

ORIGINAL ARTICLE

Potential accumulation of toxic trace elements in soils during enhanced rock weathering

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Abstract

Terrestrial enhanced rock weathering (ERW) is a carbon dioxide removal technology that aims at accelerating one of the most powerful negative feedbacks on Earth's climate, the chemical weathering of silicates. To achieve this, ERW proposes to spread ground silicate rock on agricultural soils. According to many models, global application rates of 40 tonnes of ground basaltic rock per hectare and per year would be necessary to sequester a significant amount of CO₂, representing up to 24% of the current net annual increase in atmospheric CO₂. When assessing the viability of ERW as a global geo-engineering strategy, a pivotal but overlooked question to address is whether ERW may lead to toxic trace element accumulation in soils at unauthorized and potentially harmful levels. This study evaluates the legal sustainability of ERW with regard to trace element contents in soils. We compare different trace element accumulation scenarios considering a range of rock sources, application rates and national regulatory limits. The results indicate that, at the suggested annual application rate of 40 tonnes per hectare, the first regulatory limits would be exceeded after 6 and 10 years for copper and nickel, respectively. This study argues in favour of close tailoring of ERW deployment to local conditions in order to tap into its climate mitigation potential while preserving long-term soil uses.

Highlights:

- Terrestrial enhanced rock weathering (ERW) is a carbon sequestration technology that consists in applying rock powder to soils.
- This study evaluates whether rock powder application can lead to trace metals accumulation in soils beyond regulatory limits.
- ERW can lead to the overrun of some environmental regulatory limits within less than a decade.
- Specific site and rock selection will be necessary for this method to preserve long-term soil uses.

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KEYWORDS

carbon sequestration, climate change, climate-smart agriculture, enhanced silicate weathering, enhanced weathering, heavy metals, negative emission technology, soil fertility

1 | INTRODUCTION

According to the United Nations Intergovernmental Panel on Climate Change (IPCC), the large-scale deployment of carbon dioxide removal (CDR) technologies is required to achieve the goal of limiting global warming to 1.5°C above pre-industrial levels (IPCC, 2018). Among the possible CDR technologies currently being evaluated, terrestrial enhanced rock weathering (ERW) has attracted considerable attention (Beerling et al., 2018, 2020; Fuss et al., 2018; Hartmann et al., 2013; Kantzas et al., 2022; Renforth et al., 2015; Schuiling & Krijgsman, 2006). Terrestrial ERW consists of grinding reactive silicate minerals to increase their specific surface area and adding them to soils. Atmospheric CO₂ is consumed as these minerals react with carbonic acid during the natural process of chemical weathering. Estimates of the amount of carbon this strategy could sequester vary between 0.2 and 4 Gt CO₂-equivalent annually, corresponding to up to 24% of the current atmospheric CO₂ net annual increase (Beerling et al., 2018, 2020).

Compared to other land-based CDR technologies such as afforestation, ERW presents the distinct advantage that it could be implemented without creating a conflict with current land uses. In addition, many studies point to a number of potential co-benefits of ERW, such as replenishing nutrient pools and reversing soil acidification (Beerling et al., 2018; Edwards et al., 2017; Hartmann et al., 2013). The application of silicate rock powder could therefore aid in the regeneration of deteriorated agricultural soils and, as a result, improve food security worldwide and boost other land-based CDR technologies (de Oliveira Garcia et al., 2020).

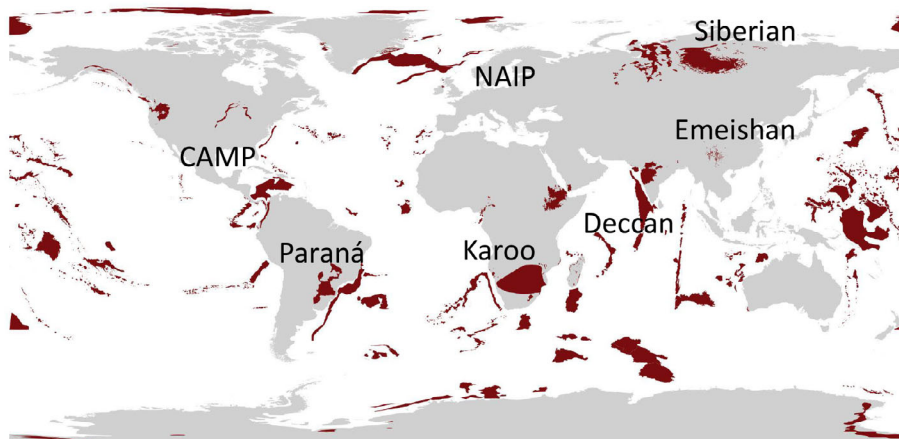
To achieve meaningful CO₂ sequestration volumes, large-scale ERW deployment scenarios generally consider application rates on agricultural soils ranging between 10 and 100 tonnes of rock per hectare and per year, with several recent studies basing their calculation on average on 40 t ha⁻¹ yr⁻¹ (Amann et al., 2020; Anda et al., 2015; Beerling et al., 2020; Renforth, 2012; Taylor et al., 2016). It is on the basis of such application rates that ERW was considered to have the potential to compete with other nature-based or technological CDR such as afforestation-reforestation or carbon capture and storage, respectively (EASAC, 2018; EASAC, 2019; Fuss et al., 2018).

Applying such large quantities of silicate rock powder raises practical questions (e.g., feedstock availability, grinding, transport and spreading equipment and costs) which may call into question the very feasibility of the practice. One additional central environmental concern is the accumulation in the soil of heavy metals—now more commonly referred to as potentially toxic trace elements (TE)—at levels that could interfere with food production and safety (Hartmann et al., 2013; Renforth, 2012).

Due to this concern, highly reactive ultramafic rocks, which were initially considered as ideal candidates for ERW (Moosdorf et al., 2014; Renforth et al., 2015; Schuiling & Krijgsman, 2006), were later dismissed for being too rich in nickel and chromium. Instead, mafic rocks such as basalts have been suggested as relatively safe sources, with nickel and chromium contents an order of magnitude lower than in ultramafic rocks (Anda et al., 2015; Beerling et al., 2018). When compared with most agricultural soils, however, basalts are still highly enriched in certain TE (Bradl, 2005; Kabata-Pendias, 2010; Lar & Gusikit, 2015). Of special concern are nickel, chromium, copper and zinc, all known for their relatively low mobility in the soil profile as well as their toxic effects at elevated concentrations on crops, soil organisms and humans (Kabata-Pendias, 2010; Sharma & Agrawal, 2005).

The need to address and quantify TE accumulation in soils has been repeatedly emphasized as a research gap (Beerling et al., 2018; Choi et al., 2021; Goll et al., 2021; Haque et al., 2020). However, a quantitative exploration of this problem has yet to be carried out. In this study, we calculate TE accumulation trajectories at the currently suggested application rates using geochemical data from different basaltic rock sources. In order to assess whether these TE accumulation trajectories could be potentially problematic, we then compare the results to the regulatory limits for TE contents in soils from top agricultural-producing countries. These regulatory limits were used as benchmarks for potentially concerning soil contamination levels, in the absence of more detailed consensual standards on soil quality. This allows us to assess the legal feasibility of ERW within existing national regulatory frameworks and to identify practical constraints and opportunities to improve the technology's viability.

FIGURE 1 Location of igneous provinces considered in this study (Source: Coffin et al., 2013).



2 | METHODS AND DATA

2.1 | TE contents in mafic and ultramafic rocks

Here, we considered the nickel, chromium, copper and zinc contents of basalts from seven large igneous provinces (LIP) with at least a partly continental occurrence. Their geographical extent makes them the most probable rock sources for ERW, and the availability of large geochemical datasets on these LIP allows for robust estimations of each TE central tendency and dispersion. The total elemental contents (parts per million) in these mafic rocks were obtained using the Geochemistry of Rocks of the Oceans and Continents (GEOROC) database from the Max Planck Institute recently migrated to Göttingen University (<https://georoc.eu/georoc/>). Basalts from the following prominent LIP across the globe were included: Central Atlantic magmatic province (CAMP), Paraná-Etendeka traps, Karoo-Ferrar LIP, North-Atlantic igneous province (NAIP), Deccan traps, Siberian traps and Emeishan traps (Figure 1). The TE contents of three major ultramafic rocks (Iherzolite, dunite and harzburgite) were also extracted from the GEOROC database and were used for the sake of comparison. For each TE and rock source, values beyond 1.5 times the interquartile range were discarded as outliers (Tukey, 1977). Mean values for each LIP and the overall mean for basalts and ultramafic rocks were calculated for each TE (Table S1).

2.2 | National legislation on maximum TE contents in soils

Despite differences between national regulations, limits for nickel, chromium, zinc and copper are found in almost all soil protection legislation. Given the central role of agricultural soils in the context of ERW

deployment, we compiled the regulatory limits for TE contents in soils from several top agricultural-producing countries (Table S2). In some countries, like the United States, legislation related to the content of toxic TE only applies to specific contexts, for example, the application of sewage sludge on agricultural land, excluding the cases relevant to ERW. In the absence of applicable legislation for the United States and the European Union, our compilation of national legislation included China, Brazil, Canada, Germany and Russia.

National regulatory values are commonly based on so-called ‘pseudo-total’ TE concentrations as determined using the *aqua regia* digestion (International Organization for Standardization, 1995; US Environmental Protection Agency, 1996). This extraction method only partially dissolves primary and secondary silicate minerals, leaving a fraction of TE unextracted and unquantified. As such, soil regulatory limits could not be directly compared with rock total TE contents classically determined by x-ray fluorescence or using a complete digestion with hydrofluoric acid.

To circumvent this methodological limitation, we adjusted the value of the regulatory limits for the four countries which use pseudo-total TE values (Brazil, Canada, Germany and Russia) to reflect the incomplete extraction power of the *aqua regia* digestion. Specifically, we divided each *aqua regia*-based national limit by the median extractability of each TE and used the resulting value to represent national upper limits. The median degree of extractability for nickel, chromium, copper and zinc used in this study (88.4%, 55.9%, 90.6% and 89.5%, respectively) were obtained from Taraškevičius et al. (2013).

As a side note, it should be pointed out that other regulations sometimes exist for maximum TE contents in products that can be applied to soils, such as fertilizers and inorganic amendments. To the best of our knowledge, these would not generally prevent the deployment

of ERW. For the countries included in this study, China has set upper chromium and nickel limits at 500 and 100 ppm, respectively, and Germany has set upper nickel limits for rock powders at 120 ppm. All these limits are higher than the mean TE contents found in basalts (Table S3). Brazil has regulated several TE such as arsenic or mercury but none of the four TE are of interest in this study. Canadian guidelines do not set absolute TE limits for products but require that their application over 45 years does not exceed certain TE amounts per hectare (e.g., 150 kg of copper ha⁻¹ over 45 years). In the case of ERW, either his regulation would allow basalt to be used but would imply that operations would need to be halted after one to a few decades or that application rates would need to be decreased. Regarding Russia, no publicly available document with TE limits for soil products could be found. As a whole, these product regulations would therefore not prevent the use of basalt in ERW thereby leaving the potential for regulatory infringements with national soil regulations.

2.3 | TE accumulation in soils

All soils in their natural state contain a certain amount of TE. World mean values for TE contents in soils extracted from Kabata-Pendias (2010) were used as starting contents before simulating TE accumulation or dilution (depending on how much TE was contained in the rock source compared with the initial natural soil). Other sources of TE such as fertilizers and pesticides were not included due to their high variability in agricultural management practices.

During ERW, some of the powdered rock components (e.g., volcanic glass, ferromagnesian minerals) undergo rapid chemical weathering while other components (e.g., feldspars) may endure in the soil for several decades or centuries. Furthermore, some products of chemical weathering may be exported out of the soil profile as dissolved species while others may re-precipitate as secondary mineral phases. The extent and rate of rock powder dissolution and the fate of the weathering products collectively determine the CO₂ removal efficiency of the ERW technology. Their estimates under field conditions are the subject of considerable uncertainty in the literature (White & Brantley, 2003). In the absence of robust quantitative estimates for these processes, we show conservative results calculated based on an annual powdered basalt dissolution rate of 1% on a mass basis. This estimate, termed *minimal weathering*, was derived from the different weathering experiments led by the Carbon Drawdown Initiative (www.carbon-drawdown.de).

In order to bracket the whole possible range of TE behaviour, we used two additional scenarios representing the upper and lower potential limits for basalt weathering kinetics. The first one, termed *maximal weathering*, essentially assumes complete dissolution of the rock powder (100%) and preferential retention of immobile TE, proportionally to their general biogeochemical lability. This is essentially the ‘worst-case’ scenario in terms of TE accumulation. The second scenario, termed *inert accumulation*, represents the lower limit of possible TE accumulation in soils. This scenario assumes no dissolution of the rock powder (0%), meaning that, with time, the topsoil TE content would asymptotically tend toward that of the basalt. Calculation details are found in the following sub-sections.

For all scenarios, yearly TE leaching rates were derived from Tyler (1978) and correspond to upper values measured at pH 4.2 under a 1000 mm precipitation regime. For nickel, copper, chromium and zinc, these rates are 16.6%, 4.9%, 2.6% and 9.8% respectively. The yearly TE removal through crop uptake was estimated by crossing world mean grain crop yields (FAOSTAT, 2020) and published TE contents of those crops (Table S4) (Kopernická et al., 2021; Mulvaney & Devkota, 2020; Rêczajska et al., 2005; Teklić et al., 2013).

2.4 | Minimal weathering

The minimal weathering scenario assumed (1) a gradual dissolution with time (1 wt% yr⁻¹) of the basalt powder; (2) the annual removal of some TE both by leaching and crop removal; and (3) the immobilization and accumulation of the remaining TE fraction, by precipitation into stable secondary phases or sorption on the reactive surfaces of soil particles. The nickel, chromium, copper and zinc content in soil after n years is the sum of the natural TE content in soil and of the TE content in the unweathered and in the weathered basalt fractions, as follows:

$$[\text{TE}_{\text{soil}}]_n = [\text{TE}_{\text{natural fraction}}]_n + [\text{TE}_{\text{unweathered fraction}}]_n + [\text{TE}_{\text{Net weathered fraction}}]_n \quad (1)$$

The meaning of each term and associated units are provided in Table 1.

The first term of Equation (1) $[\text{TE}_{\text{natural fraction}}]_n$ corresponds to the TE content in the natural soil fraction after n years:

$$[\text{TE}_{\text{natural fraction}}]_n = \frac{[\text{TE}_{\text{soil}}]_0 \times (\text{Volume}_{\text{soil}} - \text{Volume}_{\text{rock } n})}{\text{Volume}_{\text{soil}}} \quad (2)$$

where

$$\text{Volume}_{\text{rock}n} = \frac{\text{Mass}_{\text{application}}}{\text{BD}_{\text{rock}}} \times \sum_{i=1}^n \left(1 - \text{Rate}_{\text{weathering}}\right)^i \quad (2.1)$$

The second term of Equation (1) $[\text{TE}_{\text{unweathered fraction}}]_n$ corresponds to the TE content in the unweathered rock fraction after n years:

$$[\text{TE}_{\text{unweathered fraction}}]_n = \frac{[\text{TE}_{\text{rock}}] \times \text{Volume}_{\text{rock}n}}{\text{Volume}_{\text{soil}}} \quad (3)$$

The third term of Equation (1) $[\text{TE}_{\text{Net weathered fraction}}]_n$ corresponds to the TE content from the weathered rock fraction after n years once annual leaching and crop uptake have been deducted:

$$[\text{TE}_{\text{Net weathered fraction}}]_n = [\text{TE}_{\text{weathered fraction}}]_n \times (1 - \text{Rate}_{\text{leaching}}) - [\text{TE}_{\text{export}}] \quad (4)$$

where

$$[\text{TE}_{\text{weathered fraction}}]_n = \frac{[\text{TE}_{\text{rock}}] \times \text{Rate}_{\text{weathering}} \times \sum_{i=1}^n \text{Mass}_{\text{rock}i}}{\text{BD}_{\text{soil}n} \times \text{Volume}_{\text{soil}}} \quad (4.1)$$

with

$$\text{Mass}_{\text{rock}i} = \text{Rate}_{\text{weathering}} \times \text{Mass}_{\text{rock}i-1} + \text{Mass}_{\text{application}} \quad (4.2)$$

Note that

$$\text{Mass}_{\text{rock}0} = \text{Mass}_{\text{application}} \quad (4.3)$$

and

$$\text{BD}_{\text{soil}n} = \frac{\text{Volume}_{\text{rock}n} \times \text{BD}_{\text{rock}} + (\text{Volume}_{\text{soil}} - \text{Volume}_{\text{rock}n}) \times \text{BD}_{\text{soil}0}}{\text{Volume}_{\text{soil}}} \quad (4.4)$$

2.5 | Weathering boundary conditions: Maximal weathering and inert accumulation

The maximal weathering limit was calculated based on immobilization of TE in soils assuming (1) a complete dissolution of the basalt powder within the first year after each application; (2) the loss of some TE via leaching and agricultural crop removal, as above; and (3) the immobilization and accumulation of the remaining TE fraction, by precipitation or sorption on the reactive surfaces of soil particles. The maximum weathering scenario corresponds to a mathematical sequence which the general n th term equation yields:

$$[\text{TE}_{\text{soil}}]_n = [\text{TE}_{\text{soil}}]_0 + n \times \left(\frac{[\text{TE}_{\text{rock}}] \times \text{Rate}_{\text{application}}}{\text{BD}_{\text{soil}} \times \text{Volume}_{\text{soil}}} \right) \times (1 - \text{Rate}_{\text{leaching}}) - [\text{TE}_{\text{export}}] \quad (5)$$

In the inert accumulation scenario, basalt powder accumulates in the soil in the absence of any weathering reaction. Although absurd in a CDR context (without basalt dissolution, no CO_2 can be sequestered via ERW), this scenario represents a lower limit in terms of possible TE accumulation in soils where the overall TE content of the top 20 cm layer would, year after year, tend toward the basalt TE content. Mathematically, this situation corresponds to an arithmetic-geometric sequence which the general n th term equation yields:

$$[\text{TE}_{\text{soil}}]_n = [\text{TE}_{\text{rock}}] + ([\text{TE}_{\text{soil}}]_0 - [\text{TE}_{\text{rock}}]) \times \left(\frac{\text{Volume}_{\text{soil}} - \frac{\text{Rate}_{\text{application}}}{\text{BD}_{\text{rock}}}}{\text{Volume}_{\text{soil}}} \right)^n \quad (6)$$

Additional calculation details for both scenarios can be found in Supplementary Material.

TABLE 1 Meaning and units for each variable used in the trace elements (TE) accumulation scenarios. Calculations were performed on a 1 m² area.

Variable	Definition	Unit (and value if relevant)
$[\text{TE}_{\text{soil}}]_n$	TE content in soil after n years	mg of TE kg ⁻¹ of soil
$[\text{TE}_{\text{natural fraction}}]_n$	TE content from natural fraction after n years	mg of TE kg ⁻¹ of soil
$[\text{TE}_{\text{unweathered fraction}}]_n$	TE content from unweathered rock fraction after n years	mg of TE kg ⁻¹ of soil
$[\text{TE}_{\text{weathered fraction}}]_n$	TE content from weathered rock fraction after n years	mg of TE kg ⁻¹ of soil
$[\text{TE}_{\text{soil}}]_0$	Mean initial TE content in natural soils	mg of TE kg ⁻¹ of soil
$[\text{TE}_{\text{rock}}]$	Mean TE content of applied rock powder	mg of TE kg ⁻¹ of rock
$[\text{TE}_{\text{export}}]$	TE exported by crops	mg of TE kg ⁻¹ of soil
$\text{Rate}_{\text{leaching}}$	TE amount annually leached	Mass fraction (mass of leached TE/total mass of TE)
$\text{Rate}_{\text{weathering}}$	Share of rock powder being annually weathered	Mass fraction (here set at 1%)
$\text{Mass}_{\text{application}}$	Rock powder amount annually applied to the soil	kg of rock m ⁻²
$\text{Mass}_{\text{rock } n}$	Undissolved rock powder mass after n years	kg of rock m ⁻²
$\text{Volume}_{\text{soil}}$	Volume of a 20 cm soil slice m ⁻²	m ³ (here set at 0.2 m ³)*
$\text{Volume}_{\text{rock } n}$	Accumulated rock volume after n years	m ³ of rock
BD_{rock}	Rock powder bulk density	kg of rock m ⁻³ of rock (here set at 2000 kg m ⁻³)
$\text{BD}_{\text{soil } n}$	Soil bulk density after n years	kg of soil m ⁻³ of soil
$\text{BD}_{\text{soil } 0}$	Soil bulk density before the first rock powder application	kg of soil m ⁻³ of soil (here set at 1300 kg m ⁻³)

*Soil volume calculated consistently with national regulations for soil quality monitoring based on a 0–20 cm soil depth.

3 | RESULTS

First, the impact of different rock sources on TE accumulation in soil was calculated using mean TE contents for the different LIP and a fixed rock powder application rate of 40 t ha⁻¹ yr⁻¹. Different application rates ranging from 10 to 100 t ha⁻¹ yr⁻¹ were then considered, using the overall mean TE content in basalts. Finally, the *minimal weathering* scenario was compared to the two boundary scenarios (*maximal weathering* and *inert accumulation*) in order to bracket the full range of possible TE accumulation trajectories.

3.1 | Influence of rock source

The TE content of basalts varied strongly within and between the different LIP (Figure 2). While basalts typically had less nickel and chromium than ultramafic sources, they also had 2.5 times more copper and 1.4 times more zinc (see Table S1 in Supplementary Material for further data on each LIP).

When considering rock application at a rate of 40 t ha⁻¹ yr⁻¹, these differences in TE content led to significant variations in TE accumulation in soils (Figure 3). For ultramafic rocks, there would be almost immediate conflicts with national soil protection policies for nickel and chromium, supporting the emerging

consensus that these rock sources should be discarded for terrestrial ERW. The use of basalts, broadly assumed to be less problematic in terms of TE composition, would, however, only delay environmental issues. The first regulatory limits for copper and nickel would be exceeded after 6 ± 3 years and 10 ± 4 years, respectively, depending on the basalt source. Chromium and zinc accumulations appear less problematic, remaining below all examined limits for at least 38 and 94 years, respectively. Copper appears here as the most concerning TE given that its accumulation in soil would end up infringing all national limits after 36 ± 15 years depending on the basalt composition. The impact of the rock source composition would likely be even more pronounced at a regional or local scale, considering the often highly dispersed and multimodal distributions of TE contents in basaltic provinces (Figure 2).

3.2 | Influence of application rate

Higher application rates (80 to 100 t ha⁻¹ yr⁻¹) would lead to the first regulatory infringements after only 5 and 8 applications for copper and nickel, respectively. Even the most favourable national legislation would be exceeded within 25 years for copper and 48 years for nickel (Figure 4).

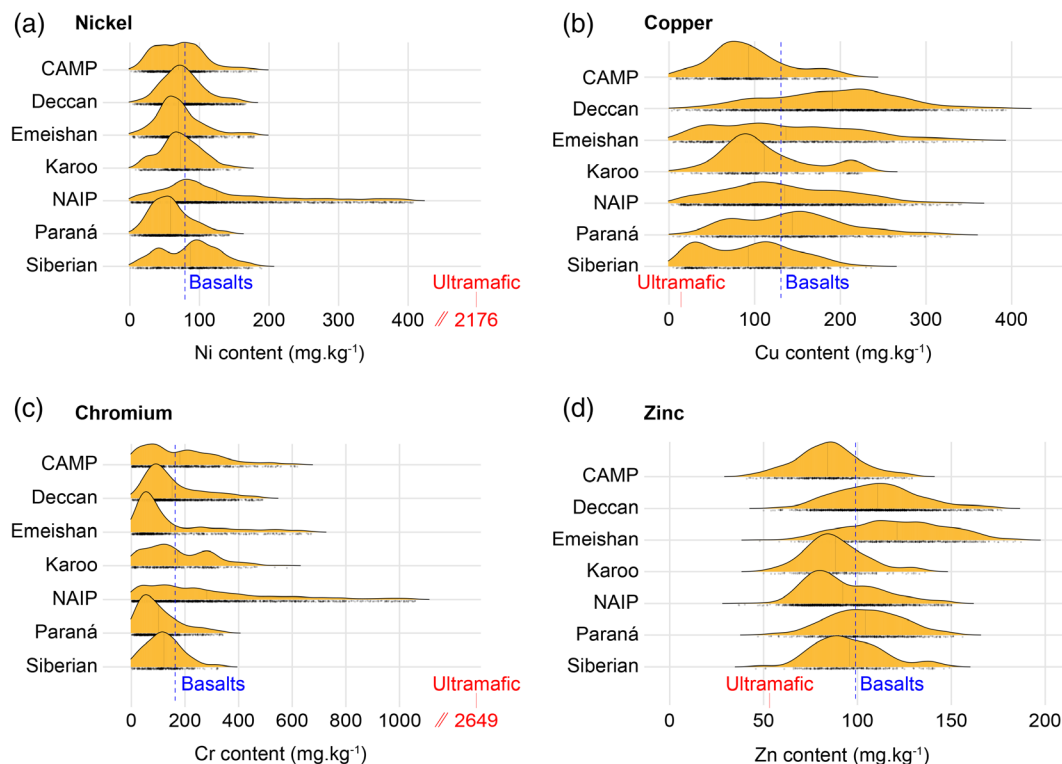


FIGURE 2 Nickel, copper, chromium and zinc content in basalts from several prominent continental large igneous provinces extracted from the GEOROC database (<https://georoc.eu/georoc/>) and represented as density plots. For each element, the mean content for basalts of each province is represented as a vertical black line and the overall basalt mean is represented as a dashed blue line. For comparison, the overall mean trace element content for common ultramafic rocks (lherzolite, dunite and harzburgite) extracted from the GEOROC database are given in red. CAMP, Central Atlantic Magmatic Province; NAIP, North Atlantic Igneous Province.

Lower application rates ($10 \text{ t ha}^{-1} \text{ yr}^{-1}$) would obviously decrease TE accumulation rates, however copper and nickel contents in soil would still exceed multiple regulatory limits after less than 15 and 35 years, respectively.

3.3 | Influence of weathering rate

The *minimal weathering* scenario, on which the previous calculations are based, represents a conservative approximation of TE accumulation in soils under realistic weathering conditions. In order to further constrain the range of possible TE accumulation in soils, we also considered two boundary scenarios representing a lower (*inert accumulation*) and upper (*maximal weathering*) potential TE accumulation trajectory (Figure 5). For copper, the first regulatory limit (Germany) would become infringed after only 3 years in the *maximal weathering* scenario, compared to 7 years under the *inert accumulation* scenario. The impact of the weathering scenario on TE accumulation becomes larger when longer time spans are considered. All considered national limits for copper would

be exceeded within 20 and 47 years under the *maximal weathering* and *inert* scenarios, respectively.

4 | DISCUSSION

The evaluation of potential TE accumulation in soils highlights potentially serious constraints on the feasibility of ERW from a regulatory standpoint. Whereas the potential TE release and accumulation in soils from ultramafic rocks during ERW has been known for years, our calculations indicate that similar issues may be encountered even when mafic rocks such as basalts are used. In several of the possible combinations of jurisdiction, rock source and scenario of TE accumulation considered, the commonly suggested application rates of $40 \text{ t ha}^{-1} \text{ yr}^{-1}$ would start leading to environmental regulatory issues after only a few rock powder applications. If confirmed, these results would stand in contrast to most current projections for ERW in which the contents of potentially toxic TE in basalts are assumed negligible (Anda et al., 2015; Beerling et al., 2018).

Our results suggest that copper may be particularly problematic. For all investigated scenarios, copper is

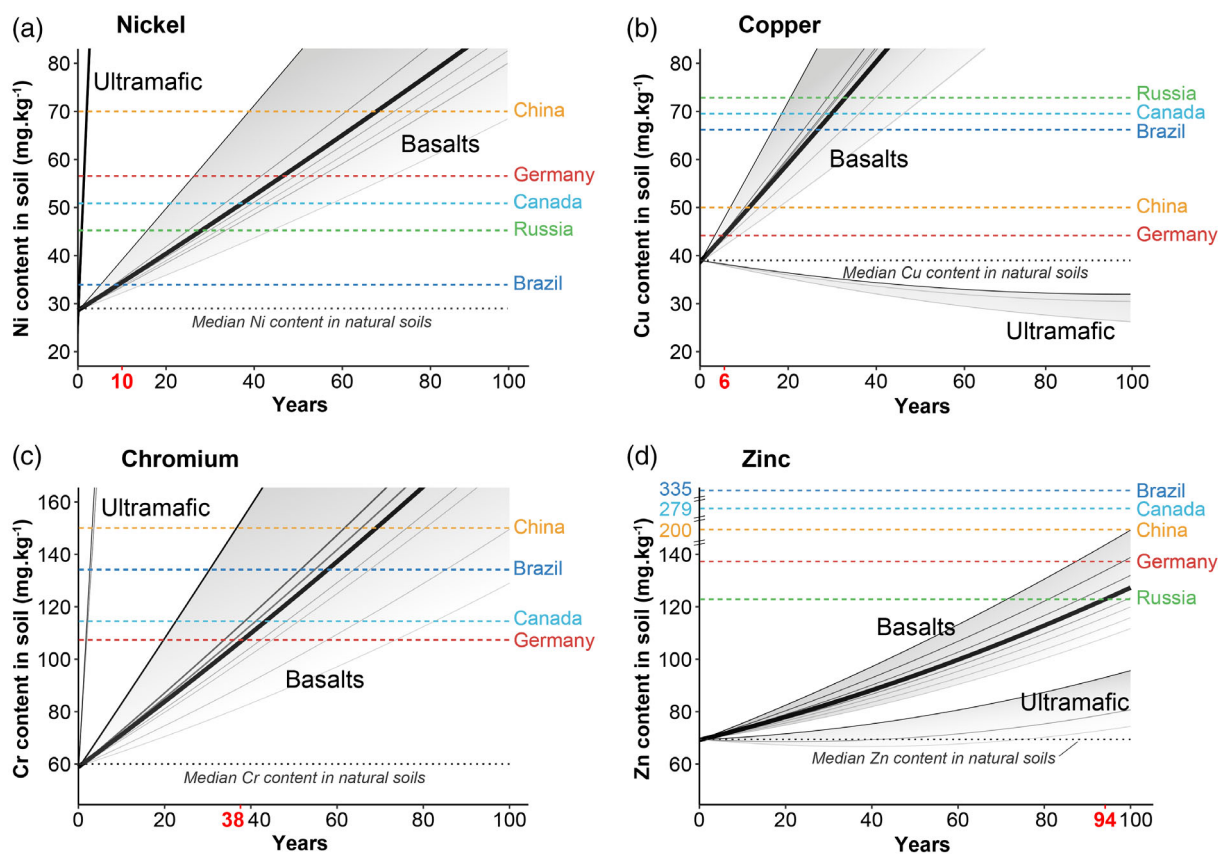


FIGURE 3 Gradual accumulation of trace elements (TE) in soils assuming an annual rock powder application rate of $40 \text{ t ha}^{-1} \text{ yr}^{-1}$. For basalts, shaded areas represent the range of TE accumulation depending on the rock source. Each grey line represents one of the large igneous provinces included in this study and the black bold line represents the overall mean. Red figures on the x-axis are the number of years after which the first regulatory limit would be exceeded, for a basalt of average composition. For ultramafic rocks, the grey lines represent TE accumulation associated with peridotite rocks (dunite, lherzolite and harzburgite). Horizontal dotted lines represent world mean TE contents in natural soils and regulatory limits for TE in soils for Brazil, Canada, China, Germany and Russia.

indeed the first element exceeding regulatory limits. Copper has a very low susceptibility to leaching and tends to strongly accumulate in the upper layers of the soil (Adrees et al., 2015; Nesbitt & Wilson, 1992). As copper-bearing primary minerals dissolve, the vast majority of soluble copper will rapidly form stable inner-sphere complexes with organic matter and, to a lesser extent, manganese and iron oxides (Karlsson et al., 2006; Oorts, 2013). Due to its very high affinity for organic matter, the effect of pH on copper solubility is limited and lower than for most other TE, thus leading to a particularly limited leaching potential (Oorts, 2013; Ponizovsky et al., 2006; Sauvé et al., 1997). The range of possible accumulation rates is therefore considerably narrower for copper than for other TE. As an example, the estimated time to reach the highest national limit ranges from about 20 to 47 years under the lower and upper boundary scenarios, respectively.

It should additionally be noted that copper contamination already represents a major issue in many of the

world's agricultural soils, limiting crop productivity (Adrees et al., 2015) and posing a potential risk to food safety (Fagnano et al., 2020). In our projections of TE accumulation, we deliberately did not account for any TE inputs not related to ERW, for example, additional anthropogenic inputs from sources such as pesticides, manure and sewage sludge (Panagos et al., 2018). The extent of additional anthropogenic copper inputs would vary considerably depending on agronomic practices but has the potential to significantly accelerate the overall accumulation of this element. In wine-making regions for instance, the long-term use of copper-based fungicides to control vine downy mildew has brought copper contents in soils to levels beyond most regulatory limits. Copper contents currently range between 70 mg kg^{-1} of soil in the south of Europe and 800 mg kg^{-1} in the Bordeaux region (Brun et al., 1998; Ruyters et al., 2013). Despite recent regulatory developments (the European Union has for instance restricted the use of copper in plant protection to a maximum of

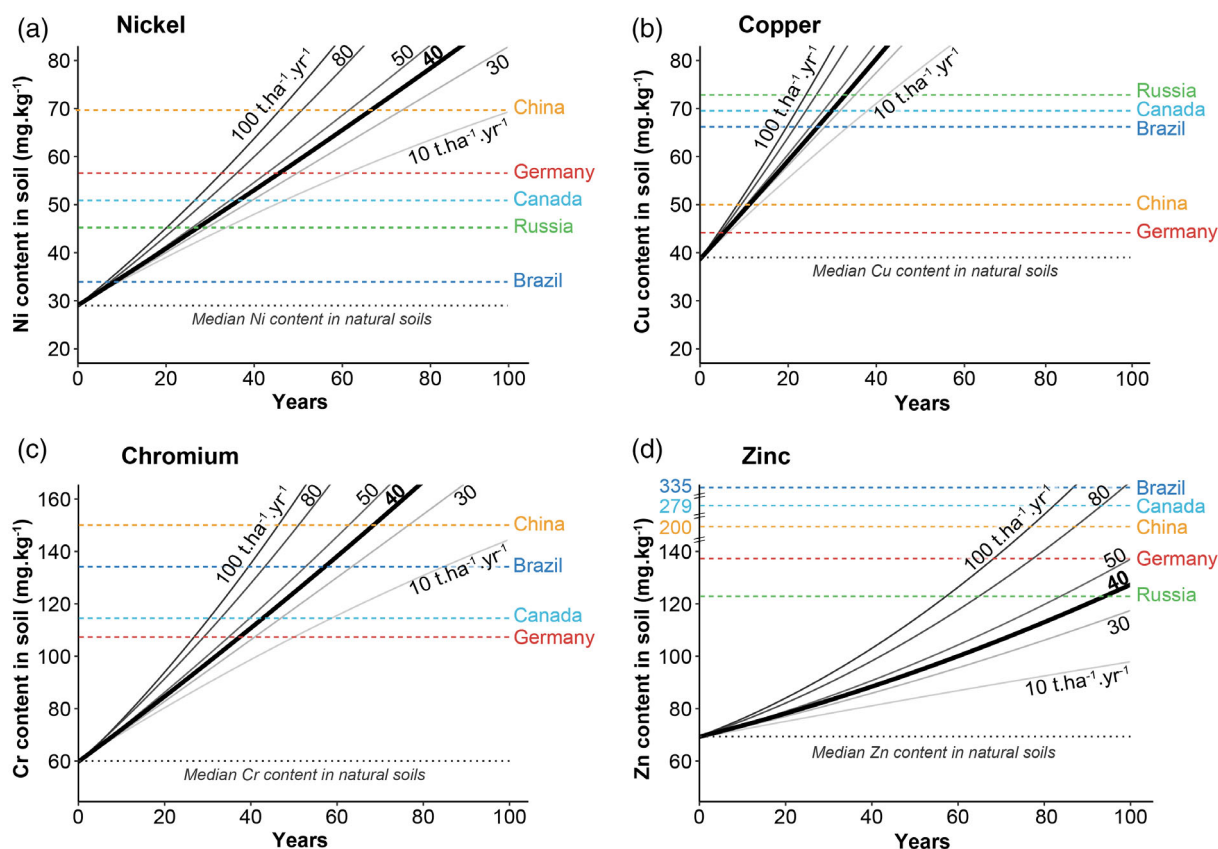


FIGURE 4 Trace elements (TE) accumulation in the soil at varying application rates of basalt powder ranging from 10 to 100 t ha⁻¹ yr⁻¹, with the most commonly recommended rate of 40 t ha⁻¹ yr⁻¹ in bold. Horizontal dotted lines represent world mean TE contents in natural soils and upper regulatory limits for different countries.

4 kg copper ha⁻¹ y⁻¹; EU Reg. 1981/2018), copper content in these soils is still estimated to increase by 1 mg kg⁻¹ per year (Fagnano et al., 2020). In such circumstances, ERW could only aggravate an already serious situation.

The methodological choices used in this study are not exempt from uncertainties. Median conversion factors compiled by Taraškevičius et al. (2013) were used in our calculations to compare rock TE total content with the pseudo-total aqua regia extraction values used in environmental regulations. Depending on the exact mineralogy, the conversion from 'pseudo-total' to total element content can of course vary. However, even when using the minimal and maximal values reported by Taraškevičius et al. (2013) for the degree of aqua regia extractability (respectively 79.2% and 97.9% for copper for a median value of 90.6%), we find that the time before reaching the different regulatory limits would only be changed by 1 to 4 years depending on countries. Additionally, crop uptake rates might be expected to increase with TE accumulation in soils, although to very variable extents depending on crop species. Yet the magnitude of TE export through crop removal remains too small to

significantly influence accumulation trends. Rock powder applications of 40 t ha⁻¹ y⁻¹ add for instance 341 more copper than crops removed under mean crop removal rates (Tables S2 and S4) and still 32 times more copper than heavily contaminated grains could remove (Sharma et al., 2012).

Our calculations were based on several additional assumptions which we argue are rather conservative, or optimistic, regarding TE accumulation potential. For instance, the analysis of several commercially available basaltic rock powders revealed that some sources have nickel or chromium contents up to 50% higher than the upper end of the range used in our study (Lewis et al., 2021). Similarly, the initial soil TE content in our study was set to be that of average natural soil. Soils with a long history of agricultural use, however, often present significantly higher TE contents. This originates, among other factors, from the use of livestock manure which can have more than 10 times the copper and zinc contents we used for basalts (Nicholson et al., 2003). Additionally, the leaching rates employed in our scenarios were derived from experiments in highly acidic conditions (Tyler, 1978). As the TE considered in this study are

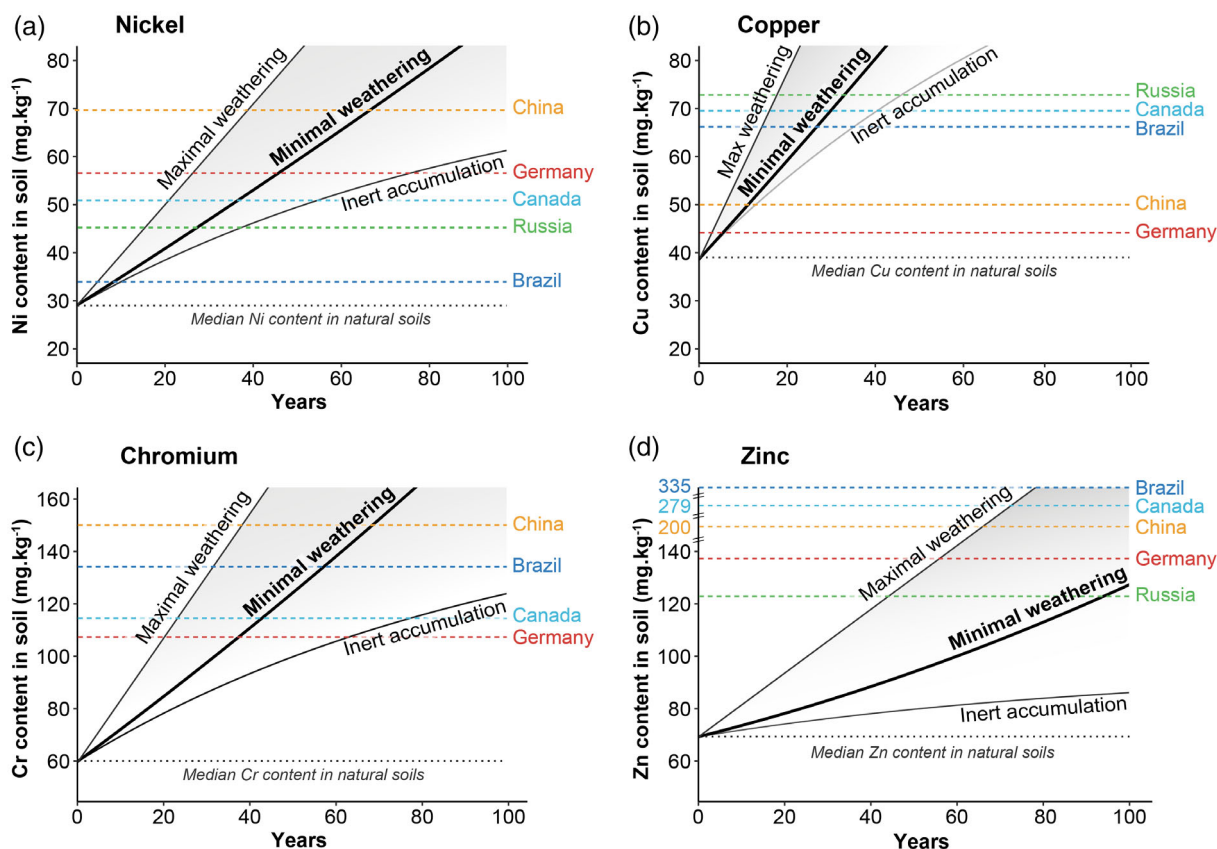


FIGURE 5 Trace element (TE) accumulation in soil assuming an application rate of $40 \text{ t ha}^{-1} \text{ yr}^{-1}$ of basalt using a minimal weathering scenario (bold) bracketed by a lower and upper accumulation trend (maximal weathering and inert accumulation, respectively). Horizontal dotted lines represent world mean TE contents in natural soils and regulatory limits for different countries.

more mobile at low pH (Kabata-Pendias, 2010), slightly acidic to circumneutral pH, which is common in agricultural lands, could result in substantially lower TE leaching rates and accordingly higher accumulation rates. Altogether, the input parameters chosen for our calculations thus represent a rather favourable ERW deployment scenario with regard to TE accumulation. In specific conditions, TE accumulation rates could far exceed our estimates. In a recent mesocosm experiment using a basalt that was particularly rich in nickel, Vienne et al. (2022) reported for instance that a single application of 50 t ha^{-1} increased the nickel content of the soil (0–50 cm) by 23 mg kg^{-1} . This increase, 48 times higher than our estimates, illustrates how TE accumulation could prevent ERW from being implemented altogether in unfavourable contexts.

Despite the potential regulatory constraints applicable to ERW with regard to soil protection policies, the practice retains an interesting potential as a CDR technology. The CO_2 sequestration potential as well as the agronomic and environmental co-benefits could still prove beneficial even when using lower application rates and/or frequencies that alleviate the potential of problematic TE

accumulations in soils as well as other potentially serious logistical challenges such as feedstock availability or mining, grinding and transport infrastructure needs. Goll et al. (2021) demonstrated that a single basalt application of 50 t ha^{-1} on selected areas could still lead to a substantial CDR of $1.3 \text{ GtCO}_2\text{eq yr}^{-1}$ over 50 years due to positive feedback on ecosystem carbon storage. This study points to the need to optimize ERW deployment by a careful selection of sites with favourable conditions, for example, nutrient-poor soils close to suitable rock sources. Additionally, the use of basalts low in problematic TE on soils with low starting TE contents might even trigger a ‘micronutrient fertilization effect’ (Wyszkowska et al., 2013). In soils depleted in zinc by the intensification of agriculture, this practice could even alleviate the health issues associated with zinc deficiency (Fischer Walker et al., 2009). The selection of rock source based on TE composition would, however, likely require compromises with regard to other selection criteria such as logistical, social acceptance and financial considerations.

Another possible measure to facilitate the deployment of ERW would be a revision of relevant soil protection policies. This would only make sense if environmental

and health risks could be safely ruled out even at higher soil TE limits, as is sometimes suggested (Fagnano et al., 2020). In a trial at the mesocosm scale, Kelland et al. (2020) showed for instance that very large applications of 100 t ha^{-1} of basalt powder did not lead to elevated contents of nickel, copper or chromium in the cereal *Sorghum bicolor*. In the experiment conducted by Vienne et al. (2022) on *Solanum tuberosum*, the addition of 50 t ha^{-1} of basalt did not lead either to an elevated nickel content in crops despite a significant increase in soil nickel. In its regulation on soil products, Germany has already moved in this direction by increasing the maximum allowable nickel content in rock powders by 50% compared with other fertilizers and additives (Table S3).

Given that TE tend to be more soluble at low pH, the ability of basaltic rock powder to increase the pH of highly weathered and acidic soils (Gillman et al., 2002) suggests that ERW operations could reduce the mobility and bioavailability of potentially toxic trace elements. This effect is well-studied for traditional liming (Goulding & Blake, 1998; Tlustos et al., 2006) and could, in different agricultural settings, offer an argument in favour of higher TE limits in soils for ERW operations. The high pH would, however, have to be maintained indefinitely, even after the ERW operations have ceased, to avoid remobilization of accumulated TE. Alternatively, ERW could be deployed in combination with other means of TE immobilization, such as biochar soil amendments, or removal through hyperaccumulator plants, with synergies previously suggested (Amann & Hartmann, 2019) but not yet tested in practice.

Another approach to evaluate the impact of ERW could consist in studying TE accumulation in natural soils developed on volcanic rocks. Many volcanic soils exceed indeed the legal limits presented in this study. In the basaltic Panyam province of Nigeria for instance, the nickel content in soils ranges between 89 and 411 ppm (Lar & Gusikit, 2015). In the basaltic Plateau of the Rio Grande do Sul (Brazil), soils display an average copper content of 101 ppm (Althaus et al., 2018) while the basaltic-derived soils from the Sabzevar Range in Iran contain between 85 and 109 ppm copper, 149–153 ppm nickel and 154–183 ppm chromium (Mazhari et al., 2017). All these soils exceed the highest regulatory thresholds presented here. Despite this, groundwater or crop contamination as well as reports of health issues are not systematic. None of such adverse effects associated with farming on these soils could for instance be found in the Plateau of the Rio Grande do Sul or in the Sabzevar Range. In the Panyam Province, health issues were mainly associated with more mobile and often unregulated TE such as selenium (Lar & Gusikit, 2015).

Conversely, in basaltic Xuyi County in eastern China, Wang et al. (2020) reported nickel accumulations in crop grains beyond national food safety recommendations associated with ingestion health risks. It should additionally be noted that metal transfer to crops is likely to differ in basaltic materials that have undergone centuries to millennia of weathering, compared to soils amended with fresh basalt powder. These examples suggest that careful geochemical and ecotoxicological assessments are now needed before opening a discussion around the adaptation of soil protection regulations in the context of ERW.

The rock weathering kinetics and the rates of TE accumulation during ERW will depend heavily on regional and local conditions including climate, soil properties and land management (Kabata-Pendias, 2010). Specific studies are now required to quantify the rate of TE accumulation under different climatic and agricultural contexts. Given the natural spatial heterogeneity of soils, the question of TE's lowest detectable difference will also need to be explored. This aspect could indeed prove to be a challenge when monitoring the very first years of ERW operations. In our calculations starting from world mean TE contents in natural soils, the currently considered annual application rates would lead copper or nickel contents in soils to double after 40 years leaving no doubt that such changes would become detectable in the long run.

The impact of basalts' mineralogical heterogeneity on TE accumulation in soils is another avenue that requires further research. The behaviour of TE varies indeed with the reactivity of the minerals that contain these TE. Depending on the chemical composition of the magma, chromium for instance could end up in chromite, a mineral known for its stability to weathering (Garnier et al., 2008), or in olivine which constitutes some of the most fast-weathering silicate minerals (Velbel, 2009). Concerning nickel, copper and zinc, the chemical composition of the magma and especially its level of saturation in sulphur will dictate whether these TE occur in sulfides or in silicate minerals each with distinct reactivities (Li & Audétat, 2012).

Future research also needs to consider other TE contained in basalts, for which precautionary values are usually not yet defined but that could nonetheless pose a serious threat to the environment and to food safety. Among the elements significantly enriched in basalts, vanadium is of special interest. Its average content in mafic rocks is approximately $250\text{--}300 \text{ mg kg}^{-1}$, higher than in any other major lithology (Turekian & Wedepohl, 1961) and four times higher than its median content in European soils (Reeder, 2006). Although the mobility of vanadium is complex due to a high redox sensitivity and multiple valence states, its adsorption on Fe

and Al hydrous oxides and organic matter is well documented (Baken et al., 2012; Gustafsson, 2019; Larsson et al., 2015; Qi et al., 2022; Shaheen et al., 2013) and could lead to its accumulation in soils in an ERW context. Significant ecotoxicological effects, especially related to soil microbiota and enzyme activity, have been reported starting at soil contents below 100 mg kg⁻¹ (Gustafsson, 2019). Mesocosm and field experiments would be of great interest to establish whether the accumulation potential of vanadium contamination could also limit the viability of ERW.

5 | CONCLUSION

All carbon sequestration technologies suggested to date come with specific institutional, technical, economic, or social challenges and terrestrial ERW is no exception. The results of our study indicate that the constraints on the sustainability of ERW, arising from TE accumulation in soils, may have been underestimated. The global, repeated applications of basalt powder at rates of several tens of tonnes per hectare, which has been proposed in several studies, appear in direct conflict with current environmental legislation. The estimated rates of TE accumulation could therefore significantly limit the overall carbon sequestration potential of ERW within existing regulatory frameworks. Modifications of the relevant regulatory limits, or alternative ERW configurations, would be required to overcome this limitation.

Our study points to the need for further research, especially regarding (1) the extent and kinetics of rock powder dissolution and TE accumulation during ERW under natural conditions; (2) crop uptake studies under different weathering and soil conditions and (3) the impact of additional TE loadings from fertilization, pesticides and other anthropogenic sources.

AUTHOR CONTRIBUTIONS

Xavier Dupla: Conceptualization; investigation; writing – original draft; methodology; visualization; software; formal analysis. **Benjamin Möller:** Conceptualization; writing – original draft; methodology; data curation; formal analysis. **Philippe C. Baveye:** Writing – original draft; validation; writing – review and editing; formal analysis. **Stéphanie Grand:** Writing – review and editing; supervision; validation; writing – original draft; formal analysis.

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CONFLICT OF INTEREST

The authors declare no competing interests.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study were derived from the following resources available in the public domain: GEOROC database, <https://georoc.eu/georoc/>

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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