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# POTENTIALLY TOXIC ELEMENTS IN FERTILIZERS: FROM A GLOBAL META-ANALYSIS TO A LOCAL CHARACTERIZATION IN THE CERRADO BIOME

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# POTENTIALLY TOXIC ELEMENTS IN FERTILIZERS: FROM A GLOBAL META-ANALYSIS TO A LOCAL CHARACTERIZATION IN THE CERRADO BIOME

Dissertação apresentada, como parte das exigências para obtenção do título de MESTRE EM AGROQUÍMICA, ao Programa de Pós-Graduação em Agroquímica do Instituto Federal de Educação, Ciência e Tecnologia Goiano – Campus Rio Verde – Área de concentração: Agroquímica Ambiental.

Orientador: Prof. Dr. Dener Márcio da Silva Oliveira
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Com todo amor do mundo, dedico ao meu sobrinho Miguel (Guelzinho).

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*"O segredo da vida é o solo, porque do solo dependem as plantas, a água, o clima e a nossa vida. Tudo está interligado. Não existe ser humano sadio se o solo não for sadio."*

(Ana Maria Primavesi)

**RESUMO GERAL**

A poluição do solo por elementos potencialmente tóxicos (EPTs) decorrente do uso inadequado de fertilizantes e corretivos ameaça a segurança alimentar e a sustentabilidade agrícola, especialmente em áreas com alta demanda de fertilizantes, como o Cerrado Brasileiro. Esse estudo combinou uma metanálise global, que sintetizou dados de 45 artigos usando abordagem de quociente de risco para organismos do solo, com uma análise regional da presença de EPTs em fertilizantes comercializados no Cerrado. As concentrações de EPTs (As, Co, Cd, Cr, Cu, Fe, Hg, Mn, Mo, Ni, Pb, Se, Zn) foram determinados pelos métodos USEPA 3051A (teor total) e solúvel em água (teor prontamente disponível), com quantificação por ICP-OES. Os resultados de ambas as abordagens projetaram riscos à biota edáfica associados principalmente aos fertilizantes orgânicos e organominerais, sobretudo devido às altas doses de aplicação. Na metanálise, foi observado valores que ultrapassaram os limites legais de EPTs para a rocha fosfática (Cd), esterco animal (Cd, Cu, Cr e Zn) e composto orgânico (Cd e As). Por sua vez, na abordagem regional, excederam os limites o superfosfato simples (As) e o FTE-BR12 (Pb). Ambos os trabalhos convergiram para a necessidade de regulamentar metais ainda não monitorados, como Mo, Zn e Cu, em fertilizantes orgânicos, além de estabelecer uma legislação ambiental baseada em ensaios ecotoxicológicos, visando à proteção da qualidade e da biota do solo.

**Palavras-chave:** Agricultura tropical; Fertilizante químico; Legislação ambiental; Metais-traço; Resíduos pecuários.

**GENERAL ABSTRACT**

Soil pollution by potentially toxic elements (PTEs) arising from the improper use of fertilizers and amendments, threatens food security and agricultural sustainability, especially in regions with high input demand, such as the Brazilian Cerrado. This study combined a global meta-analysis, which synthesized data from 45 articles using a risk quotient approach for soil organisms, with a regional assessment of PTE occurrence in fertilizers commercialized in the Cerrado. PTE concentrations (As, Co, Cd, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Se, and Zn) were determined by USEPA 3051A (total extraction) and water-soluble methods, quantified by ICP-OES. The results from both approaches projected risks to soil biota primarily associated with organic and organomineral fertilizers, mainly due to their high application rates. In the meta-analysis, legal limits were exceeded for phosphate rock (Cd), animal manure (Cd, Cu, Cr, and Zn), and organic compost (Cd and As). In the regional assessment, exceedances occurred for single superphosphate (As) and FTE-BR12 (Pb). Both studies converged to the need to regulate currently unmonitored metals, such as Mo, Zn, and Cu, in organic fertilizers, as well as to establish environmental legislation grounded in ecotoxicological assays to protect soil quality and biota.

**Keywords:** Chemical Fertilizer. Environmental legislation. Livestock waste. Trace metals. Tropical agriculture.

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## LIST OF ABBREVIATIONS AND ACRONYMS

| | |
|---|---|
| ACP / PCA | Análise de Componentes Principais (Principal Component Analysis) |
| CEC / CTC | Capacidade de Troca Catiônica (Cation Exchange Capacity) |
| EPTs / PTEs | Elementos Potencialmente Tóxicos (Potentially Toxic Elements) |
| HC5 | Concentração Perigosa para 5% das Espécies (Hazardous Concentration for 5% of the species) |
| ICP-OES | Espectroscopia de Emissão Óptica com Plasma Indutivamente Acoplado (Inductively Coupled Plasma Optical Emission Spectrometry) |
| NOEC | Concentração Sem Efeito Observado (No Observed Effect Concentration) |
| OM / MO | Matéria Orgânica (Organic Matter) |
| PEC | Concentração Ambiental Prevista (Predicted Environmental Concentration) |
| PN | Poder de Neutralização |
| PNEC | Concentração Prevista Sem Efeito (Predicted No-Effect Concentration) |
| PRISMA | Preferred Reporting Items for Systematic Reviews and Meta-Analyses (modelo de fluxo para seleção de artigos) |
| PRNT | Poder Relativo de Neutralização Total |
| RQ | Quociente de Risco (Risk Quotient) |
| SSD | Distribuição de Sensibilidade das Espécies (Species Sensitivity Distribution) |
| VBP / GPV | Valor Bruto da Produção Agropecuária (Gross Production Value) |
| FTE / MFTE | Fritted Trace Elements / Micronutrientes tipo Fritas |
| MAP / MMAP | Fosfato Monoamônico (Monoammonium Phosphate) |
| ML | Calcário (Limestone) |
| NPK / MNPK | Nitrogênio, Fósforo e Potássio (adubos minerais ternários) |
| OCM | Esterco Bovino (Cattle Manure) |
| OFC | Torta de Filtro de Cana-de-Açúcar (Sugarcane Filter Cake) |
| OMI a OMVI | Adubos Organominerais de I a VI |
| OPL | Cama de Frango / Aviária (Poultry Litter) |
| OPS | Dejeto Líquido de Suíno (Pig slurry) |
| OV | Vinhaça (Vinasse) |
| SSP / MSSP | Superfosfato Simples (Single Superphosphate) |
| TSP / MTSP | Superfosfato Triplo (Triple Superphosphate) |

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# 1 GENERAL INTRODUCTION

Projections indicate that the global population may reach 9.73 billion by 2064, declining to 8.79 billion by 2100 (Vollset et al., 2020). To meet this demand, it is estimated that global food production will need to increase by 70% by 2050, placing significant pressure on natural resources, intensifying land-use changes (Dijk, van et al., 2021), and increasing the use of agricultural inputs such as pesticides and fertilizers (Abebe et al., 2022). In this context, analyses have shown that a 1% increase in population pressure results in a 0.118% increase in fertilizer use intensity (Xiang, Malik and Nielsen, 2020).

Large-scale agriculture relies on fertilizers and soil amendments to adequately replenish lost nutrients and ensure high yields (Howe et al., 2024). This demand is particularly high in regions with acidic and nutrient-poor soils, such as the Brazilian Cerrado (Lima et al., 2022). The Cerrado alone accounts for 22% of global soybean production and 23% of sugarcane production, turning it responsible for approximately 60% of the country's agricultural output (Hurd and Silva, 2024). However, the biome is not only distinguished by its agricultural production and livestock development; it is also the most biodiverse tropical savanna in the world, both in terms of fauna and flora (Embrapa, 2020).

The improper use of fertilizers can lead to soil contamination and pollution by potentially toxic elements (PTEs) (Hou et al., 2020; Zhang et al., 2020). While contamination refers to the presence of chemical substances resulting from anthropogenic activities, i.e., at concentrations above those naturally found, pollution represents a more severe condition, in which environmental concentrations of these substances pose a risk to human health and/or biota (Ibama, 2022). It is estimated that approximately 240 million hectares of agricultural soils worldwide are contaminated with PTEs, directly affecting 0.9 to 1.4 billion people (Hou et al., 2025).

PTEs may consist of essential elements (e.g., Zn, Cu, and Cr), which play some metabolic role in organisms, or non-essential elements (e.g., As, Cd, and Cr(VI)), for which no well-defined metabolic function exists, and which can be toxic even in small amounts (Jomova et al., 2022; Balali-Mood et al., 2021). By comparison, while Zn has a reported Predicted No-Effect Concentration (PNEC) of 38.3 mg/kg, for Cd it is only 0.2 mg/kg (Wan et al., 2020; Xu et al., 2019). Although these elements may originate from natural processes such as rock weathering and volcanic eruptions, their enrichment in soils becomes more significant through anthropogenic activities, including mining, industrial operations, and agricultural practices (Jomova et al., 2025; Priya et al., 2023; Wang et al., 2023).

Recent studies have reported concentrations of As, Pb, Hg, Ni, and Cd above regulatory limits in both mineral and organic fertilizers (Sachdeva et al., 2025). In mineral fertilizers, particularly phosphate-based ones, the presence of PTEs may arise from the raw material and insufficient purification during the manufacturing process (Guilherme et al., 2020; Nziguheba and Smolders, 2008). In organic fertilizers, such as cattle, swine, and poultry manure, organic composts, biosolids, and agro-industrial residues, these elements may originate from additives used in animal feed (Arcella et al., 2019).

Soil pollution by potentially toxic elements (PTEs) directly threatens food security and environmental sustainability. Consequently, it compromises the progress of several Sustainable Development Goals (SDGs) proposed by the United Nations, including: SDG1 (No Poverty) and SDG2 (Zero Hunger and Sustainable Agriculture), by reducing agricultural productivity and income; SDG3 (Good Health and Well-being), by causing various harms, diseases, and dysfunctions due to exposure; SDG6 (Clean Water and Sanitation), due to surface runoff and leaching of toxic elements into water bodies; SDG13 (Climate Action), by reducing soil organic carbon storage potential; and SDG15 (Life on Land), by reducing soil biodiversity (Hou et al., 2020).

In view of this scenario, this dissertation seeks to investigate the presence of potentially toxic elements in fertilizers at two scales. The first chapter presents a global synthesis, through bibliometric analysis and meta-analysis, about the concentrations of these elements in fertilizers and agricultural amendments. To estimate the impact of these fertilizers on soil biota, the Risk Quotient (RQ) was calculated based on a Predicted No-Effect Concentration (PNEC). The second chapter applies this approach at the regional scale, analyzing the composition of fertilizers used in the Cerrado biome and projecting the impact of locally relevant application rates on soil biota. The ultimate goal is to generate essential information for safer and more sustainable agricultural practices, contributing to the proposal of improved public policies for the agricultural and environmental sectors.

# 2 OBJECTIVES

## *2.1 General Objective*

To evaluate the occurrence of Potentially Toxic Elements (PTEs) in fertilizers and agricultural amendments, as well as the risks associated with their application to soil, by integrating a global-scale analysis with a regional-scale study focused on the Brazilian Cerrado.

## *2.2 Specific Objectives*

- To map global research trends, scientific production, and the most relevant authors and institutions through a bibliometric review;
- To quantify, by means of a meta-analysis, the mean, minimum, and maximum concentrations of PTEs found in fertilizers and agricultural amendments worldwide;
- To estimate the impact of fertilizer use on soil biota through the Risk Quotient (RQ), based on the Predicted No-Effect Concentration (PNEC) and realistic fertilizer application rates;
- To compare the risk profile between mineral and organic sources, considering application rates and the elements most strongly correlated with the source type;
- To characterize the total and readily water-available contents of PTEs in fertilizers and agricultural amendments commercialized in the Brazilian Cerrado;
- To identify regulatory gaps and propose updates to current environmental legislation guidelines, focusing on the inclusion of new limiting elements to preserve soil quality.

# 3 CHAPTER I – POTENTIALLY TOXIC ELEMENTS IN FERTILIZERS AND ASSOCIATED RISKS FOR SOIL ORGANISMS: A BIBLIOMETRIC AND META-ANALYTICAL ASSESSMENT

RODRIGUES, J. N. M. *et al.* Potentially Toxic Elements in Fertilizers and Associated Risks for Soil Organisms: A Bibliometric and Meta-Analytical Assessment. **Land**, vol. 15, no. 8, p. 1377, 2026. DOI: https://doi.org/10.3390/land15081377. Acesso em 04 ago 2026

## RESUMO

A contaminação do solo por elementos potencialmente tóxicos (EPTs) oriundos de fertilizantes representa grande desafio para garantir a sustentabilidade agrícola e segurança alimentar global. Este trabalho conduziu uma análise bibliométrica e metanálise sobre a ocorrência de As, Cd, Cr, Cu, Pb, Zn, Ni, Co, Mn, Hg e Fe em fertilizantes e suas implicações para a biota edáfica através de uma análise de risco. Foi utilizado a *Web of Science* como banco de dados e o software R versão 4.3.2 (pacotes bibliometrix e metafor) para as análises bibliométricas (1116 documentos) e metanálise (548 artigos). Após a avaliação dos critérios de elegibilidade, 45 artigos foram incluídos na metanálise. A análise de risco foi baseada na abordagem de quociente de risco, considerando doses de aplicação altas e baixas, assim como concentrações médias e máximas dos metais. Os resultados mostraram que os limites legais foram ultrapassados pelo Cd na rocha fosfática, para Cd, Cu, Cr e Zn em estercos e para Cd e As no composto orgânico. De forma geral, fontes orgânicas apresentaram risco maior que fontes minerais, com os principais riscos associados a aplicações contínuas de médio e longo prazo. Esses achados reforçam a necessidade urgente de uma regulamentação rigorosa de contaminantes em fertilizantes e de uma legislação técnica fundamentada em dados ecotoxicológicos, visando preservar a qualidade do solo e promover a sustentabilidade agrícola.

**Palavras-chave:** Contaminantes inorgânicos; Metais tóxicos; Organismos do solo; Quociente de risco; Resíduos agrícolas.

**ABSTRACT**

Soil contamination by potentially toxic elements (PTEs) from fertilizers represents a major issue threatening agricultural sustainability and global food security. This work conducted a bibliometric analysis and a meta-analysis on the occurrence of As, Cd, Cr, Cu, Pb, Zn, Ni, Co, Mn, Hg, and Fe in fertilizers and their implications for soil biota through risk assessment. The database used was Web of Science, and R software version 4.3.2 (bibliometrix and metafor packages) was utilized to perform bibliometric analyses (1116 documents) and meta-analysis with a five-year filter (548 articles). A total of 45 articles met all the inclusion criteria for the meta-analysis. The risk assessment was based on a quotient risk approach, considering high and low fertilizer application rates, as well as the average and maximum reported metal concentrations. Legal limits were surpassed for Cd in phosphate rock; for Cd, Cu, Cr, and Zn in manure; and for Cd and As in compost. Overall, organic sources presented a higher risk than mineral ones, with major risks related to mid- to long-term applications. These findings highlight the urgent need for strict regulation of contaminants in fertilizers and technical legislation grounded on ecotoxicological data to preserve soil quality and promote agricultural sustainability.

**Keywords:** Agricultural waste. Inorganic contaminants. Risk quotient. Soil organisms. Toxic metals.

## 3.1 INTRODUCTION

Soil pollution by potentially toxic elements (PTEs) represents a major environmental issue, with 14 to 17% of the global agricultural area affected by metal pollution and up to 1.4 billion people living in high-risk areas [1]. Global evidence shows that agricultural soils can have high concentrations of metals [2,3], representing a major challenge for sustainable development and food security, especially in developing countries [4,5]. Furthermore, this scenario is likely to worsen in the coming decades due to the projected population growth to 9.4–10.1 billion by 2050 and the consequent increase in food demand [6,7] and, thus, fertilizer application [8,9].

The expression "potentially toxic element(s)" encompasses metals and metalloids that may be considered harmful to biota, depending on the level of exposure, the dose, and the receiving organism or population [10,11]. The PTEs can be classified as essential or non-essential [12]. Essential PTEs, such as zinc (Zn), copper (Cu), and iron (Fe), play a vital role in various physiological processes [13]. On the other hand, non-essential PTEs, such as arsenic (As), cadmium (Cd), and mercury (Hg), do not have known biological functions and can be toxic and carcinogenic, even at low concentrations [14].

The origin of these elements can be natural, for example, through the weathering of igneous and sedimentary rocks [15]. However, previous evidence shows that anthropogenic sources are usually more relevant to explain their environmental occurrence [16]. Major anthropogenic sources are represented by mining and metal casting [17,18], reuse of sewage sludge [19–21], chemical manufacturing [16], use of pesticides [22] and application of mineral [23] and organic fertilizers in agriculture [19,24]. Anthropogenic metal input can lead to contamination, when the metal concentration is higher than its natural occurrence, or to pollution, when the concentration is related to negative effects [25].

Mineral fertilizers, especially phosphate fertilizers, can contain impurities in the form of PTEs, such as arsenic (As), lead (Pb), mercury (Hg), nickel (Ni), and especially cadmium (Cd), caused by insufficient purification in the manufacturing process [26,27]. Organic fertilizers may also contain PTEs in their composition, depending on their origin and degree of transformation [16,28,29]. Once introduced into the soil, due to their high persistence and toxicity, PTEs can negatively affect soil quality [10,14,30], which, in turn, can harm agricultural productivity, fauna and flora, ultimately threatening human health [31,32]. Several studies evaluated the ecotoxicological risks associated with soil application of mineral and organic

fertilizers, evidencing the accumulation of metals in the soil [33], the contamination of plants and food, and human health risks [34].

Bibliometric analysis is a quantitative method for evaluating the relationships and impacts of countries, institutions, publications, and authors in a specific field, as well as determining research trends, using mathematical and statistical tools [35]. In turn, meta-analysis aims to fill gaps between traditional literature reviews and quantitative analyses, allowing quantitative comparisons based on the results of a set of studies [24]. Given the global relevance of PTEs in fertilizers and the vast amount of scientific production already available, bibliometrics and meta-analysis are useful tools to systematize and synthesize the existing knowledge.

There are bibliometric analyses exploring the development of fertilizers [36] and the application of organic amendments and biofertilizers in agriculture [37,38]. Similarly, there are meta-analyses on Cd accumulation in soils and plants through manure application [24] and on PTEs in soil amendments and fertilizers [39]. However, the present study is the first to integrate both approaches: a bibliometric analysis to identify research trends and gaps regarding the PTEs occurrence in organic and mineral fertilizers, coupled with a meta-analysis to quantify the concentrations of multiple PTEs across a range of organic and mineral fertilizers. In addition, risk assessments for soil organisms were carried out using a risk quotient (RQ) approach with simulations considering contrasting scenarios of fertilizer rates (low and high) and PTE concentrations (average and maximum).

## 3.2 MATERIALS AND METHODS

### *3.2.1. Data Collection and Processing*

The Web of Science-Core Collection database (Clarivate Analytics/Thomson Reuters) was used, where a search was applied in the field "Topic" as follows: ("heavy metals" OR "heavy metal" OR "potentially toxic elements" OR "toxic metals") AND ("contamination" OR "presence" OR "contaminated") AND ("poultry litter" OR "poultry manure" OR "chicken manure" OR "pig slurry" OR "swine manure" OR "animal waste" OR "agricultural waste" OR "mineral fertilizer" OR "organic fertilizer" OR "phosphate fertilizer" OR "organo-mineral fertilizer" OR "limestone" OR "stone meal" OR "soil remineralizer" OR "rock dust" OR "rock flour" OR "vinasse" OR "stillage" OR "filter cake") NOT ("drinking water" OR "marine" OR "aquatic" OR "animal experiment"). Articles published in English before 2025 were filtered, returning 1185 results.

For data screening, the open access web interface bibliometrix was used, known as biblioshiny, from R software, Vienna, Austria, version 4.3.2 [40,41]. The bibtex files were imported into the shiny environment, filtering the period before 2025 and the publication type "article", returning a total of 1116 results. The processes and categories for including and excluding documents are detailed in the PRISMA diagram in Figure S1.1 [42].

### *3.2.2. Bibliometric Analysis and Visualization*

Articles were imported into R software using the bibliometrix package for bibliometric analysis, using biblioshiny. Complementarily, the following packages were used: ggplot2, dplyr, extrafont, igraph, maps, mapsdata, and openxlsx [41]. The following bibliometric analyses were performed: annual article production; most productive authors, institutions, affiliations, and countries; conceptual structure analysis using multiple correspondence analysis (MCA) through keywords plus; and a thematic map from keywords plus. The author keywords and plus keywords were extracted from the database generated by Web of Science. The plus keywords are words that frequently appear in the reference titles of an article but do not appear in the article title itself.

### *3.2.3. Data Sources and Collecting for Meta-Analysis*

For meta-analysis, a five-year filter (2020–2024) was used on the 1116 articles retrieved from the bibliometric analysis, resulting in 548 articles. Articles that did not evaluate PTEs, were reviews of other studies, or did not present standard deviation or standard error were all

excluded. In addition, articles solely presenting data on PTE concentration in soil or plant tissue were excluded.

Of the 548 articles analyzed, only 45 articles fully met the meta-analysis requirements: presenting mean, standard deviation, and sample size. Standard error data were converted to standard deviation. For articles where the sample size was not described, three repetitions were adopted as a standard. Overall, 35% of the data had n imputed as three. The choice of triplicate as a standard was based on a commitment to data accuracy [43], in addition to being the most frequently reported n value among studies with non-imputed n (accounting for 67% of the data). From all references included in the meta-analysis (45 articles), data on a total of 115 sources were compiled (a single article may have information on more than one source).

Due to the high variability in the physicochemical composition of the different sources of organic and mineral fertilizers, the random effects model was chosen, with a one-arm forest plot (without control). The data were categorized into subgroups that mainly reflect their origin, detailed further below. This model assumes that, due to the heterogeneity among studies, the true effect may vary from study to study [44]. Therefore, we recognize that the variability among diets and sources (e.g., type of animal, raw materials, and production patterns) produces high $I^2$ and $\tau^2$ values (Table S1.4). Nevertheless, our assumptions are adequate for the purposes of this work, provided they are interpreted with caution considering their limitations.

The organic sources composition is directly related to the type of animal (i.e., species, weight, growth stage, sex, age, and cultivation purpose), the plant material (in the case of organic compost, chicken litter and others), the environment and the sampling procedures. Therefore, manures were grouped according to the type of animal (cattle manure, swine manure, chicken manure), while the composts (mix of animal waste + plant residue) were grouped into a single category. Sewage sludge was a separate category, including different types of municipal solid organic waste. Any residue from the industrial processing of plant, animal, or similar products was categorized as industrial waste. For mineral fertilizers, the same pattern was followed, grouping together items of similar composition. Soil conditioners without a detailed description of their composition (e.g., gypsum or limestone) were grouped into the same category.

### *3.2.4. Statistical and Ecotoxicological Risk Analysis*

GraphPad, Boston, MA, USA version 9.0 [45] was used to compare the concentrations of Cd, Cu, Ni, Cr, As, Pb, and Zn between mineral and organic sources, with an un-paired t-

test with Welch correction, at 5% significance level. Soil conditioners and industrial waste were not included due to a lack of a clear identification of their origin as organic or mineral.

The predicted environmental concentration of PTEs (PEC) in soil was estimated by considering a soil layer of 0.2 m and a bulk density of 1500 kg/m$^3$, using the average concentration (most likely scenario) and the maximum concentration (worst-case scenario) of each metal (M) and the applied dose (D). The concentration was obtained according to Equation (1):

$$PEC = (D \times M)/(3 \times 10^6) \quad (1)$$

Ecotoxicological risk analysis to soil organisms was performed using the risk quotient approach (RQ), considering predicted no-effect concentration (PNEC) data, a protective value for soil biota and the terrestrial ecosystem. The metals evaluated and their respective PNEC values (mg/kg) were: As (0.86), Cd (0.21), Cr (1.57), Pb (3.05), Hg (0.16), Cu (13.1), and Zn (38.3). The PNEC values were estimated from the hazardous concentration for 5% of the species (HC5), calculated using species sensitivity distribution (SSD) curves, according to the following references: Xu et al., 2019; Wang et al., 2015; Wang et al., 2018; and Wan et al., 2020 [46–49].

RQ was used to assess the ecotoxicological risk, representing the ratio between the PEC and the PNEC, according to Equation (2):

$$RQ = PEC/PNEC \quad (2)$$

RQ is a robust approach for assessing ecotoxicological risk and is widely used in environmental studies [50,51]. RQ was interpreted according to Bhandari et al. (2021) [52], with the following classes: no risk ($RQ < 0.01$), low risk ($0.01 \leq RQ < 0.1$), moderate risk ($0.1 \leq RQ < 1$) and high risk ($RQ \geq 1$). To estimate the years to achieve the PNEC (Y), equation 3 was used:

$$Y = PNEC/PEC \quad (3)$$

Reference fertilizer doses from studies conducted on maize crops in soils with low natural fertility were used [53–58]. The lowest and highest doses presented in the studies were used to establish the risk analysis scenarios. A unique annual application of each fertilizer was considered to estimate the application limit, with exceptions for limestone and silicon fertilizer, with applications every 4 and 2 years, respectively. The following low and high doses were considered for the risk analysis: manure, compost and sewage sludge (5 and 20 t/ha); phosphate fertilizer (133 and 667 kg/ha); phosphate rock (150 and 750 kg/ha); potassium fertilizer (150 and 400 kg/ha); nitrogen fertilizer (333 and 1000 kg/ha); silicon fertilizer (0.88 and 1.32 t/ha); and limestone (0.75–4 t/ha). Four contrasting scenarios were considered: average metal concentration with a low fertilizer dose (Most likely-Low), average metal concentration with a

high fertilizer dose (Most likely-High), maximum metal concentration with a low fertilizer dose (Worst-case-Low), and maximum metal concentration with a high fertilizer dose (Worst-case-High). Tables S1.1 and S1.2 provide more details on the values and methodologies for deriving the PNEC and on the fertilizer doses used.

It is important to highlight that several uncertainty factors need to be considered in this risk analysis. Among them, the following stand out: (i) the limitation and focus of ecotoxicological data on subtropical climates and organisms; (ii) the physical–chemical and biological variability of soils, including contrasting natural concentrations of PTEs; (iii) other pathways of PTE inputs besides fertilizer application (e.g., atmospheric deposition and pesticide use); (iv) outputs (e.g., leaching, surface runoff, and absorption by agricultural crops), which might slow down soil metal accumulation; and (v) the heterogeneity of methods for PTE quantification between countries, in which the results cannot always be directly compared. These uncertainty factors can all lead to overestimations in risk analyses.

*3.2.5. Reference Values for PTEs in Fertilizers*

Allowed values for PTEs in fertilizers were compared against the following regulations: EU 2019/1009 [59], Brazilian IN SDA Nº27/2006 (as amended by IN SDA Nº 7/2016) [60], California Code of Regulations (3 CCR § 2302) [61], and the limit values from US EPA 40 C.F.R. Part 503 [62]. The more restrictive limits of the US EPA regulation were used as a ceiling for processed organic fertilizers [63,64]. For sewage sludge, the criteria were based on EU Directive 86/278/EEC [65], US EPA-40 C.F.R. Part 503 [62], and Brazilian CONAMA Resolution Nº 498/2020 [66].

European legislation was chosen due to its restrictive nature, while U.S. and Brazilian legislations were selected because they represent major global agricultural players with high fertilizer consumption [67,68]. Chinese legislation (NY/T 525-2021) for organic fertilizers [69] was used in the specific case of total Cr, since European and Brazilian legislation only include Cr (VI), while U.S. legislation does not include Cr in its regulation. The legal limit values adopted by European, Brazilian, and U.S. legislation are presented in Table S1.3.

## 3.3 RESULTS

### *3.3.1. Bibliometric Review on PTE Sources*

Between 1982 and 2024, there was an annual growth rate of 11.89% in the number of publications (**Figure 1. 1**). A total of 4728 authors contributed to the 1116 documents, resulting in an average of 4.24 authors per document, 29.45 citations per document, and 28.49% rate of international co-authorship. A total of 3008 author keywords and 2376 keyword plus were generated. The average age of the analyzed documents is 7.52 years since publication, with an average of 3.41 citations per year.

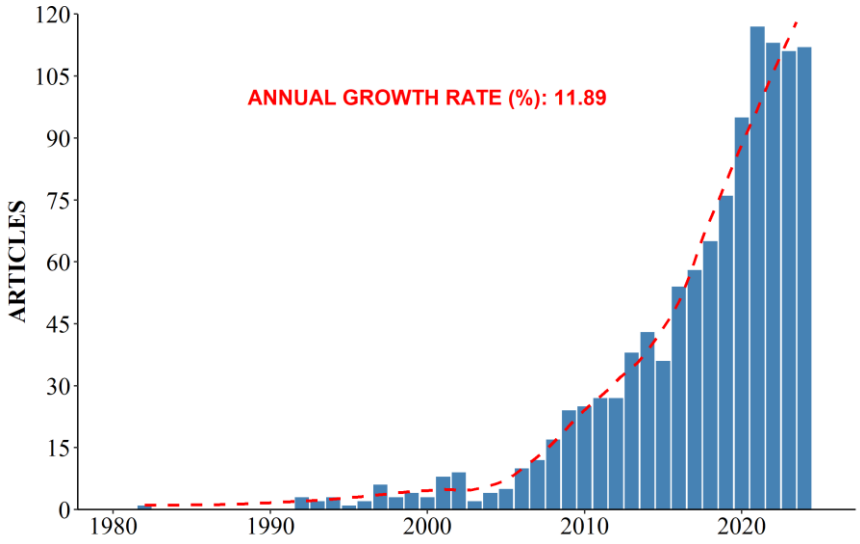


**Figure 1. 1** Annual production of articles on potentially toxic elements in mineral and organic fertilizers between 1984 and 2024.

The ten most productive authors were: Li Y (n = 30), Chen Y (n = 29), Zhang Y (n = 25), Wang X (n = 23), Li H (n = 22), Wang Y (n = 21), Wang L (n = 20), Wang J (n = 19), Liu Y (n = 19) and Liu X (n = 19), all from China. The most productive institutions were: Zhejiang University (n = 58), Sun Yat-sen University (n = 43), Institute of Soil Science from Chinese Academy of Sciences (n = 42), Federal University of Santa Maria (n = 40), and Nanjing Agricultural University (n = 32). Globally, the top five publishing countries were: China (n = 388), Brazil (n = 65), USA (n = 60), Spain (n = 57), and India (n = 46) (**Figure 1. 2**). Of these publications, China stands out for its extensive collaboration with other countries, with 86 publications, followed by Pakistan (26) and Brazil (14).

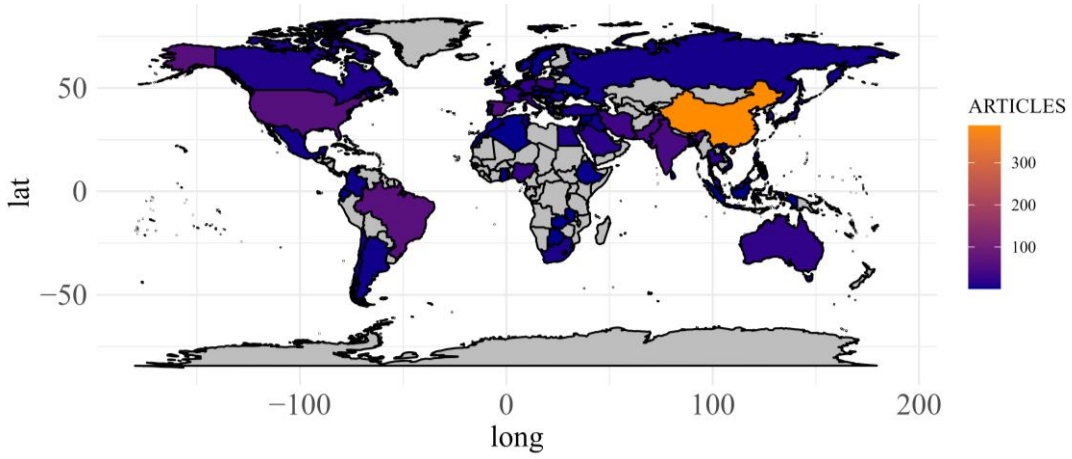


**Figure 1. 2** Global scientific production on potentially toxic elements in mineral and organic fertilizers between 1982 and 2024. The color scale corresponds to the number of articles published per region; gray areas indicate regions with no data available.

The three articles most cited were: “Biochar reduces the bioavailability and phytotoxicity of heavy metals” by Park et al. (2011) [70], with 879 citations; “Immobilization of Heavy Metal Ions (CuII, CdII, NiII, and PbII) by Broiler Litter-Derived Biochars in Water and Soil” by Uchimiya et al. (2010) [71], with 589 citations; and “Biochar application for the remediation of heavy metal polluted land: A review of in situ field trials” by O’Connor et al. (2018) [72], with 450 citations. It is important to note that these articles do not address the input of metals into the soil, focusing on remediation aspects.

The conceptual structure of the keywords plus (**Figure 1. 3**), that is, those generated by the database according to the cited references, produced five thematic clusters (number of terms = 50; number of documents = 50). The most cited articles are found in the blue and ochre clusters (**Figure 1. 3**). The blue cluster has the following key terms: biochar, heavy metal, soil, agricultural soil, pig slurry, plants, and phytoremediation. The central region of **Figure 1. 3** presents terms such as sewage sludge, compost, manure, accumulation, zinc, copper, toxicity, and trace elements. The ochre cluster has the following delimiting terms: waste, sorption, adsorption, removal, wastewater, water, biomass, and carbon, with the term “mechanisms” more centrally located.

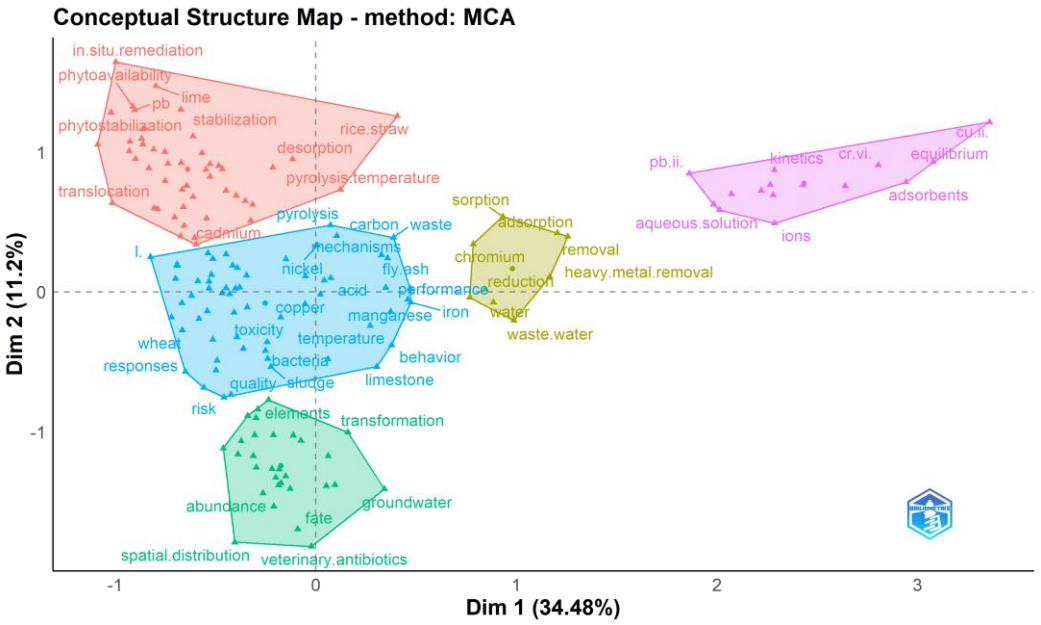


**Figure 1. 3** Conceptual structure based on keywords plus of articles on potentially toxic elements in mineral and organic fertilizers published between 1982 and 2024. The keywords are grouped into thematic clusters represented by distinct colors.

The thematic map (**Figure 1. 4**) shows the formation of two specialized groups (1: aqueous-solution; activated carbon; aqueous-solutions; biosorption; ions; and 2: swine manure; anaerobic-digestion; bacterial community; fate; microbial community); an emerging or declining group (contamination; pollution; sediments; risk-assessment; China); a driving group (cadmium; accumulation; lead; remediation; zinc); and two basic groups (1: adsorption; removal; water; sorption; wastewater; and 2: heavy-metals; copper; waste; mechanisms; biomass).

The relationship between these fields and the main research themes, the countries, and the journals is presented in **Figure 1. 5**. Based on the author keywords, it is possible to observe that heavy metals, cadmium, biochar, soil, and bioavailability are the main research drivers, with the largest contributions coming from China, Pakistan, USA, and Brazil. The major publishing journals were “Science of the Total Environment”, “Chemo-sphere”, and “Environmental Science and Pollution Research”.

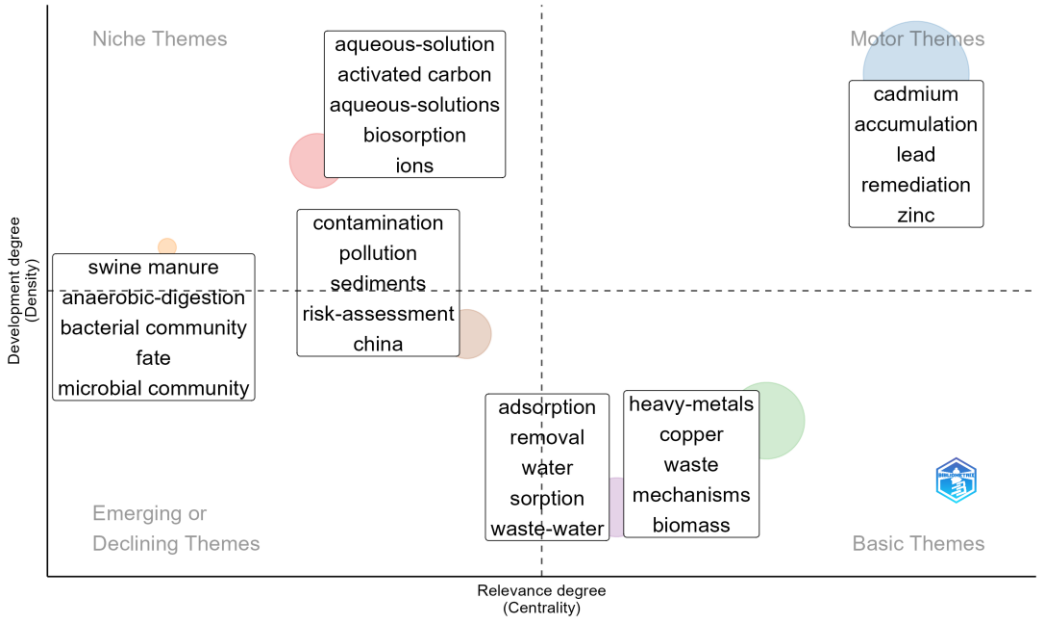


**Figure 1. 4** Thematic map based on keywords plus articles published between 1982 and 2024 on potentially toxic elements in mineral and organic fertilizers. The quadrants show the developmental degree and relevance of each topic.

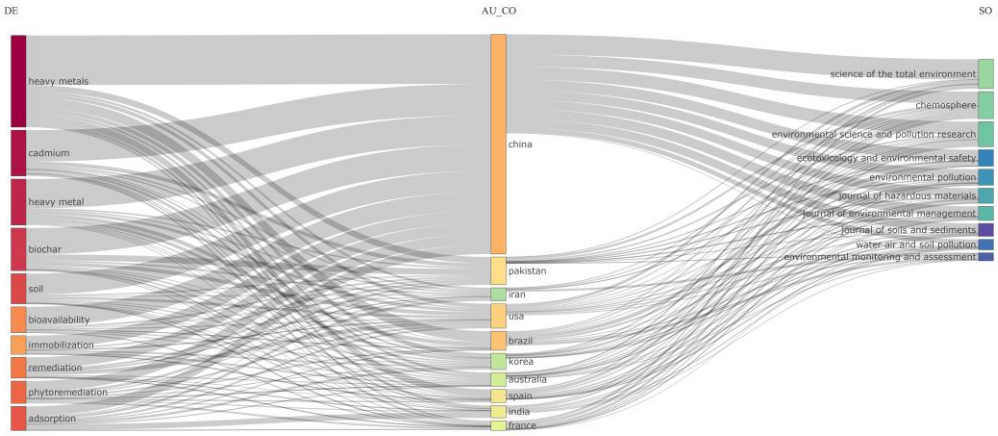


**Figure 1. 5** Three-field map (Sankey Diagram) based on the most relevant author keywords (DE), country (AU_CO), and journal (SO) in articles published between 1982 and 2024 on potentially toxic elements in mineral and organic fertilizers.

*3.3.2. Meta-Analysis*

The results evidenced a high heterogeneity and variance of the PTE concentrations in the different fertilizers, as indicated by high values of $I^2$ (above 75%), Q, and $\tau^2$ (Table S1.4). A high heterogeneity was expected due to the fertilizers' distinct origins and relevant variations in several factors, such as physicochemical composition, management patterns, edaphoclimatic conditions, as well as production processes [73]. Thus, the means and confidence intervals (**Figure 1. 6Figure 1. 7**) were described by the random effects model, which incorporates part of the variance between studies [44].

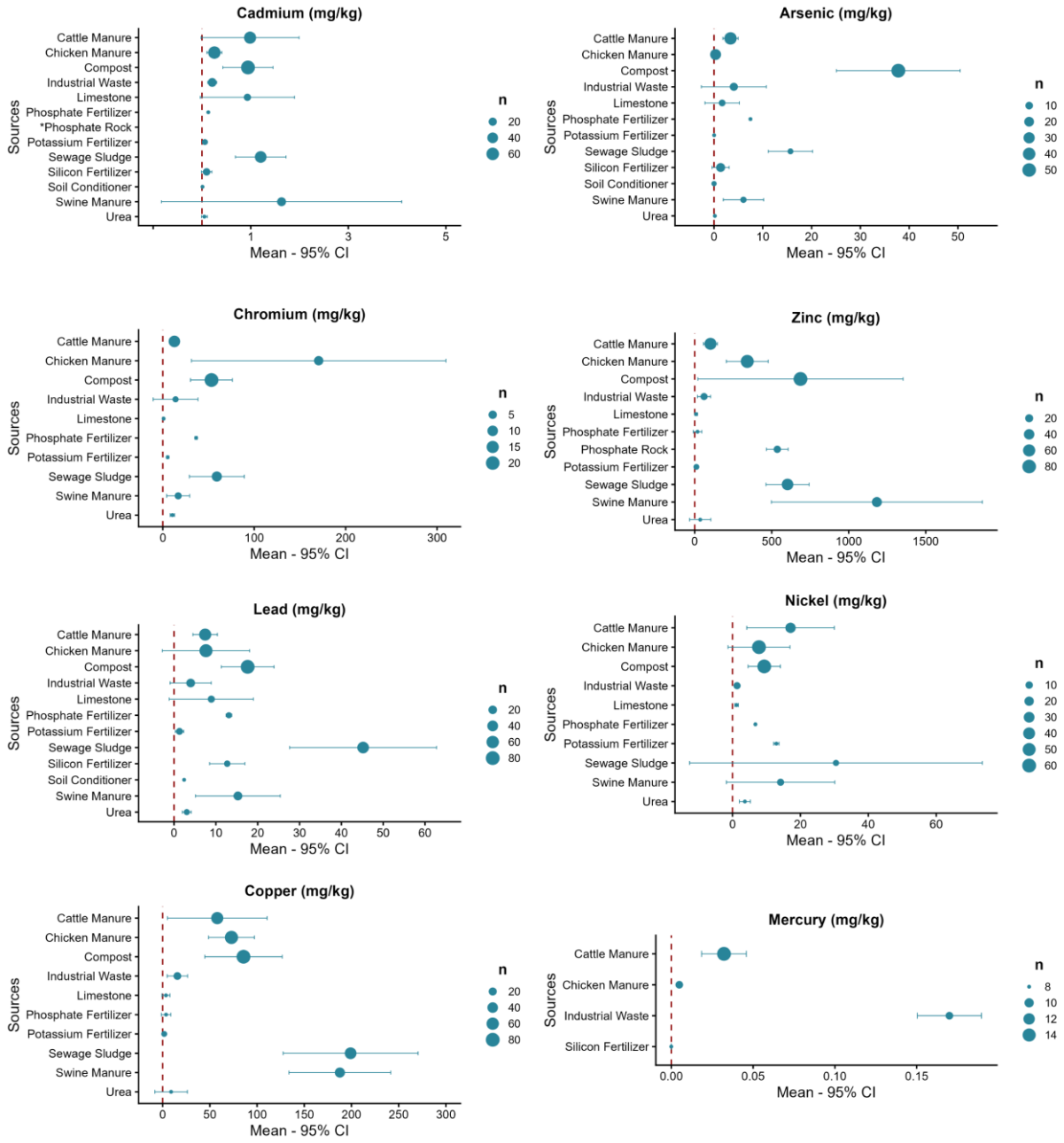


**Figure 1. 6** Average concentrations of potentially toxic elements (mg/kg) found in mineral and organic fertilizers and their confidence intervals (95%). The circle size represents the sample size (N) of each source. The asterisk in the cadmium graph indicates that the confidence interval and the average exceed the adopted scale. The data are derived from the 45 references included in the meta-analysis.

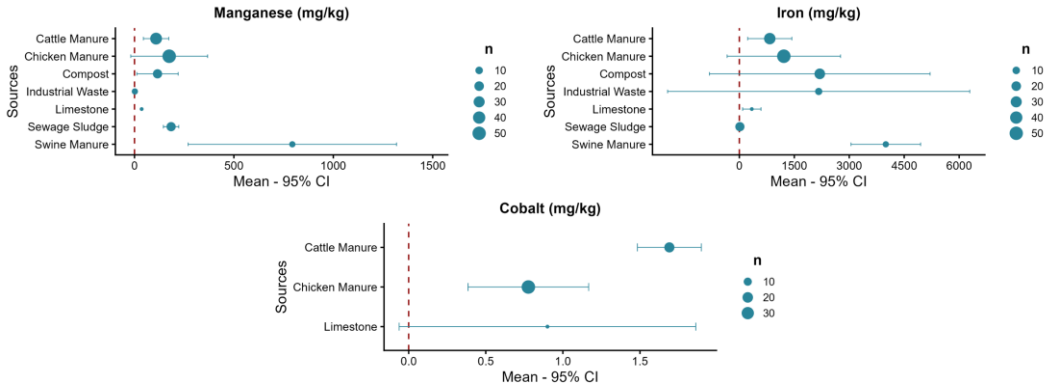


**Figure 1. 7** Average concentrations of potentially toxic elements (mg/kg) found in mineral and organic fertilizers and their confidence interval (95%). The circle size represents the sample size (N) of each source. The data are derived from the 45 references included in the meta-analysis.

3.3.2.1. PTE Occurrence on Mineral Sources

Powdered phosphate rock showed the highest cadmium concentration among all sources (74.2 mg/kg, **Table 1. 1**). This value exceeds the EU legislation limit of 60 mg/kg for fertilizers with P2O5 ≥ 5%. It is also slightly above the U.S. and Brazilian legislation limits (80 mg/kg).

**Table 1. 1** Concentration of potentially toxic elements by fertilizer source: average, minimum, maximum values and sample size. The values are expressed in mg/kg. The data are derived from the 45 references included in the meta-analysis.

| Sources | Cd | Cu | Cr | As | Pb | Zn |
|---|---|---|---|---|---|---|
| | | mean (min–max) n−mg/kg | | | | |
| Cattle Manure | 1.1 (0.0–4.4) 46 | 59.4 (1.0–423.3) 49 | 12.5 (0.0–159.3) 46 | 3.7 (0.0–7.5) 37 | 7.8 (0.0–19.6) 46 | 96.1 (10.0–371.1) 49 |
| Chicken Manure | 0.3 (0.0–0.5) 38 | 73.0 (13.1–164.2) 50 | 170.5 (0.0–418.9) 44 | 0.3 (0.22–2.7) 28 | 7.6 (0.0–53.0) 47 | 341.4 (70.0–655.8) 50 |
| Compost | 0.9 (0.0–3.1) 60 | 53.8 (0.0–132.7) 57 | 53.3 (0.0–124.2) 69 | 38.0 (1.9–63.7) 36 | 17.6 (0.0–53.3) 63 | 209.2 (0.0–425.8) 60 |
| Industrial Waste | 0.1 (0.0–0.1) 15 | 4.4 (0.3–8.6) 6 | 1.5 (0.0–1.5) 15 | 7.5 (7.5–7.5) 9 | 7.3 (0.0–12.8) 15 | 34.5 (2.0–76.5) 6 |
| Limestone | 0.9 (0.0–1.9) 15 | 3.5 (1.3–5.6) 7 | 0.9 (0.9-0.9) 3 | 1.7 (0.1-3.6) 7 | 8.9 (1.2-20.1) 11 | 9.2 (4.3–14.1) 7 |
| Mushroom Residue | 0.3 (0.2–0.3) 6 | 25.3 (19.2–31.6) 6 | - | - | 1.2 (0.6–1.9) 6 | 82.6 (37.4–127.9) 6 |
| Phosphate Fertilizer | 0.1 (0.1–0.1) 7 | 3.6 (1.0–6.1) 7 | 36.5 (36.5–36.5) 3 | 7.5 (7.5–7.5) 3 | 13.1 (0.0–13.1) 7 | 18.5 (4.5–32.5) 7 |
| Phosphate Rock | 74.2 (67.2–81.4) 6 | - | - | - | - | 536.5 (500.0–572.0) 6 |
| Potassium Fertilizer | 0.0 (0.0–0.0) 10 | 1.7 (0.5–3.2) 10 | 5.4 (5.4–5.4) 3 | 0.0 (0.0–0.0) 3 | 1.3 (0.0–1.8) 10 | 10.8 (4.3–14.2) 10 |
| Sewage Sludge | 1.2 (0.0–3.3) 50 | 199.1 (63.0–637.8) 53 | 59.1 (0.0–180.5) 32 | 15.7 (15.7–15.7) 5 | 45.2 (8.0–119.9) 50 | 602.9 (159.0–1217.0) 53 |
| Silicon Fertilizer | 0.1 (0.0–0.2) 16 | - | 4.7 (0.2–9.2) 8 | 1.3 (0.0–3.8) 16 | 12.7 (10.6–14.9) 8 | - |
| Soil Conditioner | 0.0 (0.0–0.0) 7 | - | - | 0.0 (0.0–0.0) 4 | 2.4 (2.4–2.4) 3 | - |
| Swine Manure | 1.6 (0.0–7.9) 25 | 169.8 (20.6–588.3) 30 | 16.7 (0.0–23.2) 15 | 6.0 (6.0–6.0) 6 | 15.3 (3.0–31.2) 22 | 736.0 (81.6–2404.5) 30 |
| Urea | 0.0 (0.0–0.0) 7 | 9.0 (0.2–17.8) 7 | 10.6 (10.6–10.6) 3 | 0.2 (0.2–0.2) 3 | 3.0 (2.5–3.6) 7 | 35.7 (0.7–70.8) 7 |
| | Ni | Fe | Mn | Co | Hg | |
| Cattle Manure | 19.3 (1.5–38.0) 24 | 858.2 (20.0–2962.0) 24 | 108.6 (9.0–285.7) 36 | 1.7 (0.0–1.82) 18 | 0.0 (0.0–0.1) 15 | |
| Chicken Manure | 7.8 (0.5–36.8) 39 | 1215.2 (83.0–3483.0) 30 | 174.5 (21.0–452.8) 30 | 0.7 (0.6–1.0) 17 | 0.0 (0.0–0.0) 9 | |
| Compost | 9.3 (0.0–27.6) 42 | 2861.7 (1.1–12,472.0) 27 | 115.8 (0.1–257.3) 18 | - | - | |
| Industrial Waste | 0.0 (0.0–0.0) 6 | 3189.2 (3.3–6378.0) 6 | 1.87 (0.3–3.45) 6 | - | 0.2 (0.2–0.2) 9 | |
| Limestone | 1.2 (1.2–1.2) 3 | 340.0 (340.0–340.0) 3 | 36.0 (36.0–36.0) 3 | 0.9 (0.9–0.9) 3 | - | |
| Phosphate Fertilizer | 6.7 (6.7–6.7) 3 | - | - | - | - | |
| Potassium Fertilizer | 12.9 (12.9–12.9) 3 | - | - | - | - | |
| Sewage Sludge | 30.5 (9.0–53.0) 6 | 14.7 (7.5–32.0) 18 | 184.1 (128.6–240.5) 18 | - | - | |
| Silicon Fertilizer | - | - | - | - | 0.0 (0.0–0.0) 8 | |
| Swine Manure | 14.1 (3.0–30.3) 9 | 3998.1 (3490.0–4460.0) 6 | 793.9 (525.2–1060.0) 6 | - | - | |
| Urea | 3.6 (3.6–3.6) 3 | - | - | - | - | |

Other phosphate fertilizers (e.g., calcium superphosphate, calcium magnesium phosphate) had Cd concentrations (0.12–0.14 mg/kg) much lower than phosphate rock, remaining within all the regulations considered (**Figure 1. 6**; **Table 1. 1**). Regarding Cr, the phosphate fertilizer had an average concentration of 36.47 mg/kg, the highest value among all mineral sources. California legislation does not limit Cr in phosphate fertilizers; it regulates only As, Cd, and Pb. Brazilian legislation does not evaluate Cr in fertilizers that exclusively supply micronutrients, fertilizers with secondary macronutrients and micronutrients, mixed mineral fertilizers, and complex fertilizers with guaranteed primary macro-nutrients and micronutrients. European legislation only evaluates hexavalent Cr, precluding comparison with total Cr concentrations.

3.3.2.2. PTE Occurrence on Organic Sources

Swine manure showed a maximum Cd concentration of 7.91 mg/kg, exceeding the limits of EU legislation (1.5 mg/kg), as well as the limits set by Brazilian (3 mg/kg) and U.S. (39 mg/kg) legislation. Cu and Zn were also found in high concentrations, ranging from 20.57 to 588.32 and 81.57 to 3270.00 mg/kg, respectively (**Figure 1. 6**; **Table 1. 1**). Cu and Zn maximum values are higher than the limits for Cu (300 mg/kg) and Zn (800 mg/kg) in European legislation. However, these values fall within the USEPA legislation limits for Cu (1500 mg/kg) and Zn (2800 mg/kg). Brazilian legislation does not establish limits for these elements. Swine manure also had high concentrations of manganese (525.20–1060.00 mg/kg) and Fe (3490.00–4460.00 mg/kg), which are not regulated by any legislation (Figure 1.7; Table 1.1).

Chicken manure showed variable Cr concentration (0.05–418.94 mg/kg), with a large confidence interval, demonstrating high data variability (**Figure 1. 6**; **Table 1. 1**). The chicken manure maximum Cr concentration is approximately three times higher and 18 times higher than maximum concentrations for cattle and swine manure, respectively. Chinese legislation on organic fertilizers (NY/T 525-2021) [75] sets a limit of 150 mg/kg for total Cr, which means that the maximum value for chicken manure is approximately three times higher.

Cattle manure showed a Cd maximum concentration of 4.40 mg/kg (**Figure 1. 6**; **Table 1. 1**), above the limits of European (1.5 mg/kg) and Brazilian legislation (3 mg/kg) but within USEPA legislation (39 mg/kg). The range of Ni concentrations found in cattle manure (1.52–38.00 mg/kg) stood out in relation to other animal sources; however, maximum concentrations were within the limits of European, Brazilian, and USEPA legislation (50, 70, and 420 mg/kg, respectively). For total Cr in cattle manure, the maximum value (159.30 mg/kg) was above the limit established in Chinese legislation (150 mg/kg).

The organic compost showed a Cd maximum concentration of 3.14 mg/kg, above the limits of both European (1.5 mg/kg) and Brazilian (3 mg/kg) legislation but below the USEPA legislation limit of 39 mg/kg (**Figure 1. 6**; **Table 1. 1**). Concentrations of arsenic ranged from 1.90 to 63.67 mg/kg, with high data variability and a maximum value above the limits of European (40 mg/kg), Brazilian (20 mg/kg) and USEPA legislation (41 mg/kg). Furthermore, the maximum concentrations of Cu (351 mg/kg) and Zn (5651 mg/kg) also exceeded the limits set by European legislation for organic fertilizers (300 and 800 mg/kg, respectively) and only for Zn in the case of USEPA legislation (1500 and 2800 mg/kg, respectively). Iron concentrations ranged from 1.10 to 12,472.00 mg/kg (**Figure 1. 7**; **Table 1. 1**).

Sewage sludge showed Cu concentrations between 63.00 and 637.80 mg/kg (**Figure 1. 6**; **Table 1. 1**), not exceeding European (1000–1750 mg/kg), Brazilian (1500–4300 mg/kg) or USEPA (1500–4300 mg/kg) regulations. For Zn, the values varied from 159.00 to 1217.00 mg/kg (**Figure 1. 6**; **Table 1. 1**), within the limits adopted by all legislations: European (2500–4000 mg/kg), USEPA (2800–7500 mg/kg) and Brazilian (2800–7500 mg/kg). For Ni, the values in sewage sludge (9.00–53.00 mg/kg) were higher than in other sources (maximum of 38.00 mg/kg in cattle manure) but within the limits established by European (300–400 mg/kg), USEPA (420 mg/kg) and Brazilian (420 mg/kg) legislation. High Pb concentrations were seen in sewage sludge (8.00–119.90 mg/kg) but also did not exceed the limits determined by European (750–1200 mg/kg), USEPA (300–840 mg/kg) and Brazilian (300–840 mg/kg) legislation (**Figure 1. 6**; **Table 1. 1**).

For mercury, none of the sources showed concentrations above the limits of the legislation consulted (**Figure 1. 6**; **Table 1. 1**). In general, organic sources showed higher concentrations of Mn, Fe, and Co than mineral ones (**Figure 1. 7**; **Table 1. 1**).

3.3.2.3. Other Sources

Potassium and silicon fertilizers, urea, limestone, industrial waste, and soil conditioners did not show relevant concentrations of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn, being always within the limits of all the legislations consulted. Brazilian legislation does not provide limits for Cu and Zn, although it is more restrictive than European legislation for Hg.

### *3.3.3. Mineral Versus Organic Sources*

The comparison between mineral and organic sources for PTE concentrations is shown in **Figure 1. 8**. Significant differences ($p < 0.05$) were found between the sources for copper ($p < 0.0001$), chromium ($p < 0.0001$), arsenic ($p = 0.0010$), lead ($p = 0.0004$), and zinc ($p =$

0.0008), which were all higher in organic sources. In contrast, no significant differences were found for cadmium (p = 0.1940) and nickel (p = 0.1218).

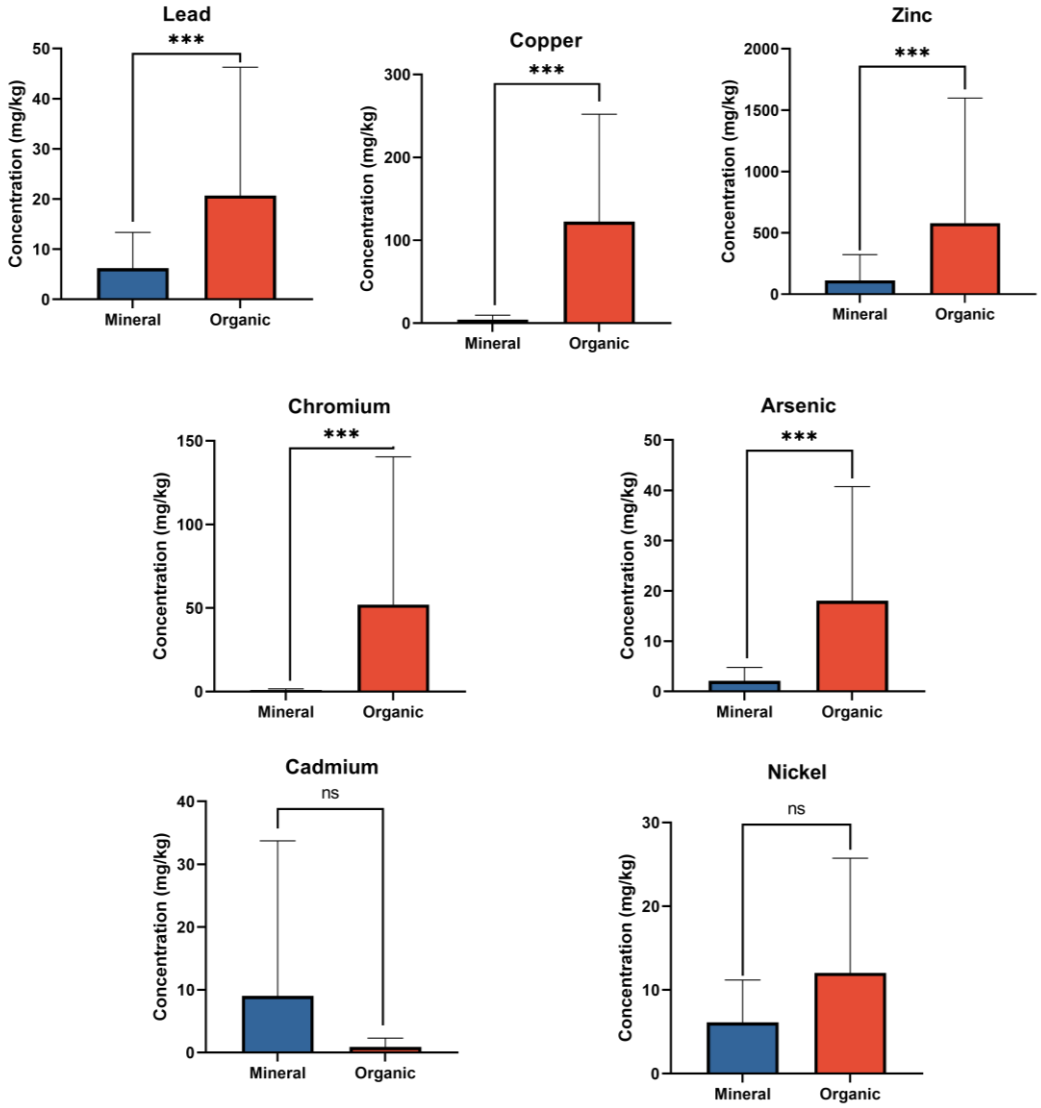


**Figure 1. 8** Average concentration and standard deviation of the most relevant potentially toxic elements among mineral and organic sources (unpaired t-test with Welch correction). ns = not significant ($p > 0.05$); *** significant at 1% ($p < 0.001$). The data are derived from the 45 references included in the meta-analysis.

*3.3.4. Ecotoxicological Risk for Soil Organisms*

Overall, organic sources showed higher RQ values than mineral ones (**Table 1. 2Table 1. 3**). Considering the most likely scenario (using the mean PTE concentration), at low application rate, only Cr (0.18, moderate risk) for chicken manure and Cd (0.02, low risk) for phosphate rock presented any risk. On the other hand, considering high application rates, other PTEs and fertilizers showed moderate risk (**Table 1. 2**): Cr—chicken manure (0.72), sewage

sludge (0.25), compost (0.23); As—compost (0.29), sewage sludge (0.12); Zn—swine manure (0.21), compost (0.12), sewage sludge (0.10); and Cu—sewage sludge (0.10).

The worst-case scenario (using the maximum reported metal concentration) showed a much more concerning picture for organic sources, with moderate risks at low application rates for: Cr—chicken manure (0.44), sewage sludge (0.19), cattle manure (0.17), compost (0.13); As—compost (0.12); and Zn—compost (0.25), swine manure (0.14). On the other hand, for mineral sources, only low risks were observed: Cd—limestone (0.02), phosphate rock (0.02); and Pb—limestone (0.01). At high application rates, the RQ for Cr in chicken manure (1.78) stood out, surpassing the safety levels as fast as in 1 year (a unique application). In this worst-case scenario, all organic sources presented moderate RQ for at least two PTEs, especially sewage sludge, which showed moderate RQ for: Cr (0.77), Cu (0.32), Pb (0.26), Zn (0.21), As (0.12), and Cd (0.10). Mineral sources presented only low risks for: Cd—phosphate rock (0.10), limestone (0.06); As—limestone (0.03); and Pb—limestone (0.04) (**Table 1. 3**).

**Table 1. 2** Ecological risk quotient (RQ) and time to PNEC (predicted no-effect concentration) for potentially toxic elements in mineral and organic fertilizers—most likely scenario (mean values).

| Mineral Sources | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Sources | Limestone | | | Silicon Fertilizer | | | Urea | | | Phosphate Rock | | | Phosphate Fertilizer | | | Potassium Fertilizer | | |
| Low Rate | Metal | Mean | RQ | Years * | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years |
| | Cd | 0.9 | 0.00 | 933 | 0.1 | 0.00 | 14,000 | 0.0 | - | - | 74.2 | 0.02 | 57 | 0.1 | 0.00 | 52,500 | 0.0 | - | - |
| | Cr | 0.9 | 0.00 | 3925 | 4.7 | 0.00 | 2279 | 10.6 | 0.00 | 1374 | - | - | - | 36.5 | 0.00 | 970 | 5.4 | 0.00 | 5815 |
| | As | 0.1 | 0.00 | 34,400 | 1.4 | 0.00 | 4195 | 0.2 | 0.00 | 39,091 | - | - | - | 7.5 | 0.00 | 2583 | 0.0 | - | - |
| | Pb | 8.9 | 0.00 | 1371 | 12.7 | 0.00 | 1637 | 3.0 | 0.00 | 9159 | - | - | - | 13.1 | 0.00 | 5250 | 1.3 | 0.00 | 46,923 |
| | Ni | 1.2 | 0.00 | 21,667 | - | - | - | 3.6 | 0.00 | 16,250 | - | - | - | 6.7 | 0.00 | 21,886 | 12.9 | 0.00 | 10,078 |
| | Cu | 3.5 | 0.00 | 14,971 | - | - | - | 9.0 | 0.00 | 13,113 | - | - | - | 3.6 | 0.00 | 81,875 | 1.7 | 0.00 | 154,118 |
| | Zn | 9.2 | 0.00 | 16,652 | - | - | - | 35.7 | 0.00 | 9664 | 536.5 | 0.00 | 1428 | 18.5 | 0.00 | 46,707 | 10.8 | 0.00 | 70,926 |
| | Sources | Limestone | | | Silicon Fertilizer | | | Urea | | | Phosphate Rock | | | Phosphate Fertilizer | | | Potassium Fertilizer | | |
| High Rate | Metal | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years |
| | Cd | 0.9 | 0.01 | 175 | 0.1 | 0.00 | 4773 | 0.0 | - | - | 74.2 | 0.09 | 11 | 0.1 | 0.00 | 9545 | 0.0 | - | - |
| | Cr | 0.9 | 0.00 | 736 | 4.7 | 0.00 | 759 | 10.6 | 0.00 | 457 | - | - | - | 36.5 | 0.01 | 193 | 5.4 | 0.00 | 2181 |
| | As | 0.1 | 0.00 | 26,061 | 1.4 | 0.00 | 1396 | 0.2 | 0.00 | 12,836 | - | - | - | 7.5 | 0.00 | 516 | 0.0 | - | - |
| | Pb | 8.9 | 0.00 | 1028 | 12.7 | 0.00 | 546 | 3.0 | 0.00 | 3050 | - | - | - | 13.1 | 0.00 | 1047 | 1.3 | 0.00 | 17,630 |
| | Ni | 1.2 | 0.00 | 4063 | - | - | - | 3.6 | 0.00 | 5417 | - | - | - | 6.7 | 0.00 | 4362 | 12.9 | 0.00 | 3779 |
| | Cu | 3.5 | 0.00 | 2807 | - | - | - | 9.0 | 0.00 | 4367 | - | - | - | 3.6 | 0.00 | 16,375 | 1.7 | 0.00 | 57,709 |
| | Zn | 9.2 | 0.00 | 3122 | - | - | - | 35.7 | 0.00 | 3218 | 536.5 | 0.00 | 286 | 18.5 | 0.00 | 9312 | 10.8 | 0.00 | 26,597 |

| Organic Sources | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Sources | Cattle Manure | | | Chicken Manure | | | Compost | | | Swine Manure | | | Sewage Sludge | | |
| Low Rate | Metal | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years |
| | Cd | 1.0 | 0.01 | 126 | 0.3 | 0.00 | 420 | 0.9 | 0.01 | 140 | 1.6 | 0.01 | 79 | 1.2 | 0.01 | 105 |
| | Cr | 12.7 | 0.01 | 74 | 170.5 | 0.18 | 6 | 53.3 | 0.06 | 18 | 16.7 | 0.02 | 56 | 59.1 | 0.06 | 16 |
| | As | 3.4 | 0.01 | 152 | 0.3 | 0.00 | 1720 | 37.8 | 0.07 | 14 | 6.0 | 0.01 | 86 | 15.7 | 0.03 | 33 |
| | Pb | 7.4 | 0.00 | 247 | 7.6 | 0.00 | 241 | 17.6 | 0.01 | 104 | 15.3 | 0.01 | 120 | 45.2 | 0.02 | 40 |
| | Ni | 17.1 | 0.00 | 228 | 7.8 | 0.00 | 500 | 9.3 | 0.00 | 419 | 14.1 | 0.00 | 277 | 30.5 | 0.01 | 128 |
| | Cu | 57.8 | 0.01 | 136 | 72.9 | 0.01 | 108 | 85.6 | 0.01 | 92 | 187.7 | 0.02 | 42 | 199.1 | 0.03 | 39 |
| | Zn | 102.3 | 0.00 | 225 | 341.4 | 0.01 | 67 | 689.9 | 0.03 | 33 | 1182.5 | 0.05 | 19 | 602.9 | 0.03 | 38 |
| | Sources | Cattle Manure | | | Chicken Manure | | | Compost | | | Swine Manure | | | Sewage Sludge | | |
| High Rate | Metal | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years | Mean | RQ | Years |
| | Cd | 1.0 | 0.03 | 31 | 0.3 | 0.01 | 105 | 0.9 | 0.03 | 35 | 1.6 | 0.05 | 20 | 1.2 | 0.04 | 26 |
| | Cr | 12.7 | 0.05 | 19 | 170.5 | 0.72 | 1 | 53.3 | 0.23 | 4 | 16.7 | 0.07 | 14 | 59.1 | 0.25 | 4 |

| | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| As | 3.4 | 0.03 | 38 | 0.3 | 0.00 | 430 | 37.8 | 0.29 | 3 | 6.0 | 0.05 | 22 | 15.7 | 0.12 | 8 |
| Pb | 7.4 | 0.02 | 62 | 7.6 | 0.02 | 60 | 17.6 | 0.04 | 26 | 15.3 | 0.03 | 30 | 45.2 | 0.10 | 10 |
| Ni | 17.1 | 0.02 | 57 | 7.8 | 0.01 | 125 | 9.3 | 0.01 | 105 | 14.1 | 0.01 | 69 | 30.5 | 0.03 | 32 |
| Cu | 57.8 | 0.03 | 34 | 72.9 | 0.04 | 27 | 85.6 | 0.04 | 23 | 187.7 | 0.10 | 10 | 199.1 | 0.10 | 10 |
| Zn | 102.3 | 0.02 | 56 | 341.4 | 0.06 | 17 | 689.9 | 0.12 | 8 | 1182.5 | 0.21 | 5 | 602.9 | 0.10 | 10 |

mg/kg—milligrams per kilogram; kg/ha/year—kilograms per hectare per year. * Annual applications, not considering losses due to leaching, surface runoff, and crop absorption (see a detailed description in Section 2.4). Assumed soil density: 1.5 g/cm$^3$; soil depth: 0.2 m.

**Table 1. 3** Ecological risk quotient (RQ) and time to PNEC (predicted no-effect concentration) for potentially toxic elements in mineral and organic fertilizers—worst-case scenario (mean values).

| Mineral Sources | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Sources | Limestone | | | Silicon Fertilizer | | | Urea | | | Phosphate Rock | | | Phosphate Fertilizer | | | Potassium Fertilizer | | |
| Low Rate | Metal | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years |
| | Cd | 1.9 | 0.02 | 66 | 0.2 | 0.00 | 7241 | 0.1 | 0.00 | 19,091 | 81.4 | 0.02 | 52 | 0.1 | 0.00 | 52,500 | 0.1 | 0.00 | 42,000 |
| | Cr | 0.9 | 0.00 | 1047 | 9.2 | 0.00 | 1164 | 10.6 | 0.00 | 1334 | - | - | - | 36.5 | 0.00 | 970 | 5.4 | 0.00 | 48,519 |
| | As | 3.6 | 0.01 | 143 | 3.8 | 0.00 | 1544 | 0.2 | 0.00 | 39,091 | - | - | - | 7.5 | 0.00 | 2583 | 0.0 | - | - |
| | Pb | 20.1 | 0.01 | 91 | 14.9 | 0.00 | 1396 | 3.6 | 0.00 | 7625 | - | - | - | 13.1 | 0.00 | 5250 | 1.8 | 0.00 | 33,889 |
| | Ni | 1.2 | 0.00 | 21,667 | - | - | - | 3.6 | 0.00 | 16,250 | - | - | - | 6.7 | 0.00 | 21,886 | 12.9 | 0.00 | 10,078 |
| | Cu | 5.6 | 0.00 | 1404 | - | - | - | 17.8 | 0.00 | 6630 | - | - | - | 6.1 | 0.00 | 48,519 | 3.2 | 0.00 | 81,875 |
| | Zn | 14.1 | 0.00 | 1630 | - | - | - | 70.8 | 0.00 | 4873 | 572.0 | 0.00 | 1339 | 32.5 | 0.00 | 26,579 | 14.2 | 0.00 | 53,944 |
| | Sources | Limestone | | | Silicon Fertilizer | | | Urea | | | Phosphate Rock | | | Phosphate Fertilizer | | | Potassium Fertilizer | | |
| High Rate | Metal | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years |
| | Cd | 1.9 | 0.06 | 17 | 0.2 | 0.00 | 2386 | 0.1 | 0.00 | 6364 | 81.4 | 0.10 | 10 | 0.1 | 0.00 | 9545 | 0.1 | 0.00 | 16,154 |
| | Cr | 0.9 | 0.00 | 262 | 9.2 | 0.00 | 388 | 10.6 | 0.00 | 444 | - | - | - | 36.5 | 0.01 | 193 | 5.4 | 0.00 | 18,194 |
| | As | 3.6 | 0.03 | 36 | 3.8 | 0.00 | 514 | 0.2 | 0.00 | 12,836 | - | - | - | 7.5 | 0.00 | 516 | 0.0 | - | - |
| | Pb | 20.1 | 0.04 | 23 | 14.9 | 0.00 | 465 | 3.6 | 0.00 | 2542 | - | - | - | 13.1 | 0.00 | 1047 | 1.8 | 0.00 | 12,708 |
| | Ni | 1.2 | 0.00 | 4063 | - | - | - | 3.6 | 0.00 | 5417 | - | - | - | 6.7 | 0.00 | 4362 | 12.9 | 0.00 | 3779 |
| | Cu | 5.6 | 0.00 | 351 | - | - | - | 17.8 | 0.00 | 2208 | - | - | - | 6.1 | 0.00 | 9661 | 3.2 | 0.00 | 30,679 |
| | Zn | 14.1 | 0.00 | 407 | - | - | - | 70.8 | 0.00 | 1623 | 572.0 | 0.00 | 268 | 32.5 | 0.00 | 5300 | 14.2 | 0.00 | 20,232 |
| Organic Sources | | | | | | | | | | | | | | | | | | | |
| | Sources | Cattle Manure | | | Chicken Manure | | | Compost | | | Swine Manure | | | Sewage Sludge | | | | | |
| Low Rate | Metal | Max | RQ | Years * | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | | | |
| | Cd | 4.4 | 0.03 | 29 | 0.5 | 0.00 | 252 | 3.1 | 0.02 | 41 | 7.9 | 0.06 | 16 | 3.3 | 0.03 | 38 | | | |
| | Cr | 159.3 | 0.17 | 6 | 418.9 | 0.44 | 2 | 124.2 | 0.13 | 8 | 10.6 | 0.01 | 89 | 180.5 | 0.19 | 5 | | | |
| | As | 7.5 | 0.01 | 69 | 2.7 | 0.01 | 191 | 63.7 | 0.12 | 8 | 6.0 | 0.01 | 86 | 15.7 | 0.03 | 33 | | | |
| | Pb | 19.6 | 0.01 | 93 | 53.0 | 0.03 | 35 | 53.3 | 0.03 | 34 | 31.2 | 0.02 | 59 | 119.9 | 0.07 | 15 | | | |
| | Ni | 38.0 | 0.01 | 103 | 36.8 | 0.01 | 106 | 27.6 | 0.01 | 141 | 30.3 | 0.01 | 129 | 53.0 | 0.01 | 74 | | | |
| | Cu | 423.3 | 0.05 | 19 | 164.2 | 0.02 | 48 | 351.0 | 0.04 | 22 | 588.3 | 0.07 | 13 | 637.8 | 0.08 | 12 | | | |
| | Zn | 371.1 | 0.02 | 62 | 655.8 | 0.03 | 35 | 5651.0 | 0.25 | 4 | 3270.0 | 0.14 | 7 | 1217.0 | 0.05 | 19 | | | |
| | Sources | Cattle Manure | | | Chicken Manure | | | Compost | | | Swine Manure | | | Sewage Sludge | | | | | |
| High Rate | Metal | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | Max | RQ | Years | | | |
| | Cd | 4.4 | 0.14 | 7 | 0.5 | 0.02 | 63 | 3.1 | 0.10 | 10 | 7.9 | 0.25 | 4 | 3.3 | 0.10 | 10 | | | |
| | Cr | 159.3 | 0.68 | 1 | 418.9 | 1.78 | 1 | 124.2 | 0.53 | 2 | 10.6 | 0.05 | 22 | 180.5 | 0.77 | 1 | | | |

| | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| As | 7.5 | 0.06 | 17 | 2.7 | 0.02 | 48 | 63.7 | 0.49 | 2 | 6.0 | 0.05 | 22 | 15.7 | 0.12 | 8 |
| Pb | 19.6 | 0.04 | 23 | 53.0 | 0.12 | 9 | 53.3 | 0.12 | 9 | 31.2 | 0.07 | 15 | 119.9 | 0.26 | 4 |
| Ni | 38.0 | 0.04 | 26 | 36.8 | 0.04 | 26 | 27.6 | 0.03 | 35 | 30.3 | 0.03 | 32 | 53.0 | 0.05 | 18 |
| Cu | 423.3 | 0.22 | 5 | 164.2 | 0.08 | 12 | 351.0 | 0.18 | 6 | 588.3 | 0.30 | 3 | 637.8 | 0.32 | 3 |
| Zn | 371.1 | 0.06 | 15 | 655.8 | 0.11 | 9 | 5651.0 | 0.98 | 1 | 3270.0 | 0.57 | 2 | 1217.0 | 0.21 | 5 |

mg/kg—milligrams per kilogram; kg/ha/year—kilograms per hectare per year. * Annual applications, not considering losses due to leaching, surface runoff, and crop absorption (see a detailed description in Section 2.4). Assumed soil density: 1.5 g/cm$^3$; soil depth: 0.2 m.

## 3.4 DISCUSSION

This study explored the state of the art regarding the presence of potentially toxic elements in organic and mineral fertilizers, encompassing information on publications, keywords, and relationships among authors, countries, and affiliations (bibliometric analyses). Moreover, quantitative information on the occurrence and concentration of these elements in fertilizers was also analyzed (meta-analysis). Overall, the results from the bibliometric analysis indicate two key findings: Firstly, the research frontier on this topic is in China, which concentrates the majority of publications, authors, and affiliations involved. Secondly, fertilizers can be a relevant source of PTEs, potentially impacting soil organisms, in some cases even in the short-term.

The conceptual structure of article keywords in the bibliometric analysis revealed that, among the most cited papers, various organic materials are used either as a source of PTEs [76,77] or as raw materials for biochar synthesis [78,79]. These studies focus on agricultural soils, particularly in relation to metal toxicity and on advancing remediation techniques in agricultural contexts [80,81]. There is also a strong influence of research related to mechanisms for the removal and sorption of metals in water or wastewater treatment [82,83].

Another way to explore the relationships among these keywords is through a thematic co-word map, which delineates clusters of keywords, using density and centrality to classify themes and map them in a two-dimensional diagram [84]. The results indicate that the metals cadmium, zinc, and lead, along with the concepts of accumulation and re-mediation, are motor themes, being the most developed and relevant within the domain. In turn, the terms contamination, pollution, risk assessment, sediments, and China represent emerging or declining themes, meaning that they are not yet fully developed or are only marginally relevant. Basic themes include heavy metals, copper, waste, biomass, and mechanisms, which reflect significant values for the domain and are transversal across its different areas. Niche themes, corresponding to themes that are highly developed but marginal to the domain, include activated carbon, biosorption, ions, and aqueous solutions.

The meta-analysis (based on 115 data lines) showed that some metals, such as Zn (n = 91), Cu (n = 89), Cd (n = 88), and Pb (n = 83), were more investigated than others, such as Co (n = 6), Hg (n = 9), Se (n = 2), and Mo (n = 2). Farid et al. (2025) [63] pointed out that, for organic fertilizers, Hg, Se, beryllium (Be), antimony (Sb), silver (Ag), and thallium (Tl) are under-investigated metals. Moreover, in the case of Hg and Se, besides being poorly

investigated, they presented high RQ, while for Sb, Ag, and Tl, risks are not clearly defined due to the limited number of available data [63].

China is dominant in this field, which is evidenced by its leading position in the number of published articles, the most productive authors, citations, co-citations, and international collaborations. This may be related to relevant metal pollution that the country has been facing in recent years [85]. For example, local surveys reported that 16% of soil samples and 19% of agricultural Chinese soils are contaminated with inorganic and/or organic pollutants, with 82.4% of the contamination attributed to metals and metalloids and 17.6% to organic contaminants [86]. This contamination poses severe risks to food security and public health, leading to the emergence of so-called “cancer villages” in some regions of China, such as Jiangsu and Guangdong provinces, where cancer morbidity rates are significantly higher than the national average. Metal contamination is directly linked to agricultural and mining activities [20].

Phosphate fertilizers are known to be a relevant source of PTEs [87], which is consistent with our meta-analysis results. Phosphate rocks are the main global source of phosphate fertilizers and occur in the Earth’s crust as igneous, metamorphic, and sedimentary deposits [88]. The geographical origin and geological characteristics of these deposits influence the concentrations of PTEs, which may also be introduced as impurities from acids used during fertilizer manufacturing [89]. In general, rock phosphates, especially those derived from sedimentary rock, contain higher Cd concentrations than other fertilizers, such as diammonium phosphate (DAP) or nitrophosphate (NP) [88]. The meta-analysis showed that phosphate rock mean Cd (74.2 mg/kg) and Zn (536.5 mg/kg) concentrations were substantially higher than any other mineral source. Moreover, the mean Cd concentration in phosphate rock was up to 700-fold higher than that of other phosphate fertilizers, while the Zn concentration was nearly 30-fold higher (**Table 1. 1**).

Lucena et al. (2025) [39] reported mean Cu concentrations of 360.08 mg/kg in super-phosphates, which is one hundred times higher than Cu in the phosphate fertilizers reported here (3.6 mg/kg). Furthermore, the authors reported a Zn concentration of 103 mg/kg in natural phosphate, a concentration much below that found in our phosphate rock (536.5 mg/kg). The authors’ meta-analysis is much different from ours, since it focuses on a very limited region (northeastern Brazil), considering fewer metals (only Cu, Zn, Fe, Mn, and Ni). Additionally, only seven sources were evaluated (limestone, agricultural gypsum, organomineral fertilizers, superphosphates, mixed fertilizers, natural phosphate, and magnesium oxides).

The organic fertilizers use can alter the soil physicochemical properties due to the addition of organic matter (OM increasing soil concentrations of N, P, K, Cu, and Zn [90,91]. Moreover, it enhances soil microbial activity and diversity, also influencing the bioavailability of metals in soils, such as Cd [92]. Because OM contains a large number of functional groups and surface charges, it can adsorb metals onto its surface and immobilize them [93], reducing their interaction with plants and animals. However, it is important to note that OM can also enhance the PTEs transport, mainly associated with dissolved organic carbon fractions [92], making monitoring of other soil properties crucial, particularly pH [94]. Under acidic conditions, the solubility of cationic metals increases, enhancing their bioavailability and expected toxicity [84].

Long-term application of organic fertilizers, such as cattle [84] and swine manure [19], may lead to the PTEs accumulation in soils [19,95]. Duan and Feng (2021) [19], when analyzing PTEs in manures, observed that swine manure (588 mg/kg) had copper concentrations 14 times higher than cattle manure (41 mg/kg). Similarly, Nookabkaew et al. (2016) [96] reported higher average Cu concentrations in swine manure (410 mg/kg) compared with cattle manure (18 mg/kg). These results are consistent with our meta-analysis, where swine manure (187.7 mg/kg) also had higher concentrations of Cu than cattle manure (57.8 mg/kg) and chicken manure (72.9 mg/kg).

High copper and zinc concentrations in manure are directly related to animal diets and mineral supplementation, as these elements are added to improve animal performance and provide protection against bacterial infections [97]. Consequently, the most effective strategy to reduce soil accumulation of these elements from manure, especially swine manure, may be lowering Cu and Zn contents through dietary management [98]. However, this approach may face resistance from the production sector, requiring reformulation of animal supplementation practices, such as the adoption of amino acid–chelated minerals as alternatives to inorganic mineral sources [99]. Chicken manure can also contain PTEs, mainly influenced by the premixed feed used in poultry feeding [100]. This highlights the importance of rigorous quality control of the raw materials used in the manufacture of animal feed [101].

In practice, untreated manure can represent a relevant fraction of the total amount of organic fertilizer applied to the soil. However, in most countries, such as in Brazil, USA and European Union, raw manure is not directly regulated for metal content, [63] which represents a major regulatory gap [63]. However, in the absence of specific legislation for metal content in raw manure, regulations available for processed organic fertilizers can serve as an approximate reference [63]. The European Union has reduced the limits of Cu and Zn in pig

feed and banned pharmacological doses of zinc oxide [102]. Considering that slightly more than 40% of the data included in this meta-analysis is from China, concerns related to Cu and Zn concentrations surpassing the European legislative limits may be overestimated for countries outside Europe with less restrictive regulations [103].

Duan and Feng (2021) [19] reported significantly higher Cr concentrations in chicken manure (153.66 ± 176.95 mg/kg) compared to swine (21.86 ± 136.62 mg/kg) and cattle (24.47 ± 34.57 mg/kg) manure, while they were similar when compared to sewage sludge (180.51 ± 35.45 mg/kg). Our results showed a similar trend, except for Cr in sewage sludge, which in our work was almost three times smaller (59.1 mg/kg) than chicken manure (170.5 mg/kg). Chromium mainly exists in two oxidation states: Cr(III) and Cr(VI). The hexavalent form is more toxic and mobile in water than the trivalent form, being classified as carcinogenic to humans [104]. Legislative frameworks are often criticized for failing to distinguish between chromium oxidation states, commonly treating both Cr(III) and Cr(VI) as equally toxic. In contrast, regulations in countries such as the United States, Brazil, and the European Union already consider limit values in organic fertilizers expressed specifically as Cr(VI) [105].

In general, for all fertilizers, European legislation is more restrictive than U.S. and Brazilian legislation (Table S1.3). For sewage sludge, a distinctive feature of Brazilian legislation is the inclusion of metals such as barium (Ba) and Cr (absent in U.S. legislation), as well as As, Cr, molybdenum (Mo), selenium (Se), and Ba (absent in European legislation). On the other hand, unlike the other legislations, the Brazilian one does not regulate Cu and Zn in manure and compost.

Like Cu, Zn, and Cr, Ni can also be present in industrial animal feed, where it is used as a catalyst in the production of specific ingredients [106]. In the meta-analysis, the highest Ni concentration were found in sewage sludge (30.5 mg/kg), cattle manure (17.1 mg/kg), swine manure (14.1 mg/kg), and potassium fertilizer (12.9 mg/kg). These results are consistent with data from the European Food Safety Authority (EFSA), which analyzed 2198 animal feed samples for Ni content and concluded that the category with the highest mean levels was “Minerals and their derived products” (n = 72; 3905 µg/kg), followed by “Compound feed” (n = 516), particularly complementary feeds for cattle fattening (n = 26; 6813 µg/kg), unspecified complementary feeds (n = 9; 5270 µg/kg), and complementary feeds for swine fattening (n = 6; 4344 µg/kg). This report also highlights that sewage sludge, widely used in agriculture, may contain appreciable levels of Ni, representing an additional source of exposure [106].

For As, organic compost was the source with the highest concentration (37.8 mg/kg) and the greatest variation (1.9–63.7 mg/kg) (**Table 1. 1**). A key aspect of organic fertilizers is

their inherent variability in physicochemical composition, which is influenced by numerous factors, such as season, location, animal diet, climate, and management practices [107]. For organic composts, numerous combinations of plant and animal components are possible, such as swine manure + cotton plant residues [29], swine manure + cereal straw [29], cattle manure + straw [108], and coffee pulp + wastewater [109]. As indicated by the bibliometric analysis, China accounts for a large proportion of the articles included in this meta-analysis. Due to the country's large cereal production, particularly rice, composting with straw is common [30,108]. Several studies conducted in China reported significant arsenic uptake by rice [110,111], which may explain the high As values found in organic composts (**Table 1. 1**, **Figure 1. 6**).

For lead, none of the fertilizers exceeded the legal limits of all legislations, indicating that fertilization is not a relevant source of soil lead contamination. Vehicle emissions, especially in soils near highways, and industrial activities are more important sources of Pb in the environment [18,112]. Nevertheless, even within regulatory limits, our data show that Pb may still cause negative impacts, with estimated application times of 40 and 4 years to achieve the PNEC, considering the most likely low and worst-case-high scenarios, respectively (**Table 1. 2Table 1. 3**).

When comparing organic and mineral sources, the results observed here are consistent with previous studies, such as a study comparing swine manure and mineral fertilizer (urea + triple superphosphate + potassium chloride) [113], where the highest Cu (285.9 ± 154 mg/kg) and Zn (455.6 ± 210 mg/kg) values were found in organic manure, whereas mineral fertilizers exhibited the lowest concentrations (29.3 and 158.5 mg/kg). On the other hand, different from our results, the mineral fertilizer analyzed by these authors contained higher concentrations of As, Cr, and Ni (8, 124, and 21 mg/kg) than the organic sources.

Concerning risk assessment, in the most likely scenario (**Table 1. 2**), only organic sources (chicken manure, compost, swine manure, and sewage sludge) presented moderate risk to soil organisms. Among these sources, chicken manure stood out, with an estimated application interval until the PNEC is achieved of only 6 years (RQ = 0.18) under a low fertilization rate (5 ton/ha). Under a high fertilization rate (20 ton/ha), safety limits would be achieved in the first year of application, with an RQ = 0.72. Among the mineral sources, only phosphate rock presented a low risk (for Cd, RQ = 0.02), with an estimated application time of 57 years.

In the worst-case scenario (**Table 1. 3**), chicken manure showed the highest RQ for Cr, with applications limited to 2 years (Moderate RQ = 0.44) under a low rate and only 1 year (High RQ = 1.78) under a high rate. In the worst-case scenario with a high rate, attention is also

drawn to Cr in cattle manure (RQ = 0.68; 1 year of application), compost (RQ = 0.53; 2 years of application), and sewage sludge (RQ = 0.77; 1 year of application). Another PTE that stood out was Zn, mainly in compost (RQ = 0.98; 1 year of application) and swine manure (RQ = 0.57; 2 years of application). Mineral sources (limestone and phosphate rock) presented only low risk in the worst-case scenario. Similarly, a study focused exclusively on animal manures (cattle, chicken, pig, and sheep) also reported RQ > 1 for Zn, Cu, As, Hg, and Se, thereby suggesting relevant ecological risks [63].

Considering the worst-case scenario under a high fertilization rate, sewage sludge (Cr = 0.77; Cu = 0.32; Pb = 0.26; Zn = 0.21; As = 0.12; Cd = 0.10) and compost (Zn = 0.98; Cr = 0.53; As = 0.49; Cu = 0.18; Pb = 0.12) stood out from the other organic sources due to the number of situations with moderate risk, indicating a multi-contamination scenario in the long-term (18 and 35 years, respectively). It is also important to highlight that sewage sludge is a relevant source of other pollutants, such as endocrine-disrupting compounds, persistent organic pollutants, microplastics, pharmaceuticals, personal care products, and pathogens [114,115]. These pollutants can accumulate in the soil or be transported to water bodies, entering the food chain and increasing risks to human health and the ecosystem [116]. Composting itself is a low-cost technology for reducing or eliminating pathogens and organic pollutants in sewage sludge, such as polycyclic aromatic hydrocarbons and antibiotic resistance genes [117,118]. However, it has little effect on PTE concentrations, as these are persistent inorganic compounds that are not biologically degraded [118].

A relevant drawback of this work is that it does not account for the dynamics of PTEs in soils and the metal losses that may occur due to different processes such as crop uptake and removal, surface runoff, and leaching [97], all of which directly affect the soil metal balance. Additionally, soil metal contamination may occur through other pathways such as atmospheric deposition and pesticide use [86,93]. However, it should be highlighted that considering metal losses will not necessarily produce relevant changes in the estimated time for metal accumulation in the soil. Taking the swine manure application as an example, with a Zn average concentration of 1182.5 mg/kg, applied to a soil with a density of 1.5 g/cm$^3$, considering a 0.2 m depth layer and a rate of 5 ton/ha, this would result in a total Zn input of 5912.5 g/ha. Considering losses due to crop harvest equal to 342 g/ha and by leaching of 39 g/ha [119], this would result in a net annual accumulation of 5531.5 g/ha. Considering the PNEC for Zn adopted in this study (38 mg/kg), this would result in an estimated maximum application time of 21 years, which is not much different than our 19-year estimate (**Table 1. 2**).

The risk assessment illustrated that, in many cases, safe thresholds could be reached within only a few years, underscoring the importance of material characterization, application rate adjustments, and derivation of safe limits to ensure sustainable and safe fertilizer use, particularly over the long term [9]. It is also crucial to recognize that natural concentrations of metals in soils can vary widely, depending on weathering processes, soil mineralogy, and volcanic activity [22]. This is a critical aspect that can strongly influence the time required to reach PNEC values. For example, in a study conducted in the state of Paraná (Brazil), natural concentrations of Cu (103.8 mg/kg) and Sb (3.7 mg/kg) already exceeded the reference values for soil quality protection [120] of 60 and 2 mg/kg, respectively. The authors emphasized the importance of the basaltic rock of this region for the high Cu and Sb natural levels [121].

For manure application in China, Zhang et al. (2026) [103] estimated that the concentration limits of Chinese legislation would be achieved in 15, 18, and 14 years for Cd, Zn, and Cu, respectively. Sun et al. (2023) [122] conducted a field monitoring of PTE accumulation in agricultural soils following continuous application of organic fertilizers, observing distinct dynamics among the analyzed metals (Cu, Zn, As, and Pb) and cropping systems (rice paddy, orchards, and vegetable garden) over a period of up to eight years. In that study, manure application significantly influenced the accumulation of Cu, Zn, and As in soils, whereas its impact on Pb accumulation was relatively minor.

Taken together, our results evidenced relevant ecological risks associated with long-term fertilizer application, especially from organic sources. These findings evidence the urgency of fertilizer management practices, the need for improved animal feed regulation, and the need for rigorous quality control of organic fertilizers. Therefore, medium- and long-term monitoring of the impacts of fertilizer application on soil PTE concentrations is essential for ecosystem sustainability [91,92,123].

## 3.5 CONCLUSIONS

Bibliometric analyses indicated an annual growth of 11.89% in publications on the topic between 1982 and 2024, driven mainly by China, Pakistan, the United States, and Brazil. Research has focused mainly on cadmium, the use of biochar, and aspects related to the bioavailability of PTEs in soils. Elements such as mercury, manganese, cobalt, and selenium have received little attention compared to cadmium, arsenic, lead, zinc and copper.

The comparison between mineral and organic sources showed that organic sources presented higher concentrations of copper, chromium, arsenic, lead, and zinc, while no differences were found for cadmium and nickel. Concentrations of zinc and copper were especially high in swine manure, with maximum concentrations of 1182 mg/kg for Zn and 588 mg/kg for Cu, respectively. Although Cu and Zn in organic fertilizers are not regulated by Brazilian legislation (IN SDA No. 27, 5 June 2006), it includes Cr and As, which are not regulated in European legislation for sewage sludge. Furthermore, some metals (such as hexavalent chromium and mercury), although included in European and Brazilian regulatory frameworks, have been poorly investigated in the scientific literature, highlighting the relevance of their inclusion in future research.

Organic sources showed higher RQ than mineral ones, representing greater risks to soil organisms. For chicken manure, even at low fertilization doses, a moderate risk for Cr was seen, with applications lasting only six years until reaching the PNEC. Although in some limited cases even short-term applications could already reach the PNEC values, for most sources, this is more likely to occur associated with medium- to long-term applications. Nevertheless, our results clearly show that fertilizers, especially organic ones, can be a relevant source of PTEs in soils.

Overall, our findings highlight the need for: (i) further studies assessing the impact of fertilizer application on soil biota under local edaphoclimatic conditions, especially in long-term application scenarios; (ii) more research to derive reference values for soil quality protection using ecotoxicological tests with native organisms, which is especially important for tropical soils, which actually show less available data; (iii) more stringent feed formulation policies; (iv) specific legislation for monitoring PTEs in raw manure; and (v) fertilization management practices that consider the pollution footprint of fertilizers. Altogether, these efforts constitute strategic means for advancing into the direction of a more sustainable agriculture.

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# 4 CHAPTER II – Potentially Toxic Elements in Fertilizers and Soil Amendments of the Brazilian Cerrado


## RESUMO

A poluição do solo por elementos potencialmente tóxicos (EPTs) é uma preocupação global, ameaçando a biota e a saúde do solo e a segurança alimentar. Áreas agrícolas do Cerrado brasileiro, a savana tropical com maior biodiversidade do mundo, dependem fortemente do uso de fertilizantes minerais e orgânicos, que podem ser uma fonte relevante de EPTs. Este estudo avaliou o teor de As, Co, Cd, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Se e Zn em fertilizantes minerais, orgânicos e organominerais comercializados no Cerrado. Os teores totais foram determinados utilizando o método de extração US-EPA 3051A, e os teores disponíveis em água utilizando uma proporção de 1 g de amostra para 100 mL de água, ambos quantificados por espectroscopia de emissão óptica com plasma indutivamente acoplado. Os riscos à biota do solo foram estimados por meio de uma abordagem de quociente de risco utilizando taxas de aplicação empregadas na região. Os limites da legislação brasileira foram excedidos para arsênio (82,3 vs. 40 mg/kg) no superfosfato simples e para chumbo no FTE-BR12 (11.553 vs. 10.000 mg/kg). Os maiores quocientes de risco foram encontrados no esterco bovino (cromo: 1,173; níquel: 0,456) e na torta de filtro de cana-de-açúcar (cromo: 0,484; níquel: 0,342). A análise de componentes principais mostrou que os quocientes de risco para molibdênio, cromo, cobre, níquel e zinco estavam mais intimamente relacionados aos fertilizantes orgânicos e organominerais, enquanto os de arsênio, cádmio e chumbo estavam mais associados aos fertilizantes minerais. Os resultados evidenciaram forte influência de altas taxas de aplicação de fertilizantes orgânicos sobre os riscos projetados à biota do solo, ressaltando a necessidade de atualizar a legislação brasileira, especialmente por meio da adoção de limites para Cu, Mo e Zn.


**Palavras-chave:** Biodisponibilidade. Metais Pesados. PNEC. Resíduos Pecuários. Sustentabilidade agrícola.

**ABSTRACT**

Soil pollution by potentially toxic elements (PTEs) is a global issue, threatening soil biota, soil health, and food security. Agricultural areas of the Brazilian Cerrado biome, the most biodiverse tropical savanna in the world, heavily rely on the use of mineral and organic fertilizers, which may be a relevant source of PTEs. This study evaluated the content of As, Co, Cd, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Se and Zn in mineral, organic and organomineral fertilizers marketed in the Cerrado biome. Total contents were determined using the US-EPA 3051A extraction method, and water-available contents using a 1 g sample to 100 mL water ratio, both quantified by inductively coupled plasma optical emission spectroscopy. Risks to soil biota were estimated through a risk quotient approach using application rates employed in the region. Brazilian legislation limits were exceeded for arsenic (82.3 vs. 40 mg/kg) in single superphosphate and for lead in FTE-BR12 (11,553 vs. 10,000 mg/kg). The highest risk quotients were found in cattle manure (chromium: 1.173; nickel: 0.456) and sugarcane filter cake (chromium: 0.484; nickel: 0.342). Principal component analysis showed that risk quotients for molybdenum, chromium, copper, nickel, and zinc were more closely related to organic and organomineral fertilizers, whereas those for arsenic, cadmium, and lead were more associated with mineral fertilizers. The results evidenced a strong influence of high application rates of organic fertilizers on the projected risks to soil biota, underscoring the need to update Brazilian legislation, especially through the adoption of limits for Cu, Mo, and Zn.

**Keywords:** agricultural sustainability. Bioavailability. Heavy metals. Livestock waste. PNEC.

## 4.1 INTRODUCTION

About 14 to 17% of the global agricultural area is affected by potentially toxic elements (PTEs) pollution, placing more than 1.4 billion people in high-risk areas (Hou et al., 2025). Soil pollution by PTEs may adversely affect soil quality by reducing the edaphic biota biodiversity and ecosystem stability (Podlasek et al., 2024). The origin of these PTEs in soil can be natural, from rock weathering, soil leaching, and volcanic eruptions (Hu et al., 2024a), or anthropogenic, from mining (Adnan et al., 2024), industrial emissions (Ahamad et al., 2021), and agricultural activities, including the use of pesticides and fertilizers (Rodrigues et al., 2026; Alengebawy et al., 2021). In general, soil contamination by anthropogenic sources is more relevant than natural ones, since they can enrich soils with PTEs over a relatively short period (Kim et al., 2026; Trujillo-González et al., 2022). This contamination scenario is likely to worsen in the coming decades, driven by the projected population growth to 9.4–10.1 billion by 2050 and the consequent increase in food demand and agro-industrial residues generation and agricultural disposal (Nogueira et al., 2026; Giller et al., 2021; Gu et al., 2021).

Brazilian agribusiness represents about 25% of the country's Gross Domestic Product (GDP), with a diversified production of grains and livestock products (CEPEA, 2022). In 2025, the country's Gross Production Value (GPV) for crop and livestock production reached approximately U$ 7.3 trillion, with 34% originating from livestock and 66% from crops (Brasil, 2026). The agriculture practiced in the Cerrado biome accounts for about 60% of the country's agricultural production, representing 22% of soybean production and 23% of sugarcane production (Hurd and Silva, 2024). In 2023, the Midwest region accounted for an inventory of 77 million head of cattle, 6 million head of swine, and 38 million of chickens. Compared to 2003, these livestock populations increased by 9.7%, 56.2%, and 121.0%, respectively (IBGE, 2024; IBGE, 2004). However, this agricultural intensity contrasts with the low natural fertility of the locally predominant highly weathered soils, characterized by high acidity and low plant nutrients availability (Lima et al., 2022).

Mineral and organic fertilizers may contain PTEs in their composition due to their origin, the raw materials used, or insufficient purification in the manufacturing process (Guilherme et al., 2020; Duan and Feng, 2021; Shan et al., 2024; Sachdeva et al., 2025; Su et al., 2025). Thus, high concentrations of arsenic (As), cadmium (Cd), copper (Cu), zinc (Zn), nickel (Ni), chromium (Cr), lead (Pb), and mercury (Hg) can be found in fertilizers (Rashid et al., 2023). PTEs concentrations in fertilizers can exceed the reference values, with legal limits being surpassed by 61.4% for Cd in Biosolids and Urban Organic Composts samples, 66.6%

for Cu in swine manure samples, 21.8% for Cd in chicken and duck manure samples, 10% for Hg in cattle manure, 24.7% for Ni in phosphate fertilizer samples, and 22% for Ni in rock phosphate powder samples (Sachdeva et al., 2025).

Accordingly, several studies show that fertilization enriches soils with these elements (Gao et al., 2021; Cabral et al., 2023; Osti et al., 2025). In 38 years of experiment about the fertilization effect on PTE accumulation in soils, Gao et al. (2021) observed that chemical fertilizer and cattle manure application led to an increase from 0.12 to 0.37 mg/kg in total Cd and from 20.4 to 35.5 mg/kg in total As, respectively. This pattern is also expected in the Cerrado agricultural soils, since high agricultural productivity demands intensive agricultural practices, such as the continuous use of fertilizers, potentially adding PTEs to soils (Cabral et al., 2023; Osti et al., 2025). Osti et al. (2025) evaluated areas subjected to ten years of pig manure application in the Cerrado biome, reporting an increase in Zn concentration from 13.7 to 67 mg/kg, from 15.6 to 24.1 mg/kg for Cu, and from 3.5 to 4.9 mg/kg for Co (control and treated areas, respectively).

One way to estimate the impact of PTE enrichment in soil resulting from fertilizer application is to compare it with a reference concentration, which may range from limits established in soil quality legislation (Brasil, 2009) to a predicted no-effect concentration (PNEC) for a given group of organisms (Xu et al., 2019; Wan et al., 2020). In this regard, Rocha et al. (2025) observed moderate ecotoxicological risk quotients (RQ) related to fertilization practices for Cd (0.21), Cu (0.23), Ni (0.49), Pb (0.10), Zn (0.56), Hg (0.43), and low risk for Cr (0.04), and As (0.006). However, for the Cerrado biome, studies estimating the risks to soil biota or human health arising from soil enrichment with PTEs from fertilizer application are still scarce.

In this context, this study evaluated the PTEs contents (As, Co, Cd, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Se, Zn) in major fertilizers marketed in the Brazilian Cerrado region. The following hypotheses were tested: i) PTE contents in fertilizers marketed in the Brazilian Cerrado are within Brazilian legal limits; ii) there are relevant differences in PTE concentrations among fertilizer sources (organic, mineral, and organomineral); and iii) the risks to soil biota posed by PTEs in fertilizers are negligible. To address these hypotheses, principal component analysis (PCA) and risk analysis (risk quotient – RQ), based on predicted no-effect concentration (PNEC) values for soil organisms, were used. Ultimately, this effort aims to sustainably optimize the health of ecosystems, animals, and people, as all are closely interconnected and interdependent.

## 4.2 MATERIALS AND METHODS

### *4.2.1 Sampling and sample storage*

The samples were all collected throughout the second half of 2025, at representative production areas with extensive use of fertilizers and soil amendments, all within the Cerrado region. A total of five samples of organic fertilizers, six organomineral fertilizers, and ten of mineral fertilizers were collected, with each sample representing a distinct material. Due to the wide diversity of organic, organomineral, and mineral fertilizers used in the region, sampling effort prioritized the largest possible number of distinct inputs, rather than multiple replicates of a single input.

The selection of the sampled materials reflects the most representative fertilizers used in the Cerrado region, which plays a fundamental role in agricultural expansion, cattle ranching, swine and poultry production (Santana et al., 2020; Santos et al., 2024), as well as in the sugarcane industry (Bufon et al., 2021). Among the most widely employed mineral fertilizers, phosphate and micronutrient sources deserve particular attention, as they have been frequently associated with elevated concentrations of potentially toxic elements in the scientific literature (Sachdeva et al., 2025; Vale and Alcarde, 2003).

Solid animal manures, sugarcane filter cake, and organomineral fertilizers I–IV were collected from piles arranged in the field, with sampling performed uniformly across the three main sections of each pile (top, middle, and base). In each section, approximately ten aliquots were collected at equidistant points using a shovel inserted into the pile at an inclined angle (Andrade and Abreu, 2006). The aliquots were homogenized in a previously cleaned bucket and subsequently packed into a plastic bag. Organomineral V and the mineral fertilizers were sampled using a probe-type sampler inserted into the valve bags stored in the companies' warehouses. The resulting sub-samples were homogenized in a previously cleaned bucket, from which an aliquot of approximately 500 g was withdrawn and packed into a sealed plastic bag.

Vinasse was collected at a sugar and ethanol mill, directly from the impermeable storage tank used to supply the trucks responsible for its field application. Liquid swine manure was collected from a settling pond at an intensive swine-rearing facility with the aid of a motor pump and suction equipment, collecting three sub-samples one minute apart. In both cases, a 5 L plastic container was used, being previously cleaned and rinsed with the sample itself. For organomineral VI, two 1 L sub-samples were collected in a plastic container from a previously homogenized 20 L commercial package. After collection, all containers holding liquid samples

were properly sealed and transported under refrigeration to the laboratory, in an insulated cooler with ice. **Table 2. 1** presents more details regarding the samples collected.

**Table 2. 1** Studied fertilizers, with their respective codes and description of their composition physico-chemical and fertility attributes.

| Code | Fertilizer | Composition Description |
|---|---|---|
| OFC | Sugarcane Filter Cake | Solid organic fertilizer, byproduct of sugarcane sugar and ethanol production. |
| OV | Vinasse | Liquid organic fertilizer, byproduct of sugarcane sugar and ethanol production. |
| OPS | Pig slurry | Liquid organic fertilizer derived from intensive swine farming. |
| OCM | Cattle Manure | Solid organic fertilizer, byproduct of intensive dairy cattle farming. |
| OPL | Poultry Litter | Solid organic fertilizer, byproduct of intensive broiler chicken farming. Composed of bedding material plus animal feces. |
| OMI | Organomineral I | Solid organomineral fertilizer. Composition: Compost barn litter, poultry litter, cattle manure, feedlot manure, and Glauconitic Siltstone. Undefined proportion. |
| OMII | Organomineral II | Solid organomineral fertilizer. Composition: Feedlot manure 56%, poultry litter 24%, and Mica Schist 20%. |
| OMIII | Organomineral III | Solid organomineral fertilizer. Composition: Separator cattle manure 75%, Glauconitic Siltstone 12.5%, and Reactive Natural Phosphate 12.5%. |
| OMIV | Organomineral IV | Solid organomineral fertilizer. Composition: Poultry litter 50% + Glauconitic Siltstone 50%. |
| OMV | Organomineral V | Solid commercial organomineral fertilizer. |
| OMVI | Organomineral VI | Liquid organomineral fertilizer, composed of peat, potassium hydroxide, phosphoric acid, water, and monoammonium phosphate. |
| MSSPI | SSP 00-19 | Solid mineral fertilizer (single superphosphate). |
| MSSPII | SSP 00-21 | Solid mineral fertilizer (single superphosphate). |
| MNPKI | NPK 04-14-08 | Solid ternary mineral fertilizer (04 Nitrogen – 14 Phosphorus – 08 Potassium). |
| MNPKII | NPK 05-25-15 | Solid ternary mineral fertilizer (05 Nitrogen – 25 Phosphorus – 15 Potassium). |
| MNPKII | NPK 06-30-10 | Solid ternary mineral fertilizer (06 Nitrogen – 30 Phosphorus – 10 Potassium). |
| MMAP | MAP 11-52 | Solid mineral fertilizer (monoammonium phosphate). |
| MTSP | TSP | Solid mineral fertilizer (Triple Superphosphate). |
| MFTE | FTE BR-12 | Solid mineral fertilizer (Micronutrient Frits). |
| MLI | Limestone I | Solid soil amendment ($CaMg(CO_3)_2$). |

| | | |
|---|---|---|
| MLII | Limestone II | Solid soil amendment ($CaMg(CO_3)_2$). |

The solid organic samples were dried in an oven at 65 ± 5 ºC until constant weight, being stored in plastic bags (Brasil, 2024). The liquid samples were frozen within the same day in which they were collected and stored frozen until be analyzed, being transported in a cooler box with ice. The sub-samples collected represented an effort to minimize the influence of local sample heterogeneity. This sampling strategy therefore enables comparisons of PTEs variability restricted to the specific samples analyzed, rather than representing the full range of variability inherent to each fertilizer group.

*4.2.2 Chemical analyses for obtaining total and water-available contents*

In this study, the term "total contents" refer to the extraction of elements by the US-EPA 3051A method (USEPA, 2007), which represents the contents that may be environmentally available, while "available contents" refer to water-soluble contents, i.e., which are readily available to plants. The US-EPA 3051A method was performed using 0.5 g of the sample extracted with nitric acid ($HNO_3$) in a microwave oven (CEM, MARS 5) and quantified by inductively coupled plasma optical emission spectrometry (ICP-OES) with axial view (Varian ES710) and ISO 17034 certified standard stock solutions. Water-soluble contents were obtained using a 1 g sample to 100 mL water ratio (m/v), following the MAPA method (Brasil, 2024), as recommended for organic and mineral foliar fertilizers, also determined by ICP-OES. Both total and available contents were determined in triplicate. The determined elements and wavelengths used were: Co (238.892), Al (308.215), As (188.980), B (208.956), Ca (315.887), Cd (214.439), Cr (267.716), Cu (327.395), Fe (238.204), Hg (184.887), Mg (383.829), Mn (293.305), Mo (202.032), Ni (231.604), P (213.618), Pb (220.353), S (181.972), Se (196.026), Zn (213.857), K (769.897), Na (568.821).

The guarantees of all fertilizers were analyzed by the official methods of the Ministry of Agriculture and Livestock (MAPA) (Brasil, 2024). The pH was determined in a calcium chloride solution ($CaCl_2$). Cation exchange capacity was determined by standardized Sodium hydroxide (NaOH) titration, using phenolphthalein indicator. Organic carbon content was obtained by the potassium dichromate ($K_2Cr_2O_7$) volumetric method. Water-soluble potassium was hot-extracted and quantified by flame photometry. Total and water-soluble nitrogen was determined by the Microraney method for mineral samples and by the Kjeldahl method for organic and organomineral samples. Finally, citric acid soluble at pH 7 and water-soluble

phosphorus were quantified by the Quimociac gravimetric method. All these analyses were performed in triplicate.

The chemical analyses were all performed in the Laboratory for Fertilizer and Waste Chemical Analysis of the Agronomic Institute of Campinas (IAC), which is accredited by the General Coordination for Accreditation (Cgcre) of the National Institute of Metrology, Quality and Technology (Inmetro), in accordance with the requirements of the ABNT NBR ISO/IEC 17025 standard. As part of this accreditation, regular equipment maintenance is carried out. The laboratory's accreditation registration number is CRL 0450 (INMETRO, 2024).

*4.2.3 Risk quotient analysis and reference values*

The Risk Quotient (RQ) approach was employed to estimate the risks to soil organisms represented by PTEs in fertilizers (Rodrigues et al., 2026; Farid et al., 2025). It was obtained according to Equation (1):

$$RQ = PEC/PNEC \quad (1)$$

Where PEC is the predicted environmental concentration in the soil and PNEC is the predicted no-effect concentration (safe level) in the soil. RQ is considered a robust method for assessing ecotoxicological risks and is widely used in environmental studies. According to Bhandari *et al.* (2021), RQ is classified as follows: no risk ($RQ < 0.01$), low risk ($0.01 \leq RQ < 0.1$), moderate risk ($0.1 \leq RQ < 1$), and high risk ($RQ \geq 1$).

PEC values were estimated considering a 0.2 m soil layer (L), a soil density of 1415 kg/m$^3$ (D), the average concentration of each metal (M) and the fertilizer application rates (A) used in production areas within the Cerrado region. The PEC concentration was obtained according to Equation (2):

$$PEC = (A \times M)/(D \times L \times 10^4) \quad (2)$$

The annual application rates used were: pig slurry (107.3 t/ha), cattle manure (20.8 t/ha), poultry litter (8.8 t/ha), organomineral I-IV (7.9 t/ha), FTE-BR (25 kg/ha), limestone (1.0 t/ha), NPK (350 kg/ha), vinasse (183.8 t/ha), sugarcane filter cake (17.5 t/ha), MAP (215 kg/ha), SSP (460 kg/ha), TSP (250 kg/ha), organomineral V (675 kg/ha), organomineral VI (135 kg/ha). These values were derived from the average of the optimal application rates reported in studies carried out in the Cerrado — that is, the rate that produced the most favorable crop response (when such data was available) — or from the single rate indicated by the study (when a range of values was not provided). This information, as well as the studies used as references, can be consulted in the material available on Zenodo (Rodrigues, 2026).

PNEC values were obtained by converting NOEC (no observed effect concentration) values using an assessment factor (AF) specific for each situation: AF = 100 for data including one trophic level (Cr, Mo, Ni, Pb); AF = 50 for data including two trophic levels (As, Cd); and AF = 10 for data including three trophic levels (Zn) (ChemSafetyPro, 2016). For Cu, the PNEC value was available in the literature, requiring no further conversions. Thus, the PNEC values (mg/kg) for each metal were: As (2.7), Cd (0.2), Cr (0.25), Cu (11), Mo (0.25), Pb (2), Ni (0.1), Zn (15.7).

NOEC values were obtained through a previous literature search using the keywords: ("latosol" OR "latossolos" OR "oxisol" OR "tropical soil") AND ("heavy metal" OR "Potentially Toxic Elements" OR "PTE") AND ("NOEC" OR "PNEC" OR "$EC_{10}$"). Only studies using standardized test organisms in natural Brazilian soils, preferably Oxisols and Latosols, were considered. The results can be viewed in the Zenodo repository (Rodrigues, 2026).

Brazilian natural soils are described as having low organic matter content (1.5–3.0%), acidic pH (4.8–5.6), and mineralogical compositions dominated by kaolinite, iron oxides, and gibbsite (Hennig *et al.*, 2026). These properties are substantially different from those of temperate region soils or artificial soils designed to simulate tropical conditions (e.g., TAS) (Hennig *et al.*, 2026), indicating that natural tropical soils offer a more realistic substrate for ecotoxicological assays.

To estimate the years to achieve the PNEC (Y), equation 3 was used:

$$Y = PNEC/PEC \quad (3)$$

It should also be stated that the risk analysis has some important limitations: it does not account for other possible sources of soil PTEs input, such as atmospheric deposition, mining activities, and industrial effluents, nor it considers the natural background concentrations of these elements in the soil (Adnan *et al.*, 2024; Hou *et al.*, 2025; Jomova *et al.*, 2025). These factors could all contribute to an underestimation of the calculated risks. It also does not consider potential losses that may occur due to different processes, such as leaching, soil erosion, surface runoff, and uptake by crops (Ali *et al.*, 2019; Hou *et al.*, 2025), a factor that can contribute to an overestimation of the projected risks.

### *4.2.4 Statistical and multivariate data analysis*

Total contents, the application rates, and RQs were subjected to the Shapiro-Wilk normality test, which showed that none of the variables followed a normal distribution, leading to data transformation using the log(1+x) method. Subsequently, principal component analysis

(PCA) was performed to visualize the relationship between fertilizer sources, total contents (Cr, Cu, Mo, Ni, Pb, and Zn), RQs (As_RQ, Cd_RQ, Cr_RQ, Mo_RQ, Ni_RQ, Pb_RQ, and Zn_RQ), and the application rates. The variables were previously selected to satisfy the Bartlett test ($p < 0.05$) and the KMO (Kaiser–Meyer–Olkin) test (Overall MSA > 0.6), which indicate that the data are suitable for inclusion in the PCA. The quality of representation was assessed by $cos^2$ values. For a similarity analysis among the materials, k-means clustering algorithm and a dendrogram using Euclidean distance were used. All analyses were performed using R software version 4.3.2 (R Core Team, 2024) and the packages "stats", "factoextra", "ggplot2", "cowplot", "dplyr", "tibble", “psych”, and "corrplot".

### *4.2.5 Legislation consulted for comparison of PTE contents in fertilizers*

Normative Instruction of the Secretariat of Agricultural Defense (SDA) No. 27 of June 5, 2006 (Brazil, 2006), amended by Normative Instruction SDA No. 7 of April 12, 2016 (Brazil, 2016) of the Republic of Brazil, was used to compare the PTEs total content in organic, organomineral, and mineral fertilizers. Since the legislation for organic fertilizers is more restrictive, the concentrations found in the organomineral fertilizers were compared with the limits applicable to organic fertilizers. The specifications of minimum nutrient guarantees for mineral fertilizers were consulted through Normative Instruction No. 39 of August 8, 2018 (Brazil, 2018).

### *4.2.6 Use of Artificial Intelligence*

For the search of topics and articles, the artificial intelligence tools LeapSpace (Elsevier, 2026) and Consensus (Consensus, 2026) were used. For translation, grammatical correction, and initial adjustments to clarity and cohesion, the generative artificial intelligence tools Claude Sonnet 5 (Anthropic, 2026) and DeepSeek-V4 (DeepSeek, 2026) were used. The text was subsequently reviewed by human reviewers fluent in English.

## 4.3 RESULTS

### *4.3.1 Total and available concentrations of PTEs*

The guarantees provided by the companies and the physico-chemical parameters analyzed in the fertilizers are detailed in **Table 2. 2**. In all cases the nutrients content where within the specifications for mineral fertilizers established by Normative Instruction Nº. 39 of August 8, 2018 (Brasil, 2018): MAP = 9% N; 48% $P_2O_5$; SSP = 18% $P_2O_5$; TSP = 41% $P_2O_5$; NPK = sum of primary macronutrients (% by weight).

The total and available concentrations of PTEs for the sampled fertilizers can be seen in **Table 2. 3 Table 2. 4**. Attention is drawn to As concentration in SSP II (00-21) (82.3 mg/kg) and the Pb concentration in FTE-BR12 (11,553 mg/kg), which are above Brazilian legislation (45 mg/kg and 10,000 mg/kg, respectively). The remaining fertilizers complied with the limits for PTEs in their respective classes.

Regarding the PTEs water-available contents, mineral fertilizers showed higher availability compared to organic and organomineral fertilizers, with the exception of Cu, Mo, and Zn (Figure S2.1). Noteworthy were the percentage of available Ni (66.3%) among mineral fertilizers, Zn (68%) among organic fertilizers, and Mo (57%) among organomineral fertilizers (Figure S2.1). Individually, TSP was the fertilizer with the highest amount of water-available PTEs contents (58.4%), followed by SSP 00-19 (44.4%) and NPK 05-25-15 (33.7%) (Figure S2.2).

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**Table 2. 2** Guaranteed versus Analyzed Chemical Composition of Mineral, Organic, and Organo-mineral Fertilizers and Soil Amendments.

| Fertilizer | Guarantee | Analyzed |
|---|---|---|
| SSPI | $P_2O_5$ = 19% | $P_2O_5$ = 17.9%; soluble $K_2O$ = 0.04% |
| SSPII | $P_2O_5$ = 21% | $P_2O_5$ = 22.4%; soluble $K_2O$ =0.16% |
| NPKI | N = 4%; $P_2O_5$ = 14%; $K_2O$ = 8% | Total N = 6.2%; soluble N = 5.5%; $P_2O_5$ = 11.6%; soluble $K_2O$ = 7.97%. |
| NPKII | N = 5%; $P_2O_5$ = 25%; $K_2O$ = 15% | Total N = 9.3%; soluble N = 8.4%; $P_2O_5$ =27.6%; soluble $K_2O$ = 6.3% |
| NPKIII | N = 6%; $P_2O_5$ = 30%; $K_2O$ = 10% | Total N = 5.4%; soluble N = 5.1%; $P_2O_5$ = 24.7%; soluble $P_2O_5$ = 25.3%; soluble $K_2O$ = 10.8% |
| MAP | N = 11%; $P_2O_5$ = 52% | Total N = 10.1%; soluble N = 9.1%; $P_2O_5$ = 48.1%; soluble $P_2O_5$ = 42%; soluble $K_2O$ = 0.03% |
| TSP | $P_2O_5$ = 46% | Total N = 0.35%; soluble N = < 0.1 % (below the detection limit); $P_2O_5$ = 45.3%; soluble $P_2O_5$ = 41.1%; soluble $K_2O$ = 0.02% |
| MFTE | B = 1.8%; Cu = 0.8%; Mn = 2%; Zn = 9%; S = 1% | Total N = 0.25%; soluble N = 0.15%; $P_2O_5$ = 0.81%; soluble $P_2O_5$ = 0.62%; soluble $K_2O$ = 0.15%; $SO_4$ = 5.07% |
| OFC | - | $CaCl_2$ pH = 7.8; Organic C = 436.7 g/kg; Cation Exchange Capacity (CEC) = 251.3 $mmol_c$/kg; total N = 21.0 g/kg |
| OV | - | $CaCl_2$ pH = 3.5; Organic C = 12.1 g/kg; total N = 1.8 g/kg |
| OPS | - | $CaCl_2$ pH = 8.2; Organic C = 4.1 g/kg; total N = 34.4 g/kg |
| OCM | - | $CaCl_2$ pH = 7.5; Organic C = 187.7 g/kg; CEC = 332 $mmol_c$/kg; total N = 14.5 g/kg; soluble N = 9.97 g/kg |
| OPL | - | $CaCl_2$ pH = 7.5; Organic C = 380 g/kg; CEC = 715.3 $mmol_c$/kg; total N = 39.1 g/kg; soluble N = 32.6 g/kg |
| OMI | - | $CaCl_2$ pH = 7.4; Organic C = 210 g/kg; CEC = 512 $mmol_c$/kg; total N = 25.4 g/kg; soluble N = 20.8 g/kg |
| OMII | - | $CaCl_2$ pH = 7.9; Organic C = 113 g/kg; CEC = 322 $mmol_c$/kg; total N = 14.9 g/kg; soluble N = 9.2 g/kg |
| OMIII | - | $CaCl_2$ pH = 7.2; Organic C = 102.3 g/kg; CEC = 280 $mmol_c$/kg; total N = 14.6 g/kg; soluble N = 10.8 g/kg |

| | | |
|---|---|---|
| OMIV | - | $CaCl_2$ pH = 7.3; Organic C = 103.7 g/kg; CEC = 284 $mmol_c$/kg; total N = 12.2 g/kg; soluble N = 8.9 g/kg |
| OMV | N = 3%; $P_2O_5$ = 15%; $K_2O$ = 7% | $CaCl_2$ pH = 5.2; Organic C = 93.7 g/kg; CEC = 243.3 $mmol_c$/kg; total N = 48.7 g/kg; soluble N = 45.7 g/kg |
| OMVI | Organic C = 6%; $P_2O_5$ = 2%; $K_2O$ = 1% | $CaCl_2$ pH = 7.5; Organic C = 127 g/kg; total N = 39.7 g/kg |
| MLI | 85 PRNT (%) | 31.00% CaO; 19.97% MgO; 103.20 PN (% $CaCO_3$ eq); 89.50 PRNT (%) |
| MLII | 85 PRNT (%) | 35.67% CaO; 17.47% MgO; 105.67 PN (% $CaCO_3$ eq); 91.50 PRNT (%) |

Values are expressed as % (w/w). Limestone, organic fertilizers, and organomineral fertilizers did not have their guarantees provided. OV – vinasse, OPS – pig slurry, OPL – poultry litter, OM – organomineral, OFC – sugarcane filter cake, MTSP – triple superphosphate, MSSP – single superphosphate, MNPK – ternary (NPK), MMAP – monoammonium phosphate, ML – limestone, MFTE – Micronutrient Frits (trace elements), OCM – cattle manure. Neutral ammonium citrate (NAC)-soluble $P_2O_5$ in phosphate fertilizers

**Table 2. 3** Total contents of Potentially Toxic Elements by fertilizer type (organic, organomineral, and mineral).

| Fertilizer | As | Cd | Cr | Cu | Mo | Ni | Pb | Zn |
|---|---|---|---|---|---|---|---|---|
| | mg/kg | | | | | | | |
| Sugarcane Filter Cake | < DL | < DL | 19.56±1.29 | 48.31±2.3 | 5.04±4.95 | 5.53±1.48 | < DL | 178.49±4.14 |
| Vinasse | < DL | < DL | < DL | 0.45±0.21 | < DL | < DL | < DL | 1.19±0.14 |
| Pig Slurry | < DL | < DL | < DL | 1.11±0.98 | < DL | < DL | < DL | 6.75±5.47 |
| Cattle Manure | < DL | < DL | 39.82±0.53 | 34.15±1.08 | 2.17±0.03 | 6.19±0.58 | < DL | 117.65±4.67 |
| Poultry Litter | < DL | 0.69±0.13 | 5.14±1.13 | 481.16±67.36 | 6.62±0.84 | 3.70±0.54 | < DL | 384.26±61.36 |
| Organomineral I | < DL | < DL | 20.37±3.75 | 53.00±2.12 | 3.55±0.24 | 4.19±0.37 | < DL | 272.52±19.03 |
| Organomineral II | < DL | < DL | 22.75±7.69 | 148.06±9.48 | 2.74±0.15 | 4.76±0.30 | < DL | 239.82±10.04 |
| Organomineral III | < DL | < DL | 23.64±4.38 | 73.62±1.20 | 1.83±0.29 | 6.57±1.14 | 21.40±1.13 | 120.10±2.29 |
| Organomineral IV | < DL | < DL | 20.64±7.81 | 36.81±3.32 | 1.94±0.23 | 9.89±2.18 | 9.59±4.01 | 223.45±5.83 |
| Organomineral V | < DL | 1.05±0.05 | 29.83±3.51 | 37.54±2.12 | 2.43±0.24 | 17.57±2.25 | 9.24±1.66 | 202.54±10.75 |
| Organomineral VI | < DL | < DL | 6.34±0.89 | 30.03±3.32 | < DL | 3.73±0.26 | < DL | 4.20±2.04 |
| SSP 00-19 | < DL | 3.69±0.12 | 72.25±0.61 | 14.53±4.36 | 4.97±0.09 | 12.96±0.25 | < DL | 166.32±0.26 |
| SSP 00-21 | 82.32±6.08 | < DL | 20.17±0.15 | 98.73±1.47 | 4.64±0.27 | 52.43±0.43 | 58.25±4.29 | 128.05±1.71 |
| NPK 04-14-08 | < DL | < DL | 5.45±0.35 | 4.39±0.35 | 1.13±0.21 | < DL | < DL | 17.67±0.44 |
| NPK 05-25-15 | 18.08±1.31 | 0.66±0.03 | 15.42±0.75 | 36.81±2.66 | 2.49±0.17 | 12.34±0.46 | 39.62±4.57 | 489.22±29.1 |
| NPK 06-30-10 | < DL | < DL | 6.65±0.44 | 4.43±0.22 | < DL | 1.53±0.22 | < DL | 11.81±0.36 |
| MAP 11-52 | < DL | 0.71±0.03 | 16.27±0.18 | 18.12±0.68 | 3.18±0.36 | 7.26±0.12 | < DL | 75.62±2.68 |
| TSP | 20.67±3.9 | 22.67±0.29 | 240.43±8.06 | 41.69±0.90 | 4.68±0.56 | 37.08±0.74 | < DL | 356.29±8.81 |
| FTE BR-12 | 111.82±7.05 | 141.85±1.04 | 600.59±86.6 | 4773.76±369.49 | 27.8±0.61 | 106.09±12.54 | 11553.53±474.17 | 84553.67±1698.66 |
| Limestone I | < DL | < DL | 1.84±0.50 | 1.02±0.20 | - | 1.35±0.04 | < DL | 10.51±1.58 |
| Limestone II | < DL | < DL | 1.53±0.12 | 7.34±11.82 | - | 1.30±0.00 | < DL | 3.55±0.32 |

< DL – Below the detection limit. Hg and Se were below the detection limit and were removed from the table for better visualization. Detection limit (mg/kg): As (<11.3), Cd (<0.4), Cr (<0.5), Hg (<15), Mo (1.3), Ni (1.3), Pb (<5), Se (<13.6). The environmentally available contents of all analyzed elements are shown in Table S2.1.

**Table 2. 4** Water-available contents of Potentially Toxic Elements by fertilizer type (organic, organomineral, and mineral).

| Fertilizer | As | Cd | Cr | Cu | Mo | Ni | Pb | Zn |
|---|---|---|---|---|---|---|---|---|
| | mg/kg | | | | | | | |
| Sugarcane Filter Cake | < DL | < DL | < DL | 2.8±0.05 | 2.42±1.46 | < DL | < DL | 178.49±4.14 |
| Vinasse | < DL | < DL | < DL | 0.41±0.17 | < DL | < DL | < DL | 1.19±0.14 |
| Pig Slurry | < DL | < DL | < DL | 0.54±0.1 | < DL | < DL | < DL | 6.75±5.47 |
| Cattle Manure | < DL | < DL | 0.50±0.00 | 2.46±0.89 | 1.81±0.24 | 6.19±0.58 | < DL | 22.37±11.56 |
| Poultry Litter | < DL | < DL | 1.25±0.04 | 281.73±1.53 | 8.06±0.08 | 3.7±0.54 | < DL | 79.77±1.40 |
| Organomineral I | < DL | < DL | 1.23±0.05 | 17.22±0.42 | 3.29±0.08 | 4.19±0.37 | < DL | 58.10±0.95 |
| Organomineral II | < DL | < DL | 0.75±0.09 | 43.62±0.13 | 2.19±0.16 | 4.76±0.30 | < DL | 31.45±0.61 |
| Organomineral III | < DL | < DL | < DL | 4.08±0.3 | 0.95±0.03 | 6.57±1.14 | < DL | 1.99±0.53 |
| Organomineral IV | < DL | < DL | 3.51±0.16 | 10.62±0.32 | 2.06±0.15 | 9.89±2.18 | < DL | 42.00±0.24 |
| Organomineral V | < DL | < DL | 1.38±0.04 | 0.28±0.15 | < DL | 17.57±2.25 | < DL | < DL |
| Organomineral VI | < DL | < DL | < DL | 0.74±0.10 | < DL | < DL | < DL | 4.20±2.04 |
| SSP 00-19 | < DL | 3.69±0.12 | 10.12±0.51 | 6.30±0.35 | < DL | 14.54±0.15 | < DL | 162.48±2.82 |
| SSP 00-21 | < DL | < DL | 2.10±0.12 | 65.95±63.81 | < DL | 42.26±0.16 | < DL | 103.35±16.04 |
| NPK 04-14-08 | < DL | < DL | 1.73±0.05 | 0.41±0.19 | < DL | < DL | < DL | < DL |
| NPK 05-25-15 | < DL | 0.66±0.03 | 4.44±0.07 | 6.82±0.21 | < DL | 10.69±0.14 | < DL | 175.71±3.35 |
| NPK 06-30-10 | < DL | < DL | 6.88±0.05 | 1.23±0.16 | < DL | 4.29±0.26 | < DL | 13.56±0.11 |
| MAP 11-52 | 15.91±1.86 | 17.32±0.77 | 28.36±1.19 | 27.33±1.66 | 2.29±0.24 | 38.39±2.06 | < DL | 314.54±15.41 |
| TSP | < DL | < DL | 3.49±0.09 | 0.65±0.13 | < DL | 1.74±0.15 | < DL | 0.47±0.17 |
| FTE BR-12 | 2.48±2.24 | 2.24±0.85 | 1.17±0.09 | 2.09±0.33 | 2.90±0.24 | 4.32±0.28 | 7.53±1.37 | 19171.8±471.57 |

< DL – Below the detection limit. Hg and Se were below the detection limit and were removed from the table for better visualization. Detection limit (mg/kg): As (<11.3), Cd (<0.4), Cr (<0.5), Hg (<15), Mo (1.3), Ni (1.3), Pb (<5), Se (<13.6). The water-available contents of all analyzed elements are shown in Table S2.2.

*4.3.2 Risk quotient (RQ) for soil organisms*

The RQ values for the analyzed fertilizers can be seen in **Figure 2. 1**. The most notable RQ were obtained for Cr in cattle manure, with a RQ = 1.173 (immediate impact, that is, a single application would be sufficient to reach the threshold value) and Ni = 0.456; and sugarcane filter cake, with Cr = 0.484 and Ni = 0.342. Another relevant organic source was poultry litter, mainly for Cu (0.136), Ni (0.115), Mo (0.082), Zn (0.076), and Cr (0.064. The organomineral fertilizers (I to IV) also showed expressive RQs for Cr and Ni, ranging from 0.228–0.265 and 0.117–0.277, respectively (**Figure 2. 1**).

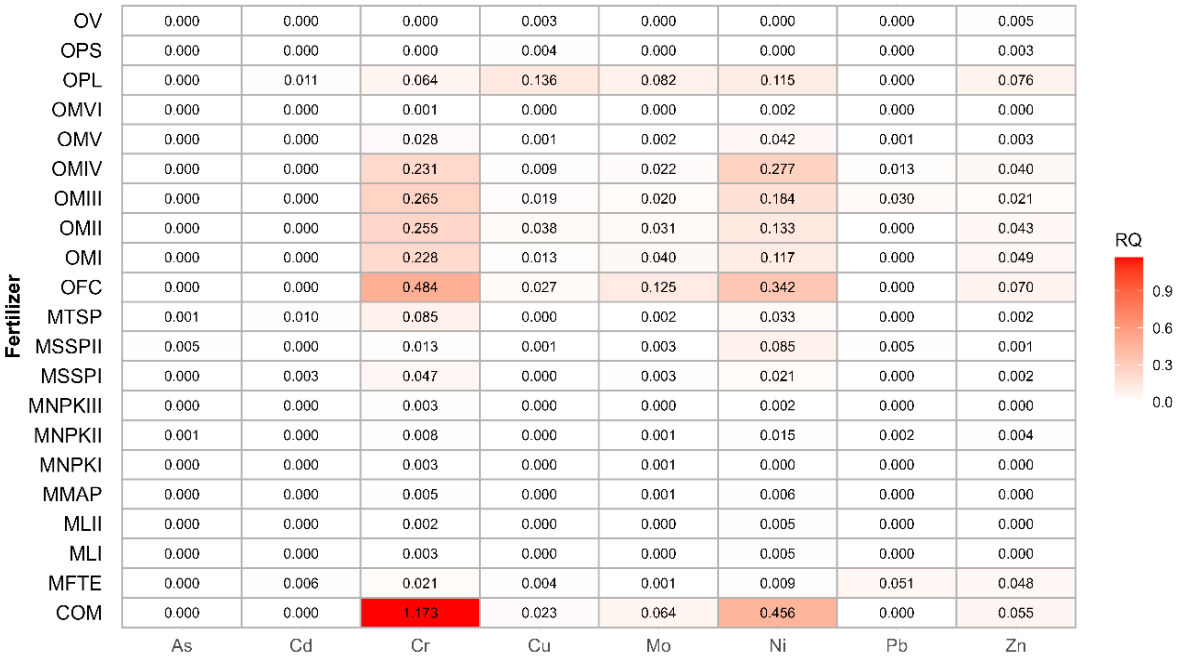


**Figure 2. 1** Risk Quotient (RQ) of Potentially Toxic Elements in fertilizers for soil biota. Darker colors indicate higher risk. RQ > 1 (dark red) indicates immediate impact. OV – vinasse, OPS – pig slurry, OPL – poultry litter, OM – organomineral, OFC – sugarcane filter cake, MTSP – triple superphosphate, MSSP – single superphosphate, MNPK – ternary (NPK), MMAP – monoammonium phosphate, ML – limestone, MFTE – Micronutrient Frits (trace elements), OCM – cattle manure.

In general, mineral fertilizers showed lower RQs than most organic and organomineral fertilizers (**Figure 2. 1**), with long periods (> 100 years) estimated to reach the PNEC (**Table 2. 5**). However, some individual values for mineral fertilizers stand out, such as Cr (0.085) in TSP, Ni (0.085) in SSP II, Cr (0.047) in SSP I, and Zn (0.048) in FTE. On the other hand, limestone, MAP, and NPK fertilizers did not show expressive RQ values.

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**Table 2. 5** Estimated number of years required to reach the PNEC based on the relationship between total concentration of Potentially Toxic Elements (PTEs), fertilizer application rate, and risk quotient (RQ) for soil biota.

| PTEs | Cake Filter | Vinasse | Pig slurry | Cattle Manure | Poultry Litter | Organomineral I | Organomineral II | Organomineral III | Organomineral IV | Organomineral V | Organomineral VI |
|---|---|---|---|---|---|---|---|---|---|---|---|
| As | - | - | - | - | - | - | - | - | - | - | - |
| Cd | - | - | - | - | 93 | - | - | - | - | - | - |
| Cr | 2* | - | - | 1 | 16 | 4 | 4 | 4 | 4 | 35 | 827 |
| Cu | 37 | 376 | 261 | 44 | 7 | 74 | 27 | 53 | 107 | 1229 | 7679 |
| Mo | 8 | - | - | 16 | 12 | 25 | 33 | 49 | 46 | 431 | - |
| Ni | 3 | - | - | 2 | 9 | 9 | 8 | 5 | 4 | 24 | 562 |
| Pb | - | - | - | - | - | - | - | 33 | 75 | 907 | - |
| Zn | 14 | 203 | 62 | 18 | 13 | 21 | 23 | 47 | 25 | 325 | 78362 |

| PTEs | SSP I | SSP II | NPK I | NPK II | NPK III | MAP | TSP | FTE BR | Limestone I | Limestone II |
|---|---|---|---|---|---|---|---|---|---|---|
| As | - | 202 | - | 1207 | - | - | 1479 | 2733 | - | - |
| Cd | 333 | - | - | 2450 | - | 3708 | 100 | 160 | - | - |
| Cr | 21 | 76 | 371 | 131 | 304 | 202 | 12 | 47 | 368 | 443 |
| Cu | 4658 | 685 | 20260 | 2416 | 20077 | 7991 | 2987 | 261 | 29205 | 4059 |
| Mo | 309 | 331 | 1789 | 812 | - | 1035 | 605 | 1018 | - | - |
| Ni | 47 | 12 | - | 66 | 528 | 181 | 31 | 107 | 201 | 208 |
| Pb | - | 211 | - | 408 | - | - | - | 20 | - | - |
| Zn | 581 | 754 | 7184 | 259 | 10749 | 2733 | 499 | 21 | 4045 | 11977 |

The RQ does not account for the soil input of PTEs from other sources, such as atmospheric deposition or the use of agrochemicals, nor does it consider losses of PTEs, such as plant uptake and removal by crops, surface runoff, and leaching. * Number of years required to reach the predicted no-effect concentration (PNEC), calculated using the formula: Years = PNEC/PEC.

*4.3.3 Principal component analysis between potentially toxic elements and fertilizers*

Figure S2.5 presents the Spearman correlation analysis between the total concentrations of the elements, the RQs, and the application rate. The correlation matrix shows that all total contents are significantly correlated with their respective RQs (Figure S2.5). As_RQ is positively correlated with Cd_RQ (0.46) and Pb_RQ (0.50). Zn_RQ and Cu_RQ showed a strong positive correlation (0.91), as well as Cr_RQ with Ni_RQ (0.96) and Mo_RQ (0.92). Regarding the total contents, Zn stands out, as it is positively correlated with all the other elements.

The explained variance percentages of the two main principal components in the PCA were 41.9% and 27.1%, totaling a cumulative variance of 69% (Figure S2.3). PC1 (Table S2.3) showed negative loadings for all evaluated PTEs and RQs (ranging from -0.38 to -0.09), while application rate showed a positive loading (0.13). Three distinct clusters were formed: i) As_RQ; ii) Cu, Zn, Ni, Pb, Cd_RQ, Mo, Cr, and Pb_RQ; and iii) application rate, Cr_RQ, Cu_RQ, Mo_RQ, Ni_RQ, and Zn_RQ (Figure S2.4). PC2 revealed a different ecotoxicological risk profile (**Figure 2. 2** and Table S2.3). Positive loadings were observed for the RQs of Mo (0.43), Ni (0.39), Zn (0.37), Cr (0.37), and Cu (0.31), as well as for application rate (0.37) (Table S2.3). Conversely, negative loadings were observed for the RQs of As (-0.17), Pb (-0.15), and Cd (-0.01) (Table S2.3).

As seen in **Figure 2. 2**, organic and organomineral fertilizers, except for organomineral V and VI, are applied at high rates, while mineral fertilizers, with the exception of limestone, are applied at lower rates (below 1 ton/ha). The RQs of Ni, Mo, Cr, Cu, and Zn are more correlated with organic and organomineral fertilizers, whereas the RQs of Cd, As, and Pb are more correlated with mineral fertilizers. Although the total Ni concentration is more correlated with mineral fertilizers, such as SSP II and FTE, the Ni RQ is more correlated with organic sources, such as cattle manure, sugarcane filter cake, and poultry litter (**Figure 2. 2**).

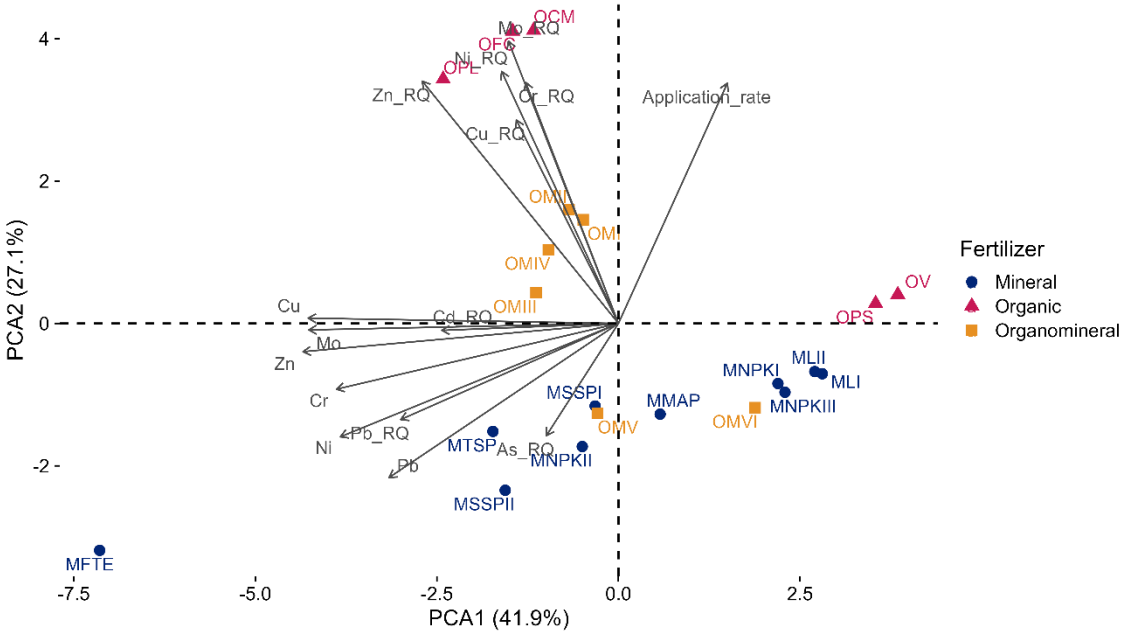


**Figure 2. 2** Principal Component Analysis (PCA) of the concentration of Potentially Toxic Elements and application rates in fertilizers and their risk quotient (RQ) for soil biota. Vectors indicate the variables, and colored symbols indicate the different fertilizer types.

*4.3.4 Cluster analysis – similarities among fertilizers*

FTE showed the greatest Euclidean distance from the other fertilizers, being the sole representative of group 1 (**Figure 2. 3**). Group 2 included poultry litter, cattle manure, and sugarcane filter cake, showing high similarity in their total composition, application rate, and

risks to soil biota. Group 3 included pig slurry, vinasse, organominerals I-VI, NPKs, limestones, MAP, TSP, and SSPs.

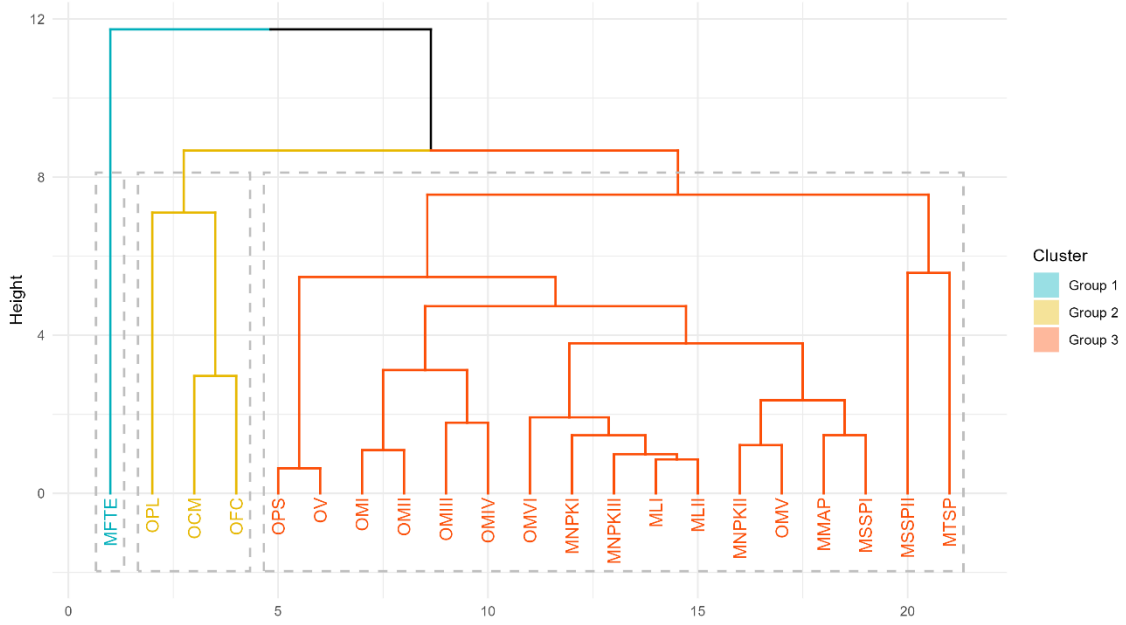


**Figure 2. 3** Cluster dendrogram using Euclidean distance of similarity among fertilizers based on their total composition, application rate, and risk quotient for soil organisms.

## 4.4 DISCUSSION

Given the extensive fertilizer and soil amendment use in agricultural areas within the Cerrado biome, it becomes important to evaluate not only the agricultural potential of these inputs but also their possible environmental implications. Overall, total PTEs content fell within the limits established by Brazilian legislation (Brasil, 2016), except for As concentrations (82.3 mg/kg) in SSP II (00-21) and Pb concentration (11,553 mg/kg) in FTE-BR12, which exceeded the reference values of 45 and 10,000 mg/kg, respectively. The total As content in the single superphosphate is considerably higher than other values reported in the literature for phosphate fertilizers:15 mg/kg in superphosphate (Liu *et al.*, 2023b), 4.09 mg/kg in triple superphosphate (Hu *et al.*, 2024b), and 26.78 mg/kg in a calcium superphosphate (Gao *et al.*, 2021). For FTE-BR12, the literature describes fritted fertilizers with similar specifications presenting Pb concentrations ranging from 3,120 mg/kg to 25,200 mg/kg (Vale and Alcarde, 2003), when extracted with concentrated hydrochloric acid.

Although reference values for PTEs in fertilizers are expressed as total contents (Brasil, 2016), this parameter constitutes a poor predictor of the element biological availability, which more accurately determines the toxicological risk (Chen *et al.*, 2023). In this context, the use of the water extraction method may be a promising tool to assess the readily available fraction of metals in the sampled fertilizers, while also preserving the natural composition of the matrix, without introducing additional chemical reagents (Liu *et al.*, 2024b). However, its use remains largely unexplored for mineral fertilizers.

For readily available contents, organic and organomineral fertilizers were shown to release proportionally less PTEs than mineral sources (Figure S2.1). The literature has already reported that chemical fertilizers dispersed 384 times faster than organic fertilizers in a soil/water mixture, taking only 120 minutes compared to 32 days (Majee *et al.*, 2023). This slow-release pattern observed in organic fertilizers stems from their more complex organic chemical structures and lower water solubility compared to mineral fertilizers, which, in turn, are more soluble and generally composed of simple ionic salts that favor dissolution in water (Islam *et al.*, 2024; Wato *et al.*, 2024).

The organic matter present in organic and organomineral fertilizers can affect PTEs availability in two contrasting ways. On one hand, it can immobilize these elements due to its large number of functional groups and surface charges (Liu *et al.*, 2024a). On the other hand, the dissolved fraction of organic matter can enhance the transport and bioavailability of these elements in soil, increasing the risks of PTEs uptake by agricultural crops (Lian *et al.*, 2022).

Thus, although organic fertilizers are generally regarded as slow-release, supplying elements in smaller amounts over an extended period (Shaji *et al.*, 2021), these particularities add greater complexity to their actual impact on the availability of toxic metals.

Moreover, other factors affect PTEs bioavailability in the soil, such as the action of roots and microbial exudates. Plant roots can release exudates capable of chelating and solubilizing PTEs (Podar and Maathuis, 2022). Like plants, soil microorganisms can also alter the bioavailability of these metals through redox reactions, chelation, acidification, complexation, and precipitation (Mishra *et al.*, 2017; Drabesch *et al.*, 2024). Moreover, soil management may also be a relevant factor, for example limestone application, which raises soil pH and decreases the availability of cationic elements through their precipitation and adsorption by variable-charge colloids (Mahmud and Chong, 2022). For elements that can be found in anionic forms in the soil, such as Se, As, Mo, and Cr(VI), rising pH levels increase their bioavailability, thereby complicating their simultaneous remediation with cations through liming (Nie *et al.*, 2020; Rokonuzzaman *et al.*, 2022; Qu *et al.*, 2023; Zhao *et al.*, 2026).

Most agricultural soils in the Cerrado regions are naturally acidic, poor in nutrients and with high aluminum saturation (Hammerschmitt *et al.*, 2021). Thus, their dependence on fertilizers and soil amendments is worrisome considering the RQs projected in this study. The most critical case is observed with Cr in cattle manure, where the RQ estimates an impact on soil biota within the first year of application (at a dose of 20.8 ton/ha and a Cr concentration of 39.82 mg/kg) (**Table 2. 5**). Considering a realistic medium-term period (fewer than fifteen years of application), major concerns identified were: filter cake (Cr, Mo, Ni, and Zn), cattle manure (Cr, Ni), poultry litter (Cu, Mo, Ni, and Zn), organomineral fertilizers I–IV (Cr and Ni), SSP II (Ni), and TSP (Cr) (**Table 2. 5**). Xia *et al.* (2026) evaluated soils under long-term agricultural use, reporting high RQs for several PTEs, with particular emphasis on Cr: Cr (128–317), Cd (0.11–2.5), Pb (10–46), Hg (1.6–16), and As (31–138).

Several short and long-term experiments have demonstrated the effects of fertilization on the PTEs accumulation in soil. Qian *et al.* (2020) analyzed soil metal contents over five years of cattle manure application, showing increases in As from 7.42 to 8.87 mg/kg, Cd from 0.22 to 0.25 mg/kg, Cr from 59 to 71.2 mg/kg, Cu from 26.9 to 32.3 mg/kg, Hg from 0.037 to 0.041 mg/kg, Pb from 26.7 to 29.1 mg/kg, Mn from 607 to 744 mg/kg, and Zn from 83 to 112 mg/kg. Similarly, Gao *et al.* (2021), in a 38-year experiment evaluating soil total PTE concentrations under fertilization, observed increases in Cd from 0.12 to 0.37 mg/kg and As from 20.4 to 35.5 mg/kg in soils fertilized with cattle manure and mineral NPK fertilizer.

Overall, according to the risk analysis, Cr and Ni were the two most concerned PTEs among the sampled fertilizers (**Figure 2. 1**). These two elements have notably restrictive PNEC values (Cr = 0.25 and Ni = 0.1 mg/kg) compared to other studies. In a Danish study, Sorensen *et al.* (2011) reported a PNEC of 3.2 mg/kg for Cr and 1.1 mg/kg for Ni. Furthermore, the predicted environmental concentrations reported by the authors were also lower (PEC Cr = 0.06 mg/kg and PEC Ni = 0.03 mg/kg) than those estimated in the present work (PEC Cr = 0.29 mg/kg and PEC Ni = 0.14 mg/kg, considering cattle manure data). These differences may reflect variations in application rates, soil physicochemical properties, and fertilizer composition that naturally exist across distinct regions.

For Brazil, several reported Cr concentrations in fertilizers can be found: sewage sludge compost (850–900 mg/kg) (Nascimento *et al.*, 2023), poultry litter (0.05–17.7 mg/kg) (Parente *et al.*, 2020, 2021), manure and sawdust compost (7.36–15.47 mg/kg) (Panisson *et al.*, 2021), cattle manure (1.46 mg/kg) (Silva and Franzini, 2024), and phosphate fertilizer (333 mg/kg) (Nascimento *et al.*, 2023). The Cr values in the studied sources exceeded those reported in the literature for poultry litter (39.8 mg/kg), organomineral compost II–IV (20.4–23.6 mg/kg), and cattle manure (6.3 mg/kg) (**Table 2. 3**).

Brazilian legislation regulates Cr based on its speciation as hexavalent chromium (Cr(VI)), so the results of this work cannot be directly compared with the reference values. Cr(VI) is significantly more toxic than Cr(III) (Sharma *et al.*, 2022). However, its determination is more complex than total Cr due to the specific extraction and speciation procedures involved (Sá *et al.*, 2023).

The dose-risk relationship discussed throughout the previous sections was also captured by the PCA results (**Figure 2. 2**). PCA allows the grouping of highly correlated variables, reducing a large dataset into two dimensions (Aidoo *et al.*, 2021). This analysis, combined with other tools, has previously been used across various soil types, either to identify sources and PTEs transport pathways in soil (Anaman *et al.*, 2022; Sojka and Jaskuła, 2022), to identify critical elements (Askari *et al.*, 2020), or to group correlated metals (Liu *et al.*, 2023a). In this study, PCA was used to correlate metals and fertilizers of distinct natures, as well as their projected risk to soil organisms and application dose.

Along PC2, a clear divergence was observed between organic/organomineral sources and mineral sources (**Figure 2. 2**). The loadings indicated a positive correlation between application doses and the RQs of Cr, Ni, Mo, and Zn, while their total contents showed a negative correlation along this axis (Table S2.3). This behavior suggests that, for these elements, the application dose is a more determinant factor of their risk potential to soil

organisms than their total concentration, being more closely associated with organic and organomineral fertilizers, which are applied at higher doses. Conversely, the RQs of As, Cd, and Pb showed negative loadings on PC2 (Table S2.3), being inversely correlated with dose and linked to mineral fertilizers, which are applied at lower doses. The stronger linear association between the RQs of Cu and Zn and the total concentration of these elements, rather than with dose, indicates that the risk to soil biota is more closely related to source composition (Figure S2.5). The fertilizers most associated with high Cu_RQ and Zn_RQ are cattle manure, poultry litter, sugarcane filter cake, and organomineral compounds (**Figure 2. 2**). While the European Union limits Cu and Zn in organic fertilizers (300/800 mg/kg) and organomineral fertilizers (600/1.500 mg/kg) (European Union, 2019), Brazilian legislation does not regulate them. Mo, which also showed elevated RQs, is not limited by either regulatory framework. The estimated risks for these elements highlight the need for Brazilian legislation to include the monitoring of Cu, Zn, and Mo in organic and organomineral fertilizers. Moreover, further studies are needed to assess metals that remain poorly investigated, such as Se and Hg, which have already been identified as high-risk elements in fertilizers (Farid *et al.*, 2025).

Thus, the results suggest that the continuous use of manures and filter cake can lead to Cu and Zn accumulation in the soil, a pattern also observed in previous studies. Lemos *et al.* (2025) evaluated soil PTEs under prolonged swine manure application, obtaining a significant increase in Cu levels (from 0.037 to 0.476 mg/kg) and Zn levels (from 0.140 to 6.487 mg/kg), indicating potential toxicity risks from Zn. Also, Zhao *et al.* (2022) evaluated the effects of swine manure application from 2018 to 2020, reporting increased Cu (from 22.4 to 54.3 mg/kg) and Zn content (from 88.4 to 149.4 mg/kg).

Unlike organic and organomineral fertilizers, mineral fertilizers, particularly phosphate fertilizers, are more strongly associated with risk to soil biota for As, Cd, and Pb (Table S2.3 and **Figure 2. 2**). However, this relationship is variable in literature. For example, Couto *et al.* (2018) compared swine manure with mineral fertilization, obtaining no relevant differences for the soil concentrations of As, Cd, Cr, Ni and Pb between these sources. Nevertheless, for Cu and Zn, concentrations reached 47.8 and 90.9 mg/kg under swine manure application, compared with 7.5 and 18.3 mg/kg under mineral fertilizer application. In turn, Hu *et al.* (2024b) observed that the prolonged use of phosphate fertilizers was directly correlated with the enrichment of contaminants in the 0–30 cm topsoil layer (Cd, Cr, Uranium (U), and Vanadium (V)), and of As in deeper horizons (above 100 cm). Although the projections in **Table 2. 5** do not identify mineral sources as those of greatest concern, studies such as Hu *et al.* (2024b) illustrate that the

prolonged, high-dose application of mineral fertilizers may also consistently increase the risks of food chain contamination by these metals.

Regarding the similarity between fertilizers, the cluster analysis (**Figure 2. 3**) showed FTE as the most distinct fertilizer. This is because it is a micronutrient fertilizer with high PTE concentrations, although applied at lower doses (25 kg/ha). Another relevant finding was that liquid swine manure, vinasse, and lime are closer to each other than to other fertilizers, largely due to the doses used (above 1 ton/ha) and a low-risk quotient compared to the other fertilizers. The remaining fertilizers are within a group that provides important insights, such as the proximity between SSP II (00-21) and TSP, which have a Ni RQ that restricts their application to 12 and 31 years, respectively (**Table 2. 5**). Still within this latter group, poultry litter, cattle manure, and filter cake form a distinct subgroup, corresponding precisely to those with the highest RQs, applied at higher doses (**Table 2. 5**). In addition, organomineral fertilizers I-IV are grouped within the same subgroup, which is justified by their shared classification as organomineral compounds. Organomineral VI, however, is positioned closer to NPKI, NPKIII, and MAP than to the other organomineral fertilizers (**Figure 2. 3**), likely due to its composition (peat, potassium hydroxide, phosphoric acid, water, and monoammonium phosphate) (**Table 2. 1**) and the low dose applied.

The PTEs occurrence in agricultural soils of the Cerrado biome has already attracted a certain degree of attention. Osti *et al.* (2025) reported Cr concentrations ranging from 94 to 515.9 mg/kg; Cu from 15.6 to 99.6 mg/kg; Hg from 2.17 to 2.9 mg/kg; Zn from 13.7 to 70.8 mg/kg; Pb from 5.7 to 11.7 mg/kg; and Ni from 8.7 to 65.4 mg/kg. Cu, Cr, Hg and Ni exceeded the threshold values for soil quality protection, which are, respectively: 60, 75, 0.5 and 30 mg $kg^{-1}$, according to the Brazilian legislation (Brasil, 2009). For Cr, in some areas, concentrations exceeded the threshold values for human health protection, which is 150 mg/kg. On the other hand, Cd and As concentrations were below the quantification limit (< 2 mg/kg).

Cabral *et al.* (2023) also reported relevant PTE concentrations in soils from the Cerrado biome, with Cu concentrations ranging from 6.16 to 418.63 mg/kg, Zn from 2.59 to 138.34 mg/kg, Ni from 3.08 to 52.45 mg/kg, Cd from 0.49 to 16.76 mg/kg, Pb from 3.11 to 28.93 mg/kg, As from 1.77 to 26.23 mg/kg, and Hg from 0.19 to 1.85 mg/kg. Moreover, 83% and 67.2% of the evaluated soil samples (n = 41) presented Cd and Hg concentrations above the soil quality thresholds, respectively. Although authors did not perform sources analyses, soil contamination by Cd was mainly associated with the use of phosphate fertilizers (Cabral *et al.*, 2023).

The use of organic and organomineral fertilizers represents a growing alternative to increase soil fertility (Han *et al.*, 2021; Iqbal *et al.*, 2023; Oliveira *et al.*, 2025), being considered a more adequate destination for the large amounts of agro-industrial and livestock waste generated compared to alternatives such as incineration or landfill disposal (Su *et al.*, 2025). However, high rates applied continuously can negatively impact the environment in many different ways apart solely from PTEs input (Wu *et al.*, 2025), including the spread of antibiotics, antibiotic resistance genes and pathogens (Lima *et al.*, 2020; Kuppusamy *et al.*, 2023), xenobiotics contamination of surface and groundwater, and greenhouse gas emissions (Goldan *et al.*, 2023; Zou *et al.*, 2026).

Furthermore, soil enrichment with PTEs may increase the uptake of these elements by agricultural crops. You *et al.* (2025) reported total PTEs (Zn, Cu, Cr, Ni, Pb, As, Cd, and Hg) accumulation of 113.2 and 63.1 mg/kg in lettuce and radish, under swine manure fertilization, compared with 78.8 and 44.6 mg/kg under mineral fertilization, respectively. Similarly, Li *et al.* (2023) reported significantly higher concentrations of Cu, Zn, Pb, and Cd ($p < 0.05$) in the edible parts of green pepper and eggplant treated with swine manure compared with the non-fertilized control. These results highlight the need for more restrictive technical criteria for the use of waste-derived organic fertilizers on agricultural soils.

In the Cerrado biome there is a gap in data regarding the impact of fertilization on PTEs accumulation in vegetables. Binde *et al.* (2026) quantified PTE concentrations in soybean grown in Cerrado agricultural areas: As (0.002 mg/kg), Cd (0.012 mg/kg), Co (0.07 mg/kg), Cr (0.06 mg/kg), Cu (9.7 mg/kg), Fe (60.66 mg/kg), Mn (21.82 mg/kg), Mo (6.44 mg/kg), Ni (0.64 mg/kg), Pb (0.01 mg/kg), Se (0.005 mg/kg), and Zn (33.72 mg/kg). Although these values are within Brazilian regulatory limits, the study did not evaluate the effect of fertilization type on PTE accumulation or include a control treatment. Therefore, further research is needed to specifically assess the role of fertilization in PTE enrichment of agricultural crops in the Cerrado biome, especially on vegetables.

Likewise, contaminants in mineral fertilizers remain a relevant concern and justify continuous monitoring by regulatory agencies, since two sampled fertilizers exceeded the legal limits established by Brazilian legislation (**Table 2. 3**) (Brasil, 2006). These findings are particularly relevant considering that Brazil is a major importer and consumer of fertilizers, accounting for 8% of global consumption, with more than 80% of this demand met by foreign sources (Brasil, 2023). In 2024 the volume of fertilizers delivered to the market reached 45.61 million tons (ANDA, 2025). The Central-West region of the country, which harbors a large portion of the Cerrado biome, alone accounts for approximately 40% of national consumption

of phosphate fertilizers, 32% of nitrogen fertilizers, and 38% of potassium fertilizers (Brasil, 2023). From this perspective, it is important to reinforce the strict enforcement of regulatory limits, in order to prevent PTE enrichment in Brazilian agricultural soils and the consequent toxicity to their edaphic biota.

It is also important to highlight some important limitations of this study: i) it should not be interpreted as being valid for all fertilizers, since it does not include a large sample size or sufficient replicates for that purpose; ii) the risk analysis does not account for PTEs soil input from different sources, such as atmospheric deposition, the use of agrochemicals or low quality irrigation water, nor does it account for PTEs losses through crop uptake and removal, surface runoff, and leaching. These are all important factors affecting PTEs dynamics in the soil and the estimated time to reach PNEC values and; iii) there is a lack of ecotoxicological assay data for all PTEs applicable to natural tropical soils, particularly Cerrado soils. Therefore, this discussion is not intended to condemn the use of fertilizers, given their importance for agricultural production and food security (Giller *et al.*, 2021), but rather to highlight the need to strictly regulate PTEs limits in fertilizers and to systematically monitor their accumulation in the soil, especially over the long-term. These are essential actions to promote sustainable fertilization management and safeguard soil functions and ecosystem health.

These findings raise concerns in regions that use high doses of organic and organomineral fertilizers, especially due to the presence of extremely weathered, nutrient-poor, and acidic soils, such as those found in the Brazilian Cerrado biome. However, this region stands out not only for its agricultural and livestock production and development, but also as the most biodiverse tropical savanna in the world, in addition to harboring the headwaters of eight of the country's twelve most important river basins (Bolfe *et* al., 2020). This concern is further heightened given the transport capacity of PTEs (Hama Aziz *et al.*, 2023), along with their high toxicity, persistence, and bioaccumulation throughout the trophic chain (Galasso *et al.*, 2025; Jomova *et al.*, 2025). Thus, this study demonstrates the importance of evaluating fertilizers beyond their agronomic role in nourishing crops, also incorporating the notion of their contamination footprint, which must be assessed, monitored, and managed.

## 4.5 CONCLUSIONS

Overall, the total PTE concentrations in the sampled fertilizers were within legal Brazilian limits, with only the single superphosphate and FTE-BR12 exceeding legislation limits for As and Pb, respectively. Regarding the readily available contents, mineral fertilizers released proportionally more PTEs than organic fertilizers. Concerning the risks to soil biota in the Cerrado biome, cattle manure showed the highest RQs (1.173 for Cr and 0.456 for Ni), projecting that the PNEC threshold would be reached within 1 and 2 years, respectively. Overall, the ecotoxicological risks of Mo, Cr, Cu, Ni, and Zn were more strongly associated with organic fertilizers, while that of As, Cd, and Pb were more strongly associated with mineral fertilizers.

Under realistic application rates used in agricultural areas of the Cerrado biome, the sampled fertilizers represented a relevant source of PTEs to the soil. For most elements, the most determining factor for soil biota risk was the application dose rather than PTEs total concentration in the fertilizer. This raises relevant concerns over the potential for PTEs soil accumulation under current fertilization practices, particularly the application of organic and organomineral fertilizers at high doses. Thus, a major finding is that regulating only total PTEs content in fertilizers is insufficient to prevent soil metal accumulation. The adoption of additional criteria, such as soil concentration monitoring and load limits, is also essential.

For future research, studies focused on ecotoxicological assays of PTEs using tropical soils and a broader range of indicator organisms are needed. Also, the Brazilian fertilizer legislation should be updated, with the adoption of limits for Cu, Mo, and Zn in organic and organomineral fertilizers representing a first step forward. Another important aspect for future regulations update would be to assess the feasibility of adopting load limits (g/ha/year) in addition to concentration limits (mg/kg) as a strategy for managing organic fertilizers applied at high rates. These are all essential steps towards a more sustainable fertilizer management in local agricultural areas, contributing to the safeguard of both ecosystem health and food security.

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# 5 GENERAL CONCLUSIONS

Both the global meta-analysis and the regional-level analysis reinforce that fertilizers represent a relevant pathway for the introduction of potentially toxic elements (PTEs) into agricultural soils. It was estimated that fertilizer use could broadly impact soil biota, that is, through multiple inorganic contaminants, over medium-term periods (< 15 years). Special attention should be given to organic and organomineral fertilizers, mainly due to their high application doses and to metal limit legislation that remains poorly restrictive. Although the global literature on the topic has expanded over the past decades, research efforts remain concentrated on elements such as Cd, As, Pb, Zn, and Cu, leaving elements such as Hg, Mn, Co, and Se on the margins.

Both studies present some limitations, such as the fact that the risk estimation considered only the input of PTEs via fertilization, disregarding other inputs, such as atmospheric deposition and natural soil concentrations, as well as outputs, such as surface runoff, erosion, plant uptake, and leaching. For the second study, there is also a lack of representativeness in the global literature regarding tropical soils and organisms for the ecotoxicological assays used in the risk analyses. It is also important to highlight the limited number of samples per fertilizer, which means the results cannot be interpreted as a broad survey of all fertilizer classes.

The results obtained in the Cerrado and the risk estimates confirm patterns identified in the literature: organic sources showed higher ecotoxicological risk, mainly due to their high application doses. It is important to emphasize that, based on an analysis of current Brazilian legislation, there are gaps that render it insufficient to preserve soil health and monitor PTE enrichment in soils. The main gaps are the absence of concentration limits for Cu, Zn, and Mo in manures, and the lack of continuous, long-term monitoring, particularly regarding cumulative soil loading and ecotoxicological thresholds. Thus, it is concluded that updates to current legislation are necessary to integrate ecosystem health and agricultural productivity. Furthermore, the development of further studies in the areas of contaminant monitoring and ecotoxicological assays are recommended, particularly considering tropical characteristics and organisms, as well as long-term periods.

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## APPENDICES

**Appendix A –** Supplementary Material: Potentially Toxic Elements in Fertilizers and Associated Risks for Soil Organisms: A Bibliometric and Meta-Analytical Assessment

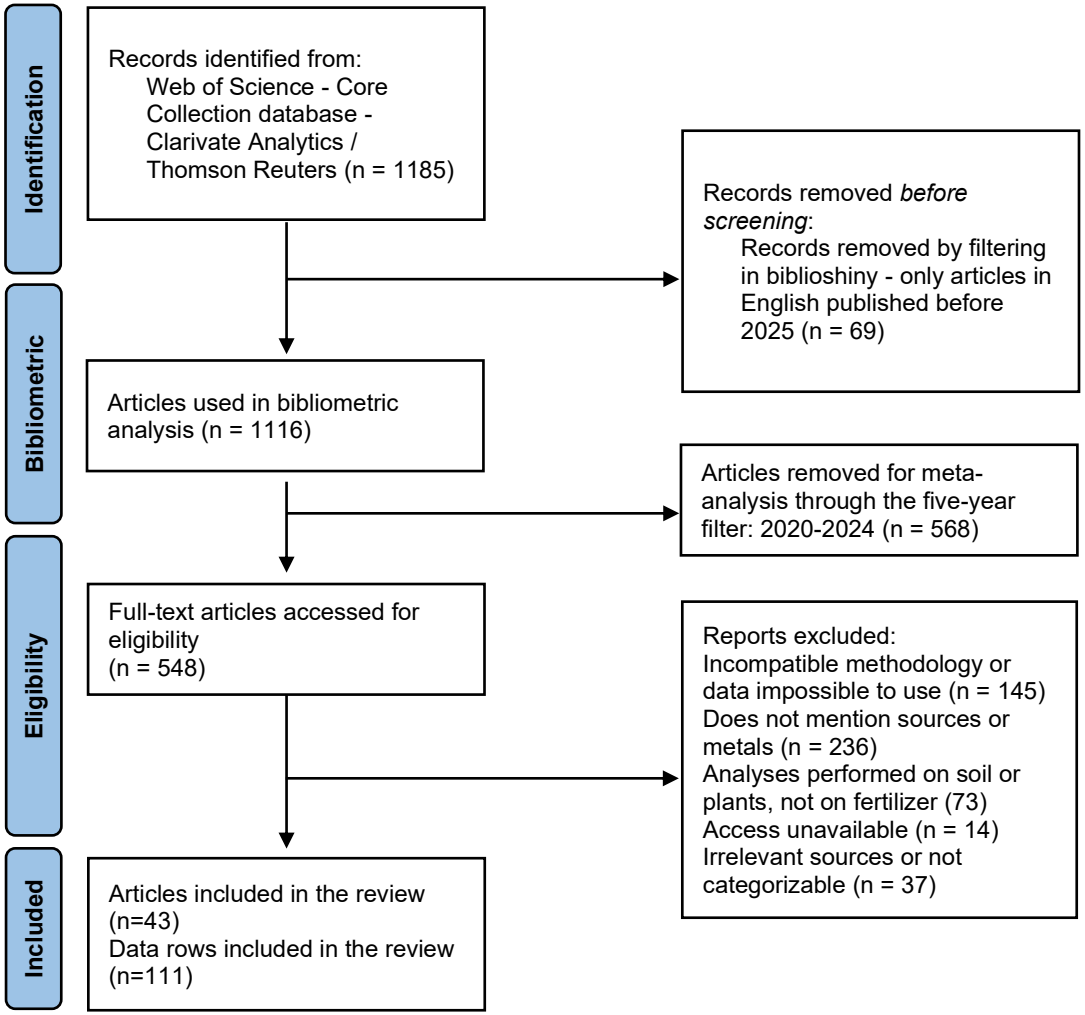


**Figure S1.1** – Flow diagram based on the PRISMA model demonstrating the process of identifying, bibliometric, eligibility, and inclusion of articles. Due to the low number of studies retrieved and the possibility that relevant information might be present in articles that could be excluded during the screening phase, it was decided to read the full text of all selected references.

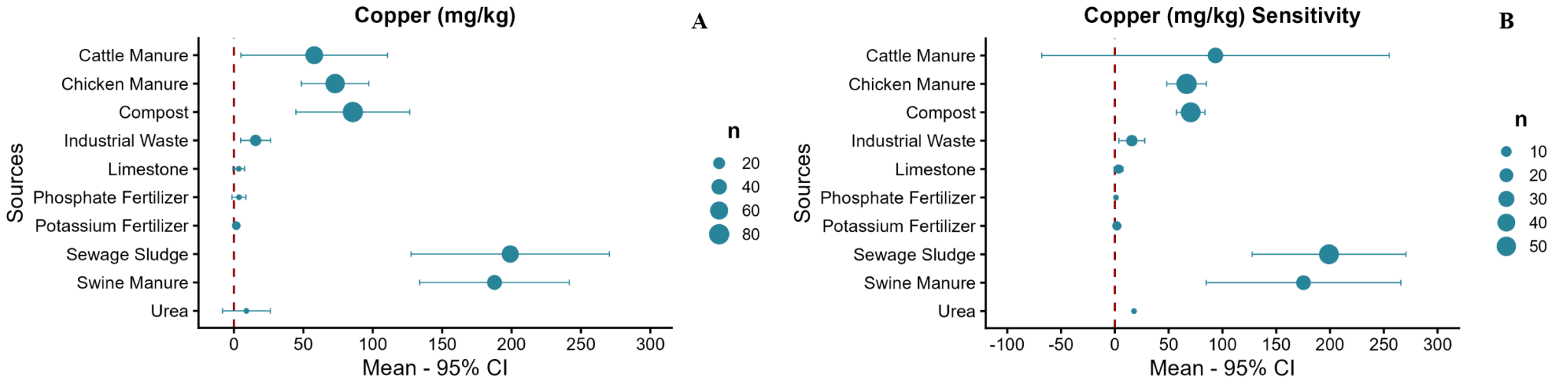


**Figure S1.2** – Sensitivity analysis comparing copper meta-analysis results with (A) and without (B) studies containing imputed sample sizes (n=3). The size of the circles represents the sample size (n), and the rods represent the 95% confidence interval.

**Table S1.1** - Reference values used in risk analysis, characterized as protective of soil biota and terrestrial ecosystems.

| Metals | PNEC* | Reference | DOI |
|---|---|---|---|
| As | 0.86 | Xu et al., 2019 | https://doi.org/10.1016/j.scitotenv.2019.05.015 |
| Cd | 0.21 | | |
| Cr | 1.57 | | |
| Pb | 3.05 | | |
| Hg | 0.16 | | |
| Ni | 6.5 | Wang et al., 2018 | https://doi.org/10.1007/s11356-017-0456-6 |
| Cu | 13.1 | Wang et al., 2015 | https://doi.org/10.1371/journal.pone.0140306 |
| Zn | 38.3 | Wan et al., 2020 | https://doi.org/10.1016/j.ecoenv.2020.110418 |

*The PNEC (Predicted No-Effect Concentration) values were estimated from the HC5 (Hazardous Concentration for 5% of the species), calculated using Species Sensitivity Distribution (SSD) curves.

**Table S1.2** – Establishing fertilizer application rates scenarios for risk assessment in maize crops under low-fertility soils. The rates are based on references.

| **Fertilizer** | **Manure and Compost** | **Phosphate** | **Potassium[c]** | **Nitrogen[d]** | **Silicon** | **Limestone** | **Sewage Sludge** |
|---|---|---|---|---|---|---|---|
| Low rate | 5 t/ha | 133 kg/ha – Other Phosphate Fertilizers[a] | 150 kg/ha | 333 kg/ha | 0.88 t/ha[e] | 3 t/ha[f] | 5 t/ha |
| | | 150 kg/ha – Phosphate Rock[b] | | | 0.44 t/ha/year | 0.75 t/ha/year | |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| High rate | 20 t/ha | 667 kg/ha – Other Phosphate Fertilizers<br>750 kg/ha – Phosphate Rock | 400 kg/ha | 1000 kg/ha | 2.64 t/ha<br>1.32 t/ha/year | 12 t/ha[e]<br>4 t/ha/year | 20 t/ha |
| Reference | Agbede (2025) | Gotz et al. (2023) | Gotz et al. (2023) | Coelho et al. (2022) | Galindo et al. (2021) | Purnomo et al. (2024) | Yada et al. (2015) |
| DOI | https://doi.org/10.1038/s41598-025-00394-8 | https://doi.org/10.3390/agronomy13010158 | https://doi.org/10.3390/agronomy13010158 | https://doi.org/10.36783/18069657rbcs20210087 | https://doi.org/10.3390/plants10071329 | https://doi.org/10.1088/1755-1315/1377/1/012113 | https://doi.org/10.1590/01000683rbcs20140728 |

[a]Considering 45% $P_2O_5$; [b]Considering 40% $P_2O_5$; [c]Considering 60% $K_2O$; [d]Considering 45% N; [e]Low rate = 50% of the reference rate; high rate = 150%. Reference rates are applied every two years.; [f]Reference rate is applied every four years.

**Table S1.3** – Limits for potentially toxic elements adopted by European and Brazilian legislation, in $mg.kg^{-1}$ per analyzed input.

| | **European legislation** | | | | | | | | **Brazilian legislation** | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Metals/ Sources** | Cd | Cr | Hg | Ni | Pb | As | Cu | Zn | As | Cd | Cr | Hg | Ni | Pb | Se | Zn | Cu | Ba | Mo |
| Cattle Manure | 1.5 | 2[a] | 1 | 50 | 120 | 40 | 300 | 800 | 20 | 3 | 2[a] | 1 | 70 | 150 | 80 | - | - | - | - |
| Swine Manure | 1.5 | 2[a] | 1 | 50 | 120 | 40 | 300 | 800 | 20 | 3 | 2[a] | 1 | 70 | 150 | 80 | - | - | - | - |
| Chicken Manure | 1.5 | 2[a] | 1 | 50 | 120 | 40 | 300 | 800 | 20 | 3 | 2[a] | 1 | 70 | 150 | 80 | - | - | - | - |
| Compost | 1.5 | 2[a] | 1 | 50 | 120 | 40 | 300 | 800 | 20 | 3 | 2[a] | 1 | 70 | 150 | 80 | - | - | - | - |
| Phosphate Rock | 60 | 2[a] | 1 | 100 | 120 | 40 | 600 | 1500 | 40 | 80 | 800 | 1 | - | 400 | - | - | - | - | - |
| Phosphate Fertilizer | 60 | 2[a] | 1 | 100 | 120 | 40 | 600 | 1500 | 250 | 57 | - | - | - | 1000 | - | - | - | - | - |
| Potassium Fertilizer | 3 | 2[a] | 1 | 100 | 120 | 40 | 600 | 1500 | 10 | 20 | 200 | 0.2 | - | 100 | - | - | - | - | - |
| Silicon Fertilizer | 200 | - | 100 | 2000 | 600 | 1000 | - | - | 10 | 20 | 200 | 0.2 | - | 100 | - | - | - | - | - |
| Urea | 3 | 2[a] | 1 | 100 | 120 | 40 | 600 | 1500 | 10 | 20 | 200 | 0.2 | - | 100 | - | - | - | - | - |
| Limestone | 1.5 | 2[a] | 1 | 100 | 120 | 40 | 300 | 800 | - | 20 | - | - | - | 1000 | - | - | - | - | - |
| Mushroom Residue | 1.5 | 2[a] | 1 | 50 | 120 | 40 | 300 | 800 | 20 | 3 | 2[a] | 1 | 70 | 150 | 80 | - | - | - | - |
| Sewage Sludge | 20-40 | - | 16-25 | 300-400 | 750-1200 | - | 1000-1750 | 2500-4000 | 41-75 | 39-85 | 1000-3000 | 17-57 | 420 | 300-840 | 36-100 | 2800-7500 | 1500-4300 | 1300 | 50-75 |

[a] Value in hexavalent chromium.

**References**

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**Table S1.4** – Characteristics of meta-analyses by subgroup: number of studies, heterogeneity ($I^2$ and Q) and variance between studies ($\tau^2$).

| Element | Fertilizer | Nº of studies (k) | $I^2$ (%) | Q | $\tau^2$ |
|---|---|---|---|---|---|
| As | Cattle Manure | 8 | 100.0 | 54818.33 | 5.05 |
| As | Chicken Manure | 5 | 85.3 | 85.04 | 0.01 |
| As | Compost | 12 | 100.0 | 186221.60 | 500.83 |
| As | Silicon Fertilizer | 4 | 99.7 | 889.88 | 3.20 |
| As | Soil Conditioner | 1 | - | 0.00 | - |
| As | Industrial Waste | 2 | 100.0 | 4794.47 | 23.12 |
| As | Limestone | 1 | - | 0.00 | - |
| As | Phosphate Fertilizer | 1 | - | 0.00 | - |
| As | Potassium Fertilizer | 1 | - | 0.00 | - |
| As | Sewage Sludge | 1 | - | 0.00 | - |
| As | Swine Manure | 1 | - | 0.00 | - |
| As | Urea | 1 | - | 0.00 | - |
| Cd | Cattle Manure | 10 | 99.9 | 11946.44 | 2.63 |
| Cd | Chicken Manure | 5 | 99.4 | 694.95 | 0.03 |
| Cd | Compost | 18 | 100.0 | 70855.81 | 1.23 |
| Cd | Industrial Waste | 4 | 95.9 | 72.52 | 0.01 |
| Cd | Limestone | 4 | 99.8 | 1387.29 | 0.94 |
| Cd | Phosphate Fertilizer | 2 | 85.4 | 6.86 | 0.00 |
| Cd | Phosphate Rock | 2 | 97.7 | 44.00 | 97.84 |
| Cd | Potassium Fertilizer | 2 | 97.9 | 47.64 | 0.00 |
| Cd | Sewage Sludge | 15 | 99.8 | 5899.38 | 1.03 |
| Cd | Silicon Fertilizer | 4 | 89.5 | 28.59 | 0.01 |
| Cd | Soil Conditioner | 1 | -- | 0.00 | -- |
| Cd | Swine Manure | 6 | 100.0 | 58394.90 | 9.47 |
| Cd | Urea | 2 | 100.0 | 2350.11 | 0.00 |
| Co | Cattle Manure | 2 | 0.0 | 0.13 | 0.00 |
| Co | Chicken Manure | 2 | 91.1 | 11.26 | 0.07 |
| Co | Limestone | 1 | - | 0.00 | - |
| Cr | Cattle Manure | 13 | 99.8 | 5951.62 | 45.18 |
| Cr | Chicken Manure | 7 | 100.0 | 160972.75 | 34499.66 |
| Cr | Compost | 21 | 100.0 | 48446.77 | 2874.11 |
| Cr | Silicon Fertilizer | 2 | 99.8 | 498.46 | 40.42 |
| Cr | Industrial Waste | 2 | 99.6 | 275.98 | 310.62 |
| Cr | Limestone | 1 | - | 0.00 | - |
| Cr | Phosphate Fertilizer | 1 | - | 0.00 | - |
| Cr | Potassium Fertilizer | 1 | - | 0.00 | - |
| Cr | Sewage Sludge | 9 | 98.7 | 620.76 | 2066.73 |
| Cr | Swine Manure | 3 | 93.4 | 30.53 | 80.98 |
| Cr | Urea | 1 | - | 0.00 | - |

| | | | | | |
|---|---|---|---|---|---|
| Cu | Industrial Waste | 6 | 99.8 | 2549.95 | 180.07 |
| Cu | Swine Manure | 11 | 100.0 | 53944.95 | 7663.20 |
| Cu | Sewage Sludge | 17 | 99.7 | 5025.71 | 22424.99 |
| Cu | Chicken Manure | 10 | 100.0 | 20926.07 | 1495.48 |
| Cu | Cattle Manure | 15 | 100.0 | 91413.10 | 10796.33 |
| Cu | Potassium Fertilizer | 3 | 99.9 | 1951.53 | 1.87 |
| Cu | Compost | 20 | 100.0 | 387632.69 | 8684.87 |
| Cu | Urea | 2 | 100.0 | 59366.33 | 154.35 |
| Cu | Phosphate Fertilizer | 2 | 99.7 | 314.48 | 13.07 |
| Cu | Limestone | 2 | 99.2 | 129.77 | 9.13 |
| Fe | Cattle Manure | 8 | 100.0 | 52238.62 | 737552.40 |
| Fe | Chicken Manure | 4 | 98.8 | 257.67 | 2474488.40 |
| Fe | Compost | 9 | 100.0 | 742196.28 | 20835913.45 |
| Fe | Industrial Waste | 3 | 99.9 | 2239.14 | 13277830.61 |
| Fe | Limestone | 1 | - | 0.00 | - |
| Fe | Sewage Sludge | 6 | 98.8 | 415.44 | 91.64 |
| Fe | Swine Manure | 2 | 94.7 | 18.74 | 445348.96 |
| Hg | Cattle Manure | 5 | 99.9 | 3484.71 | 0.00 |
| Hg | Chicken Manure | 3 | 0.0 | 0.31 | 0.00 |
| Hg | Industrial Waste | 1 | - | 0.00 | - |
| Mn | Cattle Manure | 9 | 99.5 | 1714.63 | 9453.16 |
| Mn | Chicken Manure | 4 | 99.7 | 929.05 | 38763.04 |
| Mn | Compost | 6 | 99.7 | 1668.03 | 16564.03 |
| Mn | Industrial Waste | 2 | 99.4 | 171.37 | 4.93 |
| Mn | Limestone | 1 | - | 0.00 | - |
| Mn | Sewage Sludge | 6 | 96.5 | 144.75 | 2210.35 |
| Mn | Swine Manure | 2 | 99.4 | 166.74 | 142147.88 |
| Ni | Cattle Manure | 6 | 99.9 | 4661.66 | 256.34 |
| Ni | Chicken Manure | 7 | 99.8 | 3310.54 | 150.84 |
| Ni | Compost | 13 | 99.9 | 20259.56 | 76.45 |
| Ni | Industrial Waste | 1 | - | 0.00 | - |
| Ni | Limestone | 1 | - | 0.00 | - |
| Ni | Phosphate Fertilizer | 1 | - | 0.00 | - |
| Ni | Potassium Fertilizer | 1 | - | 0.00 | - |
| Ni | Sewage Sludge | 2 | 97.5 | 39.24 | 943.33 |
| Ni | Swine Manure | 3 | 98.9 | 189.31 | 196.90 |
| Ni | Urea | 1 | - | 0.00 | - |
| Pb | Cattle Manure | 11 | 100.0 | 27227.84 | 22.96 |
| Pb | Chicken Manure | 9 | 100.0 | 27865.49 | 252.31 |
| Pb | Compost | 19 | 100.0 | 98021.88 | 195.60 |
| Pb | Silicon Fertilizer | 2 | 96.1 | 25.59 | 8.88 |
| Pb | Soil Conditioner | 1 | - | 0.00 | - |
| Pb | Industrial Waste | 4 | 98.3 | 175.73 | 23.77 |
| Pb | Limestone | 2 | 90.9 | 10.99 | 161.15 |

| | | | | | |
|---|---|---|---|---|---|
| Pb | Phosphate Fertilizer | 1 | - | 0.00 | - |
| Pb | Potassium Fertilizer | 2 | 97.2 | 35.75 | 0.51 |
| Pb | Sewage Sludge | 16 | 99.9 | 20165.13 | 1268.28 |
| Pb | Swine Manure | 6 | 100.0 | 31314.44 | 158.34 |
| Pb | Urea | 2 | 95.7 | 23.39 | 0.57 |
| Zn | Cattle Manure | 15 | 99.9 | 10202.23 | 7567.12 |
| Zn | Chicken Manure | 10 | 100.0 | 26683.99 | 47604.11 |
| Zn | Compost | 22 | 100.0 | 237640023014.46 | 2543268.42 |
| Zn | Phosphate Rock | 2 | 97.7 | 43.21 | 2532.01 |
| Zn | Industrial Waste | 5 | 99.9 | 5144.52 | 2255.98 |
| Zn | Limestone | 2 | 99.3 | 135.10 | 47.08 |
| Zn | Phosphate Fertilizer | 2 | 99.8 | 641.62 | 393.63 |
| Zn | Potassium Fertilizer | 3 | 100.0 | 4471.25 | 31.79 |
| Zn | Sewage Sludge | 17 | 99.9 | 10962.61 | 84937.97 |
| Zn | Swine Manure | 11 | 100.0 | 52143.25 | 1340599.83 |
| Zn | Urea | 2 | 100.0 | 130799.62 | 2461.19 |

**Appendix B –** Supplementary Material: Potentially Toxic Elements in Fertilizers and Soil Amendments of the Brazilian Cerrado

**Table S2.1 –** Complete data on environmentally available contents of Potentially Toxic Elements by fertilizer type (organic, organomineral, and mineral) extracted by EPA 3051A.

| Fertilizer | As | B | Cd | Cr | Cu | Mo | Ni | Pb | Zn | Co |
|---|---|---|---|---|---|---|---|---|---|---|
| | mg/kg | | | | | | | | | |
| Sugarcane Filter Cake | < DL | 12.8±0.1 | < DL | 19.6±1.3 | 48.3±2.3 | 5.1±5.0 | 5.5±1.5 | < DL | 178.5±4.1 | 40.0±5.5 |
| Vinasse | < DL | < DL | < DL | < DL | 0.5±0.2 | < DL | < DL | < DL | 1.2±0.1 | 9.7±4.2 |
| Pig slurry | < DL | < DL | < DL | < DL | 1.11±0.98 | < DL | < DL | < DL | 6.75±5.47 | 12.6±9.5 |
| Cattle Manure | < DL | < DL | < DL | 6.3±0.9 | 30.0±3.3 | < DL | 3.7±0.3 | < DL | 4.2±2.0 | 8.6±5.5 |
| Poultry Litter | < DL | 1323.9±375.6 | < DL | 39.8±0.5 | 34.2±1.1 | 2.2±0.0 | 6.2±0.6 | < DL | 117.7±4.7 | 3.9±1.2 |
| Organomineral I | < DL | 76.6±15.9 | 0.7±0.13 | 5.1±1.1 | 481.2±67.4 | 6.6±0.8 | 3.7±0.5 | < DL | 384.3±61.4 | < DL |
| Organomineral II | < DL | 40.8±3.0 | < DL | 20.4±3.8 | 53.0±2.1 | 3.6±0.2 | 4.2±0.4 | < DL | 272.5±19.0 | 10.2±1.8 |
| Organomineral III | < DL | 30.6±1.5 | < DL | 22.8±7.7 | 148.1±9.5 | 2.7±0.2 | 4.8±0.3 | < DL | 239.8±10.0 | 6.0±5.7 |
| Organomineral IV | < DL | 20.7±1.5 | < DL | 23.6±4.4 | 73.6±1.2 | 1.8±0.3 | 6.6±1.2 | 21.4±1.1 | 120.1±2.3 | 10.1±6.7 |
| Organomineral V | < DL | 28.9±1.3 | < DL | 20.6±7.8 | 36.8±3.3 | 1.9±0.2 | 9.9±2.2 | 9.6±4.0 | 223.5±5.8 | 14.4±3.1 |
| Organomineral VI | < DL | 121.8±12.5 | 1.1±0.1 | 29.8±3.5 | 37.5±2.1 | 2.4±0.2 | 17.6±2.3 | 9.2±1.7 | 202.5±10.8 | 11.3±2.0 |
| SSP 00-19 | < DL | 20.2±1.5 | 3.7±0.1 | 72.3±0.6 | 14.5±4.4 | 45.0±0.1 | 13.0±0.3 | < DL | 166.3±0.3 | 34.7±11.7 |
| SSP 00-21 | 82.3±6.1 | 14.5±1.4 | < DL | 20.2±0.2 | 98.7±1.5 | 4.6±0.3 | 52.4±0.4 | 58.3±4.3 | 128.1±1.7 | 87.4±10.7 |
| NPK 04-14-08 | < DL | 13.9±0.7 | < DL | 5.5±0.4 | 4.4±0.4 | 1.1±0.2 | < DL | < DL | 17.7±0.4 | 11.1±3.0 |
| NPK 05-25-15 | 18.1±1.3 | 34.6±0.5 | 0.7±0.0 | 15.4±0.8 | 36.8±2.7 | 2.5±0.2 | 12.3±0.5 | 39.6±4.6 | 489.2±29.1 | 32.4±3.9 |
| NPK 06-30-10 | < DL | 24.1±5.3 | 0.7±0.0 | 16.3±0.2 | 18.1±0.7 | 3.2±0.4 | 7.3±0.1 | < DL | 75.6±2.7 | 9.3±0.2 |
| MAP 11-52 | 20.7±3.9 | 89.6±21.9 | 22.7±0.3 | 240.4±8.1 | 41.7±0.9 | 4.7±0.6 | 37.1±0.7 | < DL | 356.3±8.8 | 3.6±0.1 |
| TSP | < DL | 23.9±2.3 | < DL | 6.7±0.4 | 4.4±0.2 | < DL | 1.5±0.2 | < DL | 11.8±0.4 | 3.3±0.2 |
| FTE BR-12 | 111.8±7.1 | 15022.1±916.1 | 141.9±1.0 | 600.6±86.6 | 4773.8±369.5 | 27.8±0.6 | 106.1±12.5 | 11553.5±474.2 | 84553.7±1698.7 | 155.1±12.3 |
| Limestone I | < DL | - | < DL | 1.8±0.5 | 1.0±0.2 | - | 1.4±0.0 | < DL | 10.5±1.6 | - |
| Limestone II | < DL | - | < DL | 1.5±0.1 | 7.3±11.8 | - | 1.3±0.0 | < DL | 3.6±0.3 | - |

| Fertilizer | Al | Ca | Fe | Mg | Mn | P | S |
|---|---|---|---|---|---|---|---|
| | g/kg | | | | | | |
| Sugarcane Filter Cake | 24.71±2.22 | 9.76±0.31 | 25.8±0.85 | 3.59±0.05 | 0.16±0.00 | 2.5±0.05 | 1.65±0.05 |
| Vinasse | 0.06±0.06 | 0.21±0.01 | 0.07±0.06 | 0.17±0.00 | 0.00±0.00 | 0.15±0.01 | 0.34±0.01 |
| Pig slurry | 0.01±0.00 | 0.13±0.05 | 0.01±0.01 | 0.06±0.01 | < DL | 0.05±0.02 | 0.04±0.01 |
| Cattle Manure | 9.72±1.17 | 0.07±0.01 | 0.29±0.03 | 0.04±0.00 | 0.00±0.00 | 33.23±1.18 | 72.43±9.88 |
| Poultry Litter | 17.66±0.64 | 15.38±0.57 | 22.54±1.69 | 31±1.90 | 0.49±0.01 | 3.76±0.18 | 4.38±0.35 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| Organomineral I | 0.21±0.07 | 35.5±2.10 | 0.2±0.09 | 4.21±0.66 | 0.49±0.09 | 7.19±0.4 | 6.92±0.22 |
| Organomineral II | 13.01±0.34 | 9.98±0.91 | 8.08±1.38 | 4.62±0.18 | 0.55±0.03 | 7.41±0.06 | 6.64±0.1 |
| Organomineral III | 13.86±1.20 | 1.51±0.07 | 16.87±3.45 | 4.23±0.08 | 0.41±0.10 | 4.59±0.23 | 3.59±0.16 |
| Organomineral IV | 13.94±1.10 | 14.67±0.64 | 15.14±3.11 | 2.13±0.02 | 1.85±0.09 | 10.29±0.2 | 2.73±0.06 |
| Organomineral V | 11.53±2.65 | 40.28±0.73 | 11.01±2.62 | 4.35±0.34 | 0.86±0.06 | 6.5±0.13 | 3.06±0.02 |
| Organomineral VI | 32.15±6.40 | 2.12±0.58 | 16.81±1.33 | 1.81±0.13 | 2.48±0.10 | 84.49±5.8 | 15.92±1.09 |
| SSP 00-19 | 3.11±0.05 | 213.67±2.7 | 9.75±0.11 | 1.06±0.01 | 0.26±0.00 | 91.72±0.29 | 153.1±3.87 |
| SSP 00-21 | 5.27±0.12 | 158.29±1.7 | 32.12±0.58 | 6.01±0.06 | 1.29±0.01 | 132.84±3.52 | 90.35±2.26 |
| NPK 04-14-08 | 2.1±0.09 | 174.35±0.61 | 2.6±0.06 | 6.72±0.04 | 0.13±0.01 | 64.08±1.15 | 43.82±1.51 |
| NPK 05-25-15 | 2.84±0.05 | 38.59±2.47 | 8.24±0.47 | 2.28±0.04 | 0.46±0.01 | 153.63±4.30 | 71.73±3.25 |
| NPK 06-30-10 | 3.57±0.38 | 6.39±1.08 | 5.62±0.52 | 2.86±0.04 | 0.47±0.01 | 205.32±14.03 | 26.1±1.30 |
| MAP 11-52 | 1.46±0.21 | 163.29±3.87 | 2.8±0.10 | 4.28±0.10 | 0.03±0.00 | 207.28±3.51 | 13.37±0.21 |
| TSP | 0.8±0.14 | 59.39±2.28 | 2.17±0.08 | 3.6±0.10 | 0.19±0.01 | 104.25±3.99 | 12.36±0.36 |
| FTE BR-12 | 0.16±0.01 | 30.27±1.89 | 109.75±10.43 | 14.06±0.35 | 17.1±0.25 | 3.84±0.43 | 65.15±3.84 |
| Limestone I | - | - | - | - | - | - | - |
| Limestone II | - | - | - | - | - | - | - |

< DL – Below the detection limit. Hg and Se were below the detection limit and were removed from the table for better visualization. Detection limit (mg/kg): Al (<4.5), As (<11.3), B (<12.2), Cd (<0.4), Co (<1.0), Cr (<0.5), Fe (<1), Hg (<15), Mn (<0.8), Mo (1.3), Ni (1.3), Pb (<5), Se (<13.6).

**Table S2.2 –** Complete data on Water-available contents of Potentially Toxic Elements by fertilizer type (organic, organomineral, and mineral).

| Fertilizer | As | B | Cd | Cr | Cu | Mo | Ni | Pb | Zn |
|---|---|---|---|---|---|---|---|---|---|
| | mg/kg | | | | | | | | |
| Sugarcane Filter Cake | < DL | < DL | < DL | < DL | 2.8±0 | 2.4±1.5 | < DL | < DL | 178.5±4.1 |
| Vinasse | < DL | < DL | < DL | < DL | 0.4±0.2 | < DL | < DL | < DL | 1.2±0.1 |
| Pig slurry | < DL | < DL | < DL | < DL | 0.5±0.1 | < DL | < DL | < DL | 6.8±5.5 |
| Cattle Manure | < DL | 1323.9±375.6 | < DL | 0.5±0.0 | 2.5±0.9 | 2.2±0 | 6.2±0.6 | < DL | 22.4±11.6 |
| Poultry Litter | < DL | 155.5±64.5 | < DL | 1.2±0 | 281.7±1.5 | 6.6±0.8 | 3.7±0.5 | < DL | 79.8±1.4 |
| Organomineral I | < DL | 56.3±27.8 | < DL | 1.2±0 | 17.2±0.4 | 3.5±0.2 | 4.2±0.4 | < DL | 58.1±1 |
| Organomineral II | < DL | 117.6±43.4 | < DL | 0.8±0.1 | 43.6±0.1 | 2.7±0.2 | 4.8±0.3 | < DL | 31.5±0.6 |
| Organomineral III | < DL | 55.5±1.6 | < DL | < DL | 4.1±0.3 | 1.8±0.3 | 6.6±1.1 | 21.4±1.1 | 2±0.5 |
| Organomineral IV | < DL | 61.8±1 | < DL | 3.5±0.2 | 10.6±0.3 | 1.9±0.2 | 9.9±2.2 | 9.6±4 | 42±0.2 |
| Organomineral V | < DL | 117.8±0.6 | < DL | 1.4±0 | 0.3±0.2 | 2.4±0.2 | 17.6±2.3 | 9.2±1.7 | 0.06±0.0 |
| Organomineral VI | < DL | < DL | < DL | < DL | 0.7±0.1 | < DL | < DL | < DL | 4.2±2 |
| SSP 00-19 | < DL | < DL | 3.7±0.1 | 10.1±0.5 | 6.3±0.4 | < DL | 14.5±0.1 | < DL | 162.5±2.8 |
| SSP 00-21 | < DL | < DL | < DL | 2.1±0.1 | 65.9±63.8 | < DL | 42.3±0.2 | < DL | 103.4±16 |
| NPK 04-14-08 | < DL | < DL | < DL | 1.7±0 | 0.4±0.2 | < DL | < DL | < DL | < DL |
| NPK 05-25-15 | < DL | < DL | 0.7±0 | 4.4±0.1 | 6.8±0.2 | < DL | 10.7±0.1 | < DL | 175.7±3.3 |
| NPK 06-30-10 | < DL | 38±2.3 | < DL | 3.5±0.1 | 0.7±0.1 | < DL | 1.7±0.2 | < DL | 0.5±0.2 |
| MAP 11-52 | 15,91±1,86 | 49.7±3.7 | < DL | 6.9±0 | 1.2±0.2 | < DL | 4.3±0.3 | < DL | 13.6±0.1 |
| TSP | < DL | 57.2±4.3 | 17.3±0.8 | 28.4±1.2 | 27.3±1.7 | 2.3±0.2 | 38.4±2.1 | < DL | 314.5±15.4 |
| FTE BR-12 | 2,48±2,24 | 7758.3±168.2 | 2.2±0.8 | 1.2±0.1 | 2.1±0.3 | 2.9±0.2 | 4.3±0.3 | 7.5±1.4 | 19171.8±471.6 |

| Fertilizer | Al | Ca | Co | Fe | Mg | Mn | P | S |
|---|---|---|---|---|---|---|---|---|
| | mg/kg | | | | | | | |
| Sugarcane Filter Cake | 10.8±2.4 | 467.3±8 | < DL | 13.7±1.3 | 1223.4±10.3 | < DL | 339.2±3.2 | 211.2±7.1 |
| Vinasse | < DL | 219.4±2.4 | < DL | 7.4±0.7 | 207.5±2.2 | 2.7±0.2 | 131.7±1.3 | 352.8±8.4 |
| Pig slurry | < DL | 50.3±12.6 | < DL | 0.2±0.2 | 48.2±4.3 | < DL | 28.8±4.7 | 23.2±9.4 |
| Cattle Manure | 95.9±21.3 | 360.7±5.5 | < DL | 22535.7±1688.7 | 254±16.6 | 7.8±2.9 | 390.3±24.3 | 1565.7±22.8 |
| Poultry Litter | < DL | 1777.4±33.7 | < DL | 200±91.2 | 1242.4±20.3 | 23.2±0.4 | 2528.7±24.8 | 6924.1±222 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Organomineral I | 244.9±4.1 | 728.4±2 | 1.3±0.1 | 8079.9±1380.5 | 1151.8±24.9 | 35.8±0.6 | 7409.1±62.3 | 6642.3±100.1 |
| Organomineral II | 192.7±3.9 | 280.9±4.2 | < DL | 16872.4±3452.8 | 684.4±25.4 | 11.6±0.2 | 4593.3±231.4 | 1804.5±10 |
| Organomineral III | 207.2±60.8 | 89±16.9 | < DL | 15136.1±3111 | 107.3±11 | 25.1±4.3 | 337.4±25.4 | 782.3±27.4 |
| Organomineral IV | 11531.3±2650.1 | 563.1±11.3 | 3±0 | 11011.3±2620.1 | 821.7±12.8 | 54.3±1.1 | 1395.7±16.4 | 2328.1±24.8 |
| Organomineral V | < DL | 528.8±4.1 | < DL | 16805.4±1332.6 | 381.3±7.7 | 32.4±1.1 | 84486.3±5796.7 | 15918.9±1092.7 |
| Organomineral VI | 128.6±13.4 | 19.1±10.6 | < DL | 6.4±0.6 | 4.6±2.1 | < DL | 1418.2±22.5 | 3153.8±56.4 |
| | ..... | | | | | | | |
| SSP 00-19 | 322.9±14.8 | 78122.7±1253 | 1.7±0.1 | 190.1±0.8 | 1163±17 | 235.6±5 | 75026.9±687.3 | 36192.6±1185.2 |
| SSP 00-21 | 610±32.2 | 53608.9±4459.7 | 21±0.2 | 35.4±6 | 5800±70.8 | 991.9±11.1 | 65949.5±1557.7 | 33469.5±2619.3 |
| NPK 04-14-08 | 3±0.6 | 4452.9±102.1 | < DL | < DL | 200.9±2.1 | 26.4±0.1 | 63208.6±848.1 | 45256.6±713.2 |
| NPK 05-25-15 | 291.6±6.6 | 16363.7±396.5 | 4.4±0.1 | 2.6±0.2 | 1120.8±13.6 | 199.1±1.3 | 150114.7±600 | 67279.8±2371.8 |
| NPK 06-30-10 | 3±0.2 | 2181.8±269.7 | 1.4±0.1 | < DL | 151.8±13.3 | 34.6±2.3 | 115665±949 | 13500.3±509.5 |
| MAP 11-52 | 940.4±11.4 | 1639.7±53.9 | 2.1±0 | 12.1±1.2 | 701.6±13.5 | 157.2±2.1 | 210037.7±838.8 | 25313.6±360 |
| TSP | 569.8±21.5 | 115157.7±2050.5 | 1.7±0.2 | 229.3±16.3 | 3799.9±325.1 | 24.5±0.9 | 192682.7±4448.1 | 13646.6±383.9 |
| FTE BR-12 | 1.4±0.5 | 45.3±28.5 | 3.1±0.2 | < DL | 219.6±66.1 | 59.7±28.6 | 39.7±6.3 | 53300.1±86.1 |

< DL – Below the detection limit. Hg and Se were below the detection limit and were removed from the table for better visualization. Detection limit (mg/kg): Al (<4.5), As (<11.3), B (<12.2), Cd (<0.4), Co (<1.0), Cr (<0.5), Fe (<1), Hg (<15), Mn (<0.8), Mo (1.3), Ni (1.3), Pb (<5), Se (<13.6).

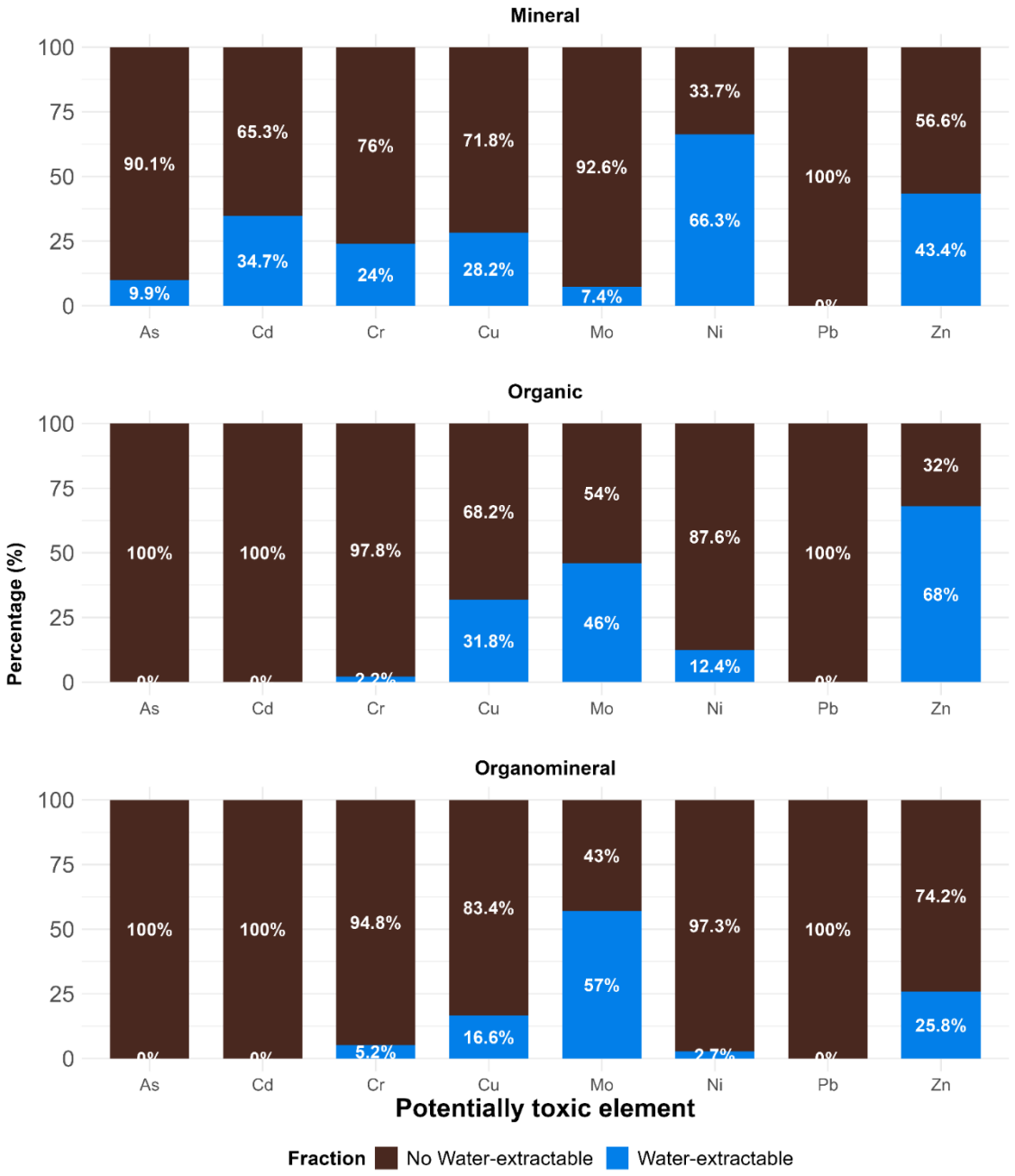


**Figure S2.1 –** Proportion of water-available and non-water-available contents of Potentially Toxic Elements, considering fertilizer groups.

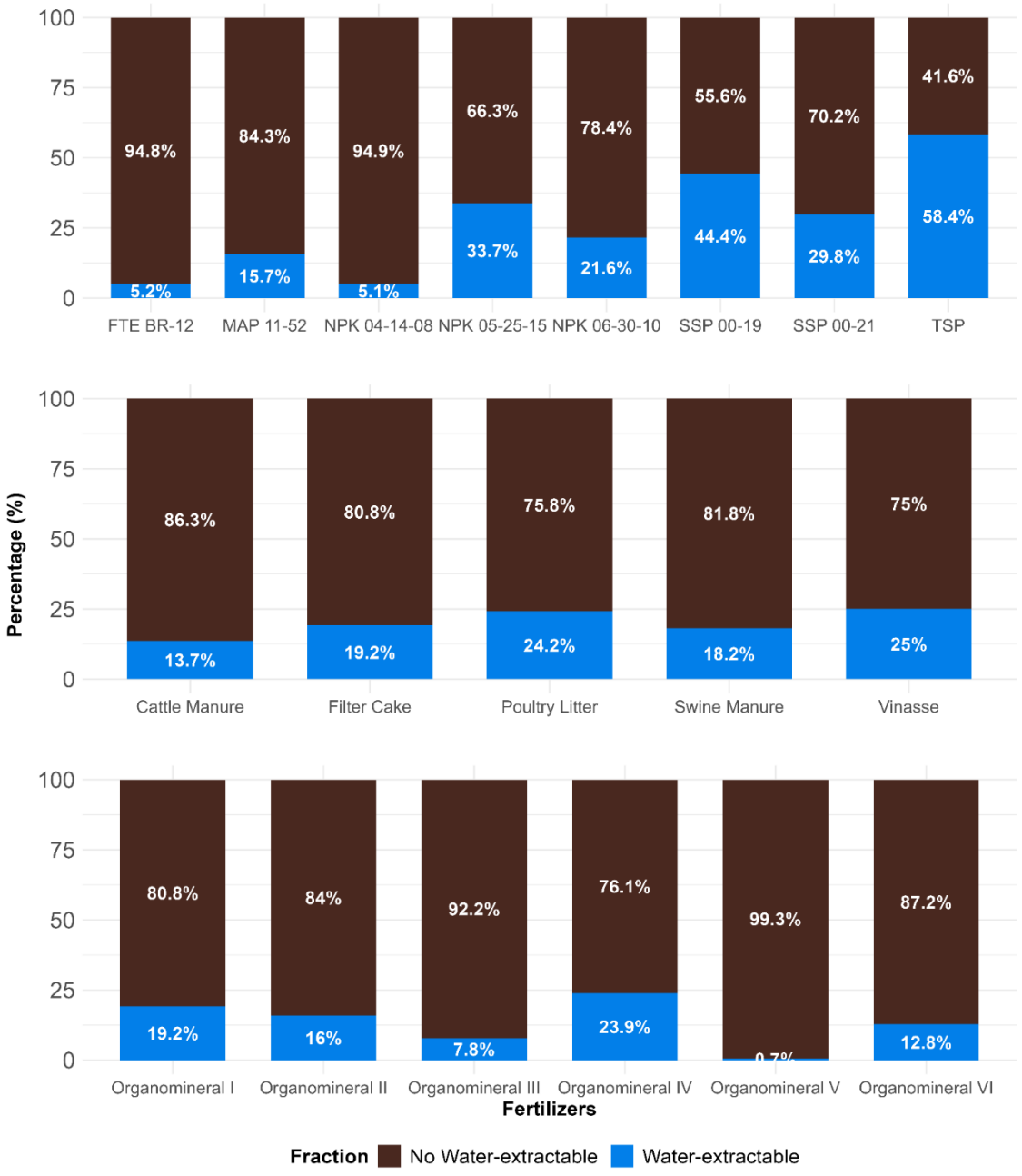


**Figure S2.2 –** Proportion of water-available and non-water-available contents of As, Cd, Cr, Cu, Mo, Ni, Pb, and Zn in individual fertilizers.

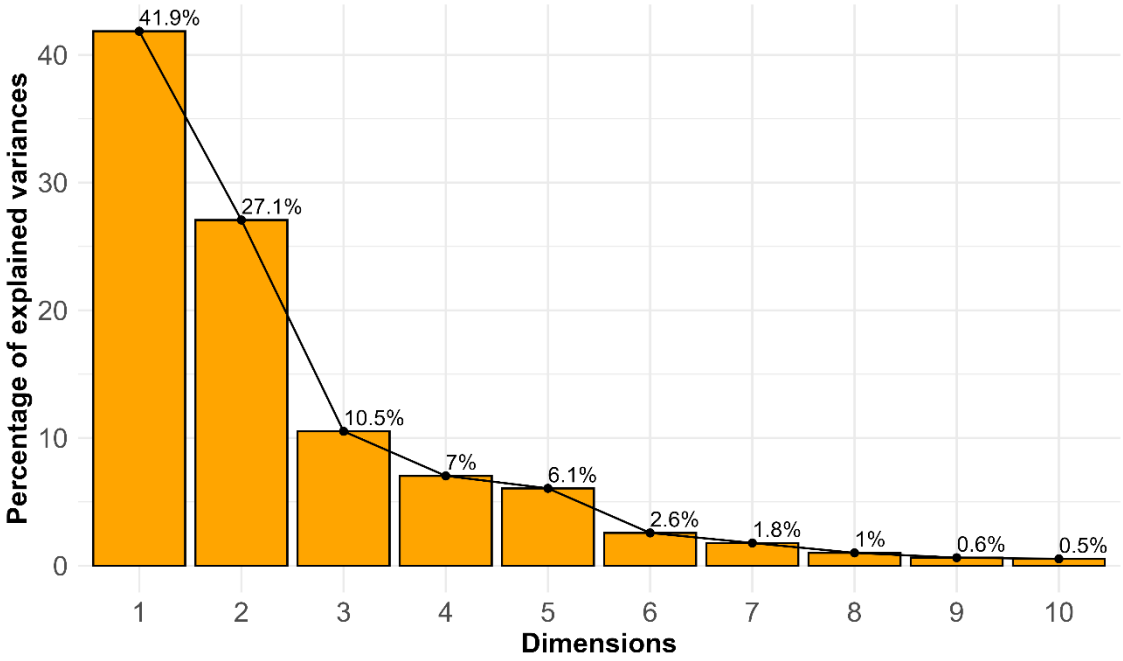


**Figure S2.3** – Percentage of variance explained by each principal component in the PCA for the variables of application rate, total contents, and risk quotient for soil organisms.

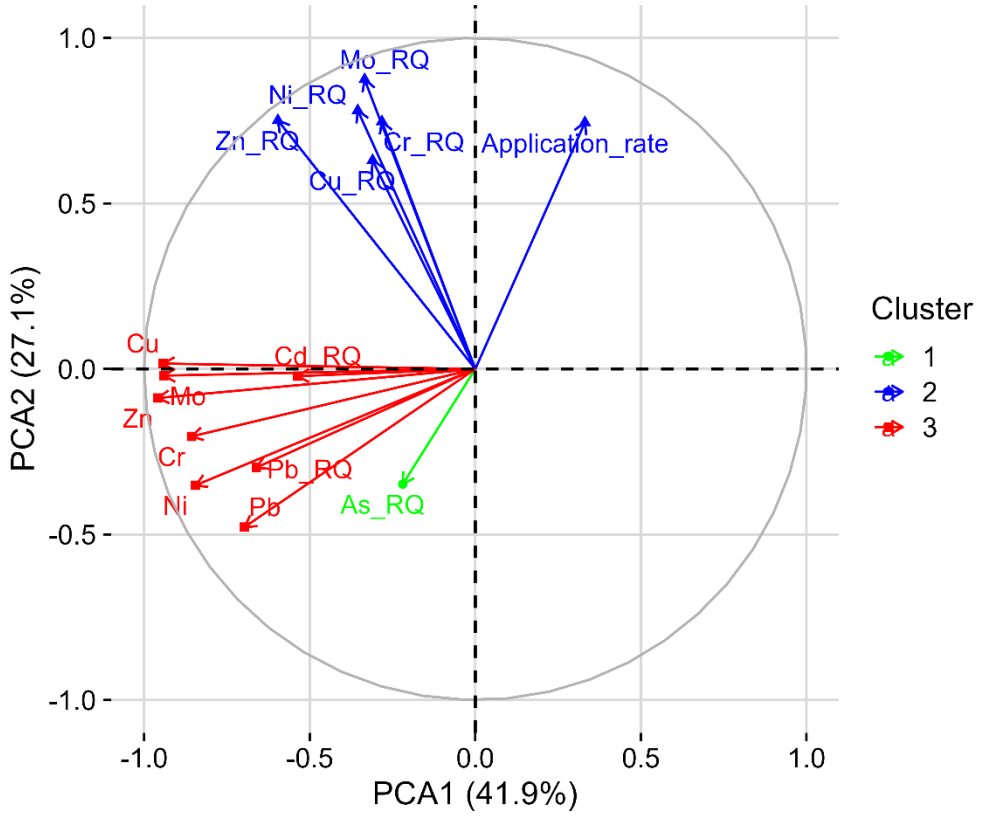


**Figure S2.4** – Clustering of variables for the sampled fertilizers. Colors differentiate the clusters formed based on similarity among variables (*k-means*).

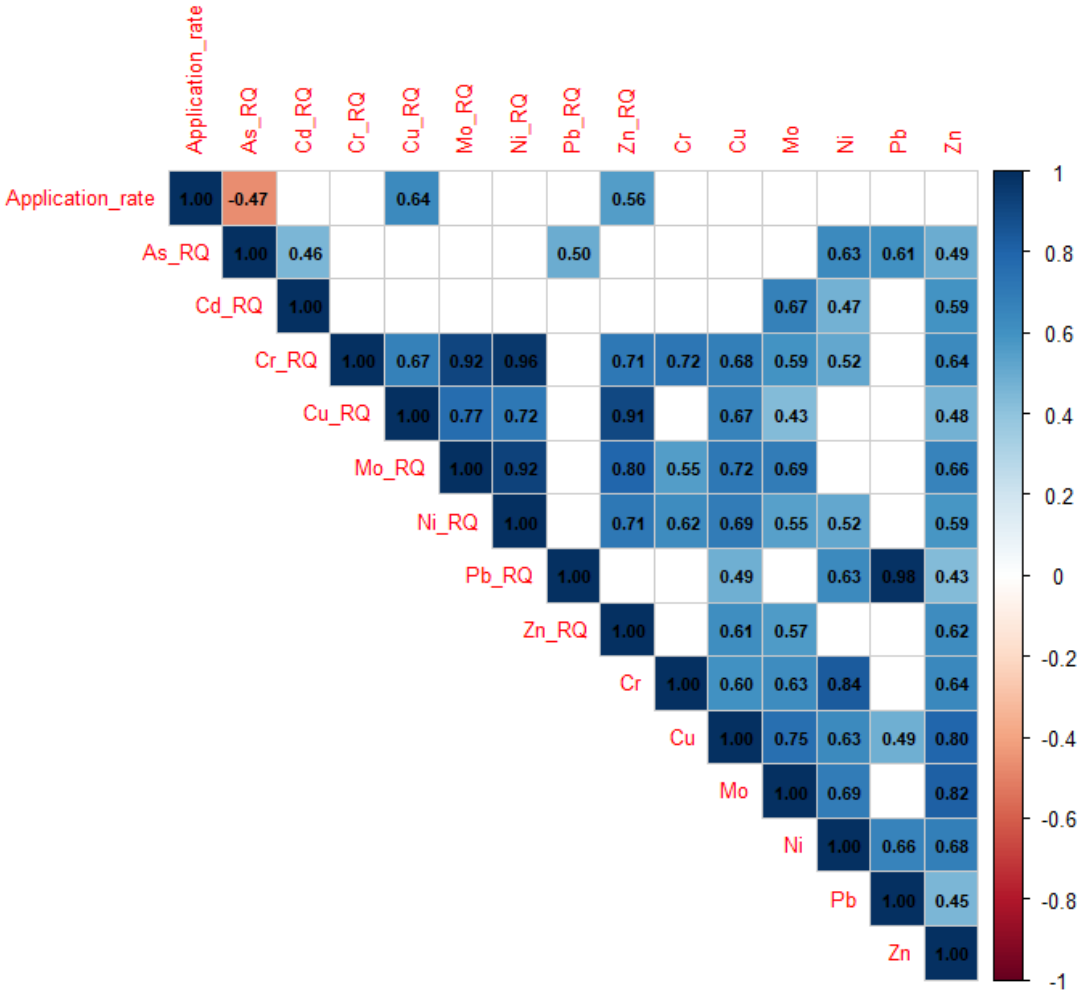


**Figure S2.5 –** Spearman correlation matrix among total PTE contents, fertilizer application rate, and risk quotient (RQ) for soil organisms. Numbers in blue indicate positive correlation, while numbers in red indicate negative correlation. White indicates non-significant correlations ($p > 0.05$).

**Table S2.3** – Loading matrix of the first two principal components for the variables: fertilizer application rate, total PTE contents, and risk quotients (RQ). Loadings were grouped by the ecotoxicological profile (PC2) of the fertilizers.

| Variable | PC1[a] | PC2[b] |
|---|---|---|
| *Mineral Fertilizer* | | |
| As_RQ | -0,09 | -0,17 |
| Cd_RQ | -0,21 | -0,01 |
| Cr | -0,34 | -0,10 |
| Mo | -0,38 | -0,01 |
| Ni | -0,34 | -0,17 |
| Pb | -0,28 | -0,24 |
| Pb_RQ | -0,26 | -0,15 |
| Zn | -0,38 | -0,04 |
| *Organic and Organomineral Fertilizers* | | |
| Application_rate | 0,13 | 0,37 |
| Cr_RQ | -0,11 | 0,37 |
| Cu | -0,38 | 0,01 |
| Cu_RQ | -0,12 | 0,31 |
| Mo_RQ | -0,13 | 0,43 |
| Ni_RQ | -0,14 | 0,39 |
| Zn_RQ | -0,24 | 0,37 |

a – PC1 reflects the relationship between PTEs and application rate in the dataset; b – PC2 reflects the ecotoxicological risk profile among fertilizers.