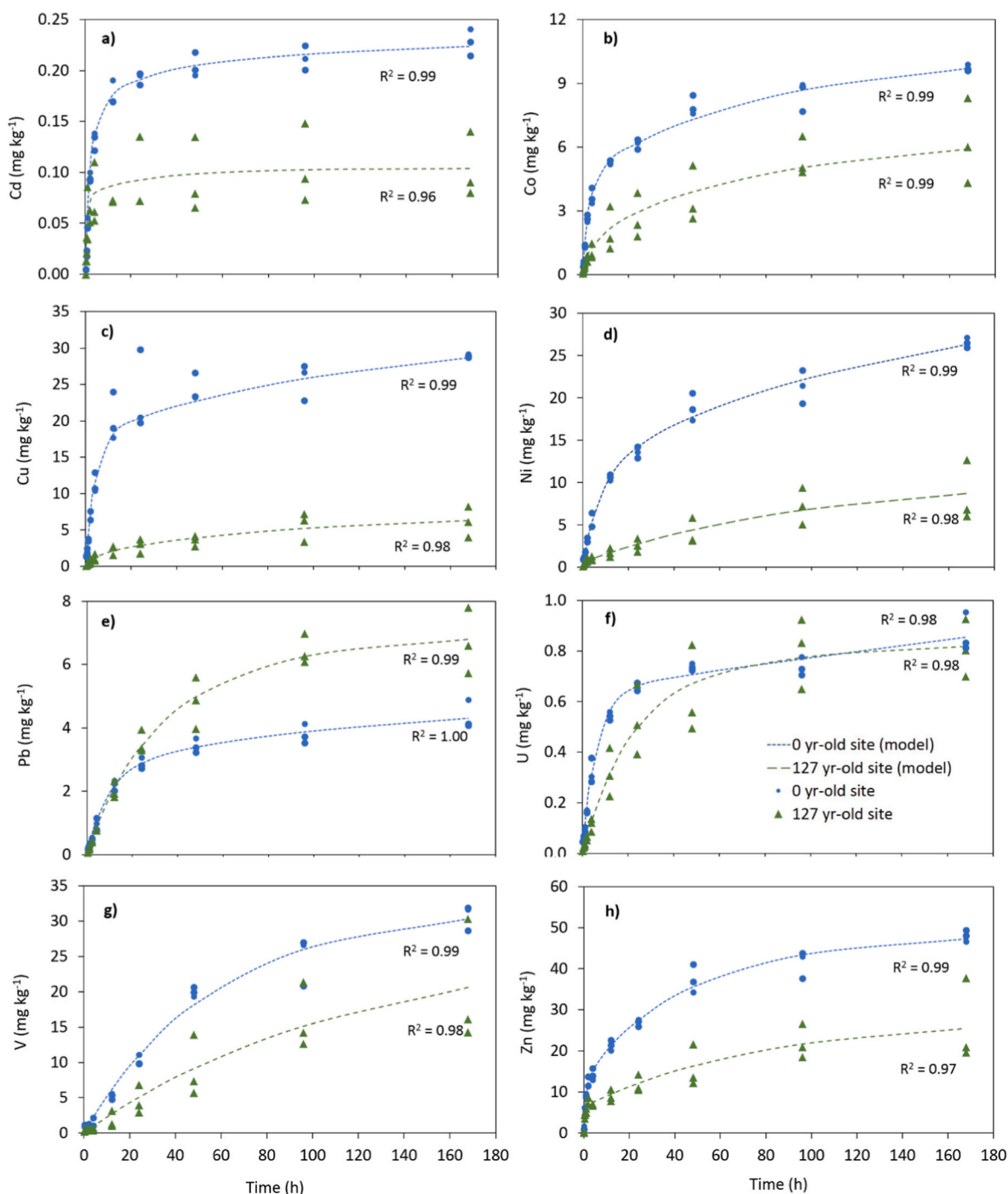


Fig. 1. Release kinetics of (a) Cd, (b) Co, (c) Cu, (d) Ni, (e) Pb, (f) U, (g) V and (h) Zn during a pH_{stat} experiment at pH 3 in mineral topsoils (0–10 cm) of the 0 yr-old (●) and the 127 yr-old (▲) sites of the Hailuoguo chronosequence. The dotted and dashed lines illustrate the biexponential fit, except for U and V, where a mono-exponential function was applied. Mind that all subfigures have individual Y axis scaling.

3. Results

3.1. Trace metal release

At the end of our pH_{stat} experiment ($t = 168$ h), the cumulative release of Cd, Co, V and Zn was approximately two times higher at the youngest (0 yr-old) than at the oldest site (127 yr-old), while Ni release

was three times higher and that of Cu even nearly five times higher (Fig. 1). In contrast to these patterns, we found a higher cumulative release of Pb at the oldest site than at the youngest site (6.7 ± 0.6 vs. 4.4 ± 0.3 mg kg⁻¹; Fig. 1e). The cumulative U release remained low and did not change with site age (~ 0.9 mg kg⁻¹; Fig. 1f).

The sizes of the fast-reacting pools of Cd, Co, Cu and Ni decreased with increasing site age, while those of Pb and Zn did not consistently

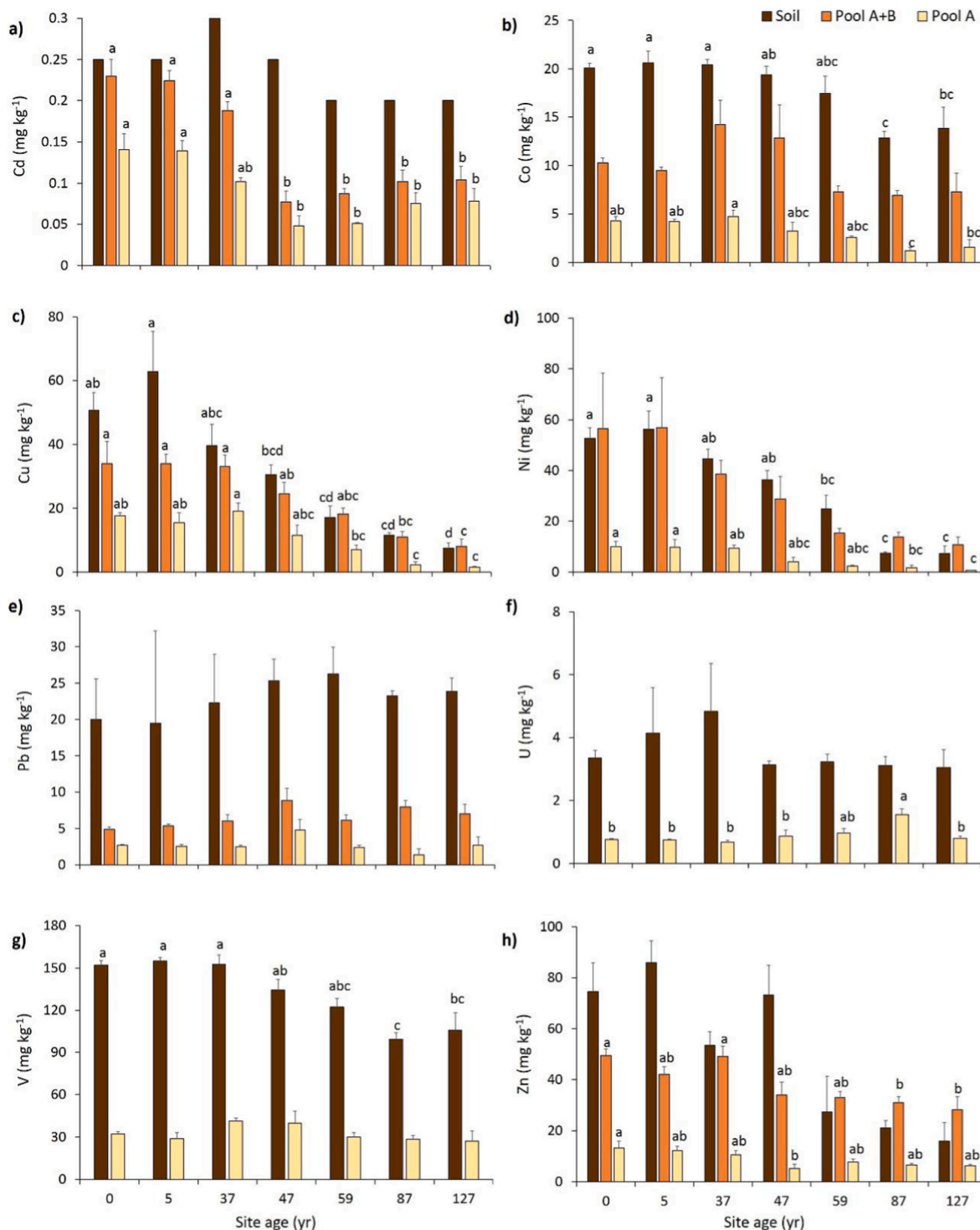


Fig. 2. Sizes of the total concentration in the soil (Soil), the fast-reacting (Pool A) and fast + slow-reacting pools (Pool A + B) of (a) Cd, (b) Co, (c) Cu, (d) Ni, (e) Pb, (f) U, (g) V, and (h) Zn in mineral topsoils (0–10 cm) along the Hailuoguo chronosequence. The release of U and V could only be modeled using a single-pool fit so that there is no slow-reacting pool. Soil Cd concentration was taken from Bing et al. (2026b). Different letters denote significant differences in the sizes of the pools among the sites ($p < 0.05$). Error bars represent standard errors ($n = 3$).

vary with site age (Fig. 2). The release of U and V could only be modeled using a single-pool rather than a two-pool fit, and the size of the singly reactive pool attributed to these elements showed no clear trend with increasing site age (Fig. 2). The fast-reacting pools of Cd and Zn were strongly correlated with each other ($r = 0.93$, $p < 0.01$; Fig. 3a), and both of them were correlated with that of Ca ($r = 0.92$, $p < 0.01$ for Cd, and $r = 0.91$, $p < 0.01$ for Zn; Fig. 3b and c).

The sizes of the slow-reacting pools of Cd, Cu, Ni and Zn decreased with increasing site age (Fig. S2), consistent with their total soil concentrations (Fig. 2). In line with these findings, the sizes of the slow-reacting pools of Cu and Ni correlated with each other ($r = 0.94$, $p < 0.001$; Fig. S2c). In contrast, the slow-reacting pool of Pb increased with site age ($r = 0.72$; $p = 0.07$) and the associated decrease in soil pH ($r = -0.92$, $p < 0.01$; Fig. 3l and m), while total Pb concentrations in soils showed substantial within-site variability and no trend with site age (Fig. 2).

The release rate constants associated with the fast- and slow-reacting pools (k_a and k_b , respectively) varied among elements and site age. Across all elements and sites, k_a ranged from 0.16 (Ni, 0 yr-old site) to 1.79 h^{-1} (Ni, 127 yr-old site), whereas k_b ranged from 0.005 (Pb, 47 yr-old site) to 0.08 h^{-1} (Cd, 5 yr-old site; Table S2). Interestingly, k_a of Ni was correlated with site age ($r = 0.93$, $p < 0.01$; Fig. 3j), while k associated with the single-pool release of V and U decreased with increasing site age ($p < 0.05$; Fig. 3q and r). Along the chronosequence, the k_a of Ca and Cd were positively correlated with each other ($r = 0.79$, $p < 0.05$), as well as the k_b of Cu with that of Co ($r = 0.76$, $p < 0.05$; Fig. 3k–p). The release rate of the slow-reacting pool of Pb was strongly influenced by both, site age ($r = 0.87$; $p = 0.01$) and total organic carbon concentrations (TOC; $r = 0.84$; $p < 0.05$), which also increased with increasing site age (Fig. 3n and o).

3.2. Trace metal stocks

Trace metal stocks varied among tree compartments and site ages (Fig. 4, Table S3). The stocks of Cd, Cu and Zn were similar between total biomass and the organic layer, whereas those of Co, Ni, Pb and V were consistently higher in the organic layer than in total biomass (Fig. 4). Element concentrations in the organic horizons generally increased in the order $O_i < O_e < O_a$. Exceptions were Cd and Zn which showed higher concentrations in the O_e than the O_a horizon along the chronosequence (Fig. S4). The latter was also true for Pb at the ≥ 87 yr-old sites (Fig. S4).

Total ecosystem stocks of Co, Cu, Ni, V and Zn declined exponentially with increasing site age (Fig. 5), with losses ranging from $39 \pm 3\%$ for Cu and Ni to $49 \pm 6\%$ for Zn. For these estimates, we corrected for the a priori lower Cu and Ni concentrations in the four oldest soils than in the three youngest ones, by adding the mean difference in the Cu and Ni stocks between the group of younger and older soils to the total ecosystem stock of the four older soils (upper release curve in Fig. 5c and d). In contrast, U stocks showed a less pronounced decline, occurring primarily during the early stage (<37 yr), after which they remained stable (Fig. 5f). Ecosystem stocks of Cd and Pb varied little across the chronosequence (Fig. 5a–e).

4. Discussion

4.1. Cumulative trace metal release in the mineral topsoil by proton buffering

In line with our first hypothesis, the cumulative element release at the end of our pH_{stat} experiment was markedly higher at the youngest (0 yr-old) than at the oldest site (127 yr-old), reflecting the progressive depletion of easily weatherable minerals with increasing site age (Fig. 1). The only exceptions were U and Pb. The cumulative release of U did not show significant differences between the youngest and the oldest site, likely indicating a small reactivity. The cumulative release of Pb

was higher at the oldest than the youngest site, suggesting an increasing influence of atmospheric inputs, because freshly deposited Pb is less strongly bound in soils than native and aged Pb (Kabata-Pendias, 2010).

Trace metal release from the mineral topsoil was quantitatively dominated by Cu, Ni, V and Zn, which are primarily hosted as trace constituents in minerals of calc-silicate rocks and granite (Fig. 6) such as biotite, hornblende and chlorite (Zhu et al., 2024). The latter three minerals accounted for more than 25% of the mineral mass along the Hailuoguo chronosequence (Zhou et al., 2016) and constituted an important primary source of nutrients during the early stage of ecosystem development (Basdediós et al., 2023, 2022b). The low and unchanged cumulative U release between the youngest and oldest sites (Fig. 1f) is consistent with its predominant occurrence in granite (Fig. 6), a common rock type in the Hailuoguo region that is highly resistant to weathering (Goldich, 1938).

4.1.1. Pool sizes and release rate constants derived from the weathering experiment

Cd and Zn: The positive correlation between the fast-reacting pools of Cd and Zn indicated that both elements were mobilized and retained through similar weathering and sorption mechanisms. In line with this, the correlation between their fast-reacting pool sizes and release rates to those of Ca likely reflected the co-association of Cd and, less strongly, Zn with calcite (CaCO_3). Divalent metals like Cd^{2+} and Zn^{2+} can be incorporated into or adsorbed onto carbonate minerals (García-Sánchez and Álvarez-Ayuso, 2002; Kim et al., 2023; Rambeau et al., 2010), which can subsequently be released during weathering processes (Liu et al., 2025). Cadmium - and, to a lesser extent, Zn - were largely released during the early stages of the Hailuoguo chronosequence (Fig. 2), when the dissolution of carbonates dominated the weathering processes (i.e., ≤ 47 yr old; Basdediós et al., 2022b; Zhou et al., 2016). In line with these findings, the analysis of calc-silicate rocks collected at the foot of the Hailuoguo glacier also revealed a Cd concentration of 0.2 mg kg^{-1} and a Zn concentration ranging from 50 to 100 mg kg^{-1} (Fig. 6). The Cd concentration is higher than on average in magmatic rocks ($50 - 100 \text{ mg kg}^{-1}$), while the Zn concentration falls into its typical range ($0.10 - 0.14 \text{ mg kg}^{-1}$; Gong et al., 1977; Wedepohl, 1972). These results suggest that the calc-silicate rocks in the Hailuoguo glacial debris may be a local source of easily weatherable Cd, while contributing to a lesser extent to Zn release.

Co, Cu and Ni: The sizes of the fast-reacting pools of Co, Cu and Ni decreased with increasing site age (Fig. 2b, c, d) and were correlated with each other ($r > 0.94$, $p < 0.01$; Fig. 3g, h, i), suggesting shared mineral sources and similar mobilization mechanisms along the chronosequence. At the same time, their release rate constants (k_a) tended to increase with increasing site age (Table S2), indicating that the fast-reacting pool became increasingly susceptible to dissolution as soil development progressed. The significant correlations between the sizes of the slow-reacting pools of Cu and Ni and the release rate constants (k_b) of the slow-reacting pools of Co and Cu illustrate some parallelism of the release of these three elements but also point at different mineral hosts and release kinetics. This assumption is corroborated by the significantly higher Cu and Ni concentrations in the stone fraction of the C horizons at the three youngest sites (0–37 yr) than the four older sites (≥ 47 yr) indicated that differences in the parent material composition (Fig. S3), which are typical of highly heterogeneous glacial debris, likely contribute to the differences in the release of these two elements (Fig. 1c and d). Similarly, Bing et al. (2016b) reported higher Cu concentrations in younger than in older soils along the chronosequence.

Pb: Interestingly, the slow-reacting Pb pool increased with site age in spite of the decrease in soil pH (Fig. 3l and m). This together with the finding that the release rate constant k_b of the slow-reacting Pb pool correlated with site age and total organic carbon concentrations suggested that the progressive accumulation of organic matter may have promoted the retention of Pb in slowly reacting pools. However, it is important to remark that only 24 – 35% of the total Pb pool in the soils

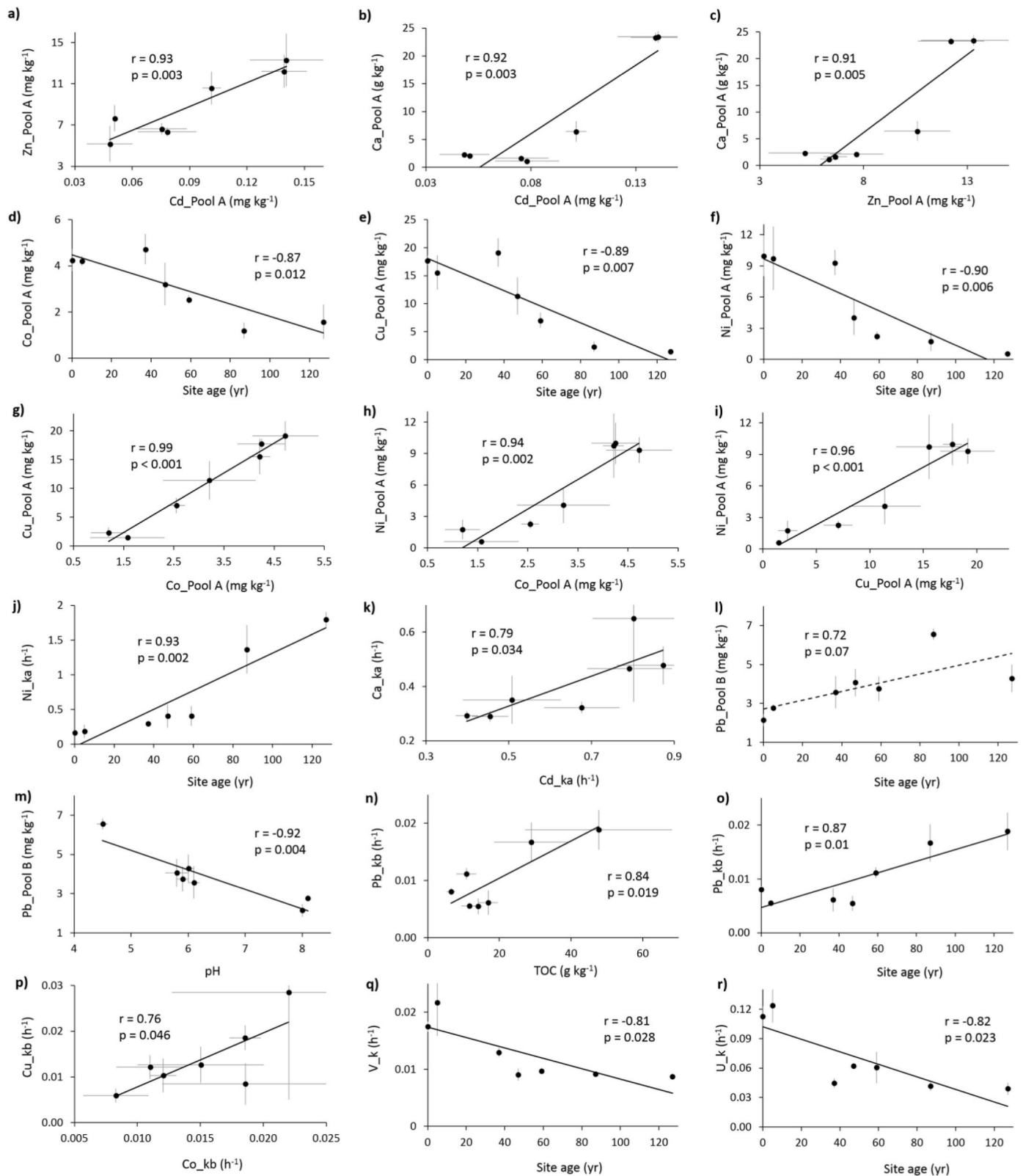


Fig. 3. Linear regression lines and results of Pearson correlation analyses of selected variables, including site age, pH, TOC, and pH_{stat}-derived parameters - the fast-reacting pool (Pool A), the slow-reacting pool (Pool B), and the release rate constants associated with the fast- and slow-reacting pools (ka and kb, respectively) - for the trace metals studied along the chronosequence. Error bars represent the standard error (n = 3). Only relationships relevant for the interpretation of the results are shown.

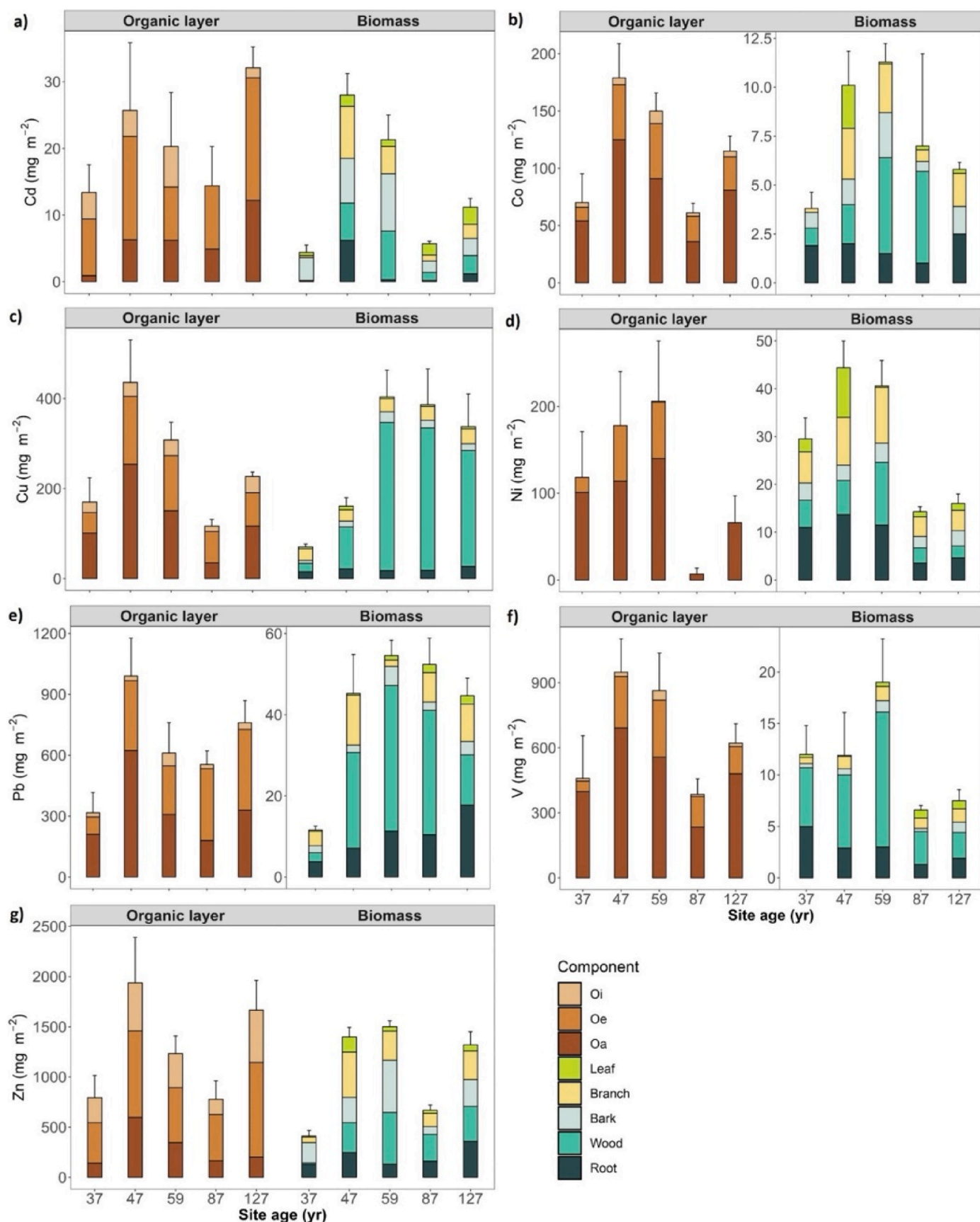


Fig. 4. Cumulative stocks of (a) Cd, (b) Co, (c) Cu, (d) Ni, (e) Pb, (f) V and (g) Zn in the organic horizons (left; organic layer: Oa, Oe and Oi) and biomass compartments (right; leaf, branch, bark, wood and root) along the Hailuoguo chronosequence. Uranium concentrations in the biomass were below the quantification limit.

and are therefore not shown. Error bars represent the standard error ($n = 3$). Mind the different y-axis scalings between organic layer and tree biomass for Co, Ni Pb and V.

was released after 168 h, indicating that a large part of Pb was still strongly bound in soil minerals.

U and V: In contrast to the other studied elements, the release of U and V could only be modeled using a single-pool fit, indicating that their release is primarily controlled by dissolution of a single dominant solid phase (Fig. 2f and g). Although the size of the single V pool did not vary significantly with site age (Fig. 2g), its release rate constant (k , Table S2) decreased with increasing site age (Fig. 3q). A similar trend was observed for U (Fig. 3r), indicating that both elements became progressively less mobile as soils developed and organic matter accumulated (Reijonen et al., 2016). Furthermore, the increasing concentrations of little crystalline, oxalate-extractable Fe and Al phases (Fe_o and Al_o) with increasing site age likely enhanced U and V adsorption (Zhou et al., 2016), reducing their mobilities. This effect may have been amplified for V by the decreasing soil pH at older sites, which generally reduces the mobility of oxyanions such as VO_4^{3-} , the dominating V species in well aerated soils (Blackmore et al., 1996). In line with this, the elevated single U pool at the 87 yr-old site may be partly associated with the lower abundance of Fe_o at this site (Zhou et al., 2013), given the role of amorphous Fe oxides as sorbents for U (VI; Ulrich et al., 2006).

4.2. Impact of the vegetation development on trace metal retention in developing soils

Trace metal stocks varied among tree compartments and site ages (Fig. 4, Table S3), with no evidence of preferential accumulation of biologically active elements (Co, Cu, Ni, V and Zn) compared to non-essential, potentially toxic elements (Cd and Pb), contrary to our second hypothesis. Although we recognize that changes in vegetation composition along the chronosequence could potentially contribute to the differential redistribution of trace elements into biomass, it is noteworthy that vegetation stocks of Ni, V, and Zn were significantly higher at the 59 yr-old site than at the 87 yr-old site (Fig. 4a–c, f), despite both sites being dominated by the same tree species (Table S1). The lower metal stocks at the 87 yr-old site suggest reduced uptake by the vegetation, consistent with previous studies showing that older trees take up nutrients more slowly (Wang et al., 2024), although metal availability (approximated as fast-reacting pool A) was similar at both sites (Fig. 2). In line with this, Li et al. (2025) recently reported that the nutrient-cycling potential of rhizosphere bacteria was strongly related to the decreasing soil pH along the Hailuoguo chronosequence but less influenced by differences in root morphology or soil nutrient availability.

4.3. Impact of the development of the organic layer on trace metal retention

In the organic horizons, element concentrations generally increased in the order $\text{O}_i < \text{O}_e < \text{O}_a$, which can be attributed to the increasing decomposition of the organic matter while the trace metals are retained and thus relatively accumulated indicating that all studied trace elements were retained in the organic layer, not only those with a particularly high affinity to organic matter in contrast to the first part of our third hypothesis (Filipinski and Grupe, 1990). The finding that Cd and Zn showed higher concentrations in the O_e than the O_a horizon along the chronosequence and Pb at the ≥ 87 yr-old sites (Fig. S4) suggested that metal deposition from the atmosphere was retained in the organic layer, again independent of the affinity for organic matter (Hahn et al., 2019; Wilcke et al., 1998, 1999). Based on measurements of bulk deposition collected at the end of the vegetation period (Table S4), we estimate that Pb deposition in the Hailuoguo area ranges from 1.4 to 1.7 $\text{mg m}^{-2} \text{yr}^{-1}$, which is consistent with the published average bulk Pb

deposition in southwest China of 1.2 $\text{mg m}^{-2} \text{yr}^{-1}$ for the year of our study (Jiang et al., 2024). Besides the input by bulk deposition, Bing et al. (2018) demonstrated that the forest canopies of the Gongga Mountain area additionally scavenged aerosol-bound Cd and Pb, contributing to the accumulation of these trace metals in the soil organic layer. Lead inputs via deposition were 2–5 times higher than losses through the stream, whereas Cd and Cu inputs were roughly equal to stream outputs, which contributes to the observed stronger retention of Pb than all other studied trace elements along the chronosequence (Table S4). Cadmium accumulation may have also been influenced by the dominance of *P. purdomii* at the 47 yr-old site, a species known to bioaccumulate Cd (Bing et al., 2016c). This species may have taken up Cd released during the earlier stages of soil development by weathering of carbonate minerals (Fig. 2a) and incorporated it into aboveground tissues (Fig. 4a). As a consequence, Cd was retained in the ecosystem and accumulated in the organic layer via litterfall (Fig. 4a).

Total ecosystem stocks of Co, Cu, Ni, V and Zn declined exponentially with increasing site age (Fig. 5), with losses ranging from $39 \pm 3\%$ for Cu and Ni to $49 \pm 6\%$ for Zn in line with our third hypothesis. For these estimates we corrected for the a priori lower Cu and Ni concentrations in the four oldest soils than in the three youngest ones, by adding the mean difference in the Cu and Ni stocks between the group of younger and older soils to the total ecosystem stock (upper release curve in Fig. 5c and d). This pattern reflects the progressive weathering of primary minerals and cumulative losses through leaching during soil development, consistent with our finding that the stream export exceeded the input by bulk deposition by approximately one order of magnitude (Table S4). Surprisingly, the decline in total ecosystem stocks of Co, Cu, Ni, V and Zn was comparable to that of Ca and Mg (44 and 30%, respectively; Basdediós et al., 2022b), despite of the common view that trace metals in soils are little mobile (Dowdy & Volk, 1983). However, the decreasing ecosystem stocks may also partly reflect some leaching from the mineral topsoil (10 cm) to greater soil depths, which we did not study so that part of the apparent ecosystem loss shown in Fig. 5 could be attributable to increasing trace metal stocks in the subsoil (Bruemmer et al., 1986). In contrast, the U stocks showed a less pronounced decline, occurring primarily during the early stage (<37 yr), after which they remained stable (Fig. 5f). This pattern reflects a transition from the carbonate-buffered young soil to an acidified, organic matter-rich forest soil. In the older study ecosystems, biological cycling and retention of trace metals from atmospheric deposition became stronger than lithogenic input by weathering.

Ecosystem stocks of Cd and Pb varied little across the chronosequence (Fig. 5a–e) in contrast to our third hypothesis. This is consistent with their inputs from the atmosphere that offset leaching losses (Table S4), again apparently unrelated with the affinity to soil organic matter which is known to be much higher for Pb than for Cd.

5. Conclusions

The cumulative element release of most elements in our weathering experiment was higher at the youngest than at the oldest site, indicating progressive depletion of easily weatherable minerals. However, although these results aligned with our first hypothesis, the pronounced change in metal release over the short period of only 127 years of soil development was striking. The only exception was Pb, for which the cumulative release increased with site age, suggesting an increasing influence of atmospheric inputs.

Vegetation development promoted the progressive redistribution of all trace elements into the biomass, not only the biologically needed ones. Consequently, our second hypothesis was only partly supported, since no clear preference for essential nutrients was observed.

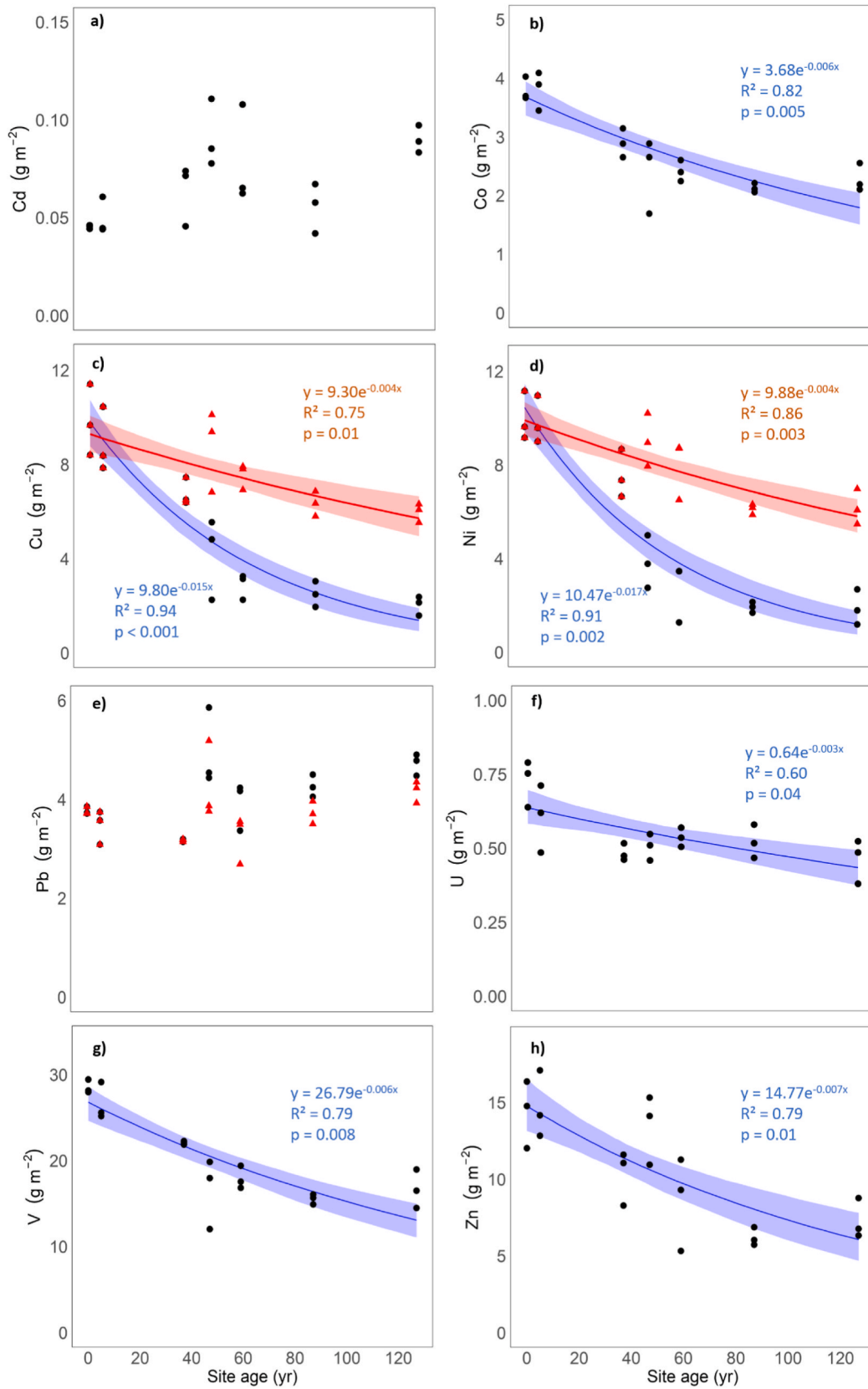


Fig. 5. Total stocks of (a) Cd, (b) Co, (c) Cu, (d) Ni, (e) Pb, (f) U, (g) V, and (h) Zn in the whole study ecosystems including the organic layer (Oa, Oe and Oi), the upper 10 cm of the mineral soil (A and C horizons) and above- and belowground biomass of the vegetation along the Hailuoguo glacier retreat chronosequence. For U, the values of the biomass were below the quantification limit and are therefore not included. The blue line represents the exponential fit of the trace metal stocks as function of site age. The red triangles in c), d), and e) show total stocks, and the red line in c) and d) represent the exponential fit after correction for the a priori lower Cu and Ni concentrations in the parent materials of the four older than three younger sites, because of the heterogeneity of the glacial debris. The shaded area represents the 95% confidence of the fits.

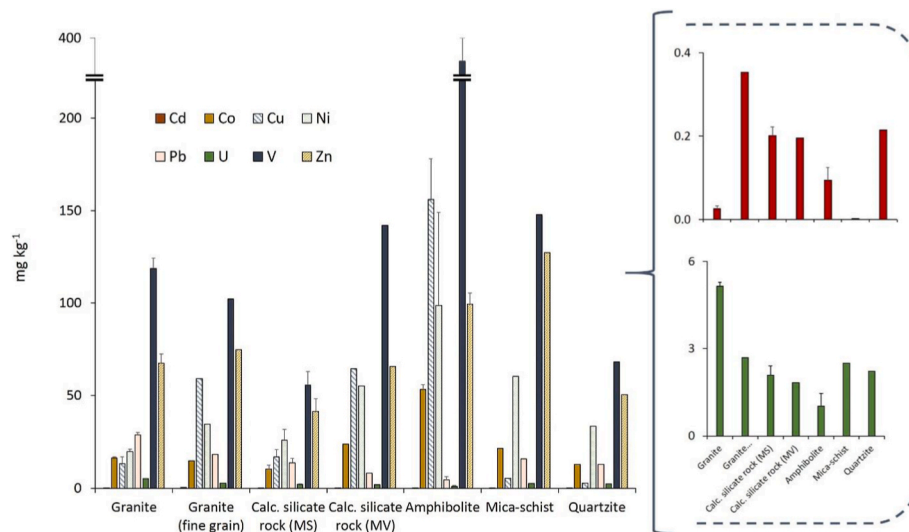


Fig. 6. Mean concentrations of Cd, Co, Cu, Ni, Pb, U, V and Zn in the sampled rock types of the Hailuoguo glacial debris. Error bars indicate the standard deviation of three (granite and amphibolite) or six (calc-silicate rocks) replicate samples. Lacking error bars indicate that only one sample was available. MS, metasedimentary rock; MV, metavolcanic rock.

In contrast to our third hypothesis the accumulation of trace metals in the organic layer and the whole ecosystem was not driven by the metal's particular affinity for organic matter but by the inputs from the atmosphere, which compensated the leaching losses of Cd and Pb irrespective of their contrasting affinities for organic matter.

Overall, we found considerable losses of Co, Cu, Ni, V and Zn (39–49%) in only 127 years of ecosystem development to at least below 10 cm of the mineral soils of which a part was exported via the stream. This illustrated an unexpectedly high mobility of these trace metals. The trace metal losses even reached a similar extent as those of the earth alkaline metals Ca and Mg in contrast to the expectation of a much smaller mobility in soils. This suggests that the trace metal cycling can respond more pronouncedly to current environmental changes including e.g., climate warming, pollutant deposition or soil acidification than up to now expected.

CRedit authorship contribution statement

Nuria Basdediós: Writing – original draft, Visualization, Validation, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Haijian Bing:** Writing – review & editing, Investigation. **Jun Zhou:** Writing – review & editing, Investigation. **Yanhong Wu:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition. **Wolfgang Wilcke:** Writing – review & editing, Visualization, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2026.129292>.

Data availability

Data will be made available on request.

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