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Pesticide resurrection

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The actual health risk and associated diseases induced by the presence of pesticides and other organic pollutants in most environmental media worldwide are largely underestimated because current analytical methods measure only the extractable fraction of the pollutant, whereas there is now ample evidence that a large part of pollutants are trapped as bound residues in complex organo-mineral matrixes, and are thus unanalyzable by current tools. Once trapped, pollutants can be stored for long periods, then released in the biosphere upon suitable conditions such as soil erosion and climate change. This ‘resurrection’ and underlying mechanisms of trapping, storage and release are poorly known. Here we discuss the limit of knowledge on bound pollutants and their later release, with examples of organochlorines in glaciers, vineyards and banana plantations. Unexpectedly, soil application of a modern pesticide such as glyphosate, can lead to the resurrection of ancient, actually banned pesticides such as chlordecone and dichlorodiphenyltrichloroethane (DDT) (Fig. 1).

Silent spring

The *Silent spring* book by Rachel Carson, first published in 1962, marks the start of public awareness of global pollution (Carson 2002; Hallmann et al. 2014). Pesticides and herbicides are used to prevent pests and weeds to damage crops. In 2018, the global agricultural use of pesticides has been estimated to exceed 4 million tons of active ingredients (FAO 2020). This massive use has led to a global soil contamination, as illustrated by the widespread pollution of soils in Europe (Silva et al. 2019). The discovery of endocrine-disrupting chemicals has also induced serious concerns for public health (Diamanti-Kandarakis et al. 2009).

Pollution ignores borders

Pollution is not limited to the location where pesticides are applied. Indeed, pesticides are partly transferred to other media, e.g., water and air, then carried to other locations. Pesticides are found in remote and supposedly pristine regions of the Earth. For instance, in the 1960’s, researchers

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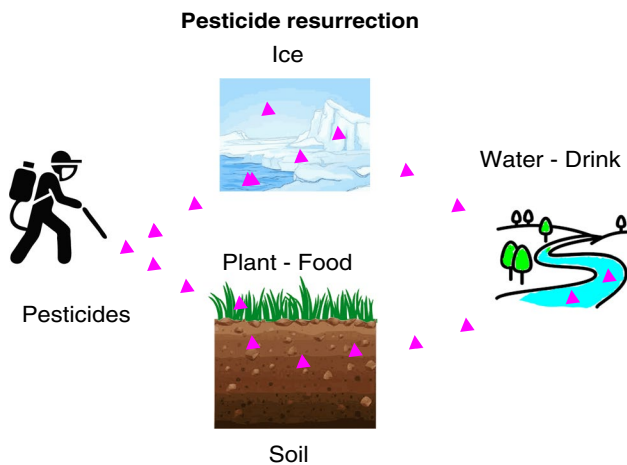


Fig. 1 Pesticides and other pollutants are trapped in soils, ice and other environmental media by unknown mechanisms for unknown time periods, then are eventually released in the biosphere, thus inducing a second toxic event

detected pesticides in Antarctica, which means that contamination has spread through air and water to the most remote and isolated parts of the Earth (Tatton and Ruzicka 1967; Potapowicz et al. 2020). Scientists have then realized that any ecosystem compartment is not closed, and that pollutants can be transferred from one compartment to another under suitable conditions. Accordingly, cropped and pastured soils are of particular concern because soils are intimately connected to surface water, groundwater, atmosphere, wildlife and even seawater (Vereecken 2005; Chang et al. 2011; Banwart et al. 2013; Della Rossa et al. 2017; Mottes et al. 2019).

Dormant pollution

Soils are also the main support for food production systems and other ecosystem services (Dominati et al. 2010). Once a pesticide molecule enters a soil, many possible phenomena can happen: the pesticide can be immobilized in minerals or organic matter, degraded by bacteria, and transferred to water, air or the pest target. Yet the underlying mechanisms are still poorly known (Lichtfouse 1997). In particular, there is few knowledge on pollutant ‘resurrection’, a process by which a pollutant stays encapsulated in the soil for a certain time—may be years or more?—then is released into the biosphere, inducing another toxic event. This soil remobilization of a pesticide has been suspected following the detection of substances that have been banned since a long time, e.g. dichlorodiphenyltrichloroethane (DDT) in French vineyards, atrazine in corn and sugarcane cropped areas, and chlordecone in banana cropped area (Dalton et al. 2014; Sabatier et al. 2014; Lesueur Jannoyer et al. 2016). There

is now ample evidence for pollution of water and sediments from water-monitoring systems in agricultural areas (Konstantinou et al. 2006; Stehle and Schulz 2015; Mottes et al. 2017; de Souza et al. 2020; Didoné et al. 2021).

Pesticide toxicity

Pesticide pollution does not disappear with the ban of pesticides. For example, DDT, chlordecone and atrazine have persisted in ecosystems at least 30 years after their ban. Farmers and population are thus exposed to secondary waves of pesticides by direct exposure, e.g., in air, when drinking polluted water or eating contaminated food (Aktar et al. 2009; Fantke et al. 2011; Toan et al. 2013; Clostre et al. 2014, 2017; Damalas and Koutroubas 2016). Pesticide residues in food and drinking water represent a serious threat to public health. Indeed, residues can compromise neuroendocrine development in unborn and newborn children and lead to chronic kidney diseases in the later life. Recent studies have also shown the influence of pesticides on breast cancers, and their toxicity for non-targeted organisms such as human populations (Gill et al. 2018; Singh et al. 2018; Xu et al. 2018; Kass et al. 2020). Biodiversity is also altered after pesticide application, as shown by the changes in arthropod behavior (de Jong et al. 2008; Brittain et al. 2010; Geiger et al. 2010; Ewald et al. 2015). Soil also influences the transport of pesticides to pollinators during tillage and bee queen hibernation in soils (Raine 2018). Biologically-significant concentrations of banned organochlorine pesticides (OCPs) still continue to be reported in wildlife, and are of particular concern for the highest trophic levels due the process of biomagnification (Humphries et al. 2021).

Bound residues

In 1989, during one day, we extracted three times soil samples that have been incubated with ¹⁴C-atrazine. As expected, the radioactivity levels of the extract decreased in the second and third extracts. But, surprisingly, when we did a fourth extraction the next day, the extract radioactivity was higher than the previous extract. Something was going on here, probably a change in soil conditions overnight, which induced the release of bound atrazine (personnal communication from Prof. Michel Schiavon in 1996).

Bound residues refer to compounds, unchanged or partly degraded, that are trapped in complex media, e.g., soil, sediment, food and living organisms, by various mechanisms such as chemical binding or physical encapsulation. Binding mechanisms were first considered as pathways for the removal of chemicals—and their associated toxicity—from

the biosphere (Karasali and Pavlidis 2021). However, actually many questions remain unanswered or partly answered about bound residues, for instance:

- What are their chemical structure?
- How are they formed and trapped?
- What percentage of the applied compound is trapped?
- Are they partly degraded or not before trapping?
- To which compounds are they bound? How?
- How much time do they stay entrapped?
- Will they be released in the short or long run?
- Where do they go upon release?
- What are the conditions needed for their release?
- What is their toxicological impact?

Some control processes are known but the scientific community remains actually unable to clearly understand and quantify bound residues, notably due the limits of analytical methods. This is for instance the case for non-extractable residues, which are commonly observed by indirect methods, e.g., ^{14}C tracing and nuclear magnetic resonance (NMR), but are not unambiguously characterized at the molecular level because the solids in which they are trapped are too complex for current analytical tools. One of the consequences of this analytical limit is that it is actually impossible to determine the true concentration of a pesticide—or any organic compound—in a complex media because a part of this pesticide is bound and unanalyzable.

When pesticides are applied to soils, they form non-extractable residues, and a fraction of these residues, which may reach 70%, is strongly bound to soil components, mainly to organic matter and clays (Barriuso et al. 2008; Woignier et al. 2012, 2019; Karasali and Pavlidis 2021). For most pesticides, the binding processes are not known, while the potential toxicity due to the presence of non-extractable pesticide residues is high (Gevao et al. 2000). Therefore, these hardly bound residues should be better taken into account to assess the environmental fate of pesticides (Schäffer et al. 2018). In addition, analytical methods should be developed to identify and quantify bound residues (Loeffler et al. 2020). For instance, molecular level ^{13}C -tracing has allowed to evidence the occurrence of temporal pools of the same organic compound in various fractions of the same soil sample (Lichtfouse et al. 1998; Lichtfouse 1999, 2012). Moreover, cleavage of sedimentary matter with Na^{18}OH has shown that DDT metabolites are bound to organic macromolecules (Kalathoor et al. 2015). A better knowledge of binding mechanisms would help to design remediation techniques such as extraction (Karasali and Pavlidis 2021).

Besides extraction, biological and chemical degradation are two other methods to remove pesticides from soil and wastewater, yet there is a lack of analytical methods and standards for quantifying degradation products (Andreu and Picó 2004).

Typically, most published articles investigating pesticide ‘degradation’ in complex media are only measuring the remaining, extractable concentration of the parent molecule to measure a ‘removal’ percentage by subtraction. Here, using the term ‘degradation’ is wrong if degradation products are not identified. Moreover, even after 100% removal of a pollutant—from the extractable fraction—a major part of the parent molecule may occur, unchanged or degraded, in the bound, unanalyzable residues. Similarly, even when the parent molecule is not detected anymore in the analyzable fraction, metabolites are still widely found, e.g., as deethylatrazine for atrazine, aminomethylphosphonic acid (AMPA) for glyphosate, dichlorodiphenyldichloroethylene (DDE) for DDT (Sabatier et al. 2014). Overall, a large fraction of pesticides and other organic pollutants is actually overlooked and most probably presents a higher toxicological risk, notably in the long run, than the risk assessed by current classical analysis.

Pesticide resurrection

Under suitable changing conditions, trapped pollutants may be released in the biosphere (Fig. 2). For example, ancient organochlorines trapped in Antarctica and Alpine glaciers

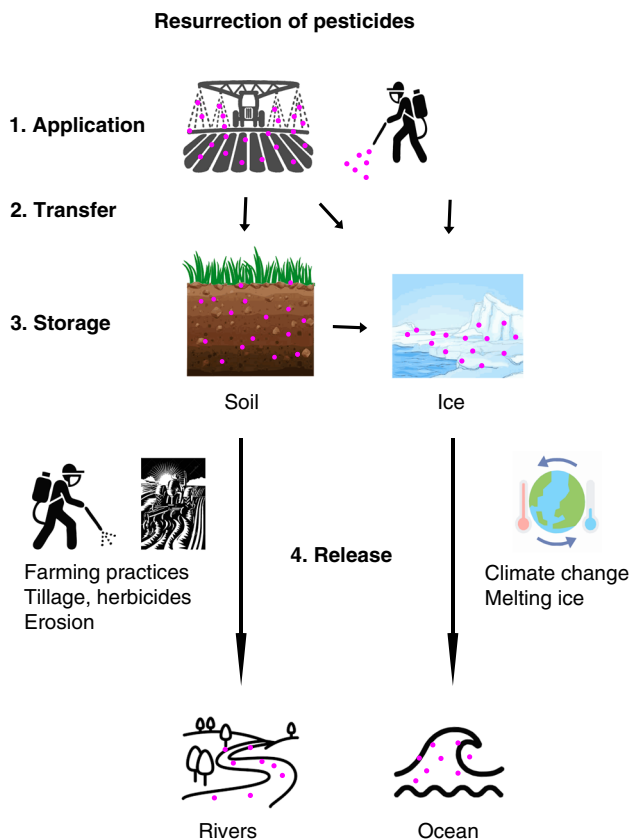


Fig. 2 Examples of pesticide resurrection pathways

may be released by ice melting in the context of climate change (Bettinetti et al. 2008; Schmid et al. 2011; Potapowicz et al. 2020). Climate seasonality is also a likely cause of pollutant release. For instance, modeling suggests that the Indian summer monsoon induces the revolatilization of soil-accumulated pollutants (Lammel et al. 2018). Another unexpected finding is the ‘resurrection’ of ancient pesticides that have been trapped in soils some time ago, e.g., DDT and chlordecone, by an actual application of a modern pesticide, e.g., glyphosate, a post-emergence herbicide, in French vineyards and banana plantations in the French West Indies (Sabatier et al. 2014, 2021). This implies that pesticides can be trapped, stored then released by glyphosate-induced soil erosion. This phenomenon is known as the soil memory effect (Foucher et al. 2020). More research is actually needed on the nature of bound pollutants, their degree of freedom and on the mechanisms ruling their trapping, storage, release and ‘resurrected toxicity’ in the biosphere. This should help to design remediation methods and farming practices that lessen health hazards.

Farming practices

In order to avoid pesticide resurrection, novel farming practices that no longer rely on the use of harmful pesticides should be designed with farmers (Fig. 3). For instance, in the French West Indies, chlordecone has been successfully replaced by pheromone traps, and by fallow management in banana cropping systems. Farmers are also better managing weeds by using cover crops, for instance, thus avoiding the use of herbicides. Tools and methods are being developed to help local stakeholders to coordinate their actions at the local scale to help reduce pesticide uses and the associated pollution (Della Rossa 2020). This demonstrates that innovation makes it possible to overcome environmental-related pesticide issues.

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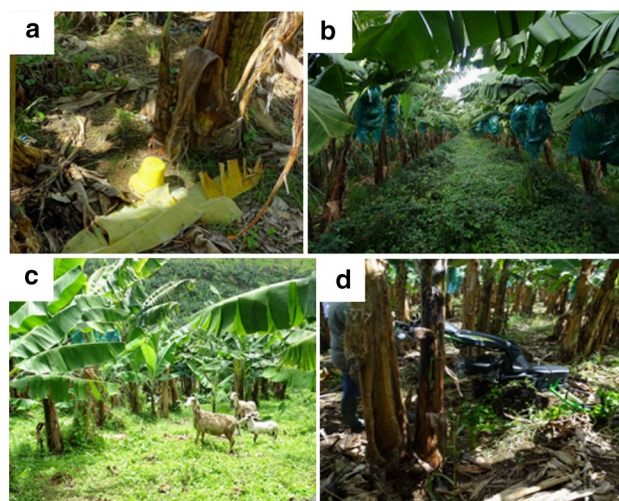


Fig. 3 Innovations developed to reduce pesticide application in the French West Indies. a) A pheromone trap has replaced chlordecone application in banana fields. b) A cover crop, *Desmodium spp.*, is grown in a banana field to avoid herbicide application. c) A banana-livestock association reduces herbicide use and uses weed as sheep fodder. d) Weeds are removed by small mechanical tools, thus limiting herbicide application. Photography credits: Charles Mottes

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