

Restoration of Soils and Groundwater Contaminated by Explosives

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Soil pollution resulting from explosives represents a critical environmental challenge. While physical methods like excavation and disposal are effective, their applicability is constrained by cost and logistical challenges for large contaminated areas. Chemical methods, such as oxidation and reduction, focus on transforming explosives into less toxic byproducts. Biological remediation utilizing plants and microorganisms emerges as a cost-effective and sustainable alternative.

Keywords: soil contamination ; explosives ; 2,4,6-trinitrotoluene (TNT) ; rapid detention explosive (RDX) ; soil remediation ; soil sustainability

1. Introduction

Soil pollution resulting from explosives represents a critical environmental challenge with far-reaching consequences for ecosystems and human health ^{[1][2]}. Explosives, commonly used in military activities, mining, and construction, release hazardous substances such as nitroaromatics and heavy metals into the soil, leading to long-lasting environmental degradation ^{[3][4]}. Explosives have the potential to cause extensive and varied soil contamination, necessitating substantial resources to delineate the contamination boundaries. It is important to note that the remediation of soil contaminated by explosives is not as advanced as the treatment of soil contamination caused by heavy metals and organic pollutants. The challenges in remediating soil contaminated by explosives can be attributed to the presence of decomposition products from energetic explosives and the emergence of new types of explosives with compositions that are difficult to define ^{[5][6]} ^[7]. Moreover, most explosives have the capacity to bind to the organic matter in the soil and to coexist with other compounds as a mixture, creating difficulties in soil remediation ^{[8][9]}.

Open burning and detonation were the most common treatment methods for waste explosives until the 1990s, when there was no safe alternative. These methods produce emissions and noise, and new regulations have restricted them ^{[10][11]}. In response to this environmental challenge, this text outlines contemporary remediation approaches and technologies that have been evaluated in pilot- and full-scale applications.

The explosive compounds that are commonly detected in polluted sites are 2,4,6-trinitrotoluene (TNT) and 1,3,5-Trinitroperhydro-1,3,5-triazine (RDX). TNT is a powerful explosive compound commonly used in military applications, construction, and mining due to its low detonation velocity ^[12]. TNT is a yellow, crystalline compound with the chemical formula $C_7H_5N_3O_6$ and remains relatively inert and does not pose an immediate danger of detonation under normal conditions, making it a suitable explosive for military and industrial applications ^[13]. It does not explode easily when subjected to mechanical shocks or high temperatures. Despite its stability, TNT is a powerful explosive when initiated by a detonator or heat. It releases a significant amount of energy upon detonation, producing a shockwave and heat that can cause destruction ^{[12][14]}. TNT is less soluble in water but more soluble in organic solvents like acetone and ethanol. This solubility affects its transport and potential for groundwater contamination ^[15]. When TNT is released into the environment, it can lead to soil contamination, which has a range of adverse effects. Soil pollution with TNT (2,4,6-trinitrotoluene) is a significant environmental concern with potentially serious consequences for both ecosystems and human health. The limit concentration of TNT is 51 mg/kg in soils for industrial uses and 0.98 µg/L in waters for domestic uses ^[16].

Hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX) is an explosive compound widely used in military applications, mining, and industrial sectors. However, its persistence in the environment has raised concerns about soil pollution ^{[17][18]}. The compound's resistance to decomposition and its limited solubility contribute to its long-term presence in soils. This soil pollution poses serious ecological risks, affecting soil quality, plant life (RDX has been shown to inhibit seed germination, root development, and overall plant growth), and microbial communities (leading to long-term soil degradation). Furthermore, RDX has been found to leach into the groundwater, posing a risk to nearby ecosystems and potentially

affecting drinking water supplies. Efforts to mitigate RDX soil pollution involve a range of remediation strategies, including bioremediation, phytoremediation, and chemical treatment methods [19]. These techniques aim to degrade or remove RDX from contaminated soils, but their effectiveness depends on site-specific conditions and challenges [20]. Regulatory measures are crucial for preventing RDX soil pollution. Many countries have established guidelines and regulations to limit the release of RDX into the environment, promoting responsible handling, storage, and disposal practices. The monitoring and enforcement of these regulations are essential to safeguarding ecosystems and public health. Most of the explosive substances and their derivatives are included in the Table of Harmonized Classifications and Labeling of Hazardous Substances (Annex VI) of European Regulation 1272/2008 [21]. RDX and HMX have been registered as hazardous in the database of the European Chemicals Agency (ECHA) in accordance with the REACH Regulation ((EC) 1907/2006) [22].

2. General Categories of Technologies for the Restoration of Polluted Soils and Groundwater

2.1. Soils

Until 1990, in the USA and internationally, only thermal methods were used as on-site techniques for treating soils contaminated with explosives. With the aim of reducing processing costs and addressing other negative characteristics, such as high energy consumption, the need for the effective control of volatiles, etc., starting in the mid-1990s, there was a shift in the search for alternative treatment methods [23][24][25]. In **Figure 1**, the main alternative methods for the restoration of soils polluted by explosives can be classified into four general categories: (i) containment, (ii) excavation and treatment or off-site disposal, (iii) in situ or on-site treatment, and (iv) monitoring of natural attenuation (**Figure 1**).

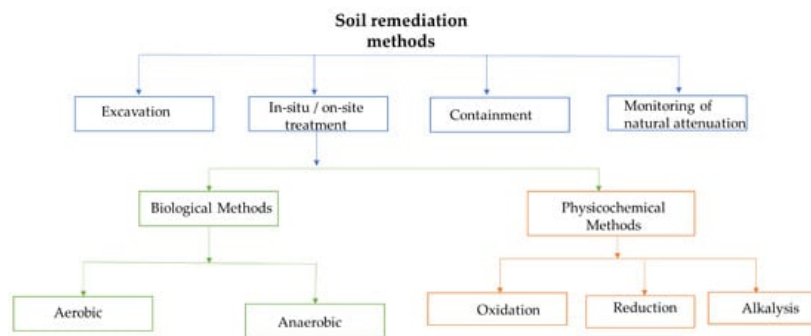


Figure 1. Remediation methods for soil contaminated with explosives.

(i) Containment

These methods aim to isolate the area where the contaminated soil is located so that there is no risk of transferring pollution to sensitive receptors through air, rainwater, or groundwater [23]. They usually involve creating impermeable surface covers or perimeter barriers, in combination with hydraulic control measures of groundwater and the collection of the leachate from the contained area. These technologies are typically applied to contaminated sites with large surface areas and large quantities of contaminated soil, particularly in remote areas (abandoned mines, uncontrolled landfills, etc.), far from populated areas and without other prospects for utilization.

(ii) Excavation and treatment or disposal of contaminated soil in facilities outside the area (off-site treatment or disposal)

In this category of methods, the excavation of the contaminated soil takes place, which is then transported to specialized facilities outside the area for the processing, cleaning, immobilization, or destruction of the pollutants (off-site treatment), with the aim of reusing it, or more commonly, for final disposal in specialized hazardous waste sites. These methods are usually feasible and economically advantageous only when the volume of contaminated soil is relatively small and is located at a shallow depth.

(iii) In Situ or on-site treatment

This category includes methods that are applied in the area where the contaminated soil is located without prior excavation (in situ) or after excavation and transport of the soil to a neighboring area that has been prepared appropriately (on-site), with the aim of processing and restoring the soil to its original position. These methods involve a wide range of biological or physicochemical technologies and will be examined in more detail later.

The monitoring of natural attenuation is usually selected in the case of organic pollutants if there are indications of their degradation, such as the detection of breakdown products. It is applied in areas with low concentrations of pollutants, and natural degradation can be monitored as a means of restoration. However, for areas with higher concentrations of pollutants, natural attenuation with in situ or on-site treatment technologies is employed. The monitoring process involves initial site characterization and the installation of monitoring wells for groundwater sampling (the samples are analyzed for concentrations of contaminants and their breakdown products, redox potential, and pH). Also, groundwater flow rates and microbial populations are recorded [26].

2.2. Waters

On-site treatment involves pumping groundwater to the surface and using biological or physical–chemical methods to clean it. In situ treatment of groundwater, on the other hand, has two main variations: the installation of a permeable reactive barrier containing materials that can absorb or degrade pollutants and the injection of suitable reactants into the aquifer to destroy or immobilize pollutants using biological or physiochemical processes.

3. Methods for Processing Soils and Waters Contaminated with Explosives

In this section, the methods that have been applied for the destruction or neutralization of explosives are presented and discussed in detail. These are methods that can be applied in situ or on-site and can be classified into two basic categories depending on the main mechanism of action: (1) biological and (2) physicochemical technologies.

In biological technologies, the breakdown of explosives can take place with bacteria under aerobic or anaerobic conditions, where the explosive compound serves as a source of carbon and/or nitrogen. Alternatively, degradation can result from metabolism, where an additional substrate serves as a source of carbon and energy. The dominant metabolites of TNT under aerobic and anaerobic conditions are 2-ADNT, 4-ADNT, 2,4-DANT, and 2,6-DANT (diamino-nitrotolouenes), while triaminotoluene can be formed under anaerobic conditions [23].

Physicochemical methods involve the addition of one or more chemical reactants (reductive, oxidative) to contaminated soils that alter their physicochemical properties, such as pH or redox potential (Eh). These methods outperform biological actions, as they are faster, can be applied to a wider range of compounds, and are more easily controllable.

Table 1 and **Table 2** compile information sourced from the international literature, highlighting the utilization of biological and physicochemical methods for treating soil and groundwater contaminated with explosives, respectively. The focus is on technologies implemented in pilot- or full-scale applications. The construction of **Table 1** and **Table 2** is informed by recent studies evaluating agents and methodologies for remediating soil and water contaminated by explosives.

Table 1. Efficient remediation case studies for soil contaminated with explosives in on-site or in situ environments: an evaluation of performance.

Method	Level of Development	Area	Main Pollutants	Observations	Reference
Biological technologies					
Windrow composting. Aerobic conditions mixing 30% soil, 70% organic material (manure, sawdust, etc.)	Full scale. 15,000 tons of soil	Umatilla Army Depot, Hermiston, Oregon, USA	TNT: 4800 mg/kg RDX: 1000 mg/kg HMX: 800 mg/kg	Processing in batches of 3000 tons within enclosed temporary structures for control of conditions. Duration: 10–12 days per batch. Reduction of pollutants below detection limits. Cost: 351 USD/t (1997).	[27][28]
Variations of aerobic composting with (a) horse manure (20% w/w), (b) Daramend (2% w/w) and ZVI (0.5% w/w), (c) only ZVI (0.5% w/w)	Lab scale	Bofors Test Center, Karlskoga, Sweden	Soil. 1: RDX 1340 mg/kg; Soil 2: RDX 28,740 mg/kg	Soil 1. 94% removal of RDX with Daramend + ZVI (75% with ZVI, 0% horse manure) after 26 weeks of treatment. Soil 2. The three variations were ineffective.	[29]

Method	Level of Development	Area	Main Pollutants	Observations	Reference
Land farming compared to bioreactor with addition of molasses	Lab scale	Louisiana Army, Ammunition Plant, USA	TNT: 4000–10,000 mg/kg; RDX: 800–1900 mg/kg; HMX: 600–900 mg/kg	Removal of TNT after 182 days: 99% in the bioreactor and 82% with Techno-Agriculture. Lower effectiveness for RDX and HMX.	[23]
Soil-pile vaccination with white-rot fungi	Pilot scale	Construction Establishment of Finnish Defense Administration, Finland	NTD: 19,000 mg/kg mixing soil (14 kg) with fertilizer (271 kg), adding pine bark with fungi (10 kg)	Soil dilution with fertilizer (1:20) to reduce toxicity. TNT degradation: 80% on a laboratory scale (in 76 days) and 70% on a pilot scale (in 49 days).	[30]
Physicochemical technologies					
Chemical Reduction					
Addition of ZVI to soil piles. Application to static soil piles. Soil pile 1: Mixing 70 kg soil, 3.5 kg ZVI, 1.05 L CH ₃ COOH. Soil pile 2: Mixing 70 kg soil, 3.5 kg ZVI, 2% w/w Al ₂ (SO ₄) ₃ .	Pilot scale	Los Alamos National Laboratory, New Mexico, USA	Soil 1: RDX: 2700 mg/kg, pH 9.9 Soil 2: RDX: 12,100 mg/kg, pH 7	98% removal of RDX	[31]
Adding sulfur compounds. Commercial product: MuniRem	Field scale	Ravenna Army Ammunition Plant, USA	TNT: 3347 mg/kg RDX: 5977 mg/kg HMX: 647 mg/kg	99.6% removal of TNT, 96.5% removal of RDX, and 97.1% removal of HMX 97.1% in 24 h (>99% in two weeks)	[32]
Alkaline Hydrolysis					
Mixing with Ca(OH) ₂ solution at pH 11 and pH 12	Lab scale	Former German ammunition factories	Soil 1: TNT: 16,000 mg/kg; Soil 2: TNT: 116 mg/kg	93–98% removal of TNT in both soils within 7 days at both pH levels. DNT and ADNT showed more efficient removal at pH 12.	[33]
Mixing soil with dry Ca(OH) ₂ 5% and moisture 0–200%	Laboratory and semi-pilot scale (2 kg)	Soils from Iowa, USA	TNT 60 mg/kg	Optimal moisture ~25% Laboratory tests: 82–92% TNT removal in 10 days. Pilot test: Slower degradation	[34]
Laboratory tests: Effect of soil chemistry, percentage of Ca(OH) ₂ , etc. Pilot tests: Type of alkali, application methods.	Lab scale and pilot scale (70 kg)	17 areas, Nebraska Ordnance Plant, USA	Pilot tests: RDX 38.4 mg/kg, HMX 4.4 mg/kg, TNT 10.8 mg/kg	Pilot tests: 82–83% removal of RDX in one week with calcium, hydroxylapatite, and complete mixing. Slower kinetics with surface application and the use of fly ash.	[35]

Table 2. Technologies for groundwater remediation in areas contaminated with explosives: evaluating their effectiveness in pilot- and large-scale applications.

Pump and treat					
Adsorption on activated carbon	Established	Nebraska Ordnance Plant, USA	Contaminated soils, TNT and RDX > 5000 mg/kg had contaminated the aquifer, RDX up to 300 µg/L	15,000 m ³ /day. Installation cost: USD 30 million. Annual operating cost: USD 800,000.	[36]
In situ methods					

Use of ZVI in permeable reactive barrier (PRB) (in situ chemical reduction)	Demonstration scale	Cornhusker Army Ammunition Plant (CAAP), Grand Island, Nebraska	RDX: 0.9 µg/L TNT: 130 µg/L	PRB dimensions: 15 m length, 4.5 m depth, and 1 m thickness. Content: ZVI (30%), sand (70%). Monitoring for 20 months. TNT below detection limits at the outlet PRB. Cost: USD 1940/m ²	[37]
Bioremediation with nutrient addition Injection of soybean oil, lactic acid, Na, and surfactants	Full-scale 2008 installation, continuous monitoring	Pantex Plant, Amarillo, Texas,	RDX: 4000 µg/L,	42 injection wells on a surface area of 40,000 m ² . Depth of the aquifer horizon: 78 m. Thickness: 4.5–6.0 m. Cost: USD 190 per m ³ of groundwater.	[38] [39]
Bioremediation with nutrient addition Injection of whey (4.7 m ³) into an aquifer (400 m ² × 2.5 m depth)	In Situ pilot tests	Czech Republic	TNT: 10 mg/L > 90%	TNT removal over a period of 17 months	[40]
Oxidation by Fenton Injection	Field application	Pueblo Chemical Depot, Colorado, USA	RDX, HMX.	Injection, 16.6 m ³ H ₂ O ₂ (12.5%)/Fe ²⁺ over a two-day period. After 26 days of treatment: 100% HMX removal, 60% RDX removal, and 72–100% removal of other nitroaromatic compounds	[41]
Oxidation by NaMnO ₄ Injection of 70 m ³ of NaMnO ₄ (10 g/L) to create a reactive zone (9.2 m × 4 m × 6 m depth)	Pilot scale	Nebraska Ordnance Plant, USA	RDX 30–70 µg/L. Problems with the uniform distribution of permanganate in the soil.	RDX concentrations reduced by 70–80% near injection wells	[36]

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