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RECENT DEVELOPMENTS IN SITE SPECIFIC RISK ASSESSMENT FOR POLLUTED SITES*

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Abstract

The present note highlights the role of geotechnical expertise for Risk Assessment of contaminated sites and illustrates specific aspects related to migration modeling. Implications of neglecting the time variable and the advantages of the direct measurements of vapors, performed by application of different techniques, are described and discussed. The comparison between direct measurements of vapor emissions and modeling outcomes show how the use of measured data is able to overcome the limitations deriving by restrictive model assumptions and effectively helps in obtaining more realistic results.

Keywords: polluted site, risk assessment, site conceptual model, migration models

1. Introduction

Among the challenges that the future poses to geotechnical engineers, the safeguard and the preservation of the environment are surely included (Viggiani, 2015). In this perspective, the study of the effects of pollution on soil and groundwater behavior is one of the main topic for geotechnical researchers. Polluted sites represents a topical problem all over the world and risk assessment (RA) for contaminated sites is the tool that allows for verifying that risks associated with contaminated soil or groundwater at a particular site are tolerable.

In Italy, as in other countries, after a triggering initial comparison with screening levels (JRC, 2007), the need of a remediation action and the related remediation goals are

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defined by means of the site-specific RA procedure. The specific procedure for RA, initially developed in the United States, was subsequently adopted in Europe, where about 2.5 millions contaminated sites were estimated in 2011 by the European commission EIONET (Panagos et al., 2013). In Italy, where about 15000 polluted sites were assessed by the National Environmental Protection Agency (ISPRA, 2012), RA was allowed by regulations on contaminated sites since 1999 (Legislative Decree 471) and became mandatory in 2006, after the Legislative Decree 152. The first version of guidelines for RA application was issued in 2005 and the latest update was released in 2014.

The RA procedure starts from the construction of the Site Conceptual Model (SCM), which consist of three components: source of contamination, migration paths and targets (or receptors) and then enables the risk/hazard index calculation. The migration mechanisms of contaminants from the source to the targets represent the core of the procedure and are modeled through analytical models in which site-specific parameters are used (Tier 2 RA) (ASTM E2081-00; APAT, 2008). The selection of appropriate migration models and the use of site-specific measurement of parameters help obtain a more realistic risk estimate.

At present, special efforts are needed from researchers, aiming at both the improvement and the refinement of the migration models, basing on solid scientific fundamentals and validating the proposals with site data. The main objective of this study is to examine the major factors affecting the assessment outcomes, by analyzing some of the existing migration models and proposing alternative models.

To this aim (1) the leaching process was analyzed by comparing stationary and transient models of migration, (2) a case study including the comparison of stationary and transient lateral transport in groundwater and sensitivity analyses is described (3) direct measurements of vapor emissions were discussed and compared with the predictions of an analytical model with reference to a second actual polluted site.

2. Site-specific health and environmental risk assessment

Risk assessment is the estimate of the effect on human health of a potentially harmful event, in terms of probability that the effects themselves occur (APAT, 2008). RA calculations with reference to polluted sites start from the definition of the Conceptual Site Model (CSM) and the description of the three components: source of contamination, migration paths and targets of contamination. Based on the CSM and through the migration models, it is possible to calculate the exposure of the targets to contamination (E). In particular, in addition to the direct exposure of targets (i.e. ingestion of contaminated soil and dermal contact), chemicals could reach the different targets via volatilization (from soil or from groundwater), leaching from soil with lateral transport in groundwater and particulate emission from surface soil. These different types of migration are described by the fate and transport factors (FT) that directly derive from the analytical models of migration and whose formulations include site-specific parameters. Each different FT, multiplied by the representative source concentration (RSC) gives the concentration at the point of exposure (C_{POE}). The product between C_{POE} and the specific exposure, EM (characteristic of each type of target) gives, in turn, the exposure, E.

The definition of "risk" related to contaminated sites is derived from the general formulation of risk as the product of the damage connected with the occurrence of an event, D, and the probability of the event to happen, P, that is equal to 1 (contamination has already happened = certain event). The damage, D, is in turn defined as the product of a factor of danger, FD, represented by the toxicity of the contaminant, T, and a contact factor, FC, represented by the exposure, E, calculated with the CSM, as previously mentioned. The specific expression of risk for contaminated sites is then:

$$R = P \cdot (FD \cdot FC) = T \cdot E \qquad (1)$$

Risk values are differentiated between risk, R (for carcinogenic effects), and hazard index, HI (for toxic not carcinogenic effects). Backward application of the RA procedure allows calculating the Clean Up Levels (CLs) by fixing maximum tolerable risk values (target values for backward application) suggested by regulations (Italian values: R=1·10-6; HI=1). Beside the risk assessment for human targets (i.e. Health RA), Italian regulations require to consider also the groundwater as a receptor, calculating the related risk as the ratio of the concentration at the site boundary to the regulatory screening level (CTC). The risk is tolerable if the ratio is lower than 1: this last calculation type is named "Environmental RA". The backward application can be also performed, calculating the CLs in soil for groundwater protection.

In other jurisdictions (e.g. USA), contaminated groundwater can reach human targets and the related risks (e.g., through ingestion of contaminated water or showering) must be taken into account, in some countries concentration limits in groundwater are prescribed to protect the ecosystem (JRC, 2007). For both the forward and the backward applications, additional criteria are defined in the guidelines to consider more than one exposure type at a time and the presence of more than one contaminant in the same site (APAT, 2008; ASTM, 2010).

3. Geotechnical aspects of the risk assessment application

The geotechnical expertise, as well as the geological and the eco-toxicological ones, is essential to build up a CSM that suits well with the framework of the Reasonable Maximum Exposure philosophy suggested by the RA main guidelines. In particular, the CSM should be set up on the basis of a site characterization that includes:

- topographic survey;
- boreholes and soil sampling following the environmental sampling procedure, to define the geotechnical model, the contaminant distribution in soil and soil chemical characteristics (e.g. fraction of organic carbon);
- in-situ tests to outline the geotechnical model such as Lefranc or Lugeon tests to assess the different type and permeability levels of an aquifer (Di Sante et al., 2012) or CPT equipped with special sensors to rapidly detect the presence of pollutant (Fratalocchi and Pasqualini, 2007);
- piezometers for groundwater sampling and hydrological measurements to know the aquifer type, groundwater flow, hydraulic gradient;
- geophysical surveys if the presence of buried waste or tanks as primary sources of contamination is suspected.

The site model for risk assessment is based on classical geotechnical investigation techniques coupled with environmental investigation and analysis methods as it also aims at quantifying the contamination and its spatial distribution both in the unsaturated zone and in groundwater. In fact, sampling procedures are different from that of typical geotechnical investigation: undisturbed sample are usually not required but the sampling must comply with protocols for contaminant substances (APAT, 2008) and each borehole represents an area defined by means of Thiessen polygons. The risk of contaminant diffusion due to investigation activities is possible (e.g. cross-contamination between two aquifers during piezometers installation), therefore, particular precautions should be taken during installation of the investigation points. In addition, if volatile compounds are present, the installation of investigation points that allow the sampling of vapors is considered worthwhile and the screening of the piezometers should be extended to the unsaturated level.

Subsoil conditions can often significantly differ from the ideal ones considered in the RA applications (Figure 1) and in these cases, the definition of the geotechnical model has a key role in the procedure.

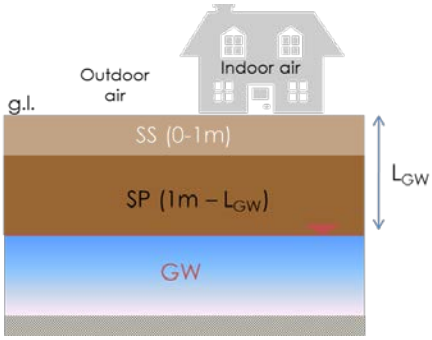


Fig. 1. Standard subsoil model for RA and related environmental media (i.e. surface soil, SS, and deep soil, SP, groundwater, GW, outdoor and indoor air); L_{GW}= depth of groundwater table from g.l.

The number of investigation points is usually much higher than that of typical geotechnical studies because they are essential to describe the contaminants distribution and to draw detailed geological sections. These sections are especially useful to (Di Sante et al., 2019):

- verify the spatial continuity of low permeability deposits affecting migration of contaminants;
- detect a non-horizontal ground surface and multi-layered aquifers, with the possible presence of lenses, that sometimes prevents adopting the simplified subsoil configuration;
- identify the possible presence of a fractured aquifer (e.g. calcareous rocks) needing the use of numerical modeling in place of the analytical ones.

In all these cases, the assessor should be aware of all the modeling possibilities offered by different computer codes to select the most suitable model to represent the actual site conditions and the possible migration modes.

In defining the representative depth of the phreatic level, L_{GW}, it is important to consider its seasonal variability. Therefore, multiple surveys are recommended, at least over a year, so that a cautionary value can be selected by the assessor for each migration model (e.g., volatilization from groundwater requires to consider the minimum L_{GW}, while lateral transport in groundwater requires to minimize the thickness of the groundwater body, thus to consider the maximum L_{GW}).

4. Fate and transport models

Analytical migration models of contaminants have the great advantage to be simple, thus easy to use. This latter feature is ensured by the simplifying assumptions on which the models are based (such as homogeneous physical, mechanical and hydraulic characteristics of the media involved, no source depletion and no biodegradation), but due to these hypothesis, analytical models may sometimes provide unrealistic or too conservative predictions (Bretti and Zanetti, 2014; Verginelli and Baciocchi, 2014). The use of site-specific parameters (i.e. measured on soil samples or derived from in-situ investigations) surely helps in making model results closer to reality.

Environmental Protection Agencies are also aware of this occurrence; in fact, the latest update (2014) of instructions for RA application allows the use of measured in situ volatilization data (with multiple lines of evidence) to verify the results of the analytical models. Moreover, the possibility to exclude the leaching path in particular conditions (i.e., geological and hydrogeological characteristics of the subsoil that prevent migration, absence of correlation between contamination in unsaturated soil and in groundwater, execution of standardized and validated leaching tests) is introduced.

5. Steady-state and transient leaching and lateral transport models

In Italy, groundwater bodies are considered of intrinsic environmental value regardless of their use and therefore, according to Italian regulations, have to be protected. If the contaminant source is located in the vadose zone, the risk assessment requires analytical modelling of contaminant leaching due to percolating rainwater and subsequent mixing with groundwater and lateral transport to the control point (located at the site boundary in the direction of groundwater flow). If the source is already dissolved in groundwater, only lateral transport considering advection, dispersion, diffusion and adsorption has to be simulated.

The guidelines and the commonly used risk assessment codes (e.g. ASTM E2081-10) adopt a steady-state approach to model transport of contaminants to/in groundwater.

If the source is located in the vadose zone, after linear equilibrium partitioning in the source, generic attenuation of the concentration is assumed along the path towards the groundwater by means of the Soil Attenuation Model coefficient (SAM < 1) which decreases with increasing path length. Dilution of the contaminant in groundwater is accounted for by a Leachate Dilution Factor (LDF > 1) which divides the source concentration and is related to the infiltration rate and to the permeability value of the aquifer.

A transient model developed by the writers and fully described in Mazzieri et al. (2016) takes into account:

- depletion of source concentration with time due to volatilization and leaching losses;
- one dimensional (vertical) advective-dispersive flux of the dissolved contaminant with linear sorption and first order biodegradation along the path;
- transient dilution of the contaminant front in groundwater.

With reference to a hypothetical site configuration, shown together with the graph in Fig. 2, the trend of the results of both the steady-state and the transient model is observable, in case of migration of benzene.

Both models predict that the screening level (CTC) for groundwater prescribed by the Italian regulations (0.001 mg/L for benzene) is exceeded. However, the transient approach (grey lines) predicts that the CTC would be exceeded after 12 years from the end of characterization, and this time interval is sufficient to design and to undertake the site remediation, in addition the peak of concentration (0.33 mg/L) at the POC (Point of Compliance) is reached after 53 years. On the contrary, by adopting the steady-state approach, no information can be obtained on the time at which the threshold concentration is exceeded, the concentration value at the POC is 0.606 mg/L (dotted black line in Figure 2). Moreover, the transient approach predicts a significant reduction in the dissolved source concentration (grey solid line in Fig. 2) whereas in the steady-state approach the source concentration is assumed to remain constant (black solid line).

Although steady-state models are simpler and easier to be applied, taking into account the time variable is essential in order to obtain information about the time span during which soil remediation activities must be concluded. If the source is located in the saturated zone, i.e. the contaminant is already dissolved in the groundwater and its concentration exceeds the CTC within the site boundary, the model suggested by the Italian guidelines to simulate the dispersion in the groundwater is the Domenico equation (Domenico and Shwarz, 1998). It generally derives the concentration distribution in a 3D domain and different solutions of the equation are possible depending on different boundary conditions applied. Beside this stationary model, in the software RISC, the migration through groundwater from an already dissolved source is modeled with a transient analytical model with a mass loading rate from the source zone calculated as a function of hydraulic conductivity (Yeh, 1981).

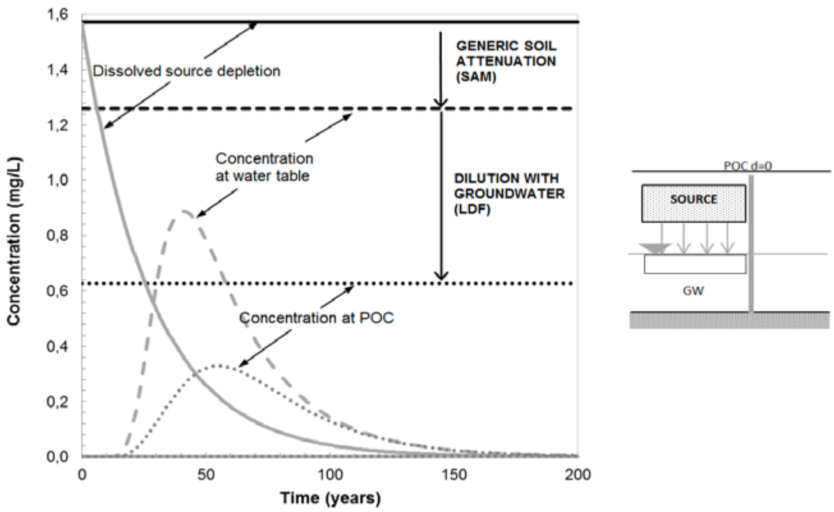
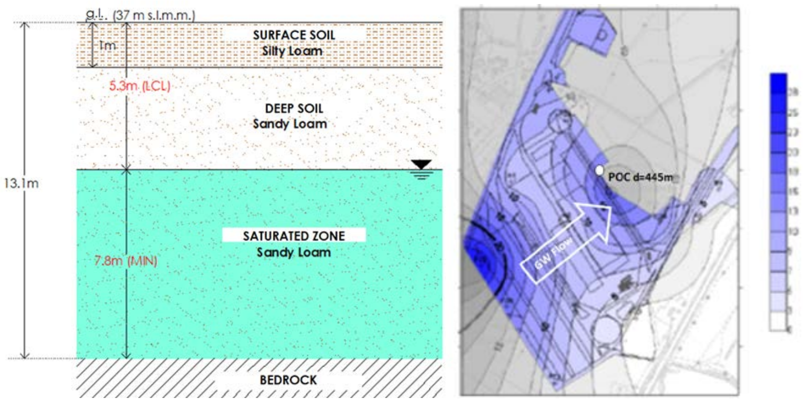


Fig. 2. Comparison between steady state model (black lines) and transient model (grey lines) referred to the shown example site configuration and to leaching of Benzene, starting from a representative soil source concentration of 1 mg/kg.

In order to compare the models' outcomes and to quantify the influence of changes in input parameters on the model results, a peculiar case study with a sensitivity analysis is considered in the following. The site is located on an alluvial deposit (left bank of a river) and the current activity developed on the site is that of an Intermodal Logistics Centre. In the past, agricultural activity was carried out in the site. During monitoring controls required by legislation to evaluate the environmental impact of new buildings to be constructed within the site boundaries, some pollutants whose concentrations exceed the screening levels (SLs) were detected in groundwater. Among them, Nickel was found to exceed the CTC of 20μg/L in 1 out of 14 available monitoring wells (6 already present in the site plus 8 boreholes equipped as wells during the characterization activities).

The subsoil conditions are schematically depicted in Fig. 3a. The permeability of the alluvial layer was characterized by means of 3 Lefranc tests. Only one test gave results for *k*, that was equal to $1.5 \cdot 10^{-5}$ m/s. During the other two tests it was not possible to measure hydraulic levels due to the high permeability of the aquifer, therefore the measured value was assumed to represent the lower boundary of permeability, not ensuring cautionary results if used in the simulation. This represent one of the major uncertainties in predicting contaminants concentration at POC, as required by the RA procedure in this case.

L_{GW} ranged from 5 to 7m and the isophreatic contours suggested that the main drainage axis direction is SW-NE (Fig. 3b). Another peculiar aspect of the site was that soil concentrations (including Nickel) were found to be lower than the threshold concentrations (CTCs) in all the taken samples. In 2011-2012 local Environmental Agencies found the same contaminant exceeding CTCs in the upstream groundwater flow during periodic monitoring, as demonstrated by publicly available records. This evidence together with the complete absence of contamination in the unsaturated soil led to the hypothesis that the contamination was entering the site with the groundwater flow.



a) b)

Fig. 3. a) Schematics of the subsoil stratigraphy adopted for the site conceptual model and b) plan view with concentration contour curves

The conceptual model for the site of concern starts form a source, represented by the polluted groundwater plume, which was geometrically defined as the most external CTC contour (drawn by means of surface modeling software Surfer ver.8.0 – Figure 3b). The representative source concentration was 28 μg/L and the physico-chemical and toxicological properties of Nickel were assumed from ISS and INAIL (Italian National Institute for Health protection of citizens and workers) (database ISS, 2013). The lateral transport in groundwater was simulated as possible migration path using both the stationary and the transient model previously described. Site-specific parameters used in the simulation are listed in Fig. 4. The only receptor considered for Nickel contamination was the groundwater at the POC (Nickel is not volatile, therefore human receptors cannot be reached through the "volatilization from groundwater" path).

The results of both the transient model of migration of Nickel implemented by RISC and of the stationary model suggested by Italian guidelines were obtained by varying the value of saturated hydraulic conductivity, *k*, from the measured value to higher ones (maximum k value= $2.3 \cdot 10^{-4}$ m/s, suggested by RISC manual for Gravel deposits). The distance, d, of the source to the control point is depicted in Fig. 3b. At this point, according to RISC results, using the *k* value from Lefranc test the contaminant will not be detected before 10000 years, while considering the maximum value of *k*, no Nickel will be detected until 3700 years. A concentration value of $2.4 \cdot 10^{-7}$ mg/L will be reached at the end of the simulation; this maximum value is far lower than the threshold limit for groundwater for Nickel, highlighting a tolerable value of risk.

Comparing the results from the two types of model (Figure 4) for the control point, the stationary model predicts a concentration of $1.58 \cdot 10^{-2}$ mg/L, 5 orders of magnitude higher than the maximum concentration value given by the transient model, but still lower than the CTC for groundwater too. This extremely slow migration is probably due to the high value of Nickel soil-water partition coefficient, k_d, that was estimated with the correlations suggested in the database ISS-INAIL (2018) as a function of pH (pH measured during characterization = 7.9 – estimated as LCL 95% of the Mean). The employed k_d is equal to 1400 ml/g, thus substantial adsorption on soil particles occurs along the migration pathway.

It is important to point out that monitoring of concentration at control point (an additional well was installed, as requested by the local Environmental Protection Agency) was scheduled and, during the subsequent 2 years, values always lower than the CTC were detected.

Parameter	units	value
Height of capillary fringe	m	0.25
Soil bulk density [a]	g/cm^3	1.7
Total porosity [a]	-	0.45
Vol water content [a]	-	0.255
Vol air content [a]	-	0.195
Soil bulk density [b]	g/cm^3	1.7
Total porosity [b]	-	0.41
Vol water content [b]	-	0.194
Vol air content [b]	-	0.216
Vol water content cap fringe[b]	-	0.288
Vol air content cap fringe[b]	-	0.057
Soil bulk density [c]	g/cm^3	1.7
Effective porosity [c]	-	0.41
Fraction of organic carbon [c]	-	0.0004
Hydraulic conductivity (x 10^{-5})	m/s	5
Hydraulic gradient	%	0.37
Dominant wind speed	m/s	0.81

[a] values for SS; [b] values for SP; [a] values for GW

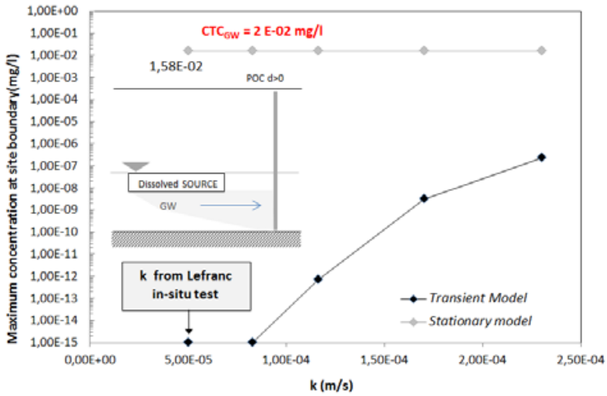


Fig. 4. Site specific parameters and comparison between maximum values of Nickel concentration predicted by the transient model and the results of stationary model at control point, referred to the depicted site configuration

6. Direct measurements and theoretical models of contaminant volatilization

Analytical models typically used to simulate the volatilization of contaminant from soil may significantly overestimate the emissions and thus the exposure of human targets (Bretti and Zanetti, 2014; MATTM, 2014; Verginelli et al., 2014). They consist in a partition of the contaminant in the source zone, a diffusion mechanism based on the Fick's law to reach the ground level and a subsequent box model to simulate the mixing of the vapors with the outdoor air. Verginelli et al. (2017) demonstrated that the assumption of considering a mixing height of 2m in the box model leads (especially in the case of large sources) to an overestimation of the risk of outdoor volatilization and developed a model able to calculate (using an "equivalent height of the mixing zone") the dispersion in the atmosphere as a function of the dimension of the source and of the atmospheric stability class.

As reported in section 5, the possibility to measure the actual vapor emissions is now permitted and standardized by Italian regulatory Agencies (SNPA, 2018). Direct measurements of vapor flux by dynamic open flux chambers allow quantifying the vapor emissions and to compare them to modelling results. Flux values up to 4 orders of magnitude lower than those predicted by volatilization models have been observed (Verginelli et al., 2018).

The assessment of volatile emissions can be carried out by measuring their concentration in the pore air (by Soil Gas Survey, SGS) or their flux from the ground surface (by open dynamic flux chambers). Finally, migration modelling can be entirely avoided by measuring the concentration of volatile compounds in outdoor air. As an alternative, the site-specific measurement of additional parameters that affect volatilization mechanism, such as the soil- water partition coefficient, k_d, can be of help to obtain a more realistic risk estimate.

Figure 5 shows a comparison between the results of direct measurements of mercury emissions and those predicted by different models for volatilization from soil medium, with reference to an actual site in Italy (Di Sante et al., 2016). In particular, Fig. 5 compares the values of mercury concentration at the Point Of Exposure (C_{POE}, i.e., in the air inhaled by the target):

- determined by direct measurement of concentration in outdoor air;
- determined by the direct measurement of flux by open dynamic flux chamber (FC),;
- predicted by the Farmer model and Jury model (APAT, 2008);
- predicted by the Farmer model implemented in the software RISC;

- predicted by the models using the measured k_d value (603 l/kg determined by leaching test) instead of the regulatory default value (52 l/kg - ISS-INAIL (2018), Database).

The value of C_{POE} from direct measurements is significantly lower than that estimated by volatilization models, particularly if the default value of k_d is used. It is important to underline that, based on the acceptable value of HI = 1 (according to the Italian legislation), the corresponding risks would result to be acceptable or unacceptable whether direct measurement or theoretical models are applied.

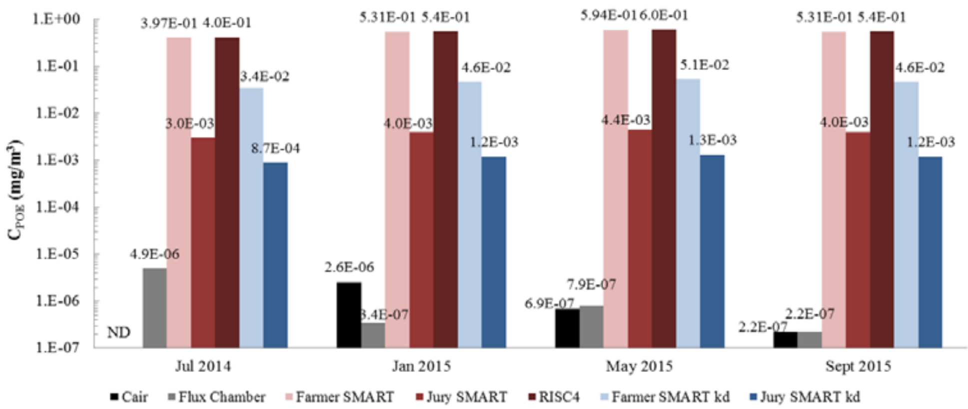


Fig. 5. Concentrations at point of exposure, CPOE

The great differences among the results of the histogram in Fig. 5 can be due to:

- the overly conservative assumptions adopted in the theoretical models
- the properties of the considered contaminant. The most volatile form of mercury is the elemental form (zero-valent). If analytical models of volatilization are applied to the total concentration (i.e. the one determined by standard chemical methods for mercury analysis in soil) all the mercury in the soil is considered to be volatile. Direct measurements of emissions offer the advantage to consider the fraction of mercury that is actually volatile, able to effectively reach the outdoor air and to pose a real hazard to human beings.

6. Concluding remarks

Geotechnical skills are essential for the application of the RA procedure because they are involved in the entire process from site characterization to migration modeling. The modeling of the migration pathways represents the core of the site conceptual modeling thus strongly affecting the RA outcomes. The environmental geotechnics expertise allows a critical view on the contaminant migration phenomena.

Although steady-state migration models are easier to apply, the presented results show that taking into account the time variable offers the advantage to know the time span during which soil remediation works must be concluded or protection measures adopted.

In addition, some of the available models to estimate contaminant volatilization may lead to overestimation of the exposure of targets; in these cases, direct measurements of vapor emissions (today admitted and standardized by the Italian Environmental Protection Agencies) can effectively help obtain more realistic results.

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