Food and Agriculture
Organization of the
United Nations

SOIL POLLUTION: A HIDDEN REALITY

GLOBAL SOIL
PARTNERSHIP

SOIL POLLUTION A HIDDEN REALITY

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FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS
Rome, 2018

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Recommended citation

Rodríguez-Eugenio, N., McLaughlin, M. and Pennock, D. 2018.
Soil Pollution: a hidden reality. Rome, FAO. 142 pp.

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ISBN 978-92-5-130505-8

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EXECUTIVE SUMMARY

“Soil pollution” refers to the presence in the soil of a chemical or substance out of place and/or present at a **higher than normal concentration** that has adverse effects on any non-targeted organism. Soil pollution often cannot be directly assessed or visually perceived, making it a hidden danger.

The Status of the World's Soil Resources Report (SWSR) identified soil pollution as one of the main soil threats affecting global soils and the ecosystems services provided by them.

Concerns about soil pollution are growing in every region. Recently, the United Nations Environmental Assembly (UNEA-3) adopted a resolution calling for accelerated actions and collaboration to address and manage soil pollution. This consensus, achieved by more than 170 countries, is a clear sign of the global relevance of soil pollution and of the willingness of these countries to develop concrete solutions to address the causes and impacts of this major threat.

The main anthropogenic sources of soil pollution are the chemicals used in or produced as byproducts of industrial activities, domestic, livestock and municipal wastes (including wastewater), agrochemicals, and petroleum-derived products. These chemicals are released to the environment accidentally, for example from oil spills or leaching from landfills, or intentionally, as is the case with the use of fertilizers and pesticides, irrigation with untreated wastewater, or land application of sewage sludge. Soil pollution also results from atmospheric deposition from smelting, transportation, spray drift from pesticide applications and incomplete combustion of many substances as well as radionuclide deposition from atmospheric weapons testing and nuclear accidents. New concerns are being raised about emerging pollutants such as pharmaceuticals, endocrine disruptors, hormones and toxins, among others, and biological pollutants, such as micropollutants in soils, which include bacteria and viruses.

Based on scientific evidence, soil pollution can severely degrade the major ecosystem services provided by soil. Soil pollution reduces food security by both reducing crop yields due to toxic levels of contaminants and by causing crops produced from polluted soils to be unsafe for consumption by animals and humans. Many contaminants (including major nutrients such as nitrogen and phosphorus) are transported from the soil to surface waters and ground water, causing great environmental harm through eutrophication and direct human health issues due to polluted drinking water. Pollutants also directly harm soil microorganisms and larger soil-dwelling organisms and hence affect soil biodiversity and the services provided by the affected organisms.

The results of scientific research demonstrate that soil pollution directly affects human health. Risks to human health arise from contamination from elements such as arsenic, lead, and cadmium, organic chemicals such as PCBs (polychlorinated biphenyls) and PAHs (polycyclic aromatic hydrocarbons), and pharmaceuticals such as antibiotics. The health risks associated with the widespread soil contamination by radionuclides from the Chernobyl disaster in 1986 are an enduring memory for many people.

Remediation of polluted soils is essential, and research continues to develop novel, science-based remediation methods. Risk assessment approaches are similar worldwide and consist of a series of steps to be taken to identify and evaluate whether natural or human-made substances are responsible for polluting the soil, and the extent to which that pollution is posing a risk to the environment and to human health. Increasingly expensive physical remediation methods such as chemical inactivation or sequestration in landfills are being replaced by science-based biological methods such as enhanced microbial degradation or phytoremediation.

FAO's *Revised World Soil Charter* recommends that national governments implement regulations on soil pollution and limit the accumulation of contaminants beyond established levels in order to guarantee human health and wellbeing, a healthy environment and safe food. Governments are also urged to facilitate remediation of contaminated soils that exceed levels established to protect the health of humans and the environment. It is also essential to limit pollution from agricultural sources by the global implementation of sustainable soil management practices.

This book aims to summarise the state of the art of soil pollution, and to identify the main pollutants and their sources affecting human health and the environment, paying special attention to those pollutants that are present in agricultural systems and that reach humans through the food chain. It concludes with some case studies of the best available techniques for assessing and remediating contaminated soils.

This book has been developed within the framework of the Global Symposium on Soil Pollution (GSOP18), identifying the main gaps in knowledge on soil pollution worldwide and serving as a basis for future discussions.

GLOSSARY

Contaminant: substance or agent present in the soil as a result of human activity (ISO, 2013).

Leaching: the dissolution and movement of dissolved substances by water (ISO, 2013).

Parent material: The original material (mineral and/or organic) from which soil developed by pedogenetic processes.

Persistent organic pollutant (POP): Synthesized carbon-based compounds from agrochemicals and industrial products that generally biodegrade very poorly and most of which will bioaccumulate in tissues of organisms. Some pesticides are POPs, as are Polychlorinated dibenzodioxins (PCDDs), Polychlorinated dibenzofurans (PCDFs), Polychlorinated biphenyls (PCBs), and Polycyclic aromatic hydrocarbons (PAHs).

Soil: the upper layer of the Earth's crust transformed by weathering and physical/chemical and biological processes. It is composed of mineral particles, organic matter, water, air and living organisms organized in genetic soil horizons (ISO, 2013).

Soil ecosystem functions: description of the significance of soils to humans and the environment. Examples are: (1) control of substance and energy cycles within ecosystems; (2) basis for the life of plants, animals and man; (3) basis for the stability of buildings and roads; (4) basis for agriculture and forestry; (5) carrier of genetic reservoir; (6) document of natural history; and (7) archaeological and paleo-ecological document (ISO, 2013).

Soil health: the continued capacity of the soil to function as a vital living system, within ecosystem and land-use boundaries, to sustain biological productivity, promote the quality of air and water environments, and maintain plant, animal, and human health (Doran, Stamatiadis and Haberern, 2002).

Soil ecosystem services: the capacity of natural processes and components to provide goods and services that satisfy human needs, directly or indirectly (Groot, 1992).

Food security: it is defined as the availability, access, utilization and stability of food supply.

Soil contamination: occurs when the concentration of a chemical or substance is higher than would occur naturally but is not necessarily causing harm (this volume).

Soil pollution: refers to the presence of a chemical or substance out of place and/or present at higher than normal concentration that has adverse effects on any non-targeted organism (this volume).

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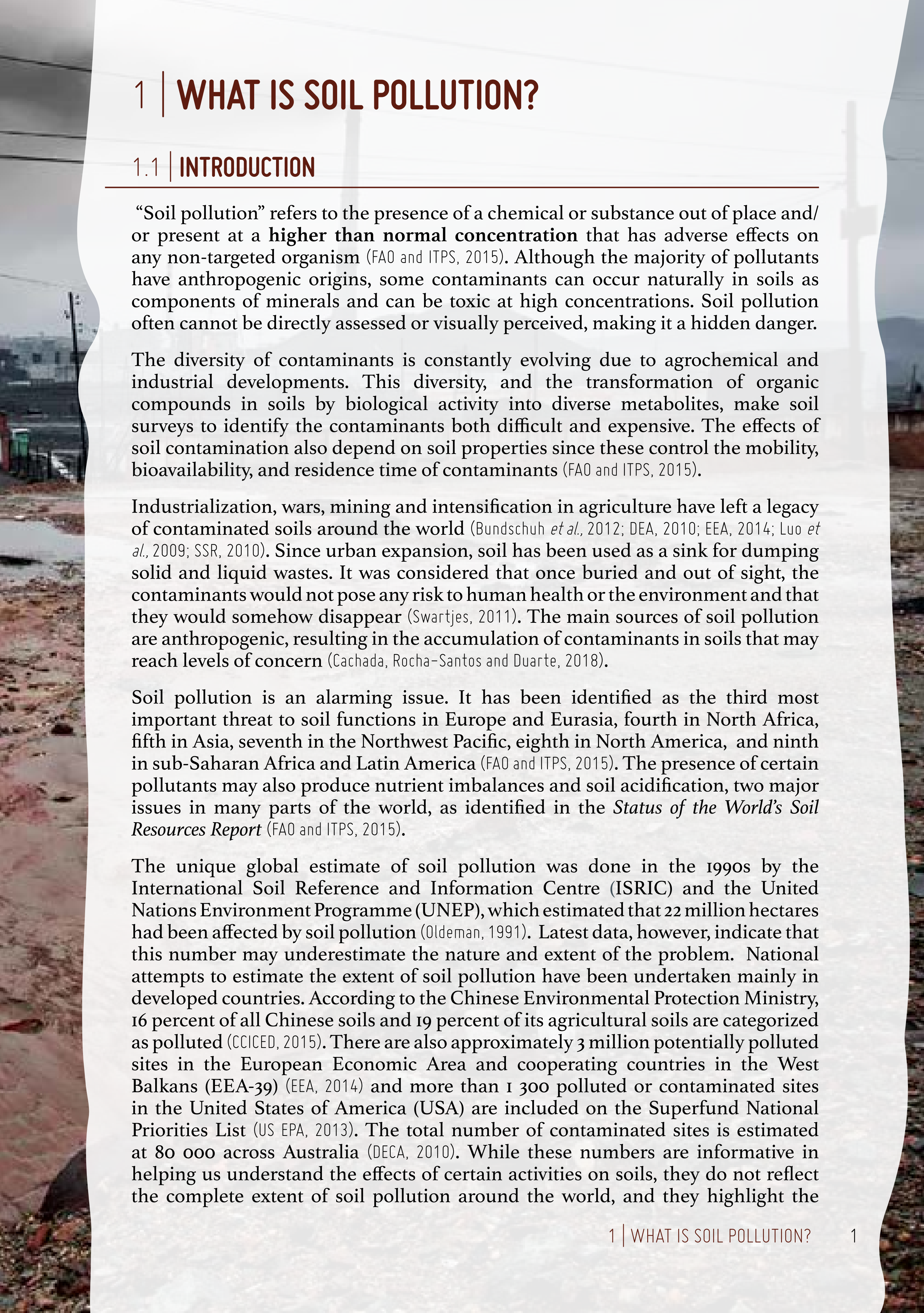

1 | WHAT IS SOIL POLLUTION?

1.1 | INTRODUCTION

"Soil pollution" refers to the presence of a chemical or substance out of place and/or present at a **higher than normal concentration** that has adverse effects on any non-targeted organism (FAO and ITPS, 2015). Although the majority of pollutants have anthropogenic origins, some contaminants can occur naturally in soils as components of minerals and can be toxic at high concentrations. Soil pollution often cannot be directly assessed or visually perceived, making it a hidden danger.

The diversity of contaminants is constantly evolving due to agrochemical and industrial developments. This diversity, and the transformation of organic compounds in soils by biological activity into diverse metabolites, make soil surveys to identify the contaminants both difficult and expensive. The effects of soil contamination also depend on soil properties since these control the mobility, bioavailability, and residence time of contaminants (FAO and ITPS, 2015).

Industrialization, wars, mining and intensification in agriculture have left a legacy of contaminated soils around the world (Bundschuh *et al.*, 2012; DEA, 2010; EEA, 2014; Luo *et al.*, 2009; SSR, 2010). Since urban expansion, soil has been used as a sink for dumping solid and liquid wastes. It was considered that once buried and out of sight, the contaminants would not pose any risk to human health or the environment and that they would somehow disappear (Swartjes, 2011). The main sources of soil pollution are anthropogenic, resulting in the accumulation of contaminants in soils that may reach levels of concern (Cachada, Rocha-Santos and Duarte, 2018).

Soil pollution is an alarming issue. It has been identified as the third most important threat to soil functions in Europe and Eurasia, fourth in North Africa, fifth in Asia, seventh in the Northwest Pacific, eighth in North America, and ninth in sub-Saharan Africa and Latin America (FAO and ITPS, 2015). The presence of certain pollutants may also produce nutrient imbalances and soil acidification, two major issues in many parts of the world, as identified in the *Status of the World's Soil Resources Report* (FAO and ITPS, 2015).

The unique global estimate of soil pollution was done in the 1990s by the International Soil Reference and Information Centre (ISRIC) and the United Nations Environment Programme (UNEP), which estimated that 22 million hectares had been affected by soil pollution (Oldeman, 1991). Latest data, however, indicate that this number may underestimate the nature and extent of the problem. National attempts to estimate the extent of soil pollution have been undertaken mainly in developed countries. According to the Chinese Environmental Protection Ministry, 16 percent of all Chinese soils and 19 percent of its agricultural soils are categorized as polluted (CCICED, 2015). There are also approximately 3 million potentially polluted sites in the European Economic Area and cooperating countries in the West Balkans (EEA-39) (EEA, 2014) and more than 1 300 polluted or contaminated sites in the United States of America (USA) are included on the Superfund National Priorities List (US EPA, 2013). The total number of contaminated sites is estimated at 80 000 across Australia (DECA, 2010). While these numbers are informative in helping us understand the effects of certain activities on soils, they do not reflect the complete extent of soil pollution around the world, and they highlight the inadequacy of available information and the differences in registering polluted sites across geographic regions (Panagiotakis and Dermatas, 2015). In low- and middle-income countries, the lack of data and information makes one of the world's biggest global problems invisible to the international community. With this overview, it is evident that there is an urgent need to implement a global assessment of soil pollution.

Fortunately, awareness on the importance of soil pollution is increasing around the world, leading to an increase in research conducted on the assessment and remediation of soil pollution (Figure 1). The *Revised World Soil Charter* (FAO, 2015b) recommends that national governments implement regulations on soil pollution and limit the accumulation of contaminants beyond established levels in order to guarantee human health and well-being. Governments are also urged to facilitate remediation of contaminated soils that exceed levels established to protect the health of humans and the environment. Soil pollution took centre stage at the Fifth Global Soil Partnership (GSP) Plenary Assembly (GSP, 2017). Recently, the United Nations Environmental Assembly (UNEA-3) adopted a resolution calling for accelerated actions and collaboration to address and manage soil pollution in the framework of Sustainable Development. This consensus, achieved by more than 170 countries, is a clear sign of the global relevance of pollution and of the willingness of these countries to develop concrete solutions to address pollution problems (UNEP, 2018). At the national level, many countries around the world have adopted or are currently adopting national regulations to protect their soils, to prevent pollution and to address historic problems of contamination. During the Estonian presidency of the Council of the European Union in the second half of 2017, soil became one of the main topics within European discussions, focusing on the key role soils play in food production. In China, soil pollution concerns have grown over the last few years, partly because the problem is directly related to human health. Other developing countries have also recently adopted regulations to prevent and control soil pollution, and to determine soil quality (Conselho Nacional do Meio Ambiente, 2009; MINAM, 2017; MMA, 2013).

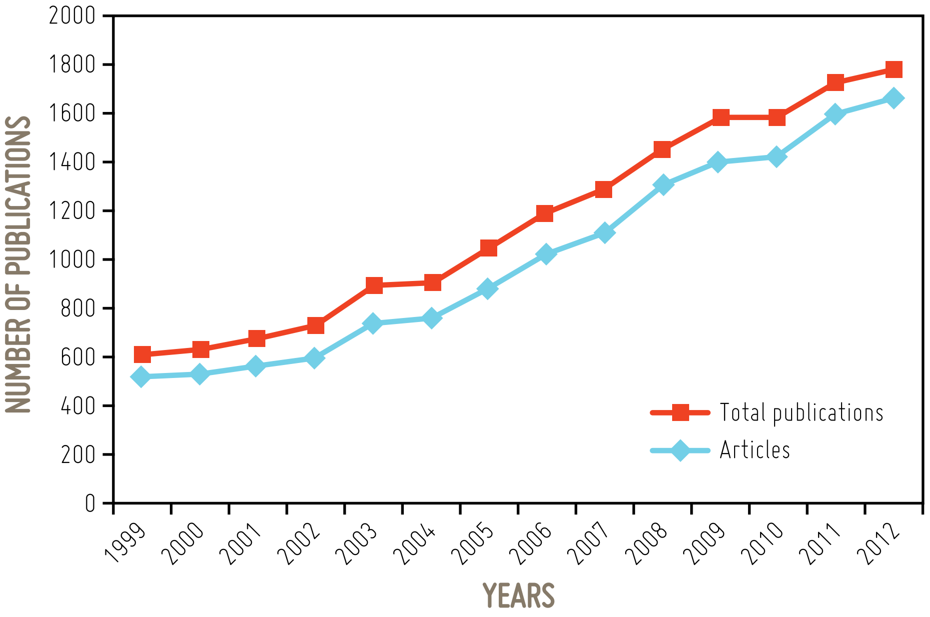

Figure 1. Number of scientific publications on soil pollution in the period of 1999–2012. Source: Guo *et al.*, 2014

The term "soil contamination" has frequently been used as a synonym for soil pollution. The Intergovernmental Technical Panel on Soils (ITPS) under the Global Soil Partnership (GSP) has formalized definitions of the two terms (FAO and ITPS, 2015). Soil contamination occurs when the concentration of a chemical or substance is higher than would occur naturally but is not necessarily causing harm. Soil pollution, on the other hand, refers to the presence of a chemical or substance out of place and/or present at a higher than normal concentration that has adverse effects on any non-targeted organism.

One issue is the difficulty in establishing a definition of "normal concentrations." It can be easier to establish hazardous concentrations for human-made substances that do not naturally occur in soils, but it can be challenging to do the same for heavy metals and metalloids, which can originate from the weathering of rocks and minerals. In that case, the parent material, climate and weathering rate need to be taken into consideration before establishing thresholds. Additionally, land use and management practices can affect background levels of substances occurring in soils. When referring to recommended levels, there are also many differences from country to country and among regions, not only about the value itself, but also about the name used to define it, including screening values, threshold values, acceptable concentrations, target values, intervention values, clean-up values, and many others (Beyer, 1990; Carlon *et al.*, 2007; Jennings, 2013). For that reason, to carry out a global study on the actual state of soil pollution and to be able to make comparisons is extremely complex. However, this is one of the main challenges when making a regional or global assessment of soil pollution.

Agreement among scientists regarding concepts and definitions would help policy-makers and stakeholders to identify other strategies and techniques used in different parts of the world to assess and to address soil pollution. Using a common and a simplified language would also lead to better understanding of the issue of soil pollution.

1.2 | POINT-SOURCE AND DIFFUSE SOIL POLLUTION

Soil pollution, as has been said, can result from both intended and unintended activities. These activities can include the direct deposition of contaminants into the soil as well as complex environmental processes that can lead to indirect soil contamination through water or atmospheric deposition (Tarazona, 2014). In the following sections, the different types of soil pollution are described.

1.2.1 | POINT-SOURCE POLLUTION

Soil pollution can be caused by a specific event or a series of events within a particular area in which contaminants are released to the soil, and the source and identity of the pollution is easily identified. This type of pollution is known as point-source pollution. Anthropogenic activities represent the main sources of point-source pollution. Examples include former factory sites, inadequate waste and wastewater disposal, uncontrolled landfills, excessive application of agrochemicals, spills of many types, and many others. Activities such as mining and smelting that are carried out using poor environmental standards are also sources of contamination with heavy metals in many regions of the world (Lu *et al.,* 2015; Mackay *et al.,* 2013; Podolský *et al.,* 2015; Strzebońska, Jarosz-Krzemińska and Adamiec, 2017). Other examples of point-source pollution are aromatic hydrocarbons and toxic metals, which are related to oil products. The sites range from leakage from tank installations in Greenland, which caused aromatic hydrocarbon and toxic metal levels that exceeded the Danish environmental quality criteria (Fritt-Rasmussen *et al.,* 2012), to accidental leakage from oil refinery storage tanks in Tehran (Bayat *et al.,* 2016).

Point-source pollution is very common in urban areas. Soils near roads have high levels of heavy metals, polycyclic aromatic hydrocarbons, and other pollutants (Kim *et al.,* 2017; Kumar and Kothiyal, 2016; Venuti, Alfonsi and Cavallo, 2016; Zhang *et al.,* 2015b). Old or illegal landfills, where waste is not disposed of properly or according to its toxicity (e.g. batteries or radioactive waste), as well as disposal of sewage sludge and wastewater, can also be important point-source pollutants (Baderna *et al.,* 2011; Bauman-Kaszubska and Sikorski, 2009; Swati *et al.,* 2014). Finally, point-source pollution caused by industrial activities can pose risks to human health. For example, over 5 000 brownfields in China are currently affecting the health of their inhabitants (Yang *et al.,* 2014). Urban brownfields, located in urban centres, are sites that once harboured industrial activities that have since been relocated.

1.2.2 | DIFFUSE POLLUTION

Diffuse pollution is pollution that is spread over very wide areas, accumulates in soil, and does not have a single or easily identified source. Diffuse pollution occurs where emission, transformation and dilution of contaminants in other media have occurred prior to their transfer to soil (FAO and ITPS, 2015). Diffuse pollution involves the transport of pollutants via air-soil-water systems. Complex analyses involving these three compartments is therefore needed in order adequately to assess this type of pollution (Geissen *et al.,* 2015). For that reason, diffuse pollution is difficult to analyze, and it can be challenging to track and to delimit its spatial extent. Many of the contaminants that cause local pollution may be involved in diffuse pollution, since their fate in the environment is not well understood (Grathwohl and Halm, 2003). Examples of diffuse pollution are numerous and can include sources from nuclear power and weapons activities; uncontrolled waste disposal and contaminated effluents released in and near catchments; land application of sewage sludge; the agricultural use of pesticides and fertilizers which also add heavy metals, persistent organic pollutants, excess nutrients and agrochemicals that are transported downstream by surface runoff; flood events; atmospheric transport and deposition; and/or soil erosion (Figure 2). Diffuse pollution has a significant impact on the environment and human health, although its severity and extent are generally unknown.

It has been widely demonstrated that the upper layers of soil are enriched in many metals and other elements that are linked to atmospheric deposition from natural and anthropogenic sources (Blaser *et al.*, 2000; Steinnes *et al.*, 1997; Steinnes, Berg and Uggerud, 2011). Almost every soil of the northern hemisphere contains radionuclides in higher concentrations than the background level, even in remote areas of North America and Eastern Asia. Due to the nuclear fallout after the catastrophic Chernobyl accident, radionuclides will be present in soils for centuries (Fesenko *et al.*, 2007). More than 50 years will be needed to reach a reduction of 50 percent of the radionuclides, such as $^{239/240}$Pu or ^{241}Am, in areas up to 200 km away from Chernobyl.

Due to these different types of pollution from diverse sources, an increase in scientific and technical efforts is needed to develop new methods for measuring, monitoring and better understanding atmospheric deposition processes and the extent of diffuse pollution.

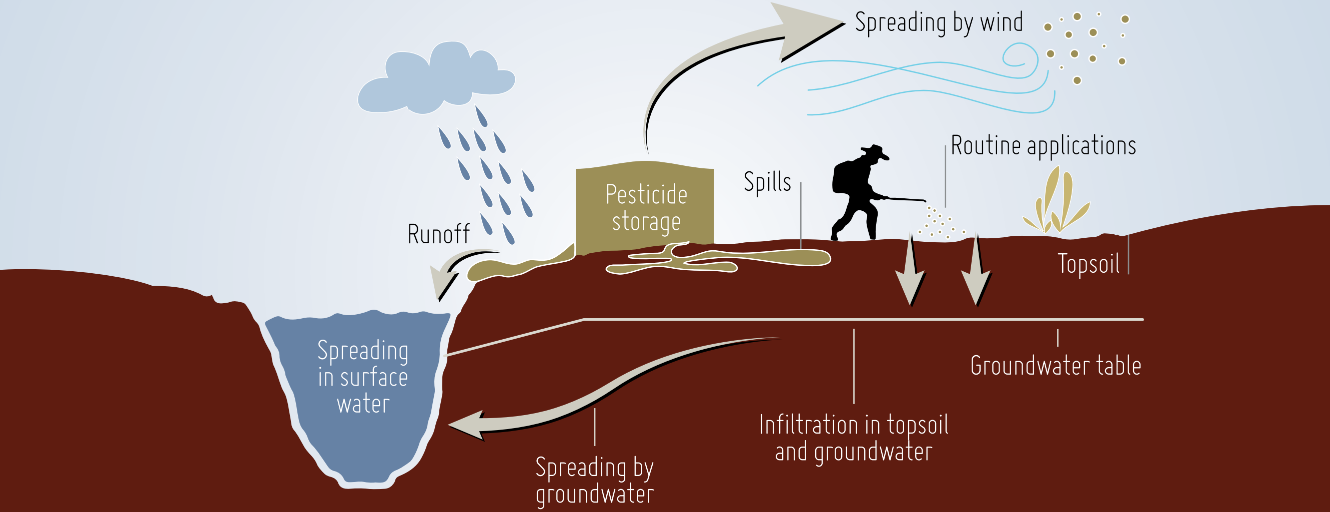

Figure 2. Transport pathway of pesticides in the environment. Source: FAO, 2000

1.3 | SOURCES OF SOIL POLLUTANTS

1.3.1 | NATURAL, GEOGENIC SOURCES

It is crucial to separate background values from baseline values when defining the extent of contamination in areas where environmental legislation has not yet established intervention limits for all environmental matrices (Albanese *et al.*, 2007). Background values indicate geogenic natural content, while baseline values indicate the actual content of an element in the superficial environment at any given point (Reimann, Filzmoser and Garrett, 2005; Salminen and Gregorauskiene, 2000).

Background concentrations in the soils of a region will be strongly related to the pedo-geochemical fraction and the dynamics of the environment that led to the formation of the soil. The use of averages or global intervals is therefore not suitable for determining background levels at the regional or local levels (Horckmans *et al.*, 2005; Paye, Mello and Melo, 2012). For example, heavy metals in soils can vary over two to three orders of magnitude, considering the natural variation in the concentration of trace metals within the parent rock type (Shacklette and Boerngen, 1984).

Several soil parent materials are natural sources of certain heavy metals and other elements, such as radionuclides, and these can pose a risk to the environment and human health at elevated concentrations. Arsenic (As) contamination is one of the major environmental problems around the world. Natural sources of As include volcanic releases (Albanese *et al.*, 2007) and weathering of As-containing minerals and ores (Díez *et al.*, 2009), but also naturally occurring mineralized zones of arsenopyrite (gossans), formed by the weathering of sulphide-bearing rock (Scott, Ashley and Lawie, 2001). Many of these minerals present a high spatial variability and many of them can be found in higher concentrations in deeper layers (Li *et al.*, 2017). However, As is slightly bioaccessible when coming from natural sources (Juhasz *et al.*, 2007).

Soils and rocks are also natural sources of the radioactive gas Radon (Rn). Radon diffusion from deeper layers to the surface is controlled, in part, by soil structure and its porosity (Hafez and Awad, 2016). High natural radioactivity is common in acidic igneous rocks, mainly in feldspar-rich rocks and illite-rich rocks (Blume *et al.*, 2016). Gregorič *et al.* found higher emissions of radon from soils containing carbonates than from any other soil or rock types (Gregorič *et al.*, 2013). Reference data for other natural radionuclides in rocks and soils are shown in Table 1.

Table 1. Specific activities of natural radionuclides in rocks and soils (given in Bq kg−1). Source: Blume *et al.*, 2016

Rock/soil	^{40}K	^{226}Ra	^{232}Th
Sandstones	461	35	4
Claystones	876	n.d.	41
Schist (Franconia)	1000	3000	60
Carbonates	97	<10	5
Acidic igneous rocks	997	37	52
Basic igneous rocks	187	10	8
Soils developed from loess	n.d.	41	54

Soils developed from granite	~1100	65–75	38–72
Soils developed from quartzite	~300	54–56	63–70
Soils developed from phyllite	n.d.	40–70	50–80

Natural events such as volcanic eruptions or forest fires can also cause natural pollution when many toxic elements are released into the environment. These toxic elements include dioxin-like compounds (Deardorff, Karch and Holm, 2008) and polycyclic aromatic hydrocarbons (PAHs). High level of heavy metals have been identified in volcanic soils in Réunion that can be associated with the active volcanic activity, mainly mercury (Hg), or with the weathering of the parent material, where high levels of chromium (Cr), copper (Cu), niquel (Ni) and zinc (Zn) have a natural pedo-geochemical origin (Dœlsch, Saint Macary and Van de Kerchove, 2006). High levels of Cr and Ni have also been reported in volcanic Indonesian soils, associated with pedo-geochemical origins (Anda, 2012). However, this natural pollution does not normally cause environmental problems due to the regenerative ability and the adaptation capacity of plants (Kim, Choi and Chang, 2011). The problems arise when the ecosystems are subject to external pressures, which alter their resilience and response ability.

Polycyclic aromatic hydrocarbons can also occur naturally in soils. They are usually of cosmogenic origin, being relatively common in cosmic dust samples and meteorites (Basile, Middleditch and Oró, 1984; Li, 2009), or derive from the diagenetic alteration processes of waxes contained in soil organic matter (Trendel *et al.*, 1989). Biogenic production of PAHs is favoured under reducing conditions (Thiele and Brümmer, 2002).

Naturally occurring asbestos (NOA) are fibrous minerals that occur naturally in soils formed from ultramafic rock, especially serpentine and amphibole. The main risk associated with NOA is inhalation exposure of humans related to extraction activities, while its natural presence in soils poses a negligible risk to the environment. However, NOA can be easily dispersed by wind erosion, and their mobilization will depend on the characteristics of the asbestos-containing materials, soil properties, humidity, and local weather conditions (Swartjes and Tromp, 2008). The environmental issues caused by NOA arise when they are released from soils close to urban areas, because asbestos is a carcinogenic substance, posing a high risk to human health from inhalation (Lee *et al.*, 2008).

1.3.2 | ANTHROPOGENIC SOURCES

Centuries of anthropogenic activities have resulted in a widespread problem of soil pollution around the world (Bundschuh *et al.*, 2012; DEA, 2010; EEA, 2014; FAO and ITPS, 2015; Luo *et al.*, 2009; SSR, 2010).

The main anthropogenic sources of soil pollution are the chemicals used in or produced as by-products of industrial activities, domestic and municipal wastes, including wastewater, agrochemicals, and petrol-derived products (Figure 3). These chemicals are released to the environment accidentally, for example from oil spills or leaching from landfills, or intentionally, as is the case with the use of fertilizers and pesticides, irrigation with untreated wastewater, or land application of sewage sludge.

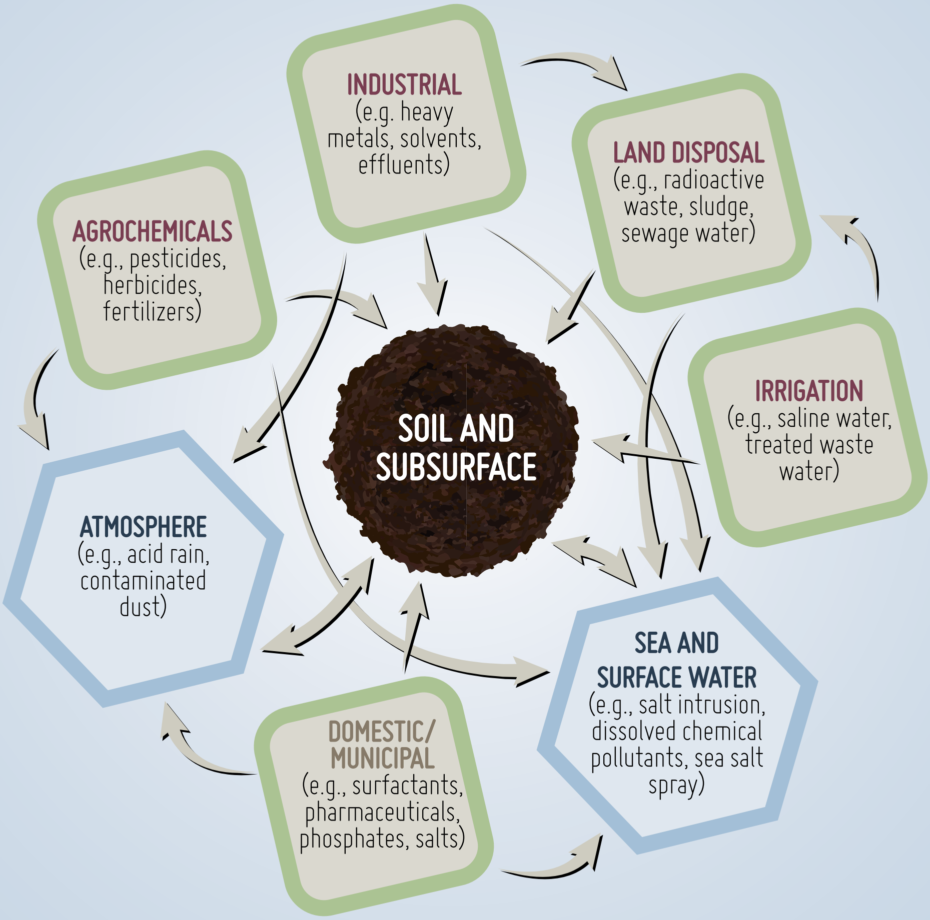

Figure 3. Potential interrelated pathways for soil-subsurface chemical contamination. Source: Yaron, Dror and Berkowitz, 2012

1.3.2.1 | INDUSTRIAL ACTIVITIES

The range of chemicals used in industrial activities is vast, as is their impact on the environment.

Industrial activities release pollutants to the atmosphere, water and soil. Gaseous pollutants and radionuclides are released to the atmosphere and can enter the soil directly through acid rain or atmospheric deposition; former industrial land can be polluted by incorrect chemical storage or direct discharge of waste into the soil; water and other fluids used for cooling in thermal power plants and many other industrial processes can be discharged back to rivers, lakes and oceans, causing thermal pollution and dragging heavy metals and chlorine that affect aquatic life and other water bodies. Heavy metals from anthropogenic activities are also frequent in industrial sites and can arise from dusts and spillages of raw materials, wastes, final product, fuel ash, and fires (Alloway, 2013).

According to the European Directive concerning integrated pollution prevention and control (IPPC) (EC, 1996), potentially polluting activities can be grouped into six main categories: 1) energy industries; 2) production and processing of metals; 3) mineral industry; 4) chemical industry and chemical installations; 5) waste management; and 6) other activities (which include paper and board production, manufacture of fibres or textiles, tanning of hides and skins, slaughterhouses, intensive poultry or pig rearing, installations using organic solvents, and the production of carbon or graphite) (García-Pérez *et al.*, 2007).

Salinization, another major threat to global soils, affects many soils which are close to certain industrial activities, mainly those associated with chlor-alkali, textiles, glass, rubber production, animal hide processing and leather tanning, metal processing, pharmaceuticals, oil and gas drilling, pigment manufacture, ceramic manufacture, and soap and detergent production (Saha *et al.*, 2017).

1.3.2.2 | MINING

Mining has had a major impact on soil, water and biota since ancient times (FAO and ITPS, 2015). Many documented examples can be found of heavily contaminated soils associated with mining activities around the world (Alloway, 2013).

Metal smelting to separate minerals has introduced many pollutants into the soil. Mining and smelting facilities release huge quantities of heavy metals and other toxic elements to the environment; these persist for long periods, long after the end of these activities (Ogundele *et al.*, 2017).

Toxic mining wastes are stocked up in tailings, mainly formed by fine particles that can have different concentrations of heavy metals. These polluted particles can be dispersed by wind and water erosion, sometimes reaching agricultural soils. For example, Mileusnić *et al.* found high levels of lead and copper in agricultural fields located near a tailings dam in Namibia (Mileusnić *et al.*, 2014). Toxic concentrations of chromium and nickel were also found in agricultural soils near an abandoned chromite-asbestos mine waste in India and in crops grown in those soils, resulting in a high risk to human and livestock health (Kumar and Maiti, 2015).

The use of phosphate rocks, which are naturally rich in radioactivity, in the production of fertilizers generates a by-product called phosphogypsum, which maintains nearly 80 percent of its original radioactivity due to ^{238}U decay products such as radon, ^{226}Ra, and polonium, ^{210}Po. These industries generate a radioactive source of pollution, which constitutes a threat to the surrounding ecosystems and organisms (Bolívar, García-Tenorio and García-León, 1995).

Significant point-source soil pollution occurs from oil and gas extraction due to spills of crude oil and brines. Brines have high salinity levels and can also contain toxic trace elements and naturally occurring radioactive materials. Brine spills are widespread: for example, Lauer *et al.* state that there have been approximately 3 900 brine spills associated with unconventional oil and gas production (including fracking) from the Bakken region of North Dakota since 2007 (Lauer, Harkness and Vengosh, 2016). Spills of crude oil from well sites and from pipelines are also a major source of soil pollution in oil producing areas.

1.3.2.3 | URBAN AND TRANSPORT INFRASTRUCTURES

The widespread development of infrastructure such as housing, roads and railways has considerably contributed to environmental degradation. Their more evident negative effects on soil are soil sealing and land consumption. Apart from these known soil threats, another major impact of infrastructure activities is the entry into the soil system of different pollutants. Despite its being a major threat, soil pollution from infrastructure activities has received very minor consideration in terms of planning and impact assessment.

Activities linked to transportation in and around urban centers constitute one of the main sources of soil pollution, not only because of the emissions from internal combustion engines that reach soils at more than a 100 m distance by atmospheric deposition and petrol spills, but also from the activities and the changes that result from them as a whole (Mirsal, 2008). Splashes generated by traffic during rainfall events and runoff, which may be significant if the drainage system is not well maintained, may translocate particles rich in heavy metals from the corrosion of metal vehicle parts, tires and pavement abrasion (Venuti, Alfonsi and Cavallo, 2016; Zhang *et al.*, 2015b) and other pollutants such as polycyclic aromatic hydrocarbons, rubber and plastic-derived compounds (Kumar and Kothiyal, 2016; Wawer *et al.*, 2015). Soil pollution associated with roads and highways is especially important in urban and peri-urban soils, and can be a major threat when food production occurs in adjacent areas. Foliar deposition and root uptake and transfer to above-ground tissues of bioavailable heavy metals are the main processes observed in roadside soil (Hashim *et al.*, 2017; Kim *et al.*, 2017; Zhang *et al.*, 2015b). Grazing in roadside soils is also quite common, and the ingestion of contaminated soil and plants constitutes potential dietary transfer of pollutants affecting animal and human health (Cruz *et al.*, 2014).

A major legacy source of soil pollution associated with transport is lead contamination of soils from leaded gasoline. Mielke and Reagan cite research that over 10 million tonnes of lead was transferred to the global environment via the motor vehicle fleet, about 5.9 million tonnes in the United States of America alone (Mielke and Reagan, 1998). The soil contamination that resulted from this was concentrated around roads and is especially high in core urban areas.

Municipal waste disposal by landfills, illegal or not, and untreated wastewater release into the environment are important sources of heavy metals, poorly biodegradable organic compounds and other pollutants which enter the soil. In most developed countries, strict regulations control the disposal and recycling of waste, solids and liquids (EC, 1986; US Federal Register, 1993), but there are countries where residue treatment and disposal are still posing a risk to the environment and to human health.

Many household chemicals, particularly those used in bulk quantities such as detergents and personal care products (PPCPs), also end up as sanitary sewage. Biosolids generated from municipal wastewater treatment can be a major sink for many PPCPs, and their land application can potentially introduce these contaminants into terrestrial and aquatic environments. The historical and continuing use of DDT for control of vector-borne diseases such as malaria has led to pollution of soils in urban and peri-urban areas (Mansouri *et al.*, 2017).

Lead-based paint is a major legacy source of lead (Pb) contamination in urban areas. Soils become contaminated when lead-based paint is pulverized into dust or small particles during renovations or demolition and then enters the environment (Mielke and Reagan, 1998). In the United States of America, approximately equal tonnages of lead were used in leaded gasoline between 1929 and 1989 as were used in white-lead paint pigments between 1884 and 1989, with peak use of lead-based paint in the 1920-29 period (Mielke and Reagan, 1998).

Plastics are also a major source of pollution. They are widely used in food packaging, shopping bags, and household items such as toothbrushes and pens, facial cleansers, and many other common items. Plastics have a strong presence in the environment globally. They are, in general, extremely persistent in the environment and they widely accumulate in oceans and landfills, but also in soils where producing factories are located. Polymers are usually considered to be biochemically inert and do not pose a threat to the environment. Unreacted residual monomers or small oligomers can, however, be found in the plastic material, since polymerization reactions are seldom complete (Araújo *et al.*, 2002). The most hazardous monomers, classified as either carcinogenic or both carcinogenic and mutagenic, are those belonging to families of polyurethanes, polyacrylonitriles, polyvinyl chloride, epoxy resins and styrenic copolymers (Lithner, Larsson and Dave, 2011). In addition, several thousand different additives such as brominated flame retardants, phthalates and lead compounds are used in the production of plastic. Many of these additives are considered harmful, with demonstrated disruptions to endocrine function, and carcinogenic and mutagenic effects on living organisms (Darnerud, 2003; Heudorf, Mersch-Sundermann and Angerer, 2007; Lithner, Larsson and Dave, 2011). All plastic, from the macro- to the nano-scale, are at risk of being leached and of adsorbing hazardous substances such as persistent organic pollutants and polycyclic aromatic hydrocarbons (Björnsdotter, 2015). They also accumulate heavy metals in high proportions (Mato *et al.*, 2001). The size and surface area are important factors influencing the leaching and adsorption behaviour: the smaller the particle, the larger the surface-volume ratio. The capacity to release or bind compounds is therefore also higher for smaller particles than for larger ones.

Plastics can reach the soil and aquatic systems via wastewater-treatment plants, but they can also be transported and suspended by wind from landfills and become airborne and widely dispersed. In agricultural fields in which plastic mulching is practised, an abundant source of plastic material is available in the soil. The presence and effects of plastic in aquatic organisms and ecosystems are well documented (Browne *et al.*, 2008; Thompson, 2004); however, the risks to human health and terrestrial ecosystems from the use of plastic polymers and products still needs to be assessed (Lithner, Larsson and Dave, 2011; Rillig, 2012; Rocha-Santos and Duarte, 2015). Almost no studies on plastics' fate in soil have been conducted.

1.3.2.4 | WASTE AND SEWAGE GENERATION AND DISPOSAL

As the global population increases, so does the generation of waste. In developing and least developed countries, high rates of population growth and increasing waste and sludge production, combined with lack of municipal services that deal with waste management, create a dangerous situation. According to a World Bank report (Hoornweg and Bhada-Tata, 2012), the global production of municipal solid waste was estimated to be 1.3 billion tonnes per year in 2012, varying from 0.45 kg per person and per day in sub-Saharan Africa to 2.2 kg per capita annually in the Organisation for Economic Co-operation and Development (OECD) countries. Future predictions are worrying, however, as waste production is expected to rise to 2.2 billion tonnes by 2025.

Municipal waste disposal in landfills and incineration are the two most common ways to manage waste. In both cases, many pollutants, such as heavy metals, polyaromatic hydrocarbons, pharmaceutical compounds, personal care products and their derivative products accumulate in the soil (Swati *et al.*, 2014), either directly from landfill leachates that may be polluting soil and under groundwater, or by ash fallout from incinerating plants (Mirsal, 2008). Baderna *et al.* discovered a complex mixture of pollutants in a landfill leachate that alters groundwater quality and in turn affects the food chain (Baderna *et al.*, 2011).

Establishments that recycle lead batteries have been identified as major sources of soil contamination around the world. This is especially the case in Africa, where the lead battery industry has notably expanded in the last few years and will continue to grow, but where regulations are weak or absent (Gottesfeld *et al.*, 2018). The proximity of lead battery industries and recycling plants to communities poses a high risk to human health, as was demonstrated by blood samples where lead levels exceeded screening level criteria (US Agency for Toxic Subtances and Disease Registry, 2011; Zahran *et al.*, 2013).

The twenty-first century has resulted in improvements in communication and important technological developments. The production of electrical and electronic equipment is growing rapidly in the world and will continue to grow, with developing countries becoming major producers within the next decade (Robinson, 2009). However, once devices become obsolete or are no longer functioning, they eventually become waste. Electronic waste, or e-waste, contains valuable elements, such as copper and gold, but also many other hazardous substances that make it impossible to treat it in a similar manner as regular urban waste. In Europe and North America, the majority of e-waste remains unrecycled (Barba-Gutiérrez, Adenso-Díaz and Hopp, 2008; Sthiannopkao and Wong, 2013), while e-waste has become a source of income in developing or emerging industrialized countries. Itai *et al.* reported high concentrations of heavy metals and of rare metalloids (In, Sb, Bi) in an e-waste recycling site in Ghana, indicating that these metalloids should be included in risk assessment approaches (Itai *et al.*, 2014). Formal recycling centres comprise only 25 percent of the industry, however, and e-waste is mostly recycled in informal sectors using primitive techniques such as burning cables for the harvesting of copper. These techniques release a multitude of hazardous substances (flame retardants, dioxin-like compounds, polycyclic aromatic hydrocarbons, heavy metals) without taking into account protective measures for the environment or for human health (Perkins *et al.*, 2014).

The use of sewage sludge to amend soils may be beneficial, as it adds organic matter and nutrients to soils. However, if that sewage sludge has not been pre-treated before its application, many pollutants such as heavy metals can accumulate in the soil and eventually enter the food chain. In Europe, the use of sewage sludge is regulated, but this is not the case everywhere.

The use of treated wastewater for agricultural irrigation is common in arid and semiarid regions as a solution to water scarcity (Jefatura del Estado, 2001; Keraita and Drechsel, 2004; Uzen, 2016). In Israel, for example, over 80 percent of municipal treated sewage is re-used (Katz, 2016), and 26 percent of Pakistan's vegetable production is irrigated using wastewater (Ensink *et al.*, 2004). The use of recycled wastewater in the arid regions of Spain has addressed the issue of water deficit, but is also a way to add nutrients, and has led to an increase in crop productivity (Dorta-Santos *et al.*, 2014). The use of wastewater can, however, be an issue in countries where water quality guidelines and legislation do not exist. Improper use of wastewater can lead to the deposition of heavy metals, salts, PPCPs and pathogens, if they are not completely removed after treatment or in cases where wastewater is left untreated (Dalkmann *et al.*, 2014; Flores-Magdaleno *et al.*, 2011; Pedrero *et al.*, 2010).

1.3.2.5 | MILITARY ACTIVITIES AND WARS

Until the twentieth century, most conflicts were of local magnitude and had relatively little impact on soils. However, modern warfare makes use of non-degradable weapons of destruction and of chemicals that can remain in the affected soils for centuries after the end of the conflict (FAO and ITPS, 2015). The nature of soils can be considerably modified by warfare activities in both wartime and times of peace due to military activities such as test-firing facilities. Total and sometimes even partial recovery of these soils can take many years, and in some cases even centuries (Certini, Scalenghe and Woods, 2013).

The First and Second World Wars left Europe with a significant heritage of pollution (land mines, remains of ammunitions and leftover chemicals, radioactive and biological toxic agents), not only in the battlefields but also in sites such as shooting areas, barracks and storage of armaments. This legacy has made the soils in some of these areas unsuitable for any kind of exploitation or service provision. There are approximately 110 million mines and other unexploded ordnance (UXO) scattered in 64 countries on all continents, remnants of wars from the early twentieth century up until today (Kobayashi, 2012).

The disposal of munitions, and the lack of care in their manufacturing caused by the urgency of the situation at the time of their production, can contaminate soils for extended periods of time. There is little published evidence on this type of contamination, largely because of restrictions placed by governments of many countries on the publication of material related to warfare. In Berlin, for example, more than a thousand hectares presented high levels of contamination (Schafer, 1995); Gruinard Island, in western Scotland, is still polluted with anthrax spores that were used as potential biological weapons, despite remediation efforts (Szasz, 1995; WHO, 2008). Mustard gas stored during the Second World War has also contaminated some sites for up to 50 years (Watson and Griffin, 1992).


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1.3.2.6 | AGRICULTURAL AND LIVESTOCK ACTIVITIES

The different agricultural sources of soil pollutants include agrochemical sources, such as fertilizers and animal manure, and pesticides (Figure 4). Trace metals from these agrochemicals, such as, Cu, Cd, Pb and Hg, are also considered soil pollutants as they can impair plant metabolism and decrease crop productivity. Water sources for irrigation can also cause soil pollution if they consist of waste water and urban sewage. Excess N and heavy metals are not only a source of soil pollution, but also a threat to food security, water quality and human health, when they enter the food chain (FAO and ITPS, 2015).

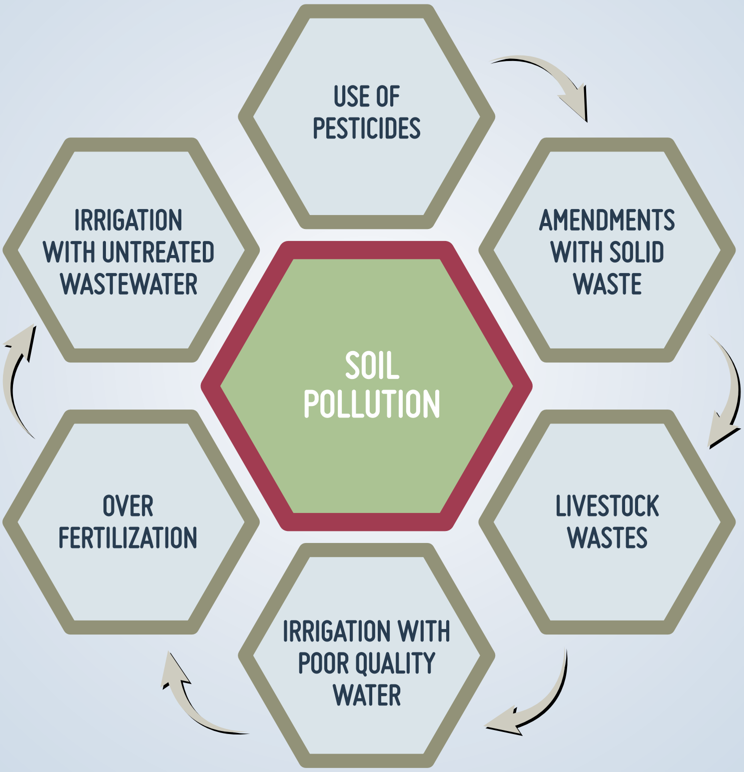


Figure 4. Agricultural sources of soil pollution

Point sources of pollution in agricultural settings include accidental spills of hydrocarbons in agricultural fields used as fuels for machines or of agrochemicals during their transportation and storage stages.

As mentioned earlier, livestock production can also be a source of pollution, especially if the waste is not properly managed and disposed of: the urine and faeces may contain parasites and medical substances that can persist and accumulate in the soil (Zhang *et al.*, 2015a). Many medical substances are lipolytic and not readily degradable and therefore have the potential to be retained in the sludge or the manure, which in turn may be used as fertilizer. Such retention can greatly affect microorganisms and other beneficial organisms in the soil (Halling-Sørensen *et al.*, 1998).

Excessive application of fertilizers and manure or inefficient use of the main nutrients (N and P) in fertilizers are the main contributors to environmental issues linked to agriculture (Kanter, 2018). These two nutrients are a source of diffuse pollution. Excess N can also be lost to the atmosphere through greenhouse gas emissions, and excess P contributes to the eutrophication of neighbouring sources of water. Excessive fertilizer usage can lead to soil salinity, heavy metal accumulation, water eutrophication and accumulation of nitrate, which can be a source of environmental pollution but also a threat to human health. The fertilizer industry is also considered to be a source of heavy metals such as Hg, Cd, As, Pb, Cu, Ni and Cu, and natural radionuclides like ^{238}U, ^{232}Th and ^{210}Po. Proper handling and management of fertilizer is crucial to avoid polluting the soil (Stewart *et al.*, 2005).

Compost and animal residues are an important source of nutrients. They contribute to achieving a circular economy, reducing the environmental impacts of waste and increasing organic matter and nitrogen content in soils while reducing external inputs in agroecosystems (Shiralipour, McConnell and Smith, 1992). Xia *et al.* reported an overall increase in crop production of 4.4 percent when manure partially substituted synthetic fertilizers (Xia *et al.*, 2017). The substitution enhanced crop uptakes of N and other nutrients, and significantly reduced N losses by volatilization, erosion and leaching, mainly due to its property of slow nutrient release and to the promotion of microbial immobilization of bioavailable N. However, yield increase depends on the manure and crop types considered (Wang *et al.*, 2016; Xia *et al.*, 2017). Furthermore, compost and manure are great sources of organic matter (Zhao *et al.*, 2014b).

A recent report by the Production and Plant Protection Division of the Food and Agriculture Organization of the United Nations (FAO) documented that global manure production from all livestock has increased by 66 percent, from 73 to 124 million tonnes of N, from 1961 to 2016, with manure applied to soils increasing from 18 to 28 million tonnes of N, and N input from manure left on pasture increasing from 48 to 86 million tonnes of N (Raffa *et al.*, 2018).

Despite their potential benefit for agriculture, there is sufficient scientific evidence of the increase in heavy metals content, pathogens, and veterinary antibiotics residues, which may lead to antimicrobial-resistant bacteria proliferation in soils amended with livestock manure. Heavy metals in livestock manure mainly come from feed (Nicholson *et al.*, 1999), while antibiotics are utilized to prevent and treat diseases and as growth promoters (Kumar *et al.*, 2005). Nicholson *et al.* conducted an inventory across agricultural fields in England and Wales to determine the main sources of heavy metals (Nicholson *et al.*, 2003). After atmospheric deposition, which is the main source, livestock manures and sewage sludge were identified as important sources of contamination. These were especially important for Zn, Cu, Ni, Pb and Cr contamination (Nicholson *et al.*, 1999; Wang *et al.*, 2016).

Pesticides are substances, or mixtures of substances, intended for preventing, destroying or controlling any pest causing harm or interfering with the production, processing, storage, transport or marketing of food, agricultural commodities, wood and wood products (FAO, 2006). Pesticides are a subset of the overall range of pesticides. They have been intentionally released into the environment at a larger scale since the Second World War, when DDT's insecticidal properties were discovered. The use of pesticides has helped provide food for an increasing population similarly to the application of nutrients; however, the over use of pesticides can have negative effects on human health and the environment (Popp, Pető and Nagy, 2013; FAO and ITPS, 2017). Negative effects on soil organisms have been widely studied (Bünemann, Schwenke and Van Zwieten, 2006; Jacobsen and Hjelmsø, 2014; Komárek *et al.,* 2010; Nguyen *et al.,* 2016; Ockleford *et al.,* 2017; Puglisi, 2012), and health problems have been related to exposure to pesticides and other agrochemicals (Bhatia *et al.,* 2005). The major threat to human health is being exposed to low dose levels over a lifetime (WHO, 1993), as the direct short-term effects of such exposure are not obvious.

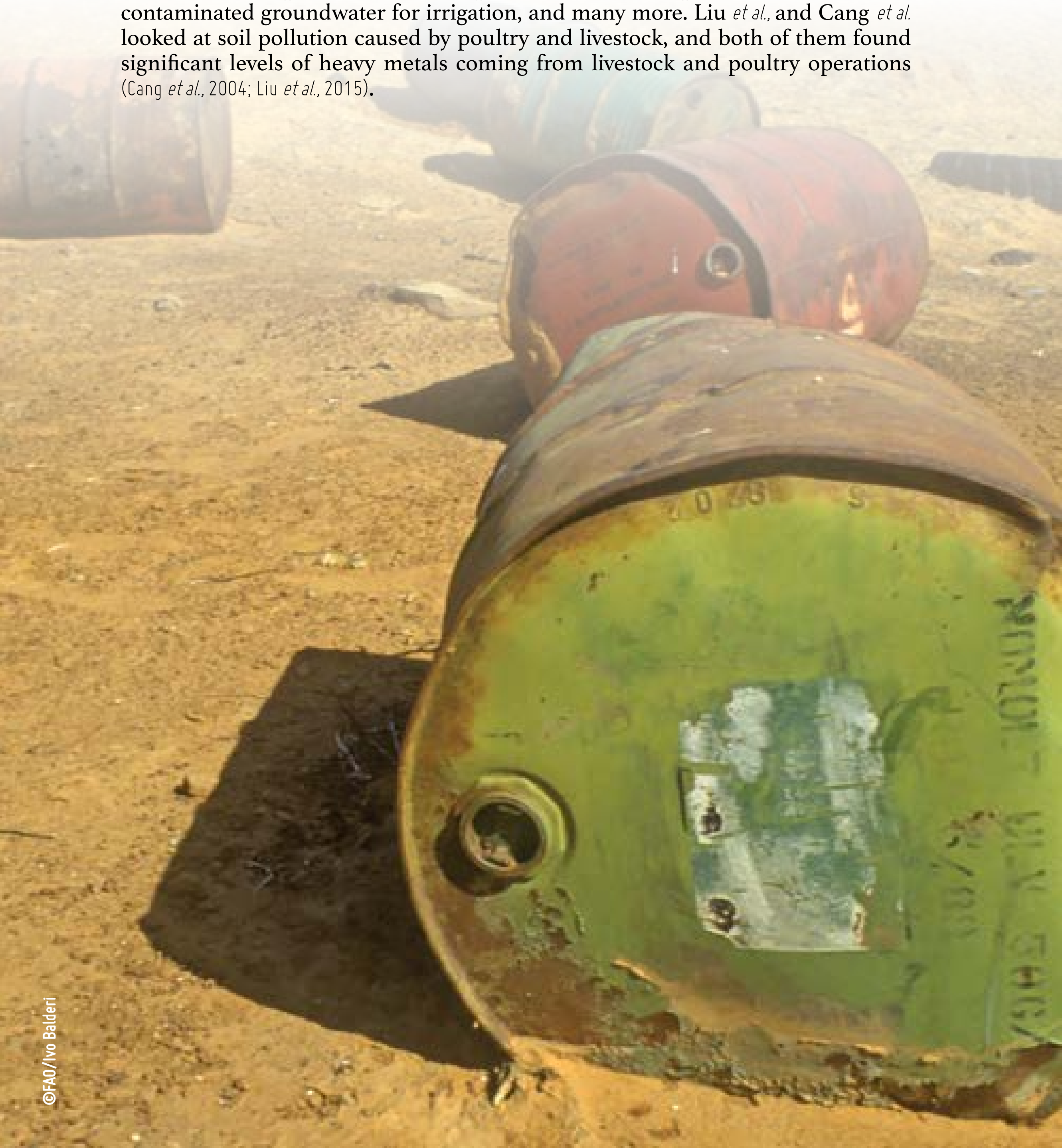
Considering the need to have coordinated efforts regarding hazardous chemicals, the Rotterdam Convention was adopted on 10 September 1998. It aims to promote shared responsibility and cooperative efforts among Parties in the international trade of hazardous chemicals and pesticides in order to protect human health and the environment from potential harm. The Convention contributes to the environmentally sound use of such chemicals, by facilitating information exchange about their characteristics, by providing for a national decision-making process on their import and export and by disseminating these decisions to Parties.

The benefit of the Convention is to prevent unwanted trade in the chemicals included in the legally binding prior informed consent (PIC) procedure. It enables member governments to alert each other to potential dangers by exchanging information on banned or severely restricted chemicals and to take informed decisions on them. It makes the international trade in hazardous chemicals more transparent and less vulnerable to abuse through its export notification provisions and by encouraging harmonized labelling of exported chemicals. The Convention also calls for the provision of technical assistance to help establish the infrastructure and capacity necessary to safely manage chemicals.

The International Code of Conduct on Pesticide Management (FAO, 2003) provides a voluntary framework and standards for managing pesticides throughout their life cycle. The Code is directed primarily at government authorities and the pesticide industry but is also relevant for other stakeholders. The Code is supported by technical guidelines and toolkits, for example the Registration Toolkit (http://www.fao.org/pesticide-registration-toolkit/en/) and the Environmental Management Toolkit for Obsolete Pesticides (EMTK).

Stocks of obsolete, unwanted and banned pesticides continue to represent a threat to human health, the environment and the sustainable development of the regions in which they are found. The reasons for the accumulation of obsolete pesticide stocks are well documented. They include poor storage and management of pesticide stocks, international bans on the use of harmful pesticides, untimely donations of pesticides, over-ordering and supply and procurement of strategic stocks of pesticides for migratory pest control, which are then not used completely or not needed. Pesticide stocks are often stored under very poor conditions, resulting in container deterioration and leakage into the surrounding environment and ultimately affecting soil and ground water quality. The volumes 1-4 of the FAO EMTK series are designed to assist countries with the risk management of obsolete pesticide stocks. Volume 6 is intended to provide practical methods to assist countries in the formulation of strategies for the management of pesticide contaminated land. As risk reduction largely depends on the accuracy of site investigation and associated risk assessment this document is meant to be used in close conjunction with EMTK Volume 5, the outputs of which are critical in the design of pesticide contaminated land environmental management plans.

Other sources of agricultural pollution include arsenic pollution due to concentrated animal feeding operations (CAFOs), plastic residues from plastic mulching, use of contaminated groundwater for irrigation, and many more. Liu *et al.*, and Cang *et al.* looked at soil pollution caused by poultry and livestock, and both of them found significant levels of heavy metals coming from livestock and poultry operations (Cang *et al.*, 2004; Liu *et al.*, 2015).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1.4 | MAIN POLLUTANTS IN SOIL

The release of pollutants to the environment, as has been mentioned, usually originates from anthropogenic processes. Even if some elements and compounds occur naturally in soils, human interventions are the main drivers of soil pollution. The following sections discuss only a small subset of the most common pollutants affecting agricultural areas, and the properties that make these compounds pollutants. Pollutants have been divided by their chemical characteristics, but some of the categories presented here overlap. Swartjes proposed a systematic categorization of pollutants (Figure 5) that may be useful in better understanding them (Swartjes, 2011).

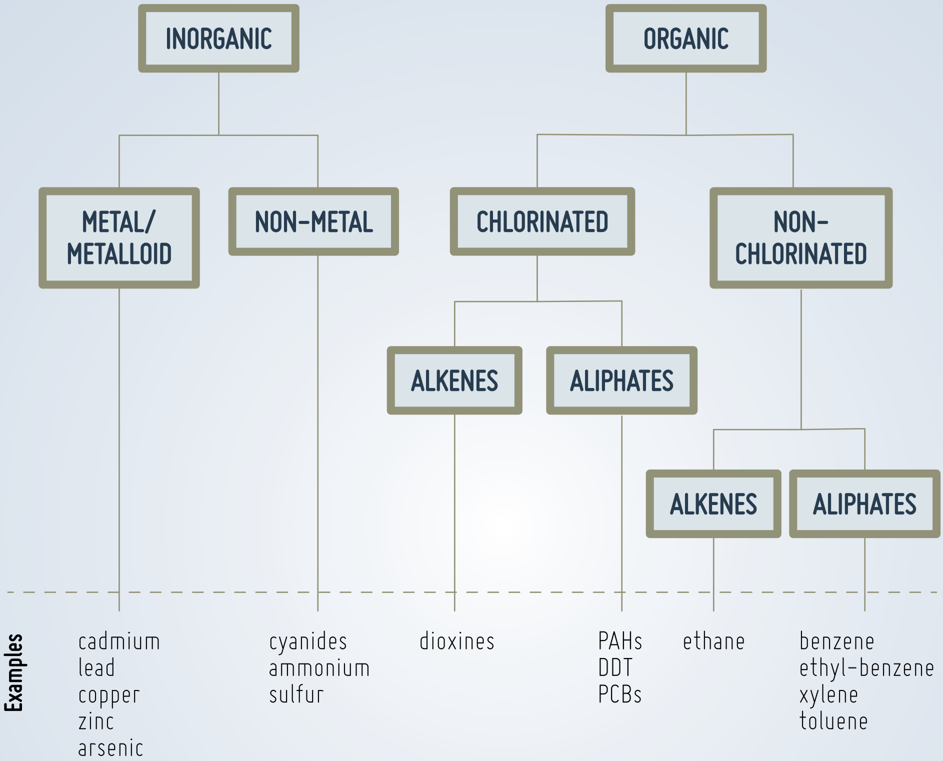

Figure 5. Systematic categorization of the main pollutants in soils. Source: Swartjes, 2011

1.4.1 | HEAVY METALS AND METALLOIDS

The term “heavy metals” refers to the group of metals and metalloids of relatively high atomic mass (>4.5 g/cm^3) such as Pb, Cd, Cu, Hg, Sn, and Zn, that can cause toxicity problems. Other non-metals that are often considered together with heavy metals include As, antimony (Sb) and selenium (Se) (Kemp, 1998). These elements naturally occur at low concentrations in soils. Many of them are essential micronutrients for plants, animals and humans, but at high concentrations may cause phytotoxicity and harm human health because of their non-biodegradable nature, which causes them to readily accumulate in tissues and living organisms.

The main anthropogenic sources of heavy metals are industrial areas, mine tailings, disposal of high metal wastes, leaded gasoline and paints, application of fertilizers, animal manures, sewage sludge, pesticides, wastewater irrigation, coal combustion residues, spillage of petrochemicals, and atmospheric deposition from varied sources (Alloway, 2013).

Heavy metals are the most persistent and complex kind of pollutants to remediate in nature. They not only degrade the quality of the atmosphere, water bodies, and food crops, but also threaten the health and well-being of animals and human beings. Metals accumulate in the tissues of living organisms because unlike most organic compounds they are not subject to metabolic breakdown. Among the heavy metals, Zn, Ni, Co, and Cu are relatively more toxic to plants, and As, Cd, Pb, Cr and Hg are relatively more toxic to higher animals (McBride, 1994).

The most important elements to consider in terms of food-chain contamination are As, Cd, Hg, Pb and Se (McLaughlin, Parker and Clarke, 1999). The main sources of As in soils are agrochemical compounds and mining and smelting activities, but they can also be introduced in manure originating from livestock feed with As-rich additives. Some parent materials are rich in As and therefore their weathering can also be a source of As in high concentrations.

Trace metals from inorganic (Cu-based) and organic pesticides represent a major environmental and toxicological concern (Komárek *et al.*, 2010). Cu is easily immobilized by soil organic matter (SOM) and Fe- and Mn-(hydro)oxides, remaining at high concentrations on upper layers of soils. However, fungicide-derived Cu has been found in large amounts in the potentially available fraction of the soil (Pietrzak and McPhail, 2004).

1.4.2 | NITROGEN AND PHOSPHORUS

Nitrogen (N) is an essential component of all living structures such as proteins, DNA, RNA, hormones, enzymes and vitamins. It occurs in both organic and inorganic forms, and in many different oxidation states. Its available forms differ depending on the specific organism. Unreactive forms such as gaseous nitrogen (N_2) can be assimilated through microbial activity. Plants need more chemically available forms, such as ammonium (NH_4^+) and nitrate (NO_3^-), while animals require complex forms, such as amino acids and nucleic acids (Yaron, Dror and Berkowitz, 2012).

Phosphorus (P) is one of the main macronutrients for all living organisms. It forms part of biological molecules, such as DNA and RNA, and it is used to transport cellular energy via adenosine triphosphate (ATP).

In order to feed the increasing population and to supply the nutrient needs of the many deficient soils around the world, additions of N, P and potassium (K) through synthetic fertilizers were widely adopted through the twentieth century (Tilman *et al.*, 2002). Fertilizer demand has increased globally (Figure 6), supported by global economic growth, leading to an excessive application of fertilizers following the idea "the more, the better." According to FAO, fertilizer global consumption by 2018 is forecast to reach 200 million tonnes, with over 50 percent of global fertilizer consumption concentrated in China, the United States of America, and India (FAO, 2015a). However, there is not a linear correlation between an increase in the amount of fertilizer applied to agricultural soils and an increase in crop production; on the contrary, increases may result in low nutrient use efficiency and in turn lower crop yields (Hossain *et al.*, 2005; Zhu *et al.*, 2005), and may cause acute environmental problems (Good and Beatty, 2011; Vitousek *et al.*, 2009; Withers *et al.*, 2014).

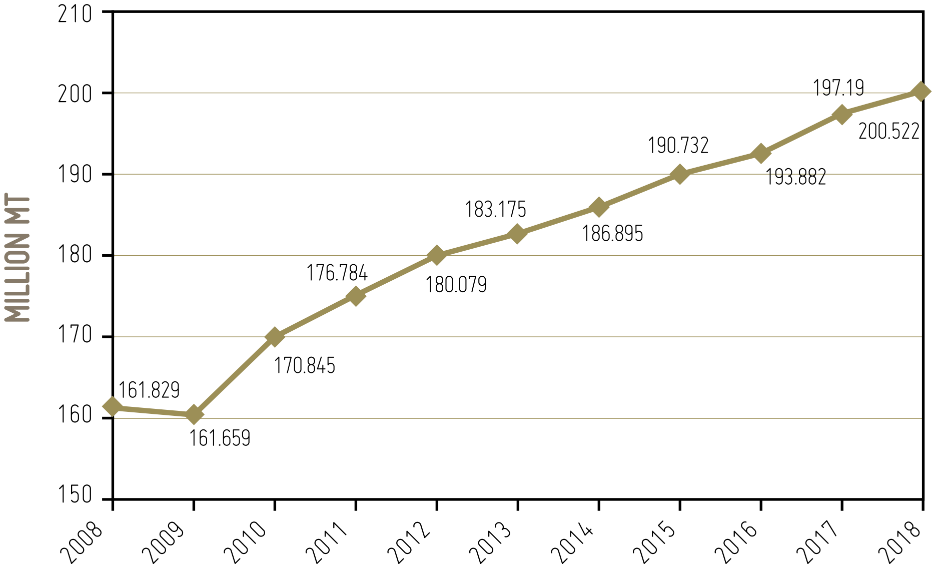

Figure 6. Global synthetic fertilizers consumption. Source: FAO, 2015a

Nitrogen and phosphorus become pollutants when they are applied in excess to agricultural soils in the form of fertilizers, or in areas of intensive livestock production (Carpenter *et al.*, 1998; Torrent, Barberis and Gil-Sotres, 2007). These nutrients are able to leach into the groundwater or be transported to surface water bodies by runoff, causing eutrophication or leading to high nitrate concentrations and related environmental and human health problems (EC, 1991; Frumin and Gildeeva, 2014; Pretty *et al.*, 2003; Yaron, Dror and Berkowitz, 2012). Many heavy metals have also been documented in phosphate and nitrate fertilizers including As, Cd, Cr, Hg, Pb, and Zn (Brevik, 2013).

While nutrients are essential to crop production, when they are applied in excess they can have negative effects on yields. Nitrogen increases chlorophyll production, and energy for flower growth and root elongation is redirected to foliage proliferation, causing disorders in plants and making them more vulnerable to pathogen attacks. It can also affect crop nutrient balance (Hao *et al.*, 2003). Nitrogen pollution influences soil organic matter decomposition, as it affects microbial community composition and activities (Bragazza *et al.*, 2006; Luo *et al.*, 2017; Shen *et al.*, 2010; Zhou and Zhang, 2014), as well as soil acidity and salinity (Han *et al.*, 2015).

1.4.3 | PESTICIDES

Pesticides are applied to reduce crop losses due to insect pests, weeds and pathogens, and thus to guarantee global food supplies (FAO and ITPS, 2017). Pesticides include but are not limited to insecticides, fungicides, herbicides, rodenticides, molluscicides, nematicides, and plant growth regulators.

Without pesticide use, crop losses have been estimated to vary from 32 percent for cereals to 78 percent in fruit production (Cai, 2008). Pesticides are applied not only on agricultural lands; they also have a great importance on human health protection, for example for sanitary pest control of vector-borne diseases. They are also used to keep infrastructures free of damaging insects and weeds, such as for preventing the attack of wood buildings by termites, or for keeping roadside and train tracks clean in order to help in avoiding accidents (Aktar, Sengupta and Chowdhury, 2009). Their use is not homogeneously distributed around the globe, mainly because of their cost and because pests vary by climatic and geographic region. According to FAOSTAT, FAO's Corporate Database for Substantive Statistical Data (FAOSTAT, 2016), some low and middle income countries have increased their consumption of pesticides in the last decade. Bangladesh, for example, has increased the use of pesticides by four times, while Rwanda and Ethiopia have increased theirs by over six times. This amount goes up to ten times in the Sudan (Figure 7). Once again, the problem arises when a misuse of pesticides occurs: when they are applied in higher amounts than needed and using practices that contribute to their spreading into the environment, such as spraying with not suitable/not maintained/not calibrated application equipment or by planes into vast regions, affecting inhabitants and non-target organisms (Carvalho, 2017).

Nonki Azariah on Unsplash

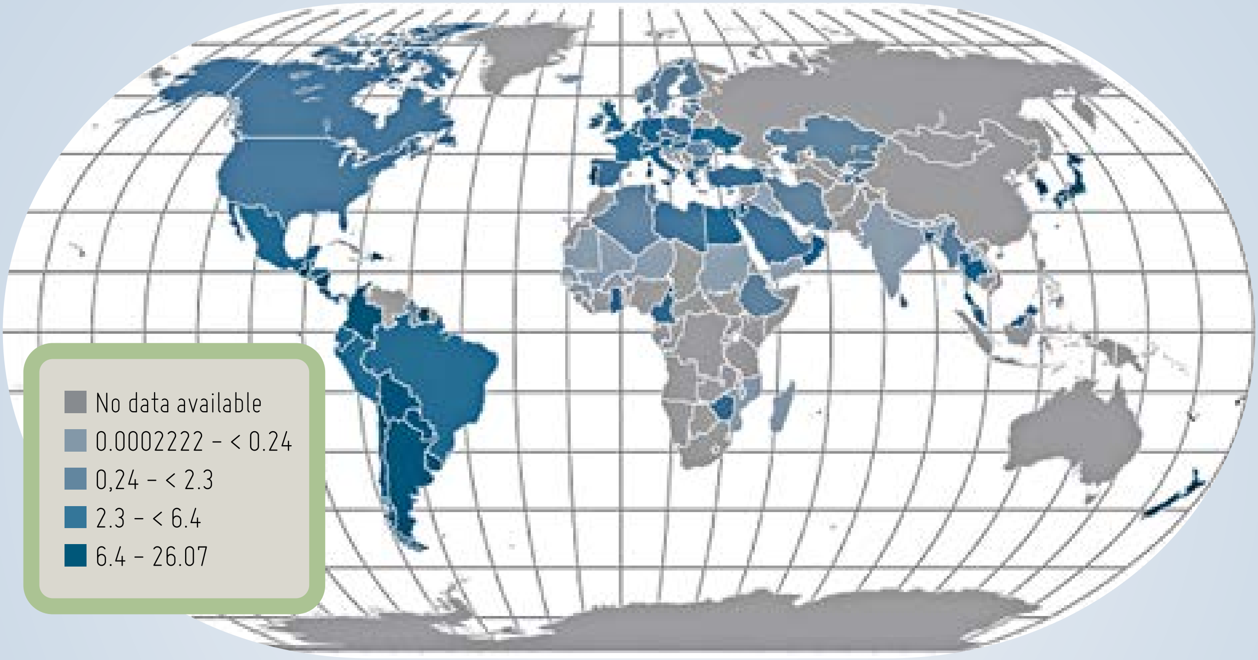

Figure 7. Use of pesticides per hectare of arable land, kg/ha, in the years 2007—2012. Source: FAO, 2015c

Pesticides can be either organic or inorganic synthetic molecules. They are classified on the basis of their chemical structures, their mode of action, their way of entry into the body, and their target organisms. Their toxicological effects on pests depend on their chemical composition, which in turn affects their interaction with soil components (Singh, 2012). According to their chemical structure, pesticides can be divided into twelve distinct groups, with the main pesticides in each group listed below:

- *organochlorine compounds*: DDT, Methoxychlor, Chlordane, Dicofol. BHC/HCH, Aldrin, Endosulfan, Heptachlor, Methoxychlor, Chlordane, Dicofol;

- *organophosphorus compounds*: Parathion, Malathion, Monocrotophos, Chlorpyrifos, Quinalphos, Phorate, Diazinon, Fenitrothion, Acephate, Dimethoate, Fenthion, Isofenfos, Phosphamidon, Temephos, Triazophos;

- *carbamates*: Aldicarb, Oxamyl, Carbaryl, Carbofuran, Carbosulfan, Methomyl, Methiocarb, Propoxur, Pirimicarb;

- *pyrethroids*: Allethrins, Deltametrin, Resmethrin, Cypermethrin, Permethrin, Fenvalerate, Pyrethrum;

- *neonicotinoids*: Acetamiprid, Imidacloprid, Nitenpyram, Thiamethoxam;

- *organotin compounds*: Triphenyltin acetate, Trivenyltin chloride, Tricyclohexyltin hydroxide, Azocyclotin;

- *organomercurial compounds*: Ethyl mercuric chloride, Phenyl mercuric bromide;

- *dithiocarbamate fungicides*: Zineb, Maneb, Mancozeb, Ziram;

- *benzimidizole compounds*: Benomyl, Carbendazim, Thiophanate methyl;

- *chlorphenoxy compounds*: 2,4-D, TCDD, DCPA, 2,4,5-T, 2,4-DB, MCPA, MCPP;

- *dipyridiliums*: Paraquat, Diquat; and

- *miscellaneous*: DNOC, Bromoxyl, Simazine, Triazamate.

Some of the pesticides listed above are also persistent organic pollutants (POPs) and are discussed further below.

Some pesticides are also associated with heavy metal contamination of soils. The recent report by the Intergovernmental Technical Panel on Soils (ITPS) on the impact of plant protection products on soil functions and ecosystem services highlighted the severe impact of copper-based fungicides on earthworms and microbial biomass. These fungicides are widely used in organic viticulture to control vine fungal diseases (FAO and ITPS, 2017).

Pesticide persistence, behaviour and mobility are also extremely varied as are the mechanisms involved in their degradation and retention in soils (Figure 8): sorption–desorption, volatilization, chemical and biological degradation, uptake by plants and leaching (Arias-Estévez *et al.*, 2008).

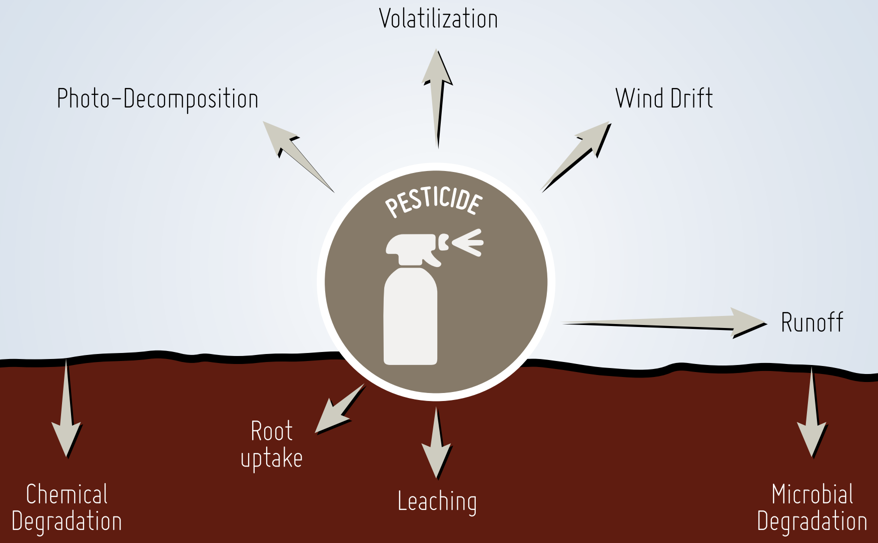

Figure 8. Behaviour of pesticides in the environment. Source: Singh, 2012

1.4.4 | POLYCYCLIC AROMATIC HYDROCARBONS

Polycyclic aromatic hydrocarbons (PAHs) are a group of persistent, semi-volatile organic pollutants.

Polycyclic aromatic hydrocarbons represent a broad group of physicochemically different molecules made of two or more unsubstituted benzene rings fused together when a pair of carbon atoms is shared between them. The most frequent PAHs are anthracene, fluoranthene, naphthalene, pyrene, phenantrene and benzopyrene (Lerda, 2011). The very low water solubility of PAHs and the slow mass-transfer rates from solid phase may limit their availability to microorganisms, thus hindering natural attenuation by microbial processes. Polycyclic aromatic hydrocarbons accumulate in soils because of their persistence and hydrophobicity and tend to be retained in the soil for long periods of time. For that reason, most PAHs are components of POPs and are widespread in air, water, soils, and sediments (Lin *et al.*, 2013). Low-molecular-weight PAHs, with two or three rings, are volatile and occur mainly in the atmosphere, whereas those of medium and high molecular weight are partitioned between the atmosphere and particles, depending on the temperature (Srogi, 2007).

Incomplete combustion of coal, gas, oil, and garbage; pyrolysis of organic materials by industries, agriculture and traffic; diagenetic alteration processes of natural organic matter (OM); long-term wastewater irrigation; reused sewage sludge; and fertilizer use in agricultural production all result in high concentrations of PAHs in agricultural soils (Conte *et al.*, 2001) (Figure 9). For example, in western German forests, brown coal strip-mining sites have been identified as the main sources of low weight PAHs (Aichner *et al.*, 2013), while Khalili *et al.* identified two- and three-ring PAHs as major products originating from multiple emission sources, including coke ovens, diesel and gasoline engines, and wood combustion (Khalili, Scheff and Holsen, 1995). Traffic emissions and fossil fuel combustion are the main identified sources of PAHs in urban areas (Fabia ska *et al.*, 2016; Keyte, Harrison and Lammel, 2013).

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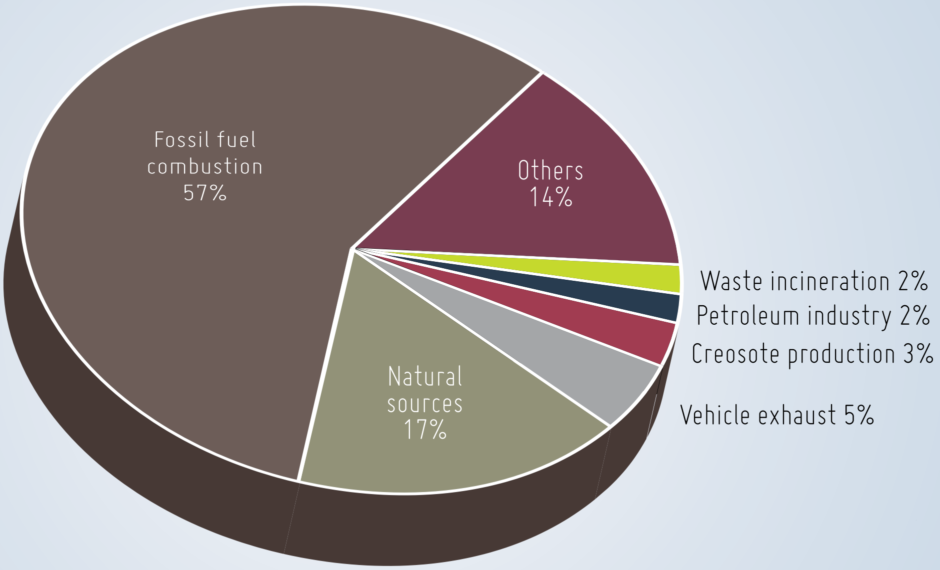

Figure 9. PAHs global emission sources. Source: Kuppusamy, *et al.*, 2017

Polycyclic aromatic hydrocarbons have attracted attention due to their high toxicity, mutagenicity, carcinogenicity, and widespread presence in the environment (EFSA, 2008). Although there are many PAHs, scientists and regulators have focused on 16 compounds that have been identified as carcinogenic (EC, 2011; US EPA, 1984). Recently, however, many different stakeholders support the implementation of broader regulations that include not only other toxic PAHs but also heterocyclic aromatic compounds and alkyl-derivatives (Andersson and Achten, 2015). In a global analysis of PAH distribution pattern in soils, Wilcke found that Germany and Czechia are more contaminated with PAHs than all other parts of the world that have been studied, such as China, the Russian Federation, Thailand, the United States of America, Brazil and Ghana (Wilcke, 2007). In a more recent study, Loganathan and Lam found higher concentrations of PAHs in soils in India compared to those in Africa, the Islamic Republic of Iran, Brazil, the Russian Federation, Canada and Australia (Loganathan and Lam, 2012). Thus, PAHs are ubiquitous contaminants in soils, but their concentration varies depending on the distance from the pollutant source, soil properties and climatic conditions.

Polycyclic aromatic hydrocarbons that enter soil may be attenuated or degraded by a number of physicochemical and biological processes such as volatilization and/or photo-oxidation to the atmosphere, irreversible sorption to soil organic matter, leaching to groundwater, abiotic loss (influence of daily seasonal temperature fluctuation), uptake by plants or microbial degradation (Okere, 2011; Šmídová *et al.*, 2012). The occurrence of PAHs in unprocessed food is very low and is governed by their relative solubility in water and organic solvents. Polycyclic aromatic hydrocarbons accumulate in the lipid tissues of plants and animals, but they do not tend to accumulate in plant tissues with a high water content. In general, limited transfer from the soil to root vegetables occurs (Abdel-Shafy and Mansour, 2016).

1.4.5 | PERSISTENT ORGANIC POLLUTANTS

Persistent organic pollutants (POPs) are chemical substances that persist in the environment, bioaccumulate through the food chain, and have adverse effects on human health and the environment (UNEP, 2001). There are many thousands of POPs, and their origins are numerous, as they have been used in agriculture, disease control, manufacturing and many industrial processes. POPs include chlorinated and brominated aromatics, such as polychlorinated biphenyls (PCBs), which have been useful in a variety of industrial applications, for example in electrical transformers and large capacitors, as hydraulic and heat exchange fluids, and as additives to paints and lubricants; and organochlorine pesticides such as DDT and its metabolites, which are still used to control mosquitoes that carry malaria in some parts of the world. Other chemicals, unintentionally produced, such as dioxins (polychlorinated dibenzo-p-dioxins and –furans), which result from some industrial processes and from combustion (municipal and medical waste incineration and backyard burning of household waste) are also included in this category (US EPA, 2014b).

Persistent organic pollutants are mainly hydrophobic and lipophilic compounds, and therefore present great affinity to organic matter and lipid membranes of cells and hence they can be stored in fatty tissue (Jones and de Voogt, 1999). The Stockholm Convention, a global treaty for protecting humans and the environment against POP contamination, has listed more than 20 POPs so far (Stockholm Convention, 2018). Persistent organic pollutants enter the food chain by accumulating in the body fat of living organisms and becoming more concentrated as they move from one organism to the next in a process known as “biomagnification” (Vasseur and Cossu-Leguille, 2006). Persistent organic pollutants also have high mobility: they can easily penetrate water in its gaseous phase during warm weather and volatilize from soils into the atmosphere. This can then lead to their deposition many miles away from the release point as temperatures cool (Schmidt, 2010). Examples of POP contamination through mobility include the discovery of significant amounts of POPs in isolated regions in the Arctic (AMAP, 1997; Muir and de Wit, 2010). As a general rule, the more chlorinated the molecule, the less water soluble and volatile it is. Polychlorinated biphenyls are poorly taken up by plants but susceptible to bioaccumulation by animals, mainly in adipose tissue and breast milk (Passatore *et al.*, 2014).

Since the publication of the book *Silent Spring* (Carson, 2002), a multitude of studies have focused on the effects of POPs on living organisms and the environment (de Boer and Fiedler, 2013; Cetin, 2016; Muir and de Wit, 2010; Mwakalapa *et al.*, 2018; Prestt, Jefferies and Moore, 1970; Ratcliffe, 1970; Vasseur and Cossu-Leguille, 2006). Yet there is still a lack of information on the presence of POPs in soils in developing countries (Fiedler *et al.*, 2013). Pollution by POPs could be expected to be significant in dumping sites of developing Asian countries, considering their poor management of municipal wastes and extensive use of such chemicals in the past (Minh *et al.*, 2006).

By the late 1970s most governments had banned PCB production, but an extensive environmental contamination still persists (Figure 10) as a consequence of accidental spills and leaks due to improper transport, storage and disposal (Jones and de Voogt, 1999; Passatore *et al.*, 2014). Despite their use and production having significantly been reduced since the adoption of the Stockholm Convention, POPs persist in the environment, affecting food security, health and the environment for many more generations (Odabasi *et al.*, 2016). Global agreement and increasing efforts and cooperation to remove POPs from soils is essential in preventing future pollution of crops and animals.

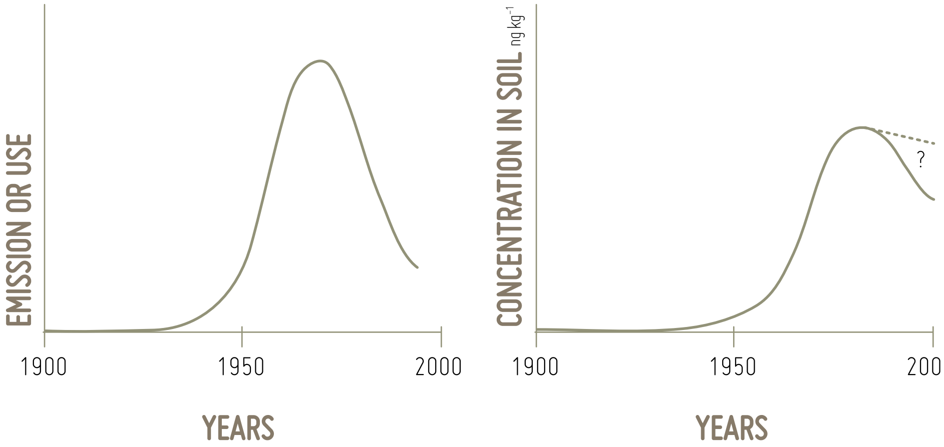

Figure 10. (a) Rate of production/use of POPs in North America and Europe and (b) POPs persistence in soils.
Source: Jones and de Voogt, 1999

Soils are the main environmental sink for these persistent pollutants. There, POPs form stable bonds with soil organic matter, where they remain in a non-extractable form. Changes in the soil environment, however, can change partitioning rates of POPs in the soil, leading them to become readily extractable. For example, an increase of 1 °C in air temperature produces an increase in the volatilization of POPs from soils by 8 percent (Komprda *et al.*, 2013). Cold temperatures favour POP deposition (Guzzella *et al.*, 2011), and forest soils can accumulate POPs over extended time periods due to their high organic carbon content (Kukučka *et al.*, 2009). The latitudinal distributions of these compounds (Figure 11) as a consequence of the temperature gradients have been described to respond to a “global distillation effect” (Wania and MacKay, 1996).

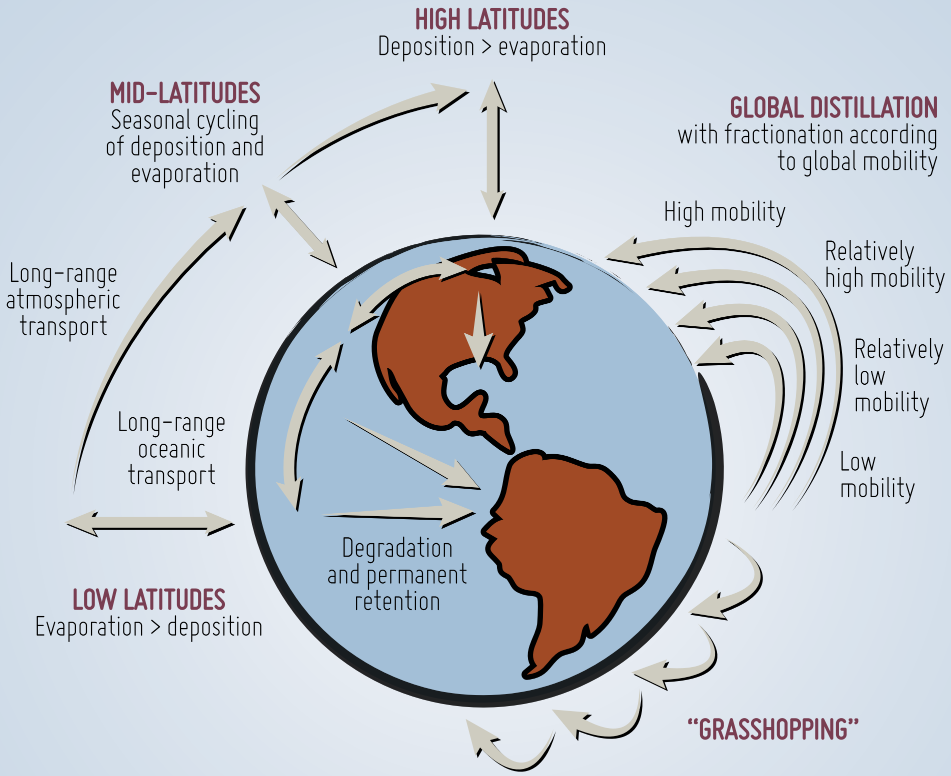

Figure 11. POPs migration processes. Source: Wania and MacKay, 1996

Per- and polyfluorinated alkyl substances (PFAS) represent a class of manmade chemicals that has been attracting more attention lately because of their abundance in the environment. Their presence has been confirmed in soils, water and human blood worldwide (Giesy and Kannan, 2001; Kannan *et al.*, 2004; Rankin *et al.*, 2016). Per- and polyfluorinated alkyl substances have been extensively used during the last few decades in military training operations (in fire-fighting foams and metal plating) (Lindstrom, Strynar and Libelo, 2011), and in commercial uses (as in the production of Gore-Tex™ and Teflon™) (Bossi, Dam and Rigét, 2015). Diverse chemical compounds are included in this category; they are highly resistant to degradation and they bioaccumulate in the food chain and result in biomagnification (Giesy and Kannan, 2001; US EPA, 2014c). Per- and polyfluorinated alkyl substances were included in the Stockholm Convention on Persistent Organic Pollutants in 2009 for their potentially harmful effects on human health and their high persistence in organisms, which has been estimated to be 100 days in laboratory rats, but over five years in humans (Wang *et al.*, 2009).

1.4.6 | RADIONUCLIDES

Radionuclides are present in the environment both as a naturally occurring substance and as one of anthropogenic origin (Mehra *et al.*, 2010; Navas, Soto and Machín, 2002). The emission of ionizing radiation during the decay of active atoms is the main contamination route of radionuclides, considering their long half-lives (Table 2).

Table 2. Characteristics of major radionuclides that occur in soil. Source: Zhu and Shaw, 2000

Isotope	Half-life (yr)	Principal radiation	Main occurrence
^{14}C	5.7×10^3	β^-	Natural and nuclear reactor
^{40}K	1.3×10^9	β^-	Natural
^{90}Sr	28	β^-	Nuclear reactor
^{134}Cs	2	β^-, γ	Nuclear reactor
^{137}Cs	30	β^-, γ	Nuclear reactor
^{239}Pu	2.4×10^4	α, X-rays	Nuclear reactor

The most common natural and anthropogenic radionuclides found in soils are ^{40}K, ^{238}U, ^{232}Th, ^{90}Sr and ^{137}Cs (Wallova, Kandler and Wallner, 2012). Anthropogenic sources of nuclear pollution include the global fallout from atmospheric nuclear weapons testing during the middle decades of the last century, operations of nuclear facilities and non-nuclear industry (e.g. coal fire power plants, nuclear waste handling and disposal, and mining of radioactive ores (Ćujić *et al.*, 2015), mineral fertilizers (Schnug and Lottermoser, 2013; Ulrich *et al.*, 2014; Van Kauwenbergh, 2010) and nuclear accidents (Three Mile Island, the United States of America (1979); Chernobyl, Ukrainian SSR (1986); Goiânia, Brazil (1987); Tokaimura (1999) and Fukushima (2011), Japan).

Radionuclides in the soil are taken up by plants, thereby becoming available for further redistribution within the food chain (Zhu and Shaw, 2000). For example, after the Fukushima accident, strict monitoring of agricultural products has been conducted to ensure food safety (Nihei, 2013). The monitoring demonstrated a fast decay in radionuclide content in vegetal products, but also discovered that radionuclides remained bioavailable in soils long after initial contamination (Absalom *et al.*, 1999; Falciglia *et al.*, 2014; Yablokov, Nesterenko and Nesterenko, 2009). Although topsoil removal is highly recommended after a major radioactive accident, it may be not possible for large areas as it would generate a huge amount of radioactive waste. For that reason, agricultural areas are often abandoned for many years. Agricultural countermeasures must be applied to reduce the transfer of radionuclides in the food chain and to facilitate the return of potentially affected soils to their agricultural use (Vandenhove and Turcanu, 2011). The transfer of radionuclides to animal-derived food products has also been analyzed (Howard *et al.*, 2009; Štrok and Smodiš, 2012), but the mechanisms involved are not yet completely clear or well understood.

1.4.7 | EMERGING POLLUTANTS

Emerging pollutants (EPs) refers to a large number of synthetic or naturally occurring chemicals that have recently appeared in the environment and are not commonly monitored (Geissen *et al.*, 2015). They have the potential to enter the environment and to cause known or suspected adverse ecological and/or human health effects. Emerging pollutants may well become pollutants of emerging concern, as new facts or information have demonstrated that they are posing a risk to the environment and human health (Sauvé and Desrosiers, 2014). Emerging pollutants encompass chemicals such as pharmaceuticals, endocrine disruptors, hormones and toxins, among others, and biological pollutants, such as micropollutants in soils, which include bacteria and viruses.

The anthropogenic production of chemicals has experienced a rapid growth globally since the 1970s. In the European Union in 2016, the chemical industry produced 319 million tonnes of hazardous and non-hazardous chemicals. Of these, 117 million tonnes were deemed to be hazardous to the environment (EUROSTAT, 2018). Global production is projected to increase annually by approximately 3.4 percent until 2030 (Figure 12), and non-OECD countries will be much greater contributors to this production in the future (OECD, 2008). Production and use of hazardous chemicals have been reduced over the last ten years; however, the uncertainties that still remain and the lack of information from many developing countries make it impossible to conclude that risks to the environment and human health have been successfully reduced.

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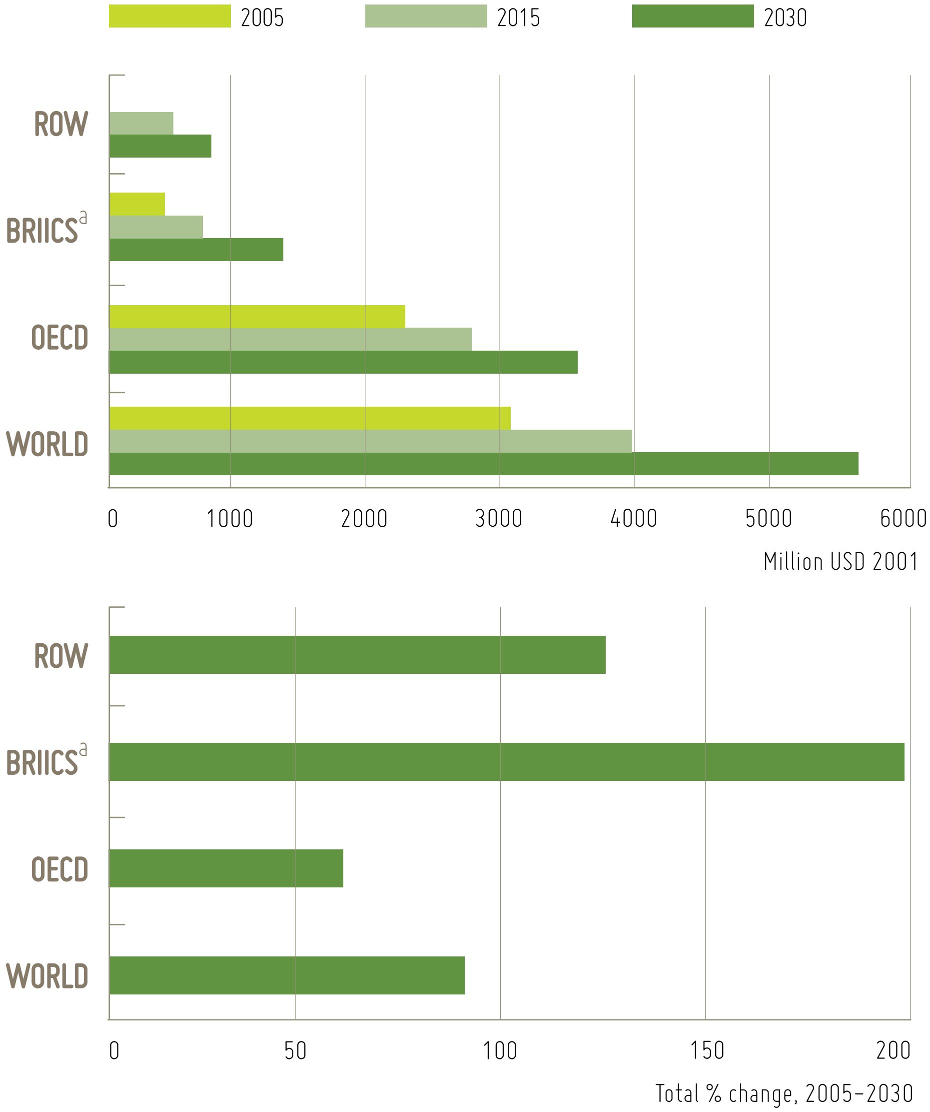


a) Includes Indonesia and South Africa.

Figure 12. Projected chemicals production by region for the period 2005-2030. Source: OECD, 2008

As an example, properties such as the adsorption behaviour of pharmaceuticals can vary vastly in different soil types as their occurrence in both ionized and un-ionized forms affects their interaction with different compounds in the soil (ter Laak *et al.*, 2006). The abundance of a high number of potentially toxic emerging pollutants in the environment reinforces the need to better understand their occurrence, fate and ecological impact (Petrie, Barden and Kasprzyk-Hordern, 2015).

Since the sources of EPs are varied and numerous, their nature, physical and chemical properties are also diverse. These include volatility, polarity, adsorption properties, persistence and their interaction with the environment. The main groups of emerging organic pollutants affecting agricultural soils are described below.

Pharmaceutical and personal care products (PPCPs) are a class of emerging chemical contaminants that have been used extensively for decades. There are more than 4 000 pharmaceutical and chemical products, including pharmaceutical drugs, diagnostic agents, cosmetics, fragrances, nutritional supplements and additives used in many household cleaning items. Many PPCPs are biologically active compounds (Boxall *et al.*, 2012), designed to interact with hormonal processes or living tissues; for this reason it is important to know their fate, effects and potential risks when they are released into the environment.

Since the late 1990s, detection of PPCPs in environmental matrices has been increasingly reported worldwide (Buser, Poiger and Müller, 1999; Hamscher *et al.*, 2004; Heberer, 2002; Jones, Voulvoulis and Lester, 2001). PPCPs enter urban wastewater streams but they are not efficiently eliminated by conventional treatment technologies (Miège *et al.*, 2009), and end up accumulating in sludge. Municipal biosolids have been found to be a sink for many PPCPs, and their land application as fertilizers may introduce these compounds into the environment, causing harm to beneficial microorganisms and affecting nutrient cycles. Limited information is available on the concentrations of PPCPs in soils amended with biosolids, even though their persistence in soil is significant (Wu, Spongberg and Witter, 2009; Wu *et al.*, 2010). Another problem associated with PPCPs is the presence of antimicrobial agents and their potential to promote bacterial resistance in the environment (Walsh *et al.*, 2003).

The possible effects of long-term PPCP exposure remain relatively unknown but may ultimately result in chronic toxicity to soil and aquatic organisms (Chalew and Halden, 2009). The presence of PPCPs has been linked to the development of antibiotic resistant bacteria, feminization of male fish, and genotoxicity in aquatic organisms (Daughton and Ternes, 1999). Further field studies are necessary to develop a reliable risk assessment for PPCPs (Walters, McClellan and Halden, 2010) and resistant bacteria and resistance genes (Sun *et al.*, 2018). The United Nations General Assembly has recognized the need for addressing antimicrobial resistance and reducing antimicrobial residues in soils (UN, 2016).

The occurrence and implications of antibiotic residues in the environment is an emerging concern. Antibiotics, fungicides, and other drugs are taken daily by humans, and they are extensively given to livestock to promote growth and reduce or prevent diseases. It is well known that pharmaceuticals, after administration, are absorbed and undergo metabolic reactions (e.g. hydroxylation, cleavage or glucuronation) to produce metabolites, which can be even more harmful than the original compounds or can be transformed back to the original active compounds (Díaz-Cruz and Barceló, 2005). Large fractions of the drugs are not assimilated or metabolized and are excreted in faeces or urine. Therefore, medicines are continuously being released into urban wastewater and animal manure. When manure and sewage sludge are applied to agricultural lands as fertilizers, or when treated wastewater is used for agricultural irrigation, crops are exposed to antibiotics that may persist in soils from a few to several hundred days. It has been documented that certain antibiotics, specifically tetracyclines, amoxicillin and fluoroquinolones, can be taken up by crop plants (Azanu *et al.*, 2016), while other PPCPs, such as miconazole (fungicide) and fluoxetine (antidepressants), are not present in crops, despite their persistence in the soil (Gottschall *et al.*, 2012).

Plasticizers are additives used to increase flexibility or plasticity, such as bisphenol A (BPA) or phthalates, and are particularly recognized as endocrine disruptors (Ghisari and Bonefeld-Jorgensen, 2009). Plasticizers have already been banned or more strictly regulated (CFR, 2017; EC, 2006; NICNAS, 1989; TwEPA, 2014). Phthalate esters (PAEs) are widely used as plasticizers and occur in many products such as lubricating oils, automobile parts, paints, glues, insect repellents, photographic films, perfumes, and food packaging. PAEs and BPA are founded in many agricultural soils close to urban or peri-urban areas, coming from sewage sludge application, agricultural use of plastic films, use of urban wastewater for irrigation or atmospheric deposition (Tran *et al.,* 2015; Zeng *et al.,* 2008). Phthalates and BPA are known to compete with endogenous hormones through binding to their specific receptors or by damaging their synthesis and metabolism (Craig, Wang and Flaws, 2011). Both phthalates and BPA have been detected in food and in humans and are listed as toxic agents in international regulations (Australian Government, 2018; EC, 2006; UNECE, 2011; US EPA, 2012; Yen, Lin-Tan and Lin, 2011).

Two other major groups of emerging contaminants are manmade nanoparticles (MNPs) and treatment by-products. The number of products that contain or require MNPs has greatly increased in the last few decades and they are present in more than a thousand products, including as additives in paints, cosmetics, textiles, papers, plastics and food (Fiorino, 2010). They are also used in textiles to produce clothes that are self-cleaning, water and dirt repellent, anti-microbial and ultraviolet and abrasion resistant. Manufactured nanoparticles are applied deliberately in soil remediation, with the purpose of reducing the impact of both organic and inorganic pollutants, and they are also released unintentionally in the soil through various other pathways (Pan and Xing, 2012).

The behaviour and interactions of MNPs with the soil matrix and with biological excretions and microorganisms (Figure 13) are not yet well understood (Nel *et al.,* 2006). This is due to the lack of available information on their properties, such as solubility, physical conformation, shape, and surface charge. Manmade nanoparticles' transformation before and after their entrance in the environment, such as surface modifications by humic acids, interactions with cations and dissolution, may control their fate in the environment (Liang *et al.,* 2013). Consequently, EP risk management remains fragmented and static (Geissen *et al.,* 2015). Treatment by-products are generated when water treatment (drinking or wastewater) generate new products from the reaction of the reagents with the components of the matrix or when reactions of the target contaminants are incomplete and some by-products that may have some residual toxicity are generated (Handy *et al.,* 2008).

Figure 13. Chemical and biological processes of MNPs in soil. Source: Pan and Xing, 2012

Manmade nanoparticles have a toxic effect on organisms through four main mechanisms: the generation of reactive oxygen species, causing oxidative stress; alteration of membrane cell permeation properties, thereby interfering with physiological activities; alteration of electron transfer process; and finally, modification of proteins conformation, which disturbs the transfer of bio-signals and gene formation (Pan and Xing, 2012).

Concerns about supply security and the broken biogeochemical phosphorus cycle have promoted the search for new, more sustainable and marketable products. New approaches to a circular economy promote the use of organic and waste-based fertilizers, such as struvite (ammonium–magnesium precipitate from the wastewater stream, slow-release fertilizer), biochar or ashes (from sewage sludge). These new fertilizer product categories aim to recycle nutrients that would otherwise have been lost. However, quality standards for these products, to assure safe application to land and crops, have yet to be established. They may contain heavy metals and residues similar to PPCPs (hormones, etc.) with adverse effects on the environment. Their production and marketability are predicted to increase globally in the next one to two decades. The European Union, for example, is addressing this new challenge by adopting a new action plan on the circular economy (EC, 2015), and is planning to regulate, amongst others, the content of contaminants in the frame of the new European Union Fertilizer Regulation (Vollaro, Galioto and Viaggi, 2017).

1.4.8 | PATHOGENIC MICROORGANISMS

Due to vast biodiversity and organism biomass, with more than 10 000 species per square metre and tonnes of bacterial biomass per hectarea (European Commission, Joint Research Centre and Global Soil Biodiversity Initiative, 2016), there is a great competitiveness for resources within the soil. Some organisms have developed chemical defences by excreting compounds that can kill or interfere with the growth of other microorganisms that encounter these compounds. Of these organisms, the vast majority pose no threat to human health, but rather function to provide numerous ecosystem services that emerge through the multitude of complex interactions between organisms within the soil and the soil itself. Some of these organisms, however, can be detrimental to humans by causing soil-borne diseases. They act either as opportunistic pathogens that take advantage of susceptible individuals, such as those who are immuno-compromised, or as obligate pathogens that must infect humans in order to complete their life cycles (Van der Putten *et al.*, 2011). Some of these organisms may be capable of surviving within the soil for extended periods of time before infecting humans who come into contact with contaminated soil, while others require the infection to occur more or less immediately after they leave the previous host.

Van der Putten *et al.* have defined soil-borne human diseases as “human diseases resulting from any pathogen or parasite, transmission of which can occur from the soil, even in the absence of other infectious individuals” (Van der Putten *et al.*, 2011). They have presented a comprehensive list of pathogens, differentiating between those that are truly soil organisms (euedaphic pathogenic organisms, EPOs) and those that can survive in soils for long periods of time as resistance structures even though they are obligate pathogens (soil transmitted pathogens, STPs) (Table 3).

Table 3. Soil borne infectious diseases (**bold**) and their causative agents (*italics*). Source: Van der Putten *et al.*, 2011

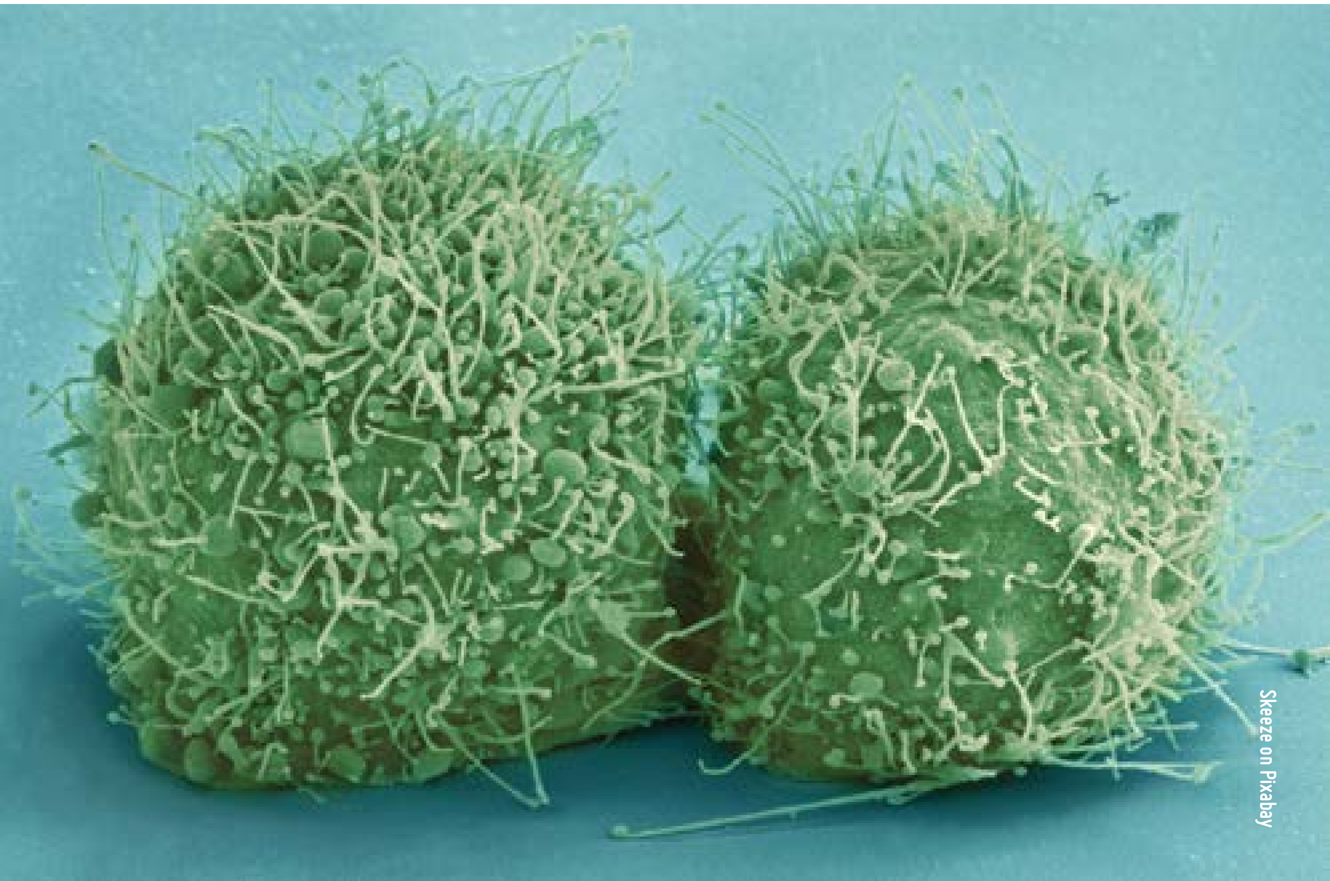
Euedaphic pathogenic organisms	Soil Transmitted Pathogens
Actinomycetoma: (e.g. *Actinomyces israelii*)	**Poliovirus**
Anthrax: *Bacillus anthracis*	**Hantavirus**
Botulism: *Clostridium botulinium*	**Q Fever:** *Coxiella burnetii*
Campylobacteriosis: e.g. *Campylobacter jejuni*	**Lyme disease:** *Borrelia* sp.
Leptospirosis: e.g. *Leptospira interrogans*	**Ascariasis:** *Ascaris lumbricoides*
Listeriosis: *Listeria monocytogenes*	**Hookworm:** e.g. *Ancylostoma duodenale*
Tetanus: *Clostridium tetani*	**Enterobiasis** (Pinworm)
Tularemia: *Francisella tularensis*	**Strongyloidiasis:** e.g. *Strongyloides stercoralis*
Gas Gangrene: *Clostridium perferingens*	**Trichuriasis** (Whipworm): *Trichuris trichiura*
Yersiniosis: *Yersinia enterocolitica*	**Echinococcosis:** e.g. *Echinococcus multicularis*
Aspergillosis: *Aspergillus* sp.	**Trichinellosis:** *Trichinella spiralis*
Blastomycosis: e.g. *Blastomyces dermatitidis*	**Amoebiasis:** *Entamoeba histolytica*
Coccidioidomycosis: e.g. *Coccidiodes immitis*	*Balantidiasis Balantidium coli*
Histoplasmosis: *Histoplasma capsulatum*	**Cryptosporidiosis:** e.g *Cryptosporidium parvum*
Sporotrichosis: *Sporothrix schenckii*	**Cyclosporiasis:** *Cyclospora cayetanensis*
Mucormycosis: e.g. *Rhizopus* sp.	**Giardiasis:** *Giardia lambila*
Mycetoma: e.g *Nocardia* sp.	**Isosporiasis:** *Isospora belli*
Strongyloidiasis: e.g. *Strongyloides stercoralis*	**Toxoplasmosis:** *Toxoplasma gondii*
	Shigellosis: e.g. *Shigella dyseneriae*, *Pseudomonas aeruginosa*, *Eschericia coli*
	Salmonellosis: e.g. *Salmonella enterica*

Some pathogens may originate from animal faeces, and soil represents the main pathway of contamination through dermal contact or contact with contaminated water and food (Ercumen *et al.*, 2017). Helminths, a type of parasitic worms, are present in human faeces, and contaminate the soil in areas with poor sanitation. The World Health Organization (WHO) has estimated that two billion people are infected by soil-transmitted helminths worldwide (WHO, 2001b). Unsafe agricultural practices such as irrigation with untreated domestic wastewater and soil amendments with improperly treated livestock manure are very commonly used by small-holder farmers mainly in developing countries, but these are also practices that can affect developed countries (Allende and Monaghan, 2015; Ongeng *et al.*, 2011; du Plessis, Duvenage and Korsten, 2015; Scallan *et al.*, 2011). The consumption of raw or minimally processed fruits and vegetables, such as lettuce, spinach, and carrots, has increased significantly due to their importance for a healthy diet. When they are produced using improper practices, however, they may become a source of enteric pathogens, as demonstrated by the increasing number of documented human infections associated with fresh produce consumption (Beuchat, 2002; Ingham *et al.*, 2004). Contact with contaminated soils has been identified as a potential source of food contamination (Khandaghi, Razavilar and Barzgari, 2010).

1.4.9 | ANTIMICROBIAL RESISTANT BACTERIA AND GENES

Bacteria are very adaptable genetically, and when confronted repeatedly with antibiotics, mutational changes can occur, changes that lead to resistance to the antibiotic. The increased and widespread use of antibiotics is therefore developing antibiotic-resistant bacteria in the environment (Rensing and Pepper, 2006). The other way microbes can become resistant is through the transference of foreign antibiotic resistant genes (Rensing and Pepper, 2006) naturally present in soils or from bacteria introduced by agricultural practices (e.g. animal husbandry, human wastewater disposal, improperly composted manures) or from domesticated and wild animal faecal droppings to the soil microbiome communities (FAO, 2016). Antimicrobial resistance (AMR) is one of the major issues facing society: AMR infections currently claim around 50 000 lives each year in Europe and the United States of America, and by 2050, if the issue is not tackled, it has been predicted that they will kill more people than cancer, and cost, globally, more than the size of the current global economy (O'Neill, 2014). The recent worldwide enrichment and spread of highly resistant pathogenic bacteria in the micro-biosphere has largely been driven by human activities, including the extensive use and misuse of antibiotics in human and veterinary medicine and in agriculture (Witte, 1998).

Antimicrobials are often administered to livestock for growth promotion, prophylaxis, and disease treatment (Joy *et al.*, 2013). Estimated global antimicrobial consumption in the livestock sector in 2010 was 63 151 tonnes (FAO, 2016). However, a substantial portion of the antimicrobials administered to livestock are not absorbed by the animals (Sarmah, Meyer and Boxall, 2006). Once animal manure is applied to the land, the fate of manure-originating antimicrobials in soil and their subsequent transport by runoff will also be affected by the compounds' sorption properties to soil particles (Sassman and Lee, 2005).



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1.5 | INTERACTION OF POLLUTANTS WITH SOIL CONSTITUENTS

One of the main ecological services provided by soil is filtering, buffering and transforming inorganic and organic contaminants. This essential function ensures good quality of groundwater and safe food production (Blum, 2005). When pollutants enter the soil, they undergo physical, physicochemical, microbiological, and biochemical processes that retain, reduce or degrade them.

Important soil characteristics that affect the behaviour of contaminants include soil mineralogy and clay content (soil texture); amount of soil organic matter (SOM); pH (acidity) of the soil; moisture levels; and temperature. The properties of the contaminants themselves are also very important and include the size, shape, molecular structure, solubility, charge distribution, and acid–base nature of the molecule (Gevao, Semple and Jones, 2000).

1.5.1 | SORPTION OF CONTAMINANTS

Sorption (or adsorption) is a process whereby the molecules of a fluid interact with a solid and are retained on the solid for a time (Navarro, Vela and Navarro, 2007). Sorption may be chemical in nature (as with ionic and hydrogen binding) or purely physical (as with van der Waals forces).

Ions or molecules that are positively charged (cationic) participate in cation exchange on charged surfaces. Soil organic matter (SOM) and clay minerals are the source of cation exchange sites in soil and the cation exchange capacity (CEC) of various SOM fractions and types of clay minerals differs greatly. Negatively charged (anionic) ions or molecules are, in general, more weakly bound in soils and react with SOM primarily through hydrogen bonding and ligand exchange (Gevao, Semple and Jones, 2000). The oxidation states of some ions or molecules can transition through cationic, neutral, or anionic states in response to the pH of the soil solution; hence their sorption in the soil is pH dependent. Additionally, some molecules can gain or lose protons and thus exhibit acidic or basic behaviour; the sorption of these molecules is also pH dependent. Finally, changes in the presence or absence of oxygen due to changing soil moisture conditions (i.e. the redox status of the soils) also causes changes in the oxidation states of some ions and compounds and can be an important control on the mobility of these contaminants.

Hydrogen bonding is an important sorption mechanism for non-ionic pesticides that exhibit polarity (i.e. asymmetric distribution of electrical charge in a molecule). Many pesticides are both non-ionic and non-polar and react with SOM through physical van der Waals forces. These forces are strongest for ions that are in close contact with the surface or can conform to the surface; hence the size and structure of the pesticide is an important control on their sorption (Gevao, Semple and Jones, 2000). As well, non-polar molecules are not attracted to polar water molecules and hence are typically insoluble in water (i.e. hydrophobic). Some non-polar and hydrophobic compounds undergo extensive and complex reactions with SOM that lead to their long-term sequestration. For example, DDT is highly insoluble and has a strong affinity for SOM, which accounts in part for the great persistence of DDT in the environment (Mansouri *et al.*, 2017).

Several well-established generalizations can be made about the sorption of contaminants and soil properties. First, the SOM content of the soil (especially the content of highly reactive humus) is the dominant control on sorption. Soil organic matter provides both charged sites for ionic reactions and highly complex structures that enhance physical sorption processes. Dissolved organic matter may also interact with nanoparticles, changing their surface properties and aggregation status, thereby increasing the mobility and bioavailability of the nanoparticles (Pan and Xing, 2012; Wang *et al.*, 2011). Second, the clay content and the nature of the clay minerals is a strong secondary control on sorption. Studies conducted worldwide have demonstrated that the fine-grained soil fraction exhibits a greater tendency for ionic adsorption than do coarse-grained soils. Since the fine fraction contains soil particles with large surface areas such as clay minerals, iron and manganese oxy-hydroxides, and humic acids (Bradl, 2004). The type of clay mineral present is important when comparing temperate-region soils to those of the tropics – soil of the tropics can be highly weathered, with low activity clays whose charge is highly pH dependent (Lewis *et al.*, 2016). Third, and following from the first two generalizations, sandy soils low in organic matter present a particular risk for leaching of contaminants due to the paucity of sorption sites.

1.5.2 | BIOAVAILABILITY, MOBILITY AND DEGRADATION OF CONTAMINANTS

Bioavailability refers to the physical, chemical and biological interactions that determine the exposure of organisms to chemicals associated with soils (Committee on Bioavailability of Contaminants in Soils and Sediments, 2002).

Metals

The soil's sorptive capacity has a major effect on the bioavailability of heavy metals and metalloids. Biological uptake of metals by soil biota and plants occurs only when the metals are in ionic form. Many metals occur as simple cationic forms (Table 4), but some like As and Cr form more complex oxyanions. Metals are adsorbed on the surface of very fine organic matter in soils (humus), clay minerals, Fe and Mn hydrous oxides and some sparingly soluble salts such as calcium carbonate (Morgan, 2013). A similar affinity has been observed between clay minerals and radionuclides (van der Graaf *et al.*, 2007). Metals also form complex compounds through interactions with organic molecules; Cu has a particular affinity to forming such compounds (Morgan, 2013).

Many sorption processes for metals are pH dependent. Sorption is highest in less acidic soils, while acidic conditions favour desorption and release of the metals back into solution. Anaerobic conditions caused by water saturation can also result in desorption of some metals.

The addition of organic and inorganic amendments is very effective in reducing the bioavailability of heavy metals in soils by increasing the number of binding sites and through modification of the soil's pH (Puschenreiter *et al.*, 2005). These amendments include compost, biosolids (sewage sludge), manure and by-products of industrial activities. Such measures can have many positive implications for the environment and the same time contribute to waste reduction. Lime application increases soil pH and reduces metal uptake by crops (Knox *et al.*, 2001).

Table 4. Dominant soil species for common soil metal pollutants. Source: Logan, 2000

Element	Symbol	Dominant Soil Species
Arsenic	As	AsO_3^{2-}, AsO_4^{3-}
Cadmium	Cd	Cd^{2+}
Chromium	Cr	Cr^{3+}, CrO_4^{2-}
Copper	Cu	Cu^{2+}
Mercury	Hg	Hg^{2+}, $(CH_3)_2Hg$
Nickel	Ni	Ni^{2+}
Lead	Pb	Pb^{2+}
Zinc	Zn	Zn^{2+}

Radionuclides

Clay content, pH and organic matter also play an important role in the sorption of ^{137}Cs and other radionuclides in soils (Absalom, Young and Crout, 1995; Rigol, Vidal and Rauret, 2002). Microorganisms play an important role in geochemical changes to radionuclides by catalyzing chemical transformations in the subsoil (Turick, Knox and Kuhne, 2013).

The Kd distribution (or partitioning) coefficient is used to describe the propensity of a radionuclide to be sorbed; low Kd values indicate lower sorption of the radionuclide.

Cesium is not very chemically reactive and has a similar behaviour in soils to that of potassium. It has enhanced bioavailability in low clay soils and in soils high in kaolinite (Kd = 240 to 290) but is strongly bound to illite clay minerals (Kd= 6 300 to 8 300) (Turick, Knox and Kuhne, 2013).

Iodine has multiple redox states and hence exhibits complex behaviour; the -1, +5 and molecular I_2 oxidation states are most relevant in environmental systems. The Kd values for mineral soils range between 0.04 and 81 (Turick, Knox and Kuhne, 2013) and hence overall sorption of I in soils is low. Sorption of iodine species appears to be controlled in part by SOM and in part by iron and aluminum oxides; adsorption increases under more acid conditions.

For uranium, the +4 and +6 states are the most important from a biological perspective. Uranium in the +4 state is mostly insoluble and largely immobile, whereas the +6 state is soluble and mobile in the environment. Uranium sorption is strongly pH dependent. For example, for reactions between U and the mineral apatite, the value for Kd is 668 at a pH of 4 and 24 660 at a pH of 7 (Turick, Knox and Kuhne, 2013).

Plutonium has a complex geochemistry and can exist in +3, +4, +5, and +6 oxidation states. It generally binds easily to surfaces in soil matrix and thereby becomes immobilized. It is also pH dependent, with maximum adsorption at pH6 (Turick, Knox and Kuhne, 2013).

Pesticides

There is a very great range in the chemical composition and structure of pesticides and hence in their interactions with soil constituents (Gevao, Semple and Jones, 2000). The distribution coefficient (Kd) is the concentration of pesticide sorbed by the soil divided by the concentration in solution. A higher value for Kd indicates that the pesticide is more strongly sorbed. In many soils the amount of organic matter present largely controls the sorption capacity of the soil, and the sorption coefficient (Koc) is calculated by dividing the Kd value by the amount of organic carbon in the soil. The higher the Koc value, the more strongly sorbed the chemical is, and hence the less mobile it is in the environment. Generally, pesticides with high Koc values like glyphosate and endosulfan and especially DDT will be relatively immobile, but other factors do come into play (Table 5). For example, Dores *et al.* observed that despite a relatively high Koc, leaching of endosulfan occurred in the Brazilian soils they examined (Dores *et al.*, 2016); they attributed this to preferential flows paths in the soils.

Table 5. Values for sorption coefficient Koc from the literature. Source: Wauchope *et al.*, 2002 except for [1] (Farenhorst *et al.*, 2008) and [2] (Dores *et al.*, 2016).

Chemical	Koc mL g^{-1}	Chemical	Koc mL g^{-1}
2,4-D	20–32	α-Endosulfan[2]	8 725–31 992
Atrazine	163–172	β-Endosulfan[2]	8186–31992
Carbofuran	29.4	Glyphosate[1]	10 891–14 863
Chlorthiamid	98–107	Lindane	1081–1340
Chlorpyrifos[2]	1 671–2 896	Malathion	20
Dicamba	2.2	Paraquat	20 000
DDT	243 000		

Microbial diversity and activity, especially bacteria and fungi, will also determine the bioavailability of pollutants, as microorganisms are able to degrade and transform certain pollutants, releasing byproducts and affecting their toxicity and mobility (Burgess, 2013). Although most pesticides have novel structures previously unseen in nature, some are able to be metabolized by microorganisms (Topp, 2003). Once microorganisms that can degrade pesticides have been identified, they can be used to inoculate and decontaminate polluted soils in the process of bioremediation.

Aerobic or anaerobic conditions have also been demonstrated to have a significant effect on pollutant persistence and bioavailability. Ying, Yu, and Kookana, for example, observed that biodegradation of triclocarban and triclosan, two antimicrobials widely present in personal care products, occurs under aerobic conditions but not under anaerobic conditions, where the persistence is much longer (Ying, Yu and Kookana, 2007). Dechlorination bacteria, responsible for anaerobic degradation of chlorinated compounds, need time to adapt to target chemicals (Brahushi *et al.*, 2004), and this adaptation seems to happen faster when the pollutant is present in high concentrations, as was observed in two adjacent polluted soils in the Netherlands (Middeldorp *et al.*, 2005).

Persistent organic pollutants

By definition the POPs are resistant to degradation in the soil due to factors such as strong sorption, hydrophobicity, or a structure that is resistant to microbial degradation. Polychlorinated biphenyls are hydrophobic, non-polar, and inert, as are the PCDDs and PCDFs. For the PAHs their degree of hydrophobicity and chemical reactivity decreases with increasing molecular weight; hence, generalization about their fate is difficult (Burgess, 2013).

Nitrogen and phosphorus

The different forms of N and their bioavailability and mobility are very well established (Cameron, Di and Moir, 2013). Nitrogen is available in four major forms in the soil: a) SOM, b) soil organisms and microorganisms, c) ammonium (NH_4^+) ions bound to soil particles, and d) mineral-N forms in soil solution including NH_4^+, nitrate (NO_3^-) and small amounts of nitrite (NO_2^-) (Cameron, Di and Moir, 2013). The available forms for plant uptake are nitrate in aerobic soils and ammonium in flooded wetlands or acidic soils (Krapp, 2015). As an anionic compound, NO_3^- is not strongly sorbed and nitrate leaching from soils is both a loss of fertility and a threat to the environment and to human health. Nitrate in drinking water has been associated with methaemoglobinaemia in babies and with cancer and heart disease (Cameron, Di and Moir, 2013). Nitrogen is also lost from soils as a gas. Ammonium volatilization is associated with N fertilizer additions and animal urine and faeces deposition, and the highly potent greenhouse gas N_2O is released at several points in the N cycle, but the greatest emissions occur from denitrification from saturated soils.

Phosphorus in the soil exists primarily in orthophosphate (HPO_4^{2-}, $H_2PO_4^-$) and organic forms; it also occurs as sorbed form, forming surface complexes with Al, Fe oxides, other minerals, and organic matter. Phosphorus is strongly retained by solid phase and transported as eroded solid particles and through transport of manure and human waste (Yuan *et al.*, 2018). Phosphorus losses from cropland through erosion and runoff are dominant contributors to aquatic systems: Yuan estimates P losses from cropland to freshwater is 10.4±5.7 Tg P/yr, and natural losses from erosion at 7.6±3.3 Tg P/yr. In their study of P sources in British rivers, Civan *et al.* found that point sources of P usually contain higher amounts of orthophosphate and more biologically available phosphorus, whereas diffuse sources generally were sorbed to particulates (from farmyard runoff, pig slurry, and erosion from fields) (Civan *et al.*, 2018). Overall, they found a 60 percent decline in mean total reactive P concentration in British rivers since a peak in 1985, which they ascribe to actions taken at sewage treatment plants.

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2 | THE IMPACTS OF SOIL POLLUTION ON THE FOOD CHAIN AND ECOSYSTEM SERVICES

The predicted world's population of over nine billion by 2050 will require the provision of enough good quality food and water (Godfray *et al.*, 2010; McBratney, Field and Koch, 2014). According to Dubois (Dubois, 2011), food production will increase by 70 percent by 2050 globally, and by 100 percent in developing countries, compared with 2009 production levels. FAO's latest projections indicate that global food production will increase by 60 percent between 2005/07 and 2050 under its baseline scenario. This represents a downward revision, based on updated data and information, from the 70 percent increase projected for the same period in 2009. (World Agriculture Towards 2030/2050: The 2012 revision ESA E Working PaperNo. 12-03 http://www.fao.org/economic/esa/esag/en/). The quantity and nutritional quality of food supports human health, and 95 percent of food production depends on soils (Oliver and Gregory, 2015; FAO, 2015). Only healthy soils can provide the needed ecosystem services and secure supplies of more food and fibre. The provision of ecosystem services has received considerable attention and can be defined as "the capacity of natural processes and components to provide goods and services that satisfy human needs, directly or indirectly" (Groot, 1992). Food security is defined as "the availability, access, utilization and stability of food supply." Soil pollution reduces food security both by reducing crop yields due to toxic levels of contaminants and by causing the produced crops to be unsafe for consumption (FAO and ITPS, 2015).

2.1 | SOIL POLLUTION, PLANT UPTAKE AND FOOD CHAIN CONTAMINATION

The pathways of contamination within the food chain by the transfer of soil pollutants through plants are shown in Figure 14.

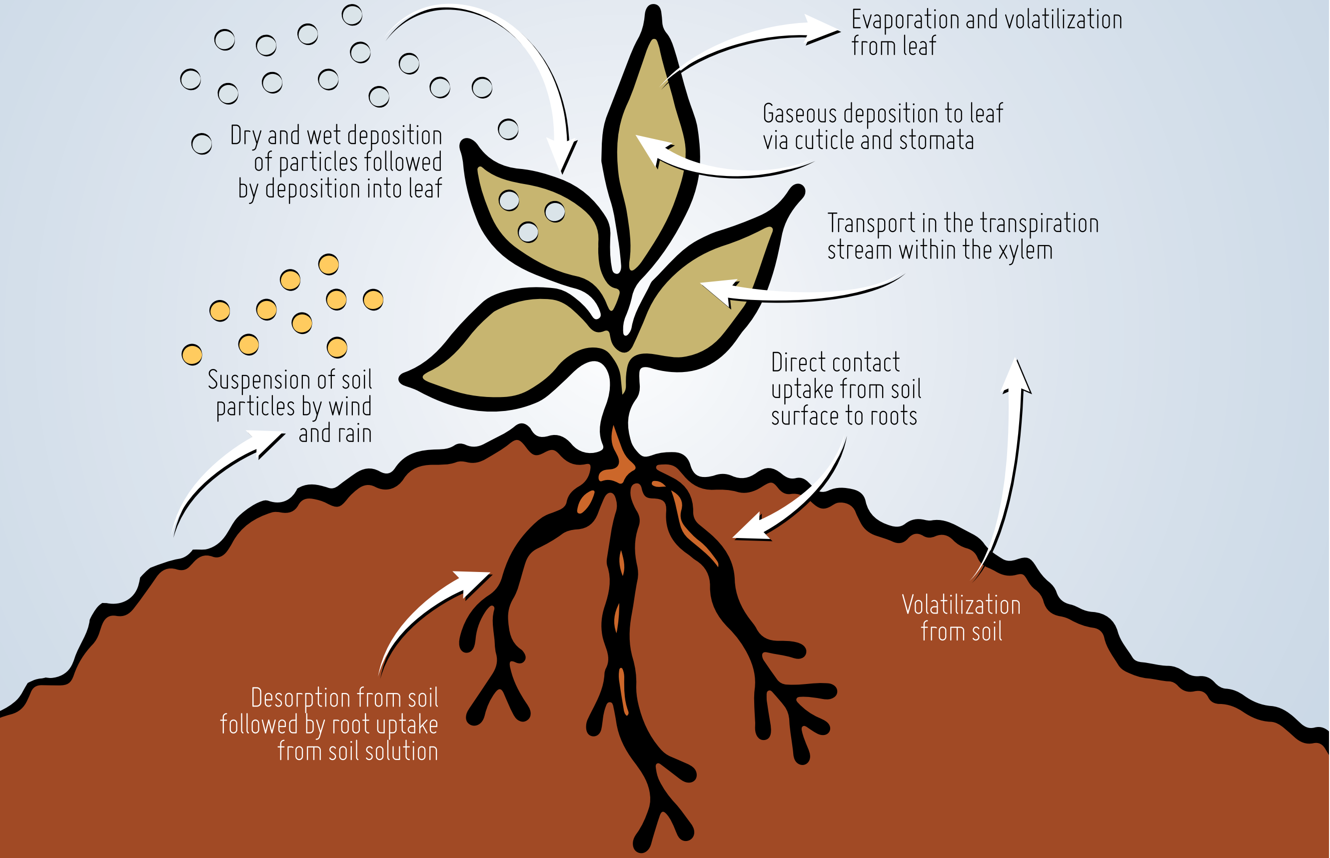

Figure 14. Principal uptake pathways for the uptake of soil contaminants by plants (adapted from Collins, Fryer and Grosso, 2006)

If a contaminant is highly toxic to plants at low concentrations and is not easily translocated to shoots, fruits or tubers to pose a hazard to animals and humans, it is unlikely to enter the food chain and become a hazard. This concept was termed the "Soil-Plant Barrier" by Chaney almost 40 years ago for metals and metalloids (Chaney, 1980). Chaney defined four groups of metals entering the food chain when sewage sludge was applied to soil, as a function of their danger to human health (Table 6).

Table 6. Metals/metalloids classified in groups according to potential food-chain risk via plant uptake.
Adapted from: Chaney, 1980

Group 1	Group 2	Group 3	Group 4
Silver (Ag)	Mercury (Hg)	Boron (B)	Arsenic (As)
Chromium (Cr)	Lead (Pb)	Copper (Cu)	Cadmium (Cd)
Tin (Sn)		Manganese (Mn)	Cobalt (Co)
Titanium (Ti)		Molybdenum (Mo)	Molybdenum (Mo)
Yttrium (Y)		Nickel (Ni)	Selenium (Se)
Zirconium (Zr)		Zinc (Zn)	Thallium (Tl)

Group 1 comprises the elements that pose a low risk of food chain contamination because they are not taken up by plants, due to their low solubility in soil, which means negligible uptake and translocation by plants. Elevated concentrations of these elements in foods usually indicate direct contamination through soil or dust accumulation. Group 2 includes elements that are strongly sorbed by soil surfaces, and while they may be absorbed by plant roots, they are not readily translocated to the edible tissues and therefore pose minimal risks to human health. These elements could, however, pose a risk to grazing animals (or humans) if contaminated soil is ingested. Group 3 comprises the elements that are readily taken up by plants, but that are phytotoxic at concentrations that pose little risk to human health. Conceptually, the "soil-plant barrier" protects the food chain from contamination by these elements. Group 4 consists of elements that are at the highest risk for food-chain contamination as they pose risks to human or animal health at plant tissue concentrations that are not generally phytotoxic. Chaney originally classified As in Group 2, but research over the last 20 years has indicated that flooded rice systems are at risk from As transfer through the food chain due to low redox conditions in flooded soils. This increases the solubility of As for uptake by rice and hence As should now be classified as a high-risk, Group 4 element. Contamination of soils by As and Cd is perhaps the most widespread risk to the food chain globally (Grant *et al.*, 1999; McLaughlin, Parker and Clarke, 1999), with large areas of South-East Asia having soils contaminated by As (Meharg, 2004) and Cd (Hu, Cheng and Tao, 2016).

In some parts of China, soils polluted by heavy metals are nevertheless used to grow grain. The grain grown in these soils is often in turn polluted with heavy metals. According to China Dialogue, an estimated 12 million tonnes of polluted grain must be disposed of each year, costing Chinese farmers up to CNY 20 billion, or about USD 2.57 billion (Lynn, 2017).

Excess heavy metals such as arsenic, cadmium, lead and mercury in soils can also impair plant metabolism and decrease crop productivity, ultimately putting pressure on arable land. When they enter the food chain, these pollutants also pose risks to food security, water resources, rural livelihoods and human health. The uptake and translocation of metals into above ground tissues are conditioned by genetic and physiological differences of plants (Chen, Li and Shen, 2004), as well as by the concentration of metals in the soil and the exposure time (Rizwan *et al.*, 2017; Tőzsér, Magura and Simon, 2017). Once metals enter plant tissues, they may interfere with several metabolic processes, reducing plant growth and causing toxicity and finally plant death. Decreases in germination rates, oxidative damage, lower roots and shoots elongation and alterations of sugar and protein metabolisms were the main effects reported (Ahmad and Ashraf, 2011). High levels of lead, for example, accelerate the production of reactive oxygen species, causing lipid membrane and chlorophyll damage that further leads to the alteration of photosynthetic processes and of the overall growth of the plant (Najeeb *et al.*, 2017). Cadmium can accumulate in different edible tissues (Baldantoni *et al.*, 2016), causing reduction of root, stem and leaf growth, decreasing net photosynthesis and water use efficiency and altering nutrient uptake (Rizwan *et al.*, 2017).

Radionuclides may also present a potential threat to food quality, through atmospheric deposition of radionuclides on soil from nuclear energy accidents, or the addition of radionuclides to the soil through fertilizers or through wastes and byproducts from the nuclear industry (Mortvedt, 1994). The potential for transfer of radionuclides from soil to plants and to the food chain was first identified in the 1950s both in restricted areas where nuclear weapons testing had taken place and more generally through deposition of fallout from bomb testing. The Chernobyl nuclear accident in 1986 caused widespread pollution of soils by radionuclides (principally ^{131}I, ^{134}Cs and ^{137}Cs) (Bell, Minski and Grogan, 1988). Uptake of pollutants from soils into forages was followed by the contamination of grazing animals and led to restrictions being placed on the sale and slaughter of sheep from affected areas in the United Kingdom of Great Britain and Northern Ireland (Smith *et al.*, 2000). Widespread contamination of the food chain by the same radionuclides also occurred after the Fukushima nuclear accident in Japan (Berends and Kobayashi, 2012).

Compared to metals, metalloids and radionuclides, the global footprint of soils that are highly contaminated by organic contaminants is much smaller, with contamination of the food chain being localized around industrial or urban centres. Contamination occurs through waste re-use on land and as a legacy of the use or disposal of persistent and bioaccumulative organic chemicals (principally organochlorines (OCs), polychlorinated dibenzo-p-dioxin (PCDD), polychlorinated dibenzofurans (PCDF) and poly- and perfluoroalkyl substances (PFAS)). Levels of soil contamination are usually lower than for metals/metalloids, generally in concentrations less than mg/kg in soil, especially for dioxins, furans and PFAS compounds.

The pathway of uptake of organic contaminants into the food chain depends on the properties of the organic contaminant – principally their volatility, hydrophobicity and solubility in water. Hydrophilic organic contaminants with low volatility (e.g. PFAS) will principally enter the food chain through root uptake and translocation to food parts (Navarro *et al.*, 2017). On the other hand, volatile hydrophobic contaminants (e.g. dioxins, furans and polychlorinated biphenyls (PCBs)) will tend to accumulate in the food chain through atmospheric uptake, as they are generally strongly sorbed in soils (Collins, Fryer and Grosso, 2006; Simonich and Hites, 1995). Some plant species, however, can accumulate these compounds through uptake from soil (Huelster, Mueller and Marschner, 1994). Sun *et al.*, were able to quantify organic pollutants and their residues in plants by measuring the uptake of ^{14}C-residues (Sun *et al.*, 2018). Many studies have demonstrated the uptake of pesticide residues through several main pathways. These residues are incorporated in plant tissues and transferred to the end consumer (Randhawa *et al.*, 2014). However, unlike metals and metalloids, the incidence and severity of adverse effects on human health caused by soil contamination by organic chemicals is much less well documented or demonstrated, likely because smaller areas of land are affected by this type of pollutant, and contamination levels are generally lower.

Atmospheric deposition and gas exchange through the stomata are the main mechanisms of absorption of POPs by plants; the pollutants are later translocated to other plant tissues and accumulated in their hydrophobic lipids and waxes (Odabasi *et al.*, 2016). Root uptake of POPs from soils is limited since POPs are tightly bound to soil particles (Collins, Fryer and Grosso, 2006). For that reason, soils can be viewed as a reservoir and as a source of POPs with low and medium volatility; these may enter the food chain after being taken up by plants from the atmosphere. Absorption efficiency of POPs across the gastro-intestinal tract and their storage and release dynamics are intimately linked to that of fat storage and metabolism (Sweetman, Thomas and Jones, 1999).

2.2 | IMPACT ON ECOSYSTEM SERVICES OF SOIL POLLUTION FROM AGRICULTURE

Entry of pollutants directly (release of effluents on land) or indirectly (use of polluted water as irrigation to crops) has been reported to contaminate vast areas of soil resources and groundwater bodies, affecting crop production as well as human and animal health through food contamination (Saha *et al.*, 2017).

Agricultural inputs such as fertilizers, pesticides, antibiotics contained in animal manure or the ones used for illness prevention and infection treatment in plants are major potential pollutants in agricultural lands and pose special challenges due to the fast-changing chemical formulas employed (GSP, 2017). Intensification of agriculture to produce enough food, fibre and biofuel has led to a heritage of polluted soils. In China, heavy metals contents have considerably increased in the last 30 years, with values oscillating between 48 percent for Zn to over 250 percent when Cd is compared with its background levels in 1990 (Zeng, Li and Mei, 2008). However, the transfer of pollutants from soil to plants is not yet well understood, and the question "is food produced in healthier soils also more nutritious?" needs stronger scientific evidence to engage policy makers, governments and land users towards sustainable and environmentally friendly practices and to leave behind more business-oriented approaches.

2.2.1 | SYNTHETIC FERTILIZERS

Modern agriculture practices accelerate soil pollution with the intensive use of fertilizer and pesticides in order to increase productivity and reduce crop losses. When pollutants reach high levels in the soil, not only do soil degradation processes take place, but crop productivity can also be affected. Therefore, in addition to endangering human health and the environment, soil pollution can also cause economic losses.

Excess N in soil has been identified as the main cause of soil acidification and salinization through nitrification and other N-transformation processes. Soils acidify very slowly under natural conditions over hundreds to millions of years (Guo *et al.,* 2010), but this process is significantly accelerated by agricultural practices, mainly excessive N fertilization, which causes reductions in soil pH by 0.26 pH units on average in different land uses (Lucas *et al.,* 2011; Tian *et al.,* 2015; Zhao *et al.,* 2014a). The analysis of acidification sources in agricultural soils in China demonstrated that anthropogenic acidification driven by N fertilization is indeed the main cause, being 10 to 100 times greater than that associated with acid deposition (Guo *et al.,* 2010).

2.2.3 | ACIDIFICATION AND CROP LOSS

Acidification of agricultural soils may contribute to further soil pollution, through the mobilization of toxic heavy metals. If the content of nitrogen applied to agricultural soils is higher than the plants' requirements, nitrification microbial activity will lead to the accumulation of nitrates (NO_3^-) that can easily leach to groundwater due to their high solubility, polluting it (Tian *et al.,* 2015). When soil nutrient availability increases, microbial biomass and activity increases as well, but the microbial biodiversity is altered, causing imbalances in the nutrient cycle (Lu and Tian, 2017).

The main risk from P fertilizers is transport to surface water bodies, which has been documented to cause eutrophication of aquatic ecosystems in many regions (Stork and Lyons, 2012; Syers, Johnson and Curtin, 2008). The P is transported to water bodies adsorbed to eroded soil particles or from excessive amounts of P fertilizer or animal manure applied when conditions are not suitable (Syers, Johnson and Curtin, 2008). Many farmlands receive more P inputs than the amount that crops are able to take up, causing a soil-P surplus, at least in the short term (Aarts, Habekotté and Keulen, 2000; Syers, Johnson and Curtin, 2008).

2.2.4 | PESTICIDES

An extensive review of scientific research about the effects of pesticide use on soil functions was recently undertaken by the Intergovernmental Technical Panel on Soil (FAO and ITPS, 2017). The main scientific-based evidence presented in this work showed an increase in the farmers' net return when they applied pesticides, however the benefits of pesticide use are usually assessed by comparing use of synthetic pesticides versus no use of pesticides rather than comparing synthetic pesticides to biological control of pests (Cai, 2008). Negative associated impacts of specific pesticides on soil organisms and soil functions have been also reported.

For example, some organochlorine pesticides suppress symbiotic nitrogen fixation, resulting in lower crop yields (Fabra, 1997; Fox *et al.*, 2007; Santos and Flores, 1995). The FAO and ITPS report also highlights the knowledge gap on the relationship between pesticides and soil health, mainly on soil pollution (FAO and ITPS, 2017). The international efforts to assess the ecotoxicological risk of pesticides and to control their use and release in the environment, through the Rotterdam and Stockholm Conventions (UNEP, 1998, 2001), constitute an important achievement in preventing and controlling soil pollution, but more information is needed regarding their specific interactions with soil components, their mobility in the soil matrix and possible plant uptake, and their effect on crop production (Arias-Estévez *et al.*, 2008). Especially for low and middle income countries not every single compound within the great variability of pesticides available in the market has been analyzed for its ecotoxicological effects before authorization (Aktar, Sengupta and Chowdhury, 2009). For example pesticides that have been taken off the market in High Income Countries (HICs) due to their severe adverse effects on human health and the environment frequently remain registered in Low and Middle Income Countries (LMICs). As pesticides residues can be found throughout the entire ecosystem, pesticide monitoring programmes about the level of residues in soils, surface and groundwater as well as and drinking water but particularly in food items are very important. However, in many low and middle income countries monitoring programmes are inexistent due to their scarcity of regulation capacity (Brodesser *et al.*, 2006).

2.2.5 | MANURE

Application of untreated manure may lead to heavy metal pollution, which not only results in adverse effects on various parameters relating to plant quality and yield, but also causes changes in the size, composition and activity of the microbial community (Yao, Xu and Huang, 2003) affecting nutrient cycling and reducing nutrient availability.

As previously discussed, a high proportion of antibiotics given to livestock is poorly assimilated in the animals' guts and is excreted in urine and faeces. Untreated manure can thus contain high amounts of veterinary antibiotics (VA) that can lead to a rapid increase in antibiotic resistance in soils (see Section 2.3.2). The fate and effects of antibiotics in soils have gained great attention in the last few years, motivated in part by the results of the O'Neill commission report (O'Neill, 2014), which estimates that antimicrobial resistant infections may become the leading cause of death in the world by 2050.

Most common intestinal pathogens that enter the soil with manure and faeces are *Salmonella*, *Campylobacter*, and *Escherichia coli* viruses. The pathogen levels decrease with time and with high temperatures that are reached during storage before land application (Garcia *et al.*, 2010). Once spread on the soil, pathogens can survive for several months or years.

2.2.6 | URBAN WASTES IN AGRICULTURE

Considering that the positive effects of sewage sludge amendment – such as waste reduction, nutrient cycling, increase of soil fertility, improvement of soil structure and water holding capacity – are significantly more important than the negative effects, efforts should focus on reducing the content of pollutants in sewage sludge and wastewaters used for irrigation. As highlighted by Petrie *et al.,* the lack of knowledge on the fate of emerging pollutants and other pollutants present in wastewater and sewage sludge can be solved only by analyzing them before land application (Petrie, Barden and Kasprzyk-Hordern, 2015).

Composting and pretreatments reduce the content of contaminants and pathogen organisms present in urban waste before their application as amendments in soils, and provide an economical and environmentally friendly approach for stabilizing animal waste and converting it into a worthy organic fertilizer. Frequently, however, high levels of heavy metals such as Pb, Cd, Cu, Zn, Cr, Ni, and salts remain in the amendments and may affect soil properties and inhibit plant growth (Bolan *et al.,* 2014; Hargreaves, Adl and Warman, 2008; Stasinos and Zabetakis, 2013; Stratton, Barker and Rechcigl, 1995). The heterogeneous composition of biosolids produced in different wastewater treatment plants requires chemical and biological investigation prior to soil application or incorporation (Bauman-Kaszubska and Sikorski, 2009; Bien, Neczaj and Milczarek, 2013). Limited bioavailability and crop uptake of metals from composted biosolids in comparison with untreated sewage sludge demonstrates the need for pretreatment before its application to soils (Smith, 2009).

2.3 | HUMAN HEALTH RISKS ASSOCIATED WITH SOIL POLLUTION

Oliver and Gregory summarise six soil-related human health risks (Oliver and Gregory, 2015). Of these, three are related to soil pollution: risks from elemental contamination (e.g. As, Cd, Pb); organic chemical contamination (e.g. PCBs, PAHs, POPs); and pharmaceutical contamination (e.g. estrogen, antibiotics). The three other risks are from soil pathogens such as anthrax and prions, micronutrient deficiencies, and under-nutrition due to degraded soils.

Long-term impacts of soil pollution on human health and the environment are still unclear, and many efforts are underway to better understand the mechanisms involved in natural attenuation and the health impacts of toxic pollutants (Bernhardt and Gysi, 2016). Urban soils deserve special attention because anthropogenic activities are concentrated on those soils, and the exposure patterns are more complex due to interactions with other health determinants such as nutrition, air quality, and access to health services for illness prevention (WHO, 2013). However, non-urban areas are also subjected to many different sources of pollution, frequently from diffuse sources, which makes it difficult to trace and to estimate their extent and risk. Future efforts related to soil pollution control and remediation should include these areas in their risk assessment approaches.

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2.3.1 | PATHWAYS OF EXPOSURE OF HUMANS TO SOIL POLLUTANTS AND THEIR EFFECTS ON HUMAN HEALTH

The main pollutants related to industrial, mining, urban and agricultural land uses have been widely discussed in the previous chapters. This section will focus on the soil pollutants that are the most relevant to human health and the risks associated with them.

The route of human exposure to a soil contaminant will vary depending on the contaminant itself and on the conditions and activities at a particular site (Shayler, McBride and Harrison, 2009). Generally, people can be exposed to contaminants present in soil through ingestion or through the consumption of plants or animals that have accumulated large amounts of soil pollutants (Khan *et al.*, 2015); through dermal exposure, from using spaces such as parks and gardens (Chaparro Leal, Guney and Zagury, 2018); or by inhaling soil contaminants that have been vapourized (Figure 15). Humans may also be affected as a result of secondary contamination of water supplies and from deposition of air contaminants (Science Communication Unit, University of the West of England., 2013); in some situations, soils play an important role as the source of contaminants in these two processes.

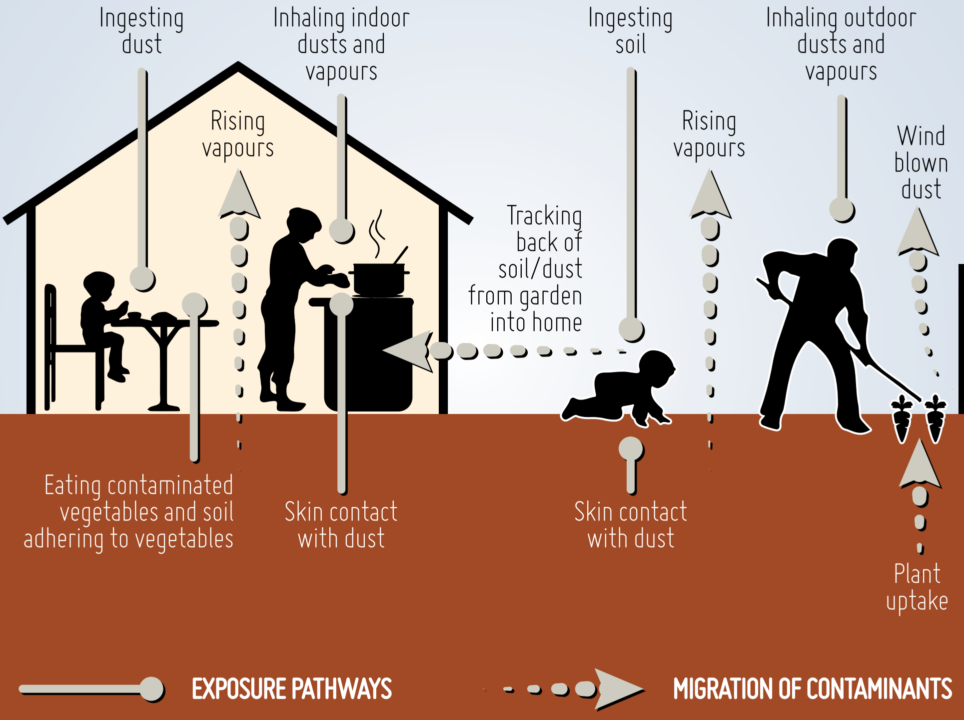

Figure 15. Possible exposure pathways of soil contamination in a residential scenario. Source: EA, 2008

Ten chemicals or groups of chemicals of major public health concern have been identified under the WHO's International Programme on Chemical Safety (WHO, 2010). The ten chemicals or groups of chemicals include soil pollutants such as, Cd, Pb and Hg; dioxin and dioxin-like substances and highly hazardous pesticides (HHP) - whose residues are transferred from contaminated soils to food and water bodies. HHPs are defined as those pesticides that are acknowledged to present particularly high levels of acute or chronic hazards to health or environment according to internationally accepted classification systems or their listing in relevant binding international agreements or conventions. In addition, pesticides that appear to cause severe or irreversible harm to health or the environment under conditions of use in a country may be considered to be and treated as highly hazardous (FAO and WHO, 2016). All of them have chronic effects due to long-term exposure and children, pregnant women and malnourished people are particularly vulnerable to pesticide exposure. Pathogens present in soil may also contaminate food, posing risks to human health. More than 200 diseases – ranging from diarrhea to cancers – are related to contaminated food intake (WHO, 2017b) and 24 percent of the world's population suffers from infections by soil-transmitted helminth, causing nutritional imbalance and chronic anemia (WHO, 2017a).

The uptake of metals by plants from soils may result in a significant risk to health (Brevik, 2013; Burgess, 2013; Jordão *et al.*, 2006). The absorption by plant roots is one of the main routes of entrance of heavy metals into the food chain, and varies according to the level of consumption (Pan *et al.*, 2010; Wagner, 1993). Cadmium and lead are the most toxic elements for man (Volpe *et al.*, 2009). Food is the main source of Cd intake by humans. A well-known case was in Japan, where ingestion of rice contaminated with Cd generated the disease known as itaí-itaí (Abrahams, 2002). Cadmium absorbed via food intake can penetrate through the placenta during pregnancy, damaging membranes and DNA and disrupting the endocrine systems, and can induce kidney, liver and bone damage (Brzóska and Moniuszko-Jakoniuk, 2005; Souza Arroyo *et al.*, 2012). The toxic effects of Pb affect several organs, causing biochemical imbalance in the liver, kidneys, spleen and lungs, and causing neurotoxicity, mainly in infants and children (Guerra *et al.*, 2012; Jaishankar *et al.*, 2014). Organomercuric compounds, especially methylmercury, are considered highly toxic. Mercury may induce changes in human neural and gastric systems and can lead to death. Arsenic is absorbed in the body orally or inhaled and is stored mainly in the liver, kidneys, heart and lungs, with smaller amounts accumulating in muscle and nerve tissue, and has been defined as carcinogenic (Brevik, 2013). It can lead to nervous systems disorders, liver and kidney failure as well as anemia and skin cancer. Nickel causes gastric, liver, and kidney defects and neurological effects (Brevik, 2013). Zinc is associated with anemia and tissue lesions, and while the negative effects of copper are rare, liver and kidney damage in infants is possible if exposure is prolonged (Brevik, 2013).

An increasing awareness in terms of the importance of vegetables and fruits to human diet and the identification of food as the main source for many contaminants suggest that the monitoring of heavy metals in food crops should be carried out frequently. The World Health Organization and FAO developed the Codex Alimentarius (WHO and FAO, 1995), which identifies safe limits for contaminants present in fruits, vegetables, fish and fishery products, and animal feed.

Aktar, Sengupta and Chowdhury presented a review of pesticide residues in food commodities in the European Union (Aktar, Sengupta and Chowdhury, 2009). Even though the amount of pesticides residues in food did not exceed the acceptable daily intake (ADI), few studies have analyzed the long term risk associated with these persistent pollutants in organisms (Kim, Kabir and Jahan, 2017; Xu *et al.,* 2017). Hernández *et al.* highlighted the need for further studies of pesticide mixture effects on human health, because current legislation considers maximum residue levels (MRL) of individual pesticides in food and water, without taking into account possible synergetic interactions at their low concentrations (Hernández *et al.,* 2013). Occupational exposure to pesticides is associated with various diseases including cancer, hormone disruption, asthma, allergies, and hypersensitivity (Burgess, 2013; Van Maele-Fabry *et al.,* 2010).

Intake of persistent organic pollutants accumulated in soils has a high relevance for human health (Figure 16). The last results of the WHO/UNEP global monitoring plan (GMP) show that levels of PCDDs, PCDFs and PCBs in human milk are still significantly above those considered toxicologically safe in many regions of the world, with a higher incidence in India and in some European and African countries (van den Berg *et al.,* 2017). Ingestion of soils (geophagia) has been a common practice in many African and South American countries (Woywodt and Kiss, 2002) among children and pregnant and breast-feeding women, and its practice has extended to western societies (Reeuwijk *et al.,* 2013). The intake of contaminated clay with POPs and heavy metals is a soil-borne source of diseases, as daily exposure levels are frequently exceeded (Odongo, Moturi and Mbuthia, 2016). Bányiova *et al.* pointed out that the main source of POP exposure in the Czech Republic is through intake of polluted food (Bányiová *et al.,* 2017). Even though POP levels in human bodies have been reduced since the introduction of the Stockholm Convention, accidents still occur and they are an important source of soil and food contamination (Hilscherova *et al.,* 2007). The presence of POPs in human milk represents a high risk to the health of newborns and fetuses, as the POPs are circulating in the mother's body (Reeuwijk *et al.,* 2013).

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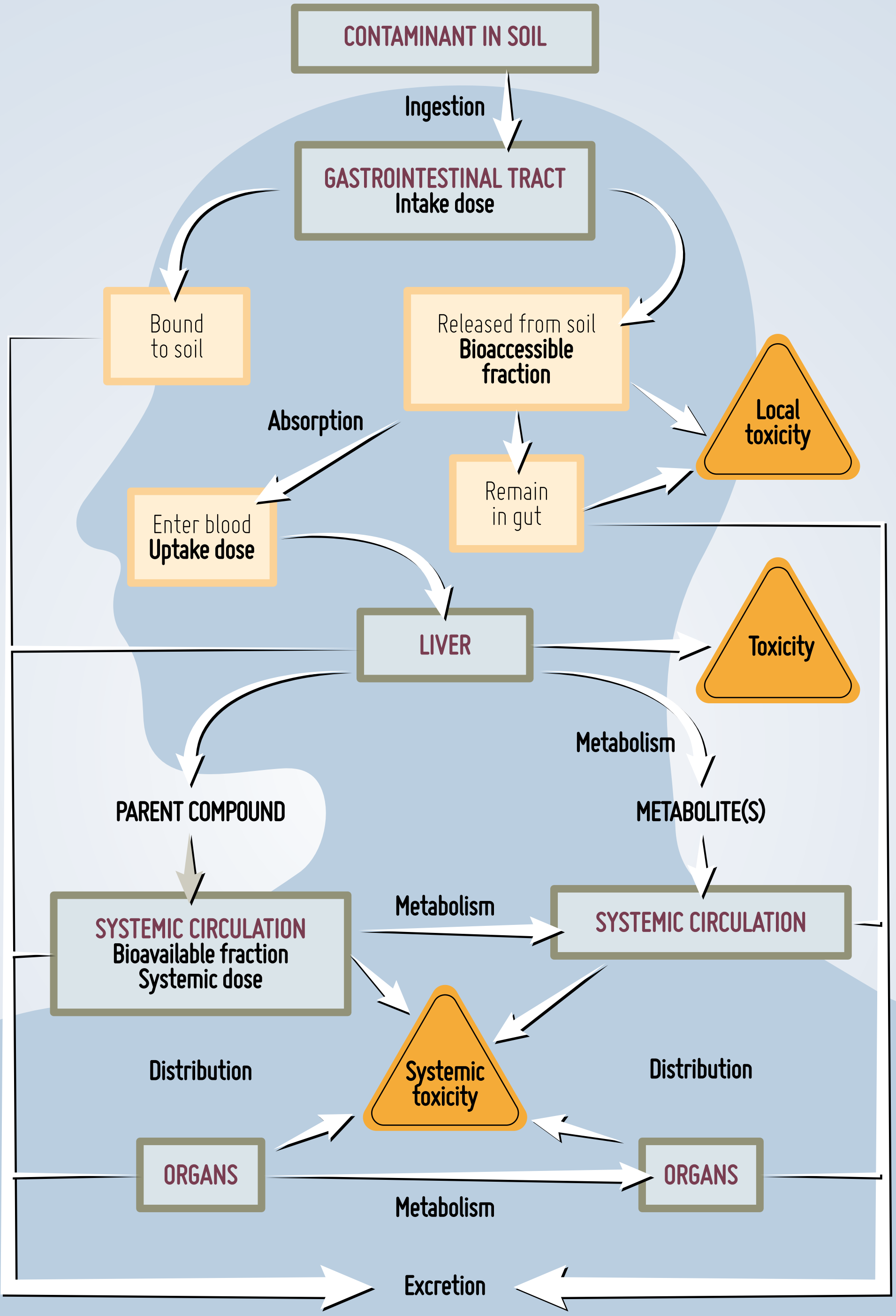

Figure 16. Simplified pathway for oral exposure to soil pollutants. Source: Hosford, 2008

The major exposure route for polycyclic PAHs is through contaminated food ingestion and they are a suspected carcinogenic risk (Brody *et al.*, 2007; Xia *et al.*, 2010). Due to the aromatic nature of PAHs, they easily penetrate cellular membranes and bind covalently with DNA molecules, where they may cause mutations (Muñoz and Albores, 2011). The establishment of a health risk assessment of PAHs would be complex because of the many uncertainties related to exposure and toxicity, which are still unsolved.

Regarding emerging pollutants, there are gaps in our understanding of how they behave in the environment, which interactions occur within the soil matrix, and their toxicity, bioaccumulation properties and transport mechanisms in human bodies, despite the already available information on exposure routes and levels in human tissues (Covaci *et al.*, 2011). Normally, these compounds appear at very low concentrations in human bodies, but the fact that many of them have only recently emerged as contaminants means that long-term studies that focus on the epidemiological aspects of this issue are needed.

Comprehensive assessments of the health effects of most forms of soil, heavy metals, and chemical pollution have not yet been published (Landrigan *et al.*, 2018). Basic toxicological data and knowledge on exposure routes and rates are needed in order to analyze the effects of soil pollutants on human health. A risk assessment based on a toxicological approach that considers tolerable doses is frequently used to establish hazard-related doses (Blume *et al.*, 2016; WHO, 2013). These acceptable doses are characterized for examining the harmful effects of individual substances on a person's health considering a conceptual exposure scenario that identifies sources, pathways and receptors.

As described in the WHO's report (WHO, 2013), there are several scientific tools that can be used to evaluate the risk posed to human health by soil pollution. Other models recently developed to assess the human exposure include BROWSE (Bystanders, Residents, Operators and WorkerS Exposure models for plant protection products) which contains more realistic scenarios (Butler Ellis *et al.*, 2017) and integrates large European guidance and regulatory databases to refine the assessment of human exposure (Lammoglia *et al.*, 2017). The real health risk assessment has not yet been well defined in the case of simultaneous exposure to two or more chemical substances, which occurs in real-life conditions and may have synergistic effects (Nicolopoulou-Stamati *et al.*, 2016).

2.3.2 | SOILS AS RESERVOIR OF ANTIMICROBIAL RESISTANT BACTERIA AND GENES

The transference of antibiotic resistance genes from the environment to human pathogens has created a great challenge due to an overall decrease in effectivity of antibiotics (Harbarth *et al.*, 2015; Thomas and Nielsen, 2005; WHO, 2018). As a result, infections persist in the body, increasing the risk of contamination of others (WHO, 2018). Each year approximately 700 000 deaths occur globally that are attributable to AMR bacteria; 25 000 deaths in Europe (EC, 2017) and around 23 000 in the United States of America (CDC, 2013). Furthermore, human health implications for intake of antibiotic residues and AMR bacteria present in food are largely unknown, although several potential adverse impacts have been observed.

These include allergic and toxic reactions or chronic toxic effects as a result of prolonged low-level exposure (McManus *et al.*, 2002; Sarmah, Meyer and Boxall, 2006). The risks of AMR are especially important in newborns, where AMR bacteria populate the newborns' guts (Brinkac *et al.*, 2017).

Soil is considered to represent a natural reservoir of antibiotic-resistant bacteria carrying a diverse set of known and unknown resistance determinants (Cytryn, 2013). Fungi and bacteria that occur naturally in the environment produce many antibiotics that humans have been using for centuries, and at the same time they have antibiotic-resistant genes against the antibiotics they produce (Hopwood, 2007). Allochthonous resistant bacteria and genes added to soil with manure or sewage sludge might not be well adapted to soil conditions as they are subject to selective pressure by native organisms (Heuer *et al.*, 2008).

When microorganisms (such as bacteria, fungi, viruses and parasites) are continuously exposed to antibiotics or another antimicrobial agent that kills or inhibits the growth of microorganisms, selection of resistant organisms occurs, even at low concentrations. As well, transference of the resistome (extrachromosomal antibiotic-resistant plasmids) or of the mutated genes inside the chromosome to other members of their own species and to other species occurs (Khachatourians, 1998).

Antibiotics are used worldwide for animal therapy and growth promotion in livestock production. A high percentage of these antibiotics are not assimilated by organisms' systems and are excreted into the environment. Assimilation rates by livestock depend on the pharmacokinetics and transformation of the antibiotics by the animal's metabolism. Heuer *et al.*, for example, found that more than 96 percent of the veterinary antibiotic sulfadiazine was excreted in its parental form or as metabolites ten days after administration to pigs (Heuer *et al.*, 2008). Excretion rates are lower for tetracyclines (Winckler and Grafe, 2001), but can reach high rates, over 90 percent for amoxicillin and difloxacine (Sukul *et al.*, 2009). Large amounts of these antibiotics end up in farm and urban wastes and soils, and they are not completely removed in wastewater treatment plants or during composting processes. Wastewater treatment plants were identified as the main source of antibiotic release into streams, with highly variable removal rates for different antibiotics (Michael *et al.*, 2013; Watkinson *et al.*, 2009).

Changes in soil microorganism communities and in crucial activities have been observed after the application of amendments to soil, promoting resistant populations (Ding *et al.*, 2014; Tian *et al.*, 2015; Tien *et al.*, 2017). Antimicrobial-resistant populations in manure may be responsible for the horizontal transference of resistant plasmid to soil-dwelling organisms, as confirmed under field conditions in manure-amended soils (Gotz and Smalla, 1997; Smit *et al.*, 1991). This transference process is promoted by the increase of nutrient sources, which activate microbial activity and population density (Ding *et al.*, 2014). Scientific evidence suggests that the presence of heavy metals, mainly Cu and Zn, in soils contributes to the co-selection of AMR (Grass, Rensing and Solioz, 2011; Hölzel *et al.*, 2012; Wales and Davies, 2015; Yu *et al.*, 2017). Mutations induced in soil microorganisms may lead to a range of metabolic phenotypes, including variations in the ability to use different carbon, nitrogen or phosphate sources (Perkins and Nicholson, 2008), altering global geochemical cycles (Allen *et al.*, 2010).

Wales and Davis also found that antimicrobial selection was enhanced by heavy metals even in the absence of antimicrobial substances when they were exposed to other biocides, such as disinfectants and antiseptics (Wales and Davies, 2015). Similar results were found in residential soils of Australia, where the high content of heavy metals stimulated the proliferation of antimicrobial-resistant genes (Knapp *et al.*, 2017). Thus, AMR becomes an even greater challenge in polluted soils. More research is needed to assess the risk of AMR spreading via sewage sludge and manure amendments (Bondarczuk, Markowicz and Piotrowska-Seget, 2016).

Antibiotic concentrations decrease rapidly after their entry in the soil through different processes (Figure 17), such as plant uptake, leaching to groundwater, sequestration in organic complexes, sorption to clay minerals or biodegradation (Jechalke *et al.*, 2014; Kuppusamy *et al.*, 2018). Despite this, an increase in resistant bacteria was observed even when antibiotic concentrations were very low (Gullberg *et al.*, 2011). Sequestration and sorption of antibiotics in soils reduce their bioavailability but increase their permanence and persistence in the environment (Jechalke *et al.*, 2014). The uptake of antibiotics by plants has been widely reported (Boxall *et al.*, 2006), and has gained attention as the food chain may be directly contributing to the spread of antibiotic resistance (Du and Liu, 2012). Furthermore, antibiotics may cause inhibition of seed germination and reduce crop growth (Du and Liu, 2012). If antibiotic-rich soils suffer any alteration that leads to changes in organic matter concentration and conformation (Gulkowska *et al.*, 2013), the sequestered antibiotics may be released in their bioavailable forms (Rosendahl *et al.*, 2011).


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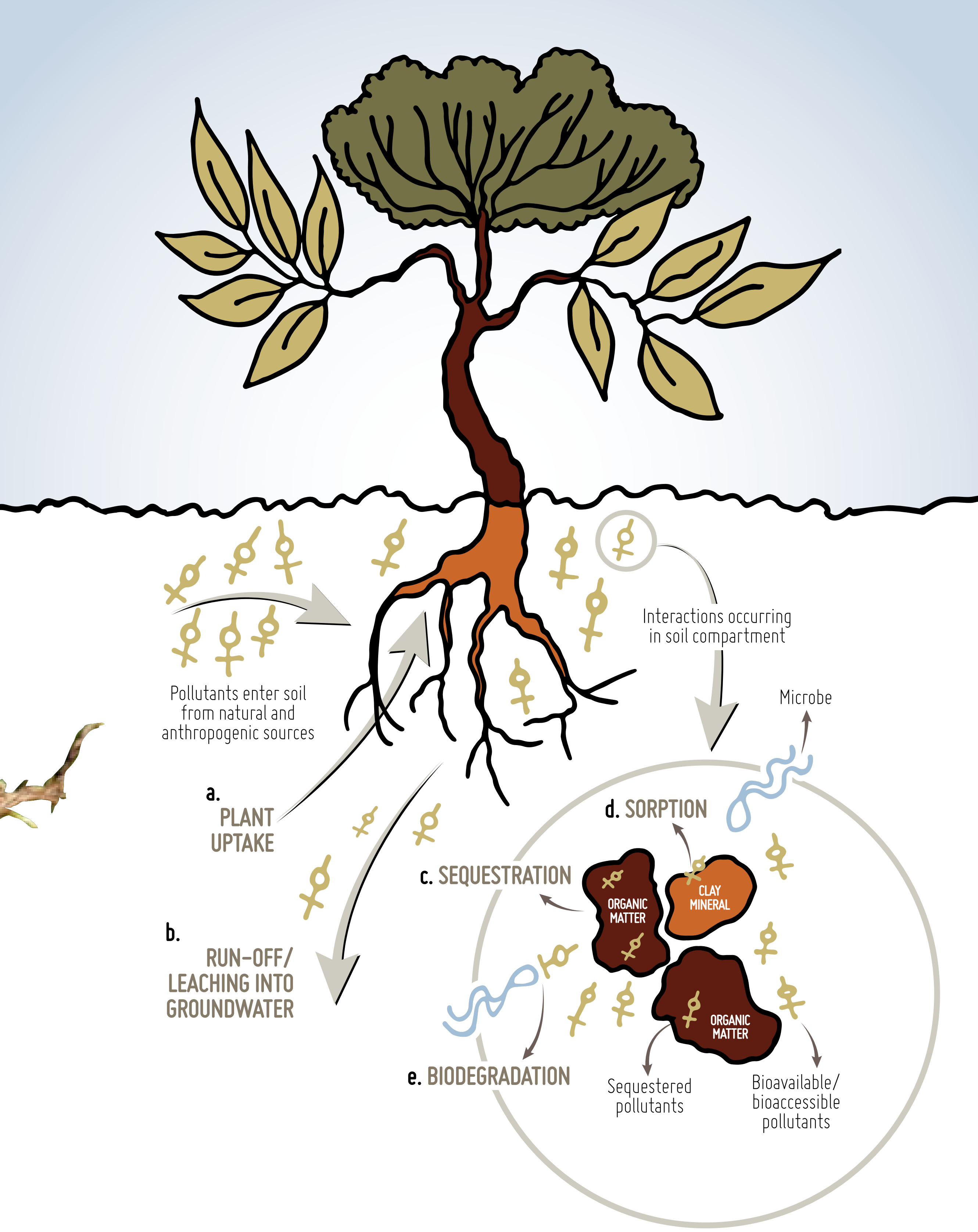

Figure 17. Fate of veterinary antibiotics in the environment. Source: Kuppusamy *et al.*, 2018

Different antibiotics have different target sites within cells; thus, organisms develop resistance to particular antibiotics and not a general resistance (Khachatourians, 1998). However, it has become more and more common to find multi-drug-resistant bacteria (CDC, 2013; EC, 2017; WHO, 2014). A recent study has shown a high co-presence of antibiotic-resistant genes and mobile genetic elements in a multi-drug-resistant bacteria community isolated from chicken manure (Yang *et al.*, 2017b). Bacteria present four main mechanisms that lead to antimicrobial resistance (Figure 18): 1) enzymatic degradation or modification of antibiotic compounds by intra and extracellular enzymes; 2) efflux pumps that actively remove antibiotic compounds outside the cell or in the periplasm; 3) modification or protection of the antibiotic binding site; and 4) natural or modified membrane permeability (Alekshun and Levy, 2007).

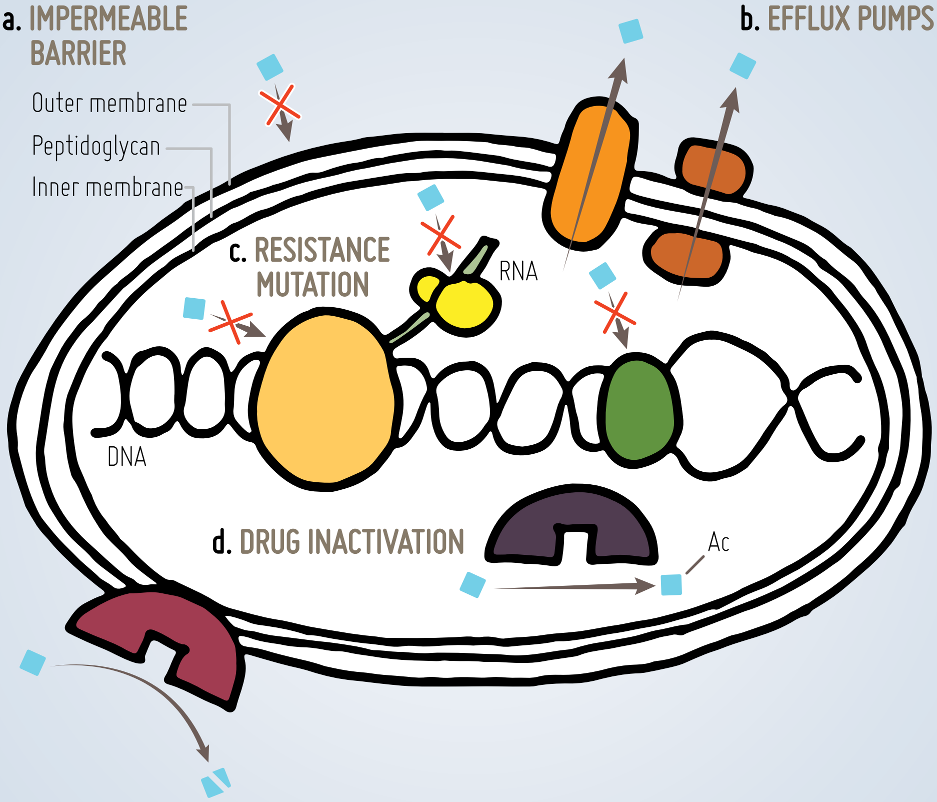

Figure 18. Main mechanisms of antimicrobial-resistance. Source: Allen *et al.*, 2010

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3 | MANAGEMENT AND REMEDIATION OF POLLUTED SOILS

The first step in the assessment and management of polluted soils is the identification of the problem; in this case, the pollutions in the soil. In general, when an area is affected by an accident such as an oil spill, a nuclear accident, or the rupture of a dam tailing, measures to control the extent and prevent further occurrences generally start immediately. However, in legacy polluted soils or where diffuse pollution could be an issue, there are often no established protocols to be followed. In some countries or regions in the world, there are national, regional or local agencies who are responsible for initiating a preliminary investigation to determine whether or not pollution is present and whether further action is needed, while there are many others where no regulation or protocols have been defined (Teh *et al.*, 2016).

In the past, criteria for land reclamation were established using standards based on background concentration and safe limits. New approaches try to adopt a more comprehensive assessment of the risk that pollutants pose to the environment, humans and food safety. The characterization of the potential risk to the environment and human health is not an easy task, due to the complexity of the matrix, the lack of knowledge on the fate of contaminants in soil and the scarcely available information of toxicological and integrated studies (Cachada *et al.*, 2016). Exposure routes for these compartments modelled taking into consideration certain land-use types (e.g. residential, industrial, and recreational) (Provoost, Cornelis and Swartjes, 2006).

3.1 | RISK ASSESSMENT APPROACHES

Assessing risks means that, based on scientific evidence, one can estimate the likelihood of a certain outcome and the gravity of that outcome, and use this knowledge to help in decision making. Uncertainties must be reduced when possible, and clearly the remaining uncertainties need to be clearly identified and explained (FAO, 2000). Risk management decisions for soils or sediments focus on identifying relevant pathways of exposure that pose a risk to human health or the environment and developing appropriate remedial measures. These could include treating or removing sources, or cutting off pathways, or both (Committee on Bioavailability of Contaminants in Soils and Sediments, 2002).

Risk assessment approaches (RAA) are similar worldwide and consist of a series of steps to be taken to identify and evaluate whether exogenous or indigenous substances have caused or are causing soil pollution, and to what extent that pollution is posing a risk to the environment and to human health (Figure 19). Risk assessment approaches are tools to enable science-based political and technical decisions and to take action when needed. Risk assessment tools often use a chemical-by-chemical approach, focusing on a single medium, a single source, and a single toxic endpoint, although integrated approaches are gaining popularity. Such approaches use models combining human exposure and effect-based environmental parameters, based on deterministic or probabilistic techniques (DEA, 2010; Hope, 2006; Provoost, Cornelis and Swartjes, 2006). The end user is interested in whether the soil is "fit for use," mainly in industrial and urban sites where local and diffuse pollution may be present. In these cases, a site-specific approach is necessary to obtain an integrated overview of exposure and risk information (Posthuma *et al.*, 2008) .

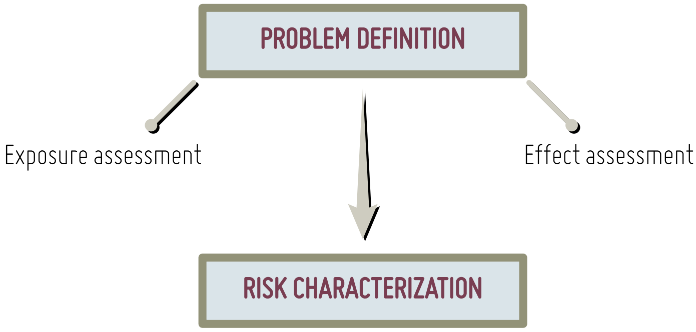

Figure 19. The "universal risk assessment paradigm". Source: Posthuma *et al.*, 2008

Once there is a suspicion of pollution, and after preliminary research on the historical use of the site, an initial assessment should be carried out to define whether exogenous substances are present, which ones are present and whether they pose any risk to the environment and human health. If pollution is confirmed and remediation measures are necessary, a detailed investigation must be accomplished to determine the extent and possible remediation measures. Risk management and/or remediation strategies are subsequently defined and implemented. After-clean-up measures are essential to confirm that the risk has been reduced and that the source of pollution has been controlled.

Worldwide, policies and regulation are based on RAA to forecast risks that cannot be directly measured (Hough, 2007). Regulations include guidelines to identify and assess soil pollution using soil quality standards, in many cases considering national characteristic of soils or site-specific conditions. Because RAA are complex and time-consuming processes, however, not every country in the world can afford to investigate pollution. This is also because no comprehensive information is available, and approaches on a site basis are frequently adopted. As Hope has pointed out, accessing documentation about ecological risk assessment and its regulatory uses is complex, especially in developing countries (Hope, 2006). In those cases, the United States Environmental Protection Agency (US EPA, 1986), Canadian guidelines (Canadian Council of Ministers of the Environment, 1999), and Netherlands guidelines (Brand, Otte and Lijzen, 2007) among others may be used as a reference, even though the characteristics of climate, soil or the local populations are not the same (Li *et al.*, 2014). Some international efforts, such as the one proposed by FAO (FAO, 2000), which provides guidelines to assess the environmental and human health risk posed by stock of obsolete pesticides with more detailed information on the steps of assessment in Environmental Management ToolKit (Volume5), or the guidelines for Integrated Risk Assessment developed by several international organizations (IAEA, 1998; Meek *et al.*, 2011; WHO, 2001a) are attempts to provide an integrated multichemical, multimedia, multiroute, and multispecies exposures analysis.

It is widely recognized that an integrative approach that includes complex mixtures of pollutants is needed to develop more precise risk assessment tools and a better understanding of the potential impacts and their extent (Reeves *et al.*, 2001). Albert launched the question "Is it possible to predict toxicity of complex mixtures?" more than 30 years ago (Albert, 1987). Since then, many researchers have tried to come up with a suitable solution or at least a more comprehensive study of interactions in complex mixtures, to determine whether additive, synergistic or antagonistic toxic effects occur when pollutant mixtures are present (Chen *et al.*, 2015). The specificity and great variability of pollutant mixtures present in each site, which depend on industrial operations or processes carried out, slow down the progress on the definition of limit values appropriate for a general risk assessment approach (Callahan and Sexton, 2007). The Dutch approach, among others, includes a protocol to analyze the risk when more than one substance is present (Cachada *et al.*, 2016). Normally, a cumulative calculation is used, considering the individual risk and the sum across the potential toxicity and risk, but it does not consider possible interactions and synergies between substances that may attenuate or increase their potential risk (Callahan and Sexton, 2007). Chen *et al.* found that the more complex the mixtures of pollutants, the greater the synergistic toxicity (Chen *et al.*, 2015). They suggest that the use of a Combination Index (CI) is more accurate to estimate the ecotoxicological risk than the conventional concentration addition (CA) or independent action (IA) models (Figure 20), not only in aquatic environments (Rosal *et al.*, 2010) but also in soils (González-Naranjo and Boltes, 2014; González-Naranjo *et al.*, 2015). The synergistic/antagonistic effect has been confirmed not only for a combination of pesticides (Yang *et al.*, 2017a) but also in other complex mixtures, such as the pollutant mixture found in landfills (Baderna *et al.*, 2011) or in railway tracks. In the latter case, Wierzbicka *et al.* found highly toxic effects of the pollutant mixture on numerous test organisms from different trophic levels, even though the single concentration of each pollutant did not exceed admissible values (Wierzbicka, Bemowska-Kałabun and Gworek, 2015). However, as explained in Sarigiannis and Hansen, combined toxicology approaches have limited applicability under specific conditions, and data cannot be generalized (Sarigiannis and Hansen, 2012).

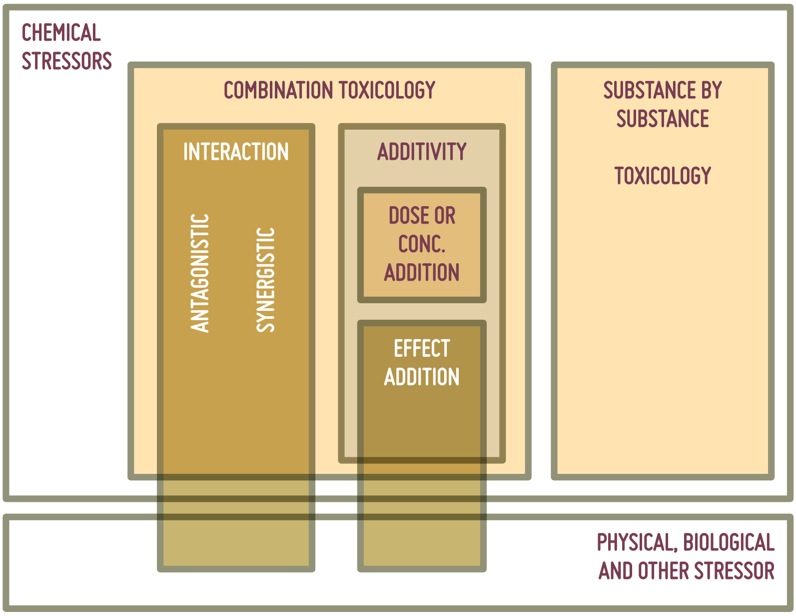

Figure 20. Risk assessment approaches based on independent action of substance by substance, additivity of doses and effects or considering interactions for the combined model. Source: Sarigiannis and Hansen, 2012

The sequence of steps to deal with polluted sites described above is a general one, and depending on national or regional approaches some steps may be omitted or others may be added (Contaminated Sites Management Working Group, 1999; DEA, 2010; FOEN, 2013; Luque, 2014).

Human health risk assessment (HHRA) can be conducted in different ways and for the purpose of meeting different objectives. This approach can be used for the following:

- derivation of soil quality standards
- site-specific risk assessment
- development of remediation objectives
- ranking of contaminated sites by priority of intervention.

Soil screening values (SSVs) are generic soil quality standards based on generic exposure pathways and scenarios (e.g. inhalation of vapours in residential or industrial areas) adopted in many countries to regulate the management of polluted soils. Soil screening values or soil quality standards are identified by different terms around the world: trigger values, reference values, target values, intervention values, cleanup values, cut-off values and others (Carlon *et al.,* 2007; Swartjes *et al.,* 2012). Furthermore, the threshold values are based on different national strategies in environmental policies and rarely take soil properties into account.

In cases of soil pollution by heavy metals, total metal concentration provides little information on the potential risk (Naidu *et al.,* 2015). It is important to identify the available and unavailable forms of the heavy metals to ensure that the soil is managed in such a way as to prevent the unavailable forms from becoming available. This can be done by using biological tests to determine the bioavailability and toxicity of metal(loid)s (Romero-Freire, Martin Peinado and van Gestel, 2015). In this case, soil quality standards or threshold values must be corrected, taking into account soil properties such as pH, soil texture and organic matter content, because it has been widely demonstrated that in many cases quality standards that do not consider soil properties under- or overestimate the actual risk (Appel and Ma, 2002; Bradl, 2004; Rodrigues *et al.,* 2012; Romero-Freire, Martin Peinado and van Gestel, 2015). In addition, by analyzing and including bioavailability during risk assessment instead of assuming that the target pollutants are 100 percent bioavailable, remediation efforts will be optimized and enhance profitability of the remediation efforts (Naidu *et al.,* 2015; Romero-Freire, Martin Peinado and van Gestel, 2015).

It is therefore crucial to develop regulations and legislation to certify the quality of food depending on its heavy metal content. The international literature contains multiple methodologies and evaluation criteria that identify permissible heavy metal values for soils that differ in magnitude (Table 7). This is generally due to the criteria considered for their establishment (Muñiz, 2008). The obtaining of reference values for soil quality in terms of heavy metal content has been established in many countries, which developed their respective environmental policies for soil protection and food safety assurance. The one developed by USEPA (US EPA, 1998, 2014a) is especially important because several other countries follow it. These standards are based on risk assessment policies and define background levels and the study of human and environmental toxicity. When it comes to food, the FAO Codex Standard is of major importance. It defines the values for contaminants and toxins (including heavy metals) permissible in food products, and it is constantly being reviewed and updated (WHO and FAO, 1995).

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Table 7. Threshold values of some heavy metals for residential land-use for various countries. Modified from Provoost, Cornelis and Swartjes, 2006

Contaminant (mg/kg dm)	Belgium[1]	France	Germany[2]	Great Britain + plant[3]	Great Britain – plant[4]	Hungary[5]	Netherlands	Poland[6]
Arsenic	110	37	50	20	20	15	55	2
Cadmium	6	20	20	8[10]	30	1	12	4
Chromium III	300	130[11]	400	130	200	75	380	--
Copper	400	190	N.A.	N.A.	N.A.	30	190	150
Mercury	15	7	20	8	8	0.5	10	--
Lead	700	400	400	450	450	100	530	100
Nickel	470	140	140	50	75	40	210	--
Zinc	1000	9000	N.A.	N.A.	N.A.	200	720	300

1 Soil Remediation Decree named Vlarebo from July 8, 2002

2 Standards applicable as national legislation for 'wirkungspad Boden-Mensch' (exposure path soil — humans)

3 Residential area with vegetable garden

4 Residential area without vegetable garden

5 Hungarian Governmental regulation number 10/2000

6 Polish soil quality standards for the top soil layer (0-30 cm), established for the group B of land use (agricultural lands, forest, residential and recreational areas) Regulation 2002

7 Royal Decree 1310/1990 of 29 October 1990 regulating the use of sewage sludge in agriculture. (B. O. E. No. 262, November 1,1990). Values for soils with pH lower or higher than 7.

8 GUIDELINE ON Investigation Levels for Soil and Groundwater. National Environment Protection (Assessment of Site Contamination) Measure as varied 2011.

9 SEPA (1995) Environmental quality standards for soils. State Environmental Protection Administration, China, GB 15618-1995

10 1/2/8 mg/kg dm related to the soil clean-up standards at pH 6, 7, 8, respectively. The clean-up standard of 8 mg/kg dm was used in this comparison.

11 Chromium total

12 Chromium (VI)

13 1000/4 related to the soil clean-up standard as total concentration and soluble concentration. The clean-up standard of 1000 mg/kg dm was used in this comparison.

14 23/6.1 describes the chlorinated mercury and organic-mercury. The clean-up standard of 23 mg/kg dm was used in this comparison.

15 HIL for lead based on blood lead models (IEUBK for HILs A, B and C and adult lead model for HIL D where 50% oral bioavailability has been considered)

16 1000/0.1 related to the soil clean-up standard as total concentration and soluble concentration. The clean-up standard of 1000 mg/kg dm was used in this comparison.

17 2000/5 related to the soil clean-up standard as total concentration and soluble concentration. The clean-up standard of 2000 mg/kg dm was used in this comparison.

Spain[7]	Sweden	Australia[8] -residential + garden	Australia[8] - residential + no soil access	Canada	China	Norway	Switzerland	U.S.A.
--	15	100	500	12	30[9]	2	N.A.	22
1-3	0.4	20	140	10	0.43	3	20	37
100-150	120	100[12]	500[12]	64	58.9	25	N.A.	100000
50-210	100	7000	30000	63	31.7	100	1000[13]	3100
1-1.5	1	200	600	6.6	--	1	N.A.	23[14]
50-300	80	300[15]	1200[13]	140	37.5	60	1000[16]	400
30-112	35	400	900	50	27.5	50	N.A.	1600
150-450	350	8000	60000	200	117.7	100	2000[17]	23000

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3.2 | MAIN TECHNIQUES FOR REMEDIATING POLLUTED SITES

Nathanail referred to sustainable remediation as "remediation that eliminates and/or controls unacceptable risks in a safe and timely manner, and which maximizes the overall environmental, social and economic benefits of the remediation work" (Nathanail, 2011). Sustainable management requires the incorporation of the best available techniques, not only during the remediation process itself, but for the whole process, including risk assessment and risk reduction. Best management practices (BMPs) are individual or combinations of management, cultural and structural practices that researchers (academic or governmental) have identified as the most effective and economical way of reducing damage to the environment (Cestti, Srivastava and Jung, 2003). Remediation is commonly done on a site-by-site basis, since for every combination of pollutant, soil property, land use, property and liability regimes and technical and economic reality of the site or area, a different technique or combination of techniques may be more appropriate (Swartjes, 2011).

Remediation techniques can be divided in two main groups: *in situ* (on the site) and *ex situ* (removal of contaminated soil for treatment off the site) remediation. Available remediation options include physical, chemical and biological treatments, and these options offer potential technical solutions to most soil pollution (Scullion, 2006). For both *in situ* and *ex situ*, the net effect on the contaminants can be categorized as reducing the concentration, reducing the bioavailability without reducing the concentration, encapsulating in an inert matrix, containment, and removal (Pierzynski, Sims and Vance, 2005). The management of polluted sites is a site-specific approach that includes characterization, risk assessment and remediation technologies selection, and therefore is mainly focused on local or point-source contamination.

Scullion presented a review of the main treatment approaches to remediate polluted soils and their effect on pollutants (Scullion, 2006), specifying whether they are degraded, separated from soil components, extracted from the matrix or stabilized (Table 8).

Table 8. Main remediation methodologies and their effects on soil pollutants (√ = main process, (√) = subsidiary process limited in extent or in the range of pollutants affected). Source: Scullion, 2006

Process treatment	Destruction/ degradation	Solid separation	Extraction/ loss	Stabilisation
Physical remediation methodologies				
Thermal	√		√	
Solidification	(√)			√
Vapour extraction			√	
Air sparging	(√)		√	
Washing/pump and treat	(√)		√	
Electroremediation	(√)		√	
Particle sorting		√		
Chemical remediation methodologies				
Oxidation	√		√	√

Process treatment	Destruction/ degradation	Solid separation	Extraction/ loss	Stabilisation
Reduction	(√)		√	√
Hydrolysis	√		√	
Solubilisation	(√)		√	
Dechlorination	(√)			
pH manipulation	(√)		√	√
Biological remediation methodologies				
Microbial activity				
Landfarming	√		(√)	√
Biopiling	√		(√)	√
Composting	√		(√)	√
Bioreactor	√			(√)
Bioleaching			√	
Plant activity				
Phytostabilisation	(√)		(√)	√
Phytoextraction	(√)		√	(√)
Phytodegradation	√		(√)	(√)

What makes many of the currently available physical methods so expensive is partially the cost of excavating and transporting large quantities of contaminated materials for *ex situ* treatment such as chemical inactivation or thermal degradation. The high cost has led to an increasing interest in alternative technologies for *in situ* applications, in particular those based on the biological remediation capability of plants and microorganisms (Chaudhry *et al.*, 2005). Bioremediation is a technology that destroys or renders harmless various contaminants, using the biological activity of certain microorganisms. Bioremediation actually relies on the microbial growth and activity; its effectiveness is highly dependent on the applied environmental parameters that influence the microbial growth and the degradation rate. Bioremediation is considered a very promising technology with great potential when dealing with certain types of contaminated sites (Zouboulis, Moussas and Nriagu, 2011). Bioremediation has been used worldwide, including in Europe, with varying success (Zouboulis, Moussas and Nriagu, 2011).

According to Alexander, several conditions must be satisfied for bioremediation by microbial activity to take place in the soil (Alexander, 1999). These include the following: 1) the organism must be present in the soil containing the pesticide; 2) an organism must have the necessary enzymes to bring about the biodegradation; 3) the pesticide must be accessible to the organism having the requisite enzymes; 4) if the initial enzyme bringing about degradation is extracellular, the bonds acted upon by that enzyme must be exposed for the catalyst to function; 5) should the enzymes catalyzing the initial degradation be intracellular, that molecule must penetrate the surface of the cell to the internal sites where the enzyme acts; and 6) because the population or biomass of bacteria or fungi acting on many synthetic compounds is initially small, conditions in the soil must be conducive to allow proliferation of the potentially active microorganisms.

Compost made from sawdust, wood chips, bark, straw, plant waste and food waste from households is another common source of organic matter to be added to the soil (Kuo *et al.*, 2004). Addition of organic matter to the soil may help to decrease the mobility of heavy metals and other pollutants (Grobelak and Napora, 2015; Wuana and Okieimen, 2011), reducing the risk to the environment and to human health.

The addition of manure and sewage sludge can be an effective bioremediation tool, but care needs to be taken to ensure that effective pre-treatment of the organic material has occurred. To attenuate the negative impacts associated with livestock manure, simple techniques such as composting can be applied before their application to the land (Zhang *et al.*, 2015a). Compared to fresh manure, composted manure generally has higher contents of lignin and polyphenol, which reduces CH_4 emission while further enhancing the potential of SOC sequestration (Xia, Wang and Yan, 2014). Lv *et al.* observed a positive effect of worms present in the composting process, resulting in the stabilization of heavy metals present in animal manure (Lv, Xing and Yang, 2016). The composting of fresh manure has been proven as an effective method for reducing various types of environmental pathogens and antimicrobial resistant bacteria (Cole, 2015; Holman *et al.*, 2016). Storing slurries for one to three months, composting at high temperatures, spreading in a manner that reduces potential volatilization and avoiding long-distance transport of manure are some of the recommendations proposed by Nicholson *et al.* in order to reduce pathogen levels in manure and slurries prior their land application (Nicholson *et al.*, 2003). Despite the observed persistence of certain antibiotics in soil and their negligible mineralization due to strong sorption to soil components, several authors highlight the importance of storage time and composting for dissipation of antibiotic compounds in manure before land application (Arikan, Mulbry and Rice, 2009; Halling-Sørensen *et al.*, 2001; Kim *et al.*, 2011; Tien *et al.*, 2017).

The planting of trees that have good resistance to high levels of toxic substances and a high capacity to collect and store pollutants can also be a good practice for bioremediation process in soils (Paz-Alberto and Sigua, 2013). According to Wisłocka *et al.*, the most popular trees exhibiting a high capacity to accumulate heavy metals are silver birch (*Betula pendula*), alder (*Alnus tenuifolia*), black locust (*Robinia pseudoacacia*), willow (*Salix sp.*), and conifer trees (Wislocka *et al.*, 2006). Selected energy crops such as *Miscanthus giganteus* have excellent adaptability to change habitat conditions, the possibility to gradually reclaim degraded lands, and the ability to prevent the migration of heavy metals into the soil and groundwater.

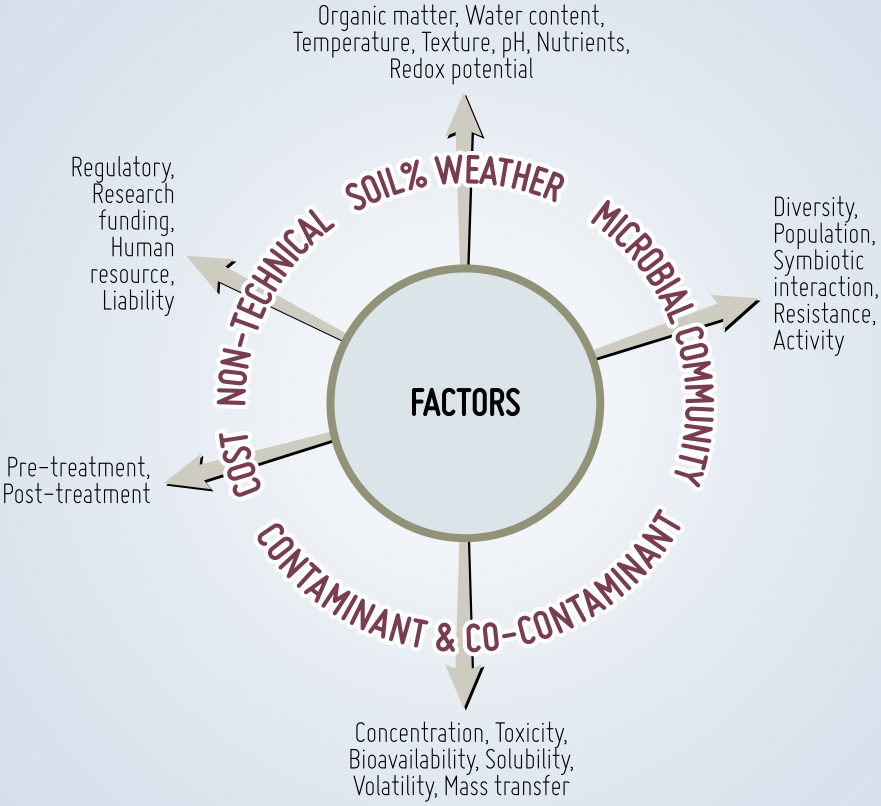


Figure 21. Factors affecting field-scale remediation of PAH-polluted soils. Source: Kuppusamy *et al.*, 2017

Interest in biochar is also growing among scientists, who are particularly interested in how the chemical and physical properties of biochar particles affect water moving through soil, remove pollutants, alter microbial communities and reduce emissions of greenhouse gases. The hope is that biochar can help farmers around the world, particularly those in developing regions who often struggle with poor soils. Biochar has ancient roots. Hundreds to thousands of years ago, residents of the Amazon produced it by heating organic matter to create rich, fertile soils called *terra preta*. The practice was abandoned around the time that European nations invaded South America, and relatively few farmers elsewhere have routinely used biochar. Scientists first took an interest in the material about a decade ago, when growing concerns over global warming led some to tout biochar as a way to store huge amounts of carbon underground. Hope for that application has faded somewhat due to the high cost of biochar, but soil scientists are now exploring its use in agriculture and in remediating soil pollution (Cernansky, 2015).

New technologies for remediation involve the application of nanoparticles for remediating polluted soils (Pan and Xing, 2012). The most widely recognized nanotechnology in soil remediation is the application of nano-zero-valent iron (nZVI) for reducing the impact of both organic and inorganic pollutants. For example, nZVI can effectively degrade chlorinated hydrocarbons and organochlorine pesticides (Singh *et al.,* 2011; Zhanqiang, 2010). Carbon nanotubes have been demonstrated to be a feasible remediation material because of their large sorption capacity for metal ions (Rao, Lu and Su, 2007), radionuclides (Ren *et al.,* 2011) and organic compounds (Pan and Xing, 2008).

Integrated approaches and emerging technologies, such as electrokinetic remediation, enzyme-mediated bioremediation, multi-process phytoremediation and vermiremediation have been employed in the treatment of PAH-contaminated soils (Kuppusamy *et al.,* 2016). The selection of the best available techniques and their success in remediating polluted soils will depend on physical, economical, regulatory and technical factors (Figure 21) (Kuppusamy *et al.,* 2017).

The critical factor affecting remediation of PCBs, PAHs and PBDEs is the strong sorption of these molecules on soil and sediments, as demonstrated by their long persistence despite heavy restrictions on their use for over 30 years. The ability to desorb these contaminants determines, in most cases, the effectiveness of remediation technologies (Gomes, Dias-Ferreira and Ribeiro, 2013). The most commonly used remediation technique for these polluted soils is "dig-and-dump," but this is not sustainable. Other techniques such as bioremediation, thermal desorption, and anaerobic dechlorination have been tested in recent years with good results (Gomes, Dias-Ferreira and Ribeiro, 2013). The technologies previously described, although aiming to destroy or transform PCB, operate in very different ways and consequently have different clean-up times, costs, breakdown products and environmental impacts. Their effectiveness is also site-specific, since each technology depends on the contaminants, the aging of the contamination, the type of soil and geomorphologic conditions, and other environmental factors such as mobility of the contaminants or sorption to soil particles (Gomes, Dias-Ferreira and Ribeiro, 2013; Wang and He, 2013).

3.3 | CHANGES IN AGRONOMIC PRACTICES TO MINIMISE FOOD-CHAIN CONTAMINATION AND IMPACTS ON ECOSYSTEM SERVICES

The Voluntary Guidelines for Sustainable Soil Management (VGSSM) aim to provide countries, farmers and other stakeholders with generally accepted, practically proven and scientifically based principles to promote sustainable soil management (SSM) (FAO, 2017). These guidelines describe SSM as follows: "Soil management is sustainable if the supporting, provisioning, regulating, and cultural services provided by soil are maintained or enhanced without significantly impairing either the soil functions that enable those services or biodiversity." SSM are related to the agronomic practices cited in this chapter. (FAO, 2017. Voluntary Guidelines for Sustainable Soil Management. Food and Agriculture Organization of the United Nations. Rome, Italy).

3.3.1 | FERTILIZERS

Integrated crop management (ICM) is a method of farming that balances the requirements of running a profitable business with responsibility and sensitivity to the environment. It presents a realistic solution to many of the problems facing agriculture. It includes practices that can be used to avoid waste, enhance energy efficiency and minimise pollution. Integrated crop management combines the best of modern technology with some basic principles of good farming practice and is a whole-farm, long-term strategy (EC, 2002).

Components of ICM for field crops are as follows:

1- Quantify nutrient source: soil reserve, manure, crop residue ;

2- Soil test: pH, lime requirement, phosphorus, potassium (calcium and magnesium optional);

3- Manure analysis: nitrogen (ammonium N, total N), phosphorus, potassium;

4- Calibration of manure and fertilizer spreaders: tonnes, 1000's gallons, lbs. per acre;

5- Fertilization plan: manure application rate, supplemental fertilizer; utilize excess manure on alternative crops (hay crops); avoid applying large amounts of manure on fields with excessive P found using soil tests; do not over apply nitrogen from manure or fertilizer, and nitrogen soil test: side- or top-dressing supplemental nitrogen fertilizer;

6- Cover crop: to reduce soil loss and nitrate leaching; consider a legume-based cover crop on vegetable farms and on distant fields on dairy farms where manure is not spread;

7- Planting plan: to ensure early harvest of crops to allow early cover crop planting on most erosion prone fields; and

8- Minimum tillage: to reduce nutrient loss through soil erosion.

Integrated Soil Fertility Management is an approach based on the following principles: 1) Neither practices based solely on mineral fertilizers nor solely on organic matter management are sufficient for sustainable agricultural production; 2) well-adapted, disease- and pest-resistant germplasm is necessary to make efficient use of available nutrients; and 3) good agronomic practices – in terms of planting dates, planting densities, and weeding – are essential for ensuring the efficient use of scarce nutrient resources (CGIAR and CCAFS, 2018). There is also a need to target nutrient resources within crop rotation cycles, preferably including legumes, thus going beyond recommendations for single crops.

Integrated nutrient management can play a role in improving plant growth. Dry matter partitioning and total crop biomass (Amanullah and Inamullah, 2016; Amanullah *et al.*, 2016), including root biomass (Amanullah and Stewart, 2013), have a significant impact on the efficiency of phytoremediation processes of degraded soils (Grobelak, 2016). Maintaining organic carbon-rich soils, restoring and improving degraded agricultural lands and, in general terms, increasing the soil carbon content all play an important role in addressing the three-fold challenge of food security, adaptation of food systems and people to climate change, and the mitigation of anthropogenic emissions (UNFCCC, 2015).

Bio-fertilizers, products containing living cells of different types of beneficial microbes (bacteria, fungi, protozoa, algae and viruses), are known to play a number of vital roles in soil fertility, crop productivity and profitability. Some of the more commonly used beneficial microbes in agriculture include *Rhizobia, Mycorrhizae, Azospirillum, Bacillus, Pseudomonas, Trichoderma, and Streptomyces* species. Beneficial microbes are essential for decomposing organic matter in the soil and for increasing the availability of essential macro-nutrients (nitrogen, phosphorus, potassium, sulfur, calcium and magnesium) and micro-nutrients (boron, copper, chlorine, iron, manganese, molybdenum and zinc) to crop plants. Beneficial microbes also play a significant role in solid waste and sewage management. Beneficial microbes increase plant tolerance to different environmental stresses (e.g. drought, heat, cold, salinity) and increase plant resistance to insects and disease. Beneficial microbes not only improve crop growth and productivity by increasing photosynthesis and producing hormones and enzymes, but also improve crop quality by controlling different insects and various plant diseases. Beneficial microbes reduce the need for the use of chemical fertilizers and thereby reduce environmental pollution caused by chemical fertilizers. They reduce the cost of production and therefore increase the grower's income and profitability. Beneficial microbes are very important in increasing crop productivity, profitability and sustainability. Applications of organic manures such as animal manure, poultry manure, green manure, composts, farm yard manure, biochar, and ash increase the beneficial microbes in the soil and improve soil health and overall sustainability (Amanullah, 2015).

3.3.2 | PESTICIDES

For achieving a pollution-free world, the Voluntary Guidelines for Sustainable Soil Management (FAO, 2017), which include integrated or organic pest management practices, are recommended worldwide.

Integrated pest management (IPM) is an approach based on prevention, monitoring, and control that offers the opportunity to eliminate or drastically reduce the use of pesticides, and thus reduce the risks of pesticide to human health and the environment. Integrated pest management does this by utilizing a variety of methods and techniques, including cultural, biological and structural strategies to control a multitude of pest problems (Beyond Pesticides, 2018). Moreover, IPM encourages the use of crop rotations, which can considerably lower the need for pesticides (García-Préchac *et al.*, 2004).

In intensive agroecosystems, the most common practice of using pesticides is the spray application, although other application systems like seed treatment, granules applied on the ground or soil drenching as well as soil fumigation. Up to 30–50 percent of the amount applied is lost by deposition on the ground, via spray drift to neighboring environmental compartments, or volatilized, not reaching the target pest (Diaconu *et al.*, 2017; Viret *et al.*, 2003). The "polluter pays" principle (adding the environmental and public health costs to the price paid by consumers) can be an effective approach to internalizing the social costs of pesticide use. The fees and taxes generated can be used to promote improved (sustainable) pest management (Popp, Pető and Nagy, 2013). Controlling the misuse of pesticides along with promoting more environmentally-friendly techniques, such as biological pest control (Popp, Pet and Nagy, 2013), can contribute to reducing contamination in agricultural fields.

Integrated weed management (IWM) is the control of weeds through a long-term management approach, using several weed management techniques such as physical control, chemical control, biological control and cultural control.

As noted in earlier sections, the most widespread type of contamination of soils that could adversely affect food quality is related to metals, metalloids and radionuclides. This has contributed to a wealth of studies examining agricultural practices to reduce food-chain contamination by these pollutants. As pollution of soils by organic chemicals is generally more restricted in areal extent, much less research has been conducted on these chemicals and they are not considered further here.

3.3.3 | METALS

Cadmium (Cd) is the most widely studied metal in terms of food-chain contamination, and there are a number of options to minimise plant uptake of Cd from soil (Grant *et al.*, 1999). They are summarised in Table 9 and can be grouped into manipulation of crops (species, cultivar and rotation), of soil conditions and of water (irrigation) attributes.

Table 9. Agronomic management practices to reduce food-chain contamination by Cd.

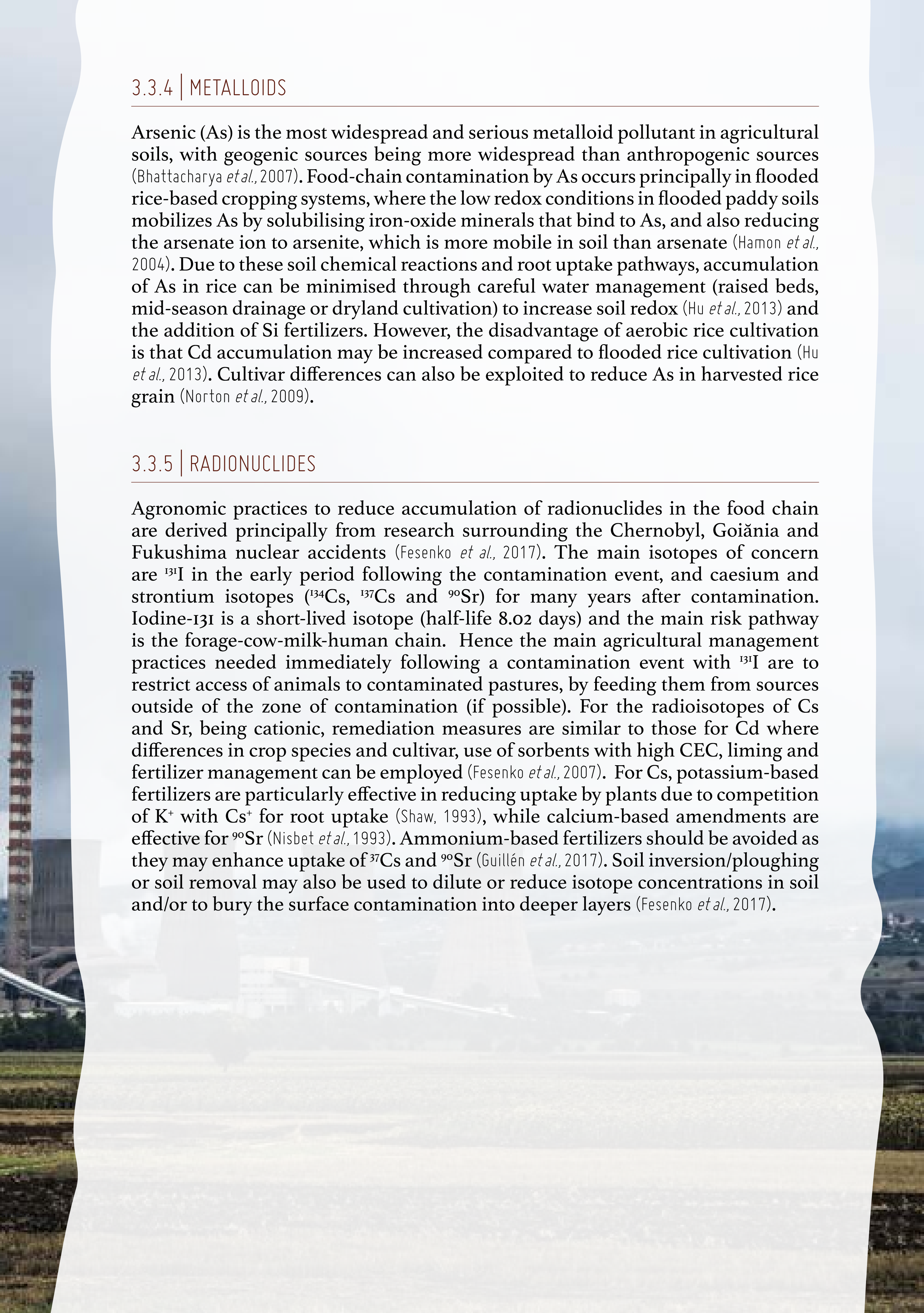
Crop manipulation	Soil manipulation	Water manipulation
Plant species	Site selection	Use low (Cl) salinity water
Plant cultivar	Cultivation (dilution/burial)	
Crop rotation	Lime addition	
Phytoextraction	Zinc addition	
	Sorbent addition	

It has been known for over 40 years that different species vary in their ability to accumulate Cd in edible portions. Leafy vegetables, for example, generally accumulate higher concentrations of Cd than do grain or fruit crops (Bingham *et al.*, 1975; Chaney and Hornick, 1977). Farmers have the option to change the type of crops grown in a specific plot of land if the soil is Cd polluted. If this is not a possibility, it is still possible to grow the same crop species if a lower-accumulating Cd cultivar is chosen. It is well known that different cultivars of the same species accumulate Cd at different rates, and this may be related to different rooting patterns, different root uptake of Cd, or different patterns of Cd translocation within the plant (Grant *et al.*, 2008). Commercialisation of specially bred low-Cd-accumulating cultivars has ensued in some countries (Clarke *et al.*, 1997), while in others, farmers can choose a low-Cd cultivar from the commercially available ones (where this information is accessible). Food-chain contamination by Cd can also be minimised by selecting an appropriate crop rotation plan: there is evidence that certain sequences of crops (e.g. wheat grown after lupin crops) (Oliver *et al.*, 1993) may encourage more Cd accumulation, although the reasons for this are not clear and may be related to the modification of soil chemical or physical conditions (e.g. changes in soil pH). Finally, farmers may also choose to grow a crop to extract available Cd from the soil (phytoextraction) and dispose of the plant material before growing a food crop (Murakami *et al.*, 2009). This strategy is now maturing to the stage of being practically possible (Abe *et al.*, 2017).

Selection or manipulation of soil chemical and physical conditions is also practised by farmers to minimise food-chain accumulation of Cd. Selection of soil conditions is effected through site selection (if possible); soils higher in pH, clay, organic matter, zinc (Zn) and lower in Cd are more likely to have minimal accumulation of Cd in crops (Grant *et al.*, 1999). If site selection is not possible, soil manipulation may be attempted. As Cd is a cationic metal, the addition of lime to raise soil pH and increase the cation-exchange capacity of soil can be used to increase soil sorption and reduce crop uptake, although effects are not consistent in field studies. Acting through similar mechanisms, sorbents can be added to soils to bind Cd more strongly and minimise its uptake by crops (Komárek, Vaněk and Ettler, 2013; Tang *et al.*, 2016), although high application rates are usually needed (tonnes per hectare) and the longevity of the remediation is unknown. The addition of Zn has also been shown to reduce crop Cd concentrations (Oliver *et al.*, 1994) through a competitive uptake of Zn over Cd for loading into edible portions (Welch *et al.*, 1999). Finally, if the Cd contamination is anthropogenic and not geogenic, it is likely that contamination is restricted to the surface soil layer. As for many contaminants, cultivation and burial or dilution of the contaminated layer can reduce Cd uptake by crops, as most crop roots are active only in the top 10–20 cm of soil.

Avoidance of irrigation waters rich in Cl will also reduce food chain contamination by Cd, due to chloro-complexation of the Cd^{2+} ion that increases mobility in soil and hence increases plant Cd uptake (McLaughlin *et al.*, 1994).


Jason Blackeye on Unsplash



3.3.4 | METALLOIDS

Arsenic (As) is the most widespread and serious metalloid pollutant in agricultural soils, with geogenic sources being more widespread than anthropogenic sources (Bhattacharya *et al.*, 2007). Food-chain contamination by As occurs principally in flooded rice-based cropping systems, where the low redox conditions in flooded paddy soils mobilizes As by solubilising iron-oxide minerals that bind to As, and also reducing the arsenate ion to arsenite, which is more mobile in soil than arsenate (Hamon *et al.*, 2004). Due to these soil chemical reactions and root uptake pathways, accumulation of As in rice can be minimised through careful water management (raised beds, mid-season drainage or dryland cultivation) to increase soil redox (Hu *et al.*, 2013) and the addition of Si fertilizers. However, the disadvantage of aerobic rice cultivation is that Cd accumulation may be increased compared to flooded rice cultivation (Hu *et al.*, 2013). Cultivar differences can also be exploited to reduce As in harvested rice grain (Norton *et al.*, 2009).

3.3.5 | RADIONUCLIDES

Agronomic practices to reduce accumulation of radionuclides in the food chain are derived principally from research surrounding the Chernobyl, Goiănia and Fukushima nuclear accidents (Fesenko *et al.*, 2017). The main isotopes of concern are ^{131}I in the early period following the contamination event, and caesium and strontium isotopes (^{134}Cs, ^{137}Cs and ^{90}Sr) for many years after contamination. Iodine-131 is a short-lived isotope (half-life 8.02 days) and the main risk pathway is the forage-cow-milk-human chain. Hence the main agricultural management practices needed immediately following a contamination event with ^{131}I are to restrict access of animals to contaminated pastures, by feeding them from sources outside of the zone of contamination (if possible). For the radioisotopes of Cs and Sr, being cationic, remediation measures are similar to those for Cd where differences in crop species and cultivar, use of sorbents with high CEC, liming and fertilizer management can be employed (Fesenko *et al.*, 2007). For Cs, potassium-based fertilizers are particularly effective in reducing uptake by plants due to competition of K^+ with Cs^+ for root uptake (Shaw, 1993), while calcium-based amendments are effective for ^{90}Sr (Nisbet *et al.*, 1993). Ammonium-based fertilizers should be avoided as they may enhance uptake of ^{37}Cs and ^{90}Sr (Guillén *et al.*, 2017). Soil inversion/ploughing or soil removal may also be used to dilute or reduce isotope concentrations in soil and/or to bury the surface contamination into deeper layers (Fesenko *et al.*, 2017).

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4 | CASE STUDIES ON SOIL POLLUTION AND REMEDIATION

4.1 | REMEDIATION BY ENHANCED NATURAL ATTENUATION OF POL POLLUTED SITES IN UN FIELD MISSIONS: A CASE STUDY ON THE UNITED NATIONS OPERATION IN CÔTE D'IVOIRE (ONUCI)[18]

The consumption of petroleum oil and lubricants (POL) in field missions is inevitable due to their use in generating electricity and operating mechanical equipment to support peacekeeping operations. Through these processes, which have a major environmental footprint, the potential of soil contamination arises. This section presents a case study of remediation work conducted by Global Service Centre/Environmental Technical Support Unit on POL polluted sites during the liquidation of a United Nations field operation in Côte d'Ivoire (ONUCI).

The goal of the project was to reduce the level of total petroleum hydrocarbon (TPH) in polluted soil (36 000 to 75 000 mg/kg) to a background TPH level of 400 to 1 000 mg/kg, providing an enabling environment for revegetation of plants. The project entailed the removal of over 1 200 tonnes of POL contaminated soil from sites and replacing it with fresh soil. The excavated contaminated soil was treated using naturally occurring materials derived locally.

The contaminated soil was deposited in a large concrete mixer to tumble and aerate in order to promote microbial growth and the breaking down of POL. Two ingredients (poultry waste and naturally occurring surface active materials (NOSAM) or palm ash soap (also known as black soap)) were added to the mix to improve the condition of the soil and to accelerate the microbial remediation.

The result showed a reduction of over 95 percent in TPH levels immediately after remediation, with natural microbial activities ensuring more reduction in TPH within a 14-day period. Native grasses were planted in the restored areas. The case study highlights the importance of implementing low cost remediation techniques in mitigating POL polluted sites within the UN field missions.

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[18] Environmental Technical Support Unit (ETSU) (GSC Environmental Technical Support Unit, Apulia, Italy)

4.2 | CONTEMPORARY APPROACHES TO REMEDIATION OF OIL-POLLUTED LANDS IN THE TAIGA ZONE OF WESTERN SIBERIA[19]

The Russian Federation occupies one of the leading places in oil production over the globe. More than 70 percent of Russian Federation oil is extracted in the Taiga zone of western Siberia. In the 1990s oil-production enterprises of this region experienced a drastic increase in pipeline accidents and oil pollution of ecosystems. Under conditions of insufficient state control over statutory compliance of environmental protection legislation, this led to a significant number of oil-polluted lands that have not been remediated for a long time, forming a so-called "historical heritage" for new companies that are currently producing oil on this territory.

Oil companies have made significant efforts to restore oil-polluted lands in the last 10–15 years, but this problem has not been completely resolved. This is mostly due to the special environmental conditions of the region: the average annual temperature ranges from -0.1 °C to -5 °C, the average temperature in January is -18 °C to -24 °C (with the recorded minimum as -62 °C); the duration of the period with a stable snow cover achieves 180–200 days; and precipitation significantly exceeds evaporation. The West Siberian lowland is a vast, weakly dissected plain, which experienced active development of swamp formation during the Holocene epoch: in some areas, swamps cover 90 percent of the territory. Spills therefore occur mainly in wetland ecosystems, which greatly complicates the use of machinery for reclamation operations.

Not only were there unfavourable weather conditions, but remediation technologies were applied that were not appropriate for wetland soils, as they were originally developed for mineral soils. Basic technological solutions included surface oil pickup (if any), agrotechnical practices (liming, mineral fertilization), biostimulation (activation of native oil-oxidizing microorganisms) or bioaugmentation (application of commercial bio-products with oil-oxidizing action), periodic loosening and phyto-melioration (sowing of meadow grasses). However, for remediation of oil-contaminated peat bog soils, some other approaches were needed.

Peat soils have a very high sorption capacity to oil. It is therefore difficult to collect spilled oil even immediately after the spill, and after thickening of the oil it is impossible. At the same time, the concentration of oil hydrocarbons in the upper, most contaminated part of the peat bog soil profile can reach 80 percent or more, which is significantly higher than the levels that oil-destructive microorganisms can consume. The above-described traditional technological solutions are therefore ineffective, even after being repeated for several processing seasons.

Effectiveness of reclamation is significantly increased if mechanical removal (shearing) of the uppermost contaminated layer (usually 10–15 cm) is performed in the oil-contaminated area first. In this layer, in addition to heavy oil hydrocarbons, a large number of resins and asphaltenes accumulate. This accumulation effectively seals the soil, preventing water and gas transfer. This in turn drastically decreases microbiological activity in the contaminated soil. At the initial stage of implementation of this technological operation, manual labour was used. This explains why despite the high efficiency of remediation on certain oil-contaminated sites, the total area of reclaimed land remained low.

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[19] Sergey Trofimov, Ruslan Kinjaev, Olga Yakimenko (Soil Science Faculty, Lomonosov Moscow State University, Moscow, Russia)

Figure 22. Work of floating excavator (Pxhere)

Later, when this technological operation started being conducted using excavators (Figure 22), it became possible to multiply the total area of oil-contaminated lands reclaimed annually.

After the removal of the upper layer, the concentration of oil hydrocarbons in soils usually does not exceed the levels at which activity of microbial oil destructors is impossible; this allows using the traditional methods of biological reclamation. However, a further decrease in oil hydrocarbons concentration up to acceptable levels is still a difficult task.

One of the most important problems is the optimization of soil acid–base regime. It is known that the optimal pH values for the activity of bacterial oil destructors are 6–8. But peat soils, as a rule, have pH values of 3.5–4.5 and are characterized by high values of exchangeable and pH-dependent acidity. The amount of lime that must therefore be added to achieve optimal pH values is so great that it makes this task technically and economically unfeasible and unreasonable.

One of the ways to solve this problem is by using biodegradation agents, which are capable of oxidizing hydrocarbons at pH 4–4.5. For effective oil destruction, however, it is necessary to provide a proper aeration of bog peat soils, which is extremely difficult to achieve in practice. To overcome this problem, it seems very promising to use a combination of bioaugmentation and phyto-melioration technologies (Glick, 2003; Khan *et al.*, 2013). This combination will provide a symbiotic interaction between microorganisms in biodegradation agent and bog plants, which have an ability to transport air to the root system via arenchyma, followed by diffusion of air oxygen into the rhizosphere, which would provide the possibility for oil oxidation by oil-destructive bacteria.

In addition to providing oxygen, plants can stimulate functioning of microbiota in the rhizosphere via root exudates (Bais *et al.*, 2006). In turn, bacteria can stimulate plant development by producing various phytohormones and anti-stress substances (Safronova *et al.*, 2006), thus allowing plants to grow even in conditions of heavy oil pollution. Moreover, bacteria can fix molecular nitrogen, mobilize hydrolysable phosphates, and produce siderophores, which can also promote plant development. Currently, however, the biodegradation agents possessing all the above functions have not been produced. This makes the task of development and practical implementation of appropriate biodegradation agents extremely urgent, as is the development of seed breeding of bog plants typical for the Taiga zone of Western Siberia.

4.3 | AIDED PHYTOSTABILIZATION: AN EFFECTIVE REMEDIATION TECHNIQUE FOR TAILINGS IN SE SPAIN[20,21]

Mining has been present in Sierra Minera de Cartagena–La Unión (Murcia, Spain) for more than 2 500 years. This activity has generated large amounts of tailings from the exploitation of mineral sulfides (mainly ZnS and PbS). Tailing ponds were abandoned after the cessation of the activity in 1991 and are of great concern due to the risk associated with the high content of toxic metal(loid)s.

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[21] Acknowledgements: This work was funded by the European Union LIFE+ project MIPOLARE (LIFE09 ENV/ES/000439).

Moreover, these tailings have low fertility, low organic matter content and high acidity. Therefore, the establishment of native vegetation is very difficult unless organic and/or inorganic amendments are applied (García and Lobo, 2013). Phytoremediation is considered an economic and environmentally-conscious method to remediate polluted soils (Wan, Lei and Chen, 2016). Among phytoremediation techniques, aided phytostabilization can be a solution to reduce the risk of pollutant dispersion (Yang *et al.,* 2016). Several amendments have been proposed to stabilize metal(loid)s in soils (Kumpiene, Lagerkvist and Maurice, 2008). Organic amendments and materials rich in carbonates have been successfully used to reduce the bioavailability of metals and to restore the ecological function of contaminated soils (Park *et al.,* 2011).

The main goal of this study was to determine the effectiveness of aided phytostabilization applied to a tailings pond from Zn/Pb mining 30 months after its reclamation. The effectiveness was evaluated by monitoring physicochemical and biochemical properties and bioavailable metal(loid) (As, Cd, Pb and Zn) contents in the tailings. In addition, the metal(loid) translocation to plant species (root, stem and leaf) and evolution of plant communities were also evaluated. The initial hypothesis was that the implementation of phytostabilization with native plant species with inorganic and organic amendments would decrease the mobility of metal(loid)s, decrease the risks for environment and public health, and increase soil quality and fertility and vegetation cover. Plants should accumulate high contents of metal(loid)s in their roots with low translocation to shoots.

The study was performed in Santa Antonieta tailings pond, located in Cartagena–La Unión mining district. The pond has a surface of 1.4 ha. Marble waste was used as a source of carbonates to neutralize acidity, immobilize metals and develop soil structure. Pig slurry and its solid phase (manure) after physical separation was used as a source of organic matter and nutrients for soil development and vegetation establishment.

The following species were planted in 2012: *Atriplex halimus* L., *Cistus albidus* L., *Helichrysum stoechas* (L.) Moench., *Hyparrhenia hirta* (L.) Stapf., *Lavandula dentata* L., *Lygeum spartum* (L.) Kunth., *Rosmarinus officinalis* L., *Phagnalon saxatile* (L.) Cass, *Piptatherum miliaceum*, *Cynodon dactylon*, *Limonium caesium*, *Sonchus tenerrimus*, and *Atriplex halimus.*

The results of the study showed that the combination of marble waste, pig slurry and manure was efficient for the reclamation of an acidic tailings pond by aided phytostabilization. The technique increased soil pH, CEC, TOC and nutrients content, improved soil structure and reduced the mobility of metals, mainly Cd, Pb and Zn up to 90–99 percent. *Lygeum spartum* and *Piptatherum miliaceum* were effective in phytostabilization of Pb, Zn and As, since they accumulated high metal concentrations in roots, with low aerial translocation. *Atriplex halimus* and *Phagnalon saxatile* presented phytotoxic concentrations of Zn in leaves. Therefore, the use of these species should be avoided in soils contaminated with high concentrations of Zn.

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SUPPORT OF

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ISBN 978-92-5-130505-8

9 789251 305058
I9183EN/1/04.18