

Chemistry

Pesticides in different environmental compartments in Brazil: a review

Agrotóxicos em diferentes compartimentos ambientais no Brasil:
um review

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ABSTRACT

Over the years Brazil has become one of the largest agricultural producers and exporters in the world. At the same time, there was a significant increase in the use of pesticides to increase productivity at harvest. In this context, there is great concern about human health, fauna, and flora, since the inputs can go through different chemical processes and migrate through various environmental compartments. In this context, this paper addresses a review of studies that have reported the presence of pesticides in various environmental compartments such as water (surface and irrigation), soil, and food. The detected concentrations, Maximum Residue Limit (MRL), current legislation, and the values of Estimated Daily Intake (ADI) or Acceptable Daily Intake (ADI) are presented. In general, it was observed that DDTs and their metabolites were reported at concentrations beyond the MRL, followed by atrazine. Regarding intake estimates, there is a greater concern with children as they are more vulnerable, due to their low body weight when compared to adults. Finally, studies that consider the cumulative effect are needed to better assess the different chemical components in the human diet and the potential adverse effects on the health of the population.

Keywords: Brazil; Pesticide; Water; Soil; Food

RESUMO

Ao longo dos anos, o Brasil se tornou um dos maiores produtores e exportadores agrícolas do mundo. Em paralelo, houve um aumento expressivo no uso de agrotóxicos para aumento da produtividade na colheita. Nesse contexto, há grande preocupação em relação a saúde humana, fauna e flora, já que os insumos podem passar por diferentes processos químicos e migrar para vários compartimentos

ambientais. Nesse contexto, este artigo aborda uma revisão de literatura acerca de estudos que têm reportado a presença de agrotóxicos em diversos compartimentos ambientais como água (superficiais e de irrigação), solo e alimentos. As concentrações detectadas, Limites Máximos de Resíduos (LMR), legislações vigentes e os valores de Ingestão Diária Estimada (IDA) ou Ingestão Diária Aceitável (IDA) são apresentadas. De forma geral, observou-se que os DDT's e seus metabólitos foram reportados em concentrações além LMR, seguido pela atrazina. Em relação às estimativas de ingestão, há maior preocupação com as crianças por serem mais vulneráveis, em virtude do baixo peso corporal quando comparadas com adultos. Por fim, são necessários estudos que considerem o efeito acumulativo para melhor avaliar os diferentes componentes químicos na dieta humana e os potenciais efeitos adversos à saúde da população.

Palavras-Chave: Brasil; Agrotóxicos; Água; Solo; Alimentos

1 INTRODUCTION

Due to its favorable location and good climatic conditions, Brazil has one of the largest agricultural sectors in the world, with a total production of over 238 million tons per year, and is currently the second-largest exporter of agricultural products in the world. The increase in agricultural production has been achieved in two ways, both through changes in land use and an increase in the quantity and variety of inputs such as chemical fertilizers and pesticides used in different crops, to guarantee maximum productivity, in addition to protection against diseases and pests (GUARDA *et al.*, 2020; NOVOTNY *et al.*, 2020; DELLA-FLORA *et al.*, 2019).

Currently, Brazil is the world's largest consumer of pesticides, accounting for approximately 20% of total global use (FIGUEIRÊDO *et al.*, 2020; SOUZA *et al.*, 2020). In addition, Brazil is considered one of the largest importers of pesticides, being allowed the use of fifteen pesticides that are prohibited in the European Union. To have an idea, only in the year 2021 the government allow the release of more than 400 pesticides, and of these, 273 products can be readily used in crops and another 138 are active principles used in the manufacture of pesticides (G1, 2021; LAGE *et al.*, 2019).

However, the intensive use of these pesticides represents a great danger to all the existing biological diversity, in addition to risks to the health of the population, since these inputs migrate to different compartments and undergo various physical, chemical, and

biological processes that define their availability and your fate in the environment (GUARDA *et al.*, 2020; SOUZA *et al.*, 2020; SOUZA *et al.*, 2019; PETTER *et al.*, 2019).

Currently, several studies have reported contamination of water resources (BARROS *et al.*, 2019; MONTAGNER *et al.*, 2019; VIEIRA *et al.*, 2017), aquatic animals (VIERA *et al.*, 2019; MARTINS; COSTA; BIACHINI, 2020), food (PEREIRA *et al.*, 2019; PAZ *et al.*, 2016; ARAÚJO *et al.*, 2015) and soil (MESQUITA *et al.*, 2017; MENDES *et al.*, 2017; CASTANHO *et al.*, 2016), due to the incorrect use of agricultural inputs or above the maximum limits established by legislation.

In general, soil contamination is usually due to the excessive use of pesticides, which can be leached, contributing negatively to the contamination of water resources. According to Galvão *et al.* (2012), fish contamination can occur either by breeding in polluted waters or by the acquired diet, leading to the bioaccumulation of these compounds in the tissues of organisms.

According to Tang and Maggi (2021), Brazil is one of the main focus of contamination by pesticides, along with Argentina, Chile, China, Malaysia, and Japan. Such facts warn that the health of the population is exposed to great risk, due to the toxicity of pesticides, as well as the fact that some compounds present greater bioaccumulation as they advance in the food chain (GUARDA *et al.*, 2020; SANTOS *et al.*, 2020; BOTARO *et al.*, 2011). Some of the problems developed in public health resulting from exposure to pesticides are the development of reproductive disorders, endocrine problems, neurotoxicity, aneuploidy, and carcinogenicity (LAGE *et al.*, 2019; MORETTO *et al.*, 2017).

In this context, it is extremely important to develop techniques that make it possible to determine and quantify pesticides in different matrices, bearing in mind their impact on the environment and society, particularly in regions where the economy is based mainly on agriculture. These tools, in addition to helping to devise more sustainable management strategies, help to better understand the behavior and interaction of pesticides in different environmental matrices (TANG; MAGGI, 2021; SILVA *et al.*, 2019; MARCATO *et al.*, 2017).

The objective of this study was to carry out a bibliometric analysis and review of current literature on the detection of pesticides in different environmental matrices in Brazil, as well as the potential risks to human health.

2 MATERIALS AND METHODS

The articles used for the theoretical foundation of this review were found by inserting the terms "PESTICIDES" AND "BRAZIL" in the Scopus database, between title, abstract, and keyword. The research period was from 2010 to September 2021.

The results of the preliminary Scopus searches were analyzed to eliminate articles with overlapping content, categorize certain articles to the method deemed most appropriate based on their content, and most importantly, exclude articles that did not fall within the scope of this article.

From the surveys and content analysis, the selected articles were explored by the authors mainly using the "Bibliometrix" tool of the RStudio® software version 7.6, as a way of systematizing the state of the art and weaving discussions that generate new knowledge about the presence of pesticides in different Brazilian environmental compartments.

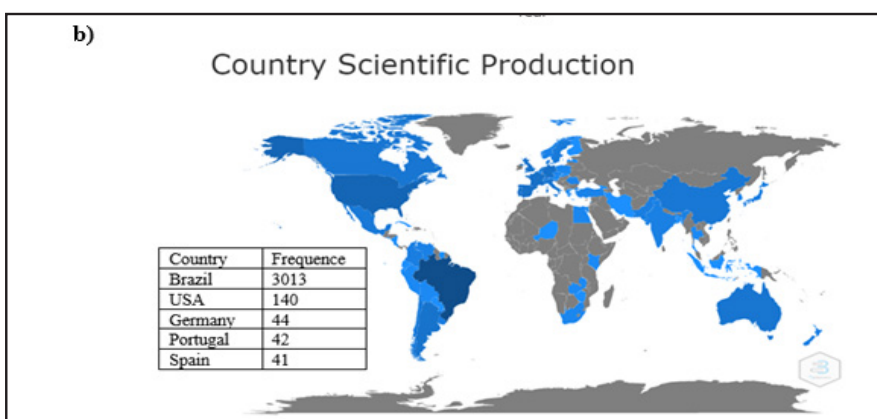
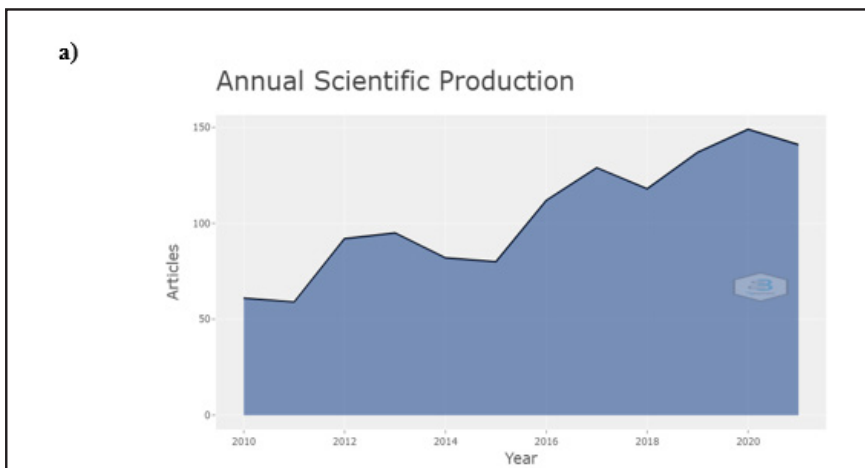
3 RESULTS AND DISCUSSION

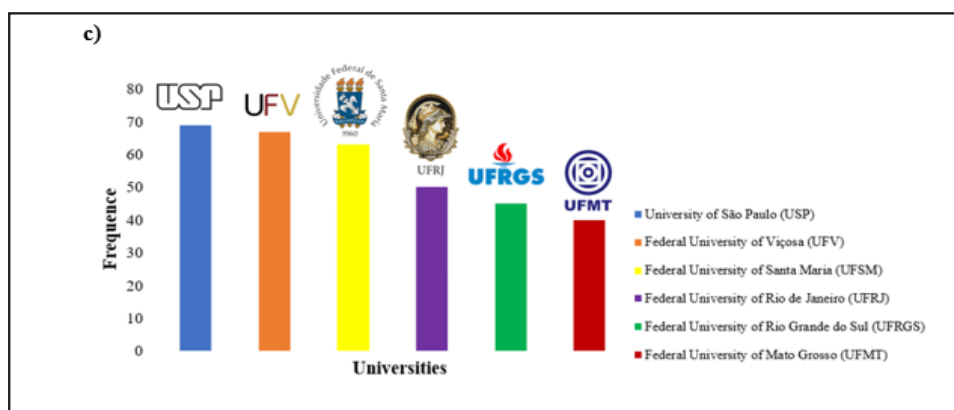
3.1 Bibliometric analysis

According to the bibliometric analysis carried out, it is noted that there is a growing interest in the topic of pesticides in Brazil, with a sharp growth from 2015 and the highest peak of publication in 2020 and an annual growth rate of 7.91% (Figure 1a). As expected, Brazil is the country that publishes the most articles on the topic, followed by the USA, Germany, Portugal, and Spain, which suggests research partnerships between these countries and Brazil (Figure 1b). Also, according to Figure 1c, the pioneer universities in publications on the topic addressed in this review are:

the University of São Paulo (USP), Federal University of Viçosa (UFV), Federal University of Santa Maria (UFSM), Federal University of Rio de Janeiro (UFRJ), Federal University of Rio Grande do Sul (UFRGS) and Federal University of Mato Grosso (UFMT), showing that the educational institutions that most study the presence of pesticides in the environment and food are concentrated mainly in southern and southeastern Brazil.

Figure 1 – a) Annual global scientific production; b) Scientific production by country; c) Universities that publish the most on the subject of this review; d) Word Cloud with the 100 most cited words in the abstracts of the analyzed articles





Source: RStudio – Bibliometrix

In the word cloud generated with the 100 most cited words in the abstracts of the articles (Figure 1d), the words circled in yellow (“environment”, “soil”, “water”, “river”, “fish”, “food” and “soybean”) indicate the main environmental compartments that pesticides were detected in Brazil and reported in studies, also, the words circled in red (“human”, “concentrations”, “residue”, “exposure”, “levels”, “ population”, “toxicity”, “health” and “risks”) suggest carrying out studies that address the risks of exposure to pesticides to the health of the Brazilian population.

In the following sections, the detection of pesticides in Brazil in different environmental compartments and food will be presented.

3.2 Detection of pesticides in environmental compartments

3.2.1 Water

Because it is an extremely important natural resource for human health, and for the survival of all living beings, water must be available for different purposes and of good quality, however, not all the population has access to drinking water, resulting in exposure to chemical contaminants present in water resources without prior treatment (GUARDA *et al.*, 2020; CARMO *et al.*, 2020).

Large water systems are often used for transportation, irrigation, recreation, fishing, and drinking water supply. However, there has been a considerable increase in the use of groundwater as an alternative source of drinking water, especially in regions where there is scarcity and degradation of surface water resources caused by human action (GONÇALVES *et al.*, 2020; SILVA; ALMEIDA; PERON, 2020; DELLA-FLORA *et al.*, 2019).

These problems related to surface and groundwater can be explained by the lack of adequate management of urban and industrial wastewater or agricultural activities, as well as the absence of stable supply and treatment infrastructure, thus affecting the quality of drinking water (CARMO *et al.*, 2020; PORTAL *et al.*, 2019).

According to Almeida *et al.* (2019), conventional water and sewage treatments usually do not eliminate certain complex substances, such as pesticides, for this reason, traces of these contaminants can reach the population. For rural families, the concern is even greater, as they can consume water without adequate treatment/quality control, coming from natural sources such as mines or wells.

Therefore, water quality monitoring is important to ensure its use in its multiple urban and rural uses. Table 1 presents the reported concentrations of pesticides detected in aqueous matrices in recent studies carried out in Brazil.

Table 1 – Detection of pesticides in aqueous matrices

(Continue)

Location of study	Amount of samples	Pesticide	Reported concentration (µg/L - mean or range)	VMP (µg/L)	Reference
Guaíba Lake (Porto Alegre/ RS)	35	Simazine	0.00906 to 0.20998	2	Perin <i>et al.</i> (2021)
		Atrazine	<0.001 to 0.232	2	
		Bifenthrin	<0.015 to 0.0487	-	
		Carbendazim	<0.001 to 0.0701	120	
		Carbofuran	0.00425 to 0.00454	7	
		Cyproconazole	<0.001 to 0.429	30	
		DDD	<0.015 to 0.0428	1	
		DDT	0.0484 to 0.25	1	
		Diuron	0.001 to 0.00418	20	
		Epoxiconazole	<0.015 to 0.0509	60	
		Imazethapyr	<0.001 to 0.578	-	
		Imidacloprid	0.031 to 0.544	-	
		Methyl thiophanate	0.0159 to 0.132	-	
		metolachlor	<0.015 to 0.138	10	
Tebuconazole	<0.001 to 0.0891	180			
Thiamethoxam	0.0184 to 0.0311	36			
Tricyclazole	0.0021 to 0.187	-			
Hydrazine maleic	<0.012 to 0.0824	-			
Hydrographic basin from Baixo Jacuí (Cachoeira do Sul/RS)	32	Atrazine	<0.02 to 0.56	2	Marins <i>et al.</i> (2021)
		Bentazone	<0.04 to 0.59	300	
		Quinclorac	0.22 to 0.5	-	
		Simazine	<0.02 to 0.051	2	
		Azinphos-ethyl	<0.02	-	
		Carbary	<0.02	0.02	
		Carbofuran	0.03 to <0.04	7	
		Fipronil	0.04 to 0.067	1.2	
		Imidacloprid	<0.02 to <0.13	-	
		Propoxur	<0.02 to 0.039	-	
		Trichlorfon	<0.02 to 1.23	-	
		Azoxystrobin	0.02 to 0.024	-	
		Dichlofluanid	0.061	-	
		Epoxiconazol	0.074	60	
Flutolanil	<0.02	-			
Metalaxyl	<0.02	-			
Propiconazol	0.074	3			
Tebuconazol	<0.04 to <0.13	180			
Tetraconazol	0.06 to 0.15	-			
Trifloxystrobin	<0.04 to 0.061	-			

Table 1 – Detection of pesticides in aqueous matrices

(Continuation)

Location of study	Amount of samples	Pesticide	Reported concentration (µg/L - mean or range)	VMP (µg/L)	Reference
Freshwater collected in the rural area of the Baía de Todos os Santos region and seawater in the city of Salvador (Salvador/BA).	7	Molinate	0.00111	6	Nascimento, Rocha and Andrade (2021)
		Atrazine	0.0174	2	
		Lambda-cyhalothrin	0.0472	-	
		Dimetoate de acaricídio	0.02 (<LOD) to 0.12	1.2	
Uruguay river (Itaqui/RS)	56	Organophosphate malathion	0.0133 (<LOD) to 0.0857	-	Gonçalves <i>et al.</i> (2020)
		Byspiribac-sodium	0.22	-	
		Imidacloprid	0.07	-	
		Quinclorac	0.59	-	
		Simazine	0.07	2	
		Malathion	0.21	0.01	
Sewage treatment plant water (Brasília/DF).	160	Propoxur	0.85, 0.26 and 0.13	-	Carvalho <i>et al.</i> (2020)
		α-BHC		-	
		p,p'-DDE		1	
		δ-BHC		-	
		Dieldrin		0.03	
		p,p'-DDT		1	
		Endosulfan I		20	
		Endosulfan sulfate	ND	-	
		Heptachlor		0.03	
Heptachlor epoxide (isomer B)		0.03			
Formoso River (Formoso do Araguaia/TO)	28	Methoxychlorine		20	Guarda <i>et al.</i> (2020)
		Clomazone	0.149 to 0.538		
		Fuazifop-p-buty	0.020		
		Flutolanil	0.020		
		Metsulfuron-Methyl	0.040	-	
		Propanil	0.020		
		Imidacloprid	0.040 to 0.065		

Table 1 – Detection of pesticides in aqueous matrices

(Continuation)

Location of study	Amount of samples	Pesticide	Reported concentration (µg/L - mean or range)	VMP (µg/L)	Reference
Rivers, shallow wells, and a hydroelectric reservoir (Santarém/PA)	58	Glyphosate	1.5 to 9.7	500	Pires <i>et al.</i> (2020)
		AMPA	0.65 to 1.9	500	
		Glufosinate	ND	-	
Rivers and streams of the Paraná hydrographic basin (Paraná/PR).	124	Glyphosate	350 to 1650	500	Mendonça <i>et al.</i> (2020)
		AMPA	550 to 750	500	
Rivers and streams (Ouro Branco/MG).	22	Acephate	ND	7	Barros <i>et al.</i> (2019)
		Difenoconazol	0.01253 to 0.09476	30	
		Phenamidone	ND	-	
		Fluazif	ND	-	
		Fluazinam	ND	-	
		Methamidophos	ND	7	
Water treatment station (Londrina/PR).	24	Atrazine	0.216 and 0.145	2	Souza <i>et al.</i> (2019)
		Azoxystrobin	0.027 and 0.023	-	
		Carbendazim	0.018 and 0.086	120	
		Imidacloprid	0.027 and 0.036	-	
		Diuron	0.024 and 0.0076	20	
		Hexazinon	0.023	-	
		Simazine	0.023	2	
		Tebuconazole	0.027	180	
Tebuthiuron	0.019	-			

Table 1 – Detection of pesticides in aqueous matrices

(Continuation)

Location of study	Amount of samples	Pesticide	Reported concentration (µg/L - mean or range)	VMP (µg/L)	Reference
São Francisco River Basin (MG)	40	4.4-DDE	0.19 to 0.49	1	Valenzuela, Menezes and Cardeal (2019)
		Propazine	0.06 to 0.43	–	
		4.4-DDD	0.08 to 2.88	1	
		Atrazine	0.96 to 10.72	2	
		Diazinone	0.13 to 0.29	–	
		Oxyfluorfen	0.44 to 16.8	–	
		δ-BHC	0.64 to 1.65	–	
		Triazophos	0.32 to 0.92	–	
		Alachlor	0.37 to 4.88	20	
		Hexaconazol	3.09 to 11.43	–	
		Lindane	0.97	2	
		Pyrimifos M.	0.10	–	
		Procymidone	2.09	–	
		Fenthion	2.54	–	
		Napropamide	1.14	–	
Dieldrin	0.54	0.03			
Kresoxim M.	2.21	–			
Carfentrazone	1.49	–			
Shallow wells in the Zumbi dos Palmares settlement (Campos dos Goytacazes and São Francisco do Itabapoana / RJ)	12	Atrazine	0.07 to 50	2	Portal <i>et al.</i> (2019)
		Carbaryl	0.361 to 50	–	
		Hexazinone	0.177 to 50	–	
		Methyl parathion	0.4 to 50	–	

Table 1 – Detection of pesticides in aqueous matrices

					(Conclusion)
Location of study	Amount of samples	Pesticide	Reported concentration (µg/L - mean or range)	VMP (µg/L)	Reference
Rivers of State of São Paulo (Campinas, Espírito Santo do Pinhal, Itatiba, Ribeirão Preto, São Paulo, Limeira, Santa Barbara D'Oeste, Rio Claro and Piracicaba / SP).	708	Ametrine	0.017	60	Montagner <i>et al.</i> (2019)
		Atrazine	0.0033 to 0.036	2	
		Azoxystrobin	0.028 to 0.001	-	
		Bromacil	0.027 to 0.0007	-	
		Carbendazim	0.158 and 0.009	120	
		Clomazone	0.010 and 0.028	-	
		Difenoconazole	0.014 and 0.002	30	
		Epoxiconazole	0.008 and 0.024	60	
		Fipronil	0.01 and 0.005	1.2	
		Flunquinconazole	0.017 and 0.012	-	
		Imidacloprid	0.013 and 0.011	-	
		Malathion	0.026 and 0.023	-	
		Picoxystrobin	0.013	-	
São Lourenço River (São Lourenço/MT)	39	Pyraclostrobin	0.003	-	Berton, Brugnera and Dores (2018)
		Simazine	0.009 and 0.01	2	
		Tebuconazole	0.039 and 0.01	180	
		Trifloxystrobin	0.004	-	
		Acetamiprid	<0.02	-	
		Carbendazim	<0.02	120	
		Carbofuran	<0.20	7	
Rivers (Nova Prata do Iguaçu, Salto do Lontra, Ampére, Santa Isabel do Oeste and Planalto/RS)	20	Diuron	<0.02	20	Vieira <i>et al.</i> (2017)
		Imidacloprid	<0.02	-	
		Malathion	<0.03	-	
		Metolachlor	<0.02	10	
		Trifluralin	<0.08	20	
		Atrazine	0.004 to 1.0	2	
		Epoxiconazole	0.040 to 0.04	60	
		Fipronil	0.0008 to 0.04	1.2	
Iprodione	0.040 to 0.07	-			
Malathion	0.004 to 0.05	60			
Penoxsulam	0.040 to 0.1	-			
Simazine	0.004 to 0.06	2			
Tebuconazole	0.040 to 0.10	180			

Source: Authors (2023)

ND= Not detected; LOQ= Quantification Limit; LOD= Detection limit; VMP= Maximum Allowed Value; (-) not stimulated or not found.

If pesticides reach water bodies, their concentration may be reduced due to processes such as sorption in sediments and chemical and/or biological degradation, in the same way, they may increase through bioaccumulation (NASCIMENTO; ROCHA; ANDRADE, 2021; GUARDA *et al.*, 2020).

The concentration of pesticides in the aqueous system is also influenced by residue mobility, water solubility, and their persistence in this environmental compartment, which help to understand the probability of a pesticide reaching water bodies and to understand the behavior of different substances in different environmental compartments (GUARDA *et al.*, 2020).

Guarda *et al.* (2020) also mention that even though pesticide residues cannot be identified, it does not mean that they are not present in the samples, but that they may be present in concentrations below the quantification limit established by the method used in the analysis.

3.2.2 Soil

Pesticides can also cause considerable stress on soil health, affecting its biota, which is responsible for maintaining its functions, and soil microfauna, such as ammonia-oxidizing bacteria, archaea, and earthworms (TANG; MAGGI, 2021; ROSA; MARQUES, 2011).

According to Arantes *et al.* (2012) and Rosa and Marques (2011), upon reaching the soil, pesticide molecules can be retained in the solid phase or remain in the soil solution, leaving the application area through volatilization, leaching, surface runoff, or "runoff", which is the absorption of pesticides by plants and removal by other organisms.

Regarding soil samples, most of the articles analyzed studied the reaction of pesticide residues in non-target animals, such as earthworms, or in a given soil exposed to a certain amount of pesticide to assess their "responses", infiltration and/or runoff content, for example. Other studies also analyzed the percentage of residues that were retained in cover crops. Thus, the studies that entered the scope of this review related to the detection of pesticides in soil are presented in Table 2.

Table 2 – Detection of pesticides in soil

Location of study	Amount of samples	Pesticide	Reported concentration (mg/kg - mean or range)	MRL (mg/kg)	Reference
Caieiras/SP	15	α -HCH	<LOQ (0.0126) to 3690	0.02	Varca <i>et al.</i> (2020)
		β -HCH	<LOQ (0.012) to 970	0.1	
		γ -HCH	<LOQ (0.0116) to 1.61	0.07	
		δ -HCH	<LOQ (0.0116) to 76.8	–	
Banks of the Formoso River/TO	28	Carbarium		–	Guarda <i>et al.</i> (2020).
		Carbofuran		0.7	
		Chlorprofam		–	
		Methomyl	ND	–	
		Propoxur		–	
		Molinate		–	
Belém/PA	11	Thiobencarb		–	Rodrigues <i>et al.</i> (2017)
		pp'-DDT	2.43 to 289.58	2	
		pp'-DDE	1.53 to 76.72	1	
		pp'-DDD	0.71 to 29.11	3	
		op'-DDT	0.76 to 37.70	2	
		op'-DDE	0.42 to 10.93	1	
op'-DDD	0.2 to 3.80	3			

Source: Authors (2023)

ND= Not detected; LOQ= Quantification Limit; LOD= Detection limit; VMP= Maximum Allowed Value; (–) not stimulated or not found.

According to Varca *et al.* (2020), several analytical methods for the determination of pesticides are used to evaluate different types of matrices, however, for the comparison of results, especially in soils, the analysis is much more complex, bearing in mind that the response of the methods is directly linked to the matrix effect. In addition, as each soil presents variations in its properties (such as pH, granulometry, texture and organic content) due to the nature of its matrix, there is a direct impact on the quantification of the analyte, being essential to carry out the characterization of the soil to understand the interactions between the matrix and the contaminant (VARCA *et al.*, 2020).

3.2.3 Food

Although pesticides are frequently used in crops to increase productivity due to the protection of crops against pests, fungi, insects, and bacteria, studies have detected the presence of chemical residues in foods, in some cases, at levels that exceed the values of the Maximum Residue Limit (MRL), established by the National Health Surveillance Agency (ANVISA), which causes great concern for food safety and consumer health (STRINGHINI *et al.*, 2021; PINHEIRO *et al.*, 2020; JARDIM *et al.*, 2018; ARAÚJO *et al.*, 2015).

This fact may be associated with the inappropriate use of residues, either due to the application technique, the use of doses above those recommended by each manufacturer, the use of restricted pesticides in Brazil, or even due to the particular chemical characteristics of the active ingredients of these pesticides (FREITAS *et al.*, 2021; STRINGHINI *et al.*, 2021; PINHEIRO *et al.*, 2020; SANTOS *et al.*, 2015).

This study shows the presence of several classes of contaminants in fruits (NAKANO *et al.*, 2016; PAZ *et al.*, 2016; KEMMERICH *et al.*, 2014), vegetables (ARAÚJO *et al.*, 2015), honey (PINHEIRO *et al.*, 2020), meats (DALLEGRAVE *et al.*, 2018), fish (MARTINS; COSTA; BIACHINI, 2020; VIERA *et al.*, 2019), eggs (PEREIRA *et al.*, 2019; DALLEGRAVE *et al.*, 2018) and bovine milk (SANTOS *et al.*, 2015; FAGNANI *et al.*, 2011).

In addition, food contamination by pesticides can occur indirectly, such as in honey, for example, which is associated with the exposure of bees to the environment contaminated with the pesticide, which can be in foliage, nectar, or pollen, and is transported by the bees to the hive, being incorporated into the honey (FREITAS *et al.*, 2021; TETTE *et al.*, 2016).

In eggs, they may come from residues present in the animal's feed or water, which after being metabolized, bioaccumulate in the egg yolk (PEREIRA *et al.*, 2019; DALLEGRAVE *et al.*, 2018). As with beef and milk, however, pesticides accumulate in muscles and fat, so, during milk removal, these residues present in adipose tissue are excreted along with the milk (SOUZA *et al.*, 2020; FAGNANI *et al.*, 2011). The presence of pesticides has also been reported in fish, which are exposed to water polluted by pesticides, and as

they are important sources of protein, this bioaccumulation can affect an entire food chain (SANTANA *et al.*, 2020; FERREIRA *et al.*, 2019; GALVAO *et al.*, 2012).

The presence of pesticide residues in organic foods is also a fact that draws a lot of attention, and that can occur due to environmental contamination, that is, the exposure of these foods to residues from neighboring areas, such as contaminated irrigation water, for example, corroborating the information reported on the topic of contamination of water resources (ARAÚJO *et al.*, 2014).

Therefore, data on monitoring pesticide residues in food are relevant to prove their occurrence in the environment, bioaccumulative effect, and possible harmful effects on human health. Table 3 presents studies that address the detection of pesticide residues in food matrices in Brazil.

Table 3 – Detection of pesticides in food

(Continue)

Location of study - Type of food	Amount of samples	Pesticide	Reported concentration (mean or range) ($\mu\text{g}/\text{kg}$) ^a or ($\mu\text{g}/\text{L}$) ^b	MRL ($\mu\text{g}/\text{kg}$) ^a or ($\mu\text{g}/\text{L}$) ^b	Reference
Santo Ângelo/ RS - Tomato	16	Chloranthraniliprole	11 ^a	300 ^a	Stringhini <i>et al.</i> (2021)
		Chlorpyrifos	17 and 22 ^a	500 ^a	
		Diflubenzuron	63 ^a	500 ^a	
		Famoxadone	23 ^a	1000 ^a	
		Imidacloprid	12 to 98 ^a	500 ^a	
		Pyraclostrobin	238 ^a	200 ^a	
		Tebuconazole	45 and 11 ^a	200 ^a	
		Thiamethoxam	11 ^a	1000 ^a	
		Azoxystrobin	10 ^a	500 ^a	
Clothianidin	10 ^a	100 ^a			
12 municipalities in the semi-arid region of Rio Grande do Norte/RN - Honey	35	Monocrotophos organophosphates	49 ^a	-	Pinheiro <i>et al.</i> (2020)
		Trichlorfon	10 to 74 ^a	-	
		Chlorpyrifos	10 ^a	20 ^a	

Table 3 – Detection of pesticides in food

(Continuation)

Location of study - Type of food	Amount of samples	Pesticide	Reported concentration (mean or range) ($\mu\text{g}/\text{kg}$) ^a or ($\mu\text{g}/\text{L}$) ^b	MRL ($\mu\text{g}/\text{kg}$) ^a or ($\mu\text{g}/\text{L}$) ^b	Reference
Lagoon of Patos/RS – Fish	18	Atrazine	0 to 11.70 ^a	–	Martins, Costa and Bianchini (2020)
		Chlorpyrifos	0 to 3.82 ^a	–	
		Dichlofluanid	0 to 10.51 ^a	–	
		Diclofenac	79.15 to 1474.25 ^a	–	
		Diuron	0 to 0.16 ^a	–	
		Methylparaben	0 to 87.85 ^a	–	
		Octocrylene	0 to 18.97 ^a	–	
		Σ Polycyclic Aromatic Hydrocarbons	833.42 to 2134.82 ^a	–	
		Trifluralin	0 to 2 ^a	–	
Rio de Janeiro/RJ – Eggs	13	Pirimiphos	4.5 ^a	–	Pereira <i>et al.</i> (2019)
		Mephosfolan	4.5 ^a	–	
		Pyraclostrobin	4.5 ^a	–	
		Spiroxamine	8.3 ^a	50 ^a	
		salinomycin	32 ^a	3 ^a	
Londrina/PR – Fish	1000	Organochlorine (ΣOCPs)	8.61 to 60.40 ^a	–	Vieira <i>et al.</i> (2019)
Rio Grande do Sul/RS – Meat	147	Chlorpyrifos	ND to 0.28 ^a	50 ^a	Dallegrave <i>et al.</i> (2018)
		Bifenthrin	0.01 to 1.04 ^a	3000 ^a	
		Cyhalothrin	0.01 to 0.04 ^a	20 ^a	
		Permethrin	ND to 0.03 ^a	50 ^a	
		Cypermethrin	0.02 to 0.51 ^a	50 ^a	
		Deltamethrin	ND to 0.025 ^a	30 ^a	
Fortaleza/CE – Guava and Graviola	8	Lindane	10.44 ^a	–	Paz <i>et al.</i> (2016)
		α -HCH	5.84 to 20.16 ^a	–	
		β HCH	6.30 ^a	–	
Rio Paranaíba/MG – Carrot	20	Linuron	9160 and 3200 ^a	1000 ^a	Araújo <i>et al.</i> (2015)
		Procymidone	5850 and 2510 ^a	1000 ^a	
		Haloxfop-methyl	480 ^a	1000 ^a	

Table 3 – Detection of pesticides in food

(Continuation)

Location of study - Type of food	Amount of samples	Pesticide	Reported concentration (mean or range) ($\mu\text{g}/\text{kg}$) ^a or ($\mu\text{g}/\text{L}$) ^b	MRL ($\mu\text{g}/\text{kg}$) ^a or ($\mu\text{g}/\text{L}$) ^b	Reference
São Paulo/SP – Orange	57	Bifenthrin	80 ^a	70 ^a	Nakano <i>et al.</i> (2016)
		Clofentezine	50 ^a	200 ^a	
		Myclobutanil	220 ^a	–	
		Azinphos-ethyl	50 ^a	–	
		Phenitrothion	60 ^a	–	
		Parathion	70 ^a	–	
		Profenophos	80 ^a	–	
Alto Vale do Itajaí/SC – Cucumber	7	Carbendazim	790 and 12.7 ^a	200 ^a	Neto and Gonçalves (2016)
		Fluopicolide	150 and 51.1 ^a	200 ^a	
		Propachlor	20.7 and 124 ^a	–	
		Propamocarb	1320 ^a	2000 ^a	
		Thiamethoxam	81.4, 21.4 and 32.9 ^a	20 ^a	
		Imidacloprid	19.4 ^a	200 ^a	
Santa Maria/RS – Fluid milk, powdered milk and cheese	113	Hexachlorobenzene	2.20, 0.25 and 1.83 ^a	10 ^a	Santos <i>et al.</i> (2015)
		α -HCH	1.85, 0.08 and 0.61 ^a	4 ^a	
		Lindane	4.60, 1.46 and 0.76 ^a	10 ^a	
		Aldrin	2.30, 0.14 and 8.68 ^a	6 ^a	
		p,p'-DDE	8.53, 0.04 and 0.56 ^a	2 ^a	
		o,p'-DDD	5.40, 0.15 and 11.49 ^a	2 ^a	
		p,p'-DDD	0.52, 0.08 and 1.23 ^a	2 ^a	
		o,p'-DDT	0.65, 0.04 and 0.97 ^a	50 ^a	
DDT	15.09, 0.30 and 14.26 ^a	50 ^a			
Espírito Santo/ES – Tomato	40	Chlorpyrifos	80 to 100 ^a	500 ^a	Santos <i>et al.</i> (2015)
		Dichlorvos	50 to 180 ^a	–	
		Methamidophos	120 ^a	–	
		Permethrin	210 to 510 ^a	300 ^a	
		Phenpropratrie	410 ^a	200 ^a	
		Carbaryl	180 to 230 ^a	100 ^a	

Table 3 – Detection of pesticides in food

(Conclusion)

Location of study - Type of food	Amount of samples	Pesticide	Reported concentration (mean or range) ($\mu\text{g}/\text{kg}$) ^a or ($\mu\text{g}/\text{L}$) ^b	MRL ($\mu\text{g}/\text{kg}$) ^a or ($\mu\text{g}/\text{L}$) ^b	Reference
Santa Maria/ RS – Pepper	20	Acetamiprid	50 ^a	700 ^a	Kemmerich <i>et al.</i> (2014)
		Azoxystrobin	10.5 to 13.6 ^a	500 ^a	
		Boscalida	45.7 ^a	500 ^a	
		Buprofezin	10.8 to 26.2 ^a	500 ^a	
		Carbendazim	14.3 to 294.4 ^a	–	
		Chlorpyrifos	10.3 to 42.8 ^a	500 ^a	
		Clothianidin	53.9 ^a	50 ^a	
		Difenoconazole	44 and 36.6 ^a	500 ^a	
		Phenpropratrie	53.6 ^a	200 ^a	
		Pyraclostrobin	10.1 to 113.5 ^a	1000 ^a	
Pernambuco/ PE – Tomato and pepper	60	Thiamethoxam	97.9 ^a	1000 ^a	Araújo <i>et al.</i> (2014)
		Acephate	12.3 ^a	200 ^a	
		Carbendazim	0 to 80 ^a	50 ^a	
		Cyromazine	25 to 30 ^a	200 ^a	
		Imidacloprid	0 to 20 ^a	30 ^a	
		Thiophanate-methyl	35 to 50 ^a	500 ^a	
		Methamidophos	20 to 20 ^a	200 ^a	
		Methomyl	0 to 90 ^a	–	
Pernambuco/ PE – Milk	30	Methomyl	20 ^a	–	Fagnani <i>et al.</i> (2011)
		Deltamethrin	60 ^a	60 ^a	
		Dithiocarbamate	60 ^a	60 ^a	
		Fenthion	400 ^a	3000 ^a	
		Fenthion	0.06 ^b	500 ^b	
		Coumaphos	0.04 ^b	500 ^b	
		Malathion	0.02 ^b	10 ^b	
Dimethoate	0.01 ^b	50 ^b			
Carbofuran	0.01 ^b	100 ^b			
Aldicarb	0.02 ^b	10 ^b			
Carbaryl	0.02 ^b	20 ^b			

Source: Authors (2023)

ND= Not detected; LOQ= Quantification Limit; NA= not analyzed; MRL= Maximum Residue Limits; (–) not stimulated or not found.

The ANVISA has been constantly reassessing the toxicology of the active ingredients of pesticides, which may lead to the restricted use of the substance or even its ban,

depending on the results. Taking this fact into account, the presence of some active ingredients reported in studies carried out by Araújo *et al.* (2014), Santos *et al.* (2015), and Nakano *et al.* (2016), identified as prohibited in Brazil for the crops studied (pepper, tomato, and orange, respectively) according to ANVISA (2012), namely Methamidophos, Methomyl, Dichlorvos, Myclobutanil, Azinphos-ethyl, Fenitrothion, Parathion, and Profenofos, due to its possible harmful effects on human health.

3.3 An overview of Brazilian legislation and established pesticide limits

According to art. 15 of Law nº 9.974, of June 6, 2000, the “production, commercialization, transport, application, and destination of waste and empty agrochemical packaging, in breach of the requirements established by the legislation, can lead to imprisonment, two to four years, plus a fine” (BRASIL, 2000).

Likewise, art. 3º of Law 7.802 of 1989, provides for pesticides, their components, and the like, which “can only be produced, exported, imported, marketed and used if previously registered with a federal agency, following with the guidelines and requirements of the agencies responsible for the health, environment and agriculture sectors.

If it fails to comply with the law, and the pesticide is not authorized by the governing legislation, it will be subject to a criminal offense of the crime provided for in Art. 334-A, which provides for imprisonment from 2 to 5 years.

In addition, PL 7.710/2017, which is still being approved by the Chamber of Deputies, provides for the expropriation of rural property that uses pesticides prohibited in Brazil.

These are laws that aim to guarantee the protection, well-being, and health of the population and the environment of a toxic, dangerous or harmful product or substance, however, the presence of prohibited agrochemicals is still observed in some studies, a fact that may be related the lack of supervision over these products, as well as the persistence of these active ingredients in the environment.

The Maximum Residue Limit (MRL) is defined as the maximum amount of residue, officially accepted in the food, expressed in parts (by weight) of the pesticide (mg/kg). In

foods of animal origin, the reference limit used is expressed in $\mu\text{g}/\text{kg}$ (in honey, eggs, fish, and meat) and $\mu\text{g}/\text{L}$ (milk), according to normative instruction n° 24, on August 9, 2011. In aqueous matrices, this limit is given as the maximum permitted value (VMP) expressed in $\mu\text{g}/\text{L}$ in potability parameters for pesticides and metabolites that pose a health risk, present in GM/MS Ordinance n° 888, of May 4, 2021. In the soil, this parameter is expressed in mg/kg of dry weight, reported in resolution n° 420, on December 28, 2009.

In Brazil, the bodies responsible for defining the MRL and VMP are the Ministry of Health, and the Ministry of Agriculture, Livestock, and Supply (MAPA), according to Decree n° 10.833/2021.

In addition, Brazilian legislation is composed of some resolutions which they regulate the presence of pesticides in the matrices addressed in this study, as shown in Table 4.

Table 4 – Some Brazilian legislation on the presence of chemical contaminants and MRLs in water, soil, and food

(Continue)

Type	Identification	Issuing body	Publication date	Subject
Ordinance	Ordinance GM/MS N° 888, 4 from May of 2021	Ministry of Health	May 7, 2021	Art. 1° provides for the procedures for controlling and monitoring the quality of water for human consumption and its potability standard; Art. 4° All water intended for human consumption is subject to water quality surveillance; Art. 5° defines in item I - water for human consumption: drinking water intended for ingestion, food preparation, and personal hygiene, regardless of its origin.

Table 4 – Some Brazilian legislation on the presence of chemical contaminants and MRLs in water, soil, and food

(Conclusion)

Type	Identification	Issuing body	Publication date	Subject
Decree	Decree N° 10.833, of October 7, 2021	Presidency of the Republic	October 7, 2021	Provides for research, experimentation, production, packaging and labeling, transport, storage, marketing, commercial advertising, use, import, export, the final destination of waste and packaging, registration, classification, control, inspection, and inspection of pesticides, their components and the like.
Normative Instruction	Normative Instruction N° 51, of December 19, 2019	Ministry of Health and ANVISA	December 26, 2019	Establishes the list of maximum residue limits (MRL), acceptable daily intake (ADI), and acute reference dose (DRfA) for active pharmaceutical ingredients (API) of veterinary drugs in food of animal origin.
Normative Instruction	Normative Instruction N° 24, of August 9, 2011.	MAPA and Agricultural Defense Department	August 09, 2011	Meat Monitoring Subprogram (Beef, Poultry, Pork and Equine), Milk, Honey, Eggs, and Fish for 2011, referring to the National Plan for the Control of Biological Residues in Products of Animal Origin – PNCRB.
Resolution	Resolution n° 420, of December 28, 2009.	Ministry of the Environment and National Council for the Environment	December 28, 2009	Provides criteria and guiding values for soil quality regarding the presence of chemical substances and establishes guidelines for the environmental management of areas contaminated by these substances as a result of anthropic activities

Source: Authors (2023)

Given the analyzed studies throughout this work, it is observed that some results do not comply with Brazilian legal norms, as reported in Table 5.

Table 5 – List of Pesticides with values above the MRL and VMP

Matrix	Pesticide	Reference
Food	Pyraclostrobin	Stringhini <i>et al.</i> (2021)
Water	Malathion	Gonçalves <i>et al.</i> (2020)
Water	Glyphosate AMPA	Mendonça <i>et al.</i> (2020)
Soil	α -HCH β -HCH γ -HCH	Varca <i>et al.</i> (2020)
Water	Atrazine Dieldrin	Valenzuela, Menezes and Cardeal (2019)
Food	Salinomycin	Pereira <i>et al.</i> (2019)
Water	Atrazine	Portal <i>et al.</i> (2019)
Soil	pp'-DDT pp'-DDE pp'-DDD op'-DDT op'-DDE op'-DDD	Rodrigues <i>et al.</i> (2017)
Food	Bifenthrin	Nakano <i>et al.</i> (2016)
Food	Carbendazim Thiamethoxam	Neto and Gonçalves (2016)
Food	Linuron Procymidone	Araújo <i>et al.</i> (2015)
Food	o,p'-DDD p,p'-DDE Aldrin	Santos <i>et al.</i> (2015)
Food	Permethrin Phenpropratrine Carbaryl	Santos <i>et al.</i> (2015)
Food	Acephate	Araújo <i>et al.</i> (2014)
Food	Clothianidin	Kemmerich <i>et al.</i> (2014)

Source: Authors (2023)

Therefore, despite the quantitative monitoring of waste by regulatory bodies, there are still chemical components above tolerable concentrations, this type of exposure can cause problems not only for humans but also for marine life and soil microbiota. (BOMBARDELLI *et al.*, 2021; PORTAL *et al.*, 2019).

3.4 Harmful health effects

The use of agrochemicals in excess or use of active ingredients prohibited due to their toxicology, as well as their application without adequate biosafety practices, can damage the health of the population, especially rural workers who frequently carry out the application and are exposed to pesticides (BOMBARDELLI *et al.*, 2021; KHAYAT *et al.*, 2013; DETÓFANO *et al.*, 2013).

Studies report higher risks of cancer mortality from prolonged exposure to pesticides, particularly in rural workers, such as esophageal cancer (MEYER *et al.*, 2011), colon cancer (MARTIN *et al.*, 2018), hepatic cancer (GUIDA *et al.*, 2021) and brain cancer (MIRANDA-FILHO *et al.*, 2014; MIRANDA-FILHO; MONTEIRO; MEYER, 2012).

To identify and assess the risks of pesticides, research is carried out to determine their potential for contamination in the different matrices (surface water, soil, air, and sediments), as well as the cumulative exposure of these compounds through oral and/or other exposure routes (JARDIM *et al.*, 2018).

Risk assessment is an estimate of human exposure to xenobiotic compounds through food consumption and provides a link between possible hazards in the food chain and the reflected risks to human health (ISHIKAWA *et al.*, 2016). It is possible to indirectly estimate the degree of exposure based on consumption data of contaminated food and the average occurrence of the contaminant. In this estimate, the degree of exposure is measured in terms of estimated daily intake (EDI) per unit of body weight and is usually expressed in ng per kg of body weight (BW) day (JAGER *et al.*, 2013).

In this sense, Dallegrave *et al.* (2018) estimated the EDI of residues of chlorpyrifos, cis-bifenthrin, cyhalothrin, permethrin, cypermethrin, and deltamethrin in food

samples of animal origin, using the average weight of 68.6 kg for a Brazilian adult, and reported maximum EDIs of 5×10^{-2} , 2×10^{-3} , 4×10^{-4} , 9×10^{-5} , 2×10^{-2} e 4×10^{-4} $\mu\text{g}/\text{kg bw}/\text{day}$, for the mentioned pesticides, respectively.

Nakano *et al.* (2016), reported on the Acceptable Daily Intake (ADI) of pesticide residues (Bifenthrin, Clofentezine, Myclobutanil, Azinphos-ethyl, Fenitrothion, Parathion, Profenofos) in oranges in the adult (60 kg) and infant (15 kg) populations, respectively, which varied from 0.04 to 6.62% for adults, and from 0.14 to 26.5% for children.

Santos *et al.* (2015), considered the age groups of the study: children (2.5 to 6 years old), adolescents (10 to 19 years old), adults (20 to 64 years old), and the elderly (over 65 years old), it was observed that about the EDI values, in dairy products, that Aldrin was the only organochlorine pesticide (OCPs) that exceeded the ADI value (0,1 ng/kg bw/day), for children and was on the edge for the elderly (0.716 and 0.100 ng/kg bw / day, respectively), this result was not observed in any other OCPs (HCB, α -HCH, lindane, aldrin, p,p'-DDE, p,p'-DDD, and o,p'-DDT) analyzed from the individual way. Furthermore, the authors observed that the EDI for all the total OC compounds studied was higher for children, compared to adolescents, adults, and the elderly, where the estimated intake by the sum of all compounds were 8.266; 0.393; 0.423; and 0.614 ng/kg/day, respectively (SANTOS *et al.*, 2015).

Pereira *et al.* (2019) esteemed the hazard indices (HIs) of chemical residues found in egg samples, for adults and children up to 27 kg, the results were 0.42 and 1.01 mg/kg bw/day, respectively, indicating that the cumulative exposure mixtures of mephosfolan, pirimiphos, pyraclostrobin, salinomycin, and spiroxamine residues pose a potential health risk to children weighing up to 27 kg.

Ferreira *et al.* (2019) performed the human risk assessment using the EDI calculations and the ADI percentage of the 21 organochlorine pesticides (OCPs). Using the average Brazilian consumption of 11.17 kg of fish/year to calculate the EDI, and considering the average weight for the Brazilian population of 60 kg for adults and 30 kg for children. The authors concluded that none of the samples analyzed

exceeded the safety limits, both for adults and children, but highlighted that children may be more vulnerable to the safety limit, bearing in mind their low body weight when compared to adults. Furthermore, they report that to reach the suggested risk limit for methoxychlor (ADI: 0.62%), which was the contaminant found in the highest concentration in this study in fish, it would be necessary to consume about 10 and 5 kg of sardines per day for adults and children respectively (FERREIRA *et al.*, 2019).

Galvão *et al.* (2012) found that the consumption of bivalves did not represent risks to human health concerning the 26 organochlorine pesticides (OCPs) and 18 polychlorinated biphenyls (PCBs) studied, being estimated the ADI levels with 12 mussels or 12 individuals of scallops, for a child of 30 kg of body weight. The authors found that to reach minimum levels of risk, it would be necessary to consume 797 mussels with the presence of p,p' DDT, and within the OCPs studied, the sum of DDT for a meal of 12 animals represented the main risks to health, with 3% of the ADI for children (GALVÃO *et al.*, 2012).

Valcke *et al.* (2017) point out that the risk of possible cancer occurrence is lower due to the consumption of fruits and vegetables possibly contaminated with residues of active ingredients of pesticides, when compared to their non-consumption, bearing in mind the benefits that daily, abundant and diversified consumption can bring the health. However, they do not consider the estimated risks to be negligible, even with uncertainties regarding the risks of exposure.

In this context, Ferreira *et al.* (2019) emphasize the need for studies that consider the cumulative effect to better assess the different chemical components in the human diet and the potential adverse effects on the health of the population.

4 CONCLUSIONS

This work addressed the detection of pesticides in several Brazilian environmental compartments, including water, food, and soil. Taking into account the reported concentrations and current Brazilian legislation, DDTs and their metabolites were

reported in concentrations beyond the MRL, followed by atrazine.

Regarding perspectives, it is noteworthy that a greater focus on research for the development and improvement of techniques for the detection/quantification of pesticides in different matrices is extremely important, to enable effective monitoring of the presence of these contaminants potentially harmful to the human health, as well as, estimating the daily intake doses by the population.

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How to quote this article

OLIVEIRA, M. P. de; SILVA, A. B. S.; RIGUETO, C. V. T.; LOSS, R. A.; GUEDES, S. F.; GERALDI, C. A. Q. Pesticides in different environmental compartments in Brazil: A review. **Ciência e Natura**, Santa Maria, v. 45, e2, 2023. DOI: <https://10.5902/2179460X70715>.

