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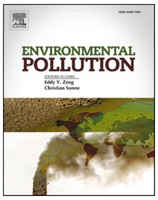

Review

Presence of pesticides in the environment, transition into organic food, and implications for quality assurance along the European organic food chain – A review[☆]

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ABSTRACT

The use of synthetic pesticides is not allowed in organic production, but traces of synthetic pesticides are regularly detected in organic food. To safeguard the integrity of organic production, organic certifiers are obliged to investigate the causes for pesticide residues on organic food, entailing high costs to the organic sector. Such residues can have various origins, including both fraud and unintentional contamination from the environment. Because the knowledge about contamination from environmental sources is scattered, this review provides an overview of pathways for unintentional and technically unavoidable contamination of organic food with synthetic pesticides in Europe. It shows that synthetic pesticides are widely present in all environmental compartments. They originate from applications in the region, in distant areas or from historical use. Transition into the food chain has been demonstrated by various studies. However, large uncertainties remain regarding the true pesticide contamination of the environment, their dynamics and the contamination risks for the food chain. Organic operators can take certain measures to reduce the risks of pesticide contamination of their products, but a certain extent of pesticide contamination is technically unavoidable. The present paper indicates that (i) a potential risk for pesticide residues exists on all organic crops and thus organic operators cannot meet a 'zero-tolerance' approach regarding pesticide residues at the moment. (ii) Applying a residue concentration threshold to distinguish between cases of fraud and unavoidable contamination for all pesticides is not adequate given the variability of contamination. More reliable answers can be obtained with a case-by-case investigation, where evidence for all possible origins of pesticide residues is collected and the likelihood of unavoidable contamination and fraud are estimated. Ultimately, for organic certification bodies and control authorities it will remain a challenge to determine whether a pesticide residue is due to neglect of production rules or technically unavoidable.

1. Introduction

1.1. Pesticide residues on organic food

According to the European Commission, "organic production is an overall system of farm management and food production that combines best environmental practices, a high level of biodiversity, the preservation of natural resources and the application of high animal welfare standards" (European Parliament, 2018). The rules for organic production cover a broad range of topics such as crop production, animal husbandry, food processing, soil management and conservation of biodiversity. One aspect of this set of rules is that the use of synthetic pesticides is not allowed in organic production. In line with consumer expectations, the organic sector aims to minimize contamination of organic produce with such substances (EU, 2018/848). However, traces of synthetic pesticides are regularly detected in organic foods in Europe (EOCC, 2019; EFSA, 2018; Ministerium für Ernährung, Ländlichen Raum und Verbraucherschutz, 2021; Schleiffer et al., 2021). For the European market, the latest survey of the European Food Safety Authority (EFSA) shows that 6% of organic produce contain pesticide residues (EFSA, 2018). For Southern Germany, a monitoring study reports pesticide residues in 28% of organic food (Ministerium für

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Ernährung, Ländlichen Raum und Verbraucherschutz, 2021). For the Swiss market, a study by Schleiffer et al. (2021) found that 9% of organic produce contain pesticide residues.

The occurrence of synthetic pesticide residues on organic food contrasts with consumer expectations. According to Stolz et al. (2022), von Meyer-Höfer et al. (2015) and Schroeder et al. (2016), avoiding pesticide residues is a main motive for purchase of organic produce among consumers. This consumer motive is driven by increasing concerns over negative health effects of pesticide residues. European legislation defines maximum residue levels (MRLs) for each combination of pesticide and food commodity (EC 396/2005). Foods with residue concentrations above the MRL may not be placed on the market while food complying with the MRL is considered to be safe (European Communities, 2008). However, the understanding of the health effects of pesticide residues on food for consumers is far from complete, also because they are difficult to disentangle from other health impacts (Kim et al., 2017). Furthermore, the interaction effects of pesticide mixtures are poorly understood yet (Roth & Wilks, 2018). Among organic traders, certifiers and authorities, however, pesticide residues below the MRL on organic produce are primarily a concern because they might indicate fraud, and not because of consumer health issues.

1.2. *Origin of pesticide residues on organic food*

Pesticides are used for a wide variety of purposes in modern society. They are applied in conventional agriculture, stock-protection, private gardening, forestry, road and railroad maintenance, conservation of industrial goods, human and veterinary medicine. In the European Union more than 333 000 tonnes of pesticides are sold every year (Eurostat, 2021) and over 450 substances are approved (as of 2021). From the application sites, pesticides have the potential to enter and disperse through the environment. For example, studies show that over half the amount of pesticides applied to crops reach the soil environment (Goulson, 2013; Navarro et al., 2007). The frequent detection of pesticides in environmental compartments such as soil, water and air (Kruse-Plaß et al., 2021; Mohaupt et al., 2020; V. Silva et al., 2018) and the findings of pesticides in non-target plants (Botías et al., 2016; Linhart et al., 2019; Linhart et al., 2021) indicate the wide occurrence of pesticides in the environment.

As a consequence, pesticide residues on organic food can have various origins. According to Bickel and Speiser (2020) important contamination pathways are: unauthorized application, conventional produce marketed as organic, cross-contamination with treated produce in transport, storage or processing facilities, spray drift and contaminated soil. Not all of these contamination pathways can be avoided by organic operators. Especially contamination from the environment or in trade facilities can be difficult to mitigate. A data collection by the European Organic Certifiers Council (EOCC) covering over 7500 organic product samples provides an overview of the most frequently identified causes for residues on organic produce by organic control bodies or control authorities (EOCC, 2019). A total of 43% of all investigated cases were due to contamination from the environment: 18% were attributed to drift, 8% to 'pollution' (transfer of pesticides to organic crops in the field from contaminated soil or water; unavoidable for the individual farmer), and 17% to 'contamination' (postharvest transfer of pesticides to organic food from contaminated equipment or installations; avoidable with appropriate preventive measures).

1.3. *Consequences of pesticide detections on organic food*

All operators along the organic production chain are required to monitor pesticide residues in the products (EU, 2018/848). The detection of pesticide residues on organic food causes an investigation, in order to fight cases of fraud and ensure the integrity of the organic market. This procedure usually involves the blocking of the affected goods, followed by labour and cost-intensive investigations of the causes for the residue (Speiser et al., 2020). The assignment of responsibilities between operators, control bodies and authorities varies between countries and is currently under debate in Europe.

Findings of pesticide residues on organic produce affect not only the owner of the concerned food lot, but also often involved suppliers and customers. Timely delivery of goods is a key factor in the food industry and the blocking of goods may severely disrupt a food chain. In addition, the legal requirements to deal with pesticide residues on organic food and to investigate cases of fraud differ in European countries (Milan et al., 2019). The EU organic regulation does not set specific threshold for residues on organic produce. In contrast, the private sector and also national legislation in some EU countries set threshold values for residues on organic produce (Speiser et al., 2020). These thresholds are usually much below the MRL. The mosaic of approaches and the blocking of goods is a challenge for international trade relations and creates a significant economic burden for organic operators (Speiser et al., 2020). Therefore, the European Union aims to harmonize the legal requirements regarding pesticide residues on organic products. However, this subject is hotly debated at the moment and a harmonized approach cannot be expected before a few years.

Adequate and fast assessment of residue cases is crucial to limit costs and to ensure timely supply of organic food. A sound understanding of pesticide occurrence in the environment and their potential to enter the food chain is important for organic operators to minimize the contamination risks. Moreover, it is a pre-requisite to distinguish between residue cases caused by unauthorized pesticide use or environmental pollution.

1.4. *Objectives of this review*

To our knowledge, there is yet no comprehensive review of the implications of environmental pesticide contamination for the organic food chain. This review elucidates some of the major pathways how synthetic pesticides (hereinafter referred to simply as pesticides) reach the main environmental compartments, how they are dispersed within and between these compartments and how they finally enter the food chain. The review focusses on the situation in Europe. However, the underlying mechanisms are of a general nature and similar patterns can be expected also elsewhere (see Benzing et al. (2021).

It is widely recognized in organic control procedures that the presence of pesticide residues on organic food might be caused by their fraudulent application, or by fraudulent marketing of conventional produce as organic (Speiser et al., 2020). By contrast, there is much less awareness about non-intentional pesticide contamination from environmental sources. For this reason, this review focuses on environmental contamination rather than fraud. Since pesticide contamination of the environment is an extensive topic, this review does not attempt to cover all pesticide compounds, regions and organic crops. Instead, we illustrate the main processes and contamination pathways.

2. Method

For this review we aimed to identify studies that provide insight into the contamination of the environment with pesticides, their dispersion within the environment and the transition from the environment to the food chain. We structured the literature search according to the environmental compartments of soil, water, air (including rainwater) and the post-harvest environment. We focussed on studies from Europe and gave preference to studies conducted within the last 20 years. For each environmental compartment and for the transition from an environmental compartment to the food chain, we aimed to identify a few studies giving a broad overview in terms of substances and geographical range. Where such studies were unavailable, we included either older studies, more detailed studies or studies from outside Europe. The keywords used for the search are listed in Table 1.

The literature search was performed in Google Scholar. Grey

Table 1
List of keywords used for literature search. The table is structured into the keywords used for the four environmental compartments and into the keywords for the three sub-topics of the review. The search on the sub-topics was performed for each environmental compartment.

	Environmental compartments
Soil	Soil
Water	Surface water; ground water; irrigation; rain; rainwater
Air	air
Post-harvest environment	Post-harvest; processing; food facilities
	Topics
Contamination of environment with pesticides	pesticide AND residues OR contamination AND Europe AND [see 'environmental compartments'] pesticide AND monitoring AND Europe AND [see 'environmental compartments']
Dynamics of pesticides in the environment	pesticide fate OR partitioning AND [see 'environmental compartments']
Transition into the food chain	Pesticide AND transition from [see 'environmental compartments'] AND food OR organic food

literature, such as governmental reports, were identified using Google and the same keywords. For the grey literature, also reports in German, French and Italian were included. The search was conducted from October 2020 until November 2021.

2.1. Occurrence of pesticides in the environment and their transition into food

2.1.1. Dynamics of pesticides in soils

Pesticides in soil can have several origins. When crops are sprayed with pesticides, the main targets are usually the leaves, shoots or flowers. Nevertheless, approximately 50% of the applied active substances are deposited on the soil (Navarro et al., 2007). In addition, pesticides deposited on the leaves are partially washed away by rainfall, and thus also carried to the soil. Furthermore, aged plant parts like wilted flowers will fall to the ground and may carry pesticides to the soil. Another important contamination pathway is the direct treatment of soil against weeds, soilborne pests or diseases (e.g. slugs, nematodes, wireworms) and the use of pesticide-treated seeds. In the case of neonicotinoids, over 90% of the active ingredient applied to seeds enter the soil (Goulson, 2013). In the soil, pesticides may bind to soil particles, volatilize, diffuse, degrade, wash away with the water, or be taken up by plants or other organisms (Navarro et al., 2007). The mobility of pesticides in soil is mainly controlled by their adsorption to soil particles, which depends on soil properties such as organic matter content, clay content, soil pH, soil porosity, water content, temperature, the microbial community and on agricultural practices, as well as the physico-chemical properties of the pesticide (Hilber et al., 2008; Vryzas, 2018). As the net result of all these processes, some pesticides may disappear from the soil ecosystem within a few days, while others persist much longer, and in the worst case for decades (Vryzas, 2018).

2.2. Contamination of conventionally managed soils

Pesticides are frequently found in agricultural soils in Europe and in general, soil and sediments are the environmental compartments with largest pesticide deposits (V. Silva et al., 2019). V. Silva et al. (2019) analysed soil samples from agricultural fields in 11 European countries. Overall, 83% of the soil samples contained at least one pesticide, and 58% contained more than one pesticide. A total of 43 different pesticidal compounds was detected. Glyphosate and its metabolite aminomethylphosphonic acid (AMPA), dichlorodiphenyldichloroethylene (DDE) (metabolite of dichlorodiphenyltrichloroethane (DDT)), boscalid, epoxiconazole, tebuconazole and phthalimide occurred most frequently. In another Europe-wide study, glyphosate, AMPA and pendimethalin were detected most frequently and in the highest concentrations (Geissen et al., 2021). A study in the Czech Republic found that 99% of the soil samples contained at least one pesticide (Hvězdová et al., 2018). In this study, triazine herbicides and conazole fungicides were found most frequently and at the highest concentrations. Studies in Switzerland show a comparable contamination level of agricultural soils (Chiaia-Hernandez et al., 2017; Humann-Guilleminot et al., 2019). Humann-Guilleminot et al. (2019) found neonicotinoids in 100% of the non-organic soils and in 93% of the samples from organic farms.

There is a correlation between the pesticides applied recently on a field and the pesticides found in soil, but there are also exceptions where pesticides are found in the soil which were applied years before the sampling. A study from Switzerland analysed soil samples from 14 sites with a known history of pesticide application over a period of 14 years (Chiaia-Hernandez et al., 2017). Overall, about 80% of the applied pesticides were detected either in their original form or as metabolites. The majority of soil samples contained 10–15 different pesticides. In 38% of the cases, triazine herbicides which were not applied during the recorded period, but probably before the recording started, were detected in soil. This corresponds with findings from the Czech Republic mentioned above (Hvězdová et al., 2018). These compounds are banned in the EU and Switzerland since 2004 and 2012 respectively. As shown by these studies, they can remain in soil for a considerable amount of time after their application.

One of the best-known examples for pesticide persistence in soil is the group of 'organochlorine pesticides' (OCPs), which comprises substances such as DDT, aldrin, dieldrin, endosulfan, lindane und hexachlorbenzene. Most of these compounds have been banned for decades. Nevertheless, their presence in soil is still reported frequently (Hilber et al., 2008; Thiombane et al., 2018).

2.3. Contamination of organically managed soils

Organically managed soils show fewer pesticides and lower levels of pesticide residues than conventionally managed soils (Geissen et al., 2021; Humann-Guilleminot et al., 2019; Riedo et al., 2021; Szekacs et al., 2015). Geissen et al. (2021) analysed the pesticide contamination of organic and conventional soils cultivated with four major crops: vegetable and orange production in Spain, grape production in Portugal and potato production in The Netherlands. This mixture of annual and perennial crops, across central and southern Europe provides insight into a broad spectrum of agricultural contexts. The organic soils had significantly fewer residues than conventional soils and 70–90% lower residue concentrations. Organic soils had a maximum of 5 substances per sample while conventional soils contained up to 16 substances per sample. In Switzerland, a total of 100 organic and conventional fields were investigated by Riedo et al. (2021), and similar patterns were found. Although pesticides were found at all sites, the number of substances was two times lower in the organic fields, and the residue levels were nine times lower. With increasing duration of organic management, pesticide residues in soil decreased significantly. However, some pesticides could still be found after 20 years of organic management. Another study in Switzerland focusing on neonicotinoids showed that organic soils contained on average 0.2 μg/kg neonicotinoids, IPM soils 0.79 μg/kg and conventional soils 2.11 μg/kg (Humann-Guilleminot et al., 2019). The authors attribute the widespread occurrence of neonicotinoids to a combination of off-field dust, runoff waters and spray drift. A study in Hungary also found lower pesticides contamination levels in organic soil samples than in conventional samples (Szekacs et al., 2015).

2.4. Transition from soil into food

Crops can be contaminated with pesticides from the soil through plant uptake and through soil particles adhering to the plant surface. Pesticide uptake happens via roots through the vapour or water phase of the soil. According to a literature review by Collins et al. (2006) the uptake process is usually passive (Cabidoche & Lesueur-Jannoyer, 2012;

Florence et al., 2015). The uptake of a compound is largely driven by its bioavailability in the soil environment. As organic matter and clay aggregates bind pesticides and other contaminants in soil, the transition of pesticides from soil to food is easier in soils with low organic matter and low clay content (Christou et al., 2019). Once inside a plant, pesticides are transported through the plant by the vascular system from where they can diffuse into adjacent tissues (Christou et al., 2019; Collins et al., 2006). The fate of pesticides depends on their physico-chemical properties, on the plant's physiology and on the transpiration rate. Environmental factors increasing the transpiration rate such as high temperature, high wind speed and low humidity, lead to an increased uptake of pollutants in plants. Consequently, crops in hot and dry regions show increased uptake of pesticides compared to plants in cool and humid regions (Christou et al., 2019). Research shows that the risk of contamination from soil is higher in root crops and leafy vegetables than in fruit and grains (Christou et al., 2019; Pullagurala et al., 2018).

The uptake of OCPs into crops from the soil environment has been studied in various cases. For the root vegetables taro, sweet potato, yam, turnip and radish, Florence et al. (2015) and Cabidoche and Lesueur-Jannoyer (2012) have demonstrated the uptake of chlordecone from soil. OCP are well-known for their accumulation in the fruits of cucurbits (Hilber et al., 2008; Wyss et al., 2012). Hilber et al. (2008) report residues of the OCPs pentachloroaniline, DDT, DDE, dieldrin, chlordane and heptachloroepoxide in cucumbers, zucchinis and pumpkins. As OCPs tend to accumulate in fats OCP concentrations in pumpkin seeds were higher than in the fruit flesh. The fat accumulation effect has also been observed for other pesticides. Han et al. (2017) found that various nut-bearing crops take up pesticides from the soil and accumulate them in the nuts. This was the case not only for several OCPs, but also for chlorpyrifos, bifenthrin, fenpropathrin, cyhalothrin, cypermethrin, fenvalerate, deltamethrin, triadimefon and buprofezin. Neonicotinoids are also known to be taken up by various crops such as cereals, beetroot and leafy vegetables (Humann-Guilleminot et al., 2019; Li et al., 2018). Scientific understanding of the uptake processes of soil pesticides into non-target plants remains limited. Based on the literature, we assume that pesticide uptake and accumulation varies significantly depending on the active substance, the soil, the environment and the crop plant.

In addition to root uptake, pesticides from the soil can also contaminate foods when soil particles adhere to the above ground plant surface. This happens as a consequence of wind erosion, rain splash, mechanical disturbance (especially during harvest) and impact of livestock (Smith & Jones, 2000). Such contamination particularly affects smaller plants with a maximum height of 50 cm high (Smith & Jones, 2000).

2.5. *Dynamics of pesticides in water*

Pesticides are not intentionally applied to water bodies, but the use of pesticides in intensive agricultural production leaves traces in the aqueous environment. Contamination patterns differs substantially for atmospheric water (clouds, rain), surface water, groundwater, and waterbodies containing wastewater. Rainwater is an intermediate case between air and water, and is discussed in Chapter "Dynamics of pesticides in the air".

The different pathways for contamination of surface water with pesticides are described by Carter (2000). Run-off from agricultural fields is particularly important in steep terrain, in areas with strong rainfall, on compacted or very dry soil or when fields are irrigated after pesticide application. Highly water-soluble pesticides are particularly prone to run-off. Other contamination pathways include lateral movement of fluids in soil or drainage systems, spray drift and also point sources such as spillage, tank washing and waste disposal (Carter, 2000).

For groundwater, the main contamination pathway is leaching from agricultural production sites. The extent of leaching depends on soil properties, pesticide physicochemical properties, formulation of commercial pesticides, distribution of rainfall events or irrigation strategy and hydrogeological processes (Tiktak et al., 2004). In soils with preferential flow channels, pesticides can reach groundwater irrespective of their physicochemical characteristics (Vryzas, 2018). Another contamination pathway for groundwater is bank infiltration from rivers and streams (BAFU, 2019; Vryzas, 2018). In general, mainly mobile and persistent pesticides and pesticide metabolites reach the groundwater. As the degradation processes are slow and water residence time is usually high, pesticide contamination of groundwater aquifers can have long term effects (BAFU, 2019).

The dissipation of pesticides in water depends on various factors. Vryzas (2018) mentions temperature, pH, sunlight, suspended materials, presence of dissolved organic material and the biota (algae, fish, zooplankton). Photo-degradation is a major dissipation pathway for pesticides in surface water. Pesticide residues may also be deposited in water sediments, but this process has not been studied intensively (Vryzas, 2018).

2.6. *Major factors influencing pesticide contamination of waters*

In many cases, there is a clear correlation between pesticide use and their occurrence in water bodies (Gauroy & Carluer, 2011; Herrero-Hernández et al., 2020; Horth & Blackmore, 2009; Szekacs et al., 2015; Vryzas et al., 2009; Wittmer et al., 2014). A variety of studies finds that the compounds most frequently detected in surface or groundwater often correspond with the substances applied in the catchment area (Ccanccapa et al., 2016; Herrero-Hernández et al., 2020; E. Silva et al., 2015). Therefore, the type of substances found in water can be regionally different (Herrero-Hernández et al., 2020; Szekacs et al., 2015). However, some pesticides can be found in surface waters even years after their application has been banned. Szekacs et al. (2015) report findings of pesticides in Hungarian surface waters early in the season, before the application starts. In the Walloon region in Belgium, several pesticides, including lindane, which were not authorized for use anymore were detected in water (Chalon et al., 2006).

Heavy rains or strong irrigation in combination with steep terrain influence leaching and run-off from agricultural areas. The run-off waters carry pesticides dissolved in the water or bound to soil particles or organic matter to surface and groundwaters, especially if precipitation onset is shortly after pesticide application (Mohaupt et al., 2020; Vryzas et al., 2009). However, Szekacs et al. (2015) highlight, that precipitation also dilutes the pesticide concentration in surface water. Thus, precipitation may increase or decrease pesticide residues in water.

2.7. *Contamination of surface water*

European streams, rivers and lakes contain residues of various pesticides, usually in the range of ng/L to mg/L. Many studies relate their findings to the threshold for safe drinking set by the EU Drinking Water Directive (Council Directive 98/83/EC, 1998) (see Table 2). The directive sets a threshold of 100 ng/L for individual compounds and of 500 ng/L for the sum of all compounds. Mohaupt et al. (2020) summarize monitoring data of 180 pesticidal substances from 6500 surface water and 14 000 groundwater sites between 2007 and 2017. At 5–15% of surface water monitoring sites, the safe drinking water limit was exceeded by herbicides and the substance with the highest exceedance rate is the herbicide glyphosate. At 3–8% of the sites, the safe drinking water limit was exceeded by insecticides. In terms of individual compounds, there are a few substances which are regularly found in water bodies all over Europe (mainly atrazine, DDT, simazine, aldrin and alachlor), while many other substances occur only sporadically or at a regional scale (Mohaupt et al., 2020).

Many surface waters contain cocktails of pesticides. In a case study with mid-sized rivers in Switzerland, 40 pesticides out of 220 analysed compounds were detected on average per sample (Wittmer et al., 2014). In another case study with streams in France, Germany, the Netherlands

Table 2

Studies on contamination of surface and ground water. Due to differing methodologies (compounds detectable, analytical sensitivity), the studies are not directly comparable.

Region	Samples containing pesticides	Samples containing >100 ng/L pesticides	Source
		Surface water	
Hungaria	2–51%	n.r.	Szekacs et al. (2015)
Czech Republic	79.8%	7.4%	Veverka & Lesinsky (2005)
Europe (only glyphosate and AMPA)	30%	23%	Horth and Blackmore (2009)
Italy	77.3%	21%	Paris et al. (2020)
Spain (Valencia region)	100%	n.r.	Ccanccapa et al. (2016)
The Netherlands	82%	n.r.	Schreiner et al. (2016)
		Groundwater	
Europe	n.r.	29%	Loos et al. (2010)
Czech Republic	28.5%	6.4%	Veverka & Lesinsky (2005)
Italy	35.9%	5.3%	(Paris et al., 2016; Paris et al., 2020)
Europe (only glyphosate and AMPA)	1.3%	0.7%	Horth & Blackmore (2009)
Belgium	65%	n.r.	DEMNA (2017)
Switzerland	>50%	2%	BAFU (2019)

Note: n.r. = not reported.

and the US, mixtures of two to five compounds were usually found (Schreiner et al., 2016). The findings of pesticide cocktails can hardly be compared between different studies, due to differences in the number of substances detected and in the sensitivity of the analytical methods.

Herbicides, insecticides and fungicides differ in their occurrence in the aquatic environment. Herbicides and their metabolites are the dominating pesticide class (Mohaupt et al., 2020; Paris et al., 2016; Schreiner et al., 2016). Their widespread occurrence in water can be attributed to their continuous application in high doses and to their high water solubility. By contrast, insecticides are less frequently found in water, which is attributed to their short, episodic use. Furthermore, most insecticides are lipophilic and bind to sediment and are therefore less frequently detected in grab samples in water. Fungicides occur in water only at low concentrations. An explanation can be found in their application pattern which leads to relatively low but continuous concentrations (Schreiner et al., 2016).

2.8. *Contamination of groundwater*

On average, groundwater contains less pesticides than surface waters (see Table 2). For example, in the Europe-wide study by Mohaupt et al. (2020), herbicides exceeded the safe drinking water limit at 7% of the groundwater sites, compared with 5–15% for surface water. Similarly, insecticides exceeded the safe drinking water limit at less than 1% of the groundwater sites, compared with 3–8% for surface water. Glyphosate and its degradation product AMPA were found 30 times more often in surface than in groundwater (Horth & Blackmore, 2009). Nevertheless, groundwater can also contain considerable pesticide residues. A European-wide study from 2008 analysed groundwater quality in 23 countries at 164 locations (Loos et al., 2010). The safe drinking water threshold for single substances was exceeded in 29% of samples, and the threshold for multiple substances was exceeded in 10% of samples. Data from Switzerland suggest that groundwater is most contaminated in regions with intensive agricultural production (BAFU, 2019).

2.9. *Transition from water into food*

Various studies documented that pesticides reach the soil-crop environment via irrigation (Christou et al., 2019; Huseth & Groves, 2014; Szekacs et al., 2015). The irrigation water can originate from surface or groundwater sources. Another pathway for pesticides in the aquatic environment to reach crop plants is after flooding events. However, the importance of this pathway is only very little studied in the scientific literature. Once in the soil-plant environment, pesticides behave similarly as pesticides taken up from the soil (Christou et al., 2019). For further information, see Chapter "Transition from soil into food".

Please note that pesticides in water may also end up in aquatic organisms used as food. However, this contamination pathway is not treated in the present review.

2.10. *Dynamics of pesticides in the air*

The air is the least understood environmental cjjompartment regarding pesticide occurrence (Van Dijk & Guicherit, 1999). Pesticides can be present in the air in liquid, solid or gaseous form (Kubiak et al., 2008). 'Droplets' are tiny particles of the liquid spray solution, while 'dust' refers to solid particles, primarily soil particles to which pesticides are bound. As the phases of pesticides in air are continuous, we will not discriminate these forms in the rest of the review.

Pesticides enter the air compartment mainly during the process of pesticide spraying. In addition, pesticides already deposited on crops or soil may enter the air compartment through volatilization. Photo-oxidation and other light induced reactions are the main transformation processes for pesticides in air. In the air, pesticides are subject to horizontal and vertical transportation in the atmosphere depending on winds and stability of the atmospheric boundary layer (Kubiak et al., 2008). Pesticides can be transported in the air over a continuous range of distances. For simplicity, however, we distinguish here between 'short-range transport' (less than 1000 m) and 'long-range transport' (more than 1000 m) based on (Kubiak et al., 2008). During rain events, pesticides are washed out of the atmosphere and reach the ground.

Most pesticides are applied to crops using hand-held sprayers, sprayers mounted on tractors, airplanes, helicopters or drones. Various factors including wind speed, humidity and temperature influence the distance of spray drift. Droplet size plays a key role, with small droplets drifting much further than large droplets (Speiser & Kretzschmar, 2021; Zimdahl, 2018). Finally, sprayers for high growing crops such as fruit trees, vineyards or hops cause more drift than sprayers for arable crops and vegetables which grow on the ground (Rautmann et al., 2001). Contamination levels usually decrease exponentially with increasing distance to the source. In the literature, the spray drift values measured by Ganzelmeier et al. (1995) are regularly referred to.

Pesticides in the air may be transported vertically to different atmospheric layers where they can be transported with regional and global winds. Such 'long range transport' can cover distances from a few kilometres up to 1000 km, as a variety of studies from the Netherlands, Denmark, Sweden, Italy and Romania show (Asman et al., 2005; Hofmann et al., 2018; Kreuger et al., 2006; Kreuger et al., 2013; Kubiak et al., 2008; Tarcau et al., 2013; Van Dijk & Guicherit, 1999). Also studies from Germany indicate long-range transport processes for several pesticides, including pendimethalin, chlorothalonil and prosulfocarb (Kruse-Plaß et al., 2021; Kruse-Plaß et al., 2020).

Not only persistent and volatile compounds can be found in air. Several studies demonstrate the occurrence of non-volatile or non-persistent pesticides in air. Hofmann et al. (2019) investigated tree barks in agricultural and remote regions of Germany. They found wide-spread occurrence of various pesticides, including also the non-volatile glyphosate. The authors conclude that glyphosate reached these places while bound to soil particles which had been transported over long distances with the wind (Hofmann et al., 2019). Long-range transport of glyphosate has also been observed in a case study in North America (Rombach et al., 2020). Not only the volatility, but also the persistence of pesticides in air can change, when pesticides are bound to soil particles or other aerosols. Socorro et al. (2016) report aerial half-lives of several days up to more than one month for pesticides bound to soil particles.

2.11. Contamination of the air

There is no recent study about the pesticide contamination of the air at European level. Some older studies reveal pesticide concentrations in air over Europe ranging from pc/m^3 to a few ng/m^3 (Van Dijk & Guicherit, 1999). In a study from 2004, the highest levels of ppDDT in the air were detected in Italy and Russia, even though these compounds were already forbidden at the time (Jaward et al., 2004).

More detailed studies exist on a national scale. In Germany, bark samples from 47 locations were analysed for the presence of pesticide residues (Hofmann et al., 2019). Pendimethalin was found at 89% of the locations, DDT at 72%, prosulfocarb at 66%, prothioconazol-desthio at 64%, and lindane and glyphosate both at 55% of the locations (Kruse-Plaß et al., 2021; Kruse-Plaß et al., 2020). Also the French pesticide monitoring program finds glyphosate and lindane in over 80% of all samples, although at low concentrations (Marliere et al., 2020). Substances with the highest concentrations are prosulfocarb and folpet. A monitoring study of the Belgian Walloon region finds triallate, pendimethaline, chlorothalonil, captane and benfluraline in over 50% of samples (Giusti et al., 2018). The number of substances was larger in agricultural regions than in remote areas. The studies further show, that pesticide concentration in the air is correlates with patterns of pesticide application (Giusti et al., 2018; Hofmann et al., 2018).

These studies show that pesticides are generally present in the air in Europe. They can be found not only in intensively farmed regions, but also in remote areas. Moreover, pesticide contamination of air is likely to vary between areas, countries and years (Kruse-Plaß et al., 2021; Kruse-Plaß et al., 2020). The evidence indicates that long-range transport of pesticides is common, however these processes are yet poorly understood and need further research in order to better understand the sources and sinks of aerial contamination.

2.12. Contamination of rainwater

Measurements of rainwater allow to estimate pesticide transport from the atmosphere to the ground. A large number of pesticides can be found in rainwater throughout Europe. Lindane and atrazine are most frequently detected substances (Kubiak et al., 2008). Mean concentrations are usually in the ng/l scale. However, peak concentrations may reach the level of μg/l rainwater. In general, there is a good correlation between application times and the occurrence of pesticides in rainwater samples (Dubus et al., 2000; Kubiak et al., 2008). As an exception, however, some compounds could still be found months after the application season, which indicates volatilization from treated soil or plants. Apart from the pesticide concentration in the atmosphere, the concentration in rainwater also depends on the amount, intensity and timing of rainfall. High pesticide concentration in rainwater is observed after long dry periods and in the beginning of a rainfall event (Kubiak et al., 2008).

Three national studies investigate the occurrence of pesticides in rainwater. Kreuger et al. (2006) and Kreuger et al. (2013) report pesticide concentrations in rainwater in Sweden in the low ng/l range, with occasional detections above 0.1 μg/L. The authors calculate that the deposited amounts correspond to 0.1–0.0004% of the applied dose. A Danish study reports deposition of pendimethalin and for desethylterbuthylazine of 0.3 g/ha per year (Asman et al., 2005). In the Greek Axios River Basin Charizopoulos and Papadopoulou-Mourkidou (1999) observed seasonal trends in the concentration of pesticides in rainwater and calculated a maximum annual deposition ranging from 51 to 395 μg/m^2 soil (=0.5–4 g/ha/yr).

2.13. Transition from air into food

Collins et al. (2006) describe three major uptake pathways for pesticides in the air to enter crops. First, pesticides in the gaseous phase can enter plants via the stomata or diffuse through the cuticula. The chemicals will then diffuse in the gaseous intercellular spaces or dissolve in the aqueous or lipophilic phases inside the plant. Lipophilic substances are taken up easier than non-lipophilic substances. A second pathway for pesticide uptake is particulate deposition on plants. Pesticides bound to particulate matter can be deposited on plant surfaces and diffuse into the interior where they are adsorbed or move further through the plant. Particulate deposition is influenced by wind speed, the properties of the particle and plant surface properties (Smith & Jones, 2000). A third uptake pathway for pesticides in the air is wet deposition via rainwater. Pesticides in rainwater can either be directly deposited on plant surfaces and enter the plant, or reach the soil. While the first pathway is likely to be of little importance (Collins et al., 2006), the storage of pesticides in soil water is probably a major contamination pathway for crops. More information regarding the uptake processes from soil water is described under section ‘Transition from soil into food’.

Only few studies trace pesticide contamination in food back to spray drift, contaminated rainwater or long-range transport of contaminated air. Spray drift has been found to cause residues of organophosphorus pesticides ranging from 3.1 to 7.6 μg/kg on non-target okra plants in Ghana (Essumang et al., 2013). The authors trace the contamination back to the application of pesticides in the neighbouring watermelon fields and subsequent spray drift. Long-range transport of pendimethalin has been found to cause residues a study conducted by Hofmann and Schlechtriemen (2015). The authors found pendimethalin residues between 0.02 and 0.06 mg/kg on fennel and kale grown in a nature reserve in Brandenburg. The sites are at least several kilometres away from the next application site of pendimethalin. The study relates the residue findings to pendimethalin concentrations in air of 4–16 ng/m^3 at the cropping sites and assumes long range transport of pendimethalin in the environment. A case study in the USA and Canada traced glyphosate levels in organic Khorasan-wheat back to contaminated rainwater and air (Rombach et al., 2020). According to this study, glyphosate is transported in the long and short range, thus causing glyphosate residues in organic wheat from 0.01 to 0.076 mg/kg. A field study in the Czech Republic analysed the uptake of polychlorinated biphenyls and OCPs in radishes. The air at both sites showed high levels of HCHs (5.1–52 ng/m^3) and HCB (1–14 ng/m^3) and the authors conclude that the pesticides were probably taken up from the air (Mikes et al., 2009).

Apart from the studies mentioned above, scientific field trials analysing uptake of pesticides in non-target crop plants from air or rainwater are scarce. More studies are needed in order to better understand the impact of pesticides in air and transition into food.

2.14. Post-harvest environment of food

In this review, we refer to equipment and installations for storage, transport and processing of food as ‘post-harvest environment’. Such equipment and installations are subject to insecticide treatments for the control of storage pests, either in the empty equipment and installations or on the conventional food. In addition, they may be polluted by pesticides adhering to conventional food, being stored, transported or processed in these facilities. The risk of such contaminations strongly depends on the foods handled previously and on the precautionary measures taken to avoid such contaminations. Moreover, the type of processing activity, installation or equipment and the food in question are also important factors. (Bickel & Speiser, 2020; Nerín et al., 2016). Experience from the organic sector indicates that post-harvest pest control, for example to control storage pests, is a major contamination pathway (Bögli & Bickel, 2018; Landau & Fassbind, 2011). In international cargo freight, containers are frequently disinfected with fumigants such as methyl bromide or phosphine (Baur et al., 2015). Nerín et al. (2007) found that the fumigant methyl bromide is able to penetrate through plastic barriers, thereby potentially contaminating the carried food. Similar behaviour is suspected for other toxic fumigants. Except for a few selected examples, the extent of pesticide contamination of foods transported in fumigated containers or in containers fumigated in earlier shipping, is barely studied in the scientific literature.

Cross-contamination between treated and non-treated lots is another major cause of pesticide contamination, which can occur in the post-harvest environment. This problem is caused by conventional crops containing pesticides, which can contaminate buildings and installations of the post-harvest environment. If later organic food is treated or stored in these buildings or installations, contamination may occur. Here, adapted cleaning measures and/or separate processing lines contribute significantly to risk reduction. In a study in Switzerland, Ortelli et al. (2005) found post-harvest fungicides on 13% of sampled organic citrus fruit. The authors suspect cross-contamination during processing or storage to be responsible for the findings. Scientific studies analysing such processes are scarce. Insect repellents used by harvesting personnel and insecticides used against household pests or for vector control are also potential sources for residues. However, these sources are mainly important under tropical conditions and have lower importance in Europe (Association of Ecological Food Producers, 2020 Bio Suisse, 2021). Overall, the knowledge on contamination processes of non-target foods in the post-harvest environment is very limited and more research is needed.

2.15. Implications for the organic sector

2.15.1. Omnipresent but variable contamination risks

The scientific literature reviewed in this study makes clear that pesticides are omnipresent in the environment. Pesticides are deliberately released to the environment by conventional production systems. Environmental migration processes distribute them to other environmental compartments including organic fields, where they remain for variable time periods. In some cases, environmental contamination can be traced to recent pesticide use in the same region (Ccanccapa et al., 2016; Chiaia-Hernandez et al., 2017). However, there are also examples where pesticide residues are caused by their use in distant areas (Hvězdová et al., 2018; Thiombane et al., 2018). Also, some pesticides such as the OCPs have been banned from use decades ago and still cause residues today. Long-range air transport has the power to distribute pesticides over significant distances to non-target sites (Hofmann et al., 2019; Kreuger et al., 2013; Kruse-Plaß et al., 2021). However, this process is yet poorly understood. Finally, pesticides are also found in the post-harvest environment such as buildings and installations for storage, transportation and processing of food (Bögli & Bickel, 2018; Landau & Fassbind, 2011). Fig. 1 provides an overview of the distribution processes of pesticides in the environment and how they can enter the organic food chain.

In addition, pesticide contamination of the environment can show very different patterns, depending on the pathways and mechanisms causing it. Spray drift typically causes short-term, small scale contamination (Ganzelmeier et al., 1995). On the other hand, persistent pesticides such as atrazine, simazine and OCPs are present over a long time period and found in a wide variety of places (Hilber et al., 2008; Paris et al., 2016; Riedo et al., 2021). This review shows that pesticide environmental contamination depends on various factors: The physico-chemical and biological properties of individual compounds, agronomic practices including the choice of crop plants, soil cultivation methods, the choice and application method of pesticides and local environmental conditions. Therefore, pesticide environmental contamination is characterized by great variability in contamination load and

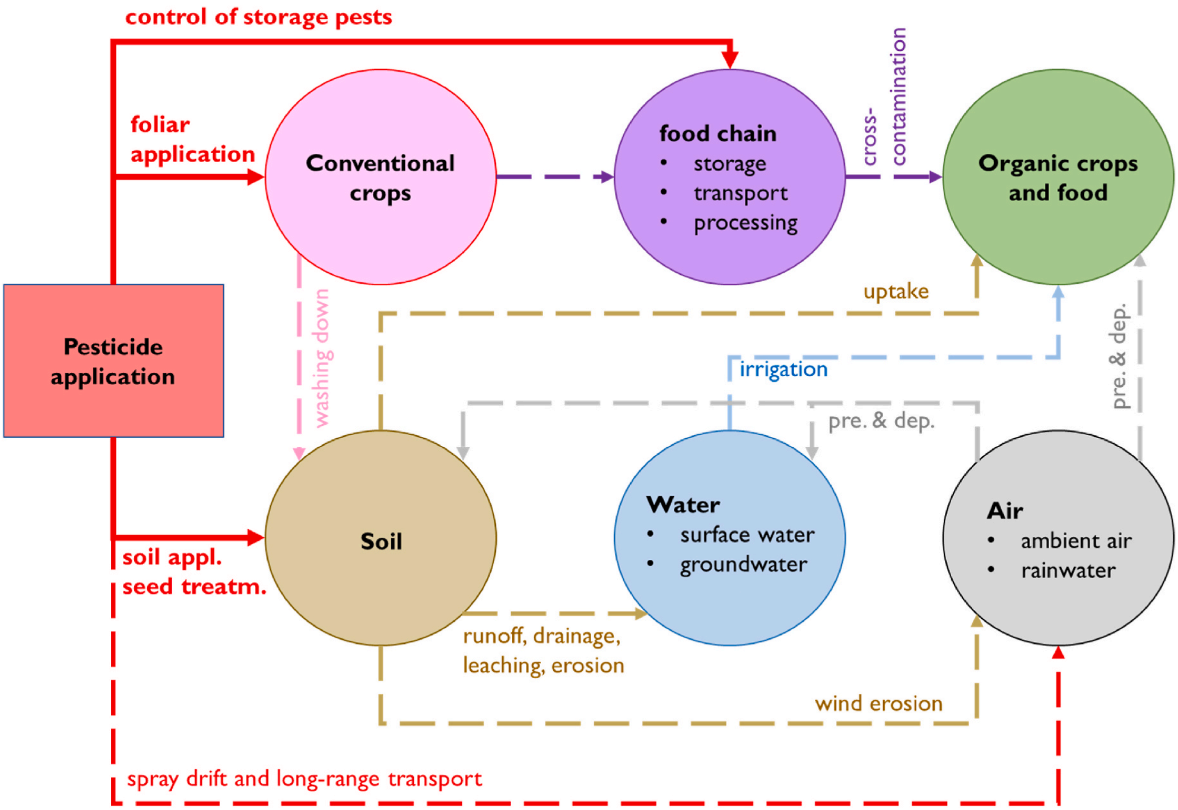

Fig. 1. Distribution of synthetic pesticides between environmental compartments and the food chain. The red square illustrates the input of pesticides into the system, circles illustrate the environmental compartments and the food chain, while arrows illustrate distribution processes. Solid arrows illustrate deliberate pesticide application, while dotted arrows illustrate unintended distribution. For simplicity, the graph is limited to organic crops and food. Abbreviations: 'soil appl.' = soil application; 'seed treatm.' = seed treatment; 'pre. & dep.' = precipitation and deposition.

type of compound.

We would like to highlight, that not all pesticides are studied to the same extent. Methods to analyse occurrence of pesticides are always limited to a selection of substances and do rarely cover all of the pesticides which have been used historically and currently. Therefore, large uncertainties remain regarding the true pesticide contamination of the environment, their dynamics and the contamination risks for the food chain.

2.16. Risk reduction in organic production

The more is known about the occurrence of pesticides in the environment, the better organic operators can avoid such risks. Based on the review, we propose various risk reduction measures for the organic sector, many of which are already implemented. The risk of residues from contaminated soil can be assessed by analysing the soil for persistent pesticides such as OCPs. This is especially advised when planting cucurbits (Hilber et al., 2008). As the contamination of spray drift decreases exponentially with the distance to the source (Ganzelmeier et al., 1995), spray drift can be managed with buffer zones and hedges. In addition, organic farmers can ask their conventional neighbours to not spray during windy or very dry conditions (Speiser & Kretzschmar, 2021). Finally, contaminations during processing can be avoided with separate lines for conventional and organic produce or with thorough cleaning between conventional and organic lots. In conclusion, organic operators have some possibilities to reduce the risk for pesticide residues, but cannot completely eliminate all contamination risks.

Prevention of pesticide residues by implementing risk reduction measures, monitoring, as well as investigating and documenting residue cases causes substantial costs to the organic sector. A non-representative survey estimates that the total costs of pesticide residues and of measures to prevent them range from 0.3% to 3.5% of the organic turnover (Speiser et al., 2020). In addition, the establishment and maintenance of adequate quality assurance systems requires highly specialised know-how and is therefore particularly a challenge for small operators. More research is needed on the effectiveness and the cost-benefit ratio of such preventive measures.

2.17. Implications for the investigation of residue cases

The findings of this review provide implications for the investigation of residue cases on organic food which aim to distinguish fraud from unavoidable contamination. First, given the ubiquitous potential for environmental contamination, a risk for pesticide residues exists on all organic crops. Consequently, residue findings in organic food may indicate the possibility of fraud, but are not evidence for it. Thus a ‘zero-tolerance’ approach for pesticide residues on organic food, as proposed by certain actors (Milan et al., 2019), cannot be met by organic operators at the moment. Second, environmental pesticide contamination is substance-dependent and subject to temporal and spatial variability. Thus, it is currently not possible to estimate the residue risk of individual farms or fields. In addition, also the type of crop and agricultural practices influence residues. Applying a residue concentration threshold to distinguish between cases of fraud and unavoidable contamination for all pesticides is not adequate in view of this variability.

More reliable answers can be obtained with an investigation, where evidence for all possible origins (fraudulent and other) of the particular residue is collected. Such an investigation needs to be tailored to the individual case, taking into account aspects such as the crop, the pesticide and the circumstances and may include (i) traceability of the lot, (ii) observations in the field (e.g. absence of weeds in case of suspected herbicide application), (iii) additional sampling of foliage, soil, equipment or crops, (iv) literature review for environmental behaviour of the substance that might lead to technically unavoidable contamination, (v) alternative sources for the compound (e.g. natural occurrence), (vi) in the case of processed food: concentration or dilution factors. Such investigations usually indicate that either fraud or technically unavoidable contamination is more likely, but they rarely give a conclusive answer. In such cases, the certification decision is a challenge for organic certification bodies and control authorities. Future research should focus on transition studies of pesticides from the environment to non-target crops. Such studies can support the decision making by organic certifiers.

3. Conclusions

As this review shows, pesticides can be found in all environmental compartments as well as in the post-harvest environment of the food chain. Considering the variability of pesticide contamination and its dependence on substances, agricultural practices and environmental factors, large uncertainties remain regarding pesticide contamination of the environment and the contamination risks for the organic food chain. Nevertheless, organic operators can take certain measures to reduce the risks of pesticide contamination of their products. However, some contamination remains technically unavoidable and risk minimization measures and monitoring of residues come at a substantial cost for the organic sector.

In Europe, a new regulation on organic production has entered into force in 2022 (EU, 2018/848). New rules regarding the handling of pesticide residues are due to be developed under this regulation, but are still controversially debated. To this discussion, the present review can contribute the following: (i) A ‘zero-tolerance’ approach cannot be met by organic production at the moment, due to the ubiquitous risks of pesticide contamination from the environment. (ii) A simple numerical threshold value for all pesticides is not adequate in view of the diversity of regions, crops and compounds concerned. Ultimately, in many residue cases, it remains a challenge to determine the precise origin of the residue and whether it is due to non-compliance with production rules or technically unavoidable. Practice-oriented research regarding the transition of pesticides from the environment into the food chain can provide useful background information that supports adequate assessment of residue cases.

Authors contributions

All authors contributed to the conception for the review article. Literature search and literature analysis was performed by Mirjam Schleiffer. A first draft of the manuscript was written by Mirjam Schleiffer. Bernhard Speiser commented on previous versions of the manuscript and double-checked references. Both authors critically revised the manuscript and approved the final version.

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Declaration of competing interest

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Data availability

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