

Review

Persistent Organic Pollutants (POPs): A Review Focused on Occurrence and Incidence in Animal Feed and Cow Milk

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Abstract: Persistent organic pollutants have particular ecotoxicological importance and they are amongst the most harmful groups of persistent pollutants. The complexity of persistent organic pollutants highlights the different sources of pollution from which they came and, depending on which, their profile could be characterized. In the first part of this review, the main characteristics of persistent organic pollutants were described, focusing on their complexity and toxic potential in relation to environmental elements. The second part of the review includes data related to the occurrence and incidence of persistent organic pollutants in different types of feed and cow’s milk, focusing on the characteristic profile of pollutants as an indicator of the sources of pollution. Moreover, a description regarding the timing and duration of the contamination of feed and milk was carried out, evaluating the distribution of pollutants within the analyzed samples and highlighting those whose presence is predominant or whose residues persist in the environment for long periods. The review concludes that the identification of pollution sources associated with different proportions of organic pollutants found in different samples could represent a suitable solution for biomonitoring the potential contamination in a geographical area.

Keywords: persistent organic pollutants; feed; milk; environment; pollution



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1. Introduction

Environmental pollution represents a threat to human health, quality of life and the natural function of ecosystems [1]. Anthropogenic actions are the main causes of environmental pollution. Whether intentional or accidental, humans can generate various pollutants with direct or indirect negative impact on the ecosystem elements, such as the soil, water, air, plants, animals and even the human body [2,3].

Environmental pollution has been widely studied and the most important properties of the most prevalent pollutants were reported. The organic pollutants are qualified among the most harmful pollutants due to the eco-toxicological risks and their slow degradability; hence, their persistence [4,5].

Recent increases in industrial and technological activities have led to high emissions of pollutants. Generated from various developing productive sectors, persistent organic pollutants (POPs) have become a real threat for modern society due to their negative effects on the environment, animal welfare, health and the safety of animal products.

Synthesized mostly by anthropogenic sources, POPs are halogenated, lipophilic, organic compounds, with a very stable chemical structure, which are resistant to photolytic and biological degradation and which have a negative impact on the environment, flora, fauna and human health [5–7].

The character of POPs facilitates their accumulation in fatty tissues [8]. Most of the studies have focused on the biological behavior of POPs due to their ability to transfer and accumulate along the food chain [9,10] through bio-amplifying and volatilization, ending in more dangerous threats [11,12], especially if slow metabolic conditions occur [5,6,8,11,13–15].

Lately, high amounts of pollutants were released into the environment and have contaminated the natural substrates and the food chain [16]. A strong network of hazards was established between pollution, vegetal production, animal production and human consumers due to the possibility of pollutants' transfer along the food chain [17–20]. Food samples that have been tested were shown to be contaminated with several types of POPs [21]. Animal-originated food as one of the main vehicles of human exposure to POPs contamination [22–24].

Feed is particularly implicated in the transfer and occurrence of such contaminants in food, especially in dairy products. Studies of POPs in the food chain have recently focused on the correlations between feed production and food safety issues [25–28] or POPs contamination and effects on the human body [29–31]. In recent years, most research has included a study of the transfer of pollutants from feed to animals, and their accumulation and excretion in milk [32].

Considering the potential negative effects of pollutants in feedstuffs on animal yields and, subsequently on public health, assessing POPs contamination level from animal feed is really important to the safety of consumers [12,19,33–39].

This review aims to describe different classes of POPs as well as to discriminate between specific sources of pollution as it relates to the concentrations of POPs in feed and milk. Thus, the sources of POPs were analyzed, as were the contamination and the proportions of the most prevalent pollutants, as well as the methods used in their monitoring and quantification.

2. Characterization of POPs

Regulations addressing the persistent organic pollutants have aimed at reducing the consequences of pollution, eliminating existing pollutants or preventing potential contamination [40]. According to the European Chemical Agency [41], at least 12 types of organic substances, the so called “dirty dozen”, were included, in 2001, under the Listing of POPs in the Stockholm Convention (Table 1). The Convention aimed to protect human health and the environment, especially by safely eliminating, reducing the production and decreasing use of the 12 dangerous substances and was signed, in the beginning, by 115 states. Currently, the Stockholm Convention is signed by 152 states and ratified by 184 states [41]. The list of banned or restricted organic pollutants currently reaches about 28 hazardous chemicals [42] and includes three categories of POPs distinguished by their originating anthropogenic process [12,40,43].

The most popular group of POPs is represented by the synthetic chemicals produced for agricultural usage in particular, which include organochlorine pesticides (OCPs) such as aldrin, dieldrin, dichloro-diphenyl-tetrachloroethane (DDT), heptachlor, mirex, chlordane, toxaphene and endrin. Some of them can be also used in dyes or plastics [5,44].

The second group of POPs refers to chemical compounds commercially produced for various industrial applications [45] or released from the chemical industry as secondary substances [46], including hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), perfluorinated compounds (PFCs) or brominated compounds (BFRs) [5,45,46].

Table 1. Characteristics of POPs listed in the Stockholm Convention.

Category	Type of POPs	Description	Source of Pollution	Ref.
 OCPs	Aldrin	Quickly converted to dieldrin; Low toxicity to plants and high toxicity to animals and humans; contamination in dairy products and meat	Industrial activities Agricultural activities	[5,44]
	Dieldrin	High concentrations (transformation of aldrin into dieldrin); residues frequently found in air, water, soil, in the bodies of birds, mammals or humans (exposed through food, especially dairy products and meat); $t_{1/2} = 5$ years		
	Chlordane	Broad spectrum of action for crops; contamination by air; $t_{1/2} = 1$ year		
	DDT	$t_{1/2} = 10$ years (more than 50 % of the initial amounts can remain in the soil after 10–15 years after application)		
	Endrin	No accumulation in fatty tissues (compared to other organic pollutants), toxicity especially for aquatic animals; $t_{1/2}$ in soil = 12 years	Industrial activities Agricultural activities	
 Industrial chemicals	Mirex	$t_{1/2} = 10$ years		
	Toxaphene	$t_{1/2}$ in soil = 12 years		
	Σ PCBs	Includes 209 different types of PCBs, of which 13 substances have particularly high toxicity, formed as a result of incomplete combustion from various industrial processes	Waste chemicals, plastic and rubber products or electrical equipment	[9,45–49]
 Combustion chemicals	HCB	Residues of the pesticide production; residue of incomplete combustion	Chemical waste	[5]
	PCDDs / PCDFs	Highly toxic chemical compounds originating from industrial processes including ~210 polychlorinated aromatic chemical compounds: PCDDs/dioxins and PCDFs/furans $t_{1/2} = \sim 7$ years; the possibility of accumulation in animal and human tissues through contaminated food; low concentrations in the environment, but persistent and easily transferred	Chemical and industrial processes (herbicides/pesticides production, metalworking) Combustion processes (waste incineration) Natural events (volcanic eruptions, wildfires) Involuntary emissions from burning plastic, wood waste or agricultural waste contaminated with pesticides Vehicle emissions	[5,34,37,50,51]
	DL-PCBs	Include 12 compounds out of 209 types of PCBs, which are formed in combustion processes and have toxic properties and potential effects similar to PCDDs/PCDFs	Incomplete combustion (natural and anthropogenic) Incineration processes: Distillation of coal Emissions from transport Wood/forest vegetation burning Volcanic eruptions Oil exploitations Biomass burning	[52]
	Σ PAHs	Organic compounds with two or more condensed aromatic cores, with specific structures and variable toxicity		[5,38,53–55]

POPs—persistent organic pollutants; OCPs—organochlorine pesticides; DDT—dichlorodifeniltricloroetano; HCB—hexachlorobenzene; PCBs—polychlorinated biphenyls; PCDDs/PCDFs—polychlorinated dibenzodioxins/dibenzofurans; DL—PCB—dioxin-like PCBs; PAHs—polycyclic aromatic hydrocarbons; $t_{1/2}$ —half time. See reference [40] for POPs category symbol shape.

Another group of chemical compounds that are formed from incomplete combustion of organic matter represents the third category of POPs: dioxins/polychlorinated dibenzodioxins (PCDDs), furans/polychlorinated dibenzofurans (PCDFs) or polycyclic aromatic hydrocarbons (PAHs) [5,34,37,38,50–55].

Because of their toxic effects, 33 PAHs were described by the European Scientific Committee on Food as having a high risk for the environment. Moreover, Shafy and Man-

sour [54] focused their attention on some PAHs with higher toxic potential. Together with the Canadian Council of Ministers of the Environment [56], they classified 7,12 dimethylbenzoanthracene (DMBA) and benzo(a)pyrene (BaP) as two of the most toxic POPs.

Considering the increasing of amounts organic pollutants in the environment, several authors have focused their attention on describing the profile of POPs. Table 1 summarizes their works, highlighting the characteristics of the main categories of OCPs, the characteristics of chemical substances from industrial processes and combustion processes and also the specific sources of pollution identified in analytical studies. Manciulea and Dumitrescu [5] have classified the POPs into three categories according to the source of pollution; the three categories of POPs mentioned by these authors [5] include the same types of pollutants listed in the Stockholm Convention [12,40,43]: the OCPs, the industrial chemical pollutants and the combustion chemicals.

OCPs are persistent organic pollutants with broad spectrum of action, high chemical stability and toxic characteristics [57]. The characteristics of OCPs were underlined in a work conducted by Rusu et al. [35], who identified residual amounts of pesticides in former areas producing OCPs, but without current productive activity (production available up to 1990s). Thus, the occurrence of OCP residues in samples, in the absence of recent contamination, highlighted their long-lasting half-life. Other authors have also observed the presence of OCP residues in the environment even after their use was reduced or was even completely banned [58–60].

Amongst the causes of pollution, agriculture, together with the industrial production of agricultural substances or plastics, paints, rubbers or electronics, are the main sources of some of the most toxic OCPs [5,44] (Table 1). Recent data presented by Chavoshani et al. [61] have confirmed that intensive agriculture, over time, has negative effects on the environment. Even if, nowadays, many of the OCPs have been banned or limited, it is important not to forget about the contamination that has existed since different OCPs were permitted.

Almost two decades since the Stockholm Convention prohibited or limited the OCP usage in agriculture, studies are still being carried out to understand their residual state and the possible risks that may appear on the environment. Khuman et al. [59] reported a general trend of decreasing levels of OCPs in urban, agricultural and industrial soils. However, the study showed that historical OCPs and their breakdown products remain a threat to the environment; therefore, the agricultural activities are still included in the category of polluting factors.

The industrial chemical pollutants group of POPs includes chemical compounds such as HCBs or PCBs, i.e., pollutants that can be generally issued as residues from plastics production, but also residues from pesticides or other chlorinated organic compounds. In some cases, HCBs or PCBs occur as a consequence of incomplete combustion of chemical waste, rubber production, electrical equipment production or other industrial processes [5,45,62,63]. Chaukura et al. [64] stated that microplastics occurring in the environment as residues of various industrial processes are also important carriers of POPs, especially of OCPs, PCBs or PAHs.

PCBs are a group of chemical compounds included in the Stockholm Convention among the 12 POPs in the “dirty dozen” category [10]. PCBs can be released from electrical equipment or construction materials [9,65,66], but they can also be released from various industrial processes [67,68]. To limit the negative effects on the environment, the intentional production of PCBs for industrial application has been limited or even banned in some countries [10,69]. The environmental negative impact of PCBs persists through the products already manufactured before the ban in the form of polluting waste [70]. With respect to the negative contribution of PCBs from various industrial processes to environmental pollution, no clear measures to control were found.

For humans, PCBs are hazardous due to their high capacity for contaminating the food chain. Although they can also enter the human body through inhalation or dermal exposure [71], foodstuffs seem to be the main route for human exposure [72].

HCb is a chemical compound with persistent organic characteristics [73], whose conscious production and use have been limited or banned in most countries. However, HCB can be produced unintentionally in the processes of incomplete combustion of various chemical substances [74]. According to Starek-Swiechowiec et al. [75], HCB has high bioaccumulation potential especially for living organisms. Different amounts of HCB residues were detected in various substrates, such as fodder crops, in food (milk, fish, vegetables, meat), in blood or in adipose tissue [75,76].

Regarding combustion chemicals, they are highly toxic chemical compounds generated by incomplete combustion from industrial processes. According to Table 1, the characteristics of these POPs have been mentioned in various works [5,38,53–55].

3. Occurrence and Incidence of POPs

3.1. Milk

Due to its nutritional qualities, milk is one of the most important animal products [77–79]. The contamination of milk with POPs has been studied by various authors [80–83]. The literature describes pollutant profiles related to the pollution sources in some areas, the relationship between some pollutants categories and the associated pollution sources, the quantitative profile, the description of the methods and analytical techniques used or the investigation of the associated risk category (Table 2).

Table 2. Summary of POPs presence in milk and sources of pollution.

Location	Collected Area	Sample	POPs	Method	POPs Level (ng/g) Mean and/or Range (Min–Max)			Annex *	References	
										
Source of the milk: FARM										
France	Vicinity of a hazardous municipal waste incinerator	Raw milk collected from animals exposed to a 10-week long-term intake of contaminated hay	ΣPCBs		GC–HRMS	-	2.22–3524.07	-	A, C	[84]
			PCDDs/TCDD			-	-	0.48	C	
			PCDDs/OCDD			-	-	2.31	C	
			PCDFs/TCDD			-	-	0.52	C	
			PCDFs/OCDD			-	-	0.16	C	
Calabria, Italy	Dairy farms near to Calabria region	36 milk samples (raw, pasteurized, semi-skimmed, whole)	ΣPAHs		HPLC–MS	-	-	5.42	A	[85]
Piedmont, Italy	No reported sources of pollution	Small and medium dairy farms	PCDDs/PCDFs		GC–HRMS	-	-	1.91	C	[37]
			DL–PCBs			-	-	5.18	A, C	
Source of the milk: MARKET										
Faisalabad, Pakistan	Urban area: dairy farms located near the cities	5 raw milk samples	α–endosulfan		GC–ECD	22.6–41.4	-	-	A	[86]
			β–endosulfan			4.06	-	-	A	
			DDE			1.52	-	-	A	
			γ–HCH			2.13	-	-	A	
Bacău, Romania	Villages around Bacău city, Comănești town, Târgu Ocna town, close to one of the greatest OCPs producers up to 1990	18 raw cow milk samples 18 pasteurized cow milk samples	α, β, γ–HCH		GS/MS	0.74–7.8	-	-	A	[35]
France	Dairy experimental station	Raw cow milk	ΣPAHs		GC–MS	-	-	0.08	A	[87]
Poland	Dairy farms located in unpolluted areas (no industry, no main roads, no chemical fertilizers) Negligible sources of pollution	Raw cow milk collected from animal exposed to contaminated feed (molassed sugar beet pellets)	PCDD/PCDFs		HRGC–HRMS	-	-	4.31	C	[34]
			DL–PCBs			-	-	0.71	A, C	
Punjab, India	Intensive dairy production, typical feeding management	Raw milk samples collected directly from the cans/cooling tanks	γ–HCH		GC–MS	0.46	-	-	A	[33]
			DDE			0.83	-	-	A	
			Endosulphan sulphate			1.01	-	-	A	

Table 2. Cont.

Location	Collected Area	Sample	POPs	Method	POPs Level (ng/g) Mean and/or Range (Min–Max)			Annex *	References	
										
Italy	Different dairy farms from Province of Taranto, near industrial area	Raw milk	PCDD/PCDFs		HRGC –HRMS	-	-	0.28	C	[88]
			DL–PCBs			-	-	0.82	A,C	
			ΣPCBs			-	2.53	-	A, C	
Catalonia region, Spain	Small local markets, supermarkets and big grocery stores	Commercial milk	ΣPAHs		HRGC /HRMS	-	-	0.47	A	[89]
Arkhangelsk Russia	Close to different urban areas from Northwest Russia	13 pasteurized milk samples purchased from supermarkets, shops and local markets; produced locally	ΣDDTs		GC	0.06–1.09	-	-	B	[90]
			ΣHCHs			0.04–0.22	-	-	A	
			ΣCHLs			0.01–0.02	-	-	A	
			Mirex			<0.01	-	-	A	
			DDE			1.4–17.0	-	-	A	
			HCB			-	0.1–0.38	-	A, C	
			ΣPCBs			-	0.17–1.1	-	A, C	
Qazvin, Iran	Marketed in urban area	7 pasteurized full-fat commercial milk samples	PCDDs/PCDFs		HRGC /HRMS	-	-	0.74	C	[91]
			DL–PCBs			-	-	0.13	A, C	
Sacramento, California	Local market area, California farms	23 commercial whole milk samples	ΣPCBs		GC –MS/MS	-	142.8–172.4	-	A, C	[36]
Source of the milk: MARKET										
Tehran, Iran	Marketed in urban area	240 samples (pasteurized; sterilized)	ΣPAHs		MSPE /GC–MS	-	-	1.42	A	[92]
Tehren, Iran	Local market area	120 pasteurized cow milk samples, collected in 2 different seasons	ΣPCBs		GC –ECD	-	18.92	-	A, C	[93]
			DL–PCBs			-	-	0.49	A, C	
China	Marketed in urban area / Traditional dairy farms	89 milk samples	ΣPAHs		GC–MS	-	-	8.85	A	[38]
Europe						-	-	9.38	A	
Australia						-	-	8.18	A	
Italy	Marketed in urban area (Southern Italy)	Whole milk	PCDD/PCDFs		HRGC –HRMS	-	-	1.19	C	[94]
			DL–PCBs			-	-	0.18	A,C	
Italy	Marketed milk	Cow milk	PCDD/PCDFs		GC –HRMS	-	-	0.155	C	[95]
			DL–PCBs			-	-	0.506	A,C	
Source of the milk: FARMS and MARKET										
* different regions: Egypt, Spain, Slovenia, Mexico	* cow milk of different origins: market zone—Egypt, Spain; dairy farms—Slovenia, Mexico.	Egypt—ns.; Spain – 94 pasteurized milk samples; Slovenia—174 raw milk samples; Mexico—355 raw milk sample	DDT		GC –ECD	15.9	-	-	B	[96]
			HCH			9.4	-	-	A	
			HCB			-	1.6	-	A, C	
Cairo, Egypt	Urban region	18 milk samples from different sources (raw milk from farm, commercial and pasteurized milk)	ΣPAHs		GC/MS	-	-	0.37–1.01	A	[97]
				A—measures for elimination production and use B—measures for restricting production and use C—measures for accidental release						
Organochlorine pesticides										
Industrial chemicals										
Combustion chemicals										

POPs—persistent organic pollutants; OCPs—organochlorine pesticides; Ind.Chem—Industrial Chemicals; Comb.—Pollutants from Combustion; HCH—hexachlorocyclohexane; DDE—dichlorodifenildicloroetano; DDT—dichlorodifeniltricloroetano; HCB—hexachlorobenzene; PCBs—polychlorinated biphenyls; PCDD/F—polychlorinated dibenzodioxins/dibenzofurans; TCDD/F—tetrachlorinated dibenzodioxins/dibenzofurans; OCDD/F—octachlorinated dibenzodioxins/dibenzofurans; DL—PCB-dioxin-like PCBs; PAHs—polycyclic aromatic hydrocarbons; GC—gas chromatography (HR/MS—high resolution/mass spectrometry; ECD—electron capture detector); HPLC/MS—high performance liquid chromatography; MS—mass spectrometry; Ns.—unspecified; CHLs—oxychlordane; cis—chlordane; trans—nonachlor. * Annex = Annexes from Stockholm Convention; this includes the list with the chemicals targeted by the Stockholm Convention: Annex A (Elimination); Annex B (Restriction); Annex C (Unintentional production). See reference [40] for POPs category symbol shape.

Most of the authors have analyzed fresh milk samples collected from farms of different sizes, with different levels of technology, located in areas with various sources of pollution [33–35,37,84–87]. Other authors have analyzed the samples of milk sold in different types of supermarkets in Egypt [96,97], Mexico [96], Spain [89,96], Russia [90], Italy [98], Iran [91–93], California [36], China or Australia [38].

For the investigation of pollutants, most of the authors have used gas chromatographic analysis coupled with mass spectrometry (GC–MS), high resolution-mass spectrometry (HRMS) or electron capture detector (ECD), and only a few authors have used high-performance liquid chromatography–mass spectrometry (HPLC–MS) [85] or HPLC with fluorescence detector [98,99].

In most of the samples, the pollutants from incomplete combustion prevailed, especially PAHs, PCDDs and PCDFs, with amounts from 0.08 to 26.6 ng/g. Some authors [33,35,86,90,96] also reported amounts of OCPs between 0.01 and 41.4 ng/g. The pollutants from industrial activity were identified only in a few cases [36,84,90,93] as being at risky levels for human health (0.09 to 3524.07 ng/g). In most of the studies, the pollutants belonged almost always to high-risk categories included in Annex A in the Listing of POPs in the Stockholm Convention (high toxic potential, regulated through measures for stopping their manufacturing and usage).

In most situations, the feed was implicated as the source of POPs in dairy animals, which ultimately resulted detectable levels of POPs in milk. Rusu et al. [35] investigated several types of milk samples (raw, pasteurized) collected from some farms in the northeast area of Romania, located close to one of the former greatest OCP producers in Romania (up to the 1990s). OCPs residues varied between 0.74 and 7.8 ng/g, suggesting long lasting persistence of OCPs in the environment and the transfer on the soil–fodder crops–grazing animals–milk chain.

Comparable results on the contamination of milk with POPs through feed was reported by Costera et al. [84] and Piskorska-Pliszczynska et al. [34]; Costera et al. [84] analyzed milk yielded by animals fed with hay produced in the vicinity of a municipal waste incinerator, and Piskorska-Pliszczynska et al. [34] analyzed molasses sugar beet pellets contaminated accidentally from different sources.

In both studies, the possibility of transferring organic pollutants such as PCDDs/PCDFs, PCBs and DL–PCBs following the long-term consumption of contaminated feed was followed. Costera et al. [84] blamed only the pollution from the municipal waste incinerator, while Piskorska-Pliszczynska et al. [34] have not identified important sources of pollution (excepting the accidental contamination) that could have modified the pollutant profile in milk. Without any exact sources of pollution, the conclusions presented by Piskorska-Pliszczynska et al. [34] have shown, therefore, that the presence of pollutants in milk (from 0.71 to 4.13 ng/g) can originated in previously contaminated feedstuffs, brought from outside the area.

Polder et al. [90] studied the contamination profile with POPs for 13 pasteurized milk samples purchased from supermarkets, shops and local markets close to different urban areas in northwest Russia. Variable amounts of OCPs, including DDT, hexachlorocyclohexane (HCH), chlordane, mirex or DDE and some pollutants from the processing of industrial substances such as HCB and PCBs, were identified. The authors emphasized the complexity of organic pollutants and how much the pollution sources associated with urban areas can influence the total amounts of POPs. Comparable results were obtained by Sajid [86] on samples of raw milk collected from farms located near cities. Important quantities of OCPs (1.52 to 41.4 ng/g) were quantified, along with high amounts of α , β –endosulphan and lower amount of HCH, dieldrin and DDE.

There were no reported sources of pollution in the study conducted by Desiato et al. [37] on milk samples collected from farms of different sizes in Italy. However, the presence of PCDDs/PCDFs (1.91 ng/g) and DL–PCBs (5.18 ng/g) was reported, and the atmospheric pollution was incriminated.

Other authors have reported OCP residues in milk, such as DDT and HCH (9.4 to 15.4 ng/g), HCB residues (1.6 ng/g) [96] and even the pollutants from incomplete combustion, especially PAHs (from 0.08 ng/g [92] to 5.42 or 5.48 ng/g) [85,99]; however, they do not mention the pollution sources.

Considering that modern pollution sources are the industrial emissions and motor vehicle emissions, some authors have particularly studied the level of contamination of milk with PCDD/PCDFs, PCBs or DL-PCBs. In Europe, Italy is one of the countries that stands out, with numerous studies carried out on this topic [82,85,88,94,95,98].

The identification of pollution sources was only possible in a few studies run on locally produced milk by animals fed with known feedstuffs. In samples of marketed milk, the identification of pollution origin was almost impossible since no information was known about the provenience of the milk; this is collected from multiple farms then subsequently processed.

Regarding the proportions of POPs found, it is important to mention that all the levels of different types of POPs presented in the studies listed are not directly comparable with enforceable actions levels or with minimum tolerance levels in milk. The levels mentioned in almost all the papers are lower than enforceable action levels or minimum tolerance levels in feed established by different authorities (e.g., European Commission, FDA), but the efforts to reduce environmental pollution are very important and must be kept constant.

3.2. Animal Feed

Table 3 summarizes the predominant POPs reported by different authors in animal feed and the distribution per pollutant according to pollution sources, the pollutant category and its classification according to the degree of risk, the applied analytical method, and the quantitative profile of the identified pollutants.

Not many articles have focused on the presence of POPs in animal feed [33,34,84,100,101], especially due to the difficulties in studying this matrix, given either by the generally physicochemical characteristics of the feeds (for example, the low-fat content), or given by the lack of analytical methods and techniques applicable to the matrix.

However, a few authors have identified the presence of POPs in animal feed in different proportions, depending on the pollution profile of the analyzed area, through highly sensitive analytical techniques, such as GC-MS, GC-HRMS or high-resolution gas chromatography (HRGC) coupled with HRMS. All the levels of different type of POPs presented in the studies listed are not directly comparable with enforceable actions levels or with minimum tolerance levels in feed; in almost all the papers, the proportions founded are lower than the enforceable action levels or minimum tolerance levels in feed as established by different authorities (e.g. European Commission, FDA).

Reported according to the degree of risk of each pollutant category, according to the Stockholm Convention [42], the pollutants identified in most of the feed samples are not part of the high-risk categories and were particularly identified pollutants from Annex C of the listing of POPs. Only accidental release reduction measures are currently applied for these.

Nevertheless, despite numerous applied pollution prevention measures, the action taken will provide effective results in the long term due to the high residues persistence. In this context, recent works [33] reported the presence of some high-risk pollutants, such as OCPs, included in Annex A of the Stockholm Convention; they were banned from production and usage since 2001, suggesting an old contamination of the area.

The behavior of pollutants in the environment is complex and aims to clarify that the interaction between chemical compounds released into the environment and the substrates (soil, water, air, plants) is different. Not all substrates react similarly in contact with pollutants due to their different sensitivities and physical and chemical particularities [95].

Table 3. Summary of POPs presence in animal feeds and sources of pollution.

Location	Collected Area	Sample	POPs	Method	POPs Level (ng/g) Mean and/or Range (Min–Max)			Annex *	References
									
France	Field located along motorways and airports, without any influence of other major sources of pollution	Grass collected from a field nearest a motorway and airport	ΣPAHs 	GC–MS	-	-	25	A	[100]
Tarragona (Catalonia region), Spain	Urban area: chemical and petrochemical industries: a municipal solid waste incinerator, a hazardous waste incinerator, a PVC production facility, highways and roads with high traffic density	Feed sample collected near important sources of pollution	PCDDs/TCDD 	HRGC /HRMS	-	-	-	C	[101]
			PCDDs/OCDD 		-	-	1680	C	
			PCDFs/TCDF 		-	-	210	C	
			PCDFs/OCDF 		-	-	430	C	
France	Vicinity of a hazardous municipal waste incinerator	Contaminated hay	ΣPCBs 	GC –HRMS	-	52.2–79.8	-	A, C	[84]
			PCDDs/TCDD 		-	-	60	C	
			PCDDs/OCDD 		-	-	1033	C	
			PCDFs/TCDF 		-	-	350	C	
			PCDFs/OCDF 		-	-	1490	C	
Poland	Dairy farms located in unpolluted area (no industry, no main roads, no chemical fertilizers) (negligible sources of pollution)	Contaminated feed (molasses sugar beet pellets)	PCDDs/PCDFs 	HRGC –HRMS	-	-	5.0–427	C	[34]
			DL–PCBs 		-	-	20–100	A, C	
Punjab, India	Intensive dairy production, typical feeding management	Feed samples collected from the animal sheds	γ–HCH 	GC–MS	2.73	-	-	A	[33]
			DDE 		1.90	-	-	A	
			Endosulfan 		2.95	-	-	A	
Poland	Marketed feed	Feeds of plant origin (sugar beet, pellets, dried alfalfa, dried apple)	PCDDs/PCDFs 	HRGC –HRMS	-	-	3.43	C	[27]
			ΣPCBs 		-	0.11	-	A, C	
					 Organochlorine Pesticides  Industrial chemicals  Combustion chemicals A—measures for elimination production and use B—measures for restricting production and use C—measures for release accidental liberation				

POPs—persistent organic pollutants; OCPs—organochlorine pesticides; Ind.Chem—Industrial Chemicals; Comb.—Pollutants from Combustion; HCH—hexachlorocyclohexane; DDE—dichlorodifenildicloroetano; PCBs—polychlorinated biphenyls; PCDDs/PCDFs—polychlorinated dibenzodioxins/dibenzofurans; TCDD/F—tetrachlorinated dibenzodioxins/dibenzofurans; OCDD/F—octachlorinated dibenzodioxins/dibenzofurans; DL—PCB—dioxin-like PCBs; PAHs—polycyclic aromatic hydrocarbons; GC—gas chromatography (HR/MS—high resolution/mass spectrometry). * Annex = Annexes from Stockholm Convention; this includes the list with the chemicals targeted by the Stockholm Convention: Annex A (Elimination); Annex B (Restriction); Annex C (Unintentional production). See reference [40] for POPs category symbol shape.

For the effective biomonitoring of different types of pollutants, the literature proposes the prediction of environmental factors that could favor the risk of contamination in order to render efficacy to control actions [102,103]. A methodological model for ranking the risks associated with pollution on livestock farms was developed by Battisti et al. [102], which is useful to estimate the probability of contamination from a certain area.

Monitoring the pollution sources and environmental factors that could increase contamination risks was also applied, without a mathematical model, in the study developed by Matei and Pop [104] to estimate the probability of contamination within a dairy farm. In addition to identifying environmental factors that may favor pollutant accumulation, Matei et al. [105] suggested that the particularities of the feed, and especially the fat content, can provide important clues about the presence of pollutants.

On the point sources of pollution, most articles mention in general the pollution from the chemical and petrochemical industry, but particularly the presence of PCDDs/PCDFs, PAHs and DL–PCBs from incomplete combustion from different industrial sectors for transport or waste burning [84,100,101]. The presence, in different proportions, of PCDDs/PCDFs and DL–PCBs was also confirmed by Piskorska-Pliszczynska et al. [34] and Bedi et al. [33], although the sources of pollution from industry and combustion were not specified.

Variable proportions of pollutants were found in animal feed, originating from incomplete combustion point sources (5–1680 ng/g), industrial point sources (52.2–79.8 ng/g) and from agriculture processes (1.90–2.95 ng/g) (see further Table 3).

Crepineau-Ducoulombier and Rychen [100] investigated the profile of contaminants in different feed samples collected from agricultural areas in the vicinity of a highway and an airport in eastern France, and they analyzed samples of fodder exposed to pollution from transport emissions (release/evacuation of gasoline, diesel, kerosene). There were 16 types of PAHs listed by the U.S. Environmental Protection Agency as hazardous. The presence of these types of POPs in different proportions in all samples has led to the association of these types of pollutants with different point sources of pollution, such as transports emissions or other substances released from incomplete combustion. The same pollution sources were reported by Schuhmacher et al. [101] in the Catalonia region (Spain) for feed samples collected from an area with intensive road traffic, highlighting the presence of organic pollutants, such as PCDD/Fs, and compounds released from incomplete combustion, similar to PAHs. Schuhmacher et al. [101] also mentioned some important point sources of pollution of PCDD/Fs, such as the incomplete combustion from chemical industry factories and waste incinerators or plastic factories, whilst Crepineau-Ducoulombier and Rychen [100] did not identify such sources for PAHs, even they belong to the same group of POPs (combustion chemicals).

Costera et al. [84] investigated the presence of PCDD/Fs in hay samples collected from the vicinity of a municipal waste incinerator in France. The same source of pollution was reported by Schuhmacher et al. [101] for soil and vegetation samples taken from area with chemical and petrochemical industries. The GC–HRMS quantification results of PCDD/Fs were also in similar ranges—60–1490 ng/g PCDD/Fs in the hay samples [84] and 210–1680 ng/g in feed samples [101]. For similar pollution sources, Costera et al. [84] also observed the presence of different congeners of PCBs pollutants at various levels such as 52.2–79.8 ng/g, which was often associated with industrial activities [45] or waste processing activities [106].

The presence of different types of POPs within a small dairy farm located in Poland in an area without relevant pollution sources (no industry, no main roads, no chemical fertilizers) was reported by Piskorska-Pliszczynska et al. [34]. Levels of 5.0–427.0 ng/g for PCDD/Fs and 20.0–100.0 ng/g for DL-PCBs were identified in samples of sugar beet pellets, most likely contaminated prior to arrival in the farm, thus highlighting the possibility of accidental contamination during the feed manufacturing. The possibility of accidental contamination was also reported by Rychen et al. [106], so the pollution sources identified within an area are not always singular but can be reinforced by previously existing potential sources.

Accidental contamination of feed from unknown pollution sources was considered by Pajurek et al. [27] in a study focused on the official control of contamination with PCDDs/PCDFs, PCBs and DL-PCBs of different types of feed sold in Poland. Mean values of PCDDs/PCDFs levels of 3.43 ng/g and 0.11 ng/g for PCBs were measured. Moreover, in 2.8% of the analyzed samples, exceedances of contamination limits were reported in relation to feed and food safety regulations. Therefore, it is crucial to consider the origin of feedstuffs or feed to maintain traceability and to identify the cause of any possible contamination. Situations when feed is not produced locally and is purchased and transported from other areas [34] can have economic consequences and negative impacts on animals and their products.

Bedi et al. [33] analyzed concentrate and green fodder samples from 55 dairy farms in Punjab, India, via HRGC–HRMS detection techniques, and the presence of three types of pollutants—OCPs–HCH, dichloro–diphenyl–dichloroethane (DDE), endosulphan sulphate—was confirmed and quantified between 1.9–2.95 ng/g, without mentioning the sources of pollution in the studied area. In conclusion, only a few authors have focused on POPs' occurrence in feedstuffs and feed, and just a few solutions have been presented for the biomonitoring of POPs and for the identification of pollution sources. Therefore, more

studies on fodder in livestock grazing or in feed used in intensive farming should be encouraged.

4. List of Reported POPs

For the global problem of pollution, the data reported in the literature were compared with the main provisions of the Stockholm Convention on POPs. Moreover, pertaining to their amount and the different measures applied to eliminate, reduce or restrict them, the potential risk was assessed.

Figure 1 includes the POPs found in feed samples in relation to the listing of POPs in the Stockholm Convention.

		Organochlorine pesticides			Industrial chemicals			Combustion chemicals		
		A	B	C	A	B	C	A	B	C
		France			Spain			France		

included in Annex C of the Stockholm Convention were also identified and only one type of pollutant identified belonged to the Annex B.

		Organochlorine pesticides			Industrial chemicals			Combustion chemicals				
		A	B	C	A	B	C	A	B	C		
		(*)										
		A/ Elimination ** B / Restriction ** C / Reduction **										
		(*)										
		France	Egypt Slovenia Mexico	Spain Russia	Italy	Iran Italy Egypt	Iran Pakistan	Romania	California Poland Iran	India	Iran	Australia Asia Europe
2001	Chlordane			1								
2001	DDE			1			1			1		
2001	DDT		1	1								
2001	Dieldrin						1					
2001	DL-PCBs ²					1	1	1	2			
2012	Endosulfan						1			1		
2001	Endrin			1								
2001	HCB [*]		1	1								
2004	HCH		1	1			1	1		1		
2001	Mirex			1								
2004	PAH			1	1	2			1		1	1
2001	PCB ³	1		1					2			
2001	PCDD	1				2	1		1			
2001	PCDF	1				2	1		1			

** included also in Annex C*

* included also in Annex C

Figure 2. POPs presence in milk (including the number of reporting articles). Classification conducted in relation to the measures applied by the Stockholm Convention (**). The dates on the Y axis indicates the year when the chemical compound was included in one of the annexes of the Stockholm Convention. Some POPs are part of Annex A and also of Annex C (*).

The analysis of POPs in relation to the potential toxicity and to one of the three annexes of the Listing of POPs in the Stockholm Convention revealed, for both milk and feed, a worrying situation regarding their safety for humans and animals. Costera et al. [84] and Polder et al. [90] highlighted that the same sources of pollution can generate pollutants with higher toxic potential, namely, those comprised in Annex A or pollutants with lower toxic potential, such as those from Annex C or Annex B.

Desiato et al. [37] and Bedi et al. [33] mentioned the presence of pollutants included in Annex A of the Stockholm Convention in the absence of eligible sources of pollution, thus underlying the need for continuous monitoring, even in apparently risk-free areas.

With respect to this review, it is important to understand that the information presented is not a representative description of all the POPs. We must consider that not all the studies used the same analytical techniques and instrumentation, and not all looked for the same list of POPs, which makes it difficult to compare the results.

5. Conclusions

Important sources of pollution were identified, namely, industrial activity, particularly the incomplete combustion of various substances originating in transportation or waste management activities. In addition to the identified pollution sources, the presence of pollutants depends on the history of pollution in the study area, on the characteristics of pollutants, on the differences regarding urbanization and industrialization and on the environmental transfer of pollutants.

Considering the temporal dimension of feed and milk contamination, it was highlighted that despite numerous efforts to prevent and eliminate POPs, their imported residues persist in the environment and create adverse effects which persist for several years after taking measures and banning the dangerous POPs via law enforcement.

Further research should focus on developing more effective strategies to reduce or to eliminate POPs contamination, especially in feed, and subsequently to decrease animal and human exposure. Additional studies might be necessary to evaluate the response of the animal body to the action of POPs, focusing on the development of physiological measures to limit the transfer of POPs from the animal body to animal products.

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