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Hotspots of soil pollution: Possible glyphosate and aminomethylphosphonic acid risks on terrestrial ecosystems and human health

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ABSTRACT

The study presents a literature review of glyphosate (GLY) occurrence and its breakdown product, aminomethylphosphonic acid (AMPA), in soils worldwide, but with a specific focus on South America. In addition, an ecological risk approach based on the ecotoxicological endpoints for key soil biota (e.g., collembolans, and earthworms) assessed the impact of GLY and AMPA on these organisms. A generic probabilistic model for human health risk was also calculated for the different world regions. For what reports the risk for edaphic species and the level of pollution under the worst-case scenario, the South American continent was identified as the region of most concern. Nonetheless, other areas may also be in danger, but no risk could be calculated due to the lack of data. Since tropical countries are the top food exporters worldwide, the results obtained in this study must be carefully examined for their implications on a global scale. Some of the factors behind the high levels of these two chemicals in soils are debated (e.g., permissive protection policies, the extensive use of genetically modified crops), and some possible guidelines are presented that include, for example, further environmental characterisation and management of pesticide residues. The present review integrates data that can be used as a base by policymakers and decision-makers to develop and implement environmental policies.

1. Introduction

Anthropogenic practices have altered three-quarters of the Earth's surface, with more than a third dedicated exclusively to crop and/or livestock production (IPBES, 2019). Despite being a huge percentage of land use, food production does not necessarily link it to a negative factor or problem to focus on. The incorrect use and management of these areas are, in fact, the main concerns and the problem that needs to be addressed. Land misuse is, directly and indirectly, the main factor that accelerates biodiversity loss (IPBES, 2019), connected with other factors such as the misuse of pesticides in agricultural practices. Despite the increasing research focused on using alternative sustainable practices

Abbreviations: ABS, dermal absorption factor; ABS_{GI}, fraction absorbed in the gastrointestinal tract; ADD_i, average daily dose by *i* exposure route pathway; AF, assessment factor; AMPA, aminomethylphosphonic acid; AT, average life span of an adult; BCF, bioconcentration factor; BW, bodyweight of the individual adult; CF, conversion factor; C_{soil}, concentration of the pollutant on soil; DAF, dermal adhesion factor; ED, exposure duration; EF, exposure frequency; ERA, ecological risk assessment; GLY, glyphosate; GMO, genetically modified organism; HI, hazard index of non-cancer; HRA, human risk assessment; ILCR, incremental lifetime carcinogenic risk; IR_i, ingestion or inhalation rate by *i* exposure route pathway; MEC, measured environmental concentration; PEF, particle emission factor; PNEC, predicated no-effect environmental concentration; RfD_i, daily maximum permissible values by *i* exposure route pathways; RQ, risk quotient; SA, surface area of the skin that contact with the soil; SDG, Sustainable Development Goals; SF_i, slope factor by *i* exposure route pathway.

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that can integrate and increase ecosystems' health, there is still no general solution. At the same time, decreasing pesticide use would also lower possible threats to non-target organisms, as already widely reported in the literature (e.g., Domínguez et al., 2016; Ferreira et al., 2015). Some public policies to reduce the application of pesticides, such as the EU Farm to Fork Strategy (Directive 2009/128/EC), have already been adopted to tackle this problem. The latest numbers of pesticide use (Table 1 - Food and Agriculture Organization of the United Nations, 2018) show what may be even the start of a reduction in herbicide use in Europe, with a 1% reduction in the past ten years. Nonetheless, even with these public policies, it is expected that the use of pesticides will continue to occur in the following years and may even increase, at least when looking into low and middle-income countries (LMICs). Such increase may be the main reason for many possible environmental impacts and human health problems, all resulting from the excessive use and misuse of these chemicals, but also due to their highly persistent in the environment or hazardousness. The annual use of pesticides for agricultural purposes worldwide is > 2.5 million tons, with China, the United States (US), Brazil, and Argentina as the countries that most use these substances. N-[phosphomethyl]-glycine ($C_3H_8NO_5P$ - GLY) based herbicides are the best-selling products worldwide due to their low cost, broad-spectrum mechanism, effectiveness, the requirement for a large number of genetically modified organisms (GMO) and the application of those products as desiccants (Duke and Powles, 2009). GLY represents the largest export in China, the most used agricultural land in the US, and the most widely used in tropical countries, responsible for a considerable food export (Jennings and Li, 2017).

Although pesticides are mainly applied in the fields, they reach far more than the targeted crops through water and wind transport. Environmental conditions may influence pesticide drift during spraying, in which droplet size, velocity and rate of liquid/air in the spray nozzles play important roles (Al Heidary et al., 2014; Gil and Sinfort, 2005; Miranda-Fuentes et al., 2018). The microbial community quickly degrades GLY in soils until its total mineralisation (la Cecilia and Maggi, 2018). Glyphosate degradation may occur through the aminomethylphosphonic acid (CH_6NO_3P – AMPA) or sarcosine pathways (Aslam et al., 2023). In contrast to the metabolites resulting from the sarcosine pathway that are easily oxidised, AMPA is the primary metabolite of GLY and has a longer half-life in soils than the precursor molecule (Aslam et al., 2023). According to EFSA (2015), GLY has a low to very high persistence (DT_{50} = 2.8 – 500.3 days), and AMPA has a moderate to high persistence in lab studies (DT_{50} = 38.98 to 300.71 days) and high to very high persistence in field studies (DT_{50} = 288.4 to > 374.9 days) being reported in some studies with a DT_{50} up to 958 days (Primost et al., 2017). GLY and AMPA are practically non-volatile, and their mobility depends on their physicochemical properties such as octanol–water coefficient, electric charge and functional groups (Gimsing et al., 2007; Okada et al., 2016; Sheals et al., 2002); soil properties such as clay content, pH and cation exchange capacity (Aparicio et al., 2018; Erban et al., 2018; Gerónimo et al., 2018; Imfeld et al., 2013; Lupi et al., 2015; Soracco et al., 2018); microbial activity (Araújo et al., 2003; Giesy et al., 2000); or even wind and water erosion (Bento et al., 2019, 2017; Ernst et al., 2018; Mendez et al., 2017; Yang et al., 2015). According to Silva et al. (2018), in sites contaminated with low GLY or AMPA concentrations (<0.5 mg/kg soil), wind erosion can remove and transport up to 1.9 g soil/ha/year, while water erosion can remove and transport up to 9.7 g soil/ha/year. As for sites with higher concentrations (>0.5 mg/kg soil), wind erosion can remove and transport up to 3 g soil/ha/year, while water erosion can remove and transport up to 47.7 g soil/ha/year.

The high public interest and comments on the draft assessment of glyphosate by EFSA and ECHA (please see https://www.efsa.europa.eu/en/news/glyphosate-consultations-over-400-submissions-collected) confirmed this subject as a hot topic to the public that requests a high level of transparency on the evaluation of active substances. As a result of the request for additional information, EFSA is conducting a reassessment, which is expected to be completed by July 2023 (EFSA, 2023). As GLY and AMPA contamination can negatively impact ecosystems, biodiversity (Gill et al., 2018), and human health (van Bruggen et al., 2021, 2018), the present study aims to: i) review the concentrations of GLY and AMPA reported in soils at a global scale, with a specific focus on South America; ii) estimate the risk for key functional soil biota; and iii) the risk for human health.

2. Material and methods

2.1. Literature review

The dataset was extracted from the literature database Scopus (02/2023) published between 1970 and 2023 using keywords in English,

Table 1

Global pesticide use. Pesticide use worldwide summarised from FAOSTAT database of Food and Agriculture Organization of the United Nations (https://www.fao.org/faostat/en/#home – FAO, 2023). The colour scheme shows reductions in pesticide consumption in green, increases in red and similar consumption in white. The darkest the colour, the higher the decrease/increase magnitude.

Region	2000		2010		2020		Δ 2010–2020 (%)		Δ 2000–2020 (%)	
	Pesticide (Mt)	Herbicide (Mt)	Pesticide (Mt)	Herbicide (Mt)	Pesticide (Mt)	Herbicide (Mt)	Pesticide	Herbicide	Pesticide	Herbicide
Global Use	2.047	0.853	2.603	1.318	2.661	1.397	2%	6%	30%	64%
Africa	0.064	0.015	0.085	0.027	0.106	0.034	25%	27%	66%	128%
Asia	0.604	0.197	0.740	0.284	0.659	0.235	−11%	−17%	9%	19%
Central America	0.064	0.024	0.106	0.039	0.090	0.032	−15%	−17%	41%	35%
Europe	0.450	0.165	0.452	0.180	0.468	0.178	4%	−1%	4%	8%
North America	0.470	0.228	0.436	0.275	0.487	0.315	12%	15%	4%	38%
Oceania	0.038	0.026	0.049	0.032	0.070	0.047	45%	46%	86%	85%
South America:	0.350	0.195	0.728	0.477	0.770	0.551	6%	16%	120%	182%
– Argentina	0.084	0.070	0.236	0.217	0.241	0.230	2%	6%	187%	230%
– Bolivia	0.004	0.002	0.013	0.007	0.019	0.012	49%	71%	412%	474%
– Brazil	0.140	0.082	0.343	0.190	0.377	0.234	10%	24%	169%	186%
– Chile	0.014	0.003	0.010	0.002	0.010	0.002	0%	0%	−31%	−43%
– Colombia	0.076	0.027	0.049	0.018	0.037	0.029	−24%	66%	−52%	8%
– Equador	0.018	0.004	0.032	0.014	0.034	0.012	8%	−18%	90%	185%
– Guyana	0.000	0.000	0.000	0.000	0.000	0.000	117%	87%	58%	53%
– Paraguay	0.004	0.002	0.021	0.013	0.020	0.011	−4%	−15%	475%	368%
– Peru	0.002	0.000	0.006	0.002	0.011	0.004	71%	98%	371%	827%
– Suriname	0.000	0.000	0.000	0.000	0.000	0.000	−21%	109%	115%	139%
– Uruguay	0.004	0.002	0.015	0.012	0.016	0.014	10%	14%	351%	484%
– Venezuela	0.004	0.001	0.004	0.001	0.004	0.001	0%	0%	0%	0%

Portuguese and Spanish to find relevant records (e.g. "glyphosate", "glifosato", "aminomethylphosphonic acid "AMPA", "soil", "suelo", "solo"). The use of three languages is mainly related to many South American publications in Spanish and Portuguese. The dataset excluded the following document types: conference paper, letter, note, editorial and retracted (Fig. 1). After removing duplicates from the different search terms, the records were screened according to the title, abstract, highlights, and keywords to remove any that were not within the scope of this review. The remaining 188 manuscripts were then checked for five criteria. Criterion 1: records must include the measurement of GLY and/or AMPA in field samples. Criterion 2: records related to the study of GLY and/or AMPA degradation and other kinetic parameters that result from the direct contamination of field samples or studies that involve the direct contamination of soil in the field (e.g. micro- or mesocosms) are excluded. Criterion 3: records that include the study of GLY and/or AMPA concentrations from field samples that accurately describe how samples were stored and preserved prior to analysis. Criterion 4: records must detail that the last application of GLY has been made at least three months before the quantification. A final screening of records was performed (Criterion 5): records with improperly described methodology, written in another language (e.g. record has a double abstract in English and Chinese), among others, were excluded. No distinction was made between land type (e.g. agriculture and forest), soil management (conventional tillage and no-tillage), sample size or soil depth. Only the maximum concentrations were considered, except for Lupi et al. (2019) and Napoli et al. (2016), which did not provide the maximum value and the average value was used. After literature analysis, only 37 studies were considered. A complete list of used publications is included in the Mendeley Data Repository (https://doi.org/10.17632/jvd9rwd8r7.1), along with the sampling region and its reference.

2.2. Geocomputation

All data analyses were performed by statistical computing in R software version 4.1.2 (R Core Team – https://www.R-project.org/). The maximum concentrations of GLY and AMPA were plotted on static maps for spatial distribution visualisation using geocomputing scripts. The "readxl" package was used to read the data frame (Wickham and Bryan, 2019). The background polygons were obtained from "ggplot2" package (Wickham, 2016) and also was used to add a shapefile layer containing the referenced data points. Cartographic elements (north symbol) were defined using "ggspatial" package (Dunnington, 2021), and the coordinate reference system was based on the World Geodetic System of 1984 (WGS84) using "sf" package (Pebesma, 2018).

2.3. Ecological risk assessment (ERA)

GLY and AMPA were incorporated into the Ecological Risk Assessment (ERA) based on the ratio between exposure and hazard factors. The ERA is based on the equations previously described by ECETOC (2004), Peake et al. (2016), and Pérez et al. (2021), and derived as follows (Eq. (1)):

$$RQ = \frac{MEC}{PNEC} \tag{1}$$

where *RQ* is the risk quotient (toxicity unit), *MEC* is the measured environmental concentration (mg/kg soil), and *PNEC* represents the predicated no-effect environmental concentration (mg/kg soil). The *PNEC* was derived as follows (Eq. (2):

$$PNEC = \frac{LC_{50}}{AF} \tag{2}$$

where LC_{50} represents the concentration that will kill 50% of the sample population (mg/kg soil). The LC_{50} values for earthworms (*Aporrectodea caliginosa*, *Eisenia andrei*, and *Eisenia fetida*), and springtail (*Folsomia*

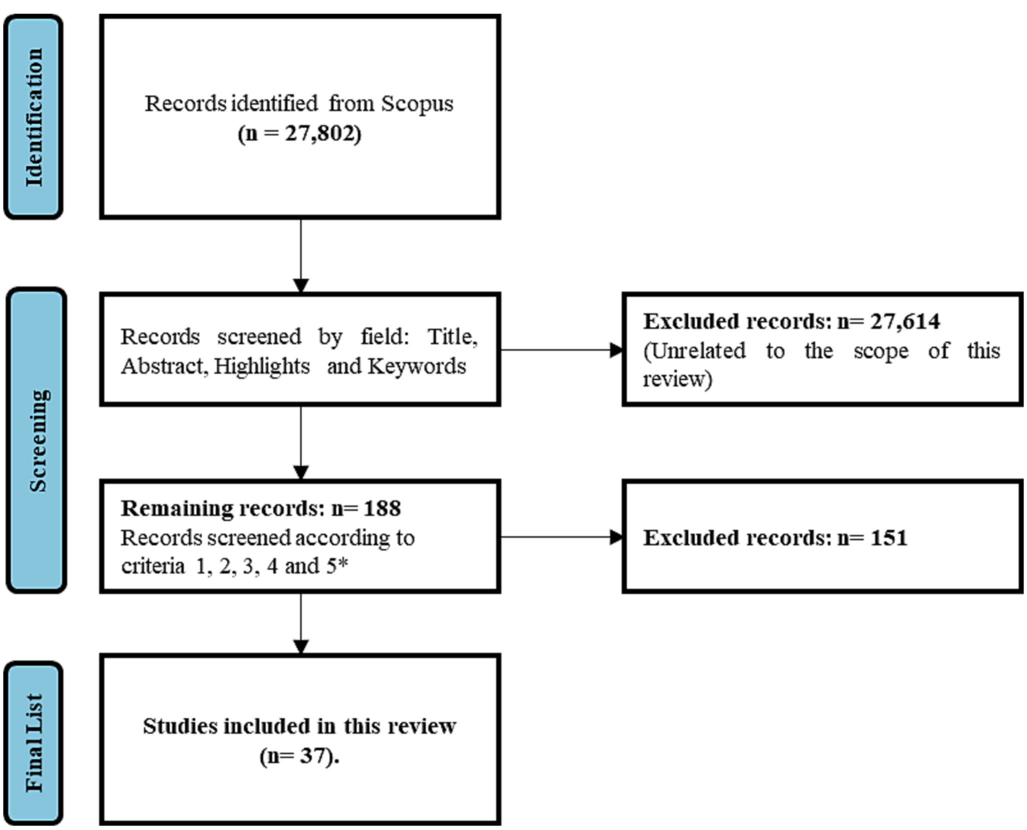


Fig. 1. Diagram of the methodology used to include/exclude records to be used in this review. * Criterion 1: records must include the measurement of GLY and/or AMPA in field samples. Criterion 2: records related to the study of GLY and/or AMPA degradation and other kinetic parameters that result from the direct contamination of field samples or studies that involve the direct contamination of soil in the field (e.g. micro- or mesocosms) are excluded. Criterion 3: records that include the study of GLY and/or AMPA concentrations from field samples that accurately describe how samples were stored and preserved prior to analysis. Criterion 4: records must detail that the last application of GLY has been made at least three months prior to the quantification. Criterion 5: records with improperly described methodology, written in another language (e.g. record has a double abstract in English and Chinese), among others, were excluded.

candida) were obtained from European Food Safety and Authority peer reviews (EFSA, 2015) and ECOTOX Knowledgebase (EPA, 2021). Since no toxicity data was available for AMPA, the LC_{50} values of the parental compound (GLY) were multiplied by a value of 20x based on the AMPA/GLY ratio for LC_{50} values of aquatic organisms described on the EFSA and ECOTOX databases. Although these calculations present some uncertainty, as the toxicity of AMPA may not be 20x higher than the toxicity of GLY, similar conservative criteria were used in previous studies (Pérez et al., 2021; Vašíčková et al., 2019). Additionally, this conservative criterion would represent a worst-case scenario. The ecotoxicological endpoints dataset is presented in Table S1 (Supplementary Data - SD). The *AF* represents an assessment factor ($AF = 50$, without a unit of measure) and was applied to reduce uncertainties about the accuracy, model errors, lack of toxicity data, and inherent variability between laboratory exposure and field conditions (Vašíčková et al., 2019). The RQ approach was classified as low ecological risk (RQ < 0.1), moderate ecological risk (0.1 < RQ < 1.0), and high ecological risk (RQ > 1.0), according to Pérez et al. (2021).

2.4. Human risk assessment (HRA)

The Human Risk Assessment (HRA) was developed according to the Exposure Factors Handbook (EPA, 2011) and Generic Exposure Routes Assumptions document (Michigan, 2014). The HRA was estimated from generic characteristics of adults. Table S2 (SD) presents details of the human lifestyle and physiological and behavioural parameters. A probabilistic risk model was used to estimate carcinogenic incidence based on the average daily dose by *i* exposure routes [ADD_i – mg/(kg/day)], being the soil ingestion (ADD_{soil}), food ingestion (ADD_{food}), dermal contact (ADD_{derm}), and dust inhalation (ADD_{inh}) expressed as follows (Eqs. (3) to (6):

$$ADD_{soil} = \frac{C_{soil} \times IR_{soil} \times ED \times EF \times CF}{BW \times AT} \tag{3}$$

$$ADD_{food} = \frac{C_{soil} \times BCF \times IR_{veget} \times ED \times EF}{BW \times AT} \tag{4}$$

$$ADD_{derm} = \frac{C_{soil} \times SA \times DAF \times ABS \times ED \times EF \times CF}{BW \times AT} \tag{5}$$

$$ADD_{inh} = \frac{C_{soil} \times IR_{air} \times ED \times EF}{PEF \times BW \times AT} \tag{6}$$

where C_{soil} is the concentration of the pollutant on soil (mg/kg soil); IR_{soil} represents the daily soil ingest rate (mg/day); *ED* is the exposure duration (year); *EF* is the exposure frequency (day/year); *CF* is the conversion factor (kg/mg); *BW* is the body weight of the individual adult (kg); *AT* is the average lifespan of an adult (day); *BCF* is the bioconcentration factor (without a unit of measure); IR_{veget} is the daily vegetable ingestion rate (kg/day); *SA* is the surface area of the skin that contact with the soil (cm^2/day); *DAF* is the soil dermal adhesion factor (mg/cm^2); *ABS* is the dermal absorption factor (without a unit of measure); IR_{air} is the air inhalation rate (m^3/day); and *PEF* is the particle emission factor (m^3/kg).

A carcinogenic slope factor (*SF*) was added to the equations to define the upper confidence limit on the increased risk from *i* exposure route. The incremental lifetime carcinogenic risk (*ILCR*) was calculated using the estimated ADD_i multiplied by the SF_i as follows (Eq. (7):

$$ILCR = \sum (ADD_i \times SF_i) \tag{7}$$

where SF_i includes oral slope factor [SF_o, – mg/(kg/day)], dermal slope factor of dermal contact [SF_{abs}, – mg/(kg/day)], and the inhalation unit risk (*IUR* – mg/m^3). Details of the parameters used to estimate the risk to human health are available in Table S2 (SD). The $ILCR < 1 \times 10^{-6}$ considers the risk to the human population acceptable, while $ILCR > 1 \times 10^{-4}$ indicates the high probability of risk over a lifetime. The non-cancer risk (hazard index - *HI*) was estimated using equation (8) (Eq. (8):

$$HI = \sum \frac{ADD_i}{SF_i} \tag{8}$$

where *RfD* [RfD_i – mg/(kg/day)] is defined as the daily maximum permissible values for the GLY of the two (*i*) exposure pathways, including reference dose for ingestion (RfD_o), and dermal contact ($RfD_{abs} = RfD_o \times ABS_{GI}$). ABS_{GI} is the fraction of GLY absorbed in the gastrointestinal tract. The daily maximum reference dose for the inhalation exposure pathway could not be calculated due to the properties of the GLY (e.g. polarity and octanol–water partition coefficient), resulting in very low vapour pressure and low volatility. An *HI* lower than unity ($HI < 1$) reflects a no non-cancer risk.

3. Results and discussion

3.1. Soil pollution

GLY and AMPA residues found in different soil matrixes (e.g. urban, agricultural and secondary forests) were plotted on a global map (Fig. 2). GLY was detected at levels ranging from 2×10^{-4} to 66.38 mg/kg soil, while AMPA was detected at 2.5×10^{-3} to 38.94 mg/kg soil. Overall, three-quarters (73%) of GLY and more than two-thirds (68%) of AMPA concentrations found in soil were quantified up to 1 mg/kg soil, with four studies for GLY (Mercado and Mactal, 2021; Newton et al., 1994, 1984; Samson-Brais et al., 2022), and four studies for AMPA (Ernst et al., 2018; Mercado and Mactal, 2021; Napoli et al., 2016; Newton et al., 1994) showing values < LOD or < LOQ. No data was available for the African and the Oceanian continent. Four studies were found for North America (Newton et al., 1994, 1984; Samson-Brais et al., 2022; Tush et al., 2018), and only three for Asia (Gunarathna et al., 2018; Jing et al., 2021; Mercado and Mactal, 2021). A possible explanation for the lack of/low data for the African and Asian continents may be related, to a certain extent, to the lack of financial and infrastructural resources in the research institutions of these continents, as the majority are included in the list of low and middle-income countries (LMICs – https://data.worldbank.org/country/XN). Nonetheless, these regions also include many countries with higher resources to monitor soil pesticide concentrations. Collecting this type of data is extremely important, especially when looking at the increase of 25% in the use of pesticides in Africa in the past ten years (Table 1). The lack of data in Africa may be mainly related to outdated information, the lack of registration of pesticide use (e.g. Nigeria does not have any data available) or even the inconsistent registration of data bringing a lot of uncertainty to the available data (e.g. South Africa - Fuhrimann et al., 2022). Other factors that may explain the lack of data may be related to challenges with chemical analyses in local laboratories (e.g., lack of specialised equipment) or lack of knowledge that concerns pesticide analysis techniques. Finally, there is also a critical gap in recording herbicide levels in soil compared with extensive reports of other types of pesticides (Paré et al., 2014; Ssebugere et al., 2010). Most African governments have encouraged pesticide use in the last decades, and these changes resulted in even less government monitoring. Therefore, inadequate regulatory policies result in the importation of banned pesticides (Sharma et al., 2019). The African market is unregulated, so the registered compounds account only for 8% of the total. Additionally, 38% of pesticides have incomplete labels, and 6% are unlabelled (van der Valk, 2003).

The intensive use of pesticides in agriculture is increasing rapidly in developing countries. Its use for controlling and eradicating malaria is common in several countries, including Asia (Kunstadter, 2007; Schreinemachers and Tipraqsa, 2012). Asia is the second-highest region that applies all commercialised pesticides worldwide (Table 1). Here, China and Japan represent the largest producers and consumers of pesticides globally (Zhang, 2018; Zhang et al., 2011).

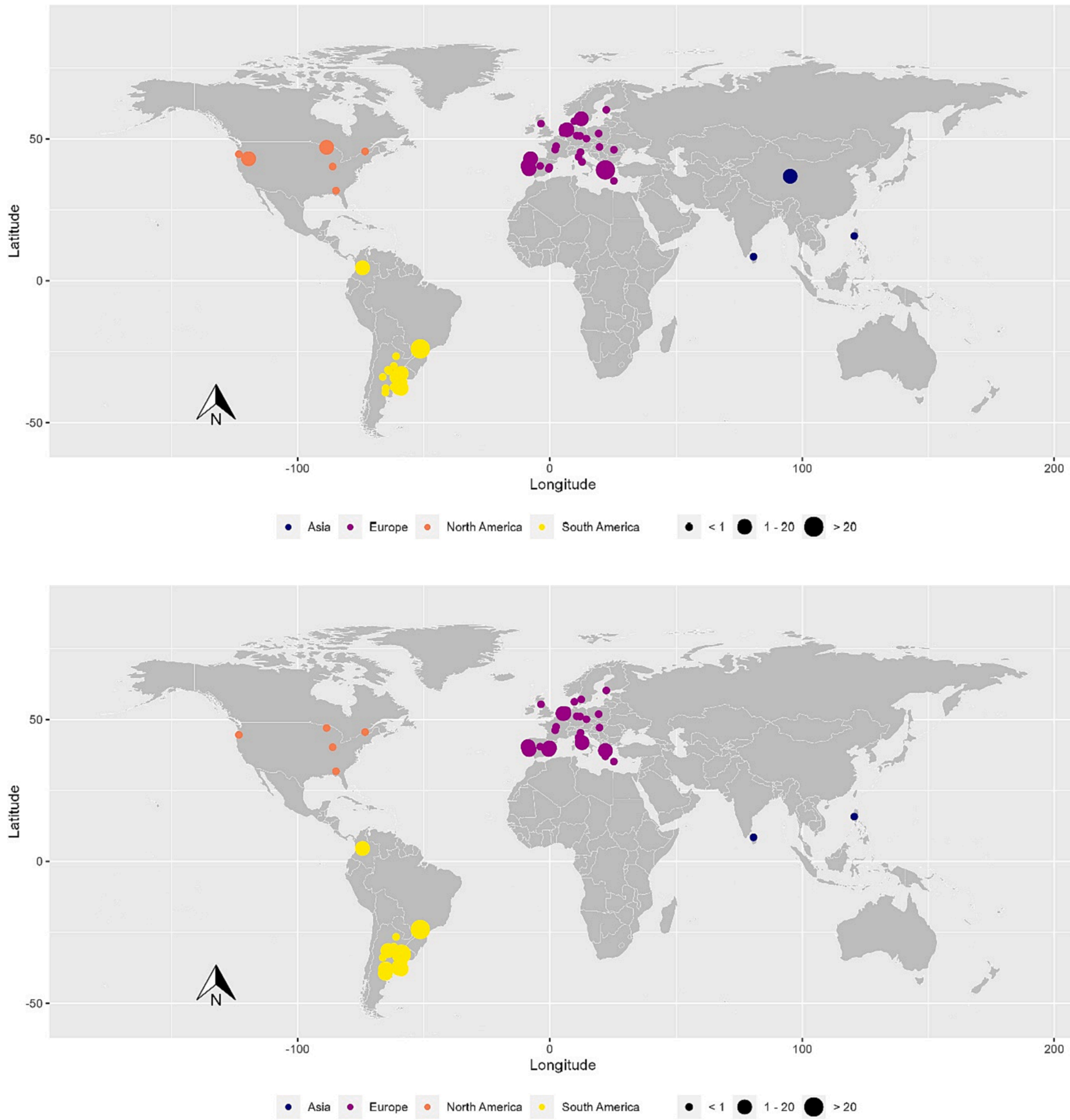


Fig. 2. Global map of GLY and AMPA in soils. Spatial distribution of glyphosate (GLY) and aminomethylphosphonic acid (AMPA).

Although the Australian region has the resources to characterise pesticides in soil, no data could be found. This may be related to the fact that most scientific resources focus on quantifying pesticides in food and water (e.g., Okada et al., 2020). Furthermore, other factors can result in low GLY concentrations, such as poor soils with high ultraviolet radiation (Papagiannaki et al., 2020). The Australian Academy of Science and Technological Engineering conducted the last major pesticide review in Australia in 2002 (Radcliffe, 2002), approximately 20 years ago. Although the document can be seen as a pervasive collection of data with great value, it can now be considered outdated. According to the models presented by Tang et al. (2021), the Australian region's pesticide levels are low compared to other high-income countries. Nonetheless, Wightwick and Allinson (2007) highlight that this region's pesticide risk to humans and the environment has received relatively little attention.

In European soils, 14 studies showed GLY and AMPA in soils (Börjesson and Torstensson, 2000; Carretta et al., 2021; Erban et al., 2018; Geissen et al., 2021; Ibáñez et al., 2005; Karanasios et al., 2018; Karasali et al., 2019; Laitinen et al., 2006; Napoli et al., 2016; Pelosi et al., 2022; Silva et al., 2018; Tauchnitz et al., 2020; Veiga et al., 2001; Vlassa et al., 2022). European countries show many different political interests with a broad public discussion on the use of pesticides, regulation and data availability. It was reported that the United Kingdom, Denmark, the Netherlands, Germany and Sweden had a very strong political and public interest in lowering the application of pesticides. On the other hand, Finland and Latvia had no specific regulations for lowering pesticide usage (Kristoffersen et al., 2008), until recently (Finland: 2018 - Reg. No. 3406/00.03.02/2018; Latvia: 2020 - Cabinet Order No 27 of 22 January 2020) when they approved new herbicides reduction strategies enforced by the EU. Despite the increase in total pesticide use in the past ten years, the pesticide application rate in

Finland and Latvia is low compared to other European countries, which may be related to the region's climate (FAO, 2023). A few European countries, including Denmark, France, Austria and the Netherlands, reduced pesticide usage, while others like Germany, Greece, Ireland, Czech Republic, Spain and Portugal increased it. In general, the local government authorities are making political efforts to improve the efficiency of pesticide regulation and environmental protection. Nonetheless, EU countries have approved the implementation of legislation on pesticide use, such as Directive No. 2009/128/EC, Regulation (EC) No. 1107/2009, and Regulation (EC) No. 396/2005. A review of the presence of pesticides in European soils has been previously performed by Silva et al. (2018), and more information can be found on it. Nonetheless, it is important to emphasise that GLY and AMPA were found in 15 European countries, mainly with levels < 1 mg/kg soil. Higher levels of GLY were mainly found in the Netherlands and Mediterranean countries (Greece, Portugal and Spain). Identically, these countries have also identified higher levels of AMPA along with Italy.

In North America, herbicides are the most prominent and widely used chemical pesticides (Canada, 2019). Herbicide usage has increased in croplands and the wildlands (EPA, 2008). GLY is the most active ingredient that harms the herbs and grasses in croplands and poses a potential threat to the surrounding native vegetation (Wagner et al., 2017). Therefore, future studies on identifying soil pollution are also needed in the region. GLY is widely sprayed in the US, the leading global GMO producer (>71.5 million hectares – ISAAA, 2018). Thus, GLY-tolerant GMOs account for about 56% of global GLY use (Benbrook, 2016). Furthermore, GLY is widely applied as a desiccant in no-tillage systems in the region, representing more than 40 million hectares in the US. Agricultural use of GLY in the US increased after the GMO adoption in 1996, which increased more than ten times between 1996 and 2017 and has steadily grown to the present day (Rifet et al., 2018). The US has a robust political framework for the use, sales, application and regulation of GLY, with data available in databases from agencies such as the Department of Agriculture (USDA – https://www.usda.gov/), Environmental Protection Agency (EPA – https://cfpub.epa.gov/ecotox/), National Agricultural Statistics Service (NASS – https://www.nass.usda.gov/), Natural Resources Conservation Service and Department of Agriculture and Geological Survey (NRCS and USRG – https://www.nrcs.usda.gov/). However, despite the study conducted by Battaglin et al. (2014) in various regions in the US, there are still not enough studies that seek to monitor herbicide residues in the soil (Benbrook, 2016).

The present review identified GLY and AMPA in 16 studies for South American soils (Alonso et al., 2018; Aparicio et al., 2018, 2013; Bento et al., 2019; Bernasconi et al., 2021; Botero-Coy et al., 2013; da Silva et al., 2021; Lupi et al., 2019; Okada et al., 2018, 2016; Pérez et al., 2021; Peruzzo et al., 2008; Primost et al., 2017; Ramirez Haberkon et al., 2021a, 2021b; Soracco et al., 2018). The high-level contamination identified in the present review in the region is in accordance with the models proposed in a previous study (Maggi et al., 2020). In their study, although the range of concentrations falls within 0.1 – 10 mg/kg of soil, the area is considered a hotspot compared with its vicinity. The South American continent is a vast area of food production that, in its majority, is exported to wealthier countries. The extensive agricultural practices for food production in those countries are associated with a great demand for pesticides. Their use results in high soil contamination rates that can be worsened with the expansion of GMO fields and no-tillage management (CETESB, 2018; ISAAA, 2018). Within South American countries, Argentina has a significant amount of information regarding pesticide levels. Still, such data is scarce for other countries. The lack of such information is of great concern for many different reasons. For example, only one study related to GLY or AMPA occurrence in soils was found for Colombia (Botero-Coy et al., 2013) despite the high agricultural practices related to a high increase in the cultivated area and the 66% increase in the consumption of herbicides in the past ten years. Similarly, Equator and Peru, in the past twenty years, have increased the consumption of herbicides by 98% and 827%, respectively, with no studies reporting GLY and AMPA levels (FAO, 2023). In addition to all the previously mentioned constraints, South American countries have an ongoing increase in the number of pesticides approved for agriculture and public health welfare through the control of disease transmission vectors (MAPA, 2022), as recently occurred in Brazil. In this country, the average number of new approvals between 2000 and 2015 was approximately 122/year, whereas, in 2022, the number reached 562/year (MAPA, 2022). Although Brazilian legislation requires a periodic review of the pesticides registered and approved for use (Brazilian Law No. 7802/1989 and Decree No. 4074/2002), these laws are not enforced. Furthermore, Brazil is also preparing other legislation less restrictive on this issue (e.g. PL 1459/2022). In addition to the non-enforcement of laws, Brazil has regulatory values for pesticide levels in food and water much higher than other regions (Bombardi, 2017). For example, GLY levels in soybean can be 200x higher than those accepted by EU legislation (10 mg/kg BR vs 0.05 mg/kg EU). Similarly, GLY levels in Brazilian waters can be 5000x higher than in EU waters (0.5 mg/L BR vs 0.0001 mg/L EU – Bombardi, 2017). Again, the urgency for the development of new environmental policies is highlighted by the lack of limits for pesticide levels in soils, and the existence of reference levels for pesticides that are now fully banned (e.g., DDT – Regulation No. 420/2009 of the Brazilian National Council for the Environment). These regulations flow towards the opposite direction of the guidelines established by the United Nations Sustainable Development Goals (SDG): food security and sustainable agriculture (SDG 2); sustainable consumption and products patterns (SDG 12); and protect, restore and promote sustainable use of soil ecosystem, reduce land degradation and biodiversity loss (SDG 15). In the EU, high efforts are being made in the European Farm to fork strategy on the sustainable use of pesticides (Directive 2009/128/EC). The strategy that claims to be at the heart of the European Green Deal aims a transition to sustainable food systems. This strategy aims, among other points (e.g. have a neutral/positive environmental impact or the mitigation of climate changes), a transition to organic farming and a reduction in pesticide use. In this sense similar stratergies should be applied worldwide.

3.2. Risk to terrestrial ecosystems

The RQ for soil was calculated for two groups of species: springtails (Collembola) and earthworms (Annelida). The RQ for GLY and AMPA are presented in Fig. 3. GLY's RQ could not be calculated for Africa and Oceania due to the nonexistence of records. Similarly, due to the low or nonexistent number of records, AMPA's RQ values for Asia, Africa and Oceania were not calculated. When comparing both species as expected, springtails show high percentages of moderate and high ecological risk values in most continents. For the springtails and specifically for GLY, the percentage of high ecological risk is between 67% and 93%, with no cases of low ecological risk scenarios. As for the AMPA RQ, the scenario is better. Still, the percentage of low ecological risk exists only for Europe and South America, with 4% and 14%, respectively. As for earthworms, the risk scenarios are much better, with GLY showing moderate and high ecological risk scenarios between 43% and 67%. These RQ scores contradicts the study of Maggi et al. (2020) which proposes a model that estimates glyphosate residue in soils and suggests no acute impact on earthworms. The RQ for AMPA shows low ecological risk scenarios of at least 86%. When looking into both groups' RQ results, as expected, earthworms show better-surviving scenarios than springtails. Although the nature of the results is not new, it renews the discussion regarding the role of the most sensitive species in ecosystems and the impact of their possible absence. Despite these alarming results (at least for springtails), it is essential to highlight that RQ values are calculated based on the highest amount of GLY and AMPA concentrations found in soils, not their bioavailable concentrations. As so, ecotoxicological tests are crucial to gather information regarding the bioavailability of these chemicals and thus to achieve a more

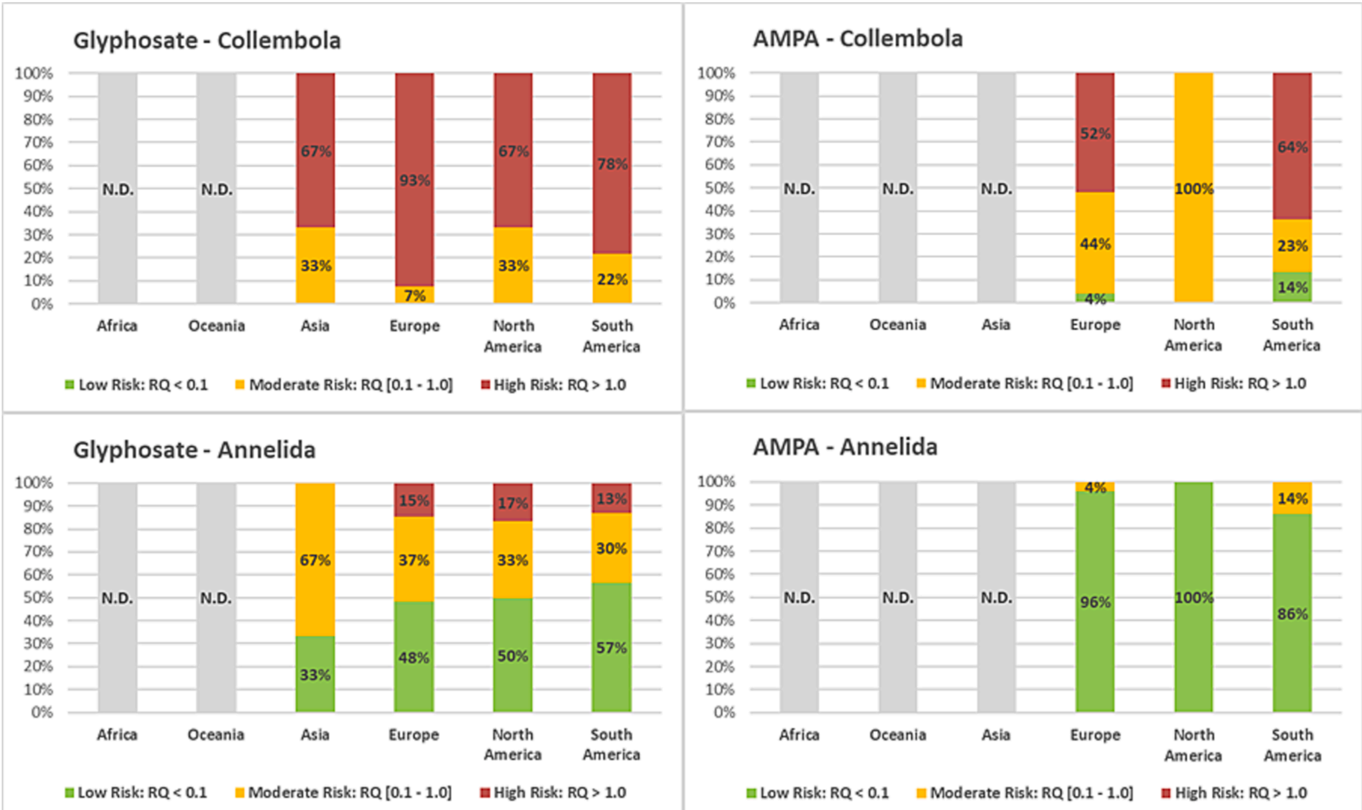


Fig. 3. Ecological risk assessment (ERA) calculated in the present study. ERA is expressed by the risk quotient (RQ) approach for soil species (Annelida, Collembola) exposure to glyphosate (GLY) and aminomethylphosphonic acid (AMPA). Figures present the percentage of soils that fall into each category (low ecological risk – RQ $<$ 0.1; moderate ecological risk – RQ [0.1 – 1.0]; high ecological risk – RQ $>$ 1.0) and its corresponding percentage.

comprehensive assessment of these soils (Niemeyer et al., 2017; Niva et al., 2016). It is also important to stress the need to improve ecotoxicological tests. The use of standardised soil with characteristics common to European and North American soils; the use of species that do not occur worldwide (or at least are uncommon); and the use of controlled conditions that do not reflect most of the areas of the globe, has been highlighted as a problem that needs to be addressed (Daam et al., 2019; Niva et al., 2016). Another important highlight is that the RQ has been based only on mortality values. However, the adverse effects can be observed at lower doses and different levels (e.g., Buch et al., 2013; Domínguez et al., 2016; Ferreira et al., 2015). These effects can then compromise the population's dynamics, decreasing reproduction and increasing mortality (O`Brien, 2017; Zhou et al., 2018). However, there are some inconsistencies in the literature about the toxicity of GLY. Some studies reported that the negative effects observed in the commercial formulation are contrary to the active ingredients usually used in ecotoxicological tests (Meftaul et al., 2020). In this sense, further studies are needed to address the chronic effects of the commercial formulation versus the active ingredient by itself.

Our results suggest a potentially harmful effect on ecosystems and their organisms in most of the revised literature for GLY and AMPA. Still, literature regarding the impact of GLY and AMPA on terrestrial organisms is scarce, as seen in Table 2. Overall, the RQ approach calculated in the present study showed the highest deleterious effect on non-target soil biota, thus highlighting the need for further environmental risk assessment and the use of this data for new policies. The RQ high values obtained for key functional organisms in this study may be a warning toward potential adverse effects on ecosystems. Nonetheless, the effects of GLY and AMPA on soil microbes (e.g. microbial activity, biomass, structure, diversity) have been reported in the literature, and their impact may be positive or negative (e.g. Bruggen et al., 2021).

3.3. *Risk to human health*

A summary of the Human Risk Assessment is shown in Fig. 4. For GLY, the ILCR index was not calculated for Africa and Oceania due to the nonexistence of records. Also, due to the low or nonexistent number of/low records, AMPA's ILCR index for Asia, Africa and Oceania were not calculated. In this study, the ILCR calculated for human health from ADD*i* exposure routes indicated an insignificant risk ($<1 \times 10^{-06}$) between 33.3% and 65.2% for GLY, and for AMPA $\sim$ 35% for Europe and South America, with North America showing an insignificant risk for all revised soils. Nonetheless, in the case of GLY, Europe and South America showed an already concerning high probability of risk over a lifetime of 3.7% and 4.3%, respectively. According to ILCR, the ADD_{food} represents the main pathway of exposure.

As for the non-cancer risk hazard index (HI), due to the physical–chemical properties of GLY (please see section 2.4), the inhalation unit risk (IUR) and the inhalation reference concentration (RfC) were considered zero. As a result, the risk from inhalation (ADD_{inh}) was also considered null. The high GLY and AMPA levels that put South America as a hotspot for soil contamination are of great concern. This region already shows a high HI ($>$1) for GLY and AMPA of 4.3% and 9.1% of the revised soils, respectively. In the same way for GLY, Europe already shows a high HI for 3.7% of the revised soils. It is also noteworthy to stress that the number of studies found for North America and Asia is low. As a result, one should look with some caution into their ILCR and HI results. Nonetheless, more restrictive legislation, similar to existing legislation, for example, in many North American or European countries, is crucial at least to maintain or decrease the current levels, as they may harm the environment and human populations. At the moment, the possible toxic effects of GLY on mammals are still under debate and different legislations, from partial ban application on field crops (e.g.,

Table 2
Adverse responses of soil pollution in key functional species. GLY and/or AMPA effects or responses from ecotoxicological studies in key soil species at different levels of biological organisation. [a] (Kronberg et al., 2018); [b] (Simões et al., 2018); [c] (Niemeyer et al., 2018); [d] (Santos et al., 2010); [e] (Hackenberger et al., 2018); [f] (Buch et al., 2013); [g] (Niemeyer et al., 2018); [h] (Domínguez et al., 2016); [i] (Santadino et al., 2014); [j] (Gaupp-Berghausen et al., 2015); [k] (Niemeyer et al., 2018); [l] (Niemeyer et al., 2006); [m] (Santos et al., 2010).

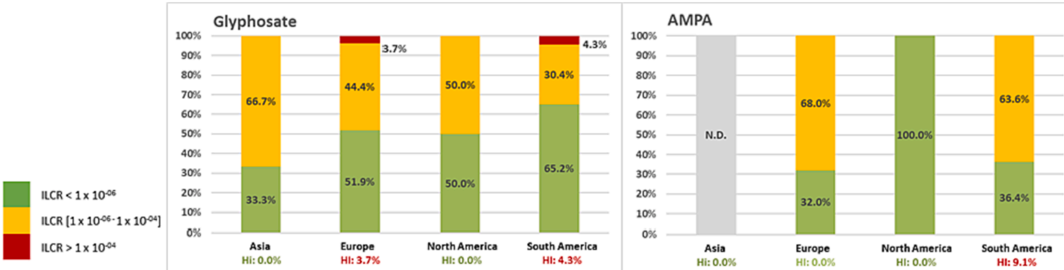
Taxon Group	Level of Biological Organization	Model Organism	Endpoints (effect / response)
Nematode	below-cell	*Caenorhabditis elegans*	**GLY formulation:** oxidative stress (increase ROS production), antioxidant defence (catalase activation), genotoxicity (increased CAT gene expression) [a]
Collembola	below-cell	*Folsomia candida*	**GLY formulation:** genotoxicity (decreased CAT gene expression, increased SOD and Cyt C gene expression, inhibiting the expression of transcripts involved in fatty acid metabolism) [b]
	organism		**GLY formulation:** behaviour (avoidance) [c]
	organism and population		**GLY formulation:** behaviour (avoidance) and reproduction (decreased number of juveniles) [d]
Annelida	below-cell	*Dendrobaena venata*	**GLY formulation:** neurotoxicity (AChE induction)oxidative stress (lipid peroxidation) [e]
	organism	*Eisenia andrei* and *Pontoscolex corethrurus*	**GLY formulation:** behaviour (avoidance) [f]
		Eisenia andrei	**GLY formulation:** behaviour (avoidance) [g]
	organism and population	*Eisenia fetida*	**AMPA pure**: body weight (biomass loss) and reproduction (increased number of cocoons and juveniles with weights decreased) [h]
	population	*Eisenia fetida*	**GLY formulation:** reproduction (increased number of cocoons with lower fertility), population dynamic (negative growth rate), development (lower survival of juveniles) [i]
		Lumbricus terrestris and *Aporrectodea caliginosa*	**GLY formulation:** reproduction (reducing percentage of cocoons) [j]
Isopoda	organism and population	*Porcellio dillatatus*	**GLY formulation:** behaviour (avoidance) [k]
		Cubaris marina	**GLY formulation:** body weight (biomass loss) [l]
	organism	*Porcellionides pruinosus*	**GLY formulation:** behaviour (avoidance) [m]

France and Germany) to high permissive levels (e.g. Brazil). Nonetheless, based on the increased incidence of liver and kidney tumours, positive correlation with non-Hodgkin lymphoma, and evidence of genotoxicity, the International Agency for Research on Cancer has classified GLY as potentially carcinogenic (Bus, 2017; van Bruggen et al., 2018). Moreover, there are positive correlations between GLY use and an increased attention deficit hyperactivity disorder, abortions, Alzheimer's and Parkinson's diseases (van Bruggen et al., 2018). This classification is based, for example, on epidemiological in vitro studies (Agostini et al., 2019). For mammals (e.g., rats and rabbits), exposure to GLY has shown an increase in malignant lymphomas, skeletal and cardiac malformations, and delayed ossification (EFSA, 2015). It is also important to highlight that the toxicity of the active ingredient by itself may be lower than the toxicity of commercial formulations that contain adjuvants such as the polyethoxylated tallow amine (POEA) that are incorporated to potentialise its effect (Meftaul et al., 2020; Tóth et al., 2020). Mesnage et al. (2015) reported more intensive toxic effects from GLY with adjuvants than isolated GLY. Despite these differences, many regulatory agencies have established toxicological parameters for human exposure based on the effects of GLY by itself (e.g. EFSA and EPA).

For what reports AMPA's toxicity, the available literature is still scarce. There is almost no evaluation within the different levels of the biological organisation except for mortality and reproduction of key species such as earthworms with a classification of low risk (EFSA, 2015). GLY and AMPA share a similar profile, and both compounds may be classified as no potential human carcinogens (Buekers et al., 2022; EFSA, 2015). Nonetheless, much data still needs to be gathered regarding the toxicological profile of AMPA which is currently critically required (Connolly et al., 2022). AMPA residues in soil, plants, or food can seep into streams and expose the general public. AMPA may cause oxidative stress (Benbrook, 2016), according to evidence from *in vitro* and animal studies mentioned by the International Agency for Research on Cancer (2014), and may be linked to breast cancer rates (Franke et al., 2021). Given that AMPA has already been found in the urine of persons who have not been exposed to it at work, frequently at a higher percentage than GLY, raising several other questions (Conrad et al., 2017; Mcguire et al., 2016; Mills et al., 2017). However, similar to the parent molecule, there are questions and ambiguities regarding the true carcinogenic risk of AMPA. It is also important to stress that AMPA can also result from the breakdown of phosphonate detergents, thus occurring at even higher levels than expected (Battaglin et al., 2014; Jaworska et al., 2002). Finally, because both substances co-exist in the soil (Buekers et al., 2022, 2021), the combined GLY + AMPA levels can increase significantly the toxicological effects and even lead to worse risk case scenarios (FAO, 2011).

4. Conclusion

Overall, the present study reviews the GLY and AMPA levels in soils across the globe, highlighting the lack of studies in different regions that may result from the lack of proper control or budget to perform such analyses (e.g. South Africa), or when the budget is not a constrain, by priorities (e.g., Australia). On the other side, even in cases where few studies are available, the levels of these two chemicals may pose serious concerns, with the worst contamination and highest risk occurring in tropical regions. South America has been defined as a hotspot of soil contamination due to pesticide spraying contents and biodiversity levels. Here is highlighted the need to improve the directive regulations, to collect more ecotoxicological data using model local soils, species and controlled parameters. Further efforts are needed to prioritise actions focused on environmental legislation and policymaking in the region. Along with these needs, it is also necessary to develop sustainable agricultural practices that reduce pesticide use and misuse. This can be achieved with, for example, initiatives such as the Farm to Fork. The overall interaction between new policies, higher interaction with the general public to disseminate more information on pesticides, and more toxicity and soil levels of GLY and AMPA around the globe is crucial to maintain and to lower the risk to human health and ecosystems. There is still a conflict of opinions about the toxicity of GLY and AMPA, and it is not up to the authors of this paper to take a side, leaving the strong conclusions up to the reader. The need to continue to monitor and increase the number of studies regarding the occurrence of GLY and AMPA around the globe is critical. Further studies such as this review are



Region	Glyphosate		AMPA	
	ILCR Average [minimum – maximum]	HI Average [minimum – maximum]	ILCR Average [minimum – maximum]	HI Average [minimum – maximum]
Asia	**4.27×10^{-06}** [6.50×10^{-08} – 8.26×10^{-06}]	**1.39×10^{-01}** [2.11×10^{-03} – 2.68×10^{-01}]	-	-
Europe	**8.63×10^{-06}** [3.44×10^{-08} -1.40×10^{-04}]	**2.70×10^{-01}** [1.08×10^{-03} – $4.37 \times 10^{+00}$]	**3.93×10^{-06}** [1.08×10^{-07} – 2.25×10^{-05}]	**1.23×10^{-01}** [3.39×10^{-03} – 7.05×10^{-01}]
North America	**3.18×10^{-06}** [3.51×10^{-08} – 1.07×10^{-05}]	**9.78×10^{-02}** [1.08×10^{-03} – 3.28×10^{-01}]	**3.85×10^{-07}** [2.05×10^{-07} – 8.18×10^{-07}]	**1.18×10^{-02}** [6.28×10^{-03} – 2.51×10^{-02}]
South America	**6.65×10^{-06}** [4.20×10^{-09} – 1.16×10^{-04}]	**2.10×10^{-6}** [1.33×10^{-04} – $3.67 \times 10^{+00}$]	**8.35×10^{-06}** [4.08×10^{-09} – 7.95×10^{-05}]	**2.64×10^{-01}** [1.29×10^{-04} – $2.51 \times 10^{+00}$]

Fig. 4. Human risk assessment (HRA). Incremental lifetime carcinogenic risk (ILCR) and hazard index (HI) calculated for GLY and AMPA from different world regions (Asia, Europe, North and South America) for human health from the average daily dose by four exposure routes: soil intake, food intake, dermal contact, and inhalation. Figures present the percentage of soils that fall into each category and the corresponding percentage of soils with a high HI (>1). Table presents the average [minimum – maximum] for each index. AMPA's ILCR and HI were not determined due to the low number of studies in the region (n = 2).

required to help policymakers and Assessment Groups on the protection of ecosystems and human health.

Data Repository

The metadata file containing the selected publications; GLY and AMPA concentrations used in the study; LC_{50} and PNEC values for soil species; RQ for GLY and AMPA; and ILCR for GLY and AMPA are available in Mendeley Data Repository (https://data.mendeley.com/datasets/jvd9rwd8r7/1).

CRediT authorship contribution statement

Nuno G.C. Ferreira: Investigation, Conceptualization, Writing – review & editing, Validation, Resources, Supervision, Formal analysis, Funding acquisition, Project administration. **Karlo Alves da Silva:** Investigation, Writing – review & editing, Formal analysis, Data curation. **Ana Tereza Bittencourt Guimarães:** Conceptualization, Validation, Data curation. **Cíntia Mara Ribas de Oliveira:** Conceptualization, Writing – review & editing, Validation, Resources, Supervision, Funding acquisition, Project administration.

Declaration of Competing Interest

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

Data availability

Data provided in Mendeley Data Repository (https://data.mendeley.com/datasets/jvd9rwd8r7/1)

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2023.108135.

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