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A comparison of models for assessing human risks of petroleum hydrocarbons in polluted soils

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ABSTRACT

Petroleum hydrocarbon products can be released to the media, affecting human health and the environment. The hydrocarbon compound mixture is typically evaluated using the Total Petroleum Hydrocarbons parameter, divided into fractions. Several risk assessment tools are available based on different models. The Risk Based Corrective Action (RBCA) Tool Kit, Risk-net and CSOIL tools were selected and compared and inter-model differences have been discussed for a real case of study. The RBCA and Risk-net risk assessment models yield nearly identical HQs, while the CSOIL model, based on different parameters, estimates lower HQs. The ingestion of contaminated water is the primary route of exposure determined using the RBCA Tool Kit and Risk-net tools; the CSOIL tool suggests that the ingestion of contaminated soil poses the highest risks. Each model better describes a different site assessment scenario. However, the CSOIL model was found to best represent the identified site-specific conditions, and thus acceptable risks were determined for this site.

1. Introduction

Soil contamination can pose risks to human health, ecosystems and groundwater (Swartjes et al., 2012), which may arise from a wide range of sources. The European Union estimated that there were approximately 3.5 million potentially contaminated sites in 2006 (COM, 2006). More than 50% of these sites were affected by mineral oil, polycyclic aromatic hydrocarbons (PAH) or volatile aromatic hydrocarbons, i.e., benzene, toluene, ethylbenzene and xylenes (BTEX) (EEA, 2011). These substances can be found in petroleum products such as gasoline, diesel and lubricants, which can be released and spread into the soil. The total petroleum hydrocarbons (TPH) parameter is broadly applied to determine the measurable amount of petroleum-based hydrocarbons in the environment, including many hazardous substances from C10 to C40 (ISO, 2004). The TPH detected in a sample is a general indicator of the presence of contamination at a site due to petroleum product leakages (ATSDR, 1999).

Soil quality is established in several countries using threshold limit values (Carlon, 2007). For example, Spanish regulations define the Reference Generic Level (RGL), set as the contaminant concentration in soil that does not result in a risk level higher than the acceptable maximum for human health or ecosystems. Regarding

TPH assessment, the RGL defined by Spanish regulation is 50 mg/kg (Presidency Ministry, 2005). When the TPH concentration exceeds 100 times the established RGL for the protection of human health (5000 mg/kg for TPH assessment), the risk level is assumed unacceptable and the site is declared to be seriously contaminated. Between these two levels, a site-specific assessment must be performed to determine the risks. Dutch regulations propose a screening Intervention Value (IV), which defines a maximum TPH limit of 5000 mg/kg (VROM, 2012). Therefore, soil quality can be established through a comparison of the measured concentrations and the screening values described in national regulations (Carlon, 2007; Ferguson, 1999; Pinedo et al., 2013a).

Total TPH measurements include compounds with very different physico-chemical and toxicological properties. Therefore, TPH is not a useful proxy for potential risks (ATSDR, 1999). In addition, volatile hydrocarbons from C5 to C10 are not included in the total TPH parameter. However, these lighter compounds have higher human and ecotoxicological risks (ISO, 2012). Previous studies (Pinedo et al., 2013b) have established a relationship between concentrations and risks for different TPH fractions to determine risk levels without performing a site-specific risk assessment in many scenarios by considering representative substances including PAH and BTEX. However, a fractionation procedure is necessary to quantitatively determine the petroleum hydrocarbon effects on humans (Lijzen et al., 2001) and the ecosystem (Verbruggen et al., 2008). Several fractionation methods have been proposed (MassDEP, 1994; NJDEP, 2010; Weisman, 1998)

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based on the environmental behavior and equivalent carbon number (EC) of the individual substances. This fractionation is based on the constituent averaging technique, which uses a composite constituent to simulate the average behavior and fate of a group of individual compounds (Gaganis et al., 2002). Aliphatic and aromatic fractions are separately defined because they have very different environmental behaviors. Some studies have been conducted with the same scope, adapting the fractionation method to the needs of the specific study (Park and Park, 2011; Pinedo et al., 2012a). Moreover, TPH fractions are considered to pose only non-carcinogenic adverse health effects (ATSDR, 1999).

Quantitative environmental models are widely used for risk assessment and subsequent environmental decision support systems (EDSS) (Bennett et al., 2013). The steps taken in the development and evaluation of environmental models are model purpose definition, modeling context specification, system conceptualization, model features selection, determination of how model structure and parameter values are to be found, estimation criteria and algorithm selection, verification (including diagnostic testing), quantification of uncertainty and model evaluation (Jakeman et al., 2006). Literature is available in regard to characterizing the performance of established environmental models (Bennett et al., 2013). EDSS can be defined as software systems in which models, databases or other decision aids are integrated for decision-maker use (Rizzoli and Young, 1997). EDSS may lead to failures when adopted by intended end users (McIntosh et al., 2011). To be successful in the decision-making step, it is necessary to obtain reliable risk assessment results.

Risk assessment determines the need of an environmental cleanup, quantifying the probability and the hazard of contaminants to human health and the environment (ISO, 2012). The Human Health Risk Assessment (HHRA) is based on predictive tools to characterize and quantify the chance of adverse effects on human health associated with the exposure to contaminated media by any particular substance on current and/or potentially future receptors. Risk assessment includes understanding the nature of the exposed chemicals, magnitude, time, transport mechanisms, routes of exposure and the sensitivity of potential receptors (USEPA, 2012).

Recently, many risk assessment models and tools for human health have been freely or commercially developed throughout Europe, such as CETOX-human and JAGG in Denmark; CLEA, SNIFFER, RAM, ConSim and RISC in the United Kingdom; CSOIL and RISC-HUMAN in The Netherlands; LUR in Spain; DESYRE, Risk-net and ROME in Italy; UMS and SISIM and CARO-PLUS in Germany; and Vlier Humaan in Belgium (APAT, 2008; ASTM, 2004; Brand et al., 2007; Carlon, 2007; Carlon et al., 2007; Cheng and Nathanail, 2009; Connor et al., 2009; Hayward and Baker, 2013; Khan and Husain, 2003; Stewart and Purucker, 2011; Swartjes et al., 2009). The RBCA Tool Kit for chemical releases was developed by the American Society for Testing Materials (ASTM) in the United States (US). However, the tool is globally utilized.

Some harmonization to improve the consistency of risk assessment tools in the European Union Member States must occur (Swartjes et al., 2009). Moreover, model uncertainties and sensitivities should be clearly known to understand the derived risks for any proposed scenario (Kumar et al., 2009; McKnight and Finkel, 2013). Therefore, a model comparison should be performed to determine model strengths and weakness. However, it is not practical to review all models and tools. As a result, the HHRA tools considered are the RBCA Tool Kit and CSOIL because they are currently commonly used in Spain. Additionally, the Risk-net tool was also included because it is based on the ASTM-RBCA model and adapted to the Italian framework. Tsai et al. (2011) defined the ASTM-RBCA model as the most effective and widely used standardized guide for risk assessment. The RBCA framework develops

site-specific environmental cleanup criteria using a tiered risk evaluation approach. This approach has been applied to geographical sites and managed under various regulatory authorities. For example, the Italian APAT-ISPRA guideline for risk analysis application (APAT, 2008) is based on the ASTM-RBCA guideline. Moreover, the CSOIL model, implemented in the Netherlands (Brand et al., 2007), is widely applied in Europe. These models have been specifically developed to analyze contaminated land. Several studies have been performed using the ASTM-RBCA model (Iturbe et al., 2005; Tsai et al., 2011; Park and Park, 2010, 2011; Pinedo et al., 2012b) and the CSOIL model (Song et al., 2012; Popescu et al., 2013). The Risk-net tool was developed in 2012 (RECONnet, 2012). Nonetheless, no study exists in which the specific features of each model were compared and assessed to understand the consequences of applying a specific model in the decision-making process.

This work contributes to the assessment of polluted soils due to the presence of petroleum hydrocarbons by means of a real case of study, comparing and evaluating a selection of available models. The study area is located near a petrol filling station in a highly populated area of the Cantabria region (Spain). In previous works (Ibáñez et al., 2010; Pinedo et al., 2014), 165 activities related to hydrocarbon storage and distribution were evaluated. In this study, three out of six soil samples at the selected site had values exceeding the RGL of 50 mg/kg, one had a TPH concentration greater than 1000 mg/kg. The main objective of this work is accomplished via two steps. The first step comprises soil sampling and analysis, including the design and implementation of sampling campaigns and analysis. The second step is performed by applying different assessment tools, evaluating the obtained results and discussing the model applications and sensitivities. The selection of a proper model is necessary to determine reliable site-specific risks to human health caused by contaminated sites and to help and support the decision-making process (Bowers, 2012; Kelly et al., 2013).

2. Methodology

The methodology is divided in two steps. The first step includes soil sample collection and laboratory analysis, with the objective to characterize the soil in terms of total TPH, fraction concentrations and VOCs when necessary. The second step is performed for the samples that have the highest concentrations. This step is focused on human health risk assessment using specific assessment tools.

2.1. Soil sampling and laboratory analysis

An exploratory investigation of in-situ soil samples was performed. The sampling campaign strategy was based on the international ISO standard 10381-1 (2005b), considering previous results (Pinedo et al., 2014) and physical limitations (i.e., paved areas and bedrock). Samples were taken near a petrol station with four underground storage tanks (UST) and five small surface deposits. Two USTs contained gasoline and the remaining two contained diesel. The surface deposits were a gas cylinder and brake fluid, antifreeze, motor oil and cleaning water tanks, respectively.

Site conditions dictated an appropriate sampling strategy. Eight soil samples were collected in locations where the soil morphological characteristics were favorable for pollutant migration and contamination (Pinedo et al., 2014). The sampling point distribution and petrol station locations are shown in Fig. 1. Soil samples were taken at depths between 10 and 30 cm. Samples were extracted with a hand auger, consisting of extendable stainless steel auger buckets with cutting heads that rotated using a handle. The auger was rotated into the soil until it was filled. The auger was then withdrawn from the hole and the soil was placed in a wide-mouth sample container using a spatula. Special care was taken not to cross-contaminate the samples. The samples were stored at a low temperature (4 °C) until analyzed.

The analytical procedure is summarized in Fig. 2. All steps followed in the laboratory analysis were based on the international ISO standards adopted in previous works (Pinedo et al., 2012a). Table 1 presents the methods applied for the individual laboratory analysis. After sample reception, the soil was pretreated via freeze-drying and sieving (ISO, 2005a). The TPH measurement method was based on international ISO standard 16703 (2004), allowing hydrocarbons in the range C10–C40 to be analyzed. The TPH analytical procedure comprised the following steps: solid–liquid extraction with acetone/n-heptane, liquid–liquid extraction to separate the organic