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REMEDIATION OF DIOXIN-CONTAMINATED SOILS THROUGH THERMAL DESORPTION AND VAPOR MANAGEMENT VIA THERMAL OXIDIZER AT BIÊN HÒA AIRBASE, VIETNAM

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Abstract

During the US-Vietnam War, millions of liters of harmful herbicides, including Agent Orange, were dumped in Vietnam, resulting in severe health issues caused by polychlorinated dibenzo-p-dioxins and dibenzofurans ("dioxins"). Even decades later, dioxin contamination continues to affect the local population. In response to this problem, the Center for Technology Environmental Treatment/Chemical Force (CTET/CF), the GAET Corporation (Vietnam), and HAEMERS Technologies SA (Belgium) have collaborated to establish a joint operation. Their aim is to assess the effectiveness of thermal desorption treatment on dioxin-contaminated soil at Biên Hòa Airbase.

The primary objective of this trial treatment is to demonstrate the capabilities of thermal treatment technology and design in meeting specific targets for dioxins at the site. The goal is to reduce the contamination level from over 17.000 ppt Toxic Equivalents (TEQ) to below 300 ppt TEQ. To achieve this, Haemers Technologies has developed a thermal treatment pilot plant comprising two main units: (i) The thermal pile of 237m³ composed of three types of materials: 187m³ of contaminated soils, 25m³ of contaminated sludges and 25m³ of soil washing cake; (ii) The vapor treatment unit composed of a thermal oxidizer where dioxins are destroyed and a quench tower that rapidly cools the vapor, preventing dioxin reformation. This unit generates no liquid or solid waste. The pilot project has successfully demonstrated that soil can be remediated by heating it to 335°C, with a pollutant removal rate of 99 wt.%. Additionally, dioxins can be destroyed at 1.100°C, achieving a destruction rate of 99.9999 wt.% in the thermal oxidizer. This zero-waste solution offers an improved method for remediating dioxin-contaminated soils, with air emission results meeting the standards set by Vietnam, the European Union (EU), and the United States (US).

Overall, this project showcases an efficient thermal treatment technology for remediating dioxin-contaminated soils. It ensures the complete destruction of all toxic components while enabling soil recycling.

Key words: agent orange, dioxins remediation, thermal conductive heating, thermal oxidizer, zero-waste

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1. Introduction

1.1. Site history and project timeline

During the US-Vietnam War, large quantities of herbicides known as the Rainbow agents were sprayed over Vietnam. Biên Hòa Airbase, which served as a joint operating base for the South Vietnam Air Force and the United States Air Force, stored thousands of barrels of Agent Orange, one of the most toxic herbicides used (USAID, 2016). It has been scientifically proven that Agent Orange causes severe health issues, including birth defects, neurological problems, and various types of cancer (Schecter et al., 2009; Young and Cecil, 2011). The composition of Agent Orange consists of 2,4-dichlorophenoxyacetic acid and 2,4,5-trichlorophenoxyacetic acid, with trace amounts of polychlorinated dibenzo-p-dioxins and dibenzofurans ("dioxins") present as impurities resulting from the production of 2,4,5-

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trichlorophenoxyacetic acid. Dioxins are highly insoluble in water, lipophilic, and extremely persistent in the environment (Kulkarni et al., 2008; Mudhoo et al., 2013).

Decades after the Vietnam War came to an end in 1975, the persistence and accumulation of dioxins continue to have adverse effects on the local population (Schecter et al., 2003; Tuyet-Hanh et al., 2015). To improve the living conditions for the residents, it became necessary to take measures to remediate the soil contaminated with dioxins.

The Center for Technology Environmental Treatment/Chemical Force (CTET/CF), the GAET Corporation, and HAEMERS Technologies SA have joined forces to establish a collaborative effort. Their primary objective is to conduct testing and evaluation activities to demonstrate the efficacy of thermal desorption treatment on dioxin-contaminated soils at Biên Hòa Airbase.

A thermal desorption treatment project for dioxin-contaminated soils has already been conducted in Đà Nẵng, Vietnam (USAID, 2010, 2015) and demonstrated the effectiveness of thermal treatment in achieving target levels of dioxins in soil. The main objective of this new operation at Biên Hòa airbase is to demonstrate the effectiveness of the Smart Burners^{TM} technology but, more importantly, to propose an improved solution compared to what has been done in the past, particularly in terms of contaminated vapor management.

Therefore, Haemers Technologies has designed a thermal treatment pilot plant composed of two main units:

- The soil pile, comprising various types of materials, is subjected to thermal treatment.
- The Vapor Treatment Unit (VTU): This unit is responsible for the destruction of dioxins. It comprises a thermal oxidizer, which ensures the complete destruction of dioxins, and prevents the formation of any liquid or solid waste during the process.

Haemers Technologies has undertaken this pilot-demonstration initiative at the Biên Hòa Airforce Base, aiming to implement its specialized technologies for the treatment of soil contaminated with Agent Orange. Although the project started in early 2020, it faced an interruption due to the COVID pandemic. Haemers Technologies returned to the site in late December 2021, resuming its activities and upholding its commitment to the demonstration project. The remediation process was restarted on February 2, 2022, and after a treatment duration of 40 days, the heating phase was concluded on March 14, 2022. The thermal pile successfully attained and sustained the target temperature of 335°C for a minimum of five days.

1.2. Technology outline

In the Haemers Technologies' process (Haemers, 2016) polluted soil is heated by conduction under the action of Smart Burners™ developed by Haemers Technologies. More precisely, steel tubes are inserted into the soil to be remediated. Hot gases, generated by Smart Burners™, circulate in the tubes in order to transfer heat to the soil. This results in the vaporization of the pollutants in the soil when their boiling temperature is reached.

The emitted vapors are extracted through perforated steel tubes, called vapor tubes, and are then either directed to a vapor treatment unit for further treatment or reintroduced into the flame generated by the Smart Burners^{TM} in the case of hydrocarbons pollution.

In *Ex situ* Thermal Desorption (ESTD) mode, heating tubes are inserted horizontally in pre-excavated soils (Fig. 1). ESTD is an on-site or near-site treatment that avoids truck traffic on public roads.

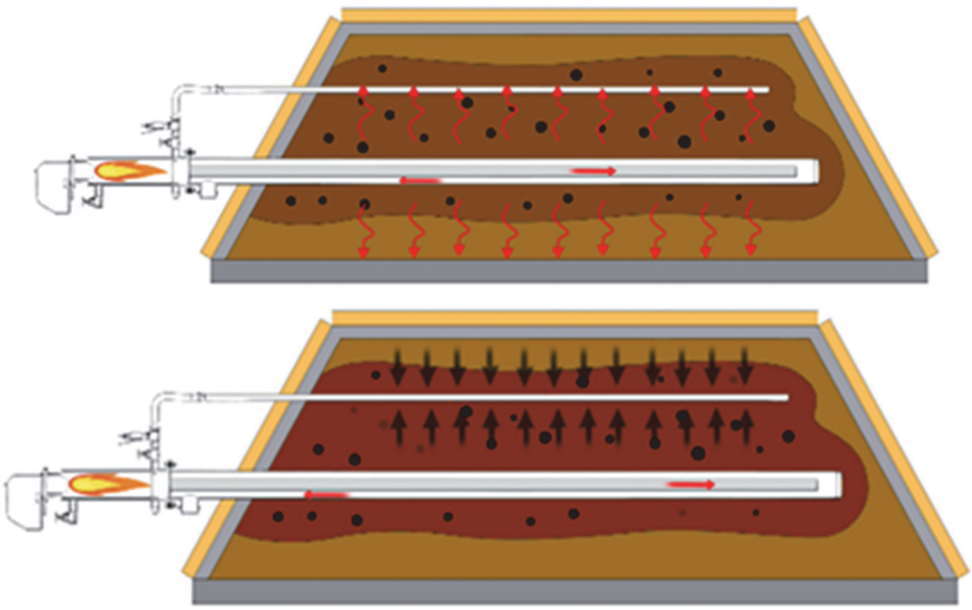


Fig. 1. *Ex situ* Thermal Desorption (ESTD) principle

2. Project design

2.1. Thermal pile design

The *Ex situ* Thermal Desorption (ESTD) takes place in a designated landfill area within Biên Hòa Airbase, where contaminated soils have been stored and restrained over the years. The ESTD is composed of 15 horizontal heating tubes and 13 exchanger tubes (Fig. 2), heating the soil by conduction. The exchanger tubes recover the combustion gases after their passage through the heating tubes, allowing a second passage through the pile and energy optimization. The fan, located at the back of the pile, drives the combustion gases from the burners through the tube network to the chimney. The combustion gases never encounter the contaminated soil and circulate through the tube network until they are released into the atmosphere. Conversely, the polluted vapors generated by the temperature rise in the pile are collected by the vapor tubes placed inside the pile and connected to a collector, placed in front of the pile, leading the polluted vapors to the vapor treatment unit.

The main goal of treatment is to lower the TCDD-TEQ concentration in contaminated soils to target objectives (Table 1), by heating the soil to the target temperature of 335°C. The top of the pile is covered with a layer of gravel, in which more vapor tubes, called secondary vapor tubes, are placed. These secondary vapor tubes are an additional security to avoid any fugitive emission or contamination of the concrete.

The whole pile is then covered with concrete and thermal insulation. The 237 cubic meters pile is made of materials from three origins: 187m^3 are contaminated soils directly extracted from the polluted area, 25m^3 are contaminated sludge and another 25m^3 are washing cake. Soil washing is a remediation technique using physical separation technique to remove contaminants from soils. Particles are separated by size. As a matter, most contaminants tend to be sorbed to clay and silt.

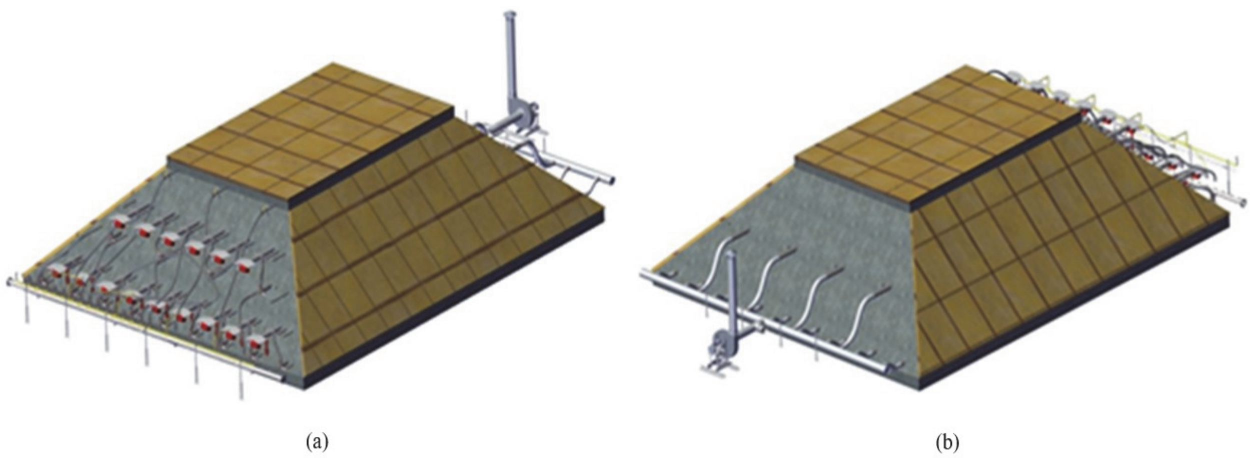
Washing separates those small particles from larger ones (sand and gravel) by breaking adhesive bonds and by mean of filtration. The smaller particles are then filtered and generate a highly contaminated filter-cake (Tran et al., 2022). To avoid the landfill disposal of this residue, the cake is treated in the ESTD pile. In this pilot project the washing cake comes from the Shimizu soil washing installation and was produced in 2019-2020 at Biên Hòa Airbase.

The sludges and washing cakes are arranged in "basins" in the pile of contaminated soil as presented in Fig. 3. The level of contamination of the different materials and their respective treatment objective are listed in Table 1. For soil and sludge materials, initial concentrations are sourced USAID environmental assessment at Biên Hòa Airbase (USAID, 2016). For washing cakes, analyses were performed by an accredited European laboratory on samples before treatment (Eigen method NEN-EN-1948).

2.2. Vapor Treatment Unit

The Vapor Treatment Unit (VTU) is specifically designed to manage the contaminated vapors generated during the soil treatment process and ensure their concentrations are reduced to comply with legal emission standards. There are two primary options available for the treatment of these vapors: condensation and adsorption on activated carbon or thermal oxidation. The first option, condensation and adsorption on activated carbon offers the advantage of low energy consumption. However, it does come with the drawback of producing liquid and solid waste.

The liquid obtained after condensation requires additional treatment, and the activated carbon used in the absorption process becomes a solid hazardous waste that needs proper disposal. In contrast, the second option, Thermal Oxidation, eliminates the need for additional treatment of liquid waste or the disposal of solid waste.



(a) (b)

Fig. 2. ESTD design front (a), back (b)

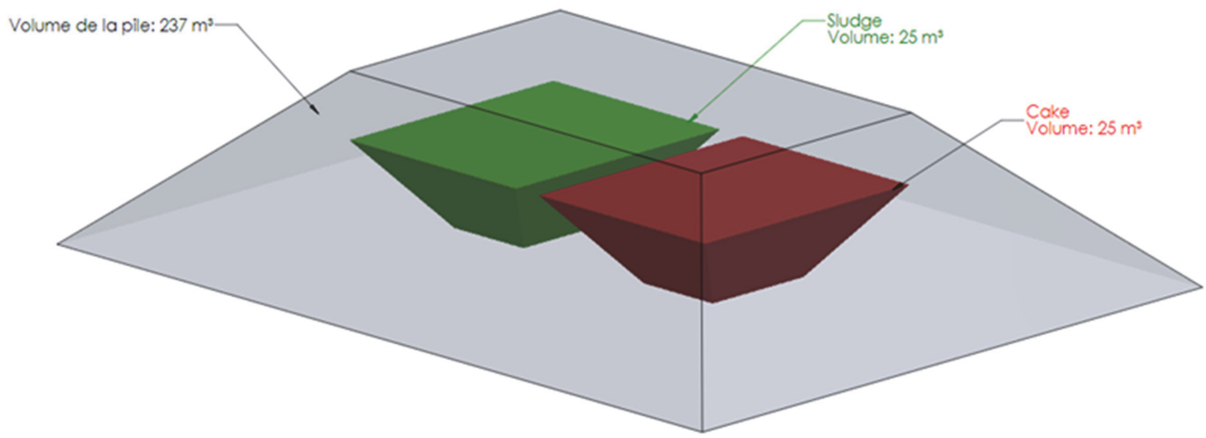


Fig. 3. ESTD composition: 187m³ of contaminated soils (in grey), 25m³ of sludges and 25m³ of washing cake

Table 1. Contaminated materials concentrations

Material	*Unit*	*Initial concentration*	*Treatment goal*	*Target DRE* (%)*
Soil	ppt TEQ	> 11,400	300 (Urban residential) 1200 (Industrial)	97.3 89.5
Sludges	ppt TEQ	> 5,410	150	97.2
Washing cake	ppt TEQ	> 17,200	300	98.2

* *Destruction Rate Efficiency*

It achieves the destruction of contaminants through high-temperature combustion. While thermal oxidation may require more energy, it eliminates the complications associated with liquid and solid waste management. An insulated piping network collects the vapours and routes them to the VTU; some condensation occurs, and the condensates are recovered and treated in the thermal oxidizer as well, leaving no contaminated waste (zero waste technology). Energy efficiency for thermal oxidation can be improved by installing a recuperative heat exchanger. The option was judged not suitable for a pilot stage. The chosen design for vapor treatment consists of a Direct Fired Thermal Oxidizer. A direct fired Thermal Oxidizer (also known as a direct-fired oxidizer or afterburner) is an air pollution control technology that is used to treat exhaust gases containing volatile organic compounds (VOCs). The process involves heating the exhaust gas stream to a high temperature in a combustion chamber with a burner and maintaining it at that temperature for a residence time sufficient to allow the oxidation of the VOCs .

In order to achieve a destruction rate efficiency over 99.99%, the following criteria must be met within the oxidation chamber (Gao et al., 2015; Temme et al., 2015):

- temperature of minimum 1.100°C (preferably 1.200°C)
- oxygen content of minimum 6% (preferably 10%)
- residence time of minimum 1 second (preferably 2 seconds)
- good mixing conditions - High Turbulence (Re>>4,000).

Dioxin compounds reformation can happen in the cooling phase, within a temperature range of 200°C to 500°C, with the highest formation occurring at 350°C. The presence of oxygen, chlorine (Cl_2), and hydrocarbons can facilitate the reformation of dioxins. Additionally, factors such as the presence of dust and/or metals can contribute to the formation of dioxins (Buekens et al., 2001; Mukherjee et al., 2016). To avoid the reformation process, the hot gases pass through a quench tower to be cooled quickly enough below 200°C before being released into the atmosphere. To comply with Vietnamese regulations, a maximum temperature of 180°C at the stack has been set. The Vapor Treatment Unit configuration is presented in Fig. 4.

3. Treatment monitoring

The performance of the thermal treatment is evaluated by various parameters: temperature profile in the thermal pile over time, pressure into the soil, emissions at the venting and soil sampling.

3.1. Soil temperature monitoring

The increase in soil temperature is the principal indicator of the progress of decontamination. It is measured at the 'cold points' in the soil. A cold point is at the center of the triangle formed by every three tubes and is therefore the furthest point from the heat source (Fig. 5).

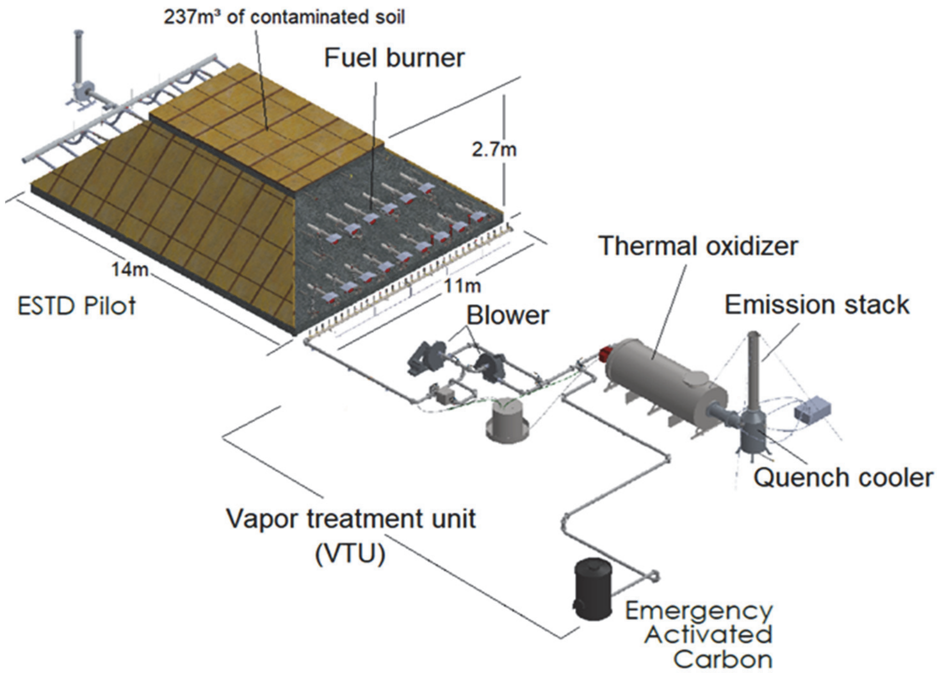


Fig. 4. Vapor Treatment Unit configuration

Temperature measurement is done through thermocouples. Thermocouples are devices consisting of two conductors that produce a temperature-dependent voltage to monitor large temperature ranges. Type-K thermocouples are used, allowing for measurements up to 800°C.

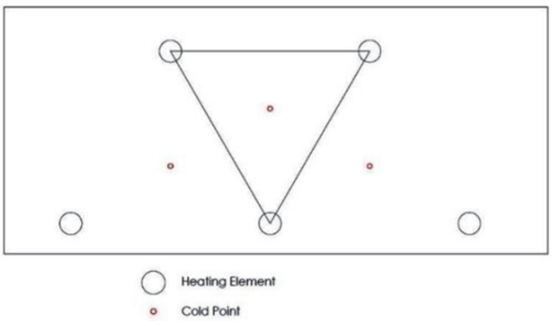


Fig. 5. Cold points

Soil temperature is measured at nine different locations, as described in Fig. 6 - T1 to T9 - and at four different depths within the thermal pile. A total of 36 temperature sensors are placed in the thermal pile.

3.2. Soil pressure monitoring

It is fundamental to control and keep track of the soil pressure during treatment. The aim of this monitoring is to ensure that a negative relative pressure in the soil ($P<P_{atm}$) is maintained, which will consequently prevent fugitive emissions during the treatment. For this project, two pressure wells (P1-P2) were installed and measurements were performed manually every day.

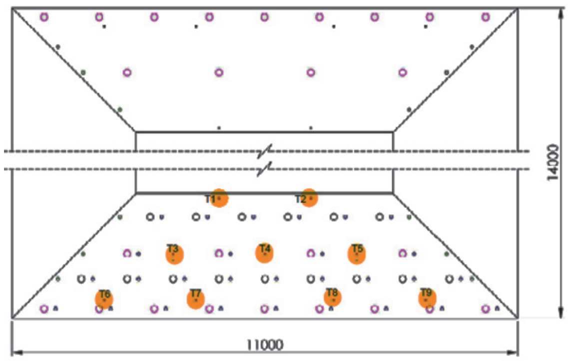


Fig. 6. Temperature monitoring locations

3.3. Emission control

The Vietnam-Russia Tropical Center (VRTC), an accredited Vietnamese laboratory, performed sampling at the outlet of the chimney during the treatment process. Six gas samples were collected between February 8 and 25, 2022, using the United States Environmental Protection Agency (USEPA) method 23 for analysis of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans. The sampling equipment consisted of a glass fiber filter, a condenser, a trap, and four impingers with different contents (EPA, 2022).

Sample extraction and analysis followed the EPA method 23 procedures, using Gas Chromatography coupled with High-Resolution Mass Spectrometry. Toxicity Equivalence Factors (TEF) were used to correlate the toxicity of various dioxin and furan compounds, with the most toxic compound, 2,3,7,8-TCDD, having a TEF of 1. Each compound's concentration was multiplied by its respective equivalence factor, and the sum of these values constituted the total toxicity relative to 2,3,7,8-TCDD (Eq. 1) (EPA, 2022).

$$Total\ Equivalent\ Toxicity\ (TEQ) = \sum_{i=1}^{n} Ci \times TEF_i \quad (1)$$

Toxicity Equivalence Factors (TEF) is established by WHO 2005 TEF (Table 2).

3.4. Soil sampling

Soil samples after treatment were collected in the cooled thermal pile and analysis by an accredited laboratory: Eurofins, Vietnam.

4. Treatment results and discussions

4.1. Soil temperature

4.1.1. Thermographies

Thermographies are heat maps representing the temperature distribution inside the thermal pile. This section presents the thermographies generated at different stages of heat treatment (10, 22, 30 and 40 days of treatment) (Fig. 7). Each thermography illustrates a vertical slice of the thermal pile: the front, the middle and the back of the pile. The evolution of the thermographies enables to visualize the evolution of the heat front in the pile.

The thermographies show that the whole mass of the thermal pile has heated up to the target temperature. After 10 and 22 days, the hot spots are localized next to the heating elements. As the treatment progresses, when 30 and 40 days of treatment are reached, the heat front gradually moves away from the heating tubes towards the so-called cold points (the most distant points from the heating elements).

4.1.2. Average temperature

As the data from thermocouples does not fully represent the global temperature evolution of the soil mass (measurement of coldest points of the thermal pile), Haemers Technologies employs the ANSYS Fluent simulation program to better determine the average temperature of the pile. Haemers Technologies has developed its own algorithm to control their sites and estimate optimal treatment stop. By comparing simulation data with previous sites, a more accurate representation of temperature evolution is achieved (Depasse et al., 2021).

The evolution of the average soil temperature is represented in the green line below. The red line represents the target temperature (335°C) (Fig. 8). The average temperature in the thermal pile has reached the target temperature of 335°C after 35 days of treatment. The target temperature was maintained for five days before stopping thermal treatment.

4.2. Pressure monitoring

Figure 9 shows the evolution of the vacuum in the pile, monitored at both wells (P1 and P2), during treatment. The vertical axis shows the pressure measured (in mbar), the horizontal axis the treatment time. Throughout the treatment process, a vacuum was maintained in the thermal pile, ensuring that the pressure remained below 0 mbar. These results provide evidence that no fugitive emissions occurred during the remediation process. The findings were further supported by ambient air monitoring conducted by the CTET: the ambient air quality remained in conformity throughout the pilot project, for all parameters monitored (dust and noise, volatile organic compounds (VOCs), chlorophenols and dioxins).

4.3. Emissions control results

The results of stack sampling analysis carried out between 08/02/2022 and 25/02/2022 are presented in Table 3 and Fig. 10. The emission standard, represented with the blue line, is 0.1 ng TEQ/Nm³ (or 100 pg TEQ/Nm³). As presented above, the norm is respected, and all emissions are compliant. These results confirm the efficiency of the thermal oxidizer and the proper oxidation of the dioxins inside.

4.4. Soil sampling results

The results of the final sampling are available in Table 4. The two treated soil analysis were respectively performed by two laboratories: Agrolab (Accredited European laboratory) and Eurofins. Treated sludges and washing cake were analysed by Eurofins. As presented in Table 4, the treatment objective of each material (soil, sludges and washing cake) were met. The Destruction Rate Efficiency is over 99% for all samples.

4. Conclusions

In conclusion, the joint operation between CTET/CF, the GAET Corporation, and HAEMERS Technologies SA has successfully demonstrated the effectiveness of thermal desorption treatment on dioxin-contaminated soil at Biên Hòa Airbase. The pilot project has shown that dioxin-contaminated soil can be remediated after being heated to 335°C, and vaporized dioxins can be destroyed at 1,100°C. The thermal treatment technology and design have met the site-specific dioxins target levels, reducing contamination levels to less than 150 ppt TEQ. Soil sampling results confirmed that the treatment objectives were met, with a destruction rate efficiency of over 99% for all samples. The chosen design for vapor treatment consists of a direct-fired thermal oxidizer, which avoids any liquid/solid waste, and ensures that emissions meet Vietnamese, European, and US standards.

Table 2. *Toxicity Equivalence Factors (TEF)* of the most toxic dioxins and furans components

List number	*Most toxic dioxins and furans components*	*WHO-TEF*
1	**2,3,7,8-TCDD**	**1**
2	1,2,3,7,8-PeCDD	1
3	1,2,3,4,7,8-HxCDD	0.1
4	1,2,3,6,7,8-HxCDD	0.1
5	1,2,3,7,8,9-HxCDD	0.1
6	1,2,3,4,6,7,8-HpCDD	0.01
7	OCDD	0.0003
8	2,3,7,8-TCDF	0.1
9	1,2,3,7,8-PeCDF	0.03
10	2,3,4,7,8-PeCDF	0.3
11	1,2,3,4,7,8-HxCDF	0.1
12	1,2,3,6,7,8-HxCDF	0.1
13	2,3,4,6,7,8-HxCDF	0.1
14	1,2,3,7,8,9-HxCDF	0.1
15	1,2,3,4,6,7,8-HpCDF	0.01
16	1,2,3,4,7,8,9-HpCDF	0.01
17	OCDF	0.0003

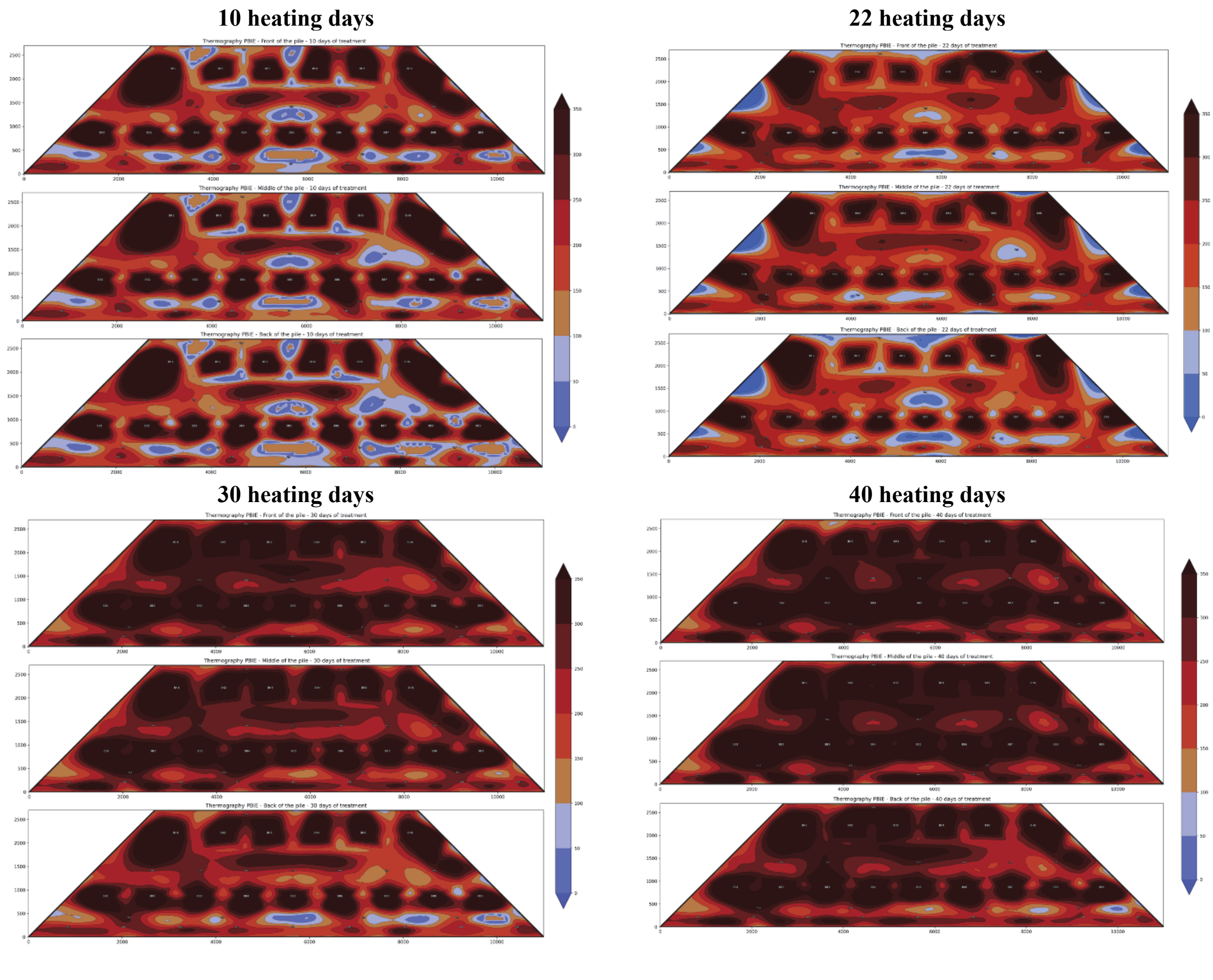


Fig. 7. Thermographies after 10, 22, 30 and 40 days of heating
(front, middle and back of the pile)

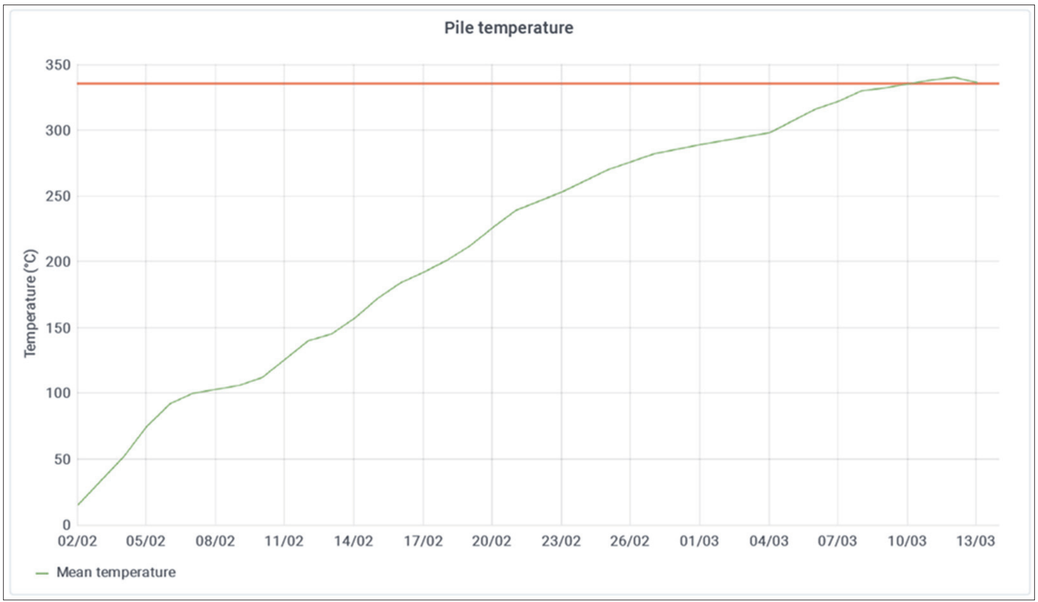


Fig. 8. Average temperature evolution in the thermal pile

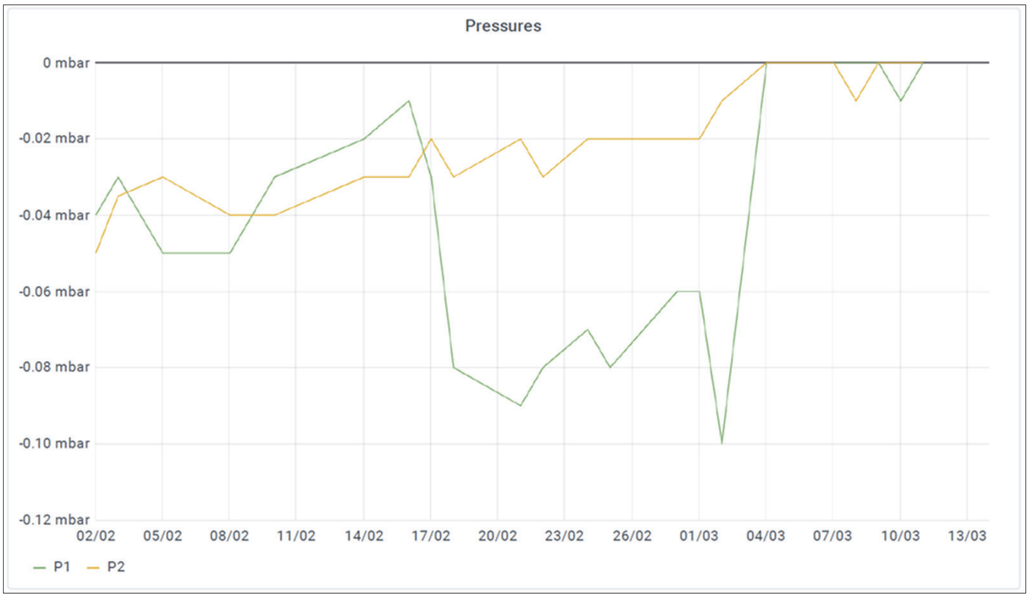


Fig. 9. Soil pressure monitoring during treatment

Table 3. Emission sampling results

Sample	*Unit*	*Concentration*	*Emission standard*	*Sampling date*
1	pg WHO-TEQ$_{2005}$/Nm^3	40.07	100	08/02/2022
2	pg WHO-TEQ$_{2005}$/Nm^3	73.52	100	11/02/2022
3	pg WHO-TEQ$_{2005}$/Nm^3	76.14	100	15/02/2022
4	pg WHO-TEQ$_{2005}$/Nm^3	1.38	100	18/02/2022
5	pg WHO-TEQ$_{2005}$/Nm^3	0.821	100	22/02/2022
6	pg WHO-TEQ$_{2005}$/Nm^3	51.38	100	25/02/2022

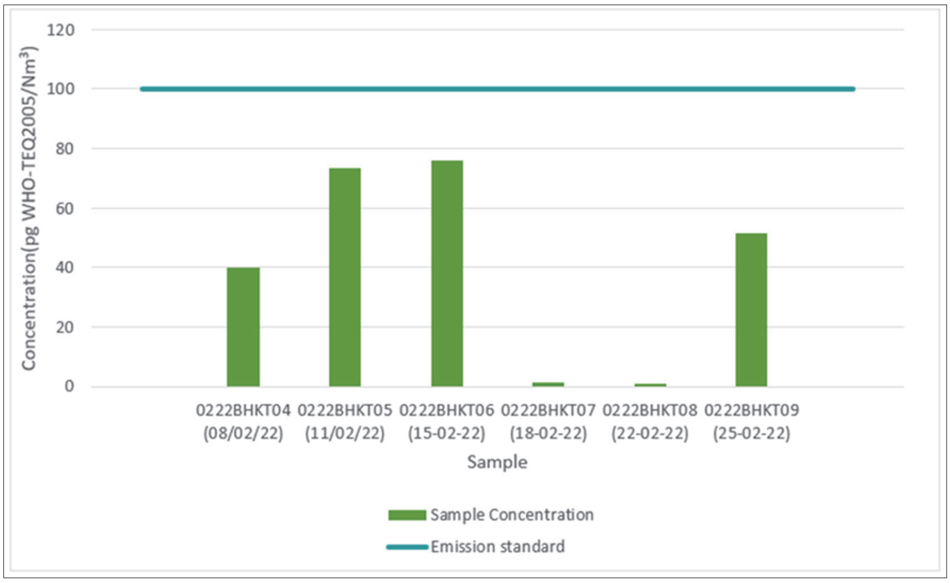


Fig. 10. Emission sampling results

Table 4. Treated soil analysis results

Material	***Unit***	***Initial concentration***	***Treatment goal***	***Treated soil results***	***DRE* (%)***
Soil	ppt WHO-TEQ2005	11,400	300 (Urban residential) 1,200 (Industrial)	4.84 96.29	99.96 99.16
Sludges	ppt WHO-TEQ2005	5,410	150	ND**	> 99.99
Washing cake	ppt WHO-TEQ2005	17,200	300	6.7	99.96

**Destruction Rate Efficiency; **Non-Detected*

Soil temperature, pressure, and emissions control were monitored throughout the treatment, and the results demonstrated the efficiency and success of the thermal treatment.

This zero-waste solution demonstrates an improved and efficient method for remediating dioxin-contaminated soils. Future design improvements could be made, such as adding a heat exchanger to the thermal oxidizer, in order to reduce the energy consumption of such a project.

Overall, the study provides a promising solution for the remediation of dioxin-contaminated soils, with the potential for broader applications in the future.

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