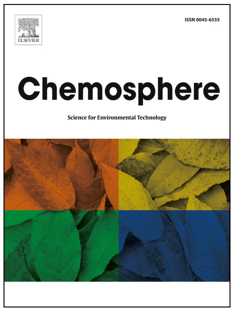

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Glyphosate application may influence the transfer of trace elements from soils to both soil solutions and plants

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Glyphosate application may influence the transfer of trace elements from soils to both soil solutions and plants

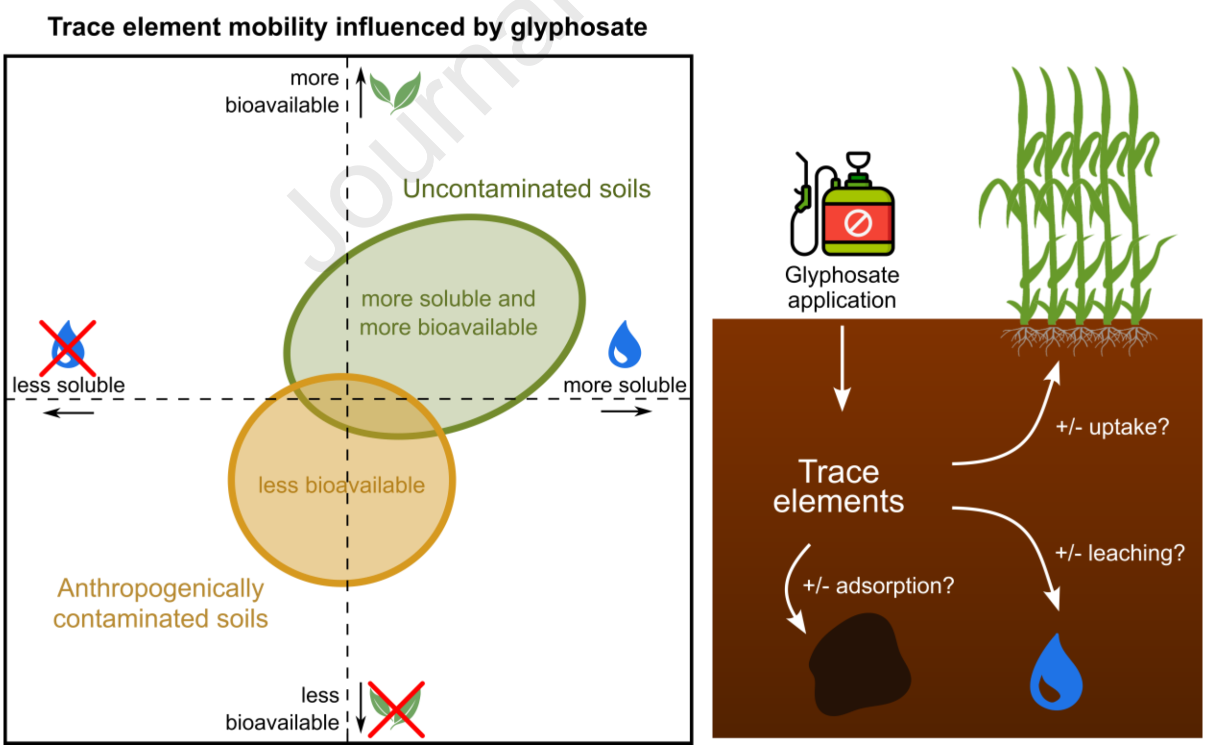
Abstract

Trace elements dynamics in the soil-plant system depends on multiple parameters, including chelating organic compounds from natural or synthetic organic matters. In this study, we evaluated the influence of one of the most common pesticides—glyphosate—on the mobility of trace elements considering contrasted soils (uncontaminated, anthropogenically contaminated, and naturally-enriched) in a greenhouse experiment. Four modalities have been tested: one control without any application, two with different glyphosate doses (1 and 3 times the authorised field dose), and one with compost addition to evaluate its potential ability to mitigate the impact of glyphosate on trace element mobility. Both, trace element and glyphosate concentrations were measured in the soil solutions and trace element contents were determined in plants at the end of the experiment. The results showed that, although glyphosate concentrations rapidly decreased in soil solutions, glyphosate application still influenced the transfer of trace elements to both soil solution (up to 12-times higher) and plant (up to 5.2-times higher). This influence was highly dependent on both the specific elements and the type of soils considered. For instance, in uncontaminated soils, glyphosate especially increased the mobilization of Mn, Co, Zn, Mo, and Pb to soil solution. This effect diminished of 2.5 times on average with increasing soil contamination. A similar trend was observed for the transfer of trace elements from soil to plant (i.e., on average 2.2-times lower in the most contaminated compared to the uncontaminated soil). However, in the naturally-enriched soil, opposing trends were noted between soil solutions and plants. The impact of compost addition on the transfer of trace elements to plants remains unclear:

compost enhanced the transfer of trace elements to soil solution in uncontaminated and naturally-enriched soils likely due to trace element input through the compost, but decreased the transfer in anthropogenically-contaminated soils likely due to adsorption processes. Therefore, glyphosate could potentially increase the exposure of trace elements through food consumption and their transfer to the ecosystem, particularly in uncontaminated and weakly contaminated soils. In highly contaminated soils, compost could mitigate the glyphosate-induced enhancement of trace element mobility to soil solutions.

Keywords: trace elements, glyphosate, soil, plant, transfer, greenhouse experiment

Graphical abstract



1. Introduction

Trace elements, whose content is below 1‰ in the Earth's crust, are naturally present in soils. Additional sources, such as direct application of exogenous organic matter or indirect inputs from industrial atmospheric deposition, may contaminate the agricultural soils (Belon et al., 2012). Although some trace elements are essential for plants and animals, all become toxic above a certain threshold, with particular health concerns for Pb, Cd, Hg, or As (Arnich et al., 2012; Underwood, 1977). Since human exposure is closely linked to agricultural products and drinking water, both soil–plant transfer and leaching to groundwater are key processes for health risk assessment (Barati et al., 2010; Kabata-Pendias, 2011).

As they encompass various chemical families—metals (e.g., Cd, Cu, Zn) and metalloids (e.g., As, Sb)—trace elements behave differently in the environment depending on their speciation and chemical properties (i.e., volatility, solubility, interaction with organic compounds, etc.; Adriano, 2001; Kabata-Pendias, 2011). Moreover, environmental characteristics, such as soil pH, redox, or cation exchange capacity (CEC), are crucial in controlling the transfer of trace elements from one compartment to another. For instance, the presence of organic compounds in soil—whether added (which may have undergone some modification) or natural—can significantly reduce their transfer due to adsorption processes (Hooda, 2010; Pathak et al., 2024). However, adding exogenous organic matter may also introduce extra trace elements to soils (e.g., compost, manure, sewage sludge; Baldantoni et al., 2010; Benke et al., 2008; Berti and Jacobs, 1998). This is especially relevant in the context of climate change, as it is increasingly recommended to add organic matter to soils for carbon storage purposes (Minasny et al., 2017). Additionally, dissolved organic matter from decomposition processes may have a chelating effect, generating a complex that is either more or less soluble than the ion itself, thus either increasing or decreasing trace element mobility

(Carrillo-González et al., 2006; Schmitt et al., 2002). Low molecular weight organic compounds can promote trace element solubility, while higher molecular weight compounds are more likely to form insoluble complexes.

Most pesticides are organic compounds, often present as residues in agricultural soils due to their widespread use (Bhandari et al., 2020; Silva et al., 2019). Some—including glyphosate, a widely used herbicide—are known to have chelating abilities (Barrett and McBride, 2006; Divisekara et al., 2018; McBride and Kung, 1989; Morillo et al., 2002). Glyphosate, along with its main degradation product, aminomethylphosphonic acid (AMPA), has shown a significant effect on the mobility of several trace elements (Barrett and McBride, 2006; Bemelmans et al., 2023; Si et al., 2013). This is primarily due to its multiple charged functional groups (Sprankle et al., 1975): phosphonate (pKa 2.6), carboxylic (pKa 5.6), and amino (pKa 10.6). Additionally, pesticides are frequently applied as formulated products, i.e. with co-formulants that improve their efficacy (Krogh et al., 2003; Leaper and Holloway, 2000). These co-formulants are known to influence the mobility of trace elements from soil to soil solution, either directly or indirectly (Bemelmans et al., 2023). Unfortunately, data on their chemical composition are unavailable due to commercial confidentiality, hindering the identification of the processes involved (Mesnage et al., 2019).

The presence of such organic compounds from pesticide application may influence the mobility of trace elements from soil to soil solution and plant. For example, Cakmak et al. (2009) suggest that the translocation of certain elements may be reduced when associated with glyphosate. Indeed, light organic acids are unable to cross cell membranes and thus must follow the apoplastic route until they reach the xylem through imperfections in the Casparian band (Häussling et al., 1988; Perry and Greenway, 1973; Schaider et al., 2006). These effects on trace element transfer are particularly concerning in contaminated soils, although glyphosate has a relatively limited additional effect compared to the initial pool of mobile elements (Bemelmans et al., 2023). Furthermore, organic compounds can affect phytohormones, which also play a role in trace element uptake (Dai et al., 2023; Kumar et al., 2024). As this research topic lies at the interface between organic and mineral soil contamination, it has been underexplored, resulting in a knowledge gap. We hypothesise that the application of glyphosate-based pesticides to soil may increase the mobility of trace elements in both soil solution and plant, but this effect should be weaker than those observed in the laboratory with concentrations higher than the doses allowed in the field.

In this study, we aimed to: (1) assess the influence of pre-sowing glyphosate application on the transfer of trace elements from soil to soil solution and plant; (2) compare the behaviour in uncontaminated and contaminated soils; and (3) evaluate the potential of exogenous organic matter to mitigate the glyphosate effect on trace element transfer. We conducted a greenhouse experiment in which a glyphosate-based pesticide was sprayed on four contrasting soils (varying in both contamination levels and types) at the maximum and 3 times the maximum dose authorised in Belgium.

2. Materials and methods

2.1. Study area

Four agricultural soils from Wallonia, Belgium, were considered in this study to include a trace element contamination gradient as described by Bemelmans et al. (2023): (1) an uncontaminated soil from Vieusart (50.6799°N, 4.6357°E; loamy soil, pH_{H_2O} = 6.5, CEC = 11.1 $cmol_c\ kg^{-1}$, organic carbon content C_{org} = 1.25%); (2) a moderately anthropogenically contaminated soil from Sainte-Walburge (50.6644°N, 5.5887°E; loamy soil, pH_{H_2O} = 7.8, CEC = 15.2 $cmol_c\ kg^{-1}$, C_{org} = 11.1%);

(3) a highly anthropogenically contaminated soil from Bressoux (50.6374°N, 5.6109°E; loamy soil, pH_{H2O} = 7.0, CEC = 26.9 $cmol_c$ kg^{-1}, C_{org} = 15.8%); and (4) a naturally-enriched soil from Aubange (49.5765°N, 5.7684°E; sandy clay loamy soil, pH_{H2O} = 7.3, CEC = 20.8 $cmol_c$ kg^{-1}, C_{org} = 2.43%). Briefly, the soils of Sainte-Walburge and Bressoux (region of Liège) were significantly impacted by industrial activities, leading to atmospheric deposition and coal-extraction debris inputs that caused trace element contamination. The Aubange soil was developed on sedimentary rocks with high natural content of trace elements (e.g., As, Ni, and Cr).

2.2. Experimental procedure

On each study site, soil from the top 30 cm was collected with a shovel and stored in 30-L plastic boxes. Soil samples were air dried at 25 °C for 15 days and sieved through a 12-mm mesh. After homogenising, the sieved soil material was slightly rehydrated before being placed in 10-L pots for the experiment. Two weeks after glyphosate application (*Glyfall Plus*® at 360 g L^{-1}, a commercial glyphosate-based pesticide from *Bayer Cropscience*), nine spikelets of spelt (*Triticum spelta*) per pot have been sown to mimic a field seeding density. In each pot, we added 20 g of NPK 9−7−14 fertiliser (i.e., 240 kg_N ha^{-1}) to ensure correct plant nutrition. The experiment was conducted in a greenhouse over a period of 93 days (from 28 July 2022 to 23 November 2022). Environmental conditions during the experiment were 17.6 to 36.1 °C for air temperature and 25 to 99% for air humidity.

Four modalities were considered in the experiment: (1) a control with no glyphosate application; (2) a single-dose application of glyphosate (3.6 kg ha^{-1}, i.e., the maximum annual dose authorised in Belgium; Phytoweb, 2015; “Max”) representing a realistic situation; (3) a triple-dose application of glyphosate (“3Max”) to study the effect of a contrasted dose in case of low glyphosate effect;

and (4) a triple-dose application of glyphosate with an addition of 245 g of green waste compost (vegetation debris <15 mm, such as grass, dead leaves and branches, from the Basse-Wavre composting platform, Walloon Brabant) mixed in the soil (i.e., 35 t ha^{-1}, "3Max&OM") to evaluate potential influence of exogenous organic matter on trace element mobility. By comparing glyphosate application modalities to the control, we considered any potential glyphosate residues present in the soil material despite no glyphosate was detected in the control modality. Note that glyphosate application is not toxic for plants when it is already present in the soil due to its foliar action. For glyphosate applications, we manually sprayed 2.5 mL (Max) or 7.5 mL (3Max and 3Max&OM) of a 10 g L^{-1} glyphosate solution using a hand sprayer.

Trace elements water extractability in soils and compost have been evaluated by agitating 1 g of 2 mm-sieved soil with 40 mL of milli-Q water for 16 h. The solutions were analysed before filtering at 0.2 µm.

2.3. Sampling procedure

During the experiment, 48 soil solution samples (one per pot) have been collected 26 days after glyphosate spraying each in 50-mL syringes with rhizon samplers (10-cm long hydrophilic polyether sulfone membrane with a porosity of 0.15 µm; 19.21.21 F, Rhizosphere, Wageningen, The Netherlands). For trace element concentrations, an aliquot of every solution was acidified (2% HNO_3) and stored in polypropylene tubes prior to ICP-MS analysis. For glyphosate concentrations, a second aliquot was stored at 4 °C (maximum 7 days) before HPLC analysis.

At the end of the experiment, plant samples were collected by isolating both aerial parts (i.e., by cutting at the collar) and root parts (i.e., by removing soil particles and cleaning thoroughly using a soft brush and demineralized water). Plant samples were oven-dried at 50 °C and then weighted and ground (Cyclotec 1093 Sample Mill, Foss A/S, Hillerød, Danemark).

2.4. Chemical analyses

Concentrations of 15 trace elements were measured by ICP-MS (iCAP Q ICP–MS, Thermo Fischer Scientific, Waltham, MA, USA) at the Earth and Life Institute analytical platform (MOCA, UCLouvain, Belgium). The SLRS-6 reference material (river water) was used for quality check; recoveries reached 100 ± 20% for all elements, except for Sb (124%), Zn (131%), and Cd and Sn that were >150% (only relative concentrations between modalities were considered). Limits of detection for soil solutions were between 0.001 and 0.01 µg kg^{-1}, except for Ti, Ni and Zn (between 0.01 and 0.1 µg kg^{-1}). Trace element concentrations were determined in both soil and plant (i.e., root and shoot) samples by ICP-MS after a $HF/HNO_3/HCl/H_2O_2$ digestion procedure in a clean room (Arbalestrie et al., 2022). Reference materials (BHVO-2 and IAEA-336) have been used for quality check of mineral and organic matrices, respectively. Recoveries reached 100 ± 20% for all certified elements, except for Sn (149%) in the BHVO-2, and Ba, Cr, Zn (80–70%), Cd, Cu, Pb (70–60%), and Mo (47%) in the IAEA-336. Limits of detection for solid samples were evaluated to <1 ng kg^{-1}, except for Cr, Cu, Mn, Pb, and V (<0.01 µg kg^{-1}), for Ba and Zn (<0.1 µg kg^{-1}), and for Ti (<1 µg kg^{-1}).

Glyphosate and AMPA were analysed in soil solution samples following the Bemelmans et al. (2023) procedure (adapted from ISO 21458 and ISO 116308). Briefly, both molecules were derivatized at pH 9 (borate buffer) by FMOC-Cl in excess before analysis by HPLC (1200 series, Agilent Technologies Santa Clara, CA, USA) using phosphate buffer/acetonitrile:methanol:Milli-Q water as elutant solutions.

2.5. Data treatment

Data were analysed using *R 4.0.2*. Results included triplicates, they were compared to the control (i.e., without glyphosate application) and expressed as tested modality/control concentration ratios for soil solution samples and tested modality/control content ratios for plant samples. Welsh t-tests were performed to evaluate the difference between results for the glyphosate application modalities and controls.

3. Results and discussion

3.1. Influence of glyphosate on trace element transfer to soil solution

In this section, we will consider indifferently the influence of the glyphosate molecule and the glyphosate-based pesticide. As the experiment was based on formulated glyphosate, one should point out that results can over- or under-estimate the actual influence of glyphosate alone, since anther formulation may provide different results (Bemelmans et al., 2023). We first focused on four contrasted elements: Cu as the best documented element to interact with glyphosate; Pb as a well-known toxic element frequently found in Belgian contaminated soils; and As and Mo as oxyanions in natural Eh-pH soil conditions (Figure 1). In the uncontaminated soil (i.e., Vieusart), the soil solutions collected 26 days after glyphosate spraying presented higher concentrations for the four selected trace elements in the Max modality than in the control: from 1.3 to 12 times, $p < 0.1$ for two of the eight cases (elements/glyphosate modalities, compost excluded). This has already been reported for few elements (e.g., Cu and Pb) under controlled conditions where glyphosate highlighted metal desorption, resulting in an increased leaching (Barrett and McBride, 2006; Divisekara et al., 2018). When considering the 15 elements studied, we observed that 80%

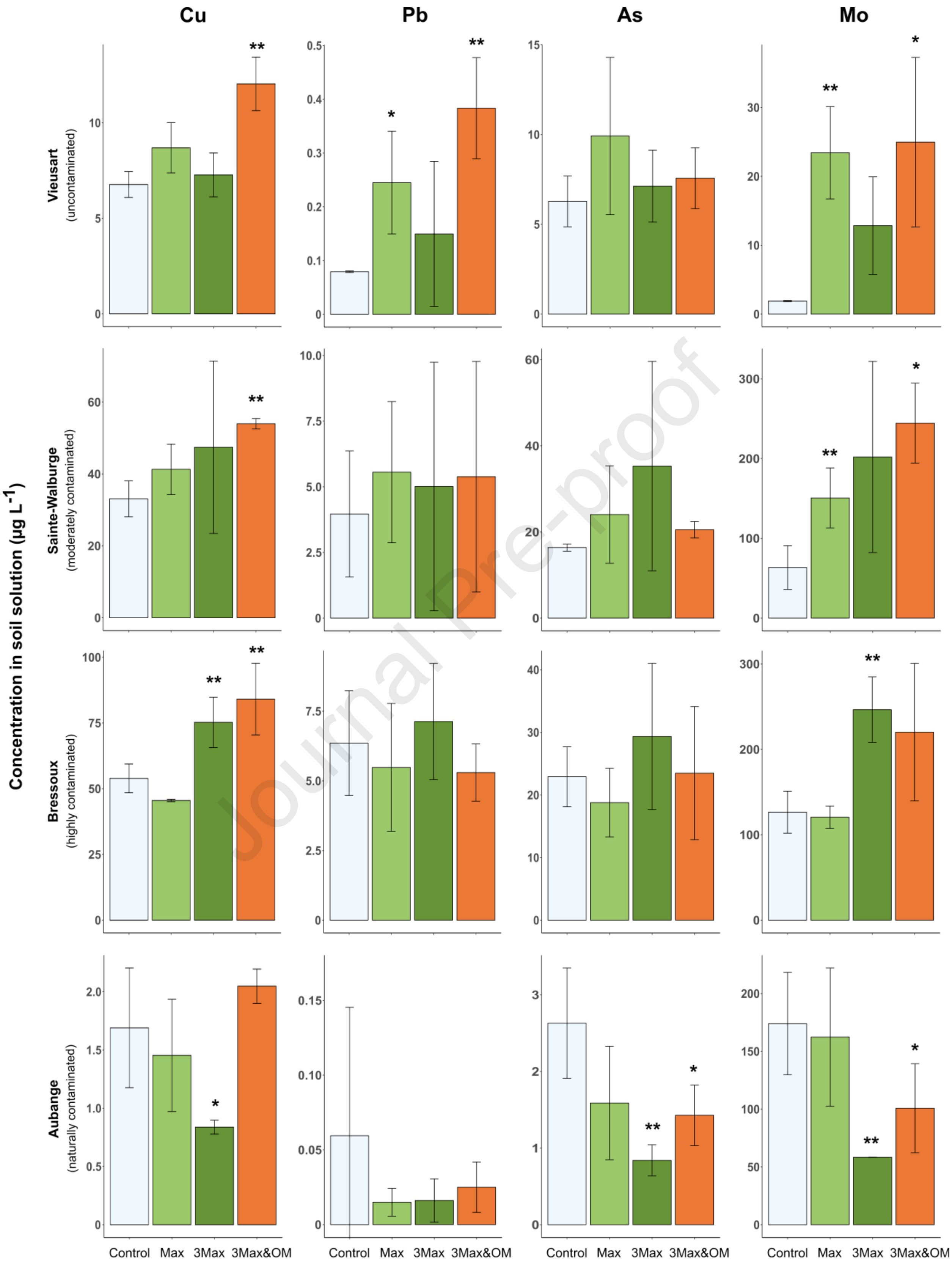
of these elements showed a mean Max/control concentration ratio >1.2 (Figure 2). This trend confirms the findings made in extractability experiment using the same soils (Bemelmans et al., 2023), supporting that the enhancement effect of glyphosate still occurs in more complex conditions using field-like concentrations and natural percolation throughout the soil. This suggests that in real conditions, glyphosate may enhance exposure to trace elements through leaching into natural waters.

When the pesticide was applied at the highest dose (i.e., 3Max), no difference was observed for As and Cu concentrations in soil solutions compared to the control, unlike Mo and, to a lesser extent, Pb with higher concentrations (up to 7 times, even if no statistically significant differences were observed; Figure 1). This contrasting result for chemically similar elements (Pb and Cu as cations and Mo and As as oxyanions; Hooda, 2010) indicates that the charge is not the only factor that drives the element behaviour. Overall, higher glyphosate concentrations (i.e., 3Max compared to Max) have less influence on trace element mobility (73% of the studied elements; Figure 1 and Figure 2), resulting in an absence of significant differences compared to the control for the 3Max modality (against 5 with $p < 0.1$ for the Max modality), as already observed in Bemelmans et al. (2023). The hypothesis is that, although chelation and complex formation can enhance trace element solubility, glyphosate may be adsorbed onto soil particles due to its multiple charges (i.e., one positive and three negative; Sprankle et al., 1975), promoting the fixation of cationic elements on the soil. The formation of ternary complexes involving glyphosate, trace elements, and soil particles has already been documented, but primarily in the context of enhancing glyphosate sorption through cationic bridge formation (Morillo et al., 2000; Wang et al., 2006). Glyphosate degradation over time leads to the AMPA formation, resulting to a reduced influence on trace element mobility (Bemelmans et al., 2023).

Similar to Vieusart, the anthropogenically contaminated soils exhibited an increased influence of glyphosate on the trace element mobility, though the effect was weaker with only three of the 16 cases (elements/glyphosate modalities, compost excluded) with $p < 0.1$ (Cu, Pb, As, and Mo; Figure 1 and Figure 2). Indeed, in the moderately contaminated soil (i.e., Sainte-Walburge), 73% of the elements studied were more concentrated in soil solutions in the Max modality than in the control with ratios >1.2 (and up to 3.0). Nonetheless, the average of concentration ratios in soil solutions was much lower, reaching 1.6 in Sainte-Walburge vs 3.2 in Vieusart. The highly contaminated soil (i.e., Bressoux), however, presented a lower influence, including negative effect: >1.2-times lower for 6 elements and >1.2-times higher for only 1 element (Figure 2). The effect decreased with the increasing anthropogenic contamination level, resulting from the higher pool of mobile elements already present on deposited anthropogenic particles in contaminated soils (Bemelmans et al., 2023). Thus, the relative contribution of glyphosate remained minor, even in conditions more realistic than extraction experiments. Note that the opposite trend was also observed for Cu in the literature (Barrett and McBride, 2006), highlighting the influence of the nature of soil considered. In contrast to Vieusart, higher pesticide dose (3Max) had no influence (Sainte-Walburge) or increased the glyphosate effect (Bressoux; on average, 1.7-times higher) on the potential leaching of trace elements in anthropogenically contaminated soils.

A stronger negative effect was observed in the naturally contaminated soil (i.e., Aubange) where statistically differences ($p < 0.05$) were observed for three of the eight cases (element/glyphosate modality, compost excluded; Figure 1). Furthermore, eight elements showed the Max modality >1.2-times lower compared to the control (up to 4.2 times) vs only three elements >1.2-times higher (up to 1.9 times; Figure 2). It is noteworthy that Aubange presented specific properties: (1) trace element less mobile than for the anthropogenically contaminated soils because there are included in the mineral matrix (Vandeuren et al., 2023); and (2) high iron oxide content promoting the sorption of trace elements alone (Suda and Makino, 2016) or glyphosate-trace element complexes onto iron oxides, limiting the trace element mobility. This negative effect was even stronger as the applied dose of glyphosate increased (average concentration ratio of 0.48 for 3Max vs 0.88 for Max), highlighting the importance of the dose considered.

In this study, only sprayed volume of similar glyphosate concentration changed between modalities Max and 3Max. Glyphosate concentrations measured in soil solutions decreased rapidly with time, resulting in concentrations mostly below the limit of detection (<0.67 µg L^{-1}). Indeed, AMPA was detected in 56% of samples when glyphosate was applied (up to 5.8 µg L^{-1}), indicating that glyphosate was degraded throughout the experiment. Since AMPA concentrations were higher in the 3Max modality (up to 2.9) than in the Max modality (up to 0.67), we assume that the influence of glyphosate should be higher in the 3Max modality. Note that AMPA may be partly responsible for the observed effects since it has already been shown to increase the extractability of trace element, although with a weaker effect (Bemelmans et al., 2023). We thus demonstrate that glyphosate may influence trace element dynamics from soil to soil solution even with field-like concentrations largely lower than concentrations used in previous experimental conditions (Bemelmans et al., 2023; Divisekara et al., 2018). Note that high air temperature and humidity in the greenhouse may accelerate the pesticide degradation compared to field conditions.


Cu
Pb
As
Mo
Concentration in soil solution (µg L-1)
Vieusart
(uncontaminated)
Sainte-Walburge
(moderately contaminated)
Bressoux
(highly contaminated)
Aubange
(naturally contaminated)
Control
Max
3Max
3Max&OM

261

Figure 1. Trace element concentrations (n = 3) in soil solutions (26 days after spraying) of the uncontaminated (Vieusart) and the three contaminated (Sainte-Walburge, Bressoux, and Aubange) soils considering various modalities: without glyphosate (Control), single dose application of glyphosate (Max), triple dose application of glyphosate (3Max), and triple dose application of glyphosate with an addition of compost (3Max&OM). Error bars represent standard deviations and y-scales were adapted for each plot. * (p < 0.1) and ** (p < 0.05) represent statistically significant differences between the modalities and the control (t-test).

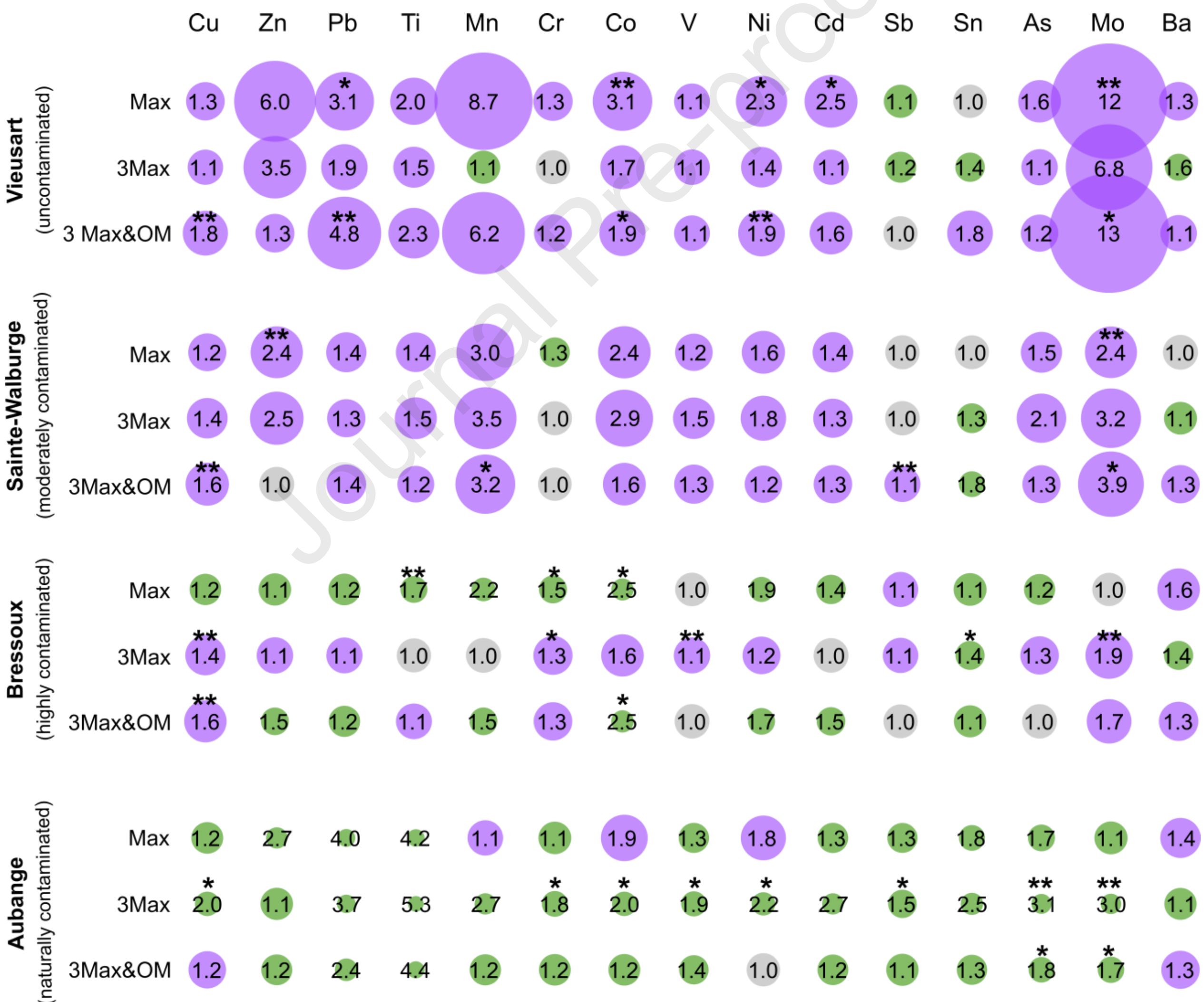


Figure 2. Summary of tested modality/control concentration ratios (n = 3) of trace elements in soil solutions of the uncontaminated (Vieusart) and the three contaminated (Sainte-Walburge, Bressoux, and Aubange) soils considering various modalities: single dose application of glyphosate (Max), triple dose application of glyphosate (3Max), and triple dose application of glyphosate with an addition of compost (3Max&OM). * ($p < 0.1$) and ** ($p < 0.05$) represent statistically significant differences between the modalities and the control (t-test).

3.2. Influence of glyphosate on trace element transfer to plant

We then studied the influence of glyphosate on trace element transfer to plant. In this section, we only considered plant shoot, as it constitutes the main agronomical interest for Poaceae. The four elements studied for soil solutions have also been selected (Figure 3): Cu, Pb, As, and Mo. Results of the uncontaminated soil (i.e., Vieusart) showed that the glyphosate application (Max modality) slightly increased the transfer of trace elements from soil to plant (from 1.4- to 2.1-times higher than in the control), despite a high heterogeneity between replicates making no significant difference ($p > 0.1$). This increasing availability for plants could result from: (1) a higher mobilisation of trace elements in soil pore water (Bruemmer et al., 1986), as illustrated through soil solutions (Figure 1); and/or (2) higher pool of trace elements in the exchangeable fraction (i.e., sorption onto soil exchange capacity without increasing concentration in soil solution). Overall, 67% of the elements studied presented a positive effect (with content ratios >1.2), including both cationic and anionic chemical forms (Figure 4). As observed for soil solutions, trace element contents in plant materials were lower in the 3Max modality than in the Max modality ($p < 0.05$ only for Cu), supporting a weaker effect on both soil solution and plant available fraction, and thus the possibility of a non-monotonic effect of pesticides on trace element mobility as a function of concentration.

When increasing the soil contamination level (Vieusart < Sainte-Walburge < Bressoux), we observed a lesser or even negative effect (average Max/control content ratios from 1.7 to 0.8; Figure 3 and Figure 4). When applying the highest dose (i.e., 3Max), results showed a decrease in content ratios, limiting positive effects in uncontaminated soil and amplifying negative effects in highly contaminated soils. The presence of coal in contaminated soils may promote the sorption of glyphosate-trace element complexes, thanks to the binding ability of both glyphosate and trace elements on coal (Ighalo et al., 2021; Swaine, 2013). However, some contrasting trends that occur between soil solution and plant reflect specific biological processes, such as barrier effect of the Casparian band for chemical complexes. This is particularly relevant in contaminated soils, where protective mechanisms are activated to prevent metal saturation in plant (Antoniadis et al., 2017; Mishra et al., 2020). In addition, this discrepancy may result from the non-representativeness of the soil solution as a bioavailable fraction to plant, but rather an indicator of possible subsequent groundwater contamination (Arbalestrie et al., 2022). Finally, most elements present in the naturally contaminated soil (i.e., Aubange) evidenced a positive effect of glyphosate on plant contents whatever the dose applied (on average, Max/control content ratio of 1.3 and 3Max/control of 1.5), including 47 and 67% of trace elements considered, respectively. Note that the heterogeneity between replicates were still important. Only Cd, Mn, Sb, and Zn were not influenced by glyphosate whatever the dose applied, likely due to their high mobility (e.g., Cd and Zn) that already promotes their leaching.

Contrary to exchanges at the soil-soil solution interface, plant uptake involves slower chemical processes that lead to lower heterogeneity between modalities in plant than in soil solution: plant content therefore constitutes an integrative compartment unlike soil solution. Indeed, element dynamics in the rhizosphere are largely dependent on root and microbial activity (including exudation and siderophores) occurring on a small scale (Neumann and Römheld, 1999), such as root acidification by proton extrusion. As low-molecular-weight organic compounds, glyphosate can form stable complexes in solution that could increase plant uptake (such as EDTA, citric acid, oxalic acid, or acetic acid; Ebrahimian and Bybordi, 2014; Qiao et al., 2020; Schmidt, 2003; Vassil et al., 1998). Yet, under a pH of 5.6, glyphosate loses one of its negative charges potentially decreasing its ability to interact with cations (Sprankle et al., 1975), which could explain the trend observed. These local acidic conditions were probably less pronounced in the naturally-enriched soil, which had a higher pH. These complexes can thus enter the plant through imperfections in the Casparian band (Schaider et al., 2006). Note that the plant uptake of glyphosate can also lead to metabolism disruption or flux saturation, resulting in variable trace element contents in plant (Degryse et al., 2009; Prasad et al., 2006; Schmidt, 2003). The complexity of those soil–plant transfer can explain some contradictory trends between plant and soil solution, as in Aubange.

In Europe, only two of the trace elements have a legally controlled content in cereals Pb and Cd with a maximum authorised content of 0.2 mg kg^{-1} of fresh material (EU, 2024). Considering ca. 80% of water content by mass for Poaceae (Glenn, 1987), shoot contents (no data for grains) exceeded in Bressoux (Cd and Pb, control modality) and Sainte-Walburge (Cd, Max modality). Note that these results were based on a single application although several pesticides are applied in several doses and a variety of pesticides is applied through the cultural period (Yang et al., 2024).

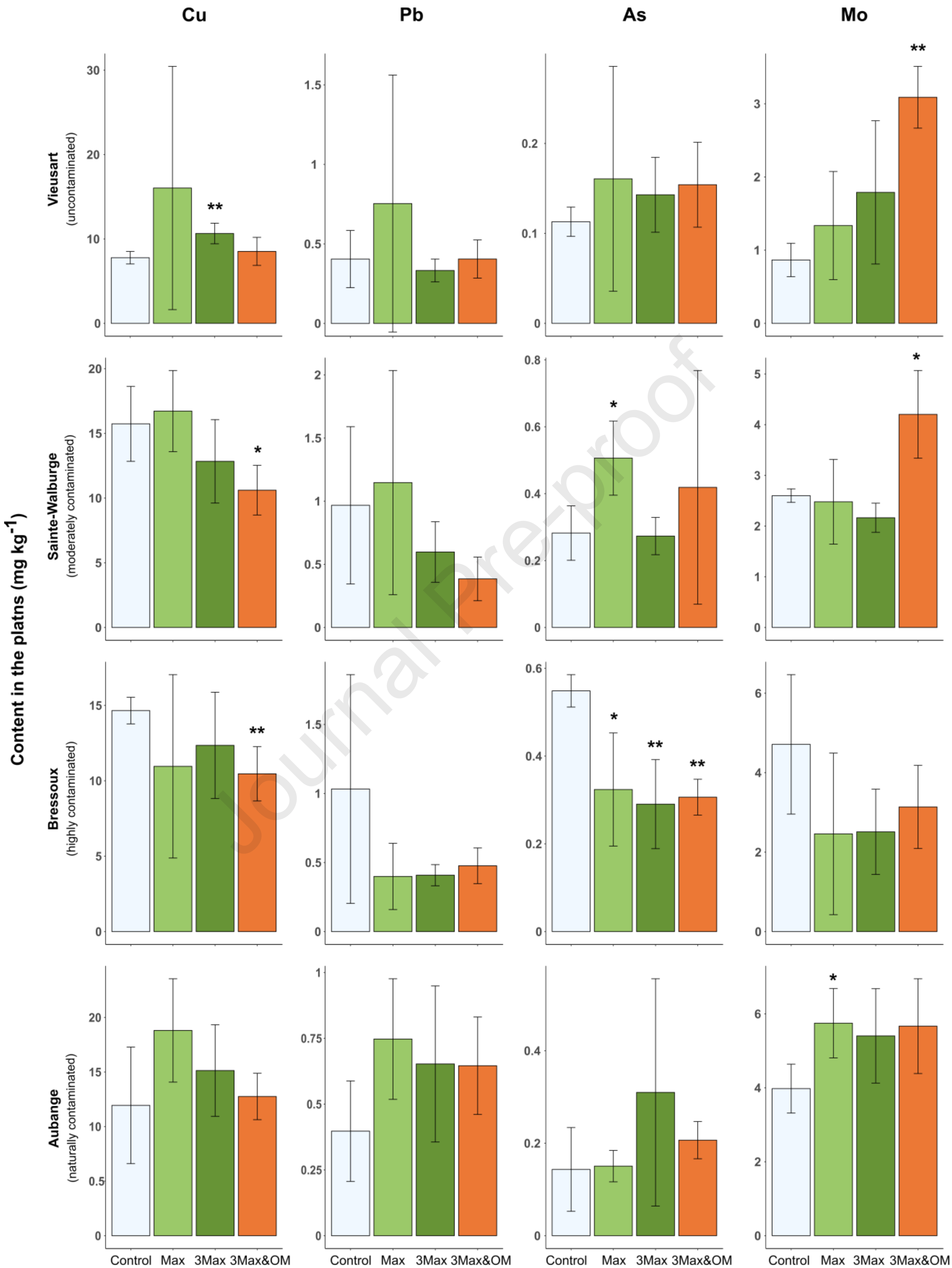

Cu
Pb
As
Mo
Content in the platns (mg kg-1)
Vieusart
(uncontaminated)
Sainte-Walburge
(moderately contaminated)
Bressoux
(highly contaminated)
Aubange
(naturally contaminated)
Control
Max
3Max
3Max&OM
*
**

Figure 3. Trace element contents (n = 3) in the plants (dry matter of shoot of spelt) that grown in the uncontaminated (Vieusart) and the three contaminated (Sainte-Walburge, Bressoux, and Aubange) soils considering various modalities: without glyphosate (Control), single dose application of glyphosate (Max), triple dose application of glyphosate (3Max), and triple dose application of glyphosate with an addition of compost (3Max&OM). Error bars represent standard deviations and y-scales were adapted for each plot. * ($p < 0.1$) and ** ($p < 0.05$) represent statistically significant differences between the modalities and the control (t-test).

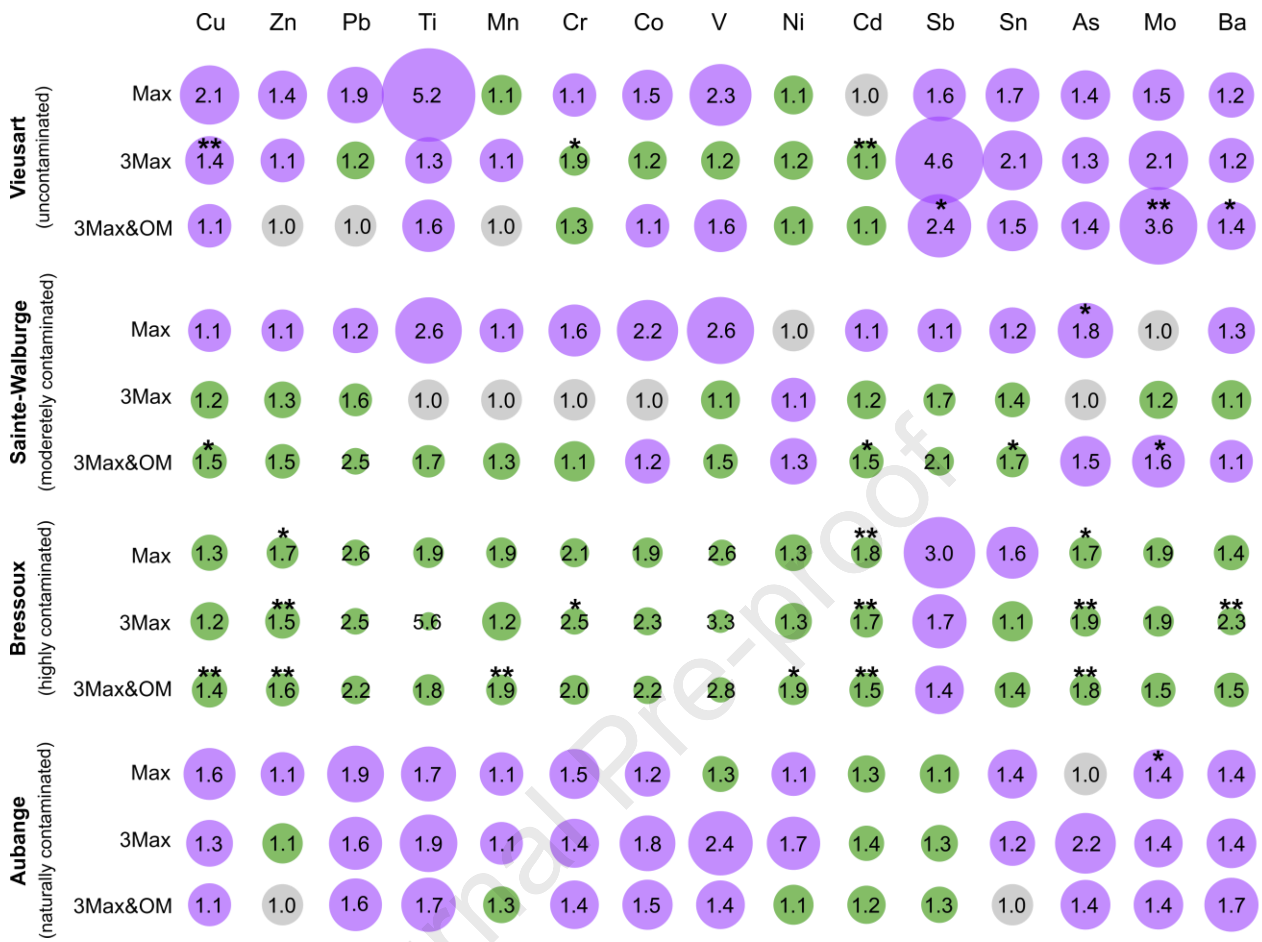


Figure 4. Summary of tested modality/control content ratios (n = 3) of trace elements in plants (dry matter of shoot of spelt) of the uncontaminated (Vieusart) and the three contaminated (Sainte-Walburge, Bressoux, and Aubange) soils considering various modalities: single dose application of glyphosate (Max), triple dose application of glyphosate (3Max), and triple dose application of glyphosate with an addition of compost (3Max&OM). * ($p < 0.1$) and ** ($p < 0.05$) represent statistically significant differences between the modalities and the control (t-test).

3.3. Role of organic matter in controlling the glyphosate influence

We finally tested the influence of exogenous organic matter (i.e., compost) input on the glyphosate effect previously evidenced on soil–water and soil–plant transfers. Considering both soils with low trace element mobility (i.e., the uncontaminated and the naturally-enriched soils), we observed overall higher concentrations in the soil solutions with compost addition (Figure 1 and Figure 2): on average, 3Max&OM/3Max content ratios of 1.8 and 1.7 for Vieusart and Aubange, respectively, including 73 and 93% of trace elements considered for which the ratios were >1.2, respectively. This was mainly observed for Mn (up to 6.5 in Vieusart), Pb, Sn, Cu, Cd, and Ni, including elements particularly toxic. Zinc, however, showed an opposite pattern with a content 2.7-times lower with compost addition, likely resulting from sorption by organic compounds. These observations may result from either (1) an additional pool of trace elements released by the compost itself or (2) soil elements mobilised by low molecular weight organic compounds from the compost via chelating properties (Ahmad and Prasad, 2012; Vaughan et al., 1993). Considering the water-extractability data, we estimated the compost contribution of the exchangeable pool between 0.2% (for Sn) and 42% (Mo) in Vieusart (<15% for 73% of the elements). In Aubange, only two elements presented a relative contribution >15% (Zn and Mo with 24 and 27%, respectively), while in the anthropogenically contaminated soils, the contribution was always <12%. We thus hypothesize that the exogenous organic matter can promote the trace element mobilisation, as for most elements their increased concentration is higher than its input. However, the transfer from soil to plant showed more divergent results (Figure 3 and Figure 4). Indeed, six elements presented 3Max&OM/3Max content ratios >1.2 (V, Mo, Cr, Co, Ti, and Pb) in Vieusart, while Sb, Sn, and Cu were less concentrated with compost addition (>1.2-times lower). Note that only negative effect was observed for five trace elements in the naturally-enriched soil (Ni, V, As, Mn, and Co). We thus consider that the compost can both adsorb or mobilise trace elements. Besides, a possible interaction may exist between organic matter and pesticides: an increased sorption of glyphosate in presence of organic matter could also favour sorption of trace elements-glyphosate complexes (Albers et al., 2009; Erban et al., 2018; Ololade et al., 2014). Since some results were inconsistent between plant and soil solution, we assume that complexes formed by mobile organic compounds are not always bioavailable for plant.

In the anthropogenically contaminated soils (i.e., Sainte-Walburge and Bressoux), elements were mainly less concentrated in the soil solution with compost addition (between 40 and 47% of elements with a negative effect, respectively, vs only 13% with a positive effect; Figure 1 and Figure 2). Cobalt, Zn, Ni, and As showed negative effect for both soils, up to 3.9-times lower for Co in Bressoux. Results showed contrasted trends for transfer to plant, including 47% of negative effect in Sainte-Walburge (vs 13% of positive effect), while Bressoux presented 27% for both positive and negative effects (Figure 3 and Figure 4). Compared to the uncontaminated soil, the addition of organic matter may partially decrease the trace element mobility in the anthropogenically contaminated soil by promoting the sorption effect over their release, which can to a certain extent counteract the effect of glyphosate in the soil solution.

3.4. Environmental fate of trace elements after a glyphosate application

Using a greenhouse experiment, glyphosate application has shown to influence the transfer of trace elements from soil to soil solution and plant. Here, we compare the two transfer pathways to assess their possible environmental fate and consequences, both in terms of hydrosystem and food contamination. We thus jointly considered the element concentration ratios between the tested modalities and the control in both soil solution (solubility) and plant (bioavailability; Figure 5). For the uncontaminated soil (i.e., Vieusart; Figure 5a), 53% of data points (glyphosate application, whatever the application dose) were in the top right corner, indicating a positive effect on the transfer to both soil solution and plant. The higher solubility may result in a greater element pool in the exchangeable fraction, and thus a higher bioavailability. The main direct risk in uncontaminated soils therefore lies in increased transfer to the plant, although the initial trace element contents are lower. This is especially concerning as plants are especially critical for human ingestion of trace elements (Antoniadis et al., 2017; Natasha et al., 2022). Indeed, contamination of hydrosystem requires crossing the barrier of deeper soil horizons before expecting to affect human health (sampling only performed on the top 10 cm of the soil).

With the increasing anthropogenic contamination level (i.e., Vieusart < Sainte-Walburge < Bressoux), however, we first observed a general decrease of the bioavailability influence, leading to antagonistic effects depending on the contamination status considered (negative effect in contaminated soil versus positive effect in uncontaminated soil; Figure 5b-c). Since the evolution on the bioavailability axis was more sensitive than that on the solubility axis, we assume that the risk will mainly be observed in natural waters. At high level of contamination, however, the effects on the soil solution are weaker, ultimately reducing the transfer of trace elements into the environment at the expense of their retention in the soil. Conversely, the behaviour observed in the naturally-enriched soil (i.e., Aubange) indicated a different trend for most elements, with decreasing the solubility effect (up to negative effect) while bioavailability remained unchanged compared to the uncontaminated soil (Figure 5d). This likely results from the strong affinity of the complexes formed with glyphosate and the mineral constituents of the soil, including iron oxides for this specific soil (Maqueda et al., 2002), limiting their solubility without impacting the more efficient root uptake (specific environmental pH and redox condition in the rhizosphere). This results in the same environmental risk as described in the uncontaminated soil without hydrosystem contamination due to higher soil retention. For both plant and soil solution, the effect of glyphosate on the element mobility, is important and mainly depends on the soil considered (including the contamination type), while the geochemistry of each individual element remains a relatively minor influencing factor.

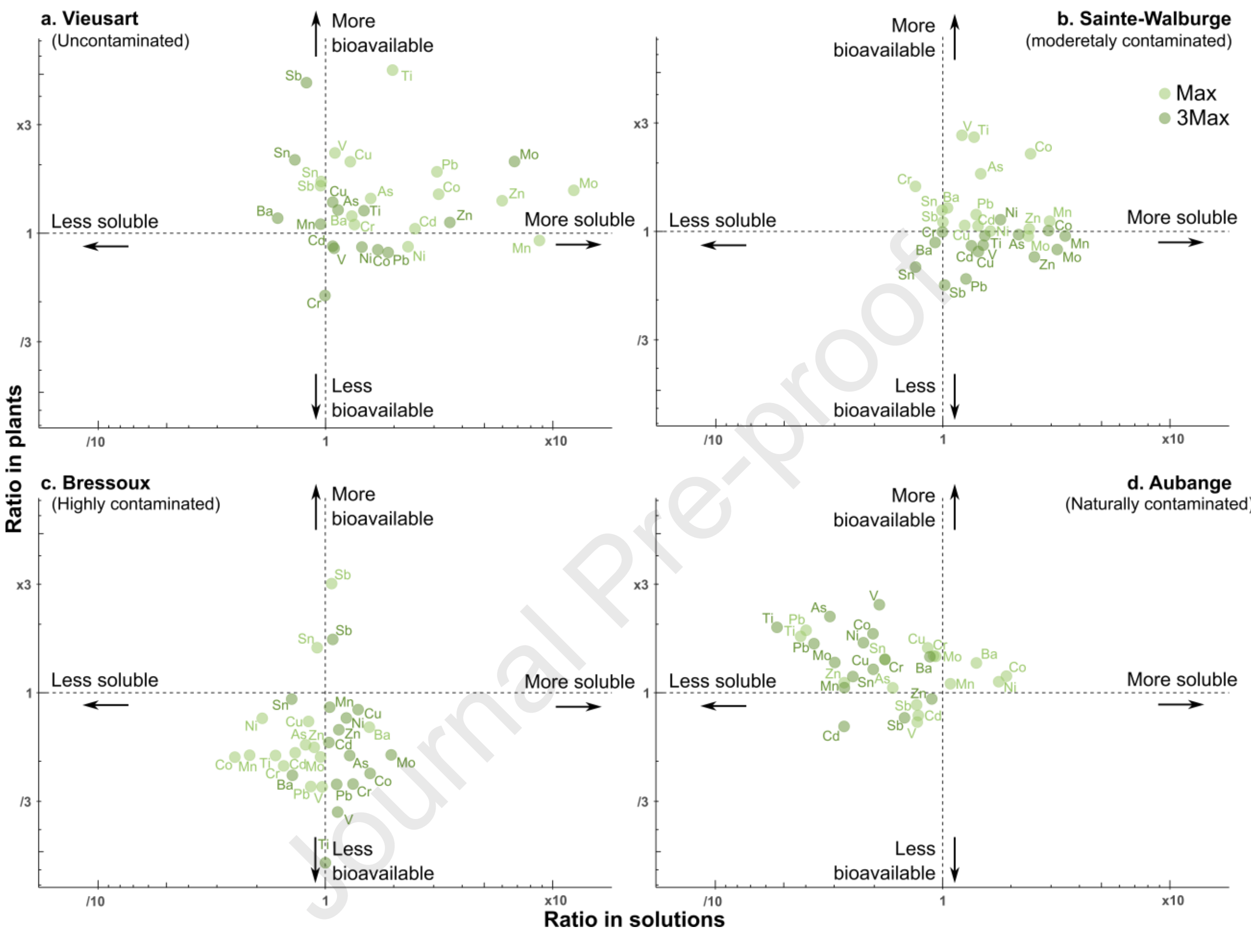


Figure 5. Influence of glyphosate application on the environmental transfer of 15 trace elements considered (i.e., tested modality/control concentration ratios in soil solutions and tested modality/control content ratios in plants) in the uncontaminated (Vieusart) and the three contaminated (Sainte-Walburge, Bressoux, and Aubange) soils considering various modalities: single dose application of glyphosate (Max, light green) and triple dose application of glyphosate (3Max, dark green).

4. Conclusions

Trace elements are a significant concern in agricultural soils worldwide, and given the widespread use of pesticides, their role could be crucial in the dynamics of trace elements in the soil–plant system and human exposure. Firstly, this study demonstrated that glyphosate-based pesticides could increase trace elements transfer in uncontaminated soil and decrease it in highly anthropogenically contaminated soils, likely due to the initially higher pool of mobile elements. Pesticide use could thus influence human exposure to harmful trace elements, but specifically in uncontaminated environments. Secondly, distinct trends were generally observed between soil solution and associated plant, particularly in contaminated environments. This requires all possible risks to be taking into account, both in foodstuffs and drinking water depending on the contamination levels. Thirdly, a non-monotonic relationship between added dose and trace element content was observed, highlighting the competition between solubilization and adsorption processes. Finally, compost addition primarily influenced soil solution concentrations and was not effective method for counteracting glyphosate-induced increases in trace element transfer in uncontaminated soils, as it tended to reinforce this effect. However, it may provide some mitigation in contaminated soils. Given the diversity of pesticides used in agriculture, there remains a knowledge gap regarding their combined effects on trace element dynamics. Also, multiple pesticides application could cumulatively affect trace element mobility and transfer to plant throughout the growing season. Due to the experimental timing, we did not analyse the edible plant parts that constitutes the main organs to consider for a risk assessment. We thus recommend continuing research in this topic, but under more realistic conditions to better assess the associated risks.

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Data availability

All the relevant data of this manuscript has been shared through the electronic supplementary materials.

Conflict of interest

The authors declare that they have no conflict of interest.

Ethical approval

This article does not contain any studies with human or animal subjects performed by any of the authors.

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Highlights

- Application of glyphosate-based pesticide modifies trace element transfer.
- Soil contamination level is a key driver for the glyphosate influence.
- Glyphosate application could increase trace element exposure through plant uptake.
- Compost could limit trace element transfer in contaminated soils.
- Compost, however, increased trace element transfer in uncontaminated soils.

Declaration of interests

☒ 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.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: