



# Human Health Risk Assessment of a landfill based on volatile organic compounds emission, immission and soil gas concentration measurements



Vicenç Martí<sup>a,c,\*</sup>, Irene Jubany<sup>a</sup>, Consol Pérez<sup>b</sup>, Xavier Rubio<sup>b</sup>, Joan De Pablo<sup>a,c</sup>, Javier Giménez<sup>c</sup>

<sup>a</sup> Fundació CTM Centre Tecnològic, Pl. de la Ciència, 2, 08243 Manresa, Spain

<sup>b</sup> Consorci Urbanístic del Centre Direccional de Cerdanyola (CUCDC), Passeig d'Horta, 66, 08290 Cerdanyola del Vallès, Spain

<sup>c</sup> Chemical Engineering Department, UPC, Universitat Politècnica de Catalunya, Avda. Diagonal, 647, 08028 Barcelona, Spain

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## ABSTRACT

A Human Health Risk Assessment (HHRA) was required for a closed landfill located in Cerdanyola del Vallès (Barcelona, Spain). The HHRA had two objectives, to evaluate the present risk of the identified receptors in the area and to safely develop the future urban planning of the area, therefore 3 scenarios for the current situation and 4 for the future situation were developed.

After reviewing the existing data and exploring the needs of information, the assessment in this study was focused on the measurement of volatile organic compounds (VOCs) fluxes from the subsoil (emission from the landfill at 5 points), concentrations of VOCs in the air (immission in 4 urban sites) and concentration of VOCs in soil–gas (measurements at 5 m below ground surface outside the landfill at 8 sites). Around 70 VOCs were analyzed by using multi-sorbent tubes and Thermal Desorption Gas Chromatography (TD–GC–MS). The VOCs that were detected and quantified include alkanes, aromatic hydrocarbons, alcohols, ketones, halocarbons, aldehydes, esters, terpenoids, ethers and some nitrogenated and sulfur compounds, furans and carboxylic acids. Specific mercury flux measurements were performed in a hot spot by using carulite tubes, that were also analyzed by using Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry.

Results showed average values of volatile emission fluxes ranging from non-detected to 331  $\mu\text{g m}^{-2} \text{day}^{-1}$  (dichlorodifluoromethane). In the case of immission, the concentration of VOCs measured in the air of populated area surrounding the landfill ranged values from non-detected to 42.0  $\mu\text{g m}^{-3}$  (acetic acid). The soil–gas measurements in piezometers around the landfill showed individual VOC values with a maximum 830  $\mu\text{g m}^{-3}$  for dichlorodifluoromethane.

With the obtained fluxes and concentrations in air and soil–gas, USEPA methodology and modeling was used to evaluate equivalent concentration in the scenarios considered. Toxicity values from IRIS database were used to finally obtain chemical risk indicators. Admissible risk indicators were obtained in all scenarios. The VOCs that contributed more to risk indexes in RH2 were trichloroethylene, trimethylbenzene, chloroform, 1,2-dichloroethane and carbon tetrachloride. The carcinogenic risk in RH7 was linked to the presence of benzene and chloroform. The comparison of the measurements of the present work with other landfills evidence that HHRA in ambient air would be needed in order to perform a correct landfill management.

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## 1. Introduction

The presence of VOCs and other trace contaminants in subsoil due to landfills, contaminated soils or contaminated groundwater is an important source of risk, as these contaminants could remain after an incomplete remediation of these sites for new uses.

In the case of the landfills, the characterization of emitted trace gases is an important topic for Risk Assessment (Butt et al., 2008) and measurements have to be focused on trace pollutants like VOCs,

\* Corresponding author at: Chemical Engineering Department, UPC, Universitat Politècnica de Catalunya, Avda. Diagonal, 647, 08028 Barcelona, Spain. Tel.: +34 93 401 09 57; fax: +34 93 401 71 50.

E-mail addresses: [vicens.marti@upc.edu](mailto:vicens.marti@upc.edu), [vicens.marti@ctm.com.es](mailto:vicens.marti@ctm.com.es) (V. Martí), [irene.jubany@ctm.com.es](mailto:irene.jubany@ctm.com.es) (I. Jubany), [cperez@parcdelalba.cat](mailto:cperez@parcdelalba.cat) (C. Pérez), [xrubio@parcdelalba.cat](mailto:xrubio@parcdelalba.cat) (X. Rubio), [joan.de.pablo@ctm.com.es](mailto:joan.de.pablo@ctm.com.es), [joan.de.pablo@upc.edu](mailto:joan.de.pablo@upc.edu) (J. De Pablo), [francisco.javier.gimenez@upc.edu](mailto:francisco.javier.gimenez@upc.edu) (J. Giménez).

mercury and sulfur species, depending on the kind of waste buried and the processes (aerobic, anaerobic) involved in the landfill.

Mercury is directly linked to wastes with batteries, fluorescents and thermometers and is usually characterized as elemental vapor form (Kim et al., 2001; Kim and Kim, 2002). Methylated species can also be characterized (Lindberg et al., 2005) but usually represent small percentage of total mercury (Lindberg et al., 2001).

Volatile sulfur compounds (VSCs) or reduced sulfur compounds (RSCs) are sometimes most dominant components in air other than trace carbon compounds (Kim et al., 2006b) and this predominance is usually linked to malodorous problems (Kim et al., 2005).

The emission of VOCs from the landfills allows to describe the release of these contaminants from a specific source, a landfill, to air. As HHRA of VOCs finally needs to evaluate the indoor and outdoor concentrations in defined scenarios, the direct immission measurements or modeled concentrations in air are needed. Emission and soil–gas measurements could be used for the modeling of air quality in scenarios linked to the landfill, while immission values in air could be directly used for HHRA and include not only the landfill emission, but also other sources of contamination that influence in these points.

In the case of contaminated soils studies, the calculation of indoor and outdoor concentrations is usually performed by modeling from the concentrations of VOCs in soil and groundwater located at a specific depth. From these concentrations equilibrium is assumed of each VOC with soil, water and soil–gas by using simple models as Henry's law or linear adsorption in soil (ASTM, 1995; WHI, 2001). From these equilibrium values, diffusion models through porous media are usually applied to finally calculate fluxes and indoor and outdoor concentrations.

In the case of a landfill, it is difficult to locate a single source of contaminants at a specific depth because the contamination could be diffuse. Furthermore, compared with contaminated soil, the material of the landfill could be more heterogeneous and cracks could be present, thus other mechanisms of transport, such as direct advection of contaminated soil–gas with VOCs could happen and make difficult the application of porous models.

In order to evaluate the concentration of VOCs in landfills, traditional approaches include the measurement of concentration of trace gases emitted from the landfill (Brosseau and Heitz, 1994) and the quality of air on the landfill (Durmusoglu et al., 2010; Kim et al., 2006a; Ding et al., 2012; Lee et al., 2002; Zou et al., 2003). From these two alternatives the second approach can be used for HHRA, but concentrations could be very dependent on the atmospheric conditions and some contaminant values could be very low. This implies that the sampling time has to be high enough to determine these low values and, thus, the equipment needs very high autonomy.

A good alternative for HHRA could be the measurement of fluxes from the landfill by using flux chambers, that have been used in some references (El-Fadel et al., 2012; Barlaz et al., 2004; Scheutz et al., 2008; Gallego et al., 2014). VOCs determination in landfills using a flux chamber consists in accumulating the emitted flux of VOCs in a chamber and displacing them to the adsorption beds by using an inert gas. The measurement of fluxes allows knowing the release of specific contaminants to the air and can be obtained in few hours, since the VOCs released are captured in the chamber. In this way the measured concentrations are more precise and higher compared with immission. Using this approach, the complex modeling of VOCs in the subsurface is avoided and flux values could be combined with single box models to estimate the concentration of VOCs on the landfill or use these data as inputs in dispersion models to evaluate the impact outside the landfill area.

The direct measurement of VOCs in outdoor air (immission) allows obtaining a value ready for HHRA. In order to be representative, these measurements have ideally to cover several moments

of the day, several days and several seasons. The VOCs obtained by using these methods are representative of all the sources that impact in the point of immission.

The measurement of VOCs in soil–gas at a specific depth is a good approach to evaluate points outside the landfill. From these VOCs values in soil–gas, the transport models based on diffusion through soil and structures around the building could be applied and, thus, air quality for future uses of soil could be calculated (Johnson and Ettinger, 1991).

All these three approaches need to use sampling devices for adsorbing VOCs into multi-sorbent tubes and release of these compounds to Gas Chromatography equipped with Mass Spectrometer to identify and quantify the individual compounds. Several applications allow determining a wide range of VOCs in air at very low concentrations (Ribes et al., 2007; USEPA, 1999).

In the proposed landfill case study, the concentration of measurements of VOCs in soil, soil–gas and groundwater from 1995 to 2008 was reviewed (IDOM, 2008a,b). These VOCs included contaminants as alkanes, aromatic hydrocarbons, halocarbons, alcohols and mercury at specific points and showed an important spatial heterogeneity, but most of the samples were measured in the landfill as concentrations and do not help to define the emission in terms of flux. Furthermore, several gaps were found as there were no immission measurements in the surrounding air of the landfill and soil–gas measurements in the subsoil outside the landfill were not determined.

In the present study, measurements of VOCs emission fluxes combined with concentrations in air and soil–gas were performed in order to obtain more accurate input values for the HHRA with respect to the typical approach. The application of this new approach will avoid all the uncertainties linked to number and type of sources, heterogeneity of the landfill subsoil and transport mechanisms.

## 2. Materials and methods

### 2.1. Case study and sampling strategy

The landfill consists of a former clay extraction activity area that caused the existence of three 16–40 m deep holes which were later filled with industrial waste during the 80s: melting salts, industrial sewage sludge, polluted soils and non-hazardous wastes. After its closure in 1995, several studies were performed in the area (IDOM, 2008a) to assess the effect of the buried wastes on the environment and concluded that VOCs were relevant. Fig. 1 shows the landfill and surroundings. The landfill has limited access to the public and is very close to a working industrial area (in SE) and to the city of Cerdanyola del Vallès (NE).

More detailed characteristics of this landfill and atmospheric conditions can be found in Gallego et al. (2014).

The determination of VOCs was planned to be quantified as emission fluxes in 5 points placed on the landfill designed as FG samples in Fig. 1. Outdoor air stations for immission measurements were placed at 4 urban sites around the landfill (PI samples in Fig. 1) that include the closest urban and industrial use of land. The composition of soil–gas was determined at 5 m below ground surface using old and new wells placed outside the landfill (8 sites designed as SG samples).

The sampling campaign was complemented by taking superficial soil samples on the landfill (SS samples) and groundwater in new wells (see Fig. 1).

### 2.2. Air sampling and analytical methods

The procedures for sampling were different for each approach, but they use the same elements and the same procedure of

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