



Literature review and collection of existing data on PPPs' distribution and accumulation in ecosystems, plants, animals, and humans

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Acronymes

AM	Arithmetic Mean
CH	Switzerland
CR	Czech Republic
CSS	Case Study Site
DK	Denmark
EFSA	European Food Safety Authority
EPAH	Ecosystem, Plant, Animal, and Human matrices
ES	Spain
FAO	Food and Agriculture Organization of the United Nations
FOEN	Federal Office for the Environment
FR	France
GM	Geometric Mean
HBM	Human Biomonitoring
HBM4EU	Human Biomonitoring for Europe
HR	Croatia
IPCHEM	Information Platform for Chemical Monitoring
IT	Italy
LOD	Limit of Detection
LOQ	Limit of Quantification
MRL	Maximum Residue level
NL	Netherlands
NMP	National Monitoring Program
PPP	Plant Protection Products
PT	Portugal
SI	Slovenia
STDDEV	Standard Deviation
UNEP	United Nation Environmental Program
WFD	Water Framework Directive
WHO	World Health Organisation
WP	Work Package



1. General introduction

This deliverable is part of WP2 entitled "PPPs' distribution and health status". The aim of this deliverable is to provide an overview on the Plant Protection Product (PPP) concentrations measured in Ecosystem, Plant, Animal, and Human matrices (EPAH). This review will be the platform for the development of an inventory of PPP distribution and accumulation in EPAH (D2.3), for the development of an inventory of PPPs' exposure and validation of the model used in WP3 (D3.4), as well as for the development of health risk assessment toolbox (D5.1). In addition, the outcomes of this deliverable will feed into D6.2 - Health damages and external costs at the regional level.

The search of the publications was motivated by the collection of data allowing spatial mapping of pesticide concentrations.

The structure of this deliverable consists of providing EPAH PPP concentrations measured in each EU country represented in SPRINT by a CSS (Spain, Portugal, France, Switzerland, Italy, Croatia, Slovenia, Czech Republic, Netherlands, Denmark). The data collected in this review will complete the ones measured at the CSS (WP2), to refine and enhance the validation status of existing models for ecosystems and plants (D3.2).

In this deliverable, we considered the 207 active substances and metabolites being analyzed in the CSS samples. This list was strongly based on interviews with farmers conducted before the start of the sampling campaign – see Silva et al., (2021) for the complete PPP component selection process.

The search criteria for the data included into this review meant for mapping were as follows:

1. We only included Plant Protection Products (PPP) concentrations for the CSS relevant 207 substances.
2. We compiled and considered the data collected from National Monitoring Programs (NMPs) of each EU- SPRINT CSS country (if possible), or European publications (e.g. EFSA, FAO). If these were not available, or if the retrieved data were limited, we considered national monitoring reports, overview studies carried out at the national scale including IPCHEM documents, the HBM4EU database, and other overview publications found using our search strategy.
3. The search of the above reports was limited to reports published from 2010 and onwards except for soil (a SPRINT review covering the last 51 years period has been published alongside this document, Sabzevari & Hofmann, 2022). We believe that the recent overview reports contain the most relevant information for our purpose, since they are related to the currently used pesticides, and with recent approvals/regulations.



4. The PPP concentration in ecosystem, plants, animals and humans of the selected studies were preferably given as a mean value (arithmetic mean) of the point measurements (e.g. plot scale), but the median, geometric mean (GM), minimum and/or maximum concentrations were also provided if available.
5. If available, the coordinates of sampling location were provided for each measurement; if not, the name of the location (city, region, country) were provided to allow a more detailed (spatial) assessment. We included only disaggregated data (not averaged at large scale) except for animal and crop because of the very few articles found.

In the Description of Action of the SPRINT project, this deliverable was described as a scoping review. However, as three systematic reviews on PPP effects are being prepared within WP3 activities, we opted for a PPP distribution database compiling the NMPs and overview studies, and this for two main reasons: (i) to account reports written in local languages, and (ii) to complement the outcomes of WP3.

As expected, while preparing this deliverable we noticed that many publications dealt only with the methods of analysis, contained highly aggregated data (average values at the country level for instance), or were related to other countries than the ones we were considering in SPRINT. Therefore, this report is rather an inventory of the availability of data in terms of NMPs and overview studies of interest for mapping purposes. We present therefore a review of the available data on PPP distribution and bioaccumulation in the terrestrial, aquatic ecosystem, crops, animals and humans across Europe.

In this deliverable document, we provided summary tables and key findings of the compiled data; the compiled datasets are stored in the SPRINT sharepoint "<https://sprint-data.eu/>" and will be used as input data for WP3 and WP5, and scientific publications after a last update. In fact, as mentioned above, the first PPP distribution review article is already out, on the PPP distribution in soils on a global scale was published (Sabzevari & Hofmann, 2022).

2. Search of national monitoring programs and overview studies

We started by asking our 11 CSS leaders (ten in Europe, and one in Argentina) to provide us information on any ongoing or past NMPs on PPP exposure and other relevant overview studies with data on PPP concentration in EPAH in their respective countries. CSS leaders were asked to help translate the key parts if NMP was in their original language (e.g. Italian, Croatia, Slovenia, and Czech Republic).

For human, relevant literature from National Human Biomonitoring (HBM) programs on PPP concentrations were identified and extracted. Literature included HBM reports or published articles presenting national HBM results. Regional biomonitoring programs within CSS countries were also included if available. After the initial HBM search and once the HBM information was provided by the CSS leaders, we used PubMed (<https://pubmed.ncbi.nlm.nih.gov>) and Web of Science (<https://www.webofscience.com/>) electronic data base for the search.



The literature search was performed for all SPRINT CSS countries using the following key terms (all fields): **"pesticide residue"** OR **"insecticide residue"** OR **"fungicide residue"** OR **"herbicide residue"** OR **"pesticide concentration"** OR **"insecticide concentration"** OR **"fungicide concentration"** OR **"herbicide concentration"** OR **"plant protection product"** combined with **"Human"** AND the country of interest (including countries with an HBM and IPCHEM). With this strategy, we therefore proposed to include all different matrices such as soil, water, air, animal, crop, and human and related biological matrices. To provide the best overview on the available PPP measurements in human matrices we have included all relevant matrices (e.g. blood, urine, breast milk, placenta, and cord blood) with published PPP measurements irrespective of whether the matrices are included in the SPRINT campaign. Due to the very few publications found for animal and plant/crop, we used data of the latest EFSA report (Carrasco Cabrera and Medina Pastor, 2021).

For the inclusion / exclusion of the articles, we first used the filters available in the platform (e.g. article attribute, article type, publication date), second, checked their relevance by reading the titles and the abstracts, and finally, we consulted the full text of the ones retained. In this search, we excluded the non-European countries, aggregated data at large scale that are not of interest for mapping purpose, and the studies with very few data (e.g. few point measurements).

For each country, all potential eligible articles were screened by the title and the year of publication. The first step was an initial limited search of a selection of relevant databases, followed by an analysis of keywords contained in the title and abstract, and of the index terms used to describe the article. A second search based on a Full-text articles was then undertaken across all included databases. A few original articles were additionally included in the PPP literature according to suggestions of the CSS leaders/SPRINT experts, or initially gathered publications. The number of the articles found, and the ones included in this report are given in **Table 1**.

Table 1. Articles found considering our search strategy and the ones considered in this report.

Country	Platform	Articles found	Overview/ Review	Considered in this report																																																																																																																																					
				Soil	Air	Water	Plant/ Crop	Animal	Human																																																																																																																																
Spain	PubMed	3406(879)	379(168)	9 (+11)	1	2	2*	1*	7																																																																																																																																
	WOS	2535(861)	87(16)							Portugal	PubMed	1306(285)	118(62)	3	0	1	2*	1*	1	WOS	695(860)	42(16)	France	PubMed	3882(690)	320(102)	6	1	12	2*	1*	5	WOS	1992(861)	118(16)	Switzerland	PubMed	1390(274)	135(45)	3	1	8	2*	1*	1	WOS	679(860)	48(16)	Italy	PubMed	4739(1173)	493(247)	2	0	3	2*	1*	2	WOS	1850(860)	141(16)	Croatia	PubMed	285(76)	21(8)	2	0	2	2*	1*	3	WOS	182(860)	14(16)	Slovenia	PubMed	276(47)	14(10)	1	0	1	2*	1*	4	WOS	156(860)	13(16)	Czech R.	PubMed	1003(183)	95(34)	7	0	1	2*	1*	2	WOS	712(860)	35(16)	Netherlands	PubMed	1702(225)	168(29)	2	0	1	2*	1*	3	WOS	746(860)	56(16)	Denmark	PubMed	1145(329)	77(46)	1	0	1	2*	1*	3	WOS	559(860)	41(16)											
Portugal	PubMed	1306(285)	118(62)	3	0	1			2*		1*	1																																																																																																																													
	WOS	695(860)	42(16)							France			PubMed	3882(690)	320(102)	6			1	12	2*	1*	5	WOS	1992(861)	118(16)	Switzerland	PubMed	1390(274)			135(45)	3	1	8	2*	1*	1	WOS	679(860)	48(16)	Italy			PubMed	4739(1173)	493(247)	2	0	3	2*	1*	2	WOS	1850(860)			141(16)	Croatia	PubMed	285(76)	21(8)	2	0	2	2*	1*	3			WOS	182(860)	14(16)	Slovenia	PubMed	276(47)	14(10)	1	0	1	2*			1*	4	WOS	156(860)	13(16)	Czech R.	PubMed	1003(183)	95(34)	7	0			1	2*	1*	2	WOS	712(860)	35(16)	Netherlands	PubMed	1702(225)	168(29)			2	0	1	2*	1*	3	WOS	746(860)	56(16)	Denmark	PubMed			1145(329)	77(46)	1	0	1	2*	1*	3	WOS	559(860)	41(16)				
France	PubMed	3882(690)	320(102)	6	1	12						2*	1*	5																																																																																																																											
	WOS	1992(861)	118(16)							Switzerland					PubMed	1390(274)			135(45)	3			1	8	2*	1*	1	WOS	679(860)			48(16)	Italy	PubMed	4739(1173)			493(247)	2	0	3	2*			1*	2	WOS	1850(860)	141(16)	Croatia			PubMed	285(76)	21(8)			2	0	2	2*	1*	3	WOS	182(860)			14(16)			Slovenia	PubMed	276(47)	14(10)	1	0	1	2*	1*	4					WOS	156(860)	13(16)	Czech R.	PubMed	1003(183)	95(34)	7	0	1			2*			1*	2	WOS	712(860)	35(16)	Netherlands	PubMed	1702(225)			168(29)	2	0			1	2*	1*	3	WOS	746(860)			56(16)	Denmark	PubMed	1145(329)	77(46)			1	0	1	2*	1*	3	WOS	559(860)
Switzerland	PubMed	1390(274)	135(45)	3	1	8								2*	1*	1																																																																																																																									
	WOS	679(860)	48(16)							Italy									PubMed	4739(1173)			493(247)	2			0	3	2*			1*	2	WOS	1850(860)			141(16)	Croatia	PubMed	285(76)					21(8)	2	0	2	2*			1*	3	WOS			182(860)	14(16)	Slovenia			PubMed	276(47)	14(10)			1			0	1	2*	1*	4	WOS	156(860)			13(16)					Czech R.	PubMed	1003(183)	95(34)	7	0	1	2*	1*	2							WOS	712(860)	35(16)	Netherlands	PubMed	1702(225)	168(29)			2	0	1			2*			1*	3	WOS			746(860)	56(16)	Denmark	PubMed	1145(329)			77(46)	1	0			1	2*	1*
Italy	PubMed	4739(1173)	493(247)	2	0	3										2*			1*	2																																																																																																																					
	WOS	1850(860)	141(16)							Croatia													PubMed	285(76)			21(8)	2					0	2	2*			1*	3	WOS	182(860)					14(16)	Slovenia	PubMed	276(47)					14(10)	1			0	1	2*			1*	4	WOS			156(860)			13(16)	Czech R.			PubMed	1003(183)	95(34)			7					0	1	2*	1*	2	WOS	712(860)			35(16)							Netherlands	PubMed	1702(225)	168(29)	2	0	1			2*	1*	3							WOS	746(860)			56(16)	Denmark	PubMed	1145(329)	77(46)			1	0	1			2*		
Croatia	PubMed	285(76)	21(8)	2	0	2														2*			1*	3																																																																																																																	
	WOS	182(860)	14(16)							Slovenia																	PubMed	276(47)					14(10)	1					0	1	2*					1*	4	WOS	156(860)					13(16)	Czech R.			PubMed	1003(183)					95(34)	7			0			1	2*			1*	2	WOS			712(860)					35(16)	Netherlands			PubMed	1702(225)	168(29)			2							0	1	2*	1*	3	WOS	746(860)					56(16)							Denmark	PubMed			1145(329)	77(46)	1	0	1			2*	1*	3					
Slovenia	PubMed	276(47)	14(10)	1	0	1																		2*			1*	4																																																																																																													
	WOS	156(860)	13(16)							Czech R.																							PubMed	1003(183)					95(34)	7							0	1	2*					1*	2			WOS	712(860)					35(16)	Netherlands			PubMed			1702(225)					168(29)	2			0					1	2*			1*	3	WOS			746(860)							56(16)	Denmark			PubMed	1145(329)	77(46)					1							0	1			2*	1*	3	WOS	559(860)					41(16)					
Czech R.	PubMed	1003(183)	95(34)	7	0	1																						2*					1*	2																																																																																																							
	WOS	712(860)	35(16)							Netherlands																													PubMed	1702(225)							168(29)	2							0			1	2*					1*	3			WOS			746(860)					56(16)	Denmark			PubMed					1145(329)					77(46)	1			0							1	2*			1*	3	WOS					559(860)							41(16)																		
Netherlands	PubMed	1702(225)	168(29)	2	0	1																												2*					1*	3																																																																																																	
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Denmark	PubMed	1145(329)	77(46)	1	0	1																																		2*							1*	3																																																																																									
	WOS	559(860)	41(16)																																																																																																																																						



WOS: Web of Science; *including the EFSA report (Carrasco Cabrera and Medina Pastor, 2021) ; values in brackets are those of Human part; in the case of Spain, additional soil studies (11) are published, but raw data are not yet received.

The European countries considered are illustrated in **Table 2**. We agreed on excluding Argentina from this deliverable because of the limited data available at this stage.

Table 2. Cropping system and livestock types of SPRINT Case Study Sites, CSS

CSS (region, country)	Cropping system	Livestock
1. Cartagena (ES)	Vegetables (broccoli)	Sheep, goat
2. Bairrada (PT)	Vineyards	No
3. Bordeaux (FR)	vineyards	Pig, chicken
4. Central zone Switzerland (CH)	Orchards	Diary
5. Po region (IT)	Vegetables	No
6. Istria (CR)	Olives	Sheep
7. Central zone Slovenia (SI)	maize	Dairy and cattle for meat
8. Central zone Czech Republic (CR)	Oil plants	Diary, chicken
9. Groningen region (NL)	Potatoes	Dairy
10. Central zone, Denmark (DK)	Cereals	Dairy

3. Overview of the human biomonitoring programs available

Table 3. Overview of the human biomonitoring programs available in the 10 EU SPRINT case study site countries shows an overview of the human biomonitoring programs available in the 10 EU SPRINT CSS countries, an overview of the studies identified from which the concentrations of PPPs for the database have been extracted (mostly reviews), and other relevant monitoring programs.

Five out of ten SPRINT CSS countries (Spain, France, Switzerland, Slovenia, and Czech Republic) have current or recent human biomonitoring programs which include PPP surveillance in human samples. Of these five countries PPP concentrations were available for four. In Switzerland, data access negotiations are still in progress with the support of the Federal Office for Environment (FOEN).

Table 3. Overview of the human biomonitoring programs available in the 10 EU SPRINT case study site countries.

CSS N°	Human biomonitoring program	Overview studies	Other
1-ES	BIOAMBIENT.ES: Ramos et al., 2017	Govarts et al., 2012 Forns et al., 2018 Corcellas et al., 2012 Katsikantami et al., 2019	HBM4EU (data not publicly available yet)



	BETTERMILK, BIOVAL, CIPAV: Yusà et al., 2021	Yusa et al., 2015	
2-PT	No	No overview studies Original article: Cruz et al., 2003	HBM4EU (data not publicly available yet)
3-FR	ENNS (2010-) Saoudi et al., 2014 ELFE (2011-) Dereumeaux et al., 2016 ESTEBAN (2016-) (Results not published)	Forns et al., 2018 Govarts et al., 2012 Saillenfait et al., 2015	HBM4EU (data not publicly available yet)
4-CH	der Schweizer Gesundheitsstudie No data available yet	van den Berg et al., 2017	HBM4EU (data not publicly available yet)
5-IT	No	Katsikantami et al., 2019	HBM4EU (data not publicly available yet)
6-CR	No	No overview studies Original articles: Klinčić et al., 2016 Želježić et al., 2018 Jovanović et al., 2019	HBM4EU (data not publicly available yet)
7-SI	Slovenia HBM Report: Horvat et al., 2015 Bravo et al., 2020 Stajanko et al., 2020	Forns et al., 2018	HBM4EU (data not publicly available yet)
8-CR	CZ-HBM Černá et al., 2012 Mikeš et al., 2012	-	HBM4EU (data not publicly available yet)
9-NL	No	OBO report (Gooijer) Roze et al., 2009 (original article) Katsikantami et al., 2019	HBM4EU (data not publicly available yet)
10-DK	No	Knudsen et al., 2017 Dalsager et al., 2018 (original article) Dalsager et al., 2019 (original article)	DEMOCOPHES HBM4EU (data not publicly available yet)

Most studies with available PPP concentrations in human matrices were found for Spain (8 studies) and France (5 studies), with data for 25 different PPP substances extracted for Spain and data for 29 different PPP substances extracted for France (see Database). For Spain, there was data available for 14 of the 207 prioritized PPPs (covering 7%). The data for the 11 additional compounds not on the prioritized list but found in literature was also included in the database. This was considered to be of added value for non-SPRINT users. For France, PPP data was available for 11 of the 207 prioritized PPPs (4%) and 18 additional compounds. The countries with the least available literature on PPP concentrations were Portugal, Switzerland, and Italy (only 1 study identified for each country) with concentrations of 12 different PPP residues for Portugal (7 of the prioritized 207 PPPs and 5 additional, 1 for Switzerland (covering only the sum of DDT residues), and 2 for Italy representing only 2 compounds not on the prioritized list. Croatia was covered with data



for 5, Slovenia for 7, Czech for 6, Netherlands for 6, and Denmark for 4 of the prioritized PPP compounds.

The most well documented PPPs represented for most countries and in most studies were the insecticide residue pp-DDE and the fungicide Hexachlorobenzene (HCB). Glyphosate or AMPA (glyphosate main metabolite) concentrations were only available for three countries (France, Slovenia, and Denmark). As the best represented country (Spain) only covering 7% of the prioritized PPP compounds, there is a real knowledge gap regarding the risk exposure of humans to PPPs in the EU countries.

All SPRINT CSS countries are included in the EU human biomonitoring project HBM4EU, which is a joint effort of 30 countries to generate knowledge on safe management of chemicals (including PPPs) to protect human health in Europe. IPCHEM data are unfortunately not publicly available yet for any of the SPRINT CSS countries – but for future research this will provide a valuable tool and basis for more knowledge and understanding of the risk to which PPPs EU general populations are exposed.

A detailed overview table of literature and compounds included in the human database is given in **Table 4**. Overview of HBM data availability and studies per country with PPP measurements extracted to the human PPP database

Table 4. Overview of HBM data availability and studies per country with PPP measurements extracted to the human PPP database

CSS N	Bio sample	Year of Sampling	Sex	N	N PPP residues analyzed	Concentrations available for	Reference	HBM data
1-ES	Serum	2009-2010	Male and female	602 male, 332 female	13	HCB and pp-DDE	Ramos et al., 2017R	Yes bioambient.es
	Urine	2014-2020	Male and female	CIPAV n=125, BETTERMILK n=116, BIOVAL n=568	26	Atrazine, 2,4-D (free), Chlorpyrifos, beta-cyfluthrin, Deltamethrin, and Dimethoate	Yusà et al., 2021	Yes (regional)
	Cord blood and serum	2004-2008	Male and female	857 female serum and 1515 cord blood	1	pp-DDE	Govarts et al., 2012	No
	Infant blood	?	Male and female	1403	2	pp-DDE, HCB	Forns et al., 2018	No
	Breast milk	2009	Female	6	5	Tetramethrin, Bifenthrin, lambda-cyhalothrin, deltamethrin, Esfenvalerate/fenvalerate	Corcellas et al., 2012	No
	Urine	2012-2013	Male and female	116 men	7	DMP, DMTP, DMDTP, DEP, DETP, DEDTP, TCPY,	Katsikantami et al., 2019	No
	Urine	2010-2012	Male and female	125-184	16	3PBA, 4F3PBA, cis-DBCA, cis-DCCA, trans-DCCA, DEDTP, DEP, DETP, DMDTP, DMP, DMTP, DEAMPY, IMPY, PNP, 2,4-D, 3,4-DCA	Yusa et al., 2015	No
2-PT	Serum	2001-2002	Male and female	203	12	α-HCH, β-HCH and γ-HCH, Aldrin Dieldrin, HE, HCB, p,p'-DDT, o,p'-DDT, p,p'-DDE, p,p'-DDD, endosulfan sulphate	Cruz et al., 2003	No
3-FR	Serum	2006-2007	Male and female	132 men, 254 women	6	HCB, pp-DDE, pp-DDT, α-HCH, β-HCH and γ-HCH)	Saoudi et al., 2014	Yes ENNS (2010-)
	Urine	2011	Female	1036-1077	30	Atrazine, Atrazine mercapturate, Atrazine desethyl, Atrazine desisopropyl, Atrazine-desethyl-desisopropyl, Atrazine-2-hydroxy, Atrazine-desethyl-2-	Dereumeaux et al., 2016	Yes ELFE (2011-)



						hydroxy, Atrazine-desisopropyl-2-hydroxy, Atrazine-desethyl-desisopropyl-2-hydroxy, Glyphosate, Aminomethylphosphonic acid, Propoxur, 2-isopropoxy-phenol, 4-monochloro-phenol, 2,4-dichloro-phenol, 2,5-dichloro-phenol, 2,4,5-trichloro-phenol, 2,4,6-trichloro-phenol, Pentachloro-phenol, Dimethyl-phosphate, Dimethyl-thiophosphate, Di-methyl-di-thiophosphate, Di-ethyl-phosphate, Di-ethyl-thiophosphate, Di-ethyl-di-thiophosphate, 3-phenoxybenzoic, 4-fluoro-3-phenoxybenzoic acid, Cis-3-(2,2dibromovinyl)-2,2-dimethylcyclopropane-carboxylic acid, Cis-3-(2,2dichlorovinyl)-2,2-dimethylcyclopropane-carboxylic acid, Trans-3-(2,2dichlorovinyl)-2,2-dimethylcyclopropane-carboxylic acid		
	Infant blood	?	Male and female	188	2	pp-DDE, HCB	Forns et al., 2018	No
	Cord blood	2004-2008	Male and female	395	1	pp-DDE	Govarts et al., 2012G	No
	Urine	2006-2009	Male and female	369	5	3-PBA, Cis-DCCA, trans_DCCA, cis-DBCA, 4f3PBA	Saillenfait et al., 2015	No
								Der Schweizer Gesundheitsstudie No data available yet
4-CH	breast milk (pooled samples)	2005-2010	Female	>50	1	sum of DDT	van den Berg et al., 2017	No
5-IT	urine	2012-2013	Male and Female	124 men and women	7	DMP, DMTP, DMDTP, DEP, DETP, DEDTP, TCPY,	Katsikantami et al., 2019	No
6-CR	Breast milk	2011	Female	33	7	HCB, pp-DDE, pp-DDT, pp-DDD, α -HCH, β -HCH and γ -HCH)	Klinčić et al., 2016	No
	Placenta	2012-2013	Female	51	7	HCB, pp-DDE, pp-DDT, pp-DDD, α -HCH, β -HCH and γ -HCH)	Želježić et al., 2018	No
	Breast milk	2011-2014	Female	79	7	HCB, pp-DDE, pp-DDT, pp-DDD, α -HCH, β -HCH and γ -HCH)	Jovanović et al., 2019J	No
7-SI	urine	2016	mixed	164 children (7-8 years) and 164 mothers	10	DEAMPY, IMPY, MDA, PNP, TCPY, CMHC, 3-PBA, 4f-3-PBA, Σ OP, Σ PYR,	Bravo et al., 2020	Yes
	Breast milk, blood		Female	Slovenia 455	2	pp-DDE, HCB	Forns et al., 2018	No
	Breast milk and serum	2011-2014	Female	521 serum, 461 breast milk	2	Sum DDT, HCB for urine	Horvat et al., 2015	Yes Report in Slovenian
	Urine	2018	Male and female	82 girls (7-10 years), 67 boys (7-10 years), 43 female 12-15 years, 54 boys 12-15 years	2	AMPA and Glyphosate	Stajanko et al., 2020	Yes
8-CR	Breast milk	1994-2009	female	5667	4	pp-DDE, pp-DDD, pp-DDT, HCB	Černá et al., 2012Č	Yes
	Breast milk	1994-2009	female	5667		pp-DDE, pp-DDD, pp-DDT, HCB	Mikeš et al., 2012	Yes
9-ND	Urine	2016-2017	Male and female	n=1-16	4	Carbendazim, Chlorotoluron, Prochloraz, Tebuconazole	Gooijer et al., 2019	No



	Urine	2012-2013	Male and Female	100 pregnant women	7	DMP, DMTP, DMDTP, DEP, DETP, DEDTP, TCPY,	Katsikantami et al., 2019	No
	Serum	2001-2002	Female	62	1	pp-DDE	Roze et al., 2009	No
	Urine	2011-2012	Male and Female	27	1	Glyphosate	Knudsen et al., 2017	No
10-DK	Urine	2010-2012	Female	858 women geatational week 28	9	TCPY, 3-PBA, 2,4-D, DMTP, DMDTP, DEP, DETP, DEDTP	Dalsager et al., 2018	No
	Urine	2010-2012	female	948	6	TCPY, 3-PBA, 4-F-3PBA, cis-DCCA, trans-DCCA, cis-DBCA	Dalsager et al., 2019	No

An overview of the content of the human database is provided in **Table 5**. It describes the content of each column, name of the variable, the unit (if applicable) and any remarks related to the variables included in the database. Additionally, a literature overview is included in the first sheet of the database including first author, publication year, country, sample type, year of sampling, sex, number of participants considered, number of PPP residues analyzed, major groups reported on, and where to find the concentrations (database). Moreover, a "read me" sheet has been included with additional information on the database.

Table 5. Overview of the content of the plant protection product database related to humans

Column	Name	Unit	Remark
A	Pesticides analyzed	/	/
B	Herbicide/fungicide/insecticide	/	/
C	Location	/	Country, Region or City
D	Bio sample (type)	/	Urine, Serum, Cord blood, breast milk
E	Samples (n)	/	/
F	Sex	/	Male, Female, Male and Female
G	LOQ ^a	ug/L	/
H	% > LOQ	%	/
I	Age group	Years	/
J	AM ^b	ug/L	Arithmetic mean (in ug/L unless otherwise stated)
K	STDDEV ^c	ug/L	AM Standard deviation (in ug/L unless otherwise stated)
L	GM ^d	ug/L	Geometric mean (in ug/L unless otherwise stated)
M	GEO 95% CI lower	ug/L	Lower limit of 95% CI for Geometric mean (in ug/L unless otherwise stated)
N	GEO 95% CI upper	ug/L	Upper limit of 95% CI for Geometric mean (in ug/L unless otherwise stated)
O	Median	ug/L	50 th percentile (median) (in ug/L unless otherwise stated)
P	Max	ug/L	Maximum concentration measured (in ug/L unless otherwise stated)
Q	Year of sampling	Years	/
R	Analytical method	/	/
S	Source	1-3	Literature source : 1 = biomonitoring program, 2 = literature, 3 = provided by CSS
T	Reference	/	/
U	Monitoring program	/	Name of monitoring program cohort

^aLOQ: Lower limit of quantification, ^bAM: Arithmetric mean, ^cSTDDEV: Standard deviation (for AM), ^dGM: Geometric mean



4. Overview of the Ecosystem monitoring programs available

Data on measured PPP concentrations in the environments were separated in three environmental categories: soil, water and air. The data collected for these 3 environmental categories are given in three separate databases. We did not consider sediment category in this report since there are no NMP nor overview studies found in the CSS countries.

An overview of the content of the soil, water and air database is provided in **Table 7** describing the content of each column, the unit (if applicable) and any remarks for each column included in the database. This information can also be found in the "Read me" sheet of the database. A literature overview is included in the second sheet of the database including first author, publication year and country. Information regarding the dataset of the studies are given in column E to I. This information indicates if the dataset is included in the database, if sample coordinates are available, crop type given, and if further steps need to be taken to collect the dataset.

The ecosystem PPP database contains 12 sheets: (i) Read me, (ii) List of references, and (iii) the data of the 10 CSSs. In the list of references sheet, there are 10 columns.

Table 6: Overview of the content of Ecosystem related plant protection product database

Column	Name	Unit	Remark
List of references			
A	Abbreviation Reference	/	First autor et al. in the presence of co-authors
B	Reference	/	Complete reference
C	Country	/	Country of the study
D	Data available	/	Yes or no
E	Coordinates available	/	Yes or no
F	Crop type	/	
G	Data in database	/	If entered completely (x), partly (if x in column I), or not (empty)
H	Comments	/	Additional information
CSS list : 1_Spain to 10_Denmark			
A2	Reference	/	Abbreviation reference
A3	Sample_ID	/	/
A4	Crop type	/	If provided in the original report
A5 and following columns	Location	/	/
B7 and following columns	GPS coordinates	/	/
C4	Crop type	/	/
C5	Latitude	/	/
C6	Longitude	/	/
C7	NUTS 2	/	/



C8	Description location	/	/
C10-218	List of active substances	/	/
D and following columns	Concentration	ppm	/

4.1. Overview of available data of PPPs in soil

In the case of soil, no NMP is known to exist for any of the CSSs, except Switzerland. However, currently we don't have access to the data due to privacy data protection. The only reports we collected for all CSS are the overview studies, which are given in **Table 7**. In this case, we considered also the publication older than 2010 to complete the overview studies. Furthermore, a separate review article was published by Sabzevari and Hofman (2022) showing an overview of the studies on monitoring currently used pesticides in agricultural soils around the world published in the last 50 years.

Table 7. Overview of available NMP and overview studies – Soil (data are given in the database)

CSS N°	NMP	IPCHEM	Overview studies / significant studies	No. of samples/sites	Number of substances analyzed / Description
1-ES	n.a.	n.a.	Silva et al., 2019	30	76
			Geissen et al., 2021	48	38/75
			Calvelo Pereira et al., 2010	252 (4 locations)	1 (HCH)
			Hildebrandt et al., 2009	34 (13 locations)	22 organochlorinated pesticides & their degradation products 16 PAH
			Tadeo et al., 2004	18	8
			Bermúdez-Couso et al., 2007	26	6
			Jesús García-Galán et al., 2010	3	9
			Luis et al., 2010L	26	122
			Vallejo et al., 2019	-	20
2-PT	n.a.	n.a.	Geissen et al., 2021	9	47
			Silva et al., 2019	17	76
3-FR	n.a.	n.a.	Silva et al., 2019	30	76
4-CH ^a	n.a.	n.a.	a.d.	a.d.	a.d.
5-IT	n.a.	n.a.	Silva et al., 2019	30	76
6-CR	n.a.	n.a.	Herceg Romanić et al., 2016	5	15 organochlorine pesticides
7-SI ^b	n.a.	n.a.	Zupan et al., 2004	34	24
8-CR	n.a.	n.a.	Hvězdová et al., 2018	75 locations	69
9-ND	n.a.	n.a.	Geissen et al., 2021	10 fields	36
			Silva et al., 2019	30	76
10-DK	n.a.	n.a.	Silva et al., 2019	30	76

n.a.: not available; a.d.: aggregated data; ^ain the case of Switzerland, the detailed information on the locations is protected by the privacy data protection (we are in contact with the federal office for Environment); ^bdata available only as LOD and LOQ (we asked for the mean concentration and are waiting for the reply); in some cases, the number of samples considered was reduced because of the type of data (e.g. aggregated data)



The CSS country with the most studies available on PPP in soils is Spain. The data of 9 studies were given in a format suitable to be inserted in the database. The authors of the other studies with aggregated or protected data will be contacted again in order to complete the database.

In all the overview studies (see **Table 7**), 77 of the 207 compounds (screened in the SPRINT project) were found in soil above the LOQ. Glyphosate (herbicide), AMPA (one of the primary degradation products of glyphosate), boscalid (fungicide), DDE p,p' (breakdown product of DDT which is an insecticide), and dieldrin (insecticide) were the compounds measured in 50 % of the CSS countries (Spain, Portugal, France, Italy, Netherlands). Atrazine (herbicide), azoxystrobin (fungicide), DDT p,p' (breakdown product of DDT which is an insecticide), dimethomorph (fungicide), imidacloprid (insecticide), and tebuconazole (fungicide) were all measured in the soils of 4 of the 10 CSS countries. Together these are the 11 substances found in the highest amount of CSS countries.

In Croatia and Slovenia, the PPP measured in soils at the largest amount of locations were the degradation products of DDT (DDD o,p', DDD p,p', DDE p,p', DDE o,p', DDT o,p', DDT p,p'). For Czech Republic, terbuthylazine-desethyl (metabolite of terbuthylazine, which is an herbicide), epoxiconazole (fungicide) and atrazine were the compounds measured in soils at the largest amount of locations.

From all the studies considered in the soil database, the study by Silva et al. (2019) analyzed the largest amount of pesticide residues, namely 76 substances.

4.2. Overview of available data of PPPs in water

For water, there were some available NMPs at the country level. However, some issues made it difficult to store the data in an appropriate way since the results were given in minimum / maximum values or were not available for data-protection reasons (e.g., Switzerland) (**Table 8**).

Table 8: Overview of available NMP and overview studies – Water (data are given in the database)

CSS N°	NMP	IPCHEM	Overview studies	No. of samples/sites	Number of substances analyzed / Description
1-ES	n.a.	n.a.	Martínez Bueno et al., 2009	1	27 analysis in marine aquaculture
2-PT	n.a.	n.a.	Gonzalez-Rey et al., 2015	2 rivers, Arade and Arade Estuary rivers	20
3-FR	n.a.	n.a.	Miège et al., 2012	2 rivers and 1 marine site	9
			Ibrahim et al., 2013	1 river	8
			Kim Tiam et al., 2014	Up- and downstream of a river	13
			Mazzella and Coste, 2014	10 sites along 3 rivers	6



			Botta et al., 2012	4 sites along 3 rivers	92
4-CH	National Surface Water Quality Monitoring Program (NAWA) NAQUA National Groundwater Monitoring	n.a.	Moschet et al., 2014	5	248 substances and 134 transformation products. Only maximum measured value are given.
			Kiefer et al., 2019		>300 pesticides and > 110 pesticides transformation products were analyzed in groundwater
5-IT	Rapporto nazionale pesticidi nelle acque - dati 2017-2018	n.a.	Masiol et al., 2018	3 (8 sites with max concentration values)	114
			di Guardo and Finizio, 2018	1	1 (glyphosate)
6-CR	NMP by Croatian water agency for surface water and groundwater	n.a.	n.a.	10	55
7-SI	NMP of drinking water (http://mpv.si/rezultati)	n.a.	n.a.	45	>70 substances (26 in SPRINT substance list)
	NMP of surface water and groundwater by Slovenian Environmental Agency (http://mpv.si/rezultati and https://www.arso.gov.si/vode/podzemne%20vode/)				
8-CR	Hydrological Yearbook of the Czech Republic, 2004 to 2019 https://www.chmi.cz/informace-pro-vas/rocnivyhodnoceni/hydrologicke-rocenky	n.a.	Hydrological Yearbook of the Czech Republic, 2004 to 2019 Hydrological	54	Data are provided in form of maps or figures. The raw data are not accessible for public use
	Hydrological balance of water quantity and quality of the Czech Republic https://www.chmi.cz/aktualni-situace/hydrologicka-situace/podzemni-vody/hydrologicka-bilance		Balance of water quantity and quality of the Czech Republic		
9-NL	n.a.	n.a.	Desmet et al., 2016	11	2 (glyphosate & AMPA)
10-DK	GRUMO, the groundwater monitoring program (https://eng.geus.dk/water-resources/monitoring-programmes/groundwater-monitoring)	n.a.	-	-	Data are provided in form of maps or figures. The raw data are not accessible for public use

n.a.: not available

Switzerland and Slovenia have the most extensive dataset on PPP measured in surface water and groundwater.



For all studies included in **Table 8**, a total of 89 of the 207 compounds (screened in the SPRINT project) were measured in water above the LOQ. Atrazine was measured in the waterbodies of 6 CSS countries (namely, in Spain, Portugal, France, Switzerland, Croatia, Slovenia), followed by azoxystrobin in 4 CSSs (Portugal, France, Switzerland, and Croatia), and diuron (herbicide) in 4 CSSs (Spain, France, Switzerland, and Croatia). Glyphosate was measured in 3 CSSs (France, Italy, and Netherlands), and isoproturon (herbicide) in 3 CSSs (France, Switzerland, Croatia). Difenconazole (fungicide), dimethomorph, metolachlor (herbicide), propiconazole (fungicide), tebuconazole, and terbutryn (herbicide) were found in 2 CSS countries only. These substances are the 11 substances found in the highest number of the CSS countries.

In Croatia the degradation products of DDT (DDD o,p', DDD p,p', DDE p,p', DDE, o,p', DDT o,p', DDT p,p') and dieldrin were measured in 24 and 10 locations respectively, compared to atrazine, diuron and isoproturon which were measured in only 6 locations in Croatia. In the Netherlands both glyphosate and AMPA were measured at 10 locations in surface waters.

The Swiss NMP is the study that includes the largest amount of pesticides and its transformation products in its screening of both surface water and groundwater.

Out of a total of 111'062 surface water bodies in EU (WFD, 2000) 199 are not achieving good chemical status for Isoproturon, and 120 for Hexachlorocyclohexane (Kristensen et al., 2018). When taking into account the periodic occurrence of pesticides and a different analyte spectrum designed to reflect current pesticide use Weisner et al., 2022 reported higher concentration values in 91 agricultural streams in Germany.

4.3. Overview of available data of PPPs in air/dust

To our knowledge, only two countries have NMP of air/dust, France and Switzerland (Swiss plateau), while for Spain, there is an overview study covering 14 locations (**Table 9**).

Table 9. Overview of available NMP and overview studies – Air/dust (data are given in the database)

CSS N°	NMP / HBM	IPCHEM	Overview studies	No. of samples /sites	Number of substances analyzed / Description
1-ES	n.a.	n.a.	Ciemat, 2018: https://www.miteco.gob.es/gl/calidad-y-evaluacion-ambiental/temas/productos-quimicos/contaminantes-organicos-persistentes-cop/plan_nacional.aspx	14	11
2-PT	n.a.	n.a.	n.a.		
3-FR	Marlière et al., 2020 Résultats de la campagne nationale exploratoire de mesure des résidus de pesticides dans l'air ambiant (2018-2019) LCSQA	n.a.	n.a.	50 sites	75
4-CH	Schläpfer et al., 2021: https://www.bafu.admin.ch/bafu/de/home/themen/luft/	n.a.	n.a.	9 regions	83 including 10 metabolites



	publikationen-studien/studien.html				
5-IT	n.a.	n.a.	n.a.	-	-
6-CR	n.a.	n.a.	n.a.	-	-
7-SI	n.a.	n.a.	n.a.	-	-
8-CR	n.a.	n.a.	n.a.	-	-
9-NL	n.a.	n.a.	n.a.	-	-
10-DK	n.a.	n.a.	n.a.	-	-

n.a.: not available

In total, 33 of the 207 compounds, screened in the SPRINT project, were measured in air above the LOQ, for Spain, France and Switzerland together. Hexachlorobenzene (fungicide) was measured at 23 different locations in Spain. The three compounds measured at the most locations in France were lindane (insecticide), metolachlor (herbicide) and pendimethalin (herbicide). For Switzerland it was cyprodinil (fungicide) that was measured at the most locations.

Only limited number of studies were found for the measurement of PPP in air for all the CSS countries. The total number of substances analyzed in air for Spain, France and Switzerland was 11, 75, and 83 respectively.

4.4. Overview of available data of PPPs in crops/plants

In the case of crop/plant, aggregated data are of these of EFSA (Carrasco Cabrera and Medina Pastor, 2021) (**Table 10**). Chlorates, the most frequently exceeding their corresponding MRL was reported in broccoli (CSS 1, Spain) and in apples (CSS 4, Switzerland, see Table 9), followed by Chlorpyrifos (the 4th highest frequency) found in olives (CSS 6, Croatia), cereals (CSS 7, SI and CSS 10, Denmark), oilseeds (CSS 8, Czech Republic), and in potatoes (CSS 9, Netherlands) (Carrasco Cabrera and Medina Pastor, 2021). Dimethoate has the 9th highest frequency found in olives (CSS 6, Croatia).



Table 10. Pesticide residues per country and substances with values exceeding MRL and matching the crops of the CSS within SPRINT, the values given are the min and max concentrations

CSS n° - SPRINT (Values from crop origin)	Substance	Residue mg/Kg	Status of a.s end of 2019 EFSA report*
CSS 1 – SPAIN			
Vegetables (broccoli)	Chlorates	0.012 – 0.049	Not approved
	Chlorfenvinphos	0.092	Not approved
	Chlorpyrifos	0.78	Approved
	Dimethoate	0.37	Not approved
	Dithiocarbamates	1.8 – 3.1	Approved
	Prothioconazole	0.079	Approved
	Pyraclostrobin	0.13-0.141	Approved
CSS 2 – Portugal			
Grape for wine	Ametoctradin	8.2	Not approved
	Carbendazim	0.67	Not approved
	Chlorpyrifos	0.012	Approved
	Cyflufenamid	0.29	Approved
	Iprodione	0.011-0.02	Not approved
	Nicotine	0.014	Not approved
	Permethrin	0.059	Not approved
	Propiconazole	0.384	Not approved
	Pyrimethanil	6.2	Approved
	Thiophanate-methyl	5.4	Approved
	Triadimefon	0.016	Not approved
CSS 3 – France			
Grape for wine	See above (Portugal)		
CSS 4 – CH			
Apple	2-Phenylphenol	0.016-0.022	Approved
	Acephate	0.02-0.03	Not approved
	Azoxystrobin	0.011	Approved
	Chlorpropham	0.018	Not approved
	Chlorpyrifos	0.011-0.28	Approved
	Dimethoate	0.094	Not approved
	Dimethomorph	0.036	Approved
	λ-Cyhalothrin	0.218	Approved
	Permethrin	0.6	Not approved
	Tetramethrin	0.014	Not approved
	Zoxamide	0.023	Approved
CSS 5 – Italy			
Spinaches	1,4-Dimethylnaphthalene		
	2-Phenylphenol	0.017	Approved
	Acetamiprid	0.036	Approved
	Aclonifen	0.77	Approved
	Benzalkonium chloride	0.011	Approved
	Biphenyl	0.14	Not approved
	Captan	0.019	Not approved
	Carbendazim	0.055	Approved
	Chlorates	0.12-0.18	Not approved
	Chlorothalonil	0.11-0.6	Not approved
	Chlorpyrifos	2.012	Not approved
	Clomazone	0.013-4.5	Approved
	Clothianidin	0.013	Approved
	Cyfluthrin	0.011-0.12	Not approved
	Cyhalothrin	0.023-0.04	Approved
	Cypermethrin	0.037	Not approved
	Deltamethrin	1.4	Approved
	Dimethoate	0.013-0.51	Approved
	Dithiocarbamates	0.038-0.16	Approved
	Diuron	0.068-2	Approved
	Dodine	0.028	Approved
	Fenitrothion	0.13	Approved
	Fluazifop-P	0.014	Not approved
	Glyphosate	0.44	Approved
	Imidacloprid	0.12	Approved
	Indoxacarb	0.078	Approved
	λ-cyhalothrin	2.2	Approved
	Lenacil	1-1.3	Approved



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	Linuron Lufenuron Methomyl Nicotine Omethoate Phenmedipham Pirimicarb Propiconazole Prosulfocarb Pyraclostrobin Fluvalinate, tau- Tebuconazole Thiamethoxam Thiophanate-methyl	0.17-0.18 0.051 0.055-0.13 0.034 0.013 0.039-0.41 0.55-3.6 0.11-6.3 0.015-0.022 0.019 1.9-3.6 0.18 0.04 0.014 0.35-0.98	Approved Not approved Approved Not approved Not approved Not approved Approved Approved Not approved Approved Approved Approved Approved Not approved Approved
Sweet potatoes	2-Phenylphenol Chlordecone	0.011 0.028-0.038	Approved Not approved
Aubergines/eggplants	4-CPA Acetamiprid Acrinathrin Chlorates Dicloran Flonicamid Spromesifen	0.018 0.23-0.32 0.051 0.011-0.12 0.04 0.54 0.76	Not approved Approved Approved Not approved Not approved Approved Approved
Cherries (sweet)	Fenvalerate Fluopicolide Folpet Metalaxyl Omethoate Phosmet Prochloraz Propamocarb Spinosad Thiofanate-Methyl Triflumuron	0.023 0.045 0.067 0.013-0.025 0.29 1.2-1.3 0.096 0.5 0.3 0.36 0.044	Approved Approved Approved Approved Not approved Approved Approved Approved Approved Approved Approved
Tomatoes	Acetamiprid Acrinathrin Chlorates Chlorfenapyr Chlorpyrifos Cyflufenamid Cyhexatin Cypermethrin Deltamethrin Dimethenamid Dimethoate Dimethomorph Dinotefuran Ethephon Fenamiphos Flonicamid Flupyradifurone Folpet Formetanate(Hydrochloride) Fosthiazate Imazalil Imidacloprid Iprodione Metaflumizone Omethoate Phosmet Pirimiphos-Methyl Pyrimethanil Spiroxamine Tau-Fluvalinate Tetramethrin Triadimefon	0.8 0.117 0.011-0.376 0.017-0.27 0.196-2.0472 0.022 0.37 0.55 0.072 0.017 0.011-0.081 0.018 0.041 6.0-18.2 0.16 0.69-0.98 1.1 11.9 0.45 0.023 9.5 0.77 0.039 0.89 0.013-0.11 0.065-0.075 0.04-0,041 1.396 0.02-0.14 0.178 0.013-0.016 0.022	Approved Approved Not approved Not approved Approved Approved Not approved Approved Approved Approved Not approved Approved Not approved Approved Approved Approved Approved Approved Approved Approved Approved Not approved Approved Approved Approved Approved Approved Not approved Not approved Not approved
CSS 6 – Croatia			
Olives for oil production	Chlorpyrifos	24.8 ¹ 0.011-0.06	Approved



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	Cyfluthrin	0.023	Approved
	Dimethoate	0.014	Not approved
	Etofenprox	0.061	Approved
	Fluopicolide	0.037	Approved
	Proquinazid	0.028	Approved
	Pyraclostrobin	0.026	Approved
	Tebufozide (RD)	1.2	Approved
	Endosulfan	26.8 ¹	/
CSS 7 – SI			
Maize (Cereals)	Acetamiprid	0.014	Approved
	Chlorpyrifos	0.013	Approved
	Tricyclazole	0.015	Not approved
CSS 8 – Czech R.			
Oil plants	<i>See Olives for oil production</i>	/	/
Sunflower	Carbendazim	0.109	Not approved
Poppy	Carboxin (RD)	0.18	Approved
	Chlorpyrifos (RD)	0.062	Approved
	Chlorpyrifos (RD)	0.032	Approved
	Picoxystrobin (RD)	0.018	Approved
	Propiconazole	0.012	Approved
Oilseed rape	2-phenylphenol	0.07	Approved
	Chlorpyrifos	0.01	Approved
mustard	n.d.	n.d.	n.d.
CSS 9 – Netherlands			
Potatoes	Chlorantraniliprole	0.083-0.24	Approved
	Chlorates	0.073	Not approved
	Chlorpropham	13	Not approved
	Chlorpyrifos	0.012-9.5	Approved
	Clothianidin	0.072	Not approved
	Cyprodinil	0.204	Approved
	Fipronil	0.02	Not approved
	Flonicamid	0.11-0.25	Approved
	Fosthiazate	0.03-0.16	Approved
	Indoxacarb	0.029	Approved
	Lufenuron	0.12-0.52	Approved
	Pirimiphos-methyl	0.012	Approved
	Propiconazole	0.768	Not approved
	Pyrimethanil	0.874	Approved
	Thiabendazole	0.21-0.77	Approved
CSS 10 – Denmark			
Wheat	Carbaryl	0.6-1.6	Not approved
	Chlorates	0.03-0.1	Not approved
	Chlorpyrifos	0.061-0.19	Not approved
	Metalaxyl	0.02	Approved
	Simazine	0.038	Not approved
	Tetramethrin	0.018-0.33	Not approved
Barley	Chlorates	0.026-0.079	Not approved
	Dikegulac	0.011-0.015	Not approved
	Permethrin	0.066	Not approved
	Pirimiphos-Methyl	5.6	Approved
	Spiroxamine	0.07	Approved
Oats	MCPA & MCPB	0.38	Approved

*EFSA report (Carrasco Cabrera and Medina Pastor, 2021); names in brackets are these reported in EFSA report; the concentration values are represented as minimum and maximum; n.d.: not determined (not considered in the analysis of EFSA); we consider the products found in the European countries

¹Amvrazi and Albanis, 2009



4.5. Overview of available data of PPPs in animal products

There was no NMP found by the CSS leaders at the national level. Therefore, we used data provided in the report of EFSA (Carrasco Cabrera and Medina Pastor, 2021) related to animal products only since no data on biological matrices are given.

DDT, hexachlorobenzene, beta-hexachlorocyclohexane and dieldrin were the most frequently quantified substances (at or above 10 samples) found in animal products since these are persistent pesticides in the environment (**Table 11. Pesticides** most frequently quantified in animal products (in absolute at or above 10 number of determinations); EFSA report (Carrasco Cabrera and Medina Pastor, 2021))

Chlorate, benzalkonium chloride and didecyldimethylammonium chloride were reported mainly in milk and muscle (Carrasco Cabrera and Medina Pastor, 2021). Their presence can be attributed to residues in treated potable water (used as ingredient and/or in cleaning equipment) and to chlorine-based solutions used in the manufacturing process. However, since these substances have been used as pesticides in the past, they still fall under the scope of the pesticide MRL regulation.

Table 11. Pesticides most frequently quantified in animal products (in absolute at or above 10 number of determinations); EFSA report (Carrasco Cabrera and Medina Pastor, 2021)

Pesticide	Eggs	Animal fat*	Animal kidney	Animal liver	Milk	Muscle	Other animal products	Total
Thiacloprid	0	0	0	0	0	0	0	173
DDT	31	24	4	23	42	10	26	160
Hexachlorobenzene	18	21	2	10	61	21	3	136
Acetamiprid	0	0	0	0	0	0	0	49
Mercury	1	0	11	0	18	1	7	38
Amitraz	0	0	0	0	0	0	0	37
Fipronil	23	8	0	0	0	0	0	31
Benzalkonium chloride	0	1	0	1	21	5	0	30
Dimoxystrobin	0	0	0	0	0	0	0	29
Chlorates	1	0	0	0	8	18	0	28
Hexachlorocyclohexane, beta-	0	0	0	23	2	1	2	28
Azoxystrobin	0	0	0	0	0	0	0	27
Pendimethalin	0	0	2	21	0	0	0	26
Diazinon	0	6	17	1	0	0	0	24
Dieldrin	2	10	0	6	1	1	0	20
Didecyldimethylammonium chloride	0	1	0	3	7	5	3	19
Glyphosate	0	0	0	0	0	0	0	17
Chlorpyrifos	0	0	14	0	0	0	0	15
Bromide Ion	9	0	0	0	3	0	0	13
Fosetyl	0	10	0	0	0	0	0	12
Coumaphos	0	0	0	0	0	0	0	10
Fonicamid	0	0	0	0	0	0	0	10

Products involves different animal species; Source: (Brancato et al., 2017)

In EFSA report (Carrasco Cabrera and Medina Pastor, 2021), 1,301 samples of honey and other apicultural products were analysed. In 265 samples (20.4%) residues at or above the LOQ but below or at the maximum residue level (MRL) were identified. Overall, 27 different pesticides were quantified. The pesticides most frequently reported in honey and other apicultural products above the LOQ were thiacloprid (173 samples), acetamiprid (49



samples), amitraz (37 samples), dimoxystrobin (29 samples), azoxystrobin (27 samples), glyphosate (17 samples), coumaphos (10 samples) and flonicamid (10 samples).

Chlorpyrifos was reported mainly in animal kidney. Its presence is likely to be due to a carryover of its use in feed. However, it is important to track this finding due to its potential genotoxicity (Rodríguez-Cortez and Menendez, 2020) and the welfare of animals.

5. Summary

In all overview studies, a total of 77 of the 207 compounds, screened in the SPRINT project, were measured in soil above the LOQ and the CSS country with the most studies available on PPP in soils was Spain.

Considering the entire dataset collected for water, 89 of the 207 compounds, were measured in water above the LOQ. Switzerland and Slovenia had the most extensive dataset on PPP measured in surface water and groundwater.

For Spain, France and Switzerland, 33 of the 207 compounds, screened in the SPRINT project, were measured in air above the LOQ. The total number of substances analysed in air in these three countries was 11, 75, and 83, respectively.

In the case of crop/plant, we used aggregated data of EFSA (Carrasco Cabrera and Medina Pastor, 2021). Since we cannot use such data for mapping, we tried to link the crops/plant to the CSSs where the type of crops exists. Chlorates, the most frequently exceeding their corresponding MRL was reported in broccoli (CSS 1, Spain) and in apples (CSS 4, Switzerland), followed by chlorpyrifos found in olives (CSS6, Croatia). The tested soils from root crops and permanent crops presented the highest pesticide contents, which is in line with the reported intensive pesticide use in these crops (Silva et al., 2019; Muthmann, 2007).

DDT, hexachlorobenzene, beta-hexachlorocyclohexane and dieldrin were found the most frequently in animal products since these substances are persistent pesticides in the environment. Their presence can be attributed to residues in treated potable water (used as ingredient and/or in cleaning equipment) and to chlorine-based solutions used in the manufacturing process. They may also result from other sources, e.g. CS₂ measured as a residue from dithiocarbamates also occurs naturally in some plants, particularly in Brassicaceae (e.g. broccoli, cauliflower) and Alliaceae (Carrasco Cabrera and Medina Pastor, 2021).

In the case of humans, the most well documented PPPs represented for the most countries and in most studies were the insecticide residue pp-DDE and the fungicide Hexachlorobenzene (HCB). Glyphosate or AMPA concentrations were only available for three countries (France, Slovenia, and Denmark). Given the small number of publications available in the case of humans, a consistent synthesis was difficult to carry out.



The studies found in the literature on the concentrations/residues of detected pesticides in EPAH, and the locations of their point measurements are hardly comparable, because of the following:

- The investigations were conducted in distinct regions with specific portfolio of pesticides following the regional crop production and plant protection products approved by regional regulatory agencies.
- The use of different LOQs and other survey parameters.
- The studies did not always report on the conditions of the selected sites in terms of cropping system and agricultural management practices.
- The majority of the studies found in the literature focused primarily on the development of analytical procedures and their calibration and validation, and less focus on soil and crop characteristics.
- Finally, time and year of sampling that is different from study to study.

The above considerations strongly biased the comparison between the studies and the conditions of the experimentation used, and consequently the comparison between the concentrations of pesticides found.

6. Gaps and steps forward

In the case of some CSSs, we encountered three main difficulties, (i) limited data / NMPs, (ii) existing of aggregated data (or only maps without data) not appropriate for our purpose (mapping), and (iii) the data are not accessible for public use (e.g. NMPs). In addition, in some cases, the reports are in the local languages and no English versions are available. These considerations make it difficult to come up with updated maps with complete data for a region and for a given aspect since the data were not provided in same format. Furthermore, we checked the data availability presented in this report (NMPs) with the corresponding CSS leaders before the final publication.

It would be of great interest for the SPRINT project to ask the national institutions to deliver missing raw data to be used within the project. The corresponding CSS leaders could start negotiation for this purpose with the support of WP2 and the coordination team.

We plan to update the actual database that will include both data gained from the literature and the one measured at the CSSs with the aim of serving as basis for a scientific publication. It will also serve for the following tasks (e.g., D2.3 – Inventory of PPP distribution and accumulation in EPAH; and D2.4 – inventory on EPAH health status in CSS).



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