

Damage to the Environment Arising From Pesticides: An Evaluation From The International Literature

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Abstract: This paper aims to make an integrative review of the literature on scientific papers of an international database on the subject of pesticides, identifying those related to the degradation of the environment, analyzing its main objectives, results, year of publication and place of study. A period of 18 years, periodicals of levels A1 and A2 of free access were used, with Digital Object Identifier System and that they fit in the area of Engineering I of the Commission of Improvement of Personnel of the Superior Level. Afterwards, the abstracts were analyzed and classified by central object of study, producing figures and comparative tables for the interpretation of the data. Only 10 articles were found that fit the inclusion criteria, being 80% of exploratory research, 7 of the 10 papers were published after 2010, showing a growth of production from one decade to another of 234%, the focus is on water, with 50% of the papers and were carried out mainly in developed countries, corresponding to 70% of the publications. The results showed that exposure to the substances was higher than allowed by the countries where the analyzes were performed and that there were several adverse effects from exposure.

Key words: pesticides, agrochemical, environment, environmental quality

1. Introduction

Pesticides, according to the current legislation (Federal Law No. 7,802, of July 11, 1989), are products and agents of physical, chemical or biological processes, intended for use in the sectors of production, storage and processing of agricultural products, on pastures, forest protection, native or implanted, and other ecosystems. The Law also deals with urban, water and industrial environments, whose purpose is to change the composition of flora or fauna, in order to preserve them from the harmful action of living beings considered noxious [1].

The concepts related to pesticides are diverse and serve the interests of those who use them. For example, the concepts for pesticide and agricultural defensives are associated with the sector that uses it: such as agrochemical companies that prefer to use the term "agricultural defensives", because in their view the products are used with the objective of protecting Agricultural production. The Brazilian National Agency of Sanitary Surveillance (ANVISA) uses the word "agrotoxics" because it conveys the idea of the potential risks of the product and alerts workers and the population.

One of the most robust sectors of the Brazilian economy, agribusiness is responsible for about 25% of the Gross Domestic Product (GDP), accounts for 20% of jobs and stands out in the country's export basket [2].

With the planting of important commodities such as soy, corn, sugar cane, coffee and cotton, the Brazilian pesticide market has grown by around 190% in the last decade [3], at a rate of expansion twice higher than that presented by the world market of 93%, in the same period [4]. In the 2017/2018 harvest, the combined consumption of herbicides, insecticides and fungicides,

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among others, moved 549,280.44 tons of active ingredients in Brazil [5].

About 487 active ingredients and 2,400 pesticide formulations are registered with the Ministry of Health, Ministry of Agriculture, Livestock and Supply (MAPA) and the Ministry of the Environment and are allowed in Brazil, according to the criteria of use and their indications. However, of the 50 most used in Brazil's crops, 22 are prohibited in the European Union [3]. Agrochemical formulations have been increasing: only in the years 2018 and 2019, 450 [5, 6] and 474 new pesticide formulations were registered by MAPA, respectively [7], a historic record.

A detail that should be taken into account for the statistics for the years 2018 and 2019 is that all products classified exclusively as adjuvants are no longer considered pesticides, and are no longer accounted for, according to Act No. 104, of November 20, 2017, from MAPA, published in the DOU of November 21, 2017 [6]. Adjuvants have always had a relevant participation in the ranking of the most commercialized products in Brazil, such as mineral and vegetable oils [6]. In this line of thought, it can be said that the number of records would be much higher than those presented in the statistics if the adjuvants continued to be part of the pesticides accounted for.

The expected action of pesticides occurs by the presence, in its composition, of a chemical molecule that affects the normal biological activity of living beings sensitive to it, inhibiting it. This component is called an active ingredient [8]. The classification of pesticides (Agronomic Classes), in relation to their use, is defined by the power of action of the active ingredient on target organisms, such as insecticides (controlling insects); acaricides (mites); nematicides (nematodes); fungicides (fungi); herbicides (weeds); growth regulators; among others [9]. However, most studies are carried out in laboratories, with animals and under control of variables; few in real situations, with workers continually exposed to the influence of a complex network of variables [3].

The complexity of evaluating the behavior of a pesticide, once applied, is due to the influence of the agents that act by causing its physical displacement and its chemical and biological transformation [10] The amount of organic matter, texture and structure, which result in the porosity of a soil, are extremely important factors for the quality of the environment. Organic matter, for example, adsorbs most of the non-polar organic compounds, while the texture and porosity parameters are determinant for the soil's ability to retain or not a solution [11]. Another factor that is influenced by the characteristics of the soil is the degradation of the active ingredient and the extent of biodegradation, since this only occurs if it is available. Thus, the half-life of a pesticide in the soil is a quantity that can vary according to environmental parameters [12].

Thus, when pesticides are used, regardless of the application method, there is great potential to reach the soil and water, mainly due to the winds and rainwater, which promote drift, washing of treated leaves, leaching and erosion. In addition, whatever the path of the pesticide in the environment, man is invariably its potential recipient [10], considering that concentrations of pesticides have been found even in breast milk [3], being found remnants of organochlorines DDT and HCH [13], and in human blood [14]. Among some diseases that can be caused by agrochemicals, we highlight those that occur due to the ingestion of nano-concentrations: a) for short periods of time in an acute manner: headache, dizziness, nausea, vomiting, muscle fasciculation, paraesthesias, disorientation, difficulty respiratory, coma; b) in a chronic form: reversible paresis and paralysis, irreversible delayed neurotoxic action, pancytopenia, neuropsychological disorders; c) for long periods of time in an acute form: hemorrhages, hypersensitivity, therapogenesis, fetal death; d) in a chronic way: irreversible brain damage, malignant tumors, testicular atrophy, male sterility, neurobehavioral changes, peripheral neurifes and contact dermatitis [15].

Pesticides and their potential metabolites have proven to be dangerous to human and environmental due to their bioaccumulation health. and biomagnification and, for this reason, studies aiming at their behavior in the environment are essential. Thus, studies and research on the effects of pesticides have focused on human beings, which is a totally opposite reality in relation to the environment, in which the studies are not conclusive. Thus, considering these aspects, this study aims to do an integrative literature review in scientific papers, identifying those that are related to the degradation of the environment quality. Still, it is intended to identify its main aims, results, separating them into categories of study (water, soil, air and bibliographic review), year of publication and place of production of the technical work.

2. Material and Methods

This study is an integrative literature review on the effects of pesticides on the environment. An integrative review is a specific review method that summarizes past empirical or theoretical literature to provide a more comprehensive understanding of a particular phenomenon or problem [11]. Integrative reviews bring together several sources of data that increase the holistic understanding of the topic of interest. Well-integrated reviews present the state of science, contribute to the development of theory and have direct applicability to practice [16].

This review was carried out from the *Science Direct* webpage of scientific papers database¹. The following advanced search filters were used: *pesticides*; *and*; *environment*; in the fields: *keywords*. These words were used, because these are the most common words for this topic and the database language is English. Also, only keywords field was used, because in the case of researches that present extensive results, the use of questions that guide the research is crucial [11]. Thus, articles that did not use the keywords correctly were not found in the database.

The timeframe used was 18 years, from the year 2002 to the date of February 6, 2020. Only the journals and topics made available on the page that fit the area of Engineering I of the Higher Education Personnel Improvement Commission (CAPES), of the Ministry of Education of Brazil, were used. This area of knowledge encompasses Civil Engineering; Transport; and Sanitary. Also, to maintain the quality of the work, only the journals that had the Classification of Journals — Qualis, from the evaluation quadrennium (2013-2016), at levels A1 and A2, which are considered the best journals used as a source of data collection is shown in Table 1.

The classification is performed by committees of consultants from each evaluation area, following criteria defined by the area, which seeks to reflect the relative importance of the different journals for a given area. The stratification of the quality of this production is done indirectly. Thus, Qualis measures the quality of papers and other types of production, based on the analysis of the quality of scientific journals. The classification of journals is carried out by the evaluation areas and undergoes an annual update process. These vehicles are framed in strata indicating quality, where A1 is the highest; A2; B1; B2; B3; B4; B5; and C of lower value [17].

The next step was the analysis of the abstracts seeking to identify the theme, aims, methodology and results found. If they did not identify with the purpose of this study, they were discarded. From this, papers were classified by central study goals, such as water, soil, air and bibliographic review. From this information, Table 2 was constructed with the synthesis of information from the papers, four figures and Table 3 (which makes the bibliographic reviews) containing the synthesis of the subjects, aims, methodologies and results.

Only journals that had free access were used, that is, references were not used that needed to be acquired by

¹ http://www.sciencedirect.com.

means of payment. Also, only references that had the *Digital Object Identifier System* — DOI were used.

For the production of comparative figures between the existing information, data from the International Monetary Fund [18] were used to determine the countries that have an advanced economy (developed countries) and developing economies (developing countries). Thus, for this study it was considered: Spain, Italy, United States of America (USA), Germany, South Korea, Portugal, Australia, United Kingdom and Japan considered as developed and India, Argentina, and Poland considered as developing. The last step was the interpretation of the data.

Qualis (2013-2016)	Journal	
A1	Atmospheric Environment	
A1	Chemosphere (Oxford)	
A1	Ecotoxicology and Environmental Safety	
A1	Environmental Research (New York, N.Y. Print)	
A1	Journal of Environmental Management	
A1	Science of the Total Environment	
A2	Marine Pollution Bulletin	
A2	Talanta (Oxford)	
	Qualis (2013-2016) A1 A2	

Table 1Journals used in this study.

3. Results and Discussion

Preliminarily, the selection process with the keywords and selected journals resulted in 56 papers selected in the database, as can be seen in the research shortcut² provided by the *Science Direct* database. After the development of the papers analysis process, this number dropped to 10 papers that fit the inclusion criteria for this study, i.e., only 17.86% of the papers preliminarily selected.

Tables 2 and 3 show the identification data of the paper, its characteristics regarding the title, authors, DOI, journal, year of publication, type of research, field of study, place where the research was developed, specific aims, scientific methodology, main results and discussions, respectively. Through these Tables, it is possible to notice that approximately 80% of the papers present an exploratory research, that is, they are

concerned with identifying the factors that determine or contribute to the occurrence of the phenomena, the relationship between cause and effect of two or more variables [19, 20], usually bringing something new to scientific knowledge. There are also papers that are bibliographic surveys, that is, reviews of the existing literature.

From Table 2, it can be seen that most studies were produced by more than four collaborators (teams), which indicates the complexity of the study on pesticides and their actions on the environment. It is also noticed that there are many studies being developed with the help of more than one country, implying that the subject is of interest and relevant for both nations involved. Still, it is noted that the titles of the papers summarize the main theme addressed in the paper and demonstrate the contribution to the construction of knowledge in this area. Seven of the ten papers were published after 2010, which points to the growing interest in the subject in this decade. Through Table 2, it was possible to generate Figs. 1 and 2 for comparison of information.

In Fig. 1, it is possible to notice that the focus of published studies is concentrated on water, with 50%

² Search results: 56 results found for pub-date > 2001 and KEYWORDS (pesticides) and KEYWORDS (environment) AND LIMIT-TO (yearnav, "2020, 2019, 2018, 2017, 2016, 2015, 2014, 2013, 2012, 2011, 2010, 2009, 2008, 2007, 2006, 2005, 2004, 2003, 2002") AND LIMIT-TO (cids, "272394, 271800, 271852, 272592, 271798, 272576, 271825, 271360", "Environmental Research, Science of The Total Environment, Chemosphere, Journal of Environmental Management, Atmospheric Environment, Eatoxicology and Environmental Safety, Marine Pollution Bulletin, Talanta").

Table 2 Characterization of scientific pape

ID	Paper title	Authors	Doi	Journal	Publication year	Type of research	Study field	Place where the research was developed
P1	Nationwide monitoring of polychlorinated biphenyls and organochlorine pesticides in sediments from coastal environment of Korea	S. H. Hong, U. H. Yim, W. J. Shim, D. H. Li, J. R. Oh	10.1016/j.chemosphere. 2005.12.056	Chemosphere	2006	Exploratory research	Water	South Korea
P2	Baseline occurrence of organochlorine pesticides and other xenobiotics in the marine environment: Caribbean and Pacific collections	Robert Menzies, Natalia Soares Quinete, Piero Gardinali, Douglas Seba	10.1016/j.marpolbul.201 3.03.003	Marine Pollution Bulletin	2013	Exploratory research	Water	USA
Р3	A comparative assessment of the transformation products of S-metolachlor and its commercial product Mercantor Gold® and their fate in the aquatic environment by employing a combination of experimental and <i>in silico</i> methods	Lukasz Gutowski, Oliver Olsson, Christoph Leder, Klaus Kümmerer	10.1016/j.scitotenv.2014 .11.025	Science of the Total Environment	2014	Exploratory research	Water	Germany
P4	Occurrence and ecological risks from fipronil in aquatic environments located within residential landscapes	Jun Wu, Jian Lu, Hai Lu, Youjian Lin, P. Chris Wilson	10.1016/j.scitotenv.2014 .12.103	Science of the Total Environment	2015	Exploratory research	Water	USA
P5	Monitoring a large number of pesticides and transformation products in water samples from Spain and Italy	⁷ N. I. Rousisa, R. Bade, L. Bijlsma, E. Zuccato, J. V. Sancho, F. Hernandez, S. Castiglioni	10.1016/j.envres.2017.0 3.013	Environmental Research	2017	Exploratory research	Water	Spain and Italy
P6	Impact assessment of treated/untreated wastewater toxicants discharged by sewage treatment plants on health, agricultural, and environmental quality in the wastewater disposal area	Kunwar P. Singh, Dinesh Mohan, Sarita Sinha, R. Dalwani	10.1016/j.chemosphere. 2003.10.050	Chemosphere	2003	Exploratory research	Water and Soil	India
P7	Effect of olive-mill waste addition to agricultural soil on the enantioselective behavior of the chiral fungicide metalaxyl	Beatriz Gámiz, Rafael Celis, M. Carmen Hermosín, Juan Cornejo	10.1016/j.jenvman.2013. 04.055	Journal of Environmental Management	2013	Exploratory research	Soil	Spain
P8	Atmospheric concentrations and deposition of organochlorine pesticides in the US Mid-Atlantic region	Rosalinda Gioia, John H. Offenberg, Cari L. Gigliotti, Lisa A. Totten, Songyan Du, Steven J. Eisenreich	10.1016/j.atmosenv.200 4.12.028	Atmospheric Environment	2005	Exploratory research	Air	Italy and USA
Р9	Application of chemometric methods to environmental analysis of organic pollutants: A review	Sílvia Mas, Anna de Juan, Romà Tauler, Alejandro C. Olivieri, Graciela M. Escandar	10.1016/j.talanta.2009.0 9.044	Talanta	2009	Literature review	Review	Spain and Argentina
P10	Solventless and solvent-minimized sample preparation techniques for determining currently used pesticides in water samples: A review	Maciej Tankiewicz, Jolanta Fenik, Marek Biziuk	10.1016/j.talanta.2011.0 8.056	Talanta	2011	Literature review	Review	Poland



Fig. 1 Distribution of publications in study fields.

Table 3	Synthesis of the su	bjects, aims.	, methodologies and	results of scientific papers.
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ID	Specific aims	Scientific methodology	Results and discussion
P1	Extensive monitor initiative regarding the sediment organochlorine (OC) pollution in the general Korean marine environment.	A total of 138 surface sediment samples, representing the entire coastal region of Korea, were collected between 1997 and 2002. The samples were fractionated using high pressure liquid chromatography (HPLC) with size-exclusion column.	Contamination status of Korean coastal sediments with regard to OCs in general showed similar OC concentrations to those of other Asian countries. There was a significant correlation between distributions of most organochlorine contaminants with each other. OC contamination is closely related to shipping and industrial activities. Of the 7 sites categorized as highly polluted, 4 are in a harbor zone. Adverse effects to benthic communities are expected at the levels of OC contamination observed from harbor and industrial areas.
P2	Report the levels of persistent organic pollutants (POPs) in sea surface slicks collected on a global circumnavigation and monitoring potential endocrine disrupters in remote áreas of the world to assess their geographical distribution in marine and aquatic environments. The focus was on chlorinated benzenes, hexachlorocyclohexanes, polychlorinated biphenyls (PCBs) and organochlorine pesticides such as DDTs and metabolites, chlordane related compounds and	A total of 33 samples were collected between 1997 and 2001 near shore coastal marine locations and oceanic islands, atolls and reefs. The method used for extraction and clean-up of samples followed the procedures described by EPA method 8081B and NOAA Technical Memorandum 130. The quantification was made on a Hewlett–Packard series II 5890 gas chromatograph equipped.	Six different groups of chlorinated aromatic compounds were detected in the slick/water samples: chlorobenzenes, hexachlorocyclohexanes (HCHs), chlordane related compounds, organochlorine pesticides and other cyclodiene pesticides, DDTs and metabolites, and polychlorinated biphenyls (PCBs). Forty-seven individual compounds were measured. The profile of the individual compounds detected and also the profiles within a group were often different from site to site. While the highest concentrations of DDT and PCBs were observed in Panama, HCHs were higher in Rio Vista, California.
Р3	other cyclodiene pesticides. Compare the impact of Mercantor Gold®'s adjuvants on the biodegradation to the pure S-metolachlor (SM) and simulate the fate of the photo transformation products (TPs) in water environment.	The primary elimination of the parent compound monitored by using HPLC–UV. The degree of mineralization was evaluated with the non-purgeable organic carbon (NPOC) analysis. This approach allowed for a comparison of the degradation and transformation potential of SM with the commercial product Mercantor Gold® (MG). The generated TPs were analyzed in terms of ready biodegradability and the observed biotransformation products in silico (QSAR) prediction tools were applied to support structure elucidation of the generated photoproducts as identified with LC–UV–MS/MS and for the assessment of toxicity of TPs.	The S-metolachlor and Mercantor Gold® were not biodegraded. HPLC–UV analysis showed higher elimination of SM in MG compared to pure SM during photolysis. A total of 10 photo-TPs of SM and MG were identified. According to MS data and in silico predictions, chemical structures were proposed for all found photo-TPs. Likewise for the parent compounds, no biodegradation has been observed for their photo-TPs. The MG formulation does not affect the biodegradation process, but it influences the photolysis efficiency and potentially might result in faster formation of TPs in the environment.
P4	Investigate the occurrence of fipronil and its metabolites in aquatic environments in residentially developed landscapes, including five canals and three retention ponds.	Surface water samples were collected from eight drainage canals and ponds located within residential areas in the Indian River Lagoon watershed. A gas chromatograph equipped with dual electron capture detectors was used for pesticide analysis. Three methods were used to estimate the ecological risks of fipronil and its metabolites on aquatic ecosystems. First, the risk quotient (RQ) screening method for different non-target organism groups at two exposure levels (median and maximum detected concentrations) was performed to identify possible acute and chronic ecological risks. For sites where potential risks were identified by RQs, a probabilistic assessment was used to further characterize risks as the probability of acute and chronic effects, as well as the potentially affected fraction of species.	Fipronil was detected at four of the sites, with concentrations of 0.5–207.3 ng L ⁻¹ . Fipronil sulfone and fipronil sulfide were detected at three sampling sites, with concentrations ranging from 0.46 to 57.75 and 0.40–26.92 ng L ⁻¹ , respectively. The deterministic method indicated no risk to certain biotic groups (i.e. aquatic plants, fish, molluscs, and algae–moss–fungi), but did indicate risks to larval insects and crustaceans.
P5	Investigate a large number of pesticides and transformation products (TPs) occurrence in wastewater (WW) (influent and effluent) and surface water (SW) in two areas with high pesticide use (Spain and Italy).	Screening method for the determination of a large number of pesticides and TPs (selected on the basis of the priority pollutant list of the EU and the United States Environmental Protection Agency and the United Nations list of persistent organic pollutants) in wastewater (WW) and surface water (SW) from Spain and Italy. Liquid chromatography coupled to high resolution mass spectrometry (HRMS) was used to screen a database of 450 pesticides and TPs. Detection and identification were based on mass accuracy, fragmentation, and comparison of retention times when reference standards were available, or a retention time prediction model when standards were not available.	Seventeen pesticides and TPs from different classes (fungicides, herbicides and insecticides) were found in WW in Italy and Spain, and twelve in SW. Generally, in both countries more compounds were detected in effluent WW than in influent WW, and in SW than WW. HRMS proved a good screening tool to determine a large number of substances in water and identify some priority compounds for further quantitative analysis.
DC	assessment of treated/untreated	a Gas Chromatograph equipped with ECD/FPD and capillary column was employed for the analysis of	in all the environmental media, suggesting a definite
P0	wastewater toxicants discharged by sewage treatment plants on health,	BHC isomers, DDT isomers and metabolites, endosulfan, malathion, methyl parathion, ethion and	adverse impact on the environmental quality of the disposal area. The critical levels of the heavy metals in the soil for

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Fig. 2 Papers published over time.

of papers, followed by literature reviews with 20%. The areas of soil, air and water together with soil have not concentrated significant amounts of production in the past 18 years. The choice for water may have been due to the fact that water invariably became the final destination of pesticides deposited in all other

environments, because through the principle of osmosis, concentrations of substances in the atmosphere are diluted and precipitated by rain water, which infiltrates and leaches the substances present in the soil towards the groundwater that will emerge in springs and rivers. Likewise occurs with pesticides applied directly to crops.

Fig. 2 shows the distribution of papers organized by decades, showing a great increase in production from one decade to another, of 234%. This great growth in interest in the study of pesticides may be due to the growing evidence of cases of diseases in humans linked to large concentrations of pesticides in blood samples and human organs. Thus, studies are necessary to increase the safety of the use of substances in food production and in the development of new substances to be used.

In Fig. 3 can be seen that the publication of papers over 18 years has a standard deviation from one publication to more or less and the linear trend line shows a slight growth in its "y" axis. However, as there was not an expressive number of publications, it can be said that the number of publications is approximately uniform among the journals that have the best quality levels, except for the journal *Ecotoxicology and Environmental Safety*, which did not present any publication in this time period.

In Fig. 4 is possible to note that over the past 18 years, studies have been carried out mainly in developed countries, corresponding to 70% of

publications. On the other hand, developing economies have only two publications (20%) and cooperation between countries with different economies is even lower, with only 10% or a single paper in this entire period. The study of pesticides mainly by developed countries occurs due to the greater barrier of use of chemical substances in food production, which implies the use of these substances by countries that will export to such places. In this way, developed countries tend to bar the entry of food that uses substances that are proven to damage the environment and the health of its inhabitants.

From Table 3 it is possible to notice that most of the studied papers aims seek to analyze the exposure of a place in relation to a certain determined substance. It is, therefore, field research, whose main method of analysis is gas chromatography, which is currently the most established method for obtaining safe results from trace substances. The results showed that in the vast majority the exposure to the substances was higher than that allowed by the countries where the analyzes were carried out and that there are several adverse effects, resulting from the exposure.



Fig. 3 Papers publication over 18 years.



Fig. 4 Distribution of paper production by the level of the countries development

4. Conclusion

Although pesticides are widely present in everyday life, the production of quality studies on their effects on the environment is not numerically significant. This is noticeable by the low availability of papers in the largest and best journals in the area. Even so, there was a great growth in scientific production, which points to the importance of the topic today.

Still, it is noted that there is greater production of papers in developed countries. This may be due to their greater concern with the quality of the environment or that they have greater technological and scientific resources for the development of this type of studies. It is also noticed that most of the researches are carried out with water, through collections in the natural environment, using gas chromatography to define product concentrations. The results of most of these studies indicate the identification of several substances in concentrations higher than those considered acceptable in relation to the protection of the environment.

The assessment of the presence of pesticides in the environment is a complex and at the same time essential process. Complex, as there are numerous methodologies for only one assessed scenario, such as water, for example, and also because it requires researchers to have a thorough grasp of all topics related to the topics of the research aim. Likewise, it is an essential process because, through the correct relationship between the aims of the research and its results, it can enable the definition and development of appropriate strategies for the reduction, or elimination, of pesticides from the environment, creating laws and regulations more restrictive.

The importance of standardizing internationally used keywords is emphasized to prevent relevant papers from being identified in the available databases. Consequently, they are no longer read and appreciated by researchers interested in their field of study.

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