**Article** **ECORFAN Journal-Republic of Nicaragua**
June, 2019 Vol.5 No.8 29-35

# Remediation methods for the removal of pesticides in wastewater

# Métodos de remediación para la remoción de pesticidas en aguas residuales

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**DOI:** 10.35429/EJRN.2019.8.5.29.35 Received March 28, 2019; Accepted May 22, 2019

**Abstract**

This work is part of one of the priorities of sustainable development that is the conservation of soil and the care of aquifers. Water is a vital liquid for human and all kind of living beings, the presence of pesticides in drinking water is a health problem that requires solution. In this paper, a review of the different methods used for the removal of pesticides in wastewater is made, such as biological remediation methods, using plants and microorganisms, remediation by physical methods by adsorption of contaminants with activated carbon, zeolites, polymers and clays and finally chemical remediation, through advanced oxidation with the production of hydroxyl radicals. A review of the most commonly used pesticides in the different agricultural areas is carried out, as well as their impact on the health of the inhabitants in these regions. Finally, a comparison of the advantages and disadvantages of these methods is made both for its effectiveness as well as for their cost.

**Remediation methods, Removal of pesticides, Wastewater**

**Resumen**

El presente trabajo se inscribe dentro de una de las prioridades del desarrollo sostenible que es la conservación del suelo y el cuidado de los mantos acuíferos. El agua es un líquido vital para el ser humano y todos los demás seres vivos, la presencia de pesticidas en el agua potable es un problema de salud que requiere solución. En este trabajo se hace una revisión de los diferentes métodos utilizados para la remoción de pesticidas en aguas residuales, tales como métodos de remediación biológicos, utilizando plantas y microorganismos, remediación por métodos físicos por adsorción de los contaminantes con carbón activado, zeolitas, polímeros y arcillas y finalmente remediación química, mediante oxidación avanzada con la producción de radicales hidroxilo. También, se hace una revisión de los pesticidas de mayor uso en las diferentes zonas agrícolas, así como su impacto en la salud de los habitantes en dichas regiones. Finalmente, se hace una comparación de las ventajas y desventajas de estos métodos tanto por su eficacia, así como por su costo

**Métodos de remediación, Aguas residuales, Remoción de pesticidas**

**Citation:** GODINEZ-GARCÍA, Andres, HERNÁNDEZ-MORALES María Guadalupe, GUIJOSA-GUADARRAMA, Santiago and DÍAZ-TECANHUEY, Pedro Jesús. Remediation methods for the removal of pesticides in wastewater. ECORFAN Journal-Republic of Nicaragua. 2019 5-8: 29-35

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## Introduction

Population growth has led to an increase in food production. And this in turn leads to an increase in the use of pesticides, with the aim of increasing the production and quality of crops. Pesticides are also used in industry, in gardening, in domestic use, livestock use and urban use, that is, they are practically present in every activity of our daily lives. However, the use of pesticides has caused health problems in animals and humans since many of them are highly toxic and also contaminate soil, water and air.

A pesticide or pesticide is a substance whose objective is to destroy, repel, attract or prevent any type of pest, be it of animal or plant origin. According to their biological activity, pesticides are classified as herbicides, insecticides, fungicides, bactericides and rodenticides (Bejarano-González edit. 2017). 7 types of pesticides, chemical, biochemical, microbial, botanical, microbial and miscellaneous are distinguished (Bejarano-González edit. 2017). In particular, chemical pesticides have had considerable development since 1940, the year in which the insecticidal properties of DDt were discovered and since then the development of new pesticides has intensified; Some of the most important groups are organophosphates, organochlorines, carbamates and pyrethroids. (Table 1).

| Group | Level of toxicity | Rapid degradation in the environment | Some commercial pesticides | Health damage |
|---|---|---|---|---|
| Organophosphates (inectidal herbicide, fungicide and acaricide) | Very toxic (I) | Very fast and easy to degrade | glyfosfato, dimetoato malation, paration | It damages lungs, liver, kidneys, bone marrow and nervous system irreversibly . |
| Organochlorines (insecticide, acaricide, herbicide and fungicide) | Very toxic (I) | Very slow | DDt,atrazin, lindano, endrin,heptacloro, endosulfan,metoxiclor,hexaclorobenceno | Cancer, central and peripheral nervous system, liver and kidney. They accumulate in fatty tissues |
| Carbamates (insecticide, fungicide, herbicides) | Moderately toxic (II) | Very fast | carbaryl, aldicarb baygon, carbofuran | Nervous and respiratory system |
| Pyrethroids (insecticide) | Moderately toxic (II) | Very fast | permetrina, cipermetrina, | Nervous system, brain |

**Table 1** Some of the most important chemical pesticides and their effects on health

Data from the 2014 National Agricultural Survey indicate that of the total hectares of production in the country only 16.7% use biological control of pests, in the remaining 83.3% chemical pesticides are used.

This tells us about the high consumption of agrochemicals in the country. One of the problems in its control is that there is no detailed information on the active ingredients used. Information from the Mexican Social Security Institute (IMSS) indicates that from 2001 to 2016 there have been 31,257 cases of acute pesticide poisoning nationwide (Guzmán-Plazola et al.2016). According to FAO data, the consumption of pesticides in Mexico, during 2014 was 98,814 t, of which 40% correspond to fungicides and bactericides, 32% to insecticides and the remaining 27% to herbicides (Bejarano-González edit. 2017) as shown in Fig. 1.

The objective of this article is to present an overview of the use and risks that pesticides present in Mexico, as well as the existing remediation methods and the options to replace chemical pesticides with biological options. For this purpose, a review of the main pesticides used in several states with a higher food production was made, which are presented in section 2.

In section 3 different methods used for the degradation of pesticides in water are presented, while that in section 4 some of the remediation processes used in Mexico are listed. Finally, section 5 corresponds to our conclusions and we discuss the scope and limitations of our work. It is important to mention that while the different hydrocarbon remediation processes are known, in the case of pesticides there is very little information about.

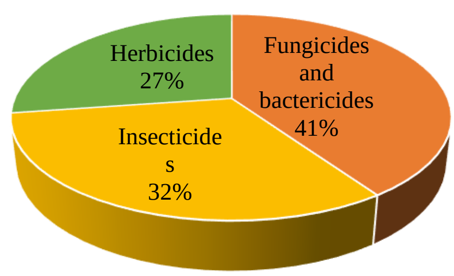


**Figure 1** Pesticide consumption in Mexico, during 2014, according to FAO data

## Pesticide use in Mexico

Mexico produces large amounts of food, both for internal consumption and for export, as well as being one of the leading producers of flowers worldwide.

The states where pesticides are most used are: Campeche, Chiapas, Chihuahua, State of Mexico, Morelos, Michoacán, Nayarit, Oaxaca, Puebla, Sinaloa, Sonora, Tabasco, Tamaulipas, Veracruz and Yucatan according to a statement from the Secretariat of Health of 2012. These states are key in the production of food for their levels of production intended for both national consumption and export (Arellano y Rendón 2016).

Sinaloa is one of the largest food producers in the country, with an area of 1,153,311.78 hectares, where corn, sorghum, tomato, green chili, potato, watermelon, forage sorghum, sugar cane, alfalfa and magician are produced. Between November 2011 and October 2012, 246 tons of pesticides were used. The most used were fungicides, herbicides and insecticides of the families of organochlorines, organophosphates, carbamates and pyrethroids (García Hernández et al. 2018). In studies conducted by Green Peace (2016) and Silvera-Gramont et al. (2018), considerable contamination was found, in soil, water and biota, due to pesticide discharges. In Sinaloa, 80 new cases of childhood cancer are presented each year and this is probably due to contamination by agricultural pesticides (Ministry of Health of the State of Sinaloa (SS, 2013)) and, on the other hand, Chaín-Castro et al. (1998) reported that 20% of Culiacán farmers, mainly those in charge of agrochemical mixtures, have presented intoxication at least once, due to contact with pesticides.

In Sonora, the Rural Development District (DDR) 144 Hermosillo has a cultivated irrigated area of 72,499 ha (SIAP 2015). The main crops are alfalfa, pumpkin, chickpea, orange, walnut, watermelon, sorghum, wheat and vine. Between 2009 and 2010, 270 t of pesticides were used, of which the most commonly used were fungicides followed by insecticides and herbicides belonging to the organophosphorus groups, dithiocarbamates, synthetic pyrethroids and organochlorines (García-Hernández et al. 2018). Regarding the use of biological pesticides, 25% was used in chickpea, while in the rest of the crops 5% was used (SENASICA 2011).

In the basin, in Valle de Santiago and Salvatierra the area of cultivated hectares is 760,921 (SIAP, 2015). The main crops are corn, sorghum, wheat, barley and vegetables.

It is estimated that in the spring-summer cycle, 9.515.3 t of pesticides were used, corresponding to approximately 10% of the national total reported by SENER (2017). Of these pesticides, 82.1% correspond to highly dangerous pesticides (PAP), among which edonsulfan, methamidophos and carbofuran, which are banned in different countries.

The municipalities of Coatepec Harina, Ixtapan de la Sal, Tenancingo, Tonatico, Villa Guerrero, Malinalco and Zumpahuacán, in the state of Mexico, are dedicated to the cultivation of vegetables and fruits, although their main activity is flowers, whose production corresponds to 50.4 % of national production and 80% of exports to the United States, Canada and Europe. In the floricultural production, organochlorine pesticides, pyrethroids, benzimidazole and triazole are mainly used (Martinez et al. 2014. When conducting studies to the farmers of Villa Guerrero, it was found that of 52 cases studied, 50% presented one or more pesticides in their waste According to statistics from the Institute of Health of the State of Mexico (ISEM), during 2006 140 cases of poisoning were recorded. (Ferrusquía-García et al. 2008).

In the area of Los Altos de Chiapas, in the municipalities of Chamula, Zinacat and Amatenango del Valle, the population is engaged in horticultural, floricultural and maize production. Producers of flowers and vegetables most frequently use highly toxic pesticides belonging to organophosphates and carbamates such as methyl parathion, methamidophos, paraquat and glyphosate among others (Díaz-Coutiño, et al. 1998; Cantoral, 2001). Due to their ignorance in the handling of pesticides and the damage they cause to the environment and their health, they are contaminating water, when disposing of, pesticide containers in areas that are recharged by aquifers. On the other hand, very high amounts of DDT (37.5 µg / L) have been found in the blood of children in locations where this pesticide was used until 2000 (Herrera-Portugal et al. (2008)).

As in other states, in Yucatan and Campeche, the consumption of agrochemicals has increased, due to the industrial planting of GM soy, corn and vegetables. According to Greenpeace studies between 1990 and 2005 there was an increase of 606 to 4800 tons of pesticides in the state of Yucatán.

The main pesticides used are glyphosate, endosulfan, paraquat, 2,4-D, carbofuran, emamectin benzoate, methamidophos, emamectin benzoate and cypermethrin. Several of these pesticides are classified as highly dangerous (PAN, 2016) and some are banned in other countries, (PAN 2017) (Rendón von Osten 2004). In fact, Campeche from 1998 to 2010, presented at national level one of the highest rates of pesticide poisoning according to the report of the Epidemiological Bulletin of the Ministry of Health (Gutiérrez, 2013)

**Methods of degradation of pesticides in water**

The process of contamination by pesticides in waters is due, on the one hand, to their use in agriculture and on the other, due to their poor management, as waste and pesticide containers are dumped in rivers, lakes, lagoons or near some freatic level. Once pesticides are released into the environment, they suffer alterations in their composition due to environmental conditions such as temperature, intensity and duration of solar radiation, pH and water composition. There are different methods for the degradation of pesticides in water, the selection of the method depends on the pesticides to be treated, their concentration and the environment. In principle, what is expected is that the degradation leads to the mineralization of the pesticides obtaining CO2, H2O, etc. But sometimes what is achieved is its degradation into compounds that may be more toxic and persistent than the initial products. Below are the main methods used in the degradation of pesticides in water.

**Remediation by biological processes or bioremediation**

The purpose of these processes is to transform dangerous organic compounds into less harmful compounds such as CO2 and H2O through the use of microorganisms, algae, worms, fungi and plants, present in the system or outside the environment. The advantage of these processes is that they are environmentally friendly and inexpensive (Nwankwegu and Onwosi 2017; Rajiv et al. 2009). Among the factors to be considered for its effectiveness are temperature, pH, pesticide concentration, chemical and physical characteristics of the substrate, nutrients, etc. that affect the development of microorganisms responsible for the mineralization of pesticides.

**Remediation by Physical processes**

These processes allow the elimination of pesticides without changing their chemical structure, through physical methods such as clays, activated carbon, zeolites and polymeric materials among others (Tan 2009).

In general, these methods work by adsorption, in which the contaminants are adhered to the surface of the materials used.

**Clays**

Mineral clays are hydrophilic and have a negative charge so they can be used to retain cationic pesticides. They can also be mixed with Fe or Ti to improve their adsorption. The important factors in the adsorption of pesticides by clays are temperature, solution pH, pesticide concentration, contact time, adsorbent mass and ionic strength.

**Activated carbon**

It is a material that adsorbs pesticides and drugs very efficiently in water. Its efficiency depends on the pH of the solution, the temperature and other compounds dissolved in the solution. There are different varieties of activated carbon such as granules and powders.

The most commonly used is dust due to its lower cost and ease of handling. There is also a carbon-rich material called biochar, which is obtained from biomass by pyrolysis (burning at very high temperatures plant material).

A disadvantage of this material is that regeneration of activated carbon is very expensive. Patricia Torres and collaborators used activated carbon in combination with a coagulation method to decrease the concentration of pentachlorophenol in the Cauca River below 1.56 µg/L (Reyes Serrano y López Alejo 2016, p.36).

**Zeolites**

These materials have been used to remove pesticides and heavy metals from water. Even though De Schmedt (2015) showed that adsorption depends on the polarity and mobility of the pesticide.

## Polymeric materials

The advantage of these materials compared to activated carbon is that they do not require so much energy to regenerate the adsorption capacity and this represents a cost advantage. The disadvantage of some organic pesticides is their low water solubility, some polymeric materials are currently being used to improve their solubility.

Among the most commonly used polymers are cyclodextrins, dendrimers and hyper-cross polymers. The dendrimers have been used with great success in the removal of pesticides in drinking water, which were not removed by nanofiltration membranes due to their low molecular weight. However, the main disadvantage of dendrimers and hypercrossed is its high cost of synthesis.

## Chemical Remediation

## Advanced Oxidation Processes

This methodology consists of a family of processes that use hydroxyl radical generation, oxidation reactions on pesticides to degrade them in inorganic salts, H2O and CO2. These processes can be catalytic or non-catalytic and with the contribution or not of external energy.

The difference between the different processes is the way in which the hydroxyl radical is generated. In general, these processes require the aggregation of hydrogen peroxide and ozone, reagents that are expensive and limit their use in the treatment of large quantities of water (Rodríguez et al. 2010). Table 2 shows the family of advanced oxidation processes. (Rodríguez et al. 2010).

| Advanced oxidation processes | | | | |
|---|---|---|---|---|
| **Heterogeneous Processes** | **Homogeneous Processes** | | | |
| | **Without energy input** | **With energy input** | | |
| | | **Electric** | **Ultrasound** | **Radiation Ultraviolet** |
| Ozonation<br>Catalytic<br>Catalytic photo<br>Heterogeneous<br>Heterogeneous photocatalysis<br>Fenton<br>Heterogeneous<br>Photofenton | $O_3$ in alkaline medium<br>$O_3/O_3H_3$<br>$Fe^{+2}/H_2O_2$ | Oxidation electrochemistry<br>Anodic oxidation<br>Electro-fenton | $O_3$/US<br>$H_2O_2$/US | $O_3$/UV<br>$H_2O_2$/UV<br>$O_3/H_2O_2$/UV<br>$Fe^{+2}$/H2O2/UV |

**Table *2*** Advanced oxidation processes. Adapted from Rodriguez et al.2010

## Dissolved oxygen processes

This methodology uses dissolved oxygen as an oxidant for pollutants and there are two methods, wet oxidation and supercritical oxidation (Rodríguez and Santos 2010). In wet oxidation, dissolved contaminants are oxidized using oxygen or air in a temperature range of (125-300 °C) and pressures of (5-200 bar) (Zimmermann et al. 1960, Himmelblau et al. 1960). The process can be carried out with or without catalysts. The advantage of using catalysts is that it improves the conversion of pollutants, decreases temperature and pressure conditions. The catalysts used are salts of Fe + 2, Cu + 2, noble metals such as (Ru, Pd, Pt) and metal oxides.

## Remediation processes used in Mexico

Knowing the problems that come with the intensive use of pesticides, various studies have been carried out to explore less harmful methods for health and the environment in the fight against pests and for the remediation of water and soil contaminated with pesticides.

In this sense, Thompson et al 2000, and Infante-Rodríguez et al. 2011, proved that the effectiveness of the spinosal naturally occurring larvicide is similar to that of the organophosphorus pesticide temefos in the fight against black fly, vector of onchocerciasis, (Infante-Rodríguez et al. 2011) and against the pest of the fly White in cotton.

On the other hand, in the state of Mexico the degradation of methyl parathion in aqueous solution was studied using heterogeneous photolysis, in the presence of titanium dioxide (TiO2) and hydrogen peroxide. In this study they obtained a removal percentage of 58% at 60 min (Ferrusquía-García et al. 2008).

While by means of a photochemical process in the presence of folic acid they obtained the degradation of methyl paration, in p-phenol and other products at a PH 2 (Manzanilla-Cano et al. 2008). In Yucatan, biological beds have been used, consisting of a mixture of soil and a component such as straw, charcoal, corn husks, etc. where pesticides are retained and degraded, Góngora-Echeverría et al. 2018 have obtained the removal of up to 99% of 2.4 dichlorophenoxyacetic acid (2,4-D), atrazine, carbofuran, diazinon and glyphosate after 40 days.

## Conclusions

In the literature there are several publications that refer to the pesticides most commonly used in the Mexican countryside, both for the production of food and flowers. As well as the damages that originate to the health of human beings, animals and the environment, contaminating soil and water. Likewise, there are publications in which they expose the main methods of chemical pesticide remediation in wastewater, but these are mainly at the laboratory or pilot plant level. However, while the different hydrocarbon remediation processes are known, in the case of pesticides we made an extensive review and found very little information about it, when it comes to its application in the field.

As for the remediation processes known in the treatment of wastewater, although the chemical remediation processes are successful in the oxidation of various pesticides, they are expensive and in many cases more toxic and persistent metabolites than the initial compound are generated, coupled with the fact that so far its use is at the laboratory or pilot plant level. As for the physical processes by membranes or activated carbon, they have the disadvantage of being expensive both in the activation of the carbon and in the cleaning and operating conditions of the membranes since high pressures are required for their operation, the advantage of these methods Remediation is that they do not add any chemical agent to the medium.

On the other hand, bioremediation processes are friendly to the environment, they are economical, it is not necessary to implement them every time an agrochemical is applied, unlike the chemical and physical processes that must be implemented every time the pesticide is applied. This means savings in energy and economic resources. In this way the most efficient and economical way to eliminate pesticides in wastewater is to make a combination of biological methods with chemical and / or physical methods.

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