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Pesticide residues in food of plant origin commercialized in Brazil from 2010 to 2020 – An update from the two national monitoring programs

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ABSTRACT

The objective of this study was to investigate the results of the two Brazilian national pesticide residue monitoring programs obtained from January 2010 to December 2020. A total of 35,321samples of 44 different food crops were analyzed, of which 55.3% tested positive for at least one compound, with pear, peach, strawberry and sweet pepper having over 90% of the analyzed samples containing residues. Approximately one-third of the positive samples had at least one irregularity, of which 86.7% due to the presence of non-authorized pesticides for the crop, 26.3% exceeding the maximum residue level, and 13.1% showing both irregularities. A total of 191 different compounds were detected, primarily organophosphorus (OP) (37.4% of positive samples, of which over 60% of cereal/flour, potatoes, and peanuts). Chlorpyrifos, acephate, pirimiphos-methyl, and methamidophos were the main OPs detected. Triazoles were present in 27.2% of the positive samples, mainly rice, and pyrethroids in 22.4% of the positive samples, mainly in popcorn. Dithiocarbamates were present in 19.7% of the positive samples, predominantly in apples, and 5.0% of the positive samples contained N-methyl carbamates, mainly in sweet peppers. Carbendazim was the most detected pesticide (30% of positive samples), mainly in papaya (18.2% of samples containing this pesticide). About 60% of positive samples contained multiple residues, primarily in sweet pepper, pear, strawberry, and orange (over 80% of positive samples). Compared to the previous decade (2001–2010), these results indicated increased percentages of positive, irregular and of samples containing multiple residues. Dithiocarbamates were no longer the most detected pesticide group, while carbendazim remained the most detected pesticide in both periods.

1. Introduction

Brazil is one of the world's major food producers and exporters, with agricultural land covering approximately 350 million hectares in 2017, primarily dedicated to soybeans, sugarcane, maize, and coffee (IBGE, 2024). Pesticides are the most widely used pest management strategy to ensure food supplies globally, and in 2020, Brazil was the largest consumer of pesticides in the world (685,746 tons of active ingredients; 10.5 kg/ha), followed by the United States of America (457,385 tons; 2.3 kg/ha), a trend that continued in 2021 (FAO, 2024).

Pesticides are developed to be toxic to pests, such as insects and fungi; however, they can also impact human health and the environment if not properly used and regulated. Brazilian pesticide regulation was recently revised (Law 14,785/2023) and includes aspects regarding research, production, importation/exportation, registration, storage, transport, and disposal (Brazil, 2023). The registration process involves the Ministry of Agriculture and Livestock (MAPA), the Institute of Environment and Natural Resources (IBAMA), and the Ministry of Health, through the National Sanitary Surveillance Agency (ANVISA). ANVISA is responsible for evaluating the impact of pesticide use on human health and establishing maximum residue levels (MRLs) based on supervised pesticide residue trials mainly conducted in the country following approved product label's good agricultural practices (GAP). As of August 2023, 289 active ingredients had MRLs established for various food commodities in the country (ANVISA, 2023a).

The presence of pesticide residues in food can be a health concern and has been extensively investigated by researchers worldwide (Gallani et al., 2020; Ngabirano, H., & Birungi, 2022; Mozzaquatro et al., 2019; Perumal et al., 2023; Jia et al., 2024) and government agencies, which conduct national or regional monitoring programs (Pappas and Foss, 2022; EFSA 2021, 2022; DAFF, 2024; USFDA, 2021, 2022. 2023, 2024). In addition to verifying compliance with national/regional MRLs, some countries use these monitoring data for conducting dietary risk assessments after chronic and acute exposure (Nougadère et al., 2014; Craig

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et al., 2020). In the United States, dietary risk assessment is conducted by the Environment Protection Agency using data collected from the Department of Agriculture (USEPA, 2011). Currently, Brazil has two programs for monitoring pesticide residues in food: the Program on Pesticide Residue Analysis in Food (PARA), implemented by ANVISA in 2001, and the National Residue and Contaminant Control Program (PNCRC), implemented for food of plant origin in 2006 by MAPA. While the objective of PARA is to monitor the irregular use of pesticides in the country and assess risks to human health (ANVISA, 2023b), legal actions may be taken by MAPA authorities when irregularities are found in the samples analyzed within the PNCRC (MAPA, 2023).

During the period from 2001 to June 2010, a total of 13,556 food samples of 22 fruits, vegetables, and beans were analyzed by PARA and PNCRC, with 48.3% of the samples containing at least one residue (Jardim & Caldas, 2012). The objective of the present work is to update these data, compiling and discussing the results obtained from January 2010 until December 2020.

2. Materials and methods

2.1. Monitoring programs

Food samples within the PARA program were collected in retail stores and supermarkets by the state sanitary surveillance agencies of the 26 Brazilian states and the Federal District (ANVISA, 2023). Five laboratories analyzed the collected samples, of which four are state laboratories. They were inspected and authorized by ANVISA to ensure compliance with ISO/IEC 17025 requirements; two laboratories are accredited according to the ISO 17025 by the competent Brazilian agency (INMETRO). The number of active ingredients investigated during the period varied between years and crops, reaching 270 active ingredients in 2017/2018. Table S (Supplementary Material) listed the 291 compounds analyzed by the PARA during the period of January 2010 to December 2019.

In the PNCRC program, samples were collected in wholesalers and supply centers in all Brazilian states and the Federal District (local production and imported). The analyses were performed by MAPA official laboratories or by private laboratories audited by MAPA, all accredited by INMETRO (MAPA, 2023). The data provided covers the period of July 2010 to December 2020. The number of investigated active ingredients also varied between years and crops, with approximately 300 compounds per year. A list of the analyzed compounds was not provided.

In both programs, dithiocarbamates were determined as CS_2 either by spectrophotometry (limit of quantification, LOQ, of 0.08–0.4 mg/kg) or by gas chromatography (GC-FPD or GC-MS; LOQ of 0.05 and 0.3 mg/kg). Multiresidue methods, including QuEChERS, were used by most laboratories, using GC-FPD, GC-ECD, CG-NPD, GC-MS/MS, or LC-MS/MS, but specific methods were used for some pesticides, including glyphosate and its main metabolite aminomethylphosphonic acid (AMPA) and 2.4 D. Limit of detection (LOD) and LOQ vary among laboratories, matrix, and compound, with a LOQ of 0.01 mg/kg in most cases.

3. Results

A total of 35,321 samples of 44 different food crops were analyzed by the Brazilian monitoring pesticide residue programs from 2010 to 2020. The PARA program (January 2010 to December 2019) analyzed 27,141 food samples of 31 different crops/food products for domestic consumption (local production and imported), ranging from 10 crops in 2011 to 25 crops in 2018; no samples were collected in 2016 and 2020. The PNCRC program analyzed 8178 food samples collected from July 2010 to December 2020.

Fig. 1 illustrates the number of samples analyzed by the programs each year, with the highest numbers occurring from 2013 to 2015, and

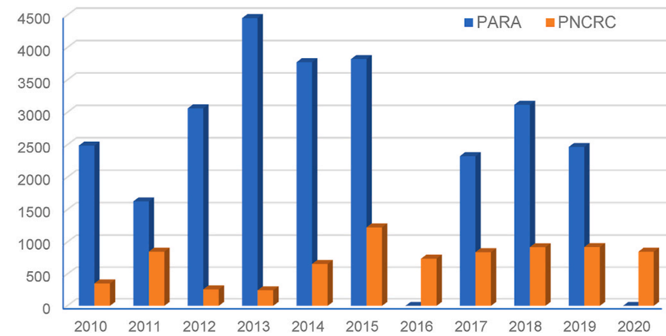

Fig. 1. Number of samples analyzed by the Brazilian pesticide residue programs from January 2010 to December 2020. PARA: Program on Pesticide Residue Analysis in Food; PNCRC: National Residue and Contaminant Control Program.

2018 with over 4000 samples analyzed annually. The PARA program analyzed most of the samples during the period, ranging from 1628 samples in 2011 up to 4455 in 2013, except in 2016 and 2020 when no samples were collected.

Table 1 presents the crops and the number of samples analyzed within the programs, along with the percentages of positive samples (≥LOD) for at least one compound and of irregularities (violations) due to the presence of non-authorized (NA) pesticide for the crop in which it was found or residues above the Brazilian maximum residue level (MRL) at the time of the analysis and both. For imported crops, the Programs also takes into consideration the Codex MRL for a given crop. The NA category includes also 47 domestic samples containing 14 pesticides not registered in the country, including trichlorfon (12 samples), which registration was canceled in August 2010, and chlorpyriphos methyl (49 samples), which was never registered in the country.

Nine crops were analyzed only by the PARA program, including minor crops such as collard green and chayote, and 11 only by the PNCRC program. Apple, rice, papaya, tomato and orange were the most analyzed crops, representing 30.6% of the total samples analyzed over the period (Table 1).

Most samples (55.3%) tested positive for at least one compound (residues ≥ LOD; Table 1), with 63% within the PARA program. Pear, peach, strawberry and sweet pepper had the highest rates of positive samples (over 90% of the analyzed samples). About 34% of the positive samples (18.9% of all samples) had at least one irregularity, mainly due to the presence of NA compounds (86.7% of irregularities).

Fig. 2 displays the % of positive samples and of irregularities according to the year of sample collection. The highest percentages of positive samples occurred in 2011 (74.9%), while the highest irregularities with NA compounds were found in 2012 (45.3% of positive samples), with the lowest values in 2016 and 2020 (about 14%). In 2012, all positive sweet pepper samples contained at least one NA compound.

A total of 191 different compounds (including metabolites) were detected in the samples analyzed by the Brazilian programs, with 176 (92%) associated with at least one irregular situation. Table 2 illustrates the percentage of positive samples for each crop according to the main pesticide classes.

Organophosphorus (OP) compounds were the most detected pesticides (37.4% of positive samples), mainly present in cereal/flour samples (corn, oat, and wheat), potatoes, and peanuts (Table 2). Chlorpyrifos, acephate, pirimiphos-methyl, and methamidophos were the main OPs detected. Triazoles (TR) were present in 27.2% of positive samples, with rice and coffee containing the highest percentages (> 50%). Pyrethroids (PY) were present in 22.4% of positive samples, mainly in popcorn, wheat flour, wheat grain, corn grain, sweet pepper, tomato, and collard greens (40-60%). Dithiocarbamates (DT) were present in 19.7% of samples, mainly in apple (57.1%), beet, and papaya

Table 1
Crops analyzed by both Brazilian Monitoring programs from January 2010 to December 2020.

Crop	Samples analyzed, n	Positive samples, %	Irregular samples			
			Total, %[b]	NA, %[c]	>MRL, %[d]	Both, %[e]
Apple	2409	89.0	7.9	58.8	48.2	7.1
Rice	2256	31.8	13.5	70.1	29.9	–
Papaya	2168	83.6	14.1	72.5	35.3	7.8
Tomato	1999	82.9	36.1	88.0	31.7	19.7
Orange	1978	72.1	13.6	90.2	12.4	2.6
Grape	1931	78.9	25.3	68.8	49.9	18.7
Bean	1922	60.1	19.2	53.2	51.8	5.0
Carrot	1544	65.7	53.8	99.8	0.7	0.5
Lettuce	1362	49.8	73.6	94.6	37.9	32.5
Cucumber[f]	1319	53.8	65.6	91.0	13.8	4.7
Sweet pepper	1209	95.0	84.8	98.0	17.2	15.3
Potato	1184	22.4	16.6	75.0	31.8	6.8
Pineapple	1160	56.8	39.6	72.4	39.8	12.3
Mango	1024	36.6	18.9	78.9	23.9	2.8
Onion	1016	11.7	41.2	91.8	8.2	–
Banana	970	26.7	17.4	46.7	53.3	–
Corn flour[f]	937	46.1	4.4	73.7	36.8	–
Beet	847	32.6	63.4	90.3	13.1	3.4
Guava	719	37.3	82.8	91.9	16.2	8.1
Strawberry	652	92.6	63.9	86.8	36.8	23.6
Zucchini[f]	650	30.0	88.2	93.0	59.3	–
Cabbage[f]	618	18.1	75.0	97.6	9.5	7.1
Wheat	581	73.8	14.9	54.7	51.6	6.3
Wheat flour	574	47.9	12.4	91.2	8.8	–
Collard green[f]	547	43.1	77.1	87.9	31.9	19.8
Soybean	507	20.5	17.3	50.0	50.0	–
Garlic	502	8.8	40.9	94.4	5.6	–
Cassava flour[f]	470	3.0	85.7	100	–	–
Pear	383	97.1	29.3	100	0.9	0.9
Sweet potato[f]	315	8.9	100	100	–	–
Chayote[f]	288	12.5	66.7	100	–	–
Oat[f]	277	61.0	97.6	100	–	–
Corn, ear	156	7.1	9.1	100	–	–
Coffee[g]	153	13.7	0	–	–	–
Corn, grain[g]	102	32.5	32.5	57.1	64.3	35.7
Peanut[g]	84	9.5	75.0	100	–	–
Lemon[g]	83	67.5	7.1	100	–	–
Melon[g]	83	51.8	23.3	50.0	50.0	–
Kiwi[g]	82	40.2	45.5	100	6.7	6.7
Corn, hominy[g]	80	13.8	0	–	–	–
Soybean bran[g]	68	26.5	5.6	100	–	–
Popcorn[g]	55	21.8	25.0	33.3	66.7	–
Peach[g]	26	96.2	80.0	85.0	25.0	10.0
Black pepper[g]	1	100	–	100	–	–
TOTAL	35,321	55.3	34.1[b]/ 18.9[h]	86.7	26.3	13.1

[a]Presence of at least one pesticide residue at levels ≥ LOD.
[b] Related to positive samples.
[c] Related to total irregular samples; NA = non-authorized pesticide.
[d] Related to total irregular samples; MRL = maximum residue level.
[e] Related to total irregular samples; both NA and > MRL.
[f] Only PARA program.
[g] Only PNCRC program.
[h] Related to total samples analyzed.

(38-39%; Table 2). Carbendazim was the most detected single pesticide (30% of positive samples and 12% of all residues; Fig. 3), mainly found in papaya, apple, and beans, followed by tebuconazole (14% of positive samples and 5.4% of all residues).

About 60% of positive samples had multiple residues (60.5%), with 38% containing 2 or 3 residues (Fig. 4A). Sweet pepper, peach, pear, strawberry, and orange had the highest percentages of samples with multiple residues (at least 80% of the positive samples (Fig. 4B). One grape sample contained 20 different residues, including triazole

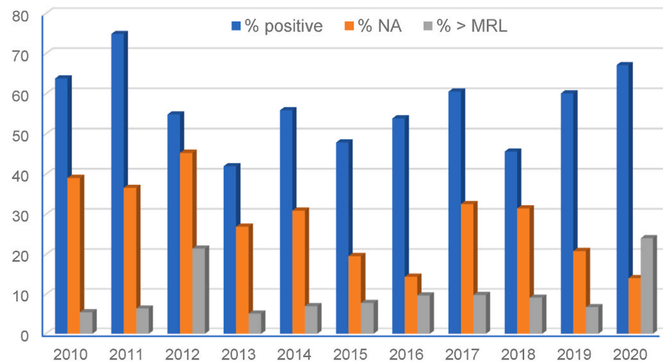

Fig. 2. Percentage of positive samples, non-authorized pesticide (NA) and at levels higher than the maximum residue level (>MRL), related to the positive samples in each year.

Table 2
Percentage of positive samples (≥LOQ) according to the chemical pesticide classe: organophosphorus (OP), triazoles (TR), pyrethroids (PY), dithiocarbamates (DT) and N-methyl carbamates (CB).

Crop	OP	TR	PY	DT	CB
Oat	97.6	–	15.4	–	–
Corn flour	97.2	–	0.3	–	0.6
Wheat flour	88.0	–	43.3	–	0.4
Corn - hominy	81.8	–	18.2	–	–
Potato	80.4	0.4	7.9	–	–
Wheat	80.4	2.1	41.7	–	0.5
Corn	79.1	–	44.2	–	–
Peanut	75.0	12.5	12.5	–	–
Corn - popcorn	66.7	–	58.3	–	–
Cassava flour	64.3	–	21.4	–	14.3
Sweet pepper	57.8	38.2	45.7	32.7	28.0
Cabbage	53.6	0.9	4.5	–	1.8
Chayote	52.8	33.3	–	–	5.6
Orange	49.1	27.0	35.6	3.0	14.8
Apple	49.0	6.0	10.3	57.1	0.8
Lemon	48.2	19.6	23.2	1.8	1.8
Guava	48.1	32.8	21.6	9.3	5.2
Peach	48.0	44.0	16.0	–	–
Carrot	47.7	36.7	1.7	2.0	5.5
Tomato	43.7	22.5	49.1	20.9	4.2
Cucumber	43.6	3.2	15.1	16.6	2.0
Beet	39.1	28.3	3.3	39.5	0.4
Onion	36.1	0.8	2.5	13.4	2.5
Zucchini	30.3	4.1	1.5	–	11.8
Strawberry	30.0	35.9	39.1	8.6	3.1
Kiwi	27.3	12.1	21.2	–	–
Rice	26.5	59.8	9.8	0.3	2.5
Collard green	21.6	16.5	41.1	–	5.5
Bean	15.8	27.2	2.9	–	1.7
Lettuce	14.9	28.1	22.4	22.2	6.1
Melon	14.0	9.3	37.2	–	–
Pear	11.6	19.4	10.5	26.1	0.5
Grape	10.6	53.3	15.4	10.8	2.7
Corn - ear	9.1	–	27.3	–	–
Pineapple	9.0	17.1	21.7	2.6	2.3
Soybean	8.7	16.3	2.9	–	–
Mango	8.5	9.9	5.1	17.6	1.1
Papaya	7.7	45.8	21.9	38.5	0.6
Garlic	6.8	20.5	4.5	6.8	2.3
Soybean bran	5.6	–	5.6	–	5.6
Sweet potato	3.6	17.9	3.6	14.3	3.6
Banana	3.1	24.7	8.5	16.2	2.3
Coffee	–	52.4	–	–	–
Total	**37.4**	**27.2**	**22.4**	**19.7**	**5.0**

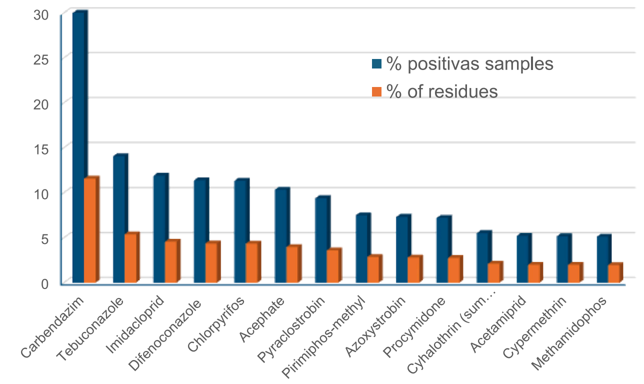

Fig. 3. The 14 pesticides most frequently detected in the samples analyzed by the Brazilian monitoring programs from January 2010 to December 2020.

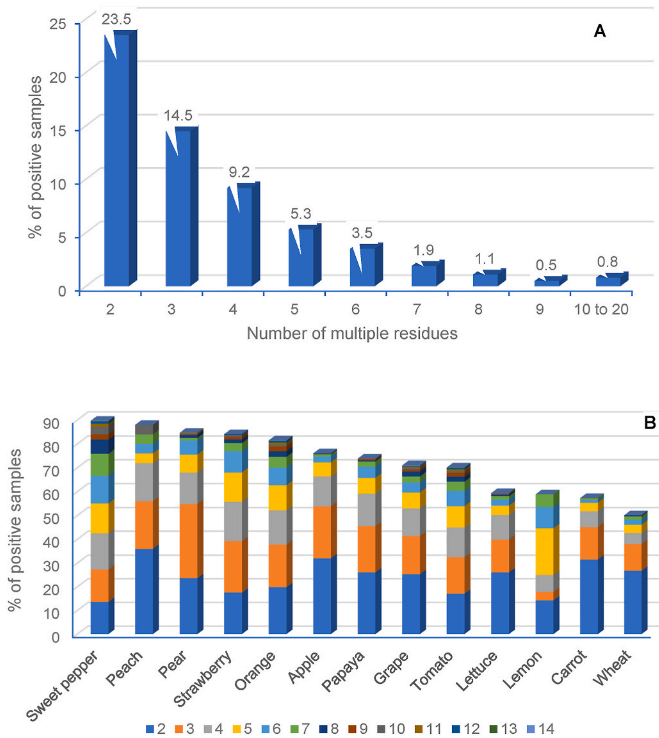

Fig. 4. (A) Percent of positive samples with multiple pesticide residues; (B) Crops containing multiple residues in at least 50% of positive samples. Four sweet pepper samples contained 15 to 19 residues and one grape sample contained 20 residues.

(cyproconazole, difenconazole, tebuconazole and tetraconazole), and strobilurin (azoxystrobin, pyraclostrobin and trifloxystrobin) fungicides and pyrethroid (lambda cyalothrin) and organophosphorus (ethephon) insecticides. Corn hominy and peanut were the only crops that did not contain multiple residues.

4. Discussion

Between 2001 and 2020, a total of 48,877 samples were analyzed by the PARA and/or PNCRC programs in Brazil. Of these, 13,556 samples were analyzed during the first decade (2001 to June 2010; Jardim and Caldas, 2012), while 35,321 samples were considered in the present study (July 2010 to December 2020). This substantial increase in sample size over time reflects the consolidation of both programs. The PARA was primarily responsible for analyzing the majority of samples in all years, except for 2016 and 2020 when the program was not active.

In the second decade, the percentage of positive samples increased by approximately 14% compared to the previous decade (48.3%). Similarly, the percentage of irregular samples increased by about 43% (from 13.2% to 18.9% of all samples). The primary reason for irregularities in both decades was the presence of NA compounds, accounting for 72.2% and 86.7% of irregularities in the first and second decades, respectively. Moreover, the percentage of samples with residues above the MRL and with both types of irregularities also increased in the second decade (20.6–26.3% and from 7.4% to 13.1% of the irregularities, respectively).

Several factors may have contributed to the higher irregularity rate observed in the second decade, including changes in MRLs, increased number of investigated pesticides per crop, lower LOQ of the analytical methods, and a higher number of crops analyzed (from 22 to 44). Additionally, some crops included in the programs after 2010 showed high rates of irregular samples, such as guava, cassava flour, zucchini, peach, sweet potato, and oat, with at least 80% of positive samples displaying irregularities.

The implementation of legislation in 2014 aimed at supporting the registration of pesticides for minor crops in Brazil (Brazil, 2014) was expected to reduce the occurrence of non-authorized pesticide use in certain crops. While some improvements may have been observed in subsequent years, the overall impact of this legislation on reducing irregularities is not yet clear, and may become more evident in the future. For instance, the presence of captan in sweet potato samples would not be considered a violation in 2023, as a MRL of 1 mg/kg was set based on the provisions of the minor crop legislation (ANVISA, 2023c).

The results found in this study showed the percentage of positive samples within the range found by the United States Food and Drug Administration (USFDA) monitoring program, but with higher violation rate. Data from 2019 showed 52.6% of positive samples, of which 15.3% with at least one violation (8% of all samples) (USFDA 2021). In 2021, due to impacts of the COVID-19 pandemic, most of the samples were imported (1067 samples), and 300 domestic samples (USFDA, 2023). In this year, about 55% of the imported samples were positive, and 10.7% were not in compliance with the legislation, while these numbers were 65 and 3.3%, respectively, for domestic samples. Like in the present work, pesticide with no set tolerance (NA compound) was the main reason for violation in USA. Liang et al. (2021) evaluate the results for over 56,000 food samples analyzed under the USFDA monitoring program between 2009 and 2017. The number of residues detected increased over time, as well as the violation rate, being 3–5 times higher for import samples, which appears to be due to targeted sampling of foods with a history of high violation rates.

Data from 32 European countries from 2020 showed that 31.5% of the 12,077 samples analyzed (domestic and imported) had quantified residues (≥LOQ), with 5.5% of positive samples exceeding the MRL (EFSA, 2022). The overall MRL exceedance rate rose from 1.7% of all samples in 2017 to 2.1% in 2020, much lower than what was found in the Brazilian data for the period of 2001–2020. Violation rate in Australia from 2013 to 2023 was also very low, ranging from 0.3 to 4% for grains (8792 samples) and from 0 to 5% for pears and apples (3363 samples) (DAFF, 2024).

The percent of positive samples containing DT (19.7%) dropped considerably compared to the first decade, when it was the main pesticide group with detected residues, present in 41.6% of the positive samples (Jardim and Caldas, 2012). This result is interesting considering that commercialization of mancozeb, the main DT used in Brazil, has increased considerably in the last decade (7000 tons in 2010 to 50,000 in 2020), being the third pesticide most commercialized in the country from 2016 to 2021 (IBAMA, 2014). On the other hand, samples containing TR, PY and CB increased significantly, at a rate of 2–2.5 times higher compared with the first decade (% of positive samples), while the rate for OP increased by about 16% (Jardim and Caldas, 2012).

Carbendazim was the single compound most detected in the samples, similar to the previous decade results (Jardim and Caldas, 2012), although during the period of the present study, registration was granted only for apple and citrus. Carbendazim is a metabolite of thiophanate-methyl, which residue definition includes carbendazim, with the total residue expressed as carbendazim (ANVISA, 2023a). The residue data from both monitoring programs showed 75.6 % of the results as carbendazim only and 24.6% includes thiophanate methyl and/or benomyl, which also metabolizes to carbendazim, but had its registration canceled in Brazil in 2005. About 15% of the carbendazim positive samples were irregular, of which 76.8% was due to NA compound, a value that may be overestimated since some samples may contain residues from the legal use of thiophanate methyl. Due to its reproductive toxicity and effects on embryo-fetal development, carbendazim registration was canceled for all crops in 2022 (ANVISA, 2022).

The pattern of irregularities during the period of the study, with most samples having NA compound, changed in 2020, when samples with residues above the MRL was higher (24.1 vs 14.1% of positive samples). This was mainly due to bean samples, for which 90.8% of irregularities were due to residues above the MRL, mainly of glyphosate.

Glyphosate is the most widely used pesticide worldwide (Finger et al., 2023), and the amount of active ingredient commercialized in Brazil almost doubled in the last decade (134,117 tons in 2010 to 266, 088 tons in 2022; IBAMA, 2024). The number of samples containing this herbicide (may include the AMPA metabolite) was low (256), representing 0.7 % of all samples (1.4% of positive samples), mainly in bean (41.4%), for which glyphosate has a MRL of 0.05 mg/kg. Glyphosate analysis was only introduced in the PARA in 2017, what probably explain this low rate. European data from 2018 to 2021 showed a higher rate, but less than 3% of samples containing glyphosate residues (EFSA, 2021; Carrasco Cabrera et al., 2023). Low residue levels of glyphosate, if any, are expected in treated crop as the product is not applied directly to the plant, unless they are genetically modified (GM). Indeed, over 90% of corn and soybean grown in USA are GM (USFDA, 2024) and monitoring data showed that 23% of corn and 98.1% of soybean samples analyzed in 2022 contained glyphosate (USDA, 2024).

ANVISA recently published the PARA results related to 1772 samples of 13 food crop/products collected in 2022 and analyzed for the presence of 311 pesticides (ANVISA, 2023). About 60% of the samples contained at least one residue, and 25% were irregular, of which 88.6% due to the presence of NA compound, similar to the results found in the present study. In addition to verify the compliance with the legislation, the monitoring data from PARA is used by ANVISA for conducting deterministic dietary risk assessment based on the FAO/WHO JMPR (Joint Meeting of Pesticide Residues) approach, as described in the RCD 295/19 (Brazil, 2019). The last chronic dietary assessment was performed using residue data from 2013 to 2022 (except 2016, 2020 and 2021, no samples analyzed) for 342 pesticides in 21,735 samples of 36 foods (ANVISA, 2023). The total exposure to each pesticide was lower than the respective acceptable daily intake (ADI), indicating no potential health risk for the population aged 10 years and over, for which Brazilian consumption data are available. For acute exposure, the dietary risk was calculated for each crop and compounds with an established Acute Reference Dose (ARfD). The intake exceeded the ARfD only for 0.17 (2022) to 1.1% (2013/2015) of the crop/compound combination (ANVISA, 2019; 2023).

Multiple pesticide residues in a single sample may be a result of application of different types of pesticides (e.g. herbicides, fungicides or insecticides) or of the same type (e.g. different fungicides), which maybe an option to avoid the development of resistant pests or diseases, soil uptake from previous treatments, spray drift from adjacent fields (EFSA, 2022), and/or bad agricultural practices. Most of the positive samples analyzed in the Brazilian monitoring programs had at least two different residues, an increase of about 27% compared to the first decade of the programs (Jardim and Caldas, 2012), reaching 20 residues of different types and chemical classes in a grape sample. In the first decade, grape was also the crop with the highest number of residues within one sample, although with lower number (up to 10; Jardim and Caldas, 2012). European data from 2018 to 2020 showed the presence of multiple residues in 27–29% of all samples, or about 60% of positive samples (EFSA 2021; 2022), similar to what was found in Brazil from 2010 to 2020. High number of residues were also found in a single sample, such as 28 residues in a dried vine fruit sample, and 35 residues in a strawberry sample.

Although many assessments are conducted for each chemical, in real life, humans are co-exposed to various chemicals in the diet, including multiple pesticides. Consider the presence of multiple pesticide residues in a single crop or in the diet in the risk assessment is to estimate the cumulative exposure. Different approaches are used to perform cumulative exposure assessment, such as to cumulate all pesticides in the diet, independent of the class or toxicological profile (Jensen et al., 2022), cumulate compounds that have a common effect on target organ/system (Craig et al., 2020), or only compounds with the same mechanism of action, such as OP, PY, CB, TR, or DT (Caldas 2023).

Cumulative dietary risk assessments using data from the PNCRC and PARA monitoring programs and probabilistic approach were conducted previously by this research group (Caldas et al., 2006a, 2006b, Jardim et al., 2018a, 2018b). In the last studies, residue data of 30,786 samples covering 30 foods analyzed between 2005 and 2015 were used to assess the acute exposure to OP, PY and CB insecticides (Jardim et al., 2018a) and the chronic exposure to DT and TR fungicides (population of 10 years or older), in addition to acute exposure of women of childbearing age to triazoles (Jardim et al., 2018b). A total of 184 foods (food-*as*-eaten) that contained one of the foods-*as*-analyzed as an ingredient were considered in the assessment. In all cases, the cumulative exposure at the 99.9th percentile of the exposure distribution did not exceed the ARfD or the ADI of the index compound (IC) for each cumulative assessment group, reaching a maximum of 59% of the ARfD for methamidophos as the IC for OP (Jardim et al., 2018a) and of 9% of the ADI for mancozeb as the IC for dithiocarbamates (Jardim et al., 2018b). Indeed, cumulative dietary risk assessments to pesticide residues using different approaches have been conducted around the word in the last 20 years, but most studies have shown potential health risks only when very conservative assumptions are used in the assessment (Caldas, 2023).

5. Conclusion

In the last decade, there has been a significant increase in the number of samples and crops analyzed for pesticide residues within both the PARA and PNCRC programs in Brazil. However, compared with the previous decade (2001–2010), the percentages of positive and irregular samples, as well as of samples containing multiple residues, have also increased, indicating the need for ongoing monitoring efforts by the Brazilian government.

In addition to evaluating the proper and legal use of pesticides, the data obtained from these monitoring programs are essential for conducting sound dietary risk assessments. These assessments support management decisions aimed at guaranteeing safe food for the population.

CRediT authorship contribution statement

Andreia Nunes Oliveira Jardim: Writing – original draft, Validation, Investigation, Formal analysis. **Eloisa Dutra Caldas:** Writing – review & editing, Supervision, Project administration, Investigation, Data curation, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Andreia Nunes Oliveira Jardim is currently working at BASF, which among another activities, is a pesticide manufacturer.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodcont.2024.110674.

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