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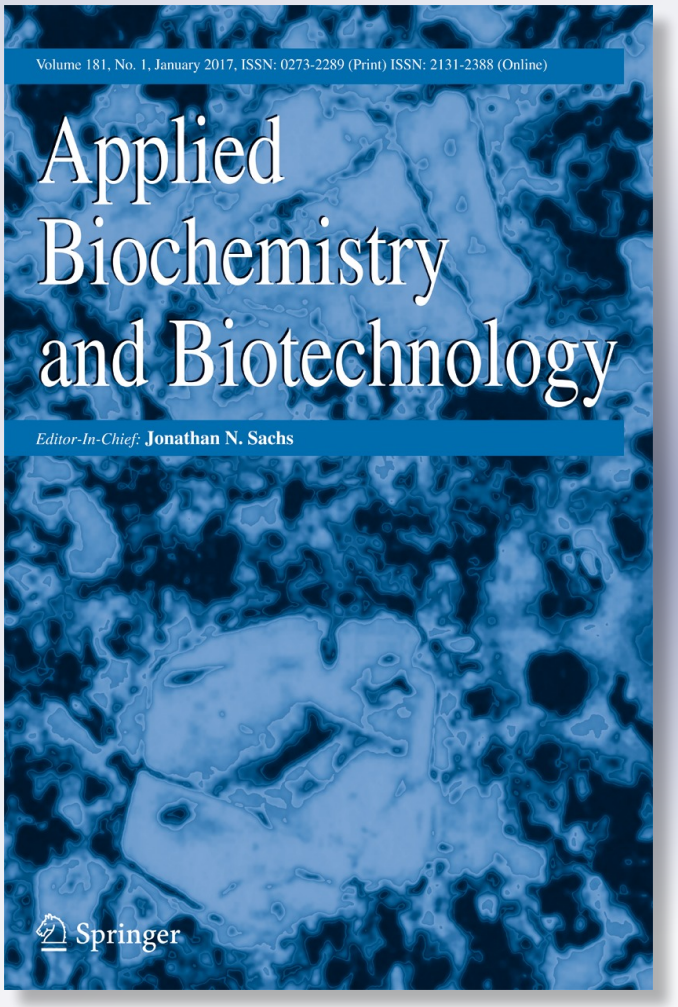

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The Environmental Issues of DDT Pollution and Bioremediation: a Multidisciplinary Review

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Abstract DDT (1,1,1-trichloro-2,2-bis(4-chlorophenyl) ethane) is probably the best known and most useful organochlorine insecticide in the world which was used since 1945 for agricultural purposes and also for vector-borne disease control such as malaria since 1955, until its banishment in most countries by the Stockholm convention for ecologic considerations. However, the World Health Organization allowed its reintroduction only for control of vector-borne diseases in some tropical countries in 2006. Due to its physicochemical properties and specially its persistence related with a half-life up to 30 years, DDT linked to several health and social problems which are due to its accumulation in the environment and its biomagnification properties in living organisms. This manuscript compiles a multidisciplinary review to evaluate primarily (i) the worldwide contamination of DDT and (ii) its (eco) toxicological impact onto living organisms. Secondly, several ways for DDT bioremediation from contaminated environment are discussed. For this, reports on DDT biodegradation capabilities by microorganisms and ways to enhance bioremediation strategies to remove DDT are presented. The different existing strategies for DDT bioremediation are evaluated with their efficiencies and limitations to struggle efficiently this contaminant. Finally, rising new approaches and technological bottlenecks to promote DDT bioremediation are discussed.

Keywords DDT · Pollution · Toxicity · Ecotoxicity · Biodegradation · Bioremediation

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Abbreviations

BRF	Brown-rot fungi
CBA	Chlorobenzoic acid
DBP	4,4′-dichlorobenzophenone
DDA	2,2-bis(*p*-chlorophenyl) acetate
DDD	1,1-dichloro-2,2-bis(*p*-chlorophenyl) ethane
DDE	1,1-dichloro- 2,2-bis(*p*-chlorophenyl) ethylene
DDM	Bis(4′-chlorophenyl)methane
DDMU	1-chloro-2-2-bis-(4′-chlorophenyl) ethylene
DDMS	1-chloro-2,2-bis(4′-chlorophenyl) ethane
DDNU	1,1-bis(4-chlorophenyl) ethylene
DDNS	2,2-bis(*p*-chlorophenyl) ethane
DDOH	2,2-bis(4′-chlorophenyl) ethanol
DDT	1,1,1-dichlorodiphenyl trichloroethane
o,*p*′-DDT	1,1,1-dichlorodiphenyl trichloroethane
p,*p*′-DDT	1,1,1-dichlorodiphenyl trichloroethane
DDTs	DDT and by-products
EPA	Environmental Protection Agency
LC_{50}	Lethal concentration 50 %
LD_{50}	Lethal dose 50 %
nRBT	Not readily biodegradable
OCP	Organochlorine pesticides
RBT	Readily biodegradable
UNEP	United Nations Environment Programme
WHO	World Health Organization
WRF	White-Rot Fungi

Introduction

In the past few decades, large quantities of xenobiotics such as chlorinated aromatic compounds have been released into the environment for industrial, agricultural, and public health purposes. Some chemicals are persistent and/or have ecotoxic properties. One of the most common chemicals is DDT [1,1,1-trichloro-2,2-bis(p-chlorophenyl) ethane] and its metabolites DDE [1,1-dichloro-2,2-bis(*p*-chlorophenyl) ethylene], and DDD [1,1-dichloro-2,2-bis(*p*-chlorophenyl) ethane] (Fig. 1), which have been used extensively in recent decades. Technical-grade DDT that was used as a pesticide is composed of 14 chemical compounds (DDTs), with 65–80 % of the active compound *p*,*p*′-DDT; the other components are distributed as follows: 15 to 21 % of *o*,*p*′-DDT (1,1,1-tricholoro-2-(*p*-chlorophenyl)-2-(*o*-chlorophenyl) ethane), up to 4 % *p*,*p*′-DDD, and up to 1.5 % of 1-(*p*-chlorophenyl)-2,2,2-trichloroethanol (Fig. 1) [1]. *p*,*p*′-DDT and *o*,*p*′-DDT are the most persistent compounds of DDTs, with a reported half-life between 2 to 15 years. Due to their toxicity, hydrophobicity, and bioaccumulative properties, DDTs have been classified in the national priority list of environmental pollutants by the US Environmental Protection Agency (EPA) as priority-persistent organic pollutants and endocrine-disrupting chemicals [2, 3].

DDT was the first synthetic insecticide to be developed and has been used worldwide since the 1940s. DDT was extensively used as an organochlorine insecticide for agricultural crops

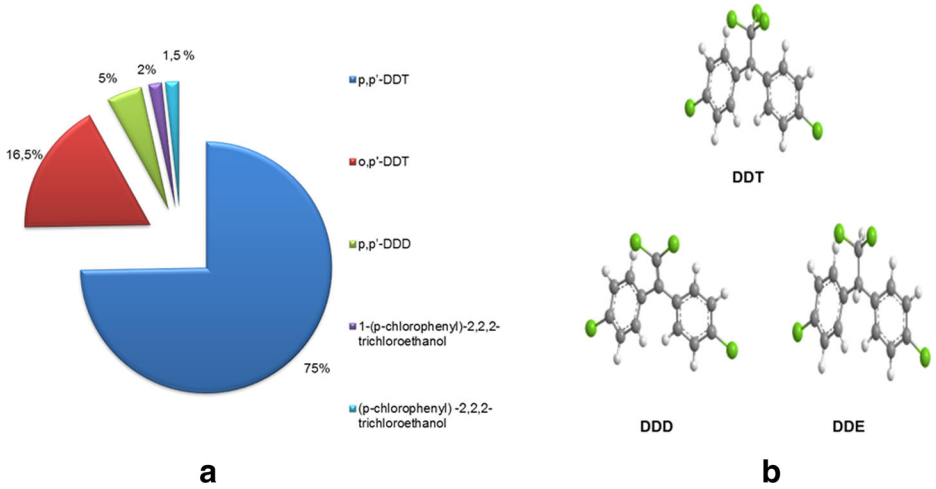


Fig. 1 Chemical composition of DDT from Metcalf (**a**) and 3D representation of DDT and its metabolites DDE and DDD (**b**) [1]. *White* hydrogen atoms, *red* oxygen atoms, *green* chlorine atoms, *black* carbons atoms

and control of vector-borne diseases, such as typhus and malaria [4, 5]. In addition, DDT was used in antifouling paint on fishing ships [6] and in industry for its transformation into dicofol, a miticide product [7].

Although DDTs have been first banned in USA since 1972, from 1945 to 1972, DDT was used for agriculture, in forests, for home use, and for controlling malarial diseases (Fig. 2). Approximately 400,000 t of DDT were used annually worldwide (70–80 % of which was used for agriculture). The United Nation Environment Programme (UNEP) estimated that world-wide consumption of DDT between 1950 and 1963 was approximately 175,000 t/year. In China, DDT was the most widely used pesticide from the 1950s to 1983. The total production of DDT was approximately 0.4 million tons, which accounted for 20 % of the total pesticides world production [8, 9]. The peak use of the compound was 550,000 t/year in 1970, which dropped to 68,800 t/year between 1971 and 1996. Moreover, between 2000 and 2010, DDTs consumption decreased from 19,677 to 4822 t/year [10, 11]. Numerous reports have noted that DDT is still produced in a few countries, mainly India (the major producer), Mexico, Italy, China, and Indonesia [12, 13].

During the Stockholm Convention on Persistent Organic Pollutants in 2001, there was an intense debate in the UNEP concerning the ban of DDTs. Despite the fact that DDTs are hazardous substances, they are the sole successful strategy to control vector-borne diseases in many countries, especially those in Africa, Asia, and Latin America in which malaria is endemic. Therefore, the World Health Organization (WHO) announced in 2000 that DDT is still needed to control malaria; that's why, it was reintroduced only in September 2006 [13] (Fig. 2).

DDTs reintroduction in Asia and Africa resulted in an immediate decrease in the number of reported malaria cases, from 42,000 in 2000 to less than 2100 in 2002. That explains why DDT is still used extensively in developing countries in Asia and Africa for vector control as well as in China, India, and Vietnam [14–17].

The purpose of this review is to evaluate the persistence of DDT worldwide and to assess its toxicity and ecotoxicity as well as to review its impact on human health and the environment.

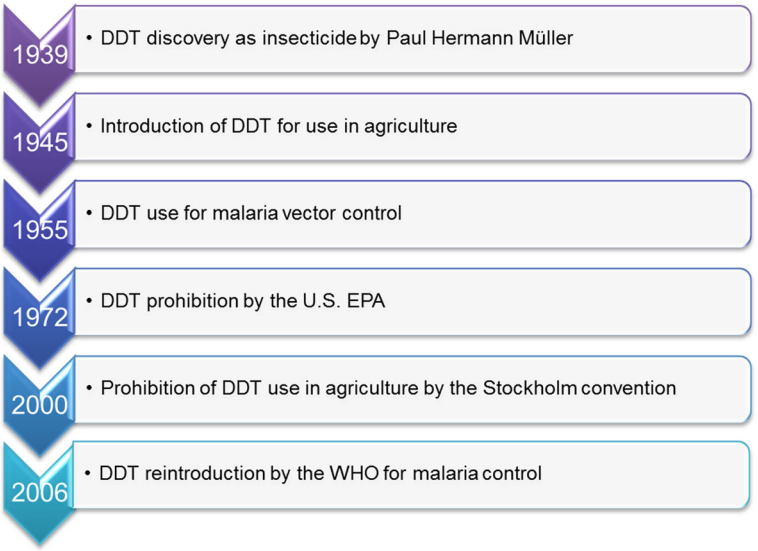


Fig. 2 Evolution of the use of DDT worldwide [13]

Additionally, we will review the possibility for and the pathways of complete biodegradation of DDT by microbial communities isolated from contaminated sites as well as focus on technologies designed to decontaminate polluted areas.

Distribution and Persistence of DDT

Due to its low solubility in water (log Kow DDT = 6.9), DDT has a strong affinity for suspended particulate matter in water, which can serve as repositories for DDT compounds and notably promote their stability and resistance to biodegradation [18]. According to its common use conditions, only 10 % of DDT used reaches its biological targets, which leads to an important release of this compound in the environment [19].

DDTs pollution in both soil and sediment can reach up to 1600 mg/kg for some highly contaminated environments [20]; this is why DDT isomers and their metabolites can still be frequently detected in soils, sediments, surface water, and groundwater leading to contamination in benthic organisms and biomagnification along the food chain [21].

DDT in Water

After treatment of crops and forests to control insects, DDT was released in large quantities into the environment and especially in water. DDTs may also evaporate from contaminated soil into the air and can then be deposited on surface water. As a result of this cycle of evaporation and deposition, DDTs can contaminate many environments that have never been treated. DDTs reach surface waters and groundwater by runoff or in some cases by direct application [10, 22–24]. In surface water, DDTs will become attached to solid particles and be deposited in the sediment where they may be taken

up by fish. In the USA, DDT has been detected in surface water samples at a median level of 1 ng/L [25, 26]. However, the highest levels of DDTs-contaminated water were detected in Nigeria, China, and India:

- In Nigeria, Asogwa and Dongo have estimated that over 130,000 tons of pesticides are applied every year in agriculture and to stop vector-borne diseases [27]. Consequently, 120 ng/L of DDTs have been recently detected in water samples [28].
- In China, according to Guo et al. [29], between 1988 and 2002, the amount of dicofol (which contains 25 % DDT) used was nearly 9000 tons, and in 2003, more than 14 tons of dicofol was directly applied in the Pearl River Delta [30]. Additionally, many boats use an antifouling paint containing DDT. As a result, most of the areas that were investigated had high levels of DDT contamination. The concentration of DDTs ranged between 1 and 250 ng/L, and the highest concentration exceeded 250 ng/L in the Taihu Basin [31].
- In India, DDT has been heavily applied in several areas. Water concentrations ranged from 3.18 ng/L in the Tighra River to 5794 mg/L in the Bhopol River [32, 33].

Although DDT has been detected in water, this matrix is not considered as optimal to assess its environmental contamination and consequently, risks posed by this chemical [25] according to the European Commission [34]; the recommended matrices for the assessment of DDT contamination are sediment and biota.

DDT in Soil and Sediment

Residues of DDTs compounds have been detected in soils in different regions worldwide, including areas distant from human activities such as the Polar Regions [35, 36]. Indeed, due to its volatility and persistence in the air, DDTs are subjected to long-range atmospheric transport. Therefore, released DDTs in the tropical and subtropical areas could be dispersed through air and water, and tend to be redistributed on a global scale [36].

Consequently, of its historic use, DDT has accumulated in soil and river sediments, and high levels have been found in these compartments ranging from 0.0086 to 1600 mg/kg [20, 21, 37]. For this reason, DDT and other organochlorine pesticides (OCP) have also been found in sediments [38, 39] from the lagoon of Bizerte, Tunisia, and Tianjin, China where *p,p′*-DDT was found to be the dominant compound, and the *o,p′*-DDT/*p,p′*-DDT concentration ratio was also found to be very high. In these ecosystems, DDTs concentrations ranged from 0.0115 to 0.93 mg/kg, which is less than the lethal concentration 50 % (LC_{50}) and lethal dose 50 % (LD_{50}) for fish [40–42]. Moreover, after the ban on the use of DDT in several regions of the world, several stocks have been established (Fig. 3). According to the DDTs embargo, several stocks of DDTs were created worldwide and account for more than 4000 t for a total of 897 known sites.

DDT in Marine Organisms

Aquatic contamination of the ecosystem is currently one of the most severe environmental problems. Because DDT and its metabolites are so hydrophobic, they are absorbed into organic particles of sediment where they can persist for many years (Table 1).

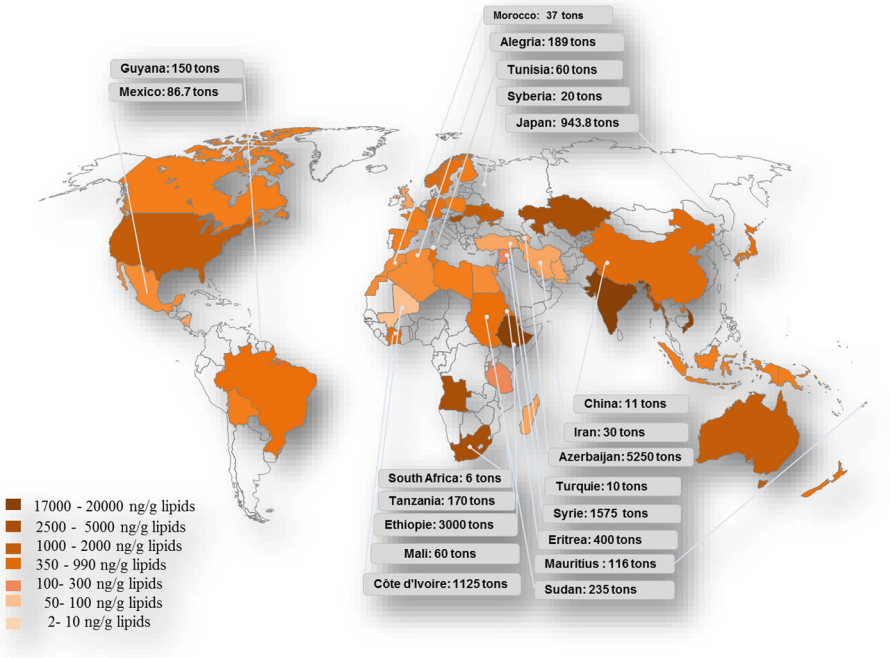


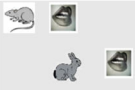
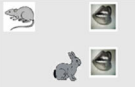
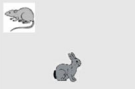
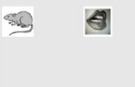
Fig. 3 The location of the main reservoirs of DDT worldwide and plasmatic concentrations of DDE in human adipose tissue [from 43–54] (Figure from the author)

In these environments, DDTs can accumulate in sediment-dwelling organisms and persist after sedimentation to the sea floor. DDT can then be transferred to higher trophic levels through biomagnification of its metabolites, mainly under the form of DDE [55–59]. For these reasons, many studies have recently been carried out assessing DDTs in aquatic ecosystems [40, 60–62]. Residues of DDT and its metabolite DDE have also been found in fish and marine mammals [63, 64]. In top-level predators, DDTs were measured at high concentrations that have a risk for human health [65], with levels ranging from 3 to 50 ng/g in various countries [46, 65–68] (Fig. 3). Obviously, these concentrations are subject to fluctuations that are due to physiological modifications of microorganisms and to the changes in water conditions.

Table 1 The half-lives of DDTs in different environments

DDTs	Environment				References
	Soil	Sediment	Water	Atmosphere	
DDT	4–30 years	1–4 years	26–56 days	1.5–3 days	[25]
ODE	151–672 days	-	1–6 days	17 h − 2 days	[25, 42]
DDD	160 days	-	190 years	4 days	[25]

Table 2 The relative toxicities of DDT and its main by-products (LC_{50} lethal concentration 50 %, LD_{50} Lethal dose 50 %, *INERIS* Institut National de l'Environnement Industriel et des Risques; http://www.ineris.fr/, *EPIWIN* Estimation Programme Interface. http://www.epa.gov/oppt/exposure/pubs/episuitedl.htm). *white* hydrogen atoms, *red* oxygen atoms, *green* chlorine atoms, *black* carbons atoms

Chemical substance					Toxicity			Ecotoxicity LC50 (µg/l)		References
Name Log KOW	Formula	Structure 3D	CAS Number	Molecular weight g.M-1	Species Rat Rabbit	Route	LD50 mg/Kg	Fish	Invertebrate	
1,1,1-dichloro Diphenyltrichloro Éthane (DDT) 6.9	$C_{14}H_9Cl_5$		50-29-3	354.49			113 1577 400	*Gambusia magna* 2 (48h) **19 (96 h)**	*Daphnia magna* 6.5 (48h) **13 (48 h)**	10 71
1,1-dichloro-2,2-bis(p-chlorophenyl) ethylene (DDE) 6	$C_{14}H_8Cl_4$		72-55-9	318.03			502 96	*Gambusia magna* 0.9 (48h) **15 (96 h)**	*Daphnia magna* 6 (48h) **14 (48 h)**	71
1,1-dichloro-2,2-bis(p-chlorophenyl) Ethane (DDD) 5.78	$C_{13}H_8Cl_2O$		72-54-8	320.05			1200 4000	*Gambusia magna* 0.06 (48 h) **71 (96 h)**	*Daphnia magna* 6 (48h) **87 mg/l (48 h)**	10 72 71
4-Chlorophenyl actetate 6.68	C_8H-ClO_2		876-27-7	170.59			No data	*Gambusia magna* **16.6 mg/l (96 h)**	*Daphnia magna* **33,7 mg/l (48h)**	10
4,4'-dichloro-Benzophenone (DBP) 4.44	$C_{13}H_8Cl_2O$		90-98-2	251.11			**200**	*Gambusia magna* **1.33 mg/l (96h)**	*Daphnia magna* **0.95 mg/l (48h)**	10
1-chloro-2-2-bis-(4'-chlorophenyl) ethylene (DDMU) 5.81	$C_{14}H_9Cl_3$		1022-22-6	283.58			**2700**	*Gambusia magna* **0.036 mg/l (96h)**	*Daphnia magna* **0.037 mg/l (48h)**	10

The EPA estimates that drinking 2 L of water per day containing 0.59 ng/L of DDT and eating 6.5 g of fish per day (containing 0.59 ng/L of DDT) would be associated with an increased cancer risk of one in one million [69].

Environmental and Human Hazards of DDT

Relative Toxicity of DDT and Human Health Hazards

Due to their high log Kow (Table 2), and their high chemical stability, DDTs tend to bioconcentrate in adipose tissue and bioaccumulate in the food chain [71–74].

Human exposure to DDT can occur by several routes, ingestion being its main source, particularly consumption of seafood from contaminated areas [75, 76]. The second source of the exposure takes place through skin absorption or through respiration, especially for workers

in agriculture, or via vaporization for malaria control. Another transmission route is accidental contamination at the workplace [2, 77, 78]. All these factors explain the relatively high levels of DDT and its metabolite DDE in adipose tissues, blood plasma, liver, brain, placenta, and breast milk [2, 79–81] and these concentrations have been extrapolated to obtain an idea of the overall environmental pollution by DDT (Fig. 3).

DDT exposure raises a serious risk to human health which is often accompanied by many harmful effects; neurological and immunodeficiency effects are the most commonly reported [10, 82], but also DDT and its derivatives are recognized as endocrine-disrupting chemicals [83]. Numerous studies have also demonstrated the carcinogenicity of DDT and its main metabolite, DDE. This was determined from multiple experimental and epidemiological studies. Many types of cancer were associated with contamination by DDT: brain cancer, pancreatic cancer, breast cancer, prostate cancer, and also testicular cancer for concentrations between 50 and 250 mg/kg [10, 84–86]. The first liver tumor in mice was observed at 50 weeks after exposure to 250 mg/kg of DDT and at 65 weeks after exposure to a concentration of 2, 10, or 50 mg/kg [87].

It is also known that DDTs compounds enhance breast cell growth and increase both tumor production and unscheduled DNA synthesis [88]. DDT can also inhibit enzymatic activities such as acetylcholinesterase [89]. The possible genotoxicity of DDT has been reported in both in vitro and in vivo studies [90]. Various studies have also shown that DDT and its metabolites (DDE and DDD) can inhibit gap junction and intercellular communications in human breast epithelial cells [91].

Ecotoxicity of DDT in Environmental Organisms

Exposure to DDT affects organisms in contaminated environments. The bioconcentration factor of DDT has been reported in aquatic species by many studies, and it is estimated to be 1000 to 1000,000 [92]. The ecotoxicity of DDT and its metabolites and degradation products were studied (Table 2). The principal metabolites were DDE, DDD, DDMU [1-chloro-2-2-bis-(4′-chlorophenyl) ethylene], and DBP [4,4′-dichlorobenzophenone]; they are highly toxic to many aquatic vertebrate and invertebrate species, and the least toxic product is chlorophenyl acetate. Compounds such as DDE and DDD were proposed to be more persistent than the parent compound DDT [54]. Their toxicity and ecotoxicity are higher than DDT [11, 93].

Considering the toxicity and ecotoxicity of DDT and its metabolites, it is necessary to address the environmental persistence of this insecticide. Moreover, the removal of DDT from contaminated sites has become an environmental priority and both physicochemical and biological remediation processes have been investigated.

Biodegradation of DDT

Biodegradation of DDT and its Metabolites

Biodegradation is a remediation method in which organic contaminants are degraded by biological means. Biodegradation of DDTs by microorganisms can lead to a wide variety of compounds, including some derivatives with a large set of chlorine atoms and a relative low hydrosolubility. DDTs biodegradation is generally incomplete and produces some easily

Table 3 Potential and predicted biodegradability of DDT and some metabolites (predicted half-life and biodegradation from EPIWIN: Estimation Programme Interface EPI Suite™ v4.11)

Molecule	Predicted half-life (Months)	Predicted for biodegradation (from EPIWIN)	Potential for biodegration	Biodegradability (from literature)	References
1,1,1-dichloro diphenyl trichloroethane (DDT)	1388	-Does not biodegrade fastly	-Does not biodegrade quickly -Aerobic degradation DDE -Anaerobic degradation DDD -Weeks	nRBT	[93, 94]
1,1-dichloro-2,2-bis(p-chlorophenyl) ethylene (DDE)	1387	-Recalcitrant -Does not biodegrade	-Does not biodegrade quickly -Anaerobic pathway: DDMU, 4-chlorobenzaldéhyde -Weeks	nRBT	[95, 96]
1,1-dichloro-2,2- bis(p-chlorophenyl) ethane (DDD)	1384	-Recalcitrant -Does not biodegrade	-Does not biodegrade quickly -Anaerobic pathway: DDMU -Terminal metabolite of anaerobic degradation of DDT -Weeks	nRBT	[97]
4, 4′-dichloro Benzophenone (DBP)	1273	-Does no biodegrade quickly	-Does no biodegrade -Weeks	nRBT	[98]
1-chloro-2-2-bis-(4′-chlorophenyl) ethylene (DDMU)	1377	-Does no biodegrade quickly	-Does no biodegrade quickly Anaerobic pathway: 1-Chloro-2, 2-bis(4′-chlorophenyl)ethane (DDMS) -Weeks	nRBT	[98]
4-Chlorophenyl acetate	1	-Biodegrades quickly	-Biodegrades quickly -Integrated in the tyrosine pathway, yield 3,4-Dihydroxyphenylacetate -Days	RBT	[98]

RBT readily biodegradable; *nRBT* not readily biodegradable

Table 4 Diversity of DDT degraders from the bacteria and fungi kingdom (*, +, ¤ those strains are from the same consortium, *NA* not available) (in bold are strains degrading completely the DDT)

Kingdom	Initial compound	DDT strain	DDT final product	Biodegradation (%)	References
Bacteria	DDT	*Aerobacter aerogenes*	DDD	NA	[107]
	DDT	*Aeromonas hydrophila*	DDD, DDE, DDMU, DDNU, DBP	73	[108]
	DDT	***Alcaligenes eutrophus* A5**	DDD, DDE, 4-CBA	NA	[109]
	DDT	*Alcaligenes* sp. DG-5	DDD, DDE	88	[108]
	DDT	*Alcaligenes* sp.	DDD, DDE	NA	[110]
	DDT	*Arthrobacter* sp.	DDA	NA	[107]
	DDT	*Bacillus* sp.	DDA	90	[107]
	DDT	***Chryseobactedum* sp. *PYR2***	DDD, DDE, DDMU, DDMS, DBP, CBA, CPA	40	[105]
	DDT	*Enterobacter cloacae*	DDD, DDM	98	[111]
	DDT	*Eubacterium limosum*	DDD	NA	[112]
	DDT	*Flavimonas oryzihabitans*	DDE, DDMU, DDOH	NA	[102]
	DDT	*Hydrogenomonas* sp.	4-chlorophenylacetate	NA	[113, 114]
	DDT	*Klebsiella pneumonia*	DDD, DDE	40	[112]
	DDE	***Pseudomonas acidovorans M3GY***	4-CBA, 4-CPA	86	[115]
	DDT	*Pseudomonas aeruginosa*	DDD	55–99	[116]
	DDT	*Pseudomonas fluorescens*	DDD, DDE	NA	[117]
	DDT	*Pseudomonas putida*	DDD, DDMS, DBP	73	[118]
	DDT	*Pseudomonas* sp.	DDD, DDE	100	[119]
	DDT, DDD, DDE	*Pseudoxanthomonas* sp.	NA	60	[120]
	DDT	*Rohodococcus* sp. Strain IITR03	DDD, DDE, DDNU	75	[119]
	DDT	*Serratia marcescens* DT-1P	NA	82	[121]
	DDT	*Sphingobacterium* sp.	DDD, DDE, DDMU, DDNS, DDA, DBP	28.48	[122]
	DDT	*Staphylococcus* sp.	DDD	69	[123]
	DDT	*Stenotrophomona* sp. DDT-1	DDD, DDE, DDMU, DDNU, DDOH, DDA, DDM	100	[106]
	DDT	*Stenotrophomona* sp. D-1	DDE	NA	[124]
	DDT	*Trichoderma viridae*	DDA	NA	[110]
Fungi	DDT	*Aspergillus niger*	DDD, DDM	NA	[125]
	DDT	*Aspergillus sydowi* Ce15 *	DDM	58	[126]
	DDT		DBP	58	[126]

Table 4 (continued)

Kingdom	Initial compound	DDT strain	DDT final product	Biodegradation (%)	References
		Aspergillus sydowii Ce19 *			
	DDT	*Bionectria sp.* Ce5 *	DDM, DBP	58	[126]
	DDT	*Boletus eduli* +	DDD, DBP	60	[127]
	DDT	*Cladosporium* sp. AJR[3] 18501	DDD	21	[128]
	DDT	*Daedalea dickinsii*	DDD, DDE, DBP	81	[93, 129]
	DDT	*Fomitopsis pinicola*	DDD, DDE, DBP	84	[93, 130]
	DDT	*Fusarium moniliforme* ¤	DDD, DBP	79.5	[130]
	DDT	*Fusarium oxysporum* ¤	DDD, DBP	94.4	[130]
	DDT	*Fusarium solani*	DDD, DDE, DDOH, DBP	34	[131]
	DDT	*Gloeophyllum trabeum*	DDT, DDE, DBP	87	[93]
	DDT	*Gomphidius viscidus* +	DDD, DBP	82	[127]
	DDT	*Glomus etunicatum*	DDD, DBP	80.3	[132]
	DDT	*Penicillium miczynskii* *	DDD	58	[127]
	DDT	*Penicillium raistrickii* *	DDD	58	[127]
	DDT	*Phanerochaete chrysosporium*	DDD, DBP	50	[99]
	DDT	*Plebia brevispora*	DDD, DDA, DBP, DBH	30	[94]
	DDT	*Plebia lindtneri*	DDD, DDA, DBP, DBH	70	[94]
	DDT	*Laccaria bicolor* +	DDD, DBP	60	[127]
	DDT	*Leccinum scabrum* +	DDD, DBP	60	[127]
	DDT	*Nectriam ariannaeae*	NA	75	[133]
	DDT	*Trichoderma* sp. *	NA	58	[127]
	DDT	*Xerocomus chrysenteron*	DDD, DDE, DBP	55	[104]

biodegradable compounds such as 4-chlorophenol-acetate and highly persistent compounds such as DDE, DDD, and DBP (Table 3).

In Vitro Biodegradation of DDT and Identification of Degraders

Numerous microorganisms and autochthonous inoculums have been isolated and characterized for their ability to metabolize DDTs, notably from contaminated sites [99–103]. However, until now and due to DDTs toxicity and complexity, only a partial degradation, generating organic by-products, has been found in the majority of microorganisms. Some cases of complete mineralization have been found in microorganisms which can metabolize the intermediates metabolites of DDT such as DDE and DDD [104–106].

Almost all microorganisms that have been described are potential DDT degraders. Among them, DDTs biodegradation abilities were found in a wide range of bacteria belonging to the phyla *Proteobacteria*, *Firmicutes*, and *Actinobacteria* (Table 4) with an efficiency of biodegradation ranging from 15 to 100 % [106, 113, 114, 117–119, 134].

Some strains, such as *Chryseobacterium* sp. PYR2 [105], *Alcaligenes eutropha* A5 [109], and *Pseudomonas acidovorans* M3GY [115], showed incredible ability to mineralize DDT compared with other environmental inoculums (Table 4). During DDT biodegradation, DDE and DDD are initially released with environmental inoculums in most cases. Thereafter, these molecules are themselves subjected to microbial biodegradation depending on the inherent capabilities of the microorganisms to metabolize these potential by-products [96]. Moreover, several biodegradation assays have also demonstrated that a broad range of compounds can be generated from the partial biodegradation of DDT, such as DDA [2,2-bis(*p*-chlorophenyl) acetate], DDOH [2,2-bis(4′-chlorophenyl) ethanol], DDNU [1,1-bis(4-chlorophenyl) ethylene], DDMS [1-chloro-2,2-bis(4′-chlorophenyl) ethane], and 4-chlorobenzoic acid (CBA) [109, 116, 121].

White-rot fungi and brown-rot fungi (respectively, WRF and BRF) are known to degrade various organic substances [98] both in cultured conditions and in the natural environment [115, 125, 133–136]. These abilities depend on their capacity to colonize these substrates and the secretion of numerous enzymes [136]. In the case of DDTs, some WRF were already reported as biodegrading strains (Table 4), e.g., *Boletus edulis*, *Gomphidius viscidus*, *Laccaria bicolor*, *Leccinum scabrum*, *Arbuscular mycorrhizal*, and *Phanerochaete chrysosporium* [125, 137, 138].

To enhance the fungal DDTs biodegradation abilities, Ortega and co-workers assessed a consortium of fungi for DDT degradation. The consortia consisted of *Aspergillus sydowii*

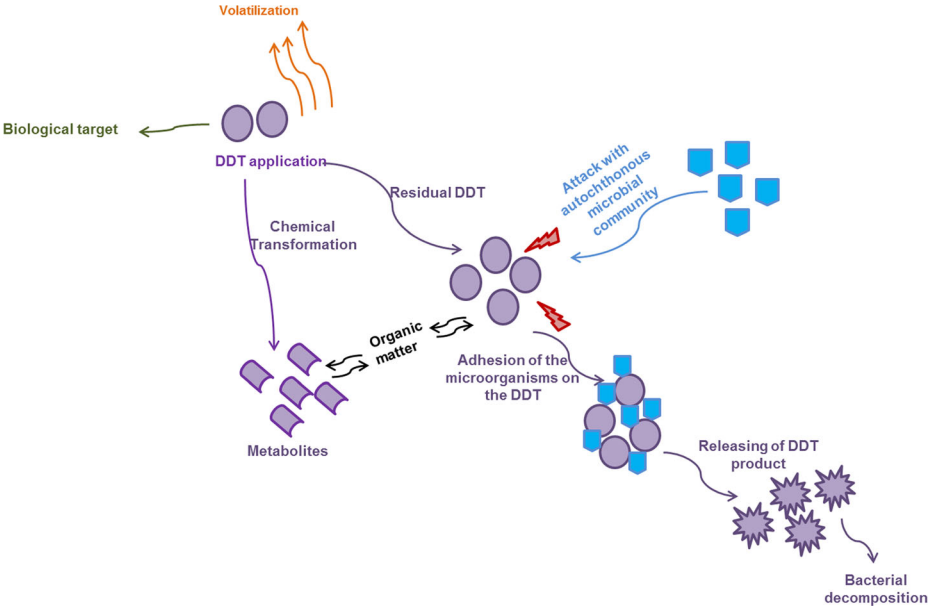


Fig. 4 DDT distribution in soil after its application (adapted from [19, 20])

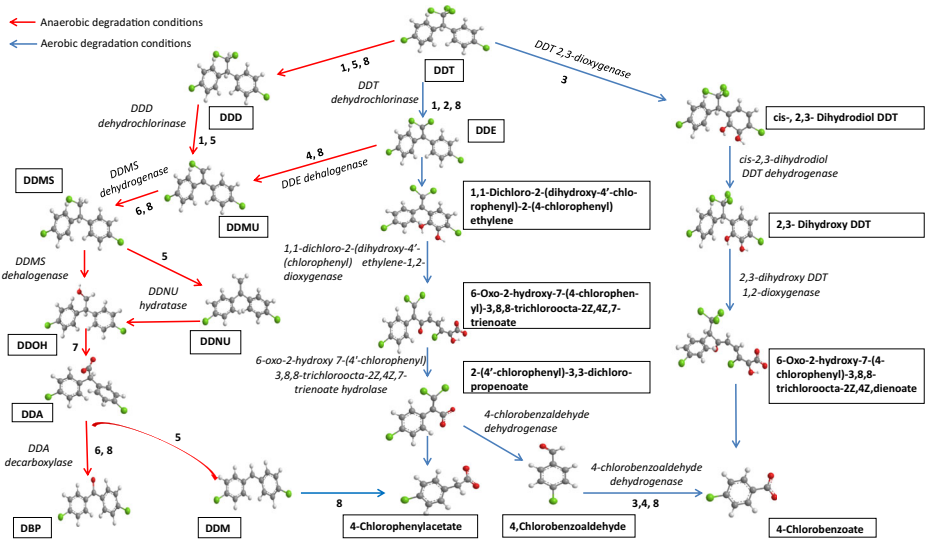


Fig. 5 Mechanistic scheme of DDT biodegradation under aerobic (blue) and anaerobic conditions (red) [95, 105, 108, 109, 113, 114, 116, 141, 142]. *White* hydrogen atoms, *red* oxygen atoms, *green* chlorine atoms, *black* carbons atoms. *DDT* 1,1,1-dichlorodiphenyl trichloroethane; *DDE* 1,1-dichloro-2,2-bis(*p*-chlorophenyl) ethylene; *DDD* 1,1-dichloro-2,2-bis(*p*-chlorophenyl) ethane; *DDMU* 1-chloro-2-2-bis-(4′-chlorophenyl) ethylene; *DDMS* 1-chloro-2,2-bis(4′-chlorophenyl) ethane; *DDNU* 1,1-bis(4-chlorophenyl) ethylene; *DDOH* 2,2-bis(4′-chlorophenyl) ethanol; *DDA* 2,2-bis(*p*-chlorophenyl) acetate; *DBP* 4,4′-dichlorobenzophenone; *DDM* bis(4′-chlorophenyl)methane. *1 Proteus vulgaris*, *2 Klebsiella pneumoniae*, *3 Ralstonia eutropha* A5, *4 Pseudomonas acidovorans* M3GY, *5 Aeromonas hydrophila*, *6 Pseudomonas putida*, *7 Bacillus* sp. *8 Chryseobacterium* sp. PYR2

Ce15, *Aspergillus sydowii* Ce19, *Bionectria* sp. Ce5, *Penicillium miczynskii*, *Trichoderma* sp., and *Penicillium raistrickii*. These fungi were able to grow in DDD concentrations ranging from 5.0 to 15.0 mg/L in solid or liquid medium with 58 % biodegradation of DDD after 14 days [126]. Although BRF has received relatively little attention from the scientific community for DDTs biodegradation compared with WRF, Purnomo and co-workers have reported that *Gloeophyllum trabeum* use hydroxyl radicals produced via the Fenton reaction for DDTs biodegradation, with 87 % degradation of DDT in only 12 days [93, 139].

Biodegradation Pathways

DDT biodegradation activity is achieved in a different manner in fungi than in bacteria. In addition to the fact that several enzymes devoted to biodegradation are found in a variety of microorganisms, it has been proposed that the ability to degrade DDT was due to the presence of enzymes that cleave the carbon-chlorine bond, a critical step in DDT biodegradation under physiological conditions [140] (Fig. 4).

The cleavage of carbon-chlorine bonds is a critical step in organochlorine degradation. Such cleavage may occur via two ways: by spontaneous dechlorination of an unstable intermediate and by enzymatic dechlorination where the carbon-chlorine bond cleavage is catalyzed by specific enzymes such as monooxygenases and dioxygenases [140]. DDTs

biodegradation is accomplished through several metabolic pathways occurring in aerobic or in anaerobic conditions (Fig. 5).

DDT Biodegradation under Anaerobic Pathways In bacteria, anaerobic degradation of DDT is achieved via a reductive dechlorination pathway, which can be divided into two types: co-metabolic and metabolic [99, 100, 143].

The metabolic conversion is carried out by halo-respiring bacteria [144]. The co-metabolic conversion is catalyzed by metal ion-containing enzymes such as coenzyme F430 and cobalamin (vitamin B12) as cofactors in anaerobic bacteria [143]. However, in fungi, the dechlorination is achieved in two different ways. In WRF, the reaction leads to a reduction of DDT to DDD: 1,1-dichloro-2,2-bis(*p*-chlorophenyl) ethane [94], except for specific biodegradation by *Phlebia* which is generally similar to anaerobic degradation by bacteria, producing DBP and DBH, but these metabolites are additionally hydroxylated, that results in ring cleavage [94]. Whereas in some species of BRF, the dechlorination leads first to DDE: 1,1-dichloro-2,2-bis(*p*-chlorophenyl) ethylene, with the exception of the biodegradation of *Fomitopsis pinicola* which produces DDD directly from DDT [93]. Thereafter, DDE is subsequently hydrogenated to DDD, the latter is dehalogenated to DDMU: 1-chloro-2-2-bis-(4′-chlorophenyl) ethylene [93, 129, 139, 145].

This pathway differs from the proposed pathways in WRF, particularly in the transformation of DDE to DDD [94] (Fig. 5). This is because BRF do not have the same metabolism; it is also possible that BRF uses an enzymatic system that differs from that of WRF.

DDT Biodegradation under Aerobic Pathway Several aerobic pathways for DDT metabolism have been proposed in different organisms [109, 116, 123]. The reports highlight the involvement of some common enzymes that are specific for 4-chlorobiphenyl degradation through an initial DDT oxidation at the *ortho* and *meta* position by dioxygenases [135]. Thereafter, DDT was dihydroxylated at the 2,3-position by biphenyl-2,3-dioxygenase via meta-cleavage, to yield a dihydrodiol-DDT derivative that undergoes meta-cleavage, ultimately yielding 4-chlorobenzoic acid (Fig. 5) [109, 135].

Considering the number of compounds involved, as well as the diversity of microorganism degrading DDT, we provide a DDT degradation pathway (Fig. 5). It is worth noting that aerobic and anaerobic metabolic pathways of DDT degradation by microorganism have many processes in common. This is why for some strains, specific metabolites of aerobic and anaerobic pathway were currently detected in the same culture and the same conditions such as for *Klebsiella pneumonia* and *Sphingobacterium* sp. [112, 122].

Genetic Biodegradation of DDT

The genetics and biochemistry of DDT biodegradation have been studied in few reports, and enzymes have been characterized for a few relevant bacteria and fungi including *Pseudomonas fluorescens* (Table 5). Despite the scientific efforts to characterize the full metabolic pathway, all the genes involved in the metabolism of DDT have not yet been elucidated. Genes that encode enzymes such as oxygenases and dehydrochlorinases have already been characterized. Among them is the *dhc* gene which encodes a protein that is responsible for the transformation of DDT to DDE by dehydrochlorination, the *rdh* gene for dechlorination of the DDT to DDD [106]. The hydrogenation and hydroxylation of DDMU to DDOH and the hydroxylation are

Table 5 Potential genes of DDT degraders

Degrader identification		Activity		Enzymatic activities						References
Kingdom	Strain	Expression	Oxygen requirement	Enzyme	Family	Molecular mass (Kda)	Optimal pH	Optima temperature (°C)	Genetic information	
Bacteria	*Pseudomonas putida* T5	Constitutive	Strictly aerobic	DDT-Dehydrohalogenase	Lyase	32	7	30	catA	[146]
	Bacillus sp.	Constitutive	Strictly aerobic	DDT-dehydrochlorinase	Lyase	36	7.2	37	–	[120]
	Pseudomonas aeruginosa	Constitutive	Strictly aerobic	DDT 2,3-dioxygenase	Oxygenase	43	7.3	30	xylE	
	Rhodococcus globerulus	Constitutive	Strictly aerobic	DDT 2,3-dioxygenase	Oxygenase	43	7.3	30	xylE	
	Klebsiella pneumonia	–	Strictly aerobic	DDT dehydrochlorinase	Lyase	36	7.2	37	–	
	Aeromonas hydrophila	–	Facultative anaerobic	DDT dehydrochlorinase	Lyase	36	7.2	37	–	[121]
	Rohodococcus sp. strain IITR03	–	Strictly aerobic	DDMS dehydrogenase	Lyase	37	7	37	–	
				DDT 1,2-dioxygenase	Oxygenase	42	7.3	30	–	
Fungi	*Phanerochaete chrysosporium*	Inductive	Strictly aerobic	DDD dehydrochlorinase	–	–	–	–	–	
				DDA decarboxylase	–	–	–	–	–	
	Fusarium solani	Inductive	Strictly aerobic	DDT dehyrochlorinase	Lyase	36	7.2	37	–	[147]
	Phlebia brevispora		Strictly aerobic	DDD dehyrochlorinase	Lyase	–	–	–	–	
			Strictly aerobic	DDD decarboxylase	–	–	–	–	–	

Table 6 Summary of studies on bioremediation of DDT-contaminated soil (ranging by references dates). (*NA* not available, *The treated soil in this case was cattle manure compost with artificial contamination with DDT)

Matrix (Initial DDTs concentration (mg/kg)	Traited volume	Strains	Method	Reactor volume	Duration of the process (efficiency of biodegradation)	Monitoring	References
Contaminated soil (NA)	10 g	Autochthonous microbial community	Batch flask 	250 mL	6 weeks (24.5)	- HPLC analysis -The effect of soil water content on the 14C DDT biodegradation - Estimation of released C02	157
Contaminated soil (82)	400 g	*Phanerochaete Chrysosporum*	Batch flask 	250 ml	4 weeks (75)	- GC–ECD analysis of DDT, DDD and DDE - CH4 and CO2 monitoring	158
Contaminated soil (5)	10 g	Autochthonous microbial community	Batch flask 	NA	12 Weeks (34)	- GLC Analysis - Effect of bioaugmentation with *Fusarium solani*	135
Contaminated soil (10)	10 g	*Ulva* sp and *Gelidium* sp	Batch flask 	150 mL	6 weeks (80)	- HPLC analysis - Microbial community analysis - Effect of seaweed addition concentration on DDT biodegradation	75
Contaminated soil (1600)	150 g	Autochthonous microbial community	Batch flask 	2 L	24 hours (50)	- GC-ECD analysis - Evaluation of H_2O_2 concentration influence on the degradation of DDT	60
Contaminated soil (32.9)	3 g	Granular sludge	Batch flask 	120 mL	8 weeks (75)	- GC-ECD analysis - Effect of temperature on DDT biodegradation	159
Contaminated soil (0.427)	10 m2	Autochthonous microbial community	In situ	NA	13 Weeks (27.8)	- GC-ECD analysis - Effet of bioaugmentation with *Sphingobacterium* sp. - Effect of temperature and additional carbon sources on DDTs biodegradation	123
Contaminated soil (0.25)	2 g	*Pleurotus ostreatus*	Batch flask 	100 mL	4 weeks (48)	- HPLC (for DDT) and GC-MS (for metabolites) analysis - Analysis of ligninolytic enzyme activities - Mineralization of [U-14C]DDT	160
Cattle manure compost with artificially contamination with DDT (0.25)	5 g	Autochthonous microbial community	Composting reactor 	NA	4 weeks (88)	- HPLC Analysis - Effect of temperature in the rate of biodegradation of DDT in compost - Microbial analysis	133
Contaminated soil (4.863)	500 g	Autochthonous microbial community	Batch flask 	NA	4 Weeks (28.1)	- GC-ECD analysis - Effect of cometabolism with laccase extraite from *Panus conchatus* on bioremediation process - Examine the effects of different atmosphere conditions (air, oxygen and nitrogen, pH) on DDT remediation	139
Contaminated soil (20)	500 g	*Pseudoxanthomonas Sp.*	Batch flask 	250 mL	3 weeks (100)	- HPLC analysis	125
Contaminated soil (0.25)	2 g	*Gloeophyllum trabeum*	Batch flask 	100 mL	4 weeks (64)	- HPLC analysis - Effect of addition Fe_{2+} on fenton reaction	151
Contaminated soil (22.35)	100 g	Autochthonous microbial community with *Phanerochaete chrysosporium*	Batch flask 	1 L	3 weeks (60)	- GC-ECD analysis - Effect of bioaugmentation with *Phanerochaete chrysosporium on DDTs biodegradation*	137
Contaminated soil landfill (NA)	50 g	Granular sludge	Batch flask 	120 mL	8 weeks (99)	- GC-MS analysis - Effect of surfactant addition in DDT biodegradation	161
Contaminated soil (4.272)	1 kg	*Flammulina velutipes*	Batch flask 	100 mL	4 weeks (66.82)	- GC-MS analysis - Effect of cometabolism with laccase (6 U g−1 soil) in DDT biodegaradtion	162
Contaminated soil (82.5)	5 g	Autochthonous microbial community	Hermetic flask 	125 ml	7 weeks 67.59	- HPLC analysis - Effect of addition of cosubstrates (phenol, hexane and toluene) in biodegradation rate of DDT, DDE and DDD - The CO2 producing - Bacterial communities evaluation - Volatile cosubstrates degradation	163
Contaminated soil (15)	1 Kg	Autochthonous microbial community	Batch flask 	2 L	15 weeks (30)	- GC-ECD analysis - Transmission electron microscopy (TEM) analysis. - pH and TOC monitoring	170
Contaminated soil (0.28)	10 g	Autochthonous microbial community	Microcosm and mesocosm 	NA	35 weeks (87)	- GC-ECD analysis - The impact of selected electron donors and electron acceptors on the anaerobic biodegradation of DDT	165
Contaminated soil (12.9)	NA	Autochthonous microbial community	Aerated composting reactor 	200 L	10 days (80.8)	- GC-ECD analysis - Temperature- and O2-monitoring - Microbial analysis	166
Contaminated soil (100)	2 kg	Autochthonous microbial community	Microcosm 	9 L	8 weeks (94)	- GC-ECD analysis - Microbial analysis - Effect of biostimulation with surfactant addition - Toxicity test - Physical and chemical properties of soil - Scanning electron microscopy of the soil	167
Contaminated aged soil (20)	NA	Autochthonous microbial community	Mesocosm 	NA	13 Weeks (85.4)	- GC-ECD analysis - Microbial diversity analysis with qPCR - Effect of biostimulation with nutrients and plants (*Orychophragmus violaceus*) - Scanning of the abundances of *linA* genes	168

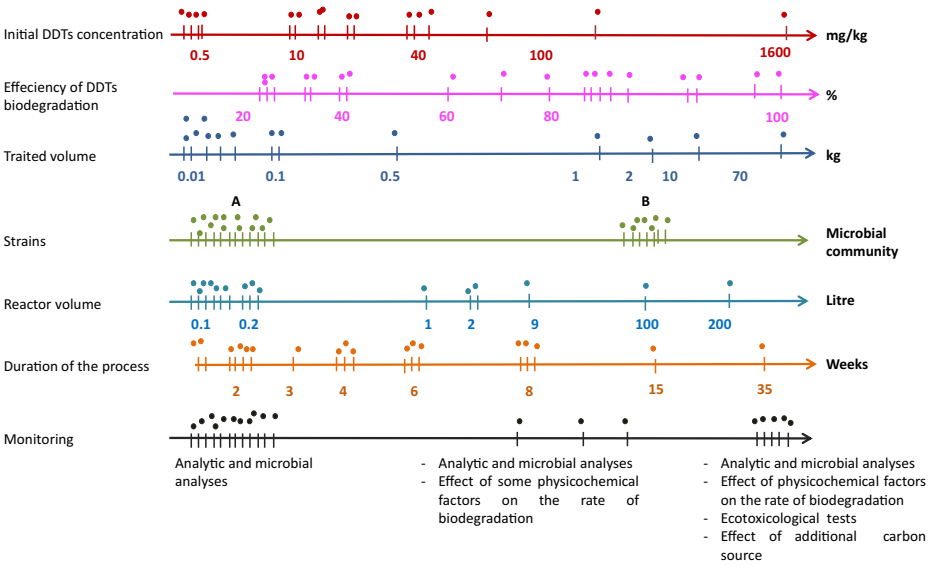


Fig. 6 Scatter representative of the main publications on the bioremediation process for DDT-contaminated soil. (*A* autochthonous microbial community; *B* bioaugmentation with other isolated strains)

encoded by the *sds* and the *dhg* genes [106], whereas the transformation of DDNU to DDOH by hydroxylation are mediated by the *hdt* gene and the *dcl* gene for the degradation of DDA to bis(4′-chlorophenyl)methane (DDM) by decarboxylation [106, 148].

Bioremediation of DDT from Lab Scale to Field Scale

A new, environmentally friendly strategy can be developed through the use of biological sources that can degrade this class of xenobiotic as shown in Table 4. For this, the use of microorganisms capable of accumulating, detoxifying, and metabolizing organic and inorganic contaminants, as well as utilizing them as a nutrient source, appears to be a prerequisite [149–151]. Several efforts have been undertaken using various bacterial and fungal strains (belonging both to WRF and BRF) that have been shown to enhance biodegradation processes against DDT (Table 6). These investigations have been performed both under laboratory conditions and in contaminated sites using both pure and mixed cultures [133, 136].

Enzyme-Linked Treatments

Yuechun and co-workers have used laccase extracts from a white-rot fungus for the bioremediation of DDT-contaminated soil [165]. After 25 days of incubation, 69 % of DDTs in soil was removed by laccase under an oxygen atmosphere [165]; later, another team demonstrated that the co-remediation of DDT in soil by the addition of white-rot fungi and laccase was more rapid and efficient than reduction using only white-rot fungi or laccase separately by nearly 14 and 16 %, respectively [158]. An optimal ratio of 5 mL/15 g of soil and 6 U of laccase per

gram of soil were used, as shown by a reduction in DDTs of 66.82 % at 28 °C [158]. Given the knowledge of the enzymatic mechanisms of the DDT degradation (Fig. 5), further optimization experiments may improve the productivity of these enzymes.

Bioremediation Using Living Cells

Due to the DDTs persistence in the environment and despite the presence of potential catalysts, knowledge of the conditions required to achieve bioremediation is an important issue. Numerous studies have demonstrated the feasibility of bioremediation of contaminated soil for organic pollutants other than DDT [160, 166]. However, numerous studies have investigated the applicability of DDT bioremoval using autochthonous microbial community. An overview is presented in Fig. 6 below and in Table 6, detailing each study addressing bioremediation of DDT. Overall, eight parameters were taken into consideration: the initial concentration of DDT, the efficiency of removal, the volume and microorganisms used, the method including the reactor volume, the follow-up duration, and especially the monitoring (Fig. 6).

The first studies on DDT bioremediation from contaminated soil appeared in the 1990s, and the studies were carried out in a batch flask reactor with few quantities of polluted soil ranging from 2 to 10 g of soil [145, 152] with processing times between 24 h and 8 weeks [154]. The monitoring was performed on only the microbial community and analytic analysis (Table 6). Several years later, the treatment process for DDT-polluted soil had improved, and a mesocosm with a treated volume up to 70 kg was used [105]. The monitoring was also more sophisticated—the effect of different physicochemical factors in the rate of DDT biodegradation, toxicity, and soil modifications following the remediation process were investigated [163]. The latest study is the only one to evaluate the toxicity of the treatment process using the luminescent bacteria *Vibrio fischeri* (Table 6). They showed a decrease in the toxicity of treated soil compared with the control [163].

DDT initial concentration ranged from 0.25 to 1600 mg/L [59, 129]. This is correlated with the efficiency of biodegradation, which ranged from 24.5 % for lower initial DDT concentrations to 100 % with an initial concentration of 20 mg/L [120, 152].

To remove DDT residues found in aged polluted soil, most bioremediation studies have been performed with microcosm-scale incubation in the laboratory to define crucial physicochemical factors and biological strains to determine the feasibility and sustainability of the process [167]. It was difficult for microorganisms to degrade DDT when it was used as the sole carbon source [168]. For this reason, in 2010, Wang and co-workers [120] isolated a strain that was denominated *Pseudoxanthomonas* sp. Wax, which was found to be an efficient degrader. They used temperatures ranging from 20 to 37 °C, with initial pH values ranging from 7 to 9, and 100 mg/L glucose as co-substrate. Under these conditions, the strain could degrade over 95 % of the total DDT, at an initial concentration of 20 mg/L in 3 days, and could degrade over 60 % of the total DDT, at an initial concentration of 100 mg/L in 6 days [120]. In both sterile and non-sterile soils, bioaugmentation with *Pseudoxanthomonas* sp. Wax cells at 10^8 CFU/g resulted in the complete removal of 20 mg/kg DDT after 3 weeks of incubation [120] (Table 6). The transformation could also be carried out by a combination of several microbial species such as *Sphingobacterium* sp. and *A. eutropha* [109, 122].

Another study has demonstrated the remediation of DDT in a compost reactor during the composting process (Table 6) where degradation of DDT at 60 °C was the most effective. Therefore, 14 strains of fungi were isolated and identified from this compost; most of them

Table 7 Summary of studies on field bioremediation of DDT-contaminated soil (*NA* not available)

Bioremediation method	Site of application	Principle concerns	Impact on removal rate (%)	References
Xenorem ®: Anaerobic-aerobic composting	Savannah River	It employed soil with large amounts of the waste to make what was effectively a huge compost heap. The soil is aerated every few weeks to enhance the effectiveness of homogenizing large quantities of soil with organic composting materials which encourage microbial communities and provided both the nutrients and a cycle of alternating anaerobic and aerobic conditions for local bacteria to degrade the pesticides	95	[172, 173]
Daramend®	Superfund Site, Montgomery, Alabama	It has been used to treat soils and sediments containing low concentrations of pesticides such as DDT, it utilizes organic amendments to create aquatic microsites where native microorganisms can grow and degrade contaminants	68	[174]
BioSite®	Arctic and the Antartic	Soil is treated in a ventilated biopile with a blend of selected microorganisms that naturally break the toxins down into less harmful compounds	NA	[175]

were *Mucor circinelloides* and *Galactomyces geotrichum*. This consortium demonstrated a high ability to degrade contaminated soil during a period of 28 days of incubation at both 30 and 60 °C with an efficiency of 87 % [155].

In a microcosm study conducted by Gohil and coworkers [161], DDTs were degraded with indigenous microorganisms supplemented with four electron donors (Table 6). The samples were incubated for 2 months in which a range of electron donors were tested including two organic acids (lactate and acetate at 20 mM) and H_2 at 100 kPa and sulfate (as K_2SO_4) as an electro-acceptor [161, 169]. The greatest losses of DDT (approximately 87 %) were observed with lactate as the electron donor. Moreover, the final concentrations of DDT and its metabolites DDE and DDD were higher after acetate supplementation than with either the lactate or H_2; the same result was also obtained at a mesocosm scale [161]. The role of lactate observed may be explained by the redox potential due to the characteristic of lactate to be fermented to other organic acids such as formate and acetate [161], which resulted in greater microbial oxygen consumption. In fact, different groups of organisms may feed on the lactate fermentation, which stimulates their biodegradation activities [170]. Recently, a study conducted by Sun and coworkers [164] showed that addition of NH_4Cl and KH_2PO_4, in a mesocosm as a nutritional supplement enhanced the biodegradation of DDT. It is important to note that the greatest obstacle to biological degradation is the low availability of the substrate. Recent studies have suggested that such

condition like adding nutrients or electron acceptors could stimulate microbial growth in uncontaminated or contaminated ecosystems [161, 170]. Although those studies have demonstrated the feasibility of the DDT remediation process on small scales [161, 170]. Nevertheless, the above-mentioned studies highlighted the need for analysis of bioremediation on a large scale. However, there were major problems:

- The control of inoculum: any microorganisms can achieve the total mineralization of DDT, but the highest majority of the isolates show partial metabolization (up to DDE and DDD). It is necessary to find new, more competent catalysts with the aim of obtaining a complete metabolization of the DDT or find other biological elements capable of detoxifying the DDT-polluted environments.
- Optimization of the remediation process and also definition of fundamental parameters, which guide the development and the improvement of bioremediation, is needed.
- Combining the various aerobics and anaerobic methods of treatment.

The improvement of the bioavailability of DDT: previous studies showed that the process is very long due to the high lipophilicity of the DDT, which leads to low availability. It is possible to increase the efficiency of the bioremediation by the stimulation of DDT bioavailability.

Recent Advances in Field-Scale DDT Bioremediation

After some remediation cases in a large scale such as the dragging operation of the sediment of San Francisco Bay in 1992, when one published study evaluated this remediation operation and which showed that it did not succeed [171]. Therefore, new remediation technologies are being developed for the in situ bioremediation of DDT-contaminated soil. It was considered as the main pathway to eliminate organic compounds such as DDT from contaminated soils (Table 7). It was demonstrated that it is necessary to establish processes required to remove the high number of DDTs-contaminated sites [172, 175].

Therefore, the objectives for developing different methods for cleaning up a contaminated area is to reduce the levels of contaminant concentrations and also identify the most effective combination of soil amendments, associated factors, and operating conditions that would achieve bioremediation and especially producing less harmful by-products.

Strategies to Promote DDTs Removal from Contaminated Environments

Although bioremediation is generally regarded as both an economically and ecologically sustainable option for the elimination of pesticides in contaminated soil, environmental factors may limit the rate and the extent of biodegradation and also the growth and activity of DDT-metabolizing microbes [176, 177]. One of the foremost factors affecting the DDT bioremediation process is its bioavailability; it has been observed that owing to their strong hydrophobicity, soil or sediment pollutant contact time increases the pollutant bioavailability [140] and consequently increase their ecotoxicological effects [178]. Therefore, it is important to determine ways of increasing the DDT bioavailability.

Various remediation processes can also use surfactants to enhance biodegradation in polluted soil. Surfactants are a class of natural and synthetic chemicals that promote the wetting, solubilization, and emulsification of various types of organic and inorganic contaminants [179].

The use of surfactants to remove DDT from soil has been described by a small number of studies [180], and there are a wide variety of surfactants that have been tested with differing organic compounds with variable results [181–183].

Baczynski and coworkers have shown that surfactant application resulted in lowered production of metabolites, which is why it enhanced the biodegradation of DDT. It also decreased the residual concentration of DDT remaining after biodegradation [157]. Different types of surfactants can be used to increase contaminant bioavailability including synthetic surfactants such as Brij 30 and C12E8 (octaethylene glycol mono *n*-dodecyl ether) [184, 185], natural surfactants [186], and biosurfactants [183, 187].

In 1995, Parfitt et al. [188] extracted DDTs from contaminated soil collected from New Zealand using two non-ionic surfactant types: triton and polypropylene glycolethoxylate. It was found that 45 % of DDT was removed using 2 % of surfactant, and the remaining DDT was associated with the silt fraction [188]. Later, Smith et al. [189] added propanol and ethanol with a low surfactant concentration to improve DDT's removal from polluted soil [189].

A large variety of surfactants have been documented for possible applications in DDT bioremediation such as Tween 80, Triton, and polypropylene glycolethoxylate [182, 188, 190]. However, surfactant concentration is another important factor to be considered; it was found that in some cases, surfactants used can inhibit DDT biodegradation. Baczynski and Pleissner have reported that in anaerobic biodegradation of DDT in contaminated soil, the use of higher doses of Tween 80 has brought DDD accumulation [190]. It was hypothesized that this effect resulted from exceeding the surfactant critical micelle concentration in the water phase, indeed when in equilibrium, the amount of solubilized DDT linearly depends on the surfactant concentration above the critical micelle concentration which was explained by the work done by Kile and coworkers [180].

In an earlier study, it was hypothesized that combinations of two surfactants may be more effective for DDT bioremediation process; especially if non-ionic and anionic properties were combined [191] (Table 8). Although, sodium dodecyl sulfate was combined with heating and low frequency ultrasound was tested as an anionic surfactant (Tab. 8) allowing efficient DDT solubilization ranging from 40 to 90 % [196]. Recently, Zheng and collaborators revealed that non-ionic surfactants including Tween 80 and Triton X-100, cosurfactant 1-pentanol and plant oils possess higher solubilizing capacities for organochlorine pesticides such as DDT [197, 198]. These surfactants caused considerable enhancement of anaerobic biodegradation of DDT in contaminated soil with lower DDD accumulation and higher DBP production [197, 198].

Conclusion

There are several regions in the world, including some countries in Africa, Asia, and Latin America, where the use of DDT represents the most efficient strategy for malaria vector control, simply because there is no alternative with equivalent success, efficiency, and feasibility. Alternative approaches will make remediation feasible, and there is an urgent need to develop strategies, not only to reduce reliance on DDT but also to accomplish its ultimate elimination and to sustain effective malaria vector control. We believe that the biodegradation processes of aged DDT-polluted sites can be the most suitable solution to reduce the health risks of this toxic compound. Biodegradation by cost-effective procedures based on bioremediation appears as a relevant solution.

Table 8 Summary of surfactant mixture to increase the bioavailability and the bioremediation of DDT

Enhancing agents	Impact on remediation	Result on removal process	Impact on removal rate (%)	References
Tween 80	Enhancing the DDT transformation	-Reducing the accumulation of persistent metabolites -Increasing the formation of terminal metabolite DBP	70	[157]
Consolvant washing (50 % 1-Propanol)	Enhance solubilization of DDT	Enhanced adsorption and accelerated transformation of DDT	42	[189]
Tween 80	Enhancing the DDT solubilization	-Increase of the DDT degradation -Higher DDT production	80	[190]
Wood sawdust with cork wastes	Acceleration of the rate of the adsorption kinetics of DDT on the low-cost adsorbents was found best	Enhanced bioavailability of DDT	NA	[191]
Reducing agent + surfactant (Triton X114 and Brij 35)	Increasing the solubility of DDT and accelerates its biodegradation	Increase the transformation of DDT with the accumulation of DDD and other products, such as DBP	28.1 (Triton X114) 84.8 (Brij 35)	[192]
Brij 30	Increase the solubilization, the bioavailability, and anaerobic biodegradability of DDT and its metabolites DDD and DDE	Significantly greater rates and extents of DDT degradation with a decrease in DDD accumulation	80	[193]
Lower molecular organic acid	Chelating inorganic ions and enhancing the bioavailability of DDT results in the dissolution of the soil structure	Lower DDE formation with oxalic and citric acid than with control soil	2.1	[194]
Vitamin B12 with ionic liquid	Acceleration of electrolytic dechlorination of DDT and DDD	Accelerated formation of final metabolites (DDNU and DDMS)	73–82	[195]
Anionic surfactant (dodecyl sulfate combined with heating and low frequency ultrasound)	Enhance and increase solubilization of DDT	Increasing solubilization and bioavaibility of DDT	40–90	[196]
Tween 80 and Trixon X-100	Enhancing the DDT solubilization	-Reducing the accumulation of persistent metabolites	72.9	[197, 198]

Several studies have proposed various bioremediation processes for DDT-contaminated areas. These studies have been conducted with the aim of developing process for bioremediation of contaminated sites. They demonstrated the feasibility of the DDT remediation process on a small scale. Nevertheless, the next challenge will be to (i) find new, more competent

catalysts able for complete mineralization of DDT, (ii) encourage further efforts to improve those catalysts to remediate polluted sites, and (iii) developing tests for monitoring the bioremediation process on a large scale.

Compliance with Ethical Standards

Conflict of Interest All authors declare no conflict of interest.

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