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Use of the glyphosate herbicide: an integrated review

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ABSTRACT. the herbicide glyphosate has a non-selective active ingredient for the functional groups carboxyl, amino, and phosphonate. Its use was accentuated with the opening of agricultural frontiers and the use of genetically modified organisms, due to the great demand for food in the world. Thus, the objective of this study was to analyze what scientific productions from 2010 to 2020 bring about the glyphosate herbicide theme and its impacts on the environment. the study comprised the search for papers in the Capes - Web of Science and Scopus Journals, covering 62 scientific studies. Europe and America stood out in scientific productions, with 38.7 and 37.1% of published works, respectively. the countries that stood out were Argentina, Brazil, United States, and France, both with 7 productions each. Regarding the researched topics, 'Biological test in a contaminated environment' stands out, with 9 publications, followed by 'Adsorption' with 8 publications. However, there is still a demand for studies, for the development of methods for determining the herbicide glyphosate and its metabolite Ampa, mainly in soil and water, as it is a potential contaminant, to guide decisions about it and propose guidelines that support the technique and scientifically their spectrum of action.

Keywords: agriculture; contamination; environment.

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Introduction

The worldwide use and trade in herbicides increases each year (Carneiro et al., 2015). Although many changes and innovations arise to meet the market needs, such as, for example, the advent of transgenic crops, the use of herbicides stands out, since most of these innovations do not prevent the emergence of weeds in the field. Allied to the opening of new agricultural frontiers, the use of genetically modified organisms and the great demand for food, there is an increase in soil contamination by such products (Ximenes, Gomes, Ximenes, & Pires, 2020).

Among the most used herbicides glyphosate - N- (phosphonomethyl) glycine stands out. as an active ingredient in Roundup formulations, glyphosate is the most used in the world, with an increasingly investigated impact (Carretta, Cardinali, Masin, Zanin, & Cederlund, 2020). It is a non-selective active ingredient for the functional groups carboxyl, amino and phosphonate, having a strong attraction with the mineral fraction of the soil (Hermansen et al., 2020) and one of the most worrying herbicides in the world today (Morales, Allegrini, Basualdo, Villamil, & Zabaloy, 2020). Their losses to the environment are undesirable due to potential environmental risks, such as soil and water contamination (Wijekoon & Yapa, 2018), where their continued use has become an environmental issue (Garba, Samsuri, Othman, & Hamdani, 2019).

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The glyphosate herbicide, despite being developed more than 40 years ago, is still present, due to its low toxicological profile and high efficiency in weed control, important for the expansion of agricultural areas. the countries that stand out in scientific research on the glyphosate herbicide are Argentina, Brazil, United States, France, Germany, Italy, Australia, Canada, China, Spain, Hungary, United Kingdom, Thailand, South Africa, India, Iran, Finland, Malaysia, New Zealand, Russia, Sri Lanka, all of which have an agricultural base in their economy.

In search of answers and to make responsible decisions about an herbicide, it is necessary to know its spectrum of action, its effects, and its final destination in the environment. Scientific research provides a foundation for managers, producers, and decision-makers, reducing uncertainties concerning the product used, always with a focus on sustainability.

The objective of this article is to analyze the national and international scientific publications on the environmental contamination issues of the herbicide glyphosate and its metabolite aminomethylphosphonic acid (Ampa). the idea is to contribute to the elaboration of inferences that may help professionals, managers, and academics in future research.

Material and methods

An exploratory literature review study was performed looking for papers between the years of 2010 and 2020, which comprises five stages covered in different ways, namely:

1) establishing the research question and objectives;
2) establishing of criteria for inclusion and exclusion of articles (screening and selection of the sample);
3) definition of the information to be extracted from the selected articles;
4) analysis and interpretation of the results and;
5) presentation of the review and synthesis of knowledge.

The review sought to identify academic studies that provided data on the use of the herbicide glyphosate. the research was carried out through access to the Capes Journals portal - Web of Science and Scopus, selecting the main works associated with contaminants aimed at the use of the herbicide glyphosate. in the search performed 83 articles were found, 42 of which through the Web of Science platform and 41 via the Scopus platform, however, 21 articles were included in both platforms, so they were counted only once, totaling 62 articles. the articles were selected by combining the descriptors ‘contaminant of soil and herbicide glyphosate’.

Of the 62 articles initially selected, all were considered relevant to the study, given their scope on the proposed theme, evaluating which met the established inclusion criteria:

1) research papers that met the proposed objectives;
2) publications between 2010 and 2020, this period being defined due to the preference for more up-to-date data on the subject at hand.

To verify the coherence with the objective of this research and for data treatment, a detailed reading of the selected research papers was done. the ideas were grouped by similarity, to compose a narrative synthesis of the results and discussion of the knowledge that served as input for discussion.

Results and discussion

The search in the scientific literature returned 62 publications of indexed journals based on pre-established criteria, distributed in several themes (Table 1). Regarding the themes worked, biological tests in environments contaminated by glyphosate was the most researched topic (14.3%), followed by adsorption of the herbicide (12.7%) (Figure 1).

Table 2 shows the temporal distribution of the publications of the works referring to the glyphosate herbicide, showing the growing interest of the academic area on the use of the herbicide.

For both platforms, the interest of several countries, mainly those with marked agricultural production, is observed in developing research related to the herbicide glyphosate. Emphasis can be on Argentina, Brazil, USA, and France, with 7 publications, followed by Germany and Italy, both with 5 publications (Figure 2). Is an evolutionary trend concerning scientific research on the topic, as it reaches all continents. Europe stood out to the research numbers, with 38.7% works, while Africa and Oceania had the lowest number of productions (1.6%).

Generally, research topics are closely related to a personal stimulus by the researcher, current problem situation, geographic location, among others. in this sense, there is a diversity of works to meet the most varied objectives. However, some topics such as biological tests, adsorption, and surface waters were given greater emphasis, showing the concern with the environmental destination of the herbicide and the search for answers to managers and agricultural and forest producers.

Table 1. Themes addressed by the authors regarding the herbicide glyphosate between the years 2010 and 2020.

Research topic	Country	Authors	Periodicals
Adsorption	Malaysia; Patagonia; Canada; Germany; Brazil; Will	Carneiro et al., 2015; Cederlund, Börjesson, Lundberg, and Stenström; 2016; Zheng et al., 2017; Flores et al., 2018; Garba et al., 2019; Xiao and Meng, 2020	Ecotoxicology and Environmental Safety Nature; Environmental Science and Pollution Research; Bulletin of Environmental Contamination and Toxicology; Water Air and Soil Pollution; Journal of Environmental Management; Electronic Journal of Environmental, Agricultural and Food Chemistry
Adsorption, leaching	Finland	Hagner et al., 2015	Journal of Environmental Management
Adsorption, leaching and mineralization	Italy	Carretta et al., 2020	Journal of Hazardous Materials
Water anda sediment	Spain	Maqueda et al., 2017	Science of the Total Environment
Water in the dilution of contaminants	Brazil	Vale, Netto, Xavier, Barreto, and Silva, 2019	Journal of Cleaner Production
Water, soil and sediment	Thailand	Maneein et al., 2011	Research Journal of Chemistry and Environment
Groundwater	Canada	Van Stempvoort, Roy, Brown, and Bickerton, 2014	Chemosphere
Surface water	Australia; Argentina; Italy; Brazil, USA	Shipitalo et al., 2011; Masiol et al., 2018; Fernandes et al., 2019; Okada, Allinson, Barral, Clarke, and Allinson, 2020; Mac Loughlin, Peluso, Aparicio, and Marino, 2020	Water Research; Science of the Total Environment; Environmental Science and Pollution Research
Surface and groundwater	Argentina; Italy; Hungary	Székács, Mörtl, and Darvas, 2015; La Cecilia et al., 2018; Lutri et al., 2020;	Science of the Total Environment; Water Research; Journal of Chemistry
Association with other herbicide	USA; UK	Jeffries et al., 2017	Horttechnology; Chemosphere
Biodegradation	Italy	La Cecilia et al., 2018	Water Research
Biodegradation in water	India	Singh et al., 2019	Journal of Environmental Chemical Engineering
Bioremediation	Russia; Sri Lanka	Bois et al., 2011; Wijekoon and Yapa, 2018	Mikologiya I Fitopatologiya; Groundwater for Sustainable Development; Journal of Soils and Sediments
Historical consumption	Argentina; Germany; USA	Mogusu, Wolbert, Kujawinski, Jochmann, and Elsner, 2015; Paudel, Negusse, and Jaisi, 2015; Morales et al., 2020	Journal of Microbiological Methods; Analytical and Bioanalytical Chemistry; Soil Science Society of America Journal
Residual effect on plants	France	Serra et al., 2019	Science of the Total Environment
Phytoremediation	South Africa	Jacklin, Brink, and Waal, 2019	Water as
Interaction with other compounds	France	Geng et al., 2021	Chemosphere
Leaching	Germany; Italy; USA; France	Napoli, Cecchi, Zanchi, and Orlandini, 2015; Ki, Ray, & Hantush, 2015; Cederlund and Börjesson, 2016;	Journal of Hazardous Materials; Journal of Environmental Quality; Water Research; Environmental Science Research
Modeling for biotic and abiotic degradation	Australia	La Cecilia and Maggi, 2020	Mathematics and Computers in Simulation
Modeling: biodegradation	Australia	Tang et al., 2020	Mathematics and Computers in Simulation
Soil and water	Thailand	Aungudornpukdee, 2019	Journal of Health Research
Soil	Argentina; Hungary	Szekacs et al., 2014	Science of the Total Environment; Carpathian Journal of Earth and Environmental Sciences
Sorption	Spain	Hermansen et al., 2020	Geoderma; Journal of Near Infrared Spectroscopy
Biological test with contaminated environment	Brazil; UK; Argentina; France; Italy	Druart, Millet, Scheifler, Delhomme, and Vaufleury, 2011; Taccari, Comitini, Casucci, and Ciani, 2011; Druart, Scheifler, Millet, and Vaufleury, 2012; Kreutz et al., 2014; Leveroni, Caffetti, and Pastori, 2017; Ximenes et al., 2020; Pochron et al., 2020	Soil & Sediment Contamination; Chemosphere; Caryologia; Brazilian Journal of Medical and Biological Research; Applied Soil Ecology; Journal of Soils and Sediments; Journal of Soils and Sediments; International Biodeterioration and Biodegradation; Journal of Hazardous Materials
Heat treatment for decomposition	Australia	Narimani and Silva, 2020	Environmental Science-Processes & Impacts

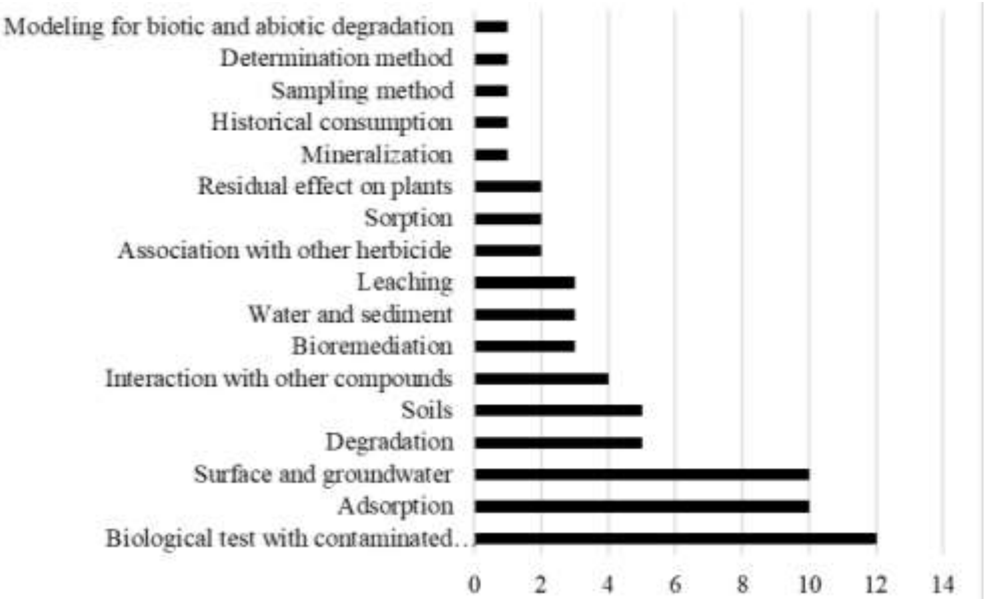

Figure 1. Distribution of articles in relation to research topics.

Table 2. Distribution of publications referring to the herbicide glyphosate between the years 2010 to 2020 on the Web of Science and Scopus platforms.

Year	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	2020
Number of papers published	1	8	1	1	5	11	3	5	4	11	12

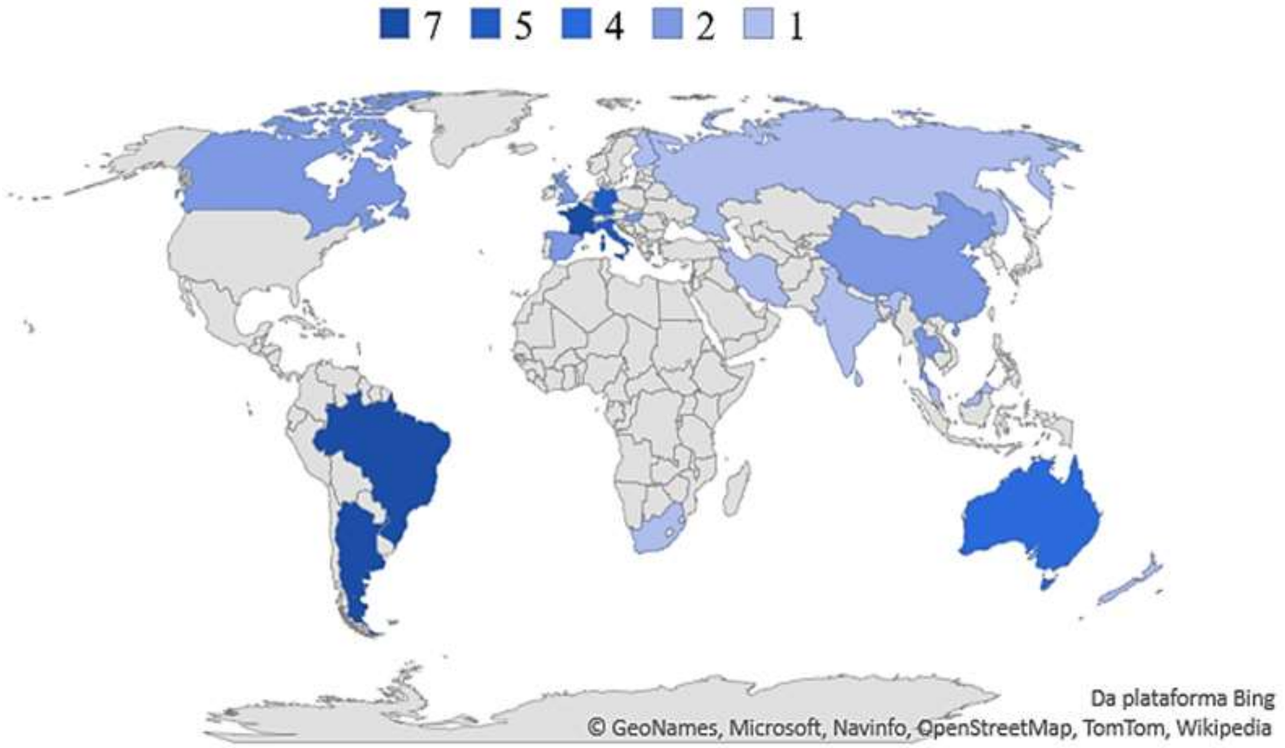

Figure 2. the number of papers published by countries that developed research on glyphosate herbicide (2010 - 2020).

Glyphosate is a non-selective and effective weed control herbicide, being used worldwide (Carneiro et al., 2015). It emerged after the development of genetically modified products, when herbicides capable of meeting the demand of producers were sought , thus reaching the worldwide proportion (Pochron et al., 2020). Its use has negative effects on the environment, such as, for example, surface water contamination (Okada et al., 2020) (Fernandes et al., 2019), groundwater contamination (Van Stempvoort et al., 2014), and residual effect on plants (Serra et al., 2019).

Biological tests

The biological tests carried out with herbicides are important to know their effects on the biota. This subject has gained global importance when it comes to the glyphosate herbicide, with the theme being the leader of the research carried out between the years 2010 to 2020 and Brazil, United Kingdom, Argentina, France, and Italy the countries involved in these studies.

Tests were carried out with *Folsomia candida* as a biological sensor in environments contaminated with the herbicide glyphosate were shown to be sensitive at concentrations of 1:50 and 1: 100 of the herbicide, since the organism has a preference for environments with no glyphosate (Ximenes et al., 2020). Another study carried out with a living organism showed that there were changes in blood cells, gills, and liver of *Piaractus mesopotamicus* (pacu-caranha) submitted to 96 hours in contact with Roundup Full II® (2.75 mg L −1). This is considered a sublethal concentration, with the three tissues showing significantly larger damage compared to the negative control (Leveroni et al., 2017).

To evaluate the embryotoxicity of some herbicides frequently used in vineyards in the solid, liquid, and gaseous phases of the soil, including glyphosate, tests were carried out with snail eggs. for the solid phase of the soil, two herbicides were tested: Corail® (tebuconazole-based fungicide) and Bypass® (glyphosate-based herbicide). Bypass® proved to be approximately 230 times less toxic than Corail®. in addition to the difference in the way the two substances act, the lower toxicity of the same in snail eggs can also be explained by the faster degradation of glyphosate in the soil (Druart et al., 2012). These data corroborate with studies that investigated its fate in the soil, the transfer, and the effects on the land snails of the herbicide glyphosate, showing a low effect of the herbicides in relevant concentrations in the soil. the detection of residues in tissues indicated a potential risk of transfer to the food chain (Druart et al., 2011).

To determine the glyphosate toxicity in the soil, the simple respirometric technique indicated that there is contamination, even at low concentrations (250 ppm), and the rate of oxygen consumption provides a better indication of microbial toxicity compared to cumulative oxygen demands. after 20 hours due to the different effects of these toxic compounds on soil microbial communities (Taccari et al., 2011). Another study with different concentrations of glyphosate in the soil points out that Eisenia fetida lost between 14.8 and 25.9% of its biomass and survived a stress test for 22.2 to 33.3% less time than the worms that live in uncontaminated soil (Pochron et al., 2020).

In a study seeking to evaluate the kinetics of antibody production in fish, it was found that, in fish exposed to atrazine and glyphosate, the secretion of specific antibodies was greater than in non-exposed inoculated fish. Such a result can be harmful, since fish that live in water contaminated with herbicides may have increased concentrations of non-specific antibodies that can mediate tissue damage (Kreutz et al., 2014).

Adsorption, leaching and mineralization

There are many techniques to mitigate or reduce the impact of herbicides on the natural environment (water, soil and plants). Among such techniques is adsorption, where alternative materials are used to remove the herbicide from the medium. Some authors have studied this technique, showing satisfactory results with materials such as cow dung and rice husk, which in addition to their availability, can be recommended as economical adsorbents for glyphosate and its metabolite. Expansive clays, such as Montmorillonite, can also be said to be glyphosate adsorbents, given the affinity between the clay molecules and the herbicide (Zheng et al., 2017; Flores, Sánchez, & Afonso, 2018; Garba et al., 2019).

Studies with resin loaded with Fe^{3+} (R – Fe^{3+}) have shown to be efficient in the adsorption of the herbicide glyphosate. the adsorption of glyphosate coordination by R – Fe^{3+} not only solves environmental problems of glyphosate but also brings important economic benefits through efficient glyphosate recovery (Xiao & Meng, 2020). Another economically viable adsorbent studied was wood-based biochar submitted to heat and iron treatment, to increase its adsorptive properties. When mixing equal parts of thermally treated biochar and biochar treated with iron salt, was produced a material with a high capacity adsorbent for glyphosate. It also tells that combining several altered biocarbon can be feasible, creating a multifunctional and comparatively inexpensive filter material, capable of adsorbing a variety of chemically different contaminating organic substances (Cederlund et al., 2016), in addition to the potential to reduce leaching soil (Hagner et al., 2015), which can reach up to one meter from the soil column (Napoli et al., 2015).

Seeking to investigate whether the presence of a surfactant based on alkyl polyglucoside, Triton CG-110, affects the adsorption, leaching, and mineralization of glyphosate in the soil (sandy and clayey), it is concluded that it reduces the adsorption of glyphosate in washed sand, while adsorption in clayey soil was not affected. Triton CG-110 did not significantly affect glyphosate leaching in washed sand or mineralization in any of the soils tested, indicating that the surfactant is unlikely to significantly affect the environmental fate of glyphosate in the soil at environmentally relevant concentrations (Carretta et al., 2020), and, in many cases, the leaching potential of glyphosate varies from low to insignificant (Ki et al., 2015).

Studies have also been carried out with alkyl polyglucoside as a foaming agent in sand columns, to verify its effects on glyphosate leaching, however, it was found that if used at the recommended concentration of 0.3% it does not significantly affect leaching, and may have effects only if used in higher concentrations (Cederlund & Börjesson, 2016).

Sorption

Sorption is the effect of gases or liquids being incorporated into a material of a different state and adhering to the surface of another molecule and plays an important role in the destination and transport of glyphosate in the environment. the sorption coefficient of glyphosate and, therefore, its mobility, varies greatly between different types of soil, since the basic properties of the soil as well as the visible near infrared spectroscopy (vis-NIRS) can predict the variation of the sorption coefficient between soil samples with high precision and, therefore, that the glyphosate sorption for a soil can be determined with vis-NIRS (Hermansen et al., 2020).

Surface and groundwater

One of the most relevant topics in studies of contaminants is associated with surface waters, given their destination. in agricultural countries, where the use of herbicides is widely used, there is a concern with surface waters, as they are potentially contaminated.

A study performed in Italy showed that the waters of the Veneto River are contaminated with glyphosate herbicide, also pointing out some critical points, precisely in the places where the vineyards are located (Masiol, Giannì, & Prete, 2018). Glyphosate was also detected in more than 90% of water samples collected in the Córrego Carnaval basin, in Buenos Aires (AR), areas marked by horticultural production (Mac Loughlin et al., 2020). A similar study carried out on the eastern slope of Las Peñas Mountain - Córdoba (AR) shows that glyphosate was detected in 66% of the surface water samples and 15.8% of the groundwater samples, whereas in 33% of the surface water and 15,8% of groundwater has detected its metabolite (Ampa), showing that application for decades under the current agricultural model exceeds the potential for soil and unsaturated zone degradation, causing groundwater contamination (Lutri et al., 2020).

A study carried out in Australia, evaluating water samples from 10 rural streams, 30 urban rainwater marshes, and 9 urban streams located in and around Melbourne, showed that the glyphosate herbicide and its metabolite are present in most water samples from urban surface waters (77%). While they were detected in only 4% of rural surface water samples, emphasizing that wetlands and streams associated with urban land use are vulnerable to glyphosate contamination (Okada et al., 2020) and may pose a risk of aquifer contamination at a maximum depth of 1.5 m within 50 years of use of glyphosate (Serra et al., 2019).

A solution for removing glyphosate from surface water was presented in a study carried out in Brazil with the use of epilithic biofilms, which pointed out that they are capable of bioaccumulating the herbicide glyphosate and its metabolite Ampa, which can be used to distinguish different anthropogenic pressures in basins hydrographic (Fernandes et al., 2019).

Still in Brazil, a study proposing to evaluate the gray water footprint for sugarcane, which takes into account the use of pesticides, the higher volumes of gray water and the higher classifications of the mixture were due to amicarbazone and hexazinone, the lowest being values were due to paraquat and glyphosate (Vale et al., 2019).

Degradation, biodegradation and bioremediation

There are model studies that can estimate the potential for biodegradation, that is, estimate the biodegraded fraction of a contaminant in the soil (Tang et al., 2020). However, there are many uncertainties about the parameters commonly used in mathematical models, uncertainties which are used as a guide of the decision-making to farmers, regulatory bodies, and managers to manage and protect the environment in a sustainable way (La Cecilia & Maggi, 2020).

The microbial degradation of glyphosate is the main dissipation mechanism acting in most environments. A quantitative polymerase chain reaction (qPCR) study was developed for a key enzyme, CeP lyase that breaks down the CeP bond, via quantification of the gene encoding phnJ, concluding that direct quantification of this gene may be an economical alternative to determine the potential for degradation of glyphosate in different matrices since it showed high efficiency and sensitivity, as well as specific detection of the target sequence in a wide range of taxonomic groups (Morales et al., 2020).

To assess the sources and degradation of the herbicide glyphosate and its metabolite Ampa, concentration measurements are inconclusive. One study presented a specific analysis of the glyphosate compound and Ampa by two-step derivatization in combination with gas chromatography/double element isotope ratio mass spectrometry (15 N/14 N, 13 C/12 C). the values of glyphosate isotopes during degradation by MnO_2 were clearly out of the commercial product range, highlighting the potential of the combined isotope analysis of carbon and nitrogen to trace sources and degradation of glyphosate (Mogusu et al., 2015), whose reaction of degradation occurs very quickly, indicating sorption as a prerequisite for degradation (Paudel et al., 2015).

The potential of plants to remove, remedy or immobilize the glyphosate herbicide in a growth matrix using natural biological, chemical or physical activities was studied in a laboratory phytoremediation system, allowing to evaluate its phytoremediation capacity, where the contaminant is present, by binding to the root structure and cell walls and hemicellulose within the cell wall, being pointed out as a simple and inexpensive method of biofiltration (Jacklin et al., 2019).

The degradation potentiation can also be done with the use of microorganisms degrading the herbicide, as a study carried out with the use of two bacterial isolates Pseudomonas sp., Bacillus sp. and mixed culture of Pseudomonas sp. and Bacillus sp. They were shown to be effective bioremediation agents for the treatment of glyphosate-contaminated soils and aquifers (Wijekoon & Yapa, 2018), and plants can help stabilize pesticides and bacteria help mitigate environments subject to conditions variables such as rainwater basins (Bois et al., 2011).

Residual effect on plants

Plants bordering places where pesticides are applied are also influenced due to their volatilization, leaching, and displacement. Such effects can be felt not only in the aerial part but also in the roots of the plants. With the increase in glyphosate dosage, the roots of Arabidopsis thaliana seedlings show a significant reduction in length, indicating that the herbicide slows down its development (Serra et al., 2019).

Soil

Soil is the environmental destination of pesticides. in analyzes carried out in a study carried out in the province of Phetchabun, Thailand, where 76.83% of the population uses the herbicide glyphosate, it was pointed out that the soils were contaminated with the herbicide and its metabolite Ampa. the analyzed water did not contain the contaminant (Aungudornpukdee, 2019).

A study carried out in Hungary, evaluating more than 2000 samples of surface water and soil between the years 1990 and 2015 did not detect glyphosate, even though the region has more than 30 records of glyphosate-based formulations, justified by its rapid degradation (Székács et al., 2015).

Conclusion

Studies indicate that there is worldwide interest in the topic, as it has agricultural and forestry use as its final destination. Although the studies carried out in the last 10 years are comprehensive, results are still lacking concerning the monitoring of the herbicide glyphosate in the soil, due to its specific properties.

More specific studies of this herbicide in different environmental conditions are important for correct use, thus reducing the direct or indirect environmental damage that this product may cause, guiding decisions about it, and proposing guidelines that technically and scientifically support its spectrum of action.

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