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Review

Advanced technologies for the remediation of pesticide-contaminated soils

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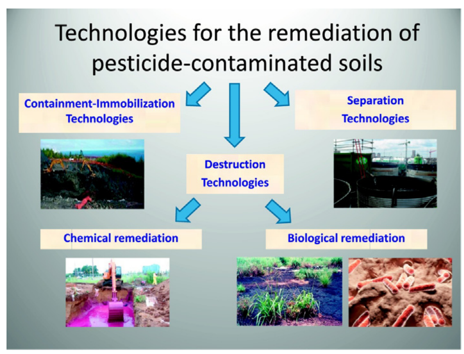
HIGHLIGHTS

- Many soils impacted by pesticide pollution due to their increasing use worldwide
- Point-source or diffuse pollution define the selected technology.
- The majority of scientific studies are still at the lab-scale stage.
- Lack of full-scale remediation technologies based on their complete degradation
- Scaling up new technologies is needed to know their real cost and efficacy.

GRAPHICAL ABSTRACT



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ABSTRACT

The occurrence of pesticides in soil has become a highly significant environmental problem, which has been increased by the vast use of pesticides worldwide and the absence of remediation technologies that have been tested at full-scale. The aim of this review is to give an overview on technologies really studied and/or developed during the last years for remediation of soils contaminated by pesticides. Depending on the nature of the decontamination process, these techniques have been included into three categories: containment-immobilization, separation or destruction. The review includes some considerations about the status of emerging technologies as well as their advantages, limitations, and pesticides treated. In most cases, emerging technologies, such as those based on oxidation-reduction or bioremediation, may be incorporated into existing technologies to improve their performance or overcome limitations. Research and development actions are still needed for emerging technologies to bring them for full-scale implementation.

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1. Fundamentals, history and general overview

Agricultural production has increased dramatically during the last decades to ensure the food supply of a population that is growing at a vertiginous rate. Greatly enhanced agricultural production has been made possible due to an increase in the use of pesticides, which have become an important part of modern agriculture. Although pesticides use is an old practice, their development and use increased dramatically after the Second World War (Gavrilescu, 2005). According to the EPA definition a pesticide is any substance or mixture of substances intended for preventing, destroying, repelling, or mitigating any pest (insects, mice and other animals, unwanted plants (weeds), or microorganisms). Although pesticides constitute an important aspect of modern agriculture, their excessive and persistent use results in damage farmland and causes serious soil pollution and deteriorated soil quality and environment. A large percentage of pesticides applied in agriculture never reach their target organisms (Niti et al., 2013). They are dispersed through soil, water and air, and detected in food for human consumption. In the case of soil-applied pesticides, their residues and metabolites can accumulate in the soil at unacceptable high levels. The potential impacts of pesticides on the environment and human health have been now recognized by governments and the public. Remediating contaminated soils to protect human health and to achieve sustainable development has become a desirable goal (Cheng et al., 2016).

In 1995, the United Nations Environment Programme Governing Council recognized only 12 Persistent Organic Pollutants (POPs) due to their adverse effects on the environment and human health. A global ban on these toxic compounds was placed, requiring taking measures to eliminate or reduce the release in the environment of these POPs. Eight of these POPs were insecticides (endrin, heptachlor, mirex, toxaphene, aldrin, chlordane, dieldrin and DDT), one of them was a fungicide (hexachlorobencene, HCB), and the rest were dioxins (some of them by-products in pesticide production), PCBs and PCDFs. As POPs, these pesticides persist in the environment for long periods of time, are toxic and biomagnify in the food chain (Ali et al., 2014). The signing of the Stockholm Convention on Persistent Organic Pollutants took effect in May 2004. In this convention the previous list was expanded in 2009 to include new POPs, being five of them other organochlorine insecticides: chlordecone, lindane, α- and β-hexachlorocyclohexane and pentachlorobencene. Endosulfans were included in 2011. This gives an idea of the importance of developing technologies to remediate pesticide contaminated sites such as soil, groundwater and aquifers, although the present review will be focused only in soils.

2. Technologies for soil pesticide remediation

As most of pesticide-contaminated soils contain complex mixtures of different compounds rather than one single contaminant, their remediation can be a complicated process. The majority of pesticides applied in agriculture are organic compounds, therefore, only their remediation will be treated in this chapter. As for the rest of organic contaminants, technologies to treat pesticide-contaminated soils fall into two categories: containment-immobilization or treatment, and treatment technologies fall into two different categories: separation and destruction.

To reduce, eliminate, isolate or stabilize a pesticide, soil remediation technologies use physical, chemical, or biological processes. The selection of appropriate technologies depends on several factors, such as site characteristics and contamination (punctual or diffuse), concentration and type of pesticides to be removed, and the end use of the contaminated media (Gavrilescu, 2009).

Depending on the technology used, techniques for soil remediation can be applied in three ways (Caliman et al., 2011): (I) *In situ*, the remediation method is applied without excavating the soil and the contaminants are treated on the place the contamination occurred; (II) *on site*, contaminated soil is excavated, treated on site and returned to the original location; (III) *ex situ*, contaminated soil is excavated and transported to another location for its treatment.

Remediation of organic pesticides can be done using any of the techniques developed for other organic pollutants with similar characteristics (Castelo-Grande et al., 2010); however, in fact, only some specific techniques are really studied and/or developed for pesticides remediation due to the particular circumstances involved in soils contaminated by these compounds. Their contamination in the majority of soils is due to diffuse pollution, and has to be treated in a different manner than in soils with point-source contamination. On the other hand, agricultural soil properties have to be maintained, and aggressive technologies used to remediate industrial polluted soils cannot be used for agricultural soils. For this reason, the aim of the present manuscript is the actualization of literature about remediation of pesticide contaminated soils in the last years, and, therefore, the majority of references collected are from the last decade, and almost all referred exclusively to pesticides.

2.1. Containment-immobilization technologies

2.1.1. Containment technologies

Pesticide contamination of soils may result not only from agricultural processes, but also from manufacturing, handling, improper storage, and disposal of pesticides and wastes. In particular, environmental contamination with organochlorine pesticides (OCPs) is widespread worldwide because, with the signing of the Stockholm Convention, many OCP production factories close to cities were abandoned (Gałuszkaa et al., 2011). Most of these contaminated sites pose a threat to the environment and face urgent land use conversion (Li et al., 2008; Yang et al., 2010; Ye et al., 2013). Due to specific properties of such pesticides, remediation efforts at macroscale in those soils where OCPs were manufactured or their waste disposed are based on containment-immobilization technologies. Such techniques comprehend excavation and landfilling both inside and outside the site, capping to limit infiltration and leaching, as well as subgrade barriers to limit lateral plume migration, and include engineering techniques to remove or isolate the