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Dissemination of Endosulfan into the Environment

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Abstract

Organochlorine pesticide pollution of surface water, groundwater, soil, sediments, and air has been recognized as a major problem in many countries because of the persistence in different environments and the consequent potential adverse health effects. This report presents a review of literature on organochlorine insecticide of endosulfan, which has been used extensively for agricultural purposes. It summarizes information on nomenclature, physical-chemical properties, production, metabolism, degradation, distribution in the environment, toxicity, regulation, and technologies for the treatment of water and soil contaminated with this pesticide.

Keywords: persistence, pesticide, pollution, toxicity

1. Introduction

Although pesticides are economically advantageous in agriculture, increasing crop yields, they do generate environmental problems [1]. Some of the most problematic pesticides are the organochlorinated ones, where chlorine is incorporated in organic molecules. Organochlorinated pesticides are highly effective and long lasting. However, these properties lead to environmental persistence and resistance to degradation, thus they are classified persistent organic pollutants. Typical organochlorinated pesticides include DDT, aldrin, dieldrin, endrin, heptachlor, endosulfan, and others.

Endosulfan has been used on a variety of crops including broccoli, potatoes, coffee, cotton, peaches, apples, nectarines, prune, lettuce, tomatoes, grapes, melons, cauliflower, carrots, cabbage, rape, strawberry, alfalfa, beans, cereals, cucumber, tobacco, tea, oil crops, and some

ornamental flowers to attack pests [2–4]. It typically enters the atmosphere through agricultural application and can be transported long distances in the air. The levels in the air vary substantially depending on the location. Rural areas tend to have higher levels. Endosulfan half-life in soils is estimated to range from 60 to 800 d, while its half-life in groundwater and sediments may increase up to 6 years [5–8].

In this chapter, a complete physicochemical description of endosulfan is presented. The degradation sub-products, toxicity, and environmental impacts are described. Finally, some technologies that have been recently applied to treat the residues are described.

2. Chemical aspects

Endosulfan is an organochlorine insecticide (6,7,8,9,10,10-hexachloro-1,5,5a, 6,9,9a-hexahydro-6, -9-methane-2,3,4-benzo-dioxathiepin-3-oxide) with a CAS Number 115-29-7 that has a cyclodiene structure. It contains a sulfite diester group that is relatively reactive and infers persistence in the environment. The condensed chemical formula is $C_9H_6Cl_6O_3S$.

This pesticide has been widely used since its introduction in the 1954 by Farbwerke Hoechst because it has a broad spectrum of activity. The commercial grade of endosulfan is a mixture of α - and β -endosulfan isomers (**Figure 1**), in a 7:3 ratio, respectively. Its main degradation product is endosulfan sulfate, which has similar properties to the compounds of origin [9–11].

3. Trade names, formulations, and use

Some of the trade names that have been given to endosulfan are Cyclodan, Thiodan, and Thiosulfan, among others that can be found elsewhere [12]. The trade formulations of endosulfan are emulsifiable, microencapsulated, powder, wettable powder, granules, technical powder, technical solid, and technical liquid, with a concentration for agricultural use of 350–500 d of active ingredient (AI)/L or kg and for industrial use the concentration is 940–970 (AI)/L or kg [13].

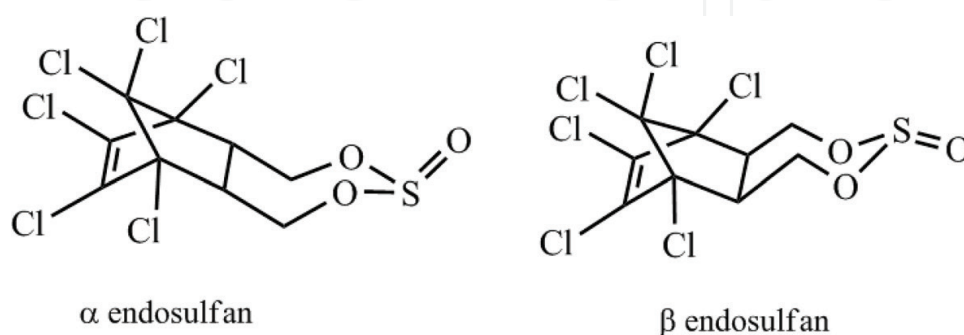


Figure 1. Structure of isomers α and β endosulfan.

4. Physicochemical properties

Table 1 shows some physicochemical properties of α -endosulfan, β -endosulfan, and endosulfan sulfate [14, 15].

Some of the environmental fates of the endosulfan are related to its physicochemical properties. For instance, the aqueous solubility of the α -isomer is higher than the β -isomer and endosulfan sulfate. Compounds whose solubility is less than 25 mg L^{-1} tend to be immobilized in soils and living organisms. The three molecules have partition coefficients octanol-water > 1 . Therefore, they are easily absorbed through biological membranes and accumulate in fatty tissues.

Another important characteristic is the vapor pressure. Endosulfan sulfate is the least volatile with a vapor pressure of 10^{-7} mm Hg , so is more persistent in soil and water. The isomers of endosulfan are semi-volatile, with similar vapor pressures to other chlorinated pesticides, making them susceptible to volatilization to the atmosphere with subsequent atmospheric transport and deposition.

One of the criteria for designing of a chemical as a persistent organic pollutant (POP) is that it has $\log K_{ow} > 5$, both endosulfan isomers and endosulfan sulfate do not exceed this value but are close to it, suggesting a potential for bioaccumulation. The relatively high K_{oc} values indicate a propensity for partitioning to the organic carbon fraction in soils and sediments for both endosulfan isomers and the sulfate [3, 15].

Properties	α -Endosulfan	β -Endosulfan	Endosulfan sulfate
Water solubility pH 5, 25°C, mg/L	0.33	0.32	0.22
Henry constant $\text{atm m}^3/\text{mol}$. 25°C	1×10^{-5}	1.91×10^{-5}	2.61×10^{-5}
Vapor pressure at 25°C mm Hg	1×10^{-5}	1×10^{-5}	1.0×10^{-11}
Log. octanol-water partition coefficient (K_{ow}) pH 5.1	3.83	3.62	3.66
Log. octanol-air partition coefficient (K_{oa})	10.29	10.29	5.18
Log. octanol-carbon partition coefficient (K_{oc})	3.5	4.1	No data
LD_{50} rats (mg/kg)	76	240	160

Table 1. Physicochemical properties of α -endosulfan, β -endosulfan, and endosulfan sulfate.

5. Production of endosulfan

The global production of endosulfan was approximately 18,000–20,000 t annually from 1958 to 2000 with a cumulative global use of 300,000 tons. Approximately one third of this was

generated in India, but it had agreed to phase out its use in 2017. China produced approximately 5000 tons per year, and the remaining endosulfan was produced by Israel, Brazil, and South Korea [16]. Regarding the consumption of endosulfan, India was the main consumer 113,000 t from 1958 to 2000, followed by the United States with 26,000 t from 1954 to 2000. In China, annual use is estimated to average 2800 t/y during the period 1998–2004 [3]. Endosulfan is prohibited in 80 countries; some countries have registered only a few commercial products containing endosulfan, other countries have registered dozens of commercial products from various formulators. Endosulfan is now still produced and widely used in the crop fields in most of the developing countries, due to its effectiveness and low application cost [17].

6. Distribution of endosulfan in the environment

Endosulfan is widely distributed in the environment and can be detected in all media, soil, sediments, air, water, and vegetation, over long distances from the application source. Levels of endosulfan in the air samples are very variable depending on the location. Recent measurements indicate that around 320 d is the half-life in the atmosphere [18].

Endosulfan is one of the most abundant organic pesticides in the atmosphere and is capable of reaching remote locations such as the Arctic. The presence of endosulfan in the Arctic at concentrations between 3 and 8 pg m⁻³ has been detected. Endosulfan concentration was obtained using an XD-based passive sampler with samples collected in monitoring stations.

It has been suggested that the transport of endosulfan occurs when the compound volatilizes from warmer regions, experiences long-range atmospheric transport, and condenses again, permitting an accumulation of the substance in temperate regions, higher mountains, and the Arctic. Endosulfan residues in snow and ocean waters in the Arctic indicate that atmospheric deposition of endosulfan is taking place, and it can promote bioaccumulation in zooplankton and various species of fish. For some biota, a biomagnification factor for α -endosulfan from fish to seal is also occurring [3, 19–21].

7. Degradation of endosulfan

Figure 2 shows the oxidation pathway of endosulfan; it has been reported that even though some hydrolysis takes place, at environmental conditions, the most common product is endosulfan sulfate [2, 16, 22]. After initial oxidation, four intermediates are detected until endosulfan lactone is reached. In further oxidation, the complete mineralization can be obtained. Endosulfan is relatively resistant to photodegradation but its metabolites, including endosulfan sulfate, are susceptible to photolysis [23]. **Figure 2** shows the main pathways of endosulfan degradation.

In aquatic systems, the α -isomer is converted more readily to endosulfan sulfate than the β -isomer. Various studies that have observed the degradation of endosulfan in aqueous phases identified hydrolysis as the dominating abiotic degradation process resulting in the

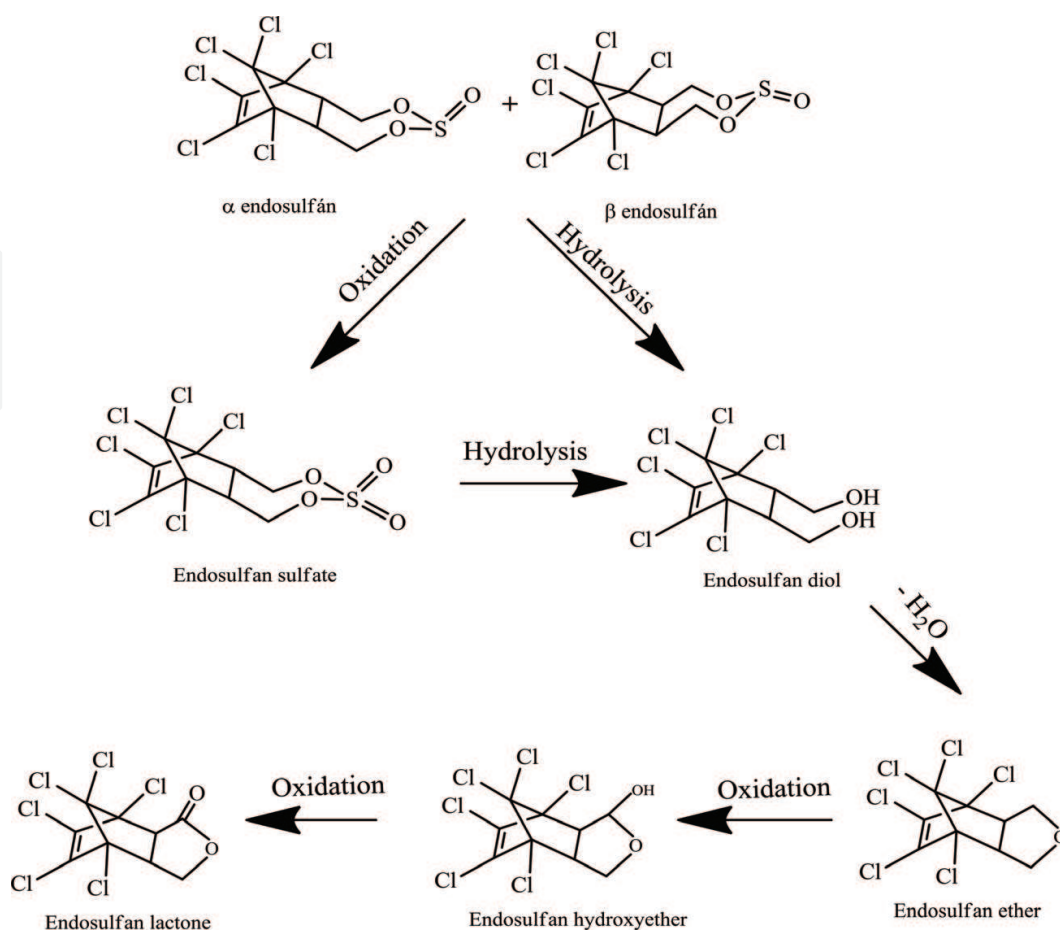


Figure 2. Degradation of isomers α - and β -endosulfan.

formation of endosulfan diol. A positive correlation between hydrolysis rate (k_{hydro}) and pH, with base-driven hydrolysis, a predominant degradation process in seawater conditions, allows a rapid degradation of endosulfan into endosulfan sulfate, whereas it takes longer in pure water.

Furthermore, hydrolysis of endosulfan sulfate to endosulfan diol is markedly slower compared to the hydrolysis of α -endosulfan.

The biodegradation of endosulfan has been reported in anaerobic conditions; these conditions predominate in sediments and the results can contradict the interpretation of the isomer deposition in freshwater sediments. At neutral pH and aerobic conditions, the degradation of endosulfan by biological oxidation has also been reported, being sulfate of endosulfan, the compound with the heaviest concentration.

It has been reported that the decomposition of endosulfan in aqueous solution can take place, but some natural environmental factors lead to the formation of sulfate indicating that some oxidation pathway is followed. The values are much lower than the persistence criteria designated for a POP, although in the colder marine waters at high latitudes, base-driven hydrolysis half-lives for endosulfan are likely to be greatly extended, compared to warmer waters in temperate or tropical regions [3, 15, 23].

There are relatively few studies that have examined the persistence of endosulfan in soil under field conditions. Degradation rates strongly depend on the soil conditions (particularly soil water content and ambient temperature).

The Environmental Protection Agency (EPA) indicates that concentrations above $0.22 \mu\text{g L}^{-1}$ (acute) and $0.05 \mu\text{g L}^{-1}$ (chronic) have adverse impacts on the health of aquatic organisms. To avoid intoxication from water consumption, fish or shellfish from water bodies near fields where the pesticide is used, concentration limits need to be established. For air concentrations, the limit recommended by the National Institute for Occupational Safety and Health (NIOSH) is 0.1 mg m^{-3} [22, 24].

8. Regulation of endosulfan

Health authorities and legislators have implemented strategies for the use of pesticides due to the thousands of deaths reported every year due to the indiscriminate use of these substances. In 1985, the code of conduct on the distribution and use of pesticides by the United Nations Food and Agriculture Organization (FAO) was published to regulate the use and availability of pesticides. For a safe use of pesticides, several actions need to be undertaken; in the case of farmers, education on the application of products needs to be provided, government inspectors require to check the application of pesticides in the crop fields within the framework of FAO's Integrated Pest Management System [25].

9. Stockholm convention

This convention that deals with persistent organic pollutants (POPs) is an international treaty for the protection of human health and the environment from the effects of chemical compounds that remain for long periods in the environment, are widely distributed, and accumulate in the fatty tissues of humans and animals. The exposure to these compounds can result in serious health problems, such as some cancers, birth defects, and deficient immune and reproductive systems, among others [12].

The POP Review Committee (POPRC) is a scientific body created to review the chemical compounds that are proposed for inclusion in the text of the Annexes A, B, or C of the convention. Endosulfan was presented for inclusion in Annex A by the European Union in November 2007. The Committee of Review issued the decision POPRC-4/5, it specifies that "the selection criteria for endosulfan have been met" and a working group was established to elaborate a draft risk profile in accordance with Annex E of the Convention [12].

The information was requested from Parties for Annex F as well as the review of additional information on the adverse effects of endosulfan on human health [12, 15]. After evaluating the risk profile and finalizing the risk management evaluation, the POPRC, at its sixth meeting held in Geneva from October, 11 to 15, 2010, decided to recommend to

the Conference of the Parties the inclusion of technical endosulfan, its α - and β -isomers as well as the main degradation product, the sulfate of endosulfan in Annex A of the Stockholm Convention for global elimination with possibility of specific and temporary exceptions [15].

10. Rotterdam Convention

This exchange under the Rotterdam Convention on “prior informed consent” applies to banned or restricted chemicals and to the severely hazardous pesticide preparations listed in Annex III. Currently, there are more than 30 chemical products to which the PIC procedure applies. At the fifth meeting of the Conference of the Parties to the Rotterdam Convention that took place in Geneva, Switzerland, between June 21 and 24, 2011, it was agreed to include the endosulfan pesticide in Annex III of the Convention.

Previously, the seventh meeting of the Chemical Review Committee, held in Rome from March 28 to April 1, 2011, recommended the inclusion of a number of new chemicals in this Annex, including the pesticide endosulfan that had recently added to the list of the Stockholm Convention of chemical substances that must be eliminated [26].

11. Recent technologies to treat endosulfan present in water and soil

Nowadays, there is a concern for the development of alternatives for the elimination of endosulfan in both water and soil. In this context, advanced oxidation processes (AOPs) are considered a highly competitive technology for the removal of those organic pollutants not treatable by conventional techniques. All AOPs produce $\bullet\text{OH}$ radicals, they are very reactive, attack most organic molecules, and are not highly selective. The generation of $\bullet\text{OH}$ radicals is through different reagent systems, which include photochemical degradation processes (UV/O_3 , $\text{UV}/\text{H}_2\text{O}_2$), photocatalysis (TiO_2/UV , photo-Fenton reactives), chemical oxidation processes (O_3 , $\text{O}_3/\text{H}_2\text{O}_2$, $\text{H}_2\text{O}_2/\text{Fe}^{2+}$), and electrooxidation (BDD electrodes) [27].

Others treatments for pesticides are biodegradation and bioremediation; both of these are based on the conversion or metabolism of pesticides by microorganisms. The difference between these two is that the biodegradation is a natural process, whereas bioremediation is a technology. One disadvantage of these processes is that they require a long time to degrade the pesticide [28].

One of the technologies most resorted to eliminate pesticides present in water is adsorption with activated carbon, as it can be applied at a large scale. Good characteristics of activated carbon include large surface area and well distributed porosity [29].

Table 2 shows some technologies that have been developed to degrade endosulfan present in water and soil.

Treatment	Matrix	Conditions	Efficiency	Reference
Electrooxidation by boron-doped diamond anode	Water	Concentration of endosulfan: 6 g/L Supporting electrolyte: NaCl (1 g/L) Current density: 60 mA cm ⁻² Reaction time: 2 h pH: 6	81% removal COD	[30]
Ozonation	Water	Concentration of endosulfan: 10 mg/L Ozone dosage: 57 mg/min Reaction time: 60 min pH: 10	93% removal endosulfan	[31]
Fenton	Water	Concentration of endosulfan: 7.5 mg/L FeSO ₄ dose: 50 mg/mL H ₂ O ₂ dose: 236 mg/mL Reaction time: 1 h pH: 3 Temperature: 30°C Continuous shaking: 130 rpm	83% removal endosulfan	[32]
Adsorption with sawdust carbon	Water	Concentration of endosulfan: 20 mg/L Adsorbent: 0.2 g Contaminated water: 200 mL pH: 7 Contact time: 250 min Agitation rate: 200 rpm	95% removal endosulfan	[33]
Biodegradation by <i>Aspergillus niger</i>	Soil	Concentration of endosulfan: 400 mg/ml Soil: 5 g Incubation time: samples were taken at 12 h interval up to 144 h Temperature: 30 ± 2°C Rotary shaker: 120 rpm pH: 6.8	Complete disappearance of endosulfan	[34]
Photocatalytic degradation with the elution of surfactants	Soil	Concentration of endosulfan: α- and β-endosulfan 12.03 and 6.87 mg/kg, respectively. Soil: 25 g Photocatalyst: nitrogen-doped anatase TiO ₂ under visible-light irradiation. Eluents: Tween 80, SDS and Na ₂ SiO ₃ Reaction time: 240 min Temperature: 25 ± 2°C in dark Rotary shaker: 200 rpm pH: 6.23	Complete degradation	[35]

Treatment	Matrix	Conditions	Efficiency	Reference
Nano-phytotechnological using zero-valent iron nanoparticles	Soil	Concentration of endosulfan: 1139 µg/Kg of soil Plants: <i>A. calcarata</i> , <i>O. sanctum</i> , <i>C. citrates</i> Temperature: 30 ± 3°C and natural light Contact time: 28 d nZVIs: 1000 mg/kg	The maximum % removal endosulfan was with <i>Alpinia calcarata</i> 94.9%	[36]

Table 2. Technologies for degradation of endosulfan present in water and soil.

12. Conclusions

One of the most important economic activities worldwide is agriculture. From the early 50s and owing to population growth, intensive agriculture practice has been carried out. In this kind of massive production, an excess of chemical substances was used, among them, pesticides. Endosulfan has been one of the most used pesticides due to its wide spectrum of activity; nevertheless, it has very negative consequences on the environment. The physicochemical properties of endosulfan favor bioaccumulation and biomagnification, and it is considered a persistent organic compound; thereby, in 2007, it was proposed to be included in the Stockholm agreement, aim that was reached in 2010. Thanks to the information gathered, a large number of countries have banned the use of endosulfan; however, many other countries still use it on their fields. It has been found that endosulfan is transported to zones considerably far from the application fields, as an example, endosulfan has been detected in the Arctic. Although a large number of countries have opted not to make use of this pesticide, research continues in order to find out how to eliminate it from soil, sediments, water, and a proper disposal of the product that that will be no longer used [37–39].

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