

Currently Used Pesticides' Occurrence in Soils: Recent Results and Advances in Soil-Monitoring and Survey Studies



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Contents

1	Introduction	2
2	Environmental Monitoring and Survey	4
3	Monitoring and Occurrence of CUPs	4
3.1	Monitoring and Survey Studies in Europe	4
3.2	Monitoring and Survey Studies in Asia	26
3.3	Monitoring and Survey Studies in America	28
3.4	Monitoring and Survey Studies in Africa	28
4	Future Perspectives	28
	References	29

Abstract The hazardous effects of pesticides on the ecosystem are indisputable. Many studies have been devoted to the monitoring of pesticides and their occurrence in various systems and the adverse effects that they impose on different parts of the environment. However, most of the efforts have been dedicated to very persistent chlorinated compounds. Other compounds such as currently used pesticides (CUPs) have not been given appropriate attention. To clarify the situation regarding recent investigations in the field of monitoring CUPs in the soil, we performed a review of the studies which have been carried out in the last 5 years worldwide. This review makes clear an acute need for bringing the status of CUPs in the soil to greater consideration and shows the current shortcomings of actions towards monitoring CUPs in soil all around the world.

Keywords Currently used pesticides, Occurrence, Soil monitoring, Transformation products

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1 Introduction

The importance of pesticides to agricultural production is vital, because without them crop yields would be beset by great loss and would be insufficient for the world's growing population. Even with current advancements in agricultural science and technology, 10–90% of all food and fibre crops are lost due to pests, diseases, weeds, and birds [1]. During the years 2015–2018 the application of pesticides on farmlands around the world was 2.65 kg ha^{-1} on average, which showed an increase in comparison with the year 2000 when the figure was 2.07 kg ha^{-1} , and moreover it stood at 1.7 times higher than the amount applied in 1990 [2]. These statistics confirm the increasing use of pesticides over the last decades. Global annual pesticide use in agriculture has increased since the emergence of these substances, yet in the last decade it has increased with gentler slope. From 2015, less than a 0.5% increase in the pesticide use has been recorded each year, and in 2018 – with 0.7% drop comparing to the previous year – it amounted to 4.1 million tons among all the regions. Asia had the highest annual use of pesticide with an average of about 2.1 million tons during the years 2015–2018 [3].

Alongside the beneficial effects that different types of pesticides bring, these chemicals can have destructive toxic effects on non-target organisms even more than on target organisms, and thus they can impact ecosystem biodiversity [4]. Many of these organisms play crucial roles in the ecosystem, including decomposing organic wastes, purifying the environment from pollution, being involved in the nitrogen cycle, preventing climate change, etc. [5, 6]. Soil can be directly contaminated by pesticides after their application. The factors that influence presence of chemicals in the soil are water solubility, affinity for pesticides to sorb to organic carbon (K_{oc}), the octanol/water partition coefficient (K_{ow}), and the half-life (DT_{50}) of the chemical in soil [7]. The pesticides degrade in the soil into transformation products (TPs), which can be as toxic as – or even more toxic than – the parent compounds. The degradation occurs through two processes: (1) the chemical process which is performed through photolysis, hydrolysis, oxidation, and reduction reactions; and (2) the microbial process which occurs with the aid of soil microorganisms [8–10].

Pesticides also enter the aquatic ecosystem via various routes such as spray drift, leaching, runoff, etc., and there they cause contamination or affect aquatic organisms [11–13]. Pesticides also find their way into the human digestive system when a person ingests contaminated water or food. Traces of contamination with pesticides were detected in water streams near lands with urban use [7]. Spray drift, volatilization, and aerial application of pesticides also cause contamination of the air by pesticides, which is a possible source of toxicity transmission to many organisms [14]. Another concern due to intensive use of pesticides is pest resistance occurring through the genetic adaptation of pests to specific pesticides due to repeated intensive use thereof, which in turn leads to the ineffectiveness of pesticide application for pest control and the need for higher application doses [15, 16]. Pesticides also jeopardize human health in various ways. The negative chronic health effects associated with pesticide exposure include dermatologic, neurologic, carcinogenic,

reproductive, genotoxic, cardiovascular, respiratory, endocrine, immunotoxicity, and gastrointestinal effects [14, 17–19].

There is evidence of long-range transport of persistent pesticides. The chemical properties of CUPs are different from earlier highly persistent pesticides. However, new reports have also shown the presence of many CUPs in remote regions, which is proof of long-range transport of these chemicals. The emergence of CUPs in the Arctic environment in both abiotic and biotic matrices has been reported frequently in papers [20–22]. These findings can be alarming; there are still unknown aspects of the behaviour of CUPs in the environment which make them travel far distances, and consequently attempts to monitor CUPs should not be limited only to specific regions or countries.

Due to the adverse effect of pesticides on human health and the ecosystem, new cultivation strategies have been introduced, such as organic farming (OF), integrated crop management, and integrated pest management (IPM), etc. [14, 23]. The benefits and drawbacks of these methods have proven controversial in the studies and subject to debate [24–27], and they are not considered to be comprehensive methods worldwide. Thus, despite all the advances, it is still crucial to assess pesticide occurrence in different compartments by monitoring them, with the aim of preventing CUPs from accumulating and posing consequent negative effects to the environment.

The term “CUP” is a very general concept and to make it clearer for the reader, it would be necessary to define which compounds were taken into consideration. For preparing this review, except for persistent chlorinated pesticides which have been banned all over the world, we considered as a CUP any pesticide related to other chemical groups. Because there is no unified global regulation regarding the prohibition of compounds, and instead they are mostly under local or national regulations, it is very probable that pesticides banned in many countries have been used in some other countries in recent years. According to the literature, the dawn of soil-monitoring studies for the active ingredients considered CUPs in this chapter was in the USA in 1969 [28]. More representative studies were carried out in the subsequent years on a continent-wide scale like Europe [29, 30] or on the scale of a whole country [31–34]. In Asian countries, representative monitoring studies have been carried out in Korea [35–38] and Saudi Arabia [39], but they are still not sufficient. In the African and American contexts an even lower number of studies have been performed on the monitoring of CUPs over the last 50 years. In this chapter, a review of the studies devoted to the survey and monitoring of CUPs in agricultural soils around the world in the last five-year period, 2016–2020, is presented.

2 Environmental Monitoring and Survey

To evaluate the quality of the environment or the influence of anthropogenic activities on different environmental compartments, environmental-monitoring techniques have been designed. In all monitoring programs, the main goal is providing an early alarm before damage to the environment reaches a critical point [40]. Regular measurement is a crucial part of environmental monitoring. Data from environmental monitoring is used for risk assessment to evaluate the health and environmental impact of pollutants and further for management planning and policy-making. However, there are many situations in which there is no historical record or adequate information on the factors causing the problem. In such cases, surveys are used. Generally, surveys include once-only observation [41, 42].

To record and assess soil changes at an early stage across both time and space, soil variables are determined and investigated. Along with many other factors, soil contamination has also been considered frequently as a threat to soil quality, because soil has the unique property of retaining and degrading contaminants [43–46]. Among all the chemicals and factors causing soil contamination, less attention has been devoted to the residue of pesticides in the soils. As will be clear from this review chapter, there is still a huge need for regular, harmonized, and effective monitoring programs all around the world in order to prevent any irrecoverable damage to this non-renewable resource.

3 Monitoring and Occurrence of CUPs

In total, monitoring data for 280 active ingredients were reported in all reviewed articles. In Table 1 these active ingredients along with their physical-chemical properties are listed. Regarding monitoring CUPs, some countries have been more active in terms of studies published. Moreover, the number of studies published during each year can serve as a helpful indication of the efforts dedicated to soil-monitoring studies by time. So, it would be practical to categorize and present this review of the studies in geographical and chronological order. The sections are arranged according to the continents with the highest number of studies on CUP monitoring in soil in the last 5 years.

3.1 *Monitoring and Survey Studies in Europe*

Karasali et al. [48] did a study on 66 soil samples of a pilot area including cotton and maize fields in Kopaida, Greece. They were monitored for eight herbicides for 3 years. The main aim was investigating the effectiveness of the Low-Input Crop Management practice in comparison with conventional farms. In total, the most

Table 1 Physical-chemical properties of the active ingredients (parent compounds and TPs) studied in all reviewed articles [47]

No.	Active ingredient	Type	Metabolite	Chemical class	CAS no.	GUS index	Solubility – in water at 20°C (mg/L)	Log P	pK _a at 25°C	Vapour pressure at 20°C (mPa)	Henry's law constant at 25°C (Pa·m ³ /mol)	Soil DT50 – field (days)	K _{oc} (mL/g)	K _{fec} (mL/g)
1	1-(4-chlorophenyl)-1H-pyrazol-3-ol	Fungicide	Yes	Pyraclostrobin metabolite	-	-	-	-	-	-	-	-	-	-
2	1-[2-chloro-4-(4-chlorophenoxy)phenyl]-2-(1H-1,2,4-triazol-1-yl)ethanol	Fungicide	Yes	Difenoconazole metabolite	-	-	-	-	-	-	-	-	-	-
3	1-[2-chloro-4-(4-chlorophenoxy)phenyl]-2-(1H-1,2,4-triazol-1-yl)ethanone	Fungicide	Yes	Difenoconazole metabolite	-	1.04	12	3.8	-	0.005	-	-	-	2,980
4	1-[3-(p-2-carboxymethylisopropyl)phenyl]-2-methylpropylpiperidine	Fungicide	Yes	Fenpropidin metabolite	-	-	-	-	-	-	-	-	-	-
5	1-[3-(p-2-hydroxymethylisopropyl)phenyl]-2-methylpropylpiperidine	Fungicide	Yes	Fenpropidin metabolite	-	-	-	-	-	-	-	-	-	-
6	1-([3-(4-tert-butylphenyl)-2-methylpropyl]amino)propan-2-ol	Fungicide	Yes	Fenpropimorph metabolite	-	-	-	-	-	-	-	-	-	-
7	2,4-D	Herbicide	No	Alkylchlorophenoxy	94-75-7	3.82	24,300	-0.82	3.4	0.009	4.00E-06	28.8	39.3	24
8	2,4-dimethylphenyl-N-methylformamide	-	Yes	-	-	-	-	-	-	-	-	-	-	-
9	2-[(2,6-dimethylphenyl)(methoxy-acetyl)amino]propanoic acid	Fungicide	Yes	Metaxyl metabolite	-	3.39	265,000	-	-	-	-	17.1	-	17.9
10	2-amino benzimidazole	Fungicide	Yes	Carbendazim metabolite	-	-	14,900	0.91	-	0.38	-	-	22	-
11	2-ethyl-7-nitro-1-propyl-1H-benzimidazole-5-sulfonamide	Herbicide	Yes	Oryzalin metabolite	-	-	-	-	-	-	-	-	-	-
12	2-ethyl-7-nitro-1-propyl-1H-benzimidazole-5-sulfonamide 3-oxide	Herbicide	Yes	Oryzalin metabolite	-	-	-	-	-	-	-	-	-	-

(continued)

Table 1 (continued)

No.	Active ingredient	Type	Metabolite	Chemical class	CAS no.	GUS index	Solubility – in water at 20°C (mg/L)	Log P	pK _a at 25°C	Vapour pressure at 20°C (mPa)	Henry's law constant at 25°C (Pa/m ³ /mol)	Soil DT50 – field (days)	K _{oc} (mL/g)	K _{fec} (mL/g)
13	3-(3-chlorophenyl)-1,1-dimethylurea	Herbicide	Yes	Diuron metabolite	–	–	–	–	–	–	–	–	–	–
14	3,3-dimethyl-1-(1H-1,2,4-triazol-1-yl)butan-2-one	Fungicide	Yes	Tebuconazole metabolite	–	–	–	–	–	–	–	–	–	–
15	3,5,6-trichloro-2-pyridinol	Insecticide	Yes	Chlorpyrifos metabolite	6515-38-4	3.89	80.9	3.21	–	137.32	1.1	75.3	149	84.5
16	3,5-dibromo-4-hydroxybenzoic acid	Herbicide	Yes	Bromoxynil metabolite	–	-0.41	250,000	–	–	5.20E-03	–	–	–	184
17	3-cyano-2,5,6-trichlorobenzamide	Fungicide	Yes	Chlorothalonil metabolite	–	–	–	–	–	–	–	–	–	–
18	3-cyano-6-hydroxy-2,4,5-trichlorobenzamide / 3-cyano-4-hydroxy-2,5,6-trichlorobenzamide	Fungicide	Yes	Chlorothalonil metabolite	–	–	–	–	–	–	–	–	–	–
19	3-phenoxybenzaldehyde (3-PBA)	Insecticide	Yes	–	–	–	–	–	–	–	–	–	–	–
20	4-cyclopropyl-6-methyl-N-(4-hydroxyphenyl)pyrimidine-2-amine	Fungicide	Yes	Cyprodinil metabolite	–	–	–	–	–	–	–	–	–	–
21	4-cyclopropyl-6-methylpyrimidine-2-amine	Fungicide	Yes	Cyprodinil metabolite	–	2.85	–	–	–	–	1.86E-09	148	–	488
22	5-tert-butyl-5-(1H-1,2,4-triazol-1-yl)methyl dihydrofuran-2(3H)-one	Fungicide	Yes	Tebuconazole metabolite	–	–	–	–	–	–	–	–	–	–
23	Acetate	Insecticide	No	Organophosphate	30560-19-1	1.14	790,000	-0.85	8.35	0.226	5.15E-08	3	302	40.3
24	Acetamiprid	Insecticide	No	Neonicotinoid	135410-20-7	0.94	2,950	0.8	0.7	1.73E-04	5.30E-08	3	200	106.5

25	Acetochlor	Herbicide	No	Chloroacetamide	34256-82-1	1.67	282	4.14	NA	2.20E-02	2.10E-03	12.1	156	285
26	Acetochlor-ESA	Herbicide	Yes	Acetochlor metabolite	-	3.73	-	-	-	-	-	-	28.8	104
27	Acetochlor-OA	Herbicide	Yes	Acetochlor metabolite	500-72-1	2.49	-	-	-	-	-	12	24.3	49.8
28	Acetyl-dinoseb-6-amino	Acaricide	Yes	Dinoseb metabolite	-	-	-	-	-	-	-	-	-	-
29	Alachlor	Herbicide	No	Chloroacetamide	15972-60-8	0.8	240	3.09	0.62	2.9	3.20E-03	14	335	1,994
30	Alachlor-hydroxy	Herbicide	Yes	Alachlor/ acetochlor metabolite	56681-55-1	-	-	1.96	-	-	-	-	278	-
31	AMBA	Herbicide	Yes	Mesotrione metabolite	-	2.65	-	0.32	-	-	-	-	77	52
32	Aminopyralid	Herbicide	No	Pyridine compound	150114-71-9	3.34	2,480	-2.87	2.56	2.59E-09	9.61E-12	2.11	-	8.3
33	AMPA	Herbicide	Yes	Glyphosate metabolite	1066-51-9	0.04	1,466,561	-1.63	-	-	0.16	419	2,002	9,664.5
34	Atrazine	Herbicide	No	Triazine	1912-24-9	2.57	35	2.7	1.7	0.039	1.50E-04	29	100	174
35	Atrazine-2-hydroxy	Herbicide	Yes	Atrazine metabolite	2163-68-0	-	5.9	2.09	-	1.131	6.36E-08	-	-	-
36	Atrazine-desethyl	Herbicide	Yes	Atrazine metabolite	6190-65-4	3.24	2,700	1.51	-	12.44	1.55E-04	45	110	-
37	Atrazine-desethyl-desisopropyl	Herbicide	Yes	Atrazine metabolite	-	-	-	-	-	-	-	-	-	-
38	Atrazine-desisopropyl	Herbicide	Yes	Atrazine metabolite	1007-28-9	-	980	1.15	-	-	980	-	130	-
39	Azoxystrobin	Fungicide	No	Strobilurin	131860-33-8	3.1	6.7	2.5	NA	1.10E-07	7.40E-09	180.7	589	423
40	Azoxystrobin acid	Fungicide	Yes	Azoxystrobin metabolite	-	-	-	-	-	-	-	-	-	-
41	Benazone	Herbicide	No	Benzothiazinone	25057-89-0	1.95	7,112	-0.46	3.51	0.17	7.20E-05	7.5	55.3	59.6
42	Bifenazate	Insecticide	No	Hydrazine carboxylate	149877-41-8	0.98	2.06	3.4	12.94	1.33E-02	1.01E-03	4.8	1,778	368
43	Bifenox	Herbicide	No	Diphenyl ether	42576-02-3	0.23	0.1	3.64	NA	0.162	1.62E-04	17.3	-	6,475

(continued)

Table 1 (continued)

No.	Active ingredient	Type	Metabolite	Chemical class	CAS no.	GUS index	Solubility – in water at 20°C (mg/L)	Log P	pK _a at 25°C	Vapour pressure at 20°C (mPa)	Henry's law constant at 25°C (Pa·m ³ /mol)	Soil DT50 – field (days)	K _{oc} (mL/g)	K _{fec} (mL/g)
44	Bifenox-acid	Herbicide	Yes	Bifenox metabolite	53774-07-5	3.23	1,000	4.55	–	–	–	–	–	143
45	Bifenox-amino	Herbicide	Yes	Bifenox metabolite	–	–	–	–	–	–	–	–	–	–
46	Bifenthrin	Insecticide	No	Pyrethroid	82657-04-3	-2.66	0.001	6.6	NA	0.0178	7.74E-05	86.8	236,610	–
47	Bitertanol	Fungicide	No	Triazole	55179-31-2	0.83	3.8	4.1	NA	1.36E-06	2.60E-07	23	–	2,461
48	Bitertanol benzoic acid	Fungicide	Yes	Bitertanol metabolite	–	–	–	–	–	–	0.16	–	–	–
49	Bitertanol ketone	Fungicide	Yes	Bitertanol metabolite	–	–	–	–	–	–	–	–	–	–
50	Boscalid	Fungicide	No	Carboxamide	188425-85-6	2.68	4.6	2.96	NA	0.00072	5.18E-05	254	–	772
51	Bromoxynil	Herbicide	No	Hydroxybenzomitrile	1689-84-5	1.71	38,000	0.27	3.86	0.12	8.70E-07	8	302	127.2
52	Bupirimate	Fungicide	No	Pyrimidinol	41483-43-6	1.11	13.06	3.68	4.4	0.057	1.35	34	–	1,882
53	Buprofezin	Insecticide	No	Unclassified	69327-76-0	0.45	0.46	4.93	NA	0.042	2.80E-02	45.6	5,363	5,334
54	Butachlor	Herbicide	No	Chloroacetamide	23184-66-9	1.23	20	4.5	–	0.24	3.74E-03	11.5	700	–
55	Cadusafos	Insecticide	No	Organophosphate	95465-99-9	2.62	245	3.85	NA	119.6	0.132	39	–	227
56	Carbaryl	Insecticide	No	Carbamate	63-25-2	2.02	9.1	2.36	10.4	0.0416	9.20E-05	–	300	211
57	Carbendazim	Fungicide	No	Benzimidazole	10605-21-7	2.21	8	1.48	4.2	0.09	3.60E-03	22	–	225
58	Carbetamide	Herbicide	No	Carbamate	16118-49-3	1.85	3,270	1.78	11.3	0.0003	1.93E-08	8	–	88.6
59	Carbofuran	Insecticide	No	Carbamate	1563-66-2	2.36	322	1.8	NA	0.08	5.00E-05	14	–	86.5

60	Chlorantraniliprole	Insecticide	No	Anthranilic diamide	50008-45-7	3.51	0.88	2.86	10.88	6.30E-09	3.20E-09	204	362	301
61	Chlorfenapyr	Insecticide	No	Pyrole	122453-73-0	-0.01	0.112	4.83	NA	9.81E-03	5.81E-04	-	12,000	-
62	Chlorfenvinphos	Insecticide	No	Organophosphate	470-90-6	1.72	145	3.8	-	0.53	-	30	680	-
63	Chloridazon	Herbicide	No	Pyridazinone	1698-60-8	2.16	422	1.19	3.38	1.00E-06	5.30E-10	34.7	120	199
64	Chloridazon-methyl-desphenyl	Herbicide	Yes	Chloridazon metabolite	-	4.39	-	-1.38	-	-	-	-	-	92
65	Chlorothaloni	Fungicide	No	Chloronitrile	1897-45-6	1.12	0.81	2.94	NA	0.076	2.50E-02	17.9	2,632	1,288
66	Chlorothalonil-4-hydroxy	Fungicide	Yes	Chlorothalonil metabolite	-	2.56	13.5	0.61	-	-	-	-	498	383
67	Chlorotoluron	Herbicide	No	Urea	15545-48-9	-	74	2.5	NA	0.005	1.44E-05	34	196	-
68	Chlorotoluron-desmethyl	Herbicide	Yes	Chlorotoluron metabolite	-	2.84	-	-	-	-	1.09E-03	-	248	-
69	Chlorotoluron-didesmethyl	Herbicide	Yes	Chlorotoluron metabolite	-	-	-	-	-	-	-	-	-	-
70	Chlorpyrifos	Insecticide	No	Organophosphate	2921-88-2	0.58	1.05	4.7	NA	1.43	0.478	27.6	5,509	3,954
71	Chlorsulfuron	Herbicide	No	Sulfonylurea	64902-72-3	3.8	12,500	-0.99	3.4	3.07E-06	3.50E-11	36.2	-	36.3
72	Clomazone	Herbicide	No	Isoxazolidinone	81777-89-1	2.72	1,212	2.58	NA	27	5.90E-03	27.3	300	128.3
73	Clopyralid	Herbicide	No	Pyridine compound	1702-17-6	3.02	7,850	-2.63	2.01	1.36E-09	1.80E-11	8.2	5	-
74	CMBA	Herbicide	Yes	Sulcotriene metabolite	53250-83-2	-	60,000	-0.2	-	-	-	8.5	-	4.76
75	Cyanophenoxy pyrimidinol	Fungicide	Yes	Azoxystrobin metabolite	-	-	-	-	-	-	-	-	-	-
76	Cyfluthrin	Insecticide	No	Pyrethroid	68359-37-5	-1.66	0.0066	6	NA	0.0003	5.30E-02	33	123,930	-
77	Cyhalothrin	Insecticide	No	Pyrethroid	68085-85-8	-2.2	0.004	6.8	9	1.00E-09	1.80E-02	-	180,000	-
78	Cyhalothrin-λ	Insecticide	No	Pyrethroid	91465-08-6	-2.09	0.005	5.5	NA	0.0002	2.00E-02	26.9	283,707	290,311

(continued)

Table 1 (continued)

No.	Active ingredient	Type	Metabolite	Chemical class	CAS no.	GUS index	Solubility – in water at 20°C (mg/L)	Log P	pK _a at 25°C	Vapour pressure at 20°C (mPa)	Henry's law constant at 25°C (Pa·m ³ /mol)	Soil DT50 – field (days)	K _{oc} (mL/g)	K _{fec} (mL/g)
79	Cymoxamil	Fungicide	No	Cyanoacetamide oxime	57966-95-7	1.47	780	0.67	9.3	0.15	3.80E-05	3.5	–	43.6
80	Cypermethrin	Insecticide	No	Pyrethroid	52315-07-8	-1.99	0.009	5.55	NA	6.78E-03	0.31	21.9	307,558	–
81	Cypermethrin- α	Insecticide	No	Pyrethroid	67375-30-8	-2.38	0.004	5.8	NA	0.00038	5.30E-02	42.6	288,735	–
82	Cyproconazole	Fungicide	No	Triazole	94361-06-5	3.04	93	3.09	NA	0.026	5.00E-05	1.29	–	364
83	Cyprodinil	Fungicide	No	Anilino-pyrimidine	121552-61-2	1.06	13	4	4.44	5.10E-01	6.60E-03	45	–	2,277
84	Deltamethrin	Insecticide	No	Pyrethroid	52918-63-5	-3.98	0.0002	4.6	NA	1.2E-05	3.10E-02	21	1E+07	1E+07
85	Desmedipham	Herbicide	No	Carbamate	13684-56-5	0.35	5.6	2.7	NA	4.00E-05	5.40E-07	8	–	4,124
86	Diazinon	Insecticide	No	Organophosphate	333-41-5	1.51	60	3.69	2.6	11.97	6.09E-02	18.4	609	643
87	Dicamba	Herbicide	No	Benzoic acid	1918-00-9	1.72	250,000	-1.88	1.87	1.67	1.00E-04	3.9	–	12.36
88	Dichlorvos	Insecticide	No	Organophosphate	62-73-7	0.69	18,000	1.9	NA	2.100	2.58E-02	–	50	–
89	Difenoconazole	Fungicide	No	Triazole	119446-68-3	0.83	15	4.36	1.07	3.33E-05	9.00E-07	85	–	3,760
90	Diffubenzuron	Insecticide	No	Benzoylurea	35367-38-5	0.17	0.08	3.89	–	0.00012	4.70E-04	–	–	4,620
91	Diffufencan	Herbicide	No	Carboxamide	83164-33-4	1.19	0.05	4.2	–	4.25E-03	1.18E-02	64.6	5,504	2,215
92	Dimethachlor	Herbicide	No	Chloroacetamide	50863-36-5	1.09	2,300	2.17	NA	0.64	1.70E-04	3.2	–	69
93	Dimethachlor-ESA	Herbicide	Yes	Dimethachlor metabolite	–	4.05	–	–	–	–	–	Soil, groundwater	–	–

Table 1 (continued)

No.	Active ingredient	Type	Metabolite	Chemical class	CAS no.	GUS index	Solubility – in water at 20°C (mg/L)	Log P	pK _a at 25°C	Vapour pressure at 20°C (mPa)	Henry's law constant at 25°C (Pa·m ³ /mol)	Soil DT50 – field (days)	K _{oc} (mL/g)	K _{fec} (mL/g)
114	Ethirimol	Fungicide	No	Pyrimidinol	23947-60-6	1.82	233	2.3	5	0.267	2.00E-04	20	–	402
115	Ethofumesate	Herbicide	No	Benzofuran	26225-79-6	3.04	50	2.7	NA	0.36	3.72E-03	37.8	–	118
116	Fenarimol	Fungicide	No	Pyrimidine	60168-88-9	2.12	13.7	3.69	–	0.065	7.00E-04	74	–	734
117	Fenazaquin	Insecticide	No	Quinazoline	120928-09-8	-0.63	0.102	5.51	2.44	1.90E-02	5.71E-02	30.5	–	26,499
118	Fenitrothion	Insecticide	No	Organophosphate	122-14-5	0.48	19	3.32	NA	0.676	9.86E-03	–	2000	791
119	Fenproprathrin	Insecticide	No	Pyrethroid	39515-41-8	-0.8	0.33	6.04	–	0.76	#####	28	5,000	35,660
120	Fenpropidin	Fungicide	No	Unclassified	67306-00-7	0.71	530	2.6	10.13	17	10.7	49.2	–	3,808
121	Fenpropidin-hydroxy	Fungicide	Yes	Fenpropidin metabolite	–	–	–	–	–	–	–	–	–	–
122	Fenpropidin-N-oxide	Fungicide	Yes	Fenpropidin metabolite	–	–	–	–	–	–	–	–	–	–
123	Fenpropimorph	Fungicide	No	Morpholine	67564-91-4	0.5	4.32	4.5	6.98	3.9	2.74E-04	25.5	–	4,382
124	Fenpropimorph carboxy	Fungicide	Yes	Fenpropimorph metabolite	–	–	–	–	–	–	–	–	–	–
125	Fenpyroximate	Insecticide	No	Pyrazolium	134098-61-6	0.26	0.023	5.01	NA	0.01	0.182	4.3	–	52,067
126	Fenthiön	Insecticide	No	Organophosphate	55-38-9	1.26	4.2	4.84	–	0.37	2.40E-02	–	1,500	–
127	Fenvalerate	Insecticide	No	Pyrethroid	51630-58-1	0.52	0.001	5.01	–	0.0192	4.20E-02	–	5,273	–
128	Florasulam	Herbicide	No	Triazolopyrimidine	145701-23-1	2.5	6.360	-1.22	4.54	0.01	4.35E-07	8.5	22	20.37
129	Fluazifop-P-acid	Herbicide	Yes	Fluazifop-p-butyl metabolite	83066-88-0	3.23	40.5	3.18	3.12	–	–	–	205	48.72

130	Fluazifop-P-butyl	Herbicide	No	Aryloxyphenoxypropionate	79241-46-6	0.43	0.93	4.5	NA	0.12	0.049	8.2	3,394	-
131	Fluazinam	Fungicide	No	Phenylpyridinamine	79622-59-6	1	0.135	4.03	7.34	7.5	25.9	16.4	16,430	1958
132	Fluazinam-3-hydroxy	Fungicide	Yes	Fluazinam metabolite		-	-	-	-	-	-	-	-	-
133	Fludioxonil	Fungicide	No	Phenylpyrrole	131341-86-1	-1.35	1.8	4.12	0	3.90E-04	5.40E-05	20.5	145,600	132,100
134	Flumequine	-	No	-		-	-	-	-	-	-	-	-	-
135	Fluometuron	Herbicide	No	Phenylurea	2164-17-2	4.24	111	2.28	NA	0.125	2.63E-04	89.8	-	67.4
136	Fluroxypyr	Herbicide	No	Pyridine compound	69377-81-7	1.03	6,500	0.04	2.94	3.8E-06	1.69E-10	51	-	68
137	Flusilazole	Fungicide	No	Triazole	85509-19-9	1.54	41.9	3.87	2.5	0.0387	2.70E-04	94	1,664	-
138	Folpet	Fungicide	No	Phthalimide	133-07-3	0.72	0.8	3.02	NA	2.10E-02	8.00E-03	3	304	-
139	Glyphosate	Herbicide	No	Organophosphate	1071-83-6	-0.29	10,500	-3.2	2.34	0.0131	2.10E-07	23.79	1,424	16,331
140	Hexaconazole	Fungicide	No	Triazole	79983-71-4	2.31	18	3.9	2.3	0.018	3.33E-04	225	1,040	-
141	Hexythiazox	Insecticide	No	Carboxamide	78587-05-0	0.03	0.1	2.67	NA	1.33E-03	1.19E-02	17.7	-	9,455
142	Imazalil	Fungicide	No	Imidazole	35554-44-0	0.26	184	2.56	6.49	0.158	0.00011	6.4	-	4,753
143	Imidacloprid	Insecticide	No	Neonicotinoid	138261-41-3	3.69	610	0.57	NA	4.00E-07	1.70E-10	174	-	225
144	Imidacloprid-guanidine	Insecticide	Yes	Imidacloprid metabolite		-	-	-	-	-	-	-	-	-
145	Imidacloprid-urea	Insecticide	Yes	Imidacloprid metabolite		-	-	-	-	-	-	-	-	-
146	Indoxacarb	Insecticide	No	Oxadiazine	173584-44-6	0.27	0.2	4.65	NA	9.80E-06	6.00E-05	5.97	4,483	-
147	Ioxynil	Herbicide	No	Hydroxybenzotrile	1689-83-4	1.06	3,034	2.2	4.1	0.00204	1.50E-05	5	-	303

(continued)

Table 1 (continued)

No.	Active ingredient	Type	Metabolite	Chemical class	CAS no.	GUS index	Solubility – in water at 20°C (mg/L)	Log P	pK _a at 25°C	Vapour pressure at 20°C (mPa)	Henry's law constant at 25°C (Pa/m ³ /mol)	Soil DT50 – field (days)	K _{oc} (mL/g)	K _{fec} (mL/g)
148	Iprodione	Fungicide	No	Dicarbonyimide	36734-19-7	0.43	6.8	3	NA	0.0005	7.00E-06	11.7	700	3,927
149	Isocarbophos	Insecticide	No	Organophosphate	24353-61-5	–	70.1	2.7	–	–	–	–	190	–
150	Isoproturon	Herbicide	No	Urea	34123-59-6	2.61	70.2	2.5	NA	5.50E-03	1.46E-05	23	–	122
151	Isoproturon-didemethyl	Herbicide	Yes	Isoproturon metabolite	–	–	–	–	–	–	–	–	–	–
152	Isoproturon-monomethyl	Herbicide	Yes	Isoproturon metabolite	34123-57-4	2.78	–	–	–	–	–	–	–	147
153	Kresoxim-methyl	Fungicide	No	Strobilurin	143390-89-0	0	2	3.4	NA	2.30E-03	3.60E-04	–	–	308
154	Lactofen	Herbicide	No	Diphenyl ether	77501-63-4	0	0.5	–	–	0.0093	4.56E-03	–	10.000	–
155	Lenacil	Herbicide	No	Uracil	2164-08-1	3.02	2.9	1.69	10.7	1.70E-06	1.30E-07	39.8	165	130
156	Lenacil-oxo	Herbicide	Yes	Lenacil metabolite	–	–	–	–	–	–	–	–	–	–
157	Linuron	Herbicide	No	Urea	330-55-2	2.11	63.8	3	NA	0.051	2.00E-04	48	842.8	559
158	Linuron-desmethyl	Herbicide	Yes	Linuron metabolite	–	–	–	–	–	–	–	–	–	–
159	Lufenuron	Insecticide	No	Benzoylurea	103055-07-8	-1.48	0.046	5.12	10.2	4.00E-03	3.41E-02	256	–	41,182
160	Maneb	Fungicide	No	Carbamate	12427-38-2	0.75	178	-0.45	NA	0.014	2.08E-05	7	2000	1,310
161	MCPA	Herbicide	No	Aryloxyalkanoic acid	94-74-6	2.98	29,390	-0.81	3.73	0.4	5.50E-05	25	–	74
162	MCPA (methyl ester)	Herbicide	No	Aryloxyalkanoic acid	–	–	–	–	–	–	–	–	–	–
163	Mecoprop	Herbicide	No	Aryloxyalkanoic acid	7085-19-0	2.29	250,000	-0.19	3.11	1.6	2.20E-04	–	47	31
164	Mecoprop-P	Herbicide	No	Aryloxyalkanoic acid	16484-77-8	2.94	250,000	-0.19	3.7	0.23	5.70E-05	21	–	59.8

165	Mesotrione	Herbicide	No	Triketone	104206-82-8	1.45	1,500	0.11	3.12	5.70E-03	5.10E-07	5	122	83.3
166	Metaxalyl	Fungicide	No	Phenylamide	57837-19-1	2.06	8,400	1.75	0	0.75	1.60E-05	38.7	162	162.3
167	Metaxalyl-M	Fungicide	No	Phenylamide	70630-17-0	2.42	26,000	1.71	NA	3.3	3.50E-05	14.1	-	78.9
168	Metamitron	Herbicide	No	Triazinone	41394-05-2	2.16	1770	0.85	NA	7.44E-04	8.95E-08	11.1	77.7	86.4
169	Metamitron-desamino	Herbicide	Yes	Metamitron metabolite	-	2.97	399.9	-	-	-	-	31.1	-	103
170	Metazachlor	Herbicide	No	Chloroacetamide	67129-08-2	1.75	450	2.49	NA	0.093	5.90E-05	6.8	54	79.6
171	Metazachlor-deschloro	Herbicide	Yes	Metazachlor metabolite	-	-	-	-	-	-	-	-	-	-
172	Metazachlor-ESA	Herbicide	Yes	Metazachlor metabolite	-	6.8	-	-	-	-	-	115	8.8	5
173	Metazachlor-hydroxy	Herbicide	Yes	Metazachlor metabolite	-	-	-	-	-	-	-	-	-	-
174	Metazachlor-OA	Herbicide	Yes	Metazachlor metabolite	-	5.18	-	-	-	-	-	96.3	18.9	24.6
175	Metconazole	Fungicide	No	Triazole	125116-23-6	2.03	30.4	3.85	11.38	2.10E-05	2.21E-07	134.7	-	1,116
176	Methamidophos	Insecticide	No	Organophosphate	10265-92-6	2.41	200,000	-0.79	-	2.3	1.60E-06	4	1	-
177	Methoxyfenozide	Insecticide	No	Diacylhydrazine	161050-58-4	3	3.3	3.72	12.2	1.33E-02	1.64E-04	68	402	231
178	Methyl-N-(3-hydroxyphenyl) carbamate	Herbicide	Yes	Phenmedipham metabolite	13683-89-1	1.62	-	1.3	-	-	-	-	-	48
179	Metolachlor	Herbicide	No	Chloroacetamide	51218-45-2	2.36	530	3.4	NA	1.7	2.40E-03	21	120	163
180	Metolachlor-demethyl	Herbicide	Yes	Metolachlor metabolite	-	-	-	-	-	-	-	-	-	-
181	Metolachlor-ESA	Herbicide	Yes	Metolachlor metabolite	171118-09-5	7.22	212,461	-1.89	-	-	-	-	9	-
182	Metolachlor-OA	Herbicide	Yes	Metolachlor metabolite	152019-73-3	6.88	238	-	-	-	-	127.5	17	7.26
183	Metribuzin	Herbicide	No	Triazinone	21087-64-9	2.96	10,700	1.75	1.3	0.121	2.50E-05	19	-	37.92
184	Metribuzin-desamino	Herbicide	Yes	Metribuzin metabolite	-	1.25	475	-	-	6.50E-03	-	-	-	43.7

(continued)

Table 1 (continued)

No.	Active ingredient	Type	Metabolite	Chemical class	CAS no.	GUS index	Solubility – in water at 20°C (mg/L)	Log P	pK _a at 25°C	Vapour pressure at 20°C (mPa)	Henry's law constant at 25°C (Pa/m ³ /mol)	Soil DT50 – field (days)	K _{oc} (mL/g)	K _{fec} (mL/g)
185	Metribuzin-diketo	Herbicide	Yes	Metribuzin metabolite	56507-37-0	3.61	3,300	0.9	–	0.081	–	–	99	48
186	Mevinphos	Insecticide	No	Organophosphate	7786-34-7	1.12	600,000	0.127	–	17	6.30E-06	–	44	–
187	MNBA	Herbicide	Yes	Mesotrione metabolite	110964-79-9	1.86	–	–1.3	–	–	–	–	6	3.2
188	Monocrotophos	Insecticide	No	Organophosphate	6923-22-4	4.02	818,000	–0.22	–	0.29	–	30	19	–
189	Myclobutanil	Fungicide	No	Triazole	88671-89-0	1.99	132	2.89	2.3	0.198	4.33E-04	35	–	517
190	N-(2,6-dimethylphenyl)-N-(methoxyacetyl)alanine	Fungicide	Yes	Metaxyl metabolite	–	4.38	–	–	–	–	4.54	51	38	27.2
191	Napropamid	Herbicide	No	Alkanamide	15299-99-7	1.96	74	3.3	NA	2.20E-02	8.10E-05	72	839	885
192	Napropamide-didesethyl	Herbicide	Yes	Napropamide metabolite	–	–	–	–	–	–	–	–	–	–
193	Nicosulfuron	Herbicide	No	Sulfonylurea	111991-09-4	3.44	7,500	0.61	4.78	8.00E-07	1.48E-11	19.3	30	21
194	N-propyl-N-(2-(2,4,6-trichlorophenoxy)ethyl)urea	Fungicide	Yes	Prochloraz metabolite	–	–	–	–	–	–	–	–	–	–
195	N-propyl-N-(2-(2,4,6-trichlorophenoxy)-ethyl)amine	Fungicide	Yes	Prochloraz metabolite	–	–	–	–	–	–	–	–	–	–
196	Omethoate	Insecticide	No	Organophosphate	1113-02-6	2.73	500,000	–0.9	–	19	4.62E-09	14	41.3	–
197	Orbencarb	Herbicide	No	Thiocarbamate	34622-58-7	–	24	4.17	–	4.26	1.33E-01	–	–	–
198	Orbencarb-desethyl	Herbicide	Yes	Orbencarb metabolite	–	–	–	–	–	–	–	–	–	–
199	Oryzalin	Herbicide	No	Dinitroamine	19044-88-3	2.27	1.13	3.73	9.4	1.10E-07	3.37E-08	98.2	949	724

Table 1 (continued)

No.	Active ingredient	Type	Metabolite	Chemical class	CAS no.	GUS index	Solubility – in water at 20°C (mg/L)	Log P	pK _a at 25°C	Vapour pressure at 20°C (mPa)	Henry's law constant at 25°C (Pa/m ³ /mol)	Soil DT50 – field (days)	K _{oc} (mL/g)	K _{fec} (mL/g)
219	Procymidone	Fungicide	No	Dicarboximide	32809-16-8	3.3	2.46	3.3	NA	0.023	2.65E-03	208.3	378	–
220	Profenofos	Insecticide	No	Organophosphate	41198-08-7	0.59	28	1.7	NA	2.53	1.65E-03	7	2,016	–
221	Prometryn	Herbicide	No	Triazine	7287-19-6	0.59	33	3.34	4.1	0.13	1.20E-03	–	400	4,330
222	Propachlor	Herbicide	No	Chloroacetamide	1918-16-7	1.47	580	1.6	–	30.6	3.65E-03	–	80	–
223	Propachlor-ESA	Herbicide	Yes	Propachlor metabolite	–	–	–	–	–	–	–	–	–	–
224	Propachlor-OA	Herbicide	Yes	Propachlor metabolite	–	–	–	–	–	–	–	–	–	–
225	Propamocarb	Fungicide	No	Carbamate	24579-73-5	–	900,000	0.84	9.5	730	1.50E-04	–	–	–
226	Propaquizafop	Herbicide	No	Aryloxyphenoxypyronate	111479-05-1	2.67	0.63	4.78	NA	4.39E-07	9.20E-08	85	–	–
227	Propiconazole	Fungicide	No	Triazole	60207-90-1	1.58	150	3.72	1.09	0.056	9.20E-05	35.2	1,086	955
228	Propoxur	Insecticide	No	Carbamate	114-26-1	3.65	1800	0.14	–	1.3	1.50E-04	28	30	–
229	Prosulcarb	Herbicide	No	Thiocarbamate	52888-80-9	0.76	13.2	4.48	NA	0.79	0.0152	9.8	–	1,693
230	Prothioconazole	Fungicide	No	Triazolothione	178928-70-6	-0.07	22.5	2	6.9	7.40E-06	1.10E-08	0.77	1,765	2,556
231	Pyraclostrobin	Fungicide	No	Strobilurin	175013-18-0	0.05	1.9	3.99	NA	2.60E-05	5.31E-06	33.3	9,304	9,315
232	Pyridaben	Insecticide	No	Pyridazinone	96489-71-3	-1.2	0.022	6.37	NA	0.001	0.3	29	–	66,503
233	Pyrifenoxy	Fungicide	No	Pyridine	88283-41-4	1.84	300	3.4	4.61	1.7	5.80E-03	66	980	–

234	Pyrimethanil	Fungicide	No	Anilinyrimidine	53112-28-0	2.17	110	2.84	3.52	1.1	2.20E-03	31.4	-	355.7
235	Quinalphos	Insecticide	No	Organophosphate	13593-03-8	1.1	17.8	4.44	-	0.346	4.70E-03	-	1,465	-
236	Quinmerac	Herbicide	No	Quinoline	90717-03-6	2.05	107,000	-1.41	4.31	1.00E-07	1.00E-10	9.8	86	86
237	Quinoxifen	Fungicide	No	Quinoline	124495-18-7	-0.8	0.047	5.1	NA	0.012	3.08E-02	169.3	-	22,929
238	Quizalofop-P-ethyl	Herbicide	No	Aryloxyphenoxypionate	100646-51-3	0.19	0.61	4.61	NA	1.10E-04	6.70E-05	1.8	-	1,816
239	Resmethrin	Insecticide	No	Pyrethroid	10453-86-8	-1.48	0.01	5.43	-	0.0015	8.93E-02	-	100,000	-
240	Simazine	Herbicide	No	Triazine	122-34-9	2.2	5	2.3	1.62	0.00081	5.60E-05	90	130	750
241	Simazine-2-hydroxy	Herbicide	Yes	Simazine metabolite	2599-11-3	-	32.8	1.67	-	-	-	-	5	-
242	S-metolachlor	Herbicide	No	Chloroacetamide	87392-12-9	2.32	480	3.05	NA	3.7	2.20E-03	21	-	226.1
243	S-metolachlor-ESA	Herbicide	Yes	S-metolachlor metabolite	-	-	-	-	-	-	-	-	-	-
244	S-metolachlor-OA	Herbicide	Yes	S-metolachlor metabolite	-	-	-	-	-	-	-	-	-	-
245	Spinosad	Insecticide	No	Micro-organism derived	168316-95-8	-	-	-	-	-	-	-	-	-
246	Spirodiclofen	Insecticide	No	Tetronic acid	148477-71-8	-0.42	0.05	5.83	-	3.00E-04	2.00E-02	-	31,037	-
247	Spiroxamine	Fungicide	No	Morpholine	118134-30-8	-0.28	405	2.89	6.9	3.5	3.80E-03	57.5	-	14,567
248	Spiroxamine acid	Fungicide	Yes	Spiroxamine metabolite	-	-	-	-	-	-	-	-	-	-
249	Spiroxamine desethyl acid	Fungicide	Yes	Spiroxamine metabolite	-	-	-	-	-	-	-	-	-	-
250	Spiroxamine-desethyl	Fungicide	Yes	Spiroxamine metabolite	-	0.49	14.8	-	-	-	-	-	-	4,816
251	Spiroxamine-despropyl	Fungicide	Yes	Spiroxamine metabolite	-	0.58	46.6	-	-	-	-	-	-	4,165
252	Sulcotrione	Herbicide	No	Triketone	99105-77-8	1.36	165	-1.7	3.13	5.00E-03	6.00E-07	3.6	-	36

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270	Thiamethoxam	Insecticide	No	Neonicotinoid	153719-23-4	3.58	4,100	-0.13	NA	6.60E-06	4.70E-10	39	56.2	-
271	Thiaram	Fungicide	No	Carbamate	137-26-8	0.02	18	1.84	8.19	2.00E-02	1.39E-04	15	-	9,629
272	Thiophanate-methyl	Fungicide	No	Benzimidazole	23564-05-8	0.5	18.5	1.4	7.28	9.00E-03	1.67E-04	2	-	220
273	Triadimefon	Fungicide	No	Triazole	43121-43-3	1.59	70	3.18	-	0.02	9.00E-05	-	300	749
274	Triadimenol	Fungicide	No	Triazole	55219-65-3	2.44	72	3.18	NA	0.0005	3.50E-06	64.9	750	273
275	Triasulfuron	Herbicide	No	Sulfonylurea	82097-50-5	4.59	815	-0.59	4.64	0.0021	8.00E-05	38.5	60	12.8
276	Triazophos	Insecticide	No	Organophosphate	24017-47-8	1.38	35	3.55	-	1.33	4.90E-03	9	358	-
277	Trifloxystrobin	Fungicide	No	Strobilurin	141517-21-7	0.15	0.61	4.5	NA	3.40E-03	2.30E-03	1.69	-	2,287
278	Trifloxystrobin acid	Fungicide	Yes	Trifloxystrobin metabolite	252913-85-2	3.57	21,000	-	-	5.50E-03	-	70	-	116
279	Trifluralin	Herbicide	No	Dinitroaniline	1582-09-8	0.13	0.221	5.27	NA	9.5	10.2	170	15,800	8,765
280	Trinexapac-ethyl	Plant growth regulator	No	Cyclohexanecarboxylate derivative	95266-40-3	1.81	10,200	-0.29	4.57	2.16	5.40E-04	14.6	-	280

“-” no available, NA no applicable

frequently detected herbicides were ethalfuralin (39% of samples, up to $260 \mu\text{g kg}^{-1}$), fluometuron (35% of samples, up to $385 \mu\text{g kg}^{-1}$), trifluralin (29% of samples, up to $95 \mu\text{g kg}^{-1}$), S-metolachlor (21% of samples, up to $210 \mu\text{g kg}^{-1}$), and pendimethalin (4% of samples, up to $29.8 \mu\text{g kg}^{-1}$). Quizalofop, fluazifop, and chlorpyrifos were not detected in any of the fields. Overall, the occurrence of herbicides after implementation of the practice was claimed to be lower.

In Switzerland, Chiaia-Hernandez et al. [34] analysed 29 archived soil samples taken over 14 years from agricultural lands and investigated the long-term persistency of 170 CUPs including 80 polar pesticides and 93 TPs. The most prevalent CUPs were simazine (97%, up to $80 \mu\text{g kg}^{-1}$), atrazine (86%, up to $249 \mu\text{g kg}^{-1}$), tebutam (70%, up to $22 \mu\text{g kg}^{-1}$), and carbendazim (72%, up to $61 \mu\text{g kg}^{-1}$), and of TPs were atrazine-2-hydroxy (100%, up to $220 \mu\text{g kg}^{-1}$), simazine-2-hydroxy +terbutylazine-desethyl-2-hydroxy (93%, up to $680 \mu\text{g kg}^{-1}$), terbutylazine-desethyl (90%, up to $2 \mu\text{g kg}^{-1}$), and atrazine-desisopropyl (86%, up to $9 \mu\text{g kg}^{-1}$). Of the pesticides which were applied on lands and detected in archived samples, 45% were detected while applied, and 16% were those applied but not detected. Moreover, 38% of parent pesticides were detected, although they had never been applied on land. It was claimed that even parental pesticides and/or their TPs with lower half-life showed persistence higher than what was expected. In addition, some compounds may occur as impurities in other formulations or as a contamination from adjacent sites. Of the TPs related to applied pesticides, 47% were detected in the soil samples.

A study in the Czech Republic performed by Scherr et al. [49] on soil samples collected from 75 intensively used agricultural fields for residues of 12 chlorotriazine herbicides and their TPs found that the parent compound with the highest maximum concentration and detection frequency (DF) was terbutylazine with values of $37.6 \mu\text{g kg}^{-1}$ and 17%, respectively. Atrazine was detected in concentrations less than the limit of quantification (LOQ) and simazine was detected in one sample with a concentration of $8.7 \mu\text{g kg}^{-1}$. Major cases of contamination were related to TPs such as 2-hydroxyterbutylazine (83%, $74.5 \mu\text{g kg}^{-1}$), 2-hydroxysimazine (44%, $24.4 \mu\text{g kg}^{-1}$), 2-hydroxyatrazine (39%, $123 \mu\text{g kg}^{-1}$), and deethylsimazine (12%, $31 \mu\text{g kg}^{-1}$). According to the application history of parental pesticides, which stood at 6 months to one decade before sampling for the various compounds, it was stated that the degradation of parent pesticides in soil will produce TPs which are even more persistent than their parent compounds. This result showed the importance of monitoring for TPs besides chemicals used as pesticides in the soil, as they may be more accumulative and persistent than their parent compounds.

In another study, Karasali et al. [50] on 27 soil samples from Greece's central cotton fields, the occurrence and distribution of trifluralin, ethalfuralin, and pendimethalin was investigated. Trifluralin in 44% of the samples ($10\text{--}210 \mu\text{g kg}^{-1}$) and pendimethalin in 7% of the samples ($10\text{--}48 \mu\text{g kg}^{-1}$) were detected. Ethalfuralin was not detected in concentrations higher than the LOQ in any of the samples. These results were obtained while prior to the sampling time, ethalfuralin, trifluralin, and pendimethalin had not been applied to the land for 3 years, 1 year, and 3 months, respectively. This result reveals that some CUPs can be considered as

highly persistent in soil and require regular monitoring programs to help improve the pesticide-application system.

Aznar et al. [51] investigated the presence of ten pyrethroid insecticides in 33 samples taken in two sampling periods in plow and cultivation time and two sampling depths of 0–40 and 40–60 cm in a paddy-field area located on the Mediterranean coast (Valencia, Spain). Very high contamination was reported, especially during the rice-cultivation period, which was associated with the wastewater treatment plant used as the source of irrigation of the field. The contamination rate in the rice-production period was higher in comparison with the plow period. The highest contamination was reported for resmethrin and esfenvalerate with 62 and 57 $\mu\text{g kg}^{-1}$ in the rice-production and plow period, respectively. Unexpectedly, it was the subsurface soil layer that was more contaminated by pesticides, rather than the topsoil layer, as resmethrin, rifenthrin, fenpropathrin, ryfluthrin, lambda-cyhalothrin, alpha-cypermethrin, and esfenvalerate were detected in 97–100% and 3–100% of the subsoil samples during the cultivation and plow period, respectively. This is a very firm reason for monitoring soil not only in the plow layer but also in deeper layers of the soil.

Barchanska et al. [52] monitored mesotrione, sulcotrione, and atrazine in 24 agricultural and forest soil samples in Poland. Due the prohibition on atrazine imposed in Poland 7 years prior to the sampling time, it was stated that atrazine did not occur in the soil samples. However, its TPs were detected abundantly in soil samples: deethylatrazine was detected in 46% of soil samples (up to 180 $\mu\text{g kg}^{-1}$), deisopropylatrazine in 17% (up to 1,640 $\mu\text{g kg}^{-1}$), and hydroxyatrazine in 42% soil samples, but in concentrations lower than the LOQ. Other compounds which were detected in the soil samples were as follows: sulcotrione in 75% of samples with concentration up to 730 $\mu\text{g kg}^{-1}$ and its TP 2-chloro-4-(methylsulfonyl) benzoic acid (CMBA) in 37% of samples with concentration up to 60 $\mu\text{g kg}^{-1}$. Neither mesotrione nor its TPs were detected in soil samples. This study was also proof of the importance of monitoring TPs in the soil.

A very broad study on the distribution and occurrence of glyphosate and aminomethylphosphonic acid (AMPA) was performed by Silva et al. [29] on 317 topsoil samples from 11 European Union countries. The countries were chosen from among those with the highest percentage of agricultural area with different crops and the highest amounts of pesticide used per hectare. 21% and 42% of the soil samples contained concentrations higher than LOQ of glyphosate and AMPA in the range of 50–2050 and 50–1920 $\mu\text{g kg}^{-1}$, respectively. Northern soils were detected with higher DFs and southern soils with higher concentrations for both compounds. On the other hand, in eastern soils a lower frequency of glyphosate and in the southern soils a lower frequency of AMPA were detected. Portugal had the highest DF of glyphosate (53%) and Denmark the highest DF of AMPA (80%). Although no noticeable effect of the crop system was seen, the DF of glyphosate and AMPA was higher in areas under permanent and root crops (30 and 52%) and lower in dry pulses and fodder crops (5 and 29%). Off-site transportation of glyphosate and AMPA is claimed to be a matter of concern as they not only contaminate their surrounding area but can also find their way into all environmental compartments. This is alarming

and displays a need for further worldwide programs for monitoring agricultural soils in order to evaluate the occurrence and special distribution of glyphosate and AMPA.

The same soil samples which were subject to analysis for chlorotriazine herbicides by Scherr et al. [49], were analysed for the residue of other active ingredients (53 CUPs and 9 TPs) of different chemical groups by Hvezdova et al. [53] in 75 arable soils in the Czech Republic (CR) several months after the last pesticide application. Almost all the 68 soil samples (99%) were contaminated with at least one of the analysed pesticides or their TPs. Of the 68 analysed pesticides, the residue of 68% was detected in soil samples. Another 32% were absent. Conazole fungicides were the most frequently detected compounds (73% of the soils, up to $65 \mu\text{g kg}^{-1}$) in the region. Epoxiconazole (48% of the soils, up to $31 \mu\text{g kg}^{-1}$) and tebuconazole (36% of the soils, up to $28 \mu\text{g kg}^{-1}$), followed by flusilazole (23% of the soils, up to $19 \mu\text{g kg}^{-1}$), prochloraz (21% of the soils, up to $28 \mu\text{g kg}^{-1}$), propiconazole (13% of the soils, up to $12 \mu\text{g kg}^{-1}$), cyproconazole (8% of the soils, up to $23 \mu\text{g kg}^{-1}$), and difenoconazole (7% of the soils, up to $11 \mu\text{g kg}^{-1}$) were frequently detected in the soil samples. Chloroacetanilides such as S-metolachlor, metazachlor, or their TPs were other detected chemicals in 25% of the samples (up to $74 \mu\text{g kg}^{-1}$). The low-to-moderate water solubility, low pK_a , and high DT_{50} values reported for conazoles were in accordance with the high DF of these compounds. It is claimed that chloroacetanilides and their TPs represented short-term soil contamination due to very short half-lives in the soil and very high mobility. Detected pesticides related to other chemical groups were fenpropidin (20% of soils), diflufenican (17% of soils), urea herbicides (15% of soils), and carbendazim (11% of soils) with maximum concentrations of 62, 51, 92, and $21 \mu\text{g kg}^{-1}$, respectively. Chemicals like chlorpyrifos and pendimethalin, which are used in great amounts, were expected to be detected in the samples but were not. The reason was associated with the higher LOD of the analysing method which was reported as $10 \mu\text{g kg}^{-1}$.

Glyphosate and AMPA were assessed in a 3-year monitoring study in southern Greece [54]. A total of 170 soil samples from glyphosate-treated as well as non-treated and organic olive farms were analysed for residues. Sampling in conventional sites was done 13–276 days after pesticide application and the mean concentration of glyphosate and AMPA was 14.8 and $54.8 \mu\text{g kg}^{-1}$ and their DF was 13% and 63%, respectively. In non-treated and organic farms just one sample was contaminated by glyphosate and 11 samples contaminated by AMPA residues. The maximum concentration of AMPA was almost 2 times higher than glyphosate, 650 vs $350 \mu\text{g kg}^{-1}$, which indicates that AMPA's degradation was generally slower than its parent compound because it is more likely to be adsorbed to soil.

Silva et al. [30] analysed 74 other pesticides and TPs on the same European agricultural soils evaluated in their previous study [29]. Of all examined soil samples, 83% were contaminated with residues of at least one pesticide or TPs (concentrations $>\text{LOQ}$) and in 53% of the samples with multiple residues of pesticides and/or TPs. In total, 57% of the individual compounds investigated in this study were detected in the soil samples. However, 166 pesticide combinations were presented in the samples. Generally, the most frequently detected CUPs were

boscalid (27% up to $410 \mu\text{g kg}^{-1}$), epoxiconazole (24% up to $160 \mu\text{g kg}^{-1}$), and tebuconazole (12% up to $190 \mu\text{g kg}^{-1}$). More frequently detected multi residues were glyphosate + AMPA and glyphosate + AMPA + phthalimide, present in 2% of the samples. Of the pesticide residues detected in the soils, 60% were non-persistent compounds or moderately persistent compounds with DT_{50} less than 30 and 30–100 days, respectively. The soils from root crops and permanent crops had the highest contamination with pesticides, which was in accordance with the reported pesticide use in these crops.

In Northern Portugal, Bragança et al. [55] investigated eight pyrethroids in 18 conventional agricultural sites in two different sampling periods in summer and winter. Deltamethrin was the only pyrethroid detected in soil samples (8% of soil samples) with concentration up to $101.7 \mu\text{g kg}^{-1}$. It occurred only in the summer sampling.

Kosubova et al. [33] investigated 53 CUPs in 136 soil samples from 34 sites in Czech Republic. It was part of a 4-year monitoring program for the occurrence of 60 pesticides and four TPs across space and time. Of the analysed compounds, 19 were not detected in any of the samples mainly due to the ban or restrictions on their use prior to the sampling time or their short half-lives. These compounds included some important compounds such as atrazine and its TP desethylatrazine, acetochlor, metalaxyl-M, promethryn, and 2,4-D. On the other hand, more than 50% of the samples contained a mixture of 2–7 chemicals in 116 different multiple-residue combinations. In all the sampling years, atrazine TPs were present and detected in 44–71% of the soil samples which, it is claimed, was due to the previous applications on the sampling area. Current application of terbuthylazine products also caused contamination of atrazine, because atrazine can occur as an impurity in terbuthylazine formulations. This result again emphasizes that TPs may be more persistent than their parent compounds. Pendimethalin (up to $310 \mu\text{g kg}^{-1}$), diflufenican (up to $160 \mu\text{g kg}^{-1}$), tebuconazole (up to $140 \mu\text{g kg}^{-1}$), chlorotoluron (up to $90 \mu\text{g kg}^{-1}$), and linuron (up to $80 \mu\text{g kg}^{-1}$) were the compounds with the highest residue concentrations in the soil samples. High DFs in each sampling year were reported for epoxiconazole with a DF range of 57–62% (up to $32 \mu\text{g kg}^{-1}$) and tebuconazole with a DF range of 35–47% (up to $140 \mu\text{g kg}^{-1}$) and not in accordance with their rate of application to the soil. The occurrence of the banned pesticide carbendazim was attributed to its long half-life in soil (DT_{90} of up to 257 days in soil) and degradation of thiophanate-methyl.

In the upper part of the river Elbe, Germany, Karlsson et al. [56] investigated 20 soil samples from the floodplain area for pesticides of different chemical groups. The only detected CUPs were simazine (100%, up to $0.061 \mu\text{g kg}^{-1}$) and ethofumesate (30%, up to $23.25 \mu\text{g kg}^{-1}$), along with two TPs 2-hydroxy-atrazine (100%, up to $6.61 \mu\text{g kg}^{-1}$) and 2-hydroxyterbuthylazine (100%, up to $0.75 \mu\text{g kg}^{-1}$). The two parent compounds were also present in the water-monitoring database. Atrazine, known as a relatively persistent compound, was not found in the soil, while its main TPs were present. This was related to the periodical floods, which led to suspending or dissolving contaminations from the stream into the floodplain soil.

Ukalska-Jaruga et al. [57] examined 216 arable soil samples from Poland for the presence of organochlorine and non-chlorinated pesticides. Higher concentrations and DFs were reported for carbaryl and atrazine with values equal to 28.07 and 15.85 $\mu\text{g kg}^{-1}$ and 20 and 80%, respectively. Maneb was absent in all the soil samples. The total residue of non-chlorinated pesticides was reported to be up to 43.92 $\mu\text{g kg}^{-1}$.

In Belgium, soil samples from 18 fruit orchards (apple and cherry) were monitored for 70 active ingredients, including 35 insecticides, 22 fungicides, and 13 herbicides, of which 66% were detected in the soil samples [58]. Boscalid, carbendazim, and difenoconazole were detected in all soil samples (DF = 100%) with concentrations up to 10,990, 3,279, and 1,480 $\mu\text{g kg}^{-1}$, respectively. Other compounds frequently detected in the soil samples were tebuconazole (99%, up to 1,472 $\mu\text{g kg}^{-1}$), imidacloprid (97%, up to 112 $\mu\text{g kg}^{-1}$), linuron (97%, up to 1,280 $\mu\text{g kg}^{-1}$), diuron (89%, up to 225 $\mu\text{g kg}^{-1}$), and pyraclostrobin (86%, up to 203 $\mu\text{g kg}^{-1}$).

3.2 *Monitoring and Survey Studies in Asia*

Liu et al. [59] analysed the residues of 29 pesticides including CUPs in 46 soil samples from persimmon and jujube farms in China. Of 10 detected CUPs, the most frequently detected were fenpropathrin (50%, up to 400 $\mu\text{g kg}^{-1}$) deltamethrin (47%, up to 46.9 $\mu\text{g kg}^{-1}$), cyhalothrin (47%, 16 $\mu\text{g kg}^{-1}$), cypermethrin (45%, 44.7 $\mu\text{g kg}^{-1}$), bifenthrin (41%, 18.3 $\mu\text{g kg}^{-1}$), triadimefon (28%, 94.5 $\mu\text{g kg}^{-1}$), and chlorpyrifos (8%, 25.5 $\mu\text{g kg}^{-1}$). In this study, corresponding fruit samples were also analysed for the residues. Comparison showed that the DF of detected CUPs in soil was higher than in fruit samples. Multiple residues were frequently observed in soil samples, whereas up to 14 combinations were detected in some soils due to the higher persistence of compounds in the soil and secondary contamination from neighbouring areas.

In an article by Rafique et al. [60] high soil contamination with CUPs was reported. This study was carried out on 90 samples collected from cotton/wheat farmlands in Okara district, Pakistan. The main contaminations were imidacloprid and chlorpyrifos with DFs equal to 74 and 66%, and maximum concentrations equal to 1950 and 1993 $\mu\text{g kg}^{-1}$, respectively. MCPA (methyl ester) (29%, up to 1,531 $\mu\text{g kg}^{-1}$), deltamethrin (34%, up to 1,184 $\mu\text{g kg}^{-1}$), bifenthrin (20%, up to 884 $\mu\text{g kg}^{-1}$), and α -cypermethrin (49%, up to 774 $\mu\text{g kg}^{-1}$) were other CUPs with higher residue concentration.

In the same region studied by Liu et al. [59], 38 nut-planted soils of China were analysed for pesticides of different chemical groups by Han et al. [61]. The abundantly detected pesticides were pyrethroids, such as triadimefon (71%, up to 193.7 $\mu\text{g kg}^{-1}$), bifenthrin (63%, up to 156.3 $\mu\text{g kg}^{-1}$), cypermethrin (60%, up to 70.4 $\mu\text{g kg}^{-1}$), buprofezin (52%, up to 807.4 $\mu\text{g kg}^{-1}$), fenvalerate (47%, up to 884 $\mu\text{g kg}^{-1}$), and fenpropathrin (39%, up to 143.8 $\mu\text{g kg}^{-1}$). Chlorpyrifos was the

only detected organophosphorus pesticide in 5% of the samples up to $77.2 \mu\text{g kg}^{-1}$. Chlorpyrifos and fenvalerate were two pesticides widely used in the region according to local farmers. This was in accordance with their occurrence in the soil samples. Multiple-residue pesticides were detected in 73.7% of the samples.

Kailani et al. [62] reported that in monitoring 448 CUPs in 100 soil samples from southern Jordan, metalaxyl (37%, up to $2,660 \mu\text{g kg}^{-1}$), difenoconazole (24%, up to $1700 \mu\text{g kg}^{-1}$), imidacloprid (22%, up to $2,380 \mu\text{g kg}^{-1}$), and azoxystrobin (17%, up to $460 \mu\text{g kg}^{-1}$) were the most frequently detected CUPs. The compounds with the highest concentration were oxyfluorfen (2%, up to $6,490 \mu\text{g kg}^{-1}$), pyridaben (12%, up to $5,820 \mu\text{g kg}^{-1}$), and chlorfenapyr (12%, up to $4,990 \mu\text{g kg}^{-1}$).

Pan et al. [63] studied 530 soil samples from orchard and vegetable farms of a major agricultural area in North China for residue of 47 CUPs. The most frequently detected CUPs were tebuconazole with DFs of about 60%, then difenoconazole, chlorpyrifos, and thiamethoxam with DFs of approximately 25 to 30%. In concentrations above $100 \mu\text{g kg}^{-1}$, atrazine was the pesticide present in the highest number of soils (3.4%). Orchard fields were revealed to have the most contaminated soil (60% of the most contaminated locations). This proved higher pesticide use on orchards in comparison with other crops.

In central China, Pan et al. [64] collected 60 soil samples from two soil layers. Chlorpyrifos with a maximum concentration up to $5.58 \mu\text{g kg}^{-1}$ was the only CUP occurring in both layers. The high evaporation and water solubility of analysed CUPs were considered as the main reason for the obtained result.

Bhandari et al. [65] presented the concentration and distribution of residues of 23 pesticides analysed in 147 soil samples from IPM and conventional vegetable-growing lands in Gaidahawa rural municipality Nepal. Seventy-five per cent of conventional farms and 15% of IPM farms were found to be contaminated by pesticides. The lower contamination of IPM fields with pesticides was attributed to the efficiency of the IPM method, yet it was also stated that due to lower organic carbon content in IPM fields, chemicals were more likely to leach into the groundwater. It was stated that 15 pesticides and/or TPs in 39 combinations were present in the 60% of the soil samples, of which 60% were non-persistent or moderately persistent.

Pico et al. [66] did a study on the occurrence of 59 CUPs in the soil of lagoon wetland in Saudi Arabia. The region was said to be affected by wastewater discharge from agricultural activities. The residue of five compounds was detected in ten soil samples as follows: chlorpyrifos (100%, up to $0.84 \mu\text{g kg}^{-1}$), chlorfenvinphos (40%, $0.84 \mu\text{g kg}^{-1}$), fenitrothion (10%, up to $56.1 \mu\text{g kg}^{-1}$), carbendazim (10%, $0.04 \mu\text{g kg}^{-1}$), and imidacloprid (10%, $0.28 \mu\text{g kg}^{-1}$).

In the soil of 11 randomly collected soil samples from tea-plantation sites in the wet and dry season in Indonesia, Ariyani et al. [67] analysed the residues of five pyrethroids (lambda-cyhalothrin, permethrin, cypermethrin, fenvalerate, and deltamethrin) using a validated proposed method. The only detected compounds were permethrin and deltamethrin with DFs equal to 68 and 50% in all samples and a maximum concentration of 360 and $120 \mu\text{g kg}^{-1}$, respectively. Mostly, the concentration of permethrin was higher than deltamethrin in the soil samples and the

average concentrations of both were higher in the wet season. In the dry season no residue of deltamethrin was found, a fact which was associated with its shorter half-life in comparison with other pyrethroids.

3.3 Monitoring and Survey Studies in America

Bortolozzo et al. [68] monitored five CUPs in 108 soil samples taken at three depths from Ponta Grossa peatland. The major contaminants were atrazine (17%, up to $1 \mu\text{g kg}^{-1}$), chlorpyrifos (14%, up to $0.6 \mu\text{g kg}^{-1}$), and lambda-cyhalothrin (10%, up to $0.5 \mu\text{g kg}^{-1}$). In a deeper layer (over 30 cm) no residue of analysed CUPs was detected. The number of detections in the soil samples was 1.6 times more than water well samples. This was due to the high tendency of the analysed pesticides to bond to the soil's organic carbon.

In southeast of the Entre Rios Province, Argentina, Primost et al. [69] found that 100% of the soil samples collected from 17 agricultural farms had residues of both glyphosate and AMPA in concentrations up to 8,105 and 38,939 $\mu\text{g kg}^{-1}$, respectively. In this study, lower DFs were reported for surface-water samples in comparison with soil samples and no detection was reported for groundwater samples.

3.4 Monitoring and Survey Studies in Africa

The only study determining pesticide residues in African soils during the last 5 years was carried out in Sokoto state, Nigeria [70]. In the 17 number of soil samples, 13 organophosphate and organochlorin pesticides were analysed. It found that chlorpyrifos was the most abundant CUP, and it was detected in high concentrations in soil samples with a DF of 88% and maximum concentration of $2,870 \mu\text{g kg}^{-1}$. Dimethoate (59%, up to $1870 \mu\text{g kg}^{-1}$), dichlorvos (53%, up to $1,180 \mu\text{g kg}^{-1}$), mevinphos (53%, up to $1,000 \mu\text{g kg}^{-1}$), and methyl-parathion (53%, up to $1910 \mu\text{g kg}^{-1}$) were other CUPs with high DFs and concentrations.

4 Future Perspectives

According to the findings of this review, the level of soil contamination with CUPs in many regions is worrying. In some of them the situation is even worse, and countries still show a significant disparity in the inspecting strategies applied. Also, no significant improvement in the number of studies dedicated to the monitoring of CUPs in the soil has been observed after some time has passed. For instance, in Africa in which great amounts of CUPs were present, a lower number of monitoring studies have been performed during the last few years. Moreover, due to the

unknown complex behaviour of CUPs in different compartments and their ability to undergo long-range transportation, a wise response to CUP contamination would be regular monitoring of the soil not only in specific regions, but also at a global scale. The results also showed that under specific conditions, CUPs' properties in the soil differ of some expectations, i.e. a half-life longer than expected was reported for some compounds, which makes these chemicals more persistent and accumulative in the soil. Furthermore, the results presented that the degradation of CUPs such as atrazine led to TPs, which were even more persistent than their parental compounds. The results showed that monitoring of TPs in the soil is as important as tracking their parent compounds and must be included in monitoring programs. Clearly, there is insufficient research dedicated to monitoring CUPs in soil and there is an enormous need worldwide. As a result, future research is recommended to meet this need on the occurrence and fate of CUPs in the soil.

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