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Sanitary and environmental risk assessment for a potentially contaminated aquifer system: a case study

Evaluation de risques pour la santé et pour les environs à cause d'un système aquifère potentiellemet contaminé: Cas étudié

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ABSTRACT: Contaminated sites are a current problem all over the world. In accordance with regulations, risk assessment is required in order to verify that health risks associated with contaminated sites are tolerable and to protect the environment. The paper presents a case study regarding a potentially contaminated site with the presence of chlorinated solvents and Nickel in groundwater. After extensive characterization of the site of concern, the distribution of contaminants in groundwater is identified by means of concentration contour maps, statistic data analysis for the source representative concentration was performed and the site conceptual model for risk assessment was defined. Site specific parameters are derived mainly from geotechnical investigation. Besides health risk assessment, we performed environmental risk assessment considering also groundwater as a target of nickel contamination, as particularly required by Italian regulations. We found tolerable values for health risk. In addition, sensitivity analysis to permeability values of the aquifer was performed.

RÉSUMÉ: Pour ce qui concerne les sites contaminés , il s'agit d'un problème qui intéresse notre époque et le monde entier. Conformément à la loi, l'on requiert une évaluation des risques afin de vérifier que les sites contaminés ne soient pas dangereux pour la santé et afin de protéger aussi les environs. Le document concerne l'étude d'un cas rélatif à un site potentiellement contaminé à cause de solvants chlorés et de nickel dissous dans les eaux souterraines.Après une caractérisation approfondie du site en question, on a identifié par des cartes de courbes de concentration, la distribution des contaminantes dans les eaux souterraines, on a réalisé une analyse des données statistiques pour la concetration représentative de la source et, enfin, on a défini le model conceptuel du site pour l'évaluation des risques. Les paramètres spécifiques concernants le site sont dérivés surtout après des investigations géotechniques. Outre l'évaluation des risques pour la santé, on a effectué aussi une évaluation des risques pour les environs, même en considerant les eaux souterraines comme une cible de la contamination par le nickel, conformément au réglement italien.On a détecté des valeurs tolérables pour la santé. En plus, on a réalisé une anlyse de sensibilité aux valeurs de perméabilité de l'aquifère.

Keywords: Contaminated sites; Risk Assessment; Hazard index; Cancer risk; Groundwater

1 INTRODUCTION

Contaminated sites are a current problem all over the world. In order to quantify, evaluate and manage this problem, risk assessment (RA) for contaminated sites was applied to verify that risks associated with contaminated soils or groundwater of a particular site are tolerable.

In Italy, RA plays a key role in the management of contaminated sites; it is defined as an estimate of the effects of prolonged exposure to the contaminated environmental media (soil, subsoil and groundwater) on human health (Italian Legislative Decree 152/06). Initially developed in the United States; the specific procedure for RA was subsequantly adopted in Europe and in Italy, RA was allowed by regulations on contaminated sites since 1999 (Legislative Decree 471). The first handbook for the implementation was published by UNICHIM association in 2002. The Italian National Environmental Protection Agency (APAT, now named ISPRA) has published the first guidelines for RA for contaminated sites in 2005 and two later revisions in 2006 and 2008 together with some recent accessory recommendations in 2014.

RA calculations starts from the definition of the Conceptual Site Model (CSM) and the description of its three components: source of contamination, migration paths and targets (or receptors) of contamination. Based on the CSM it is possible to calculate the Exposure of targets to contamination (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 is in turn defined as the product of a factor of danger, FD, represented by the toxicity of the contaminant, T, and a factor of contact, FC, represented by the exposure, E, calculated with the CSM, as previously written. The specific expression of risk for contaminated sites therefore can be derived:

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

Risk values are differentiated in risk, R (for carcinogenic effects) or hazard index, HI (for toxic not carcinogenic effects).

To calculate risk using eq.1, taking into account site-specific features, the E must be calculated by means of transport factors of contaminants whose formulations include site-specific parameters. Analytical migration models of contaminants may sometimes provide unrealistic predictions the use of site-specific parameters (i.e. measured on soil samples or derived from in-situ investigations) helps in making model results as close as possible to reality (Di Sante et al., 2013). Backward application of the described procedure allows to calculate Clean Up Levels (CLs) by fixing maximum risk values suggested by regulations (Ialian values: target $R=1\cdot10^{-6}$;target HI=1).

Italian regulations requires also to consider the groundwater as a receptor, calculating the related risk as the ratio of the concentration at the site boundary to the regulatory screening level, tolerable values are lower than 1.

Aim of the present paper is to present: (1) a case study of characterization by geotechnical investigation and RA of a potentially contaminated site and (2) a sensitivity analysis to permeability values of the aquifer.

2 SITE DESCRIPTION

The site of concern is located on an alluvial deposit and the current activity developed on the site is that of an Intermodal Logistics Centre. In the past agricultural activity was developed in the site. During monitoring controls required by legislation to evaluate the environmental impact of new buildings to be constructed within the site boundaries, concentrations exceeding screening levels (SLs) for chlorinated solvents and Nickel were detected in groundwater.

The site has a total area of 54 ha and is currently in use, therefore workers and people living in the neighborhood respresent the human target of contamination. The present paper is related to the worker receptors only.

3 METHODOLOGY

3.1 Site characterization

The property displays a very irregularly shaped area and is located approximately at 35 m m.s.l. on the left bank of a river. In order to investigate the subsoil condition and collect soil samples, 11 boreholes were drilled with a roughly regular grid (200x200m). Eight of the boreholes were converted into groundwater monitoring wells (named with A and B in Figure 1) in addition to the 6 already present in the site (named with PZ in Figure 1) . Also 4 wells located outside the site were used for the phreatic contours determination only.

The subsoil conditions are schematically depicted in Figure 2. The upper layer is made by clayey and sandy silt of 0.7-4.8m thickness. Underneath this layer the alluvial deposit made of gravel in silty matrix is found. The thickness of these layer ranges between 13 and 20 m moving from the north side to the south east side of the site. The permeability of the alluvial layer was studied by means of 3 Lefranc tests. In only one of the three tests permeability measurement was achievable and equal to 1,5 x 10^{-5} m/s. During the other two tests it was not possible to measure hydrulic levels due to the high permeability of the aquifer, therefore this represent one of the major task and decision to be made for the site with the aim to develop proper RA. Underneath the alluvial layers a stratified marly-arenaceous bedrock (Plio-Pleistocenic formation) is found.

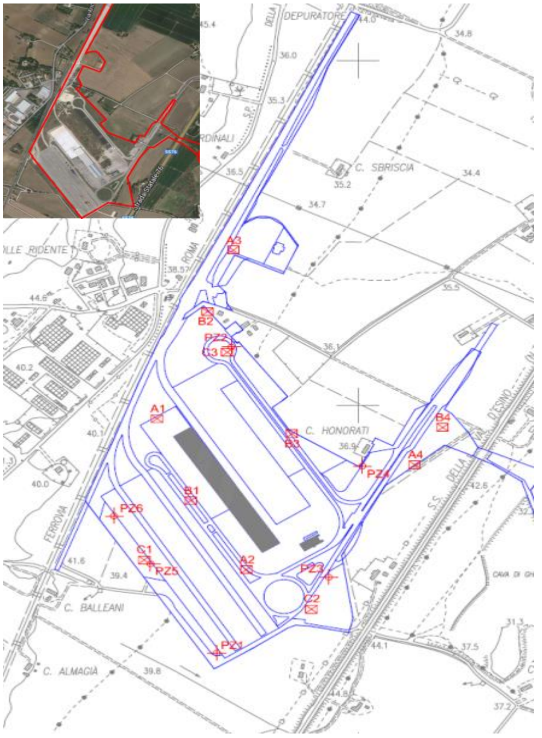

Figure 1. Survey points location map

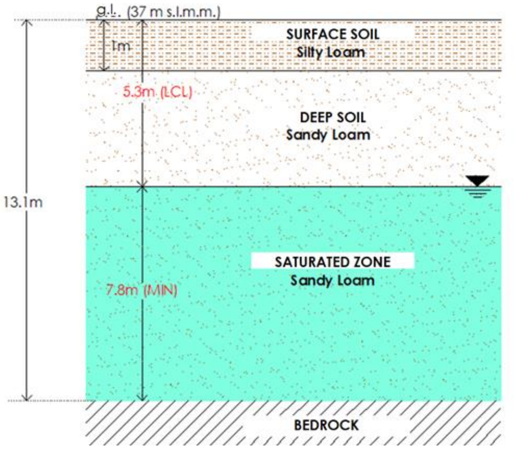

Figure 2. Schematics of the subsoil stratigraphy adopted for the site conceptual model and reference soil textures from grain size analysis.

Water table was located at a depth of 5-7m from ground level, the isophreatic asset highlights the presence of a main drainage axis whose direction is SW-NE. A minimum of three samples from each borehole were analyzed for chemicals (34 samples in total). Three soil samples were tested to determine the fraction of organic carbon and to perform particle size analysis. Fourteen groundwater samples were extracted from monitoring wells and tested. Table 1 summarizes the analytical results for groundwater samples. Trichloromethane, tetrachloroethylene and Nichel were identified as potential contaminants, exceeding CTCs in groundwater. Soil concentrations were found to be lower than the threshold concentrations (CTCs) in all samples. Public available records prove that in 2011-2012 local Environmental Agencies found the same contaminants (trichloromethane, tetrachloroethylene and Nichel) exceeding CTCs in the upstream ground water flow during periodic control surveys. This evidence together with the complete absence of unsaturated soil contamination lead to the hypothesis that the groundwater contamination was incoming in the site from upstream. A particular case is represented by Nickel, whose origin is not identifiable (could be also natural background).

During validation activities, the Environmental Protection agency found in two samples also 1,1-Dichloroethilene exceeding CTC therefore it is required to add the chemical in the RA procedure as a potential contaminant.

Table 1. Analytical results for groundwater samples

Contaminants	MIN µg/l	MAX µg/l	CTC µg/l	C>CTC
Trichloromethane	<0.1	0,30	0,15	6/14
Tetrachloroethylene	2,50	17,00	1,10	11/14
Nickel	2,10	28,00	20,00	1/14

3.2 Site Conceptual Model

The site conceptual model represents the core of the risk assessment procedure. It includes three components: sources, migration pathways and receptors.

In the present case, the sources are polluted plumes located in the saturated zone and they were geometrically defined as the most external CTC contour among all the CTC contours of contaminants of the same family (CTC contours highighted as thick lines in Figure 3). Concentration contours were drawn by means of surface modeling software (Surfer ver.8.0). If the data number was sufficiently high, a statistical procedure (UCL of the data mean) was used to calculate the representative source concentration (RSC) otherwise the maximum concentration was selected as the representative value. ”Non-detect” values were replaced by the detection limits of the analytical method in the RSCs calculations, as recommended by the Italian regulatory agency (APAT, 2008). All the RSCs are summarized in Table 2.

Table 2. Representative Source Concentrations

Contaminants	RSC µg/l	Calculation type
Trichloromethane	1.75E-01	95% Modified-t UCL (Johnson-1978)
Tetrachloroethylene	10.7	95% Student's-t UCL
1,1-Dichloroethilene	3.88E-02	95% Chebyshev(Mean, Sd) UCL
Nickel	2.80	95% Modified-t UCL (Johnson-1978)

The physico-chemical and toxicological properties of contaminants are those suggested by ISS and INAIL (Italian National Institute for Health protection of citizens and workers) (ISS, 2013).

Migration pathways included volatilization from groundwater for the chlorinated solvents (Nichel is not considered as volatile). Migration with groundwater to the downstream site bounday was considered for Nichel only because other chemicals already exceeded the CTCs at the site boundary and their outside origin is already proved (see section 3.1). The extensive data set collected during the characterization allowed us to determine or derive by correlation most of the site-specific parameters necessary for the analytical migration models and they are summarized in Table 3.

The remaining data were collected from various sources. In analogy with the RSCs, site-specific parameters were statistically determined if the data number was sufficiently high (>10), otherwise the most precautionary value was adopted. The on-site receptors considered were adult workers, as foresees by the guidelines for an industrial scenario. In addition, as requested by Italian regulations, also the groundwater at the site boundaries was considered as a receptor but only for Nickel, for the reasons discussed in section 3.1. Exposure parameters (e.g. exposure duration, frequency, age of receptor, body weight, amount inhaled), were assigned to receptors following the RME (Reasonable Maximum Exposure) philosophy according to Italian guidelines (Table 4)

3.3 Risk Evaluation Tool

$RISC_5$ ® (Risk Integrate Software for Clean-ups ver. 5.05) was used to evaluate the potential risks. It gives an estimation of potential adverse impact on human health from different exposure pathways (Spence et al., 2011).

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Table 3. Environmental parameters

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

[a] values for surface soil; [b] values for deep soil; [a] values for saturated zone (see Figure 2)

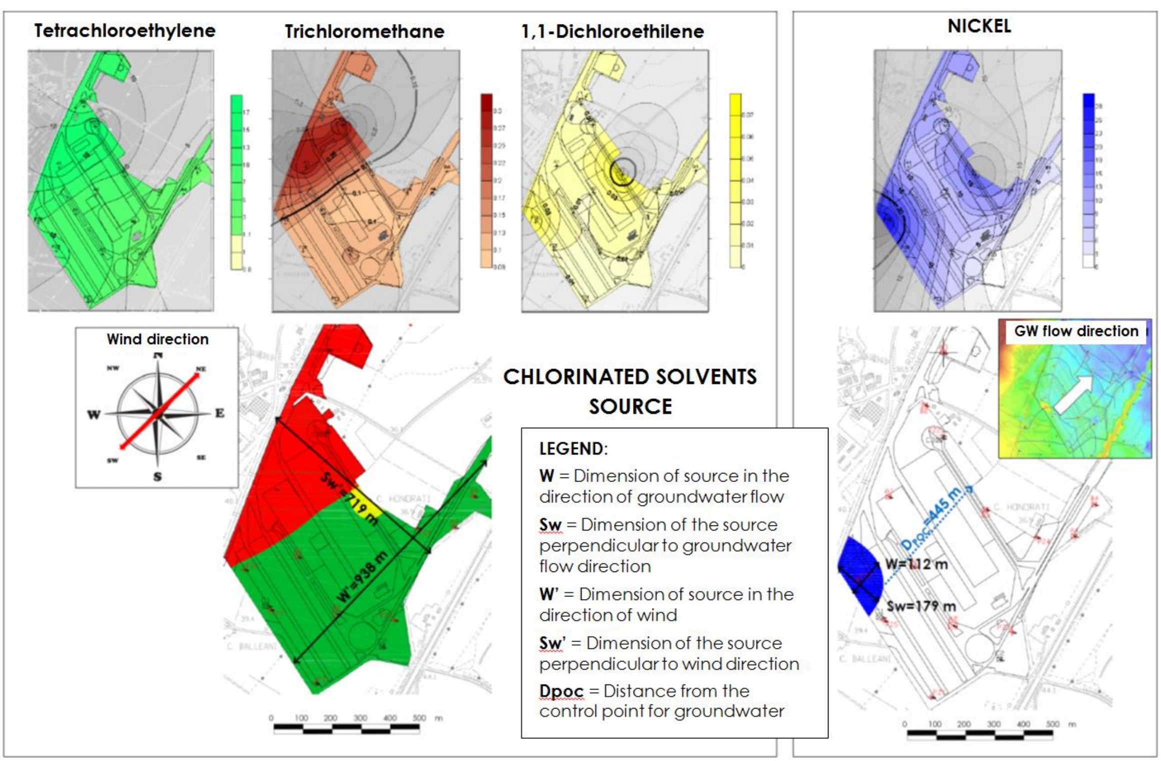

Figure 3 Concentration contours and source geometry definition and parameters

Table 4. Exposure parameters (APAT, 2008)

EXPOSURE FACTORS	Units	Worker Hard work	Worker Sedentary
Body weight	kg	70	70
Carcinogen average time	years	70	70
Non-carcinogen average time	years	ED	ED
Inhalation of air			
Exposure duration	years	25	25
Exposure frequency	days/year	250	250
Daily exposure frequency	h/day	8	8
Inhalation	m^3/h	2.5	0.9

The options of the software include the poosibilty of modifying contaminant properties in order to use Italian ISS-INAIL data-base values as well as the possibility to take into account the presence of soil lenses in the volatilization pathway. This last possibility is extremely useful for the site of concern given the presence of a surface layer of soil with grain size characteristics different from the remaining unsaturated soil portion (see Figure 2). In the event of contaminants with carcinogenic effects, an added receptor is considered with exposure parameters averaged between childhood and adulthood following also the suggestions of the Italian Regulatory Agency. Moreover, the software offers the possibility to insert a maximum allowed concentration value in groundwater (MCL, set as Italian CTC in groundwater) and uses it as a target value to comply with, in calculations of CLs. Migration in groundwater is modeled by the software with a transient analytical model with a mass loading rate from the source zone calculated as a function of hydraulic conductivity (Yeh, 1981). It is important to uderline that Italian guidelines contain a stationary analytical model for the dispersion in groundwater (Domenico e Schwartz, 1998).

4 RESULTS

Results of RA include risk values and hazard indexes for forward applications and clean up levels for backward application.

4.1 Forward assessment

Both human and environmental receptors are taken into account and separate risk values obtained.

4.1.1 Health risk results

Forward health risk assessment was performed for both carcinogenic and non-carcinogenic effects. In particular, two worker types are considered as receptors: one performing hard working activity (loading and unloading) and one doing a sedentary job because both service areas and offices are present in the site of concern. In Figure 4 risk results are summarized. The hazard index and the risk values show that hard workers are, obviously, the most sensitive receptors.

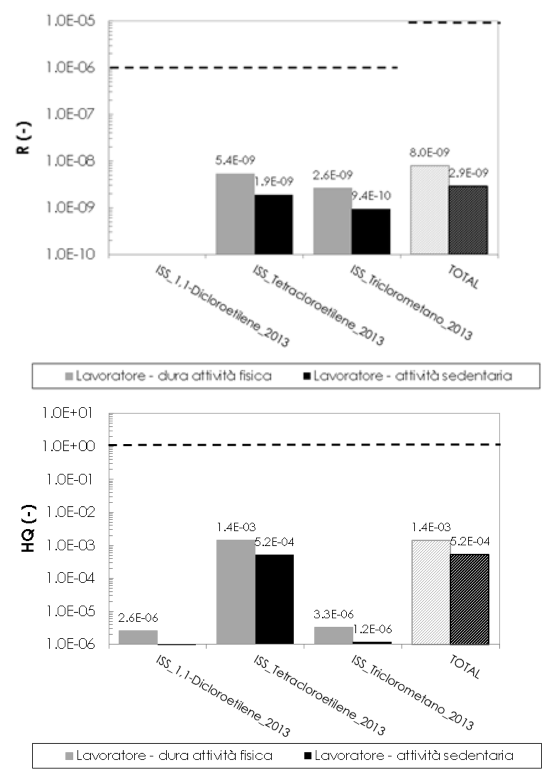

Figure 4. Cancer risk and non-cancer hazard results for human receptors (workers)

Cumulative and individual hazard indexes and cancer risks does not exceed regulatory acceptable levels. For both effects, Tetrachloroethylene is the chemical causing highest risk values.

The effect of the different characteristics of the surface soil from the remaining unsaturated zone is taken into account. This was possible thanks to the calculation of an overall effective diffusion coefficient as a depth-weighted avarage of the effective diffusion coefficients (Millington and Quirk, 1961) in the capillary fringe, the vadose zone above the capillary fringe and the surface silty loam (Figure 2) that, in the case of concern, was simulated as the lens. It is important to point out that smaller air-filled porosity will reduce the overall diffusion coefficient significantly.

4.1.2 Environmental risk results

In taking into account the environmental receptor (i.e. groundwater at the downgradient site bounday), results of both the transient model of migration of Nickel implemented by $RISC_5$ and of the stationary model suggested by Italian guidelines were studied varying the value of saturated hydraulic conductivity, k.

In particular, given the difficulties caused by the high hydraulic conductivity during Lefranc tests (section 3.1), different values of k were used in the simulation starting from the measured value to higher ones (maximum k value= 2.3E-04 m/s, suggested by $RISC_5$ manual for Gravel deposits).

The distance of source to the control point is depicted in Figure 3 (Dpoc), and concentration trends with time are observable in Figure 5.

At this point, according to $RISC_5$ results, no Nickel will be detected in 10000 years using k value from Lefranc test. With reference to a $k=1.16\times10^{-4}$ m/s, the contaminant is supposed to arrive after more than 7500 years and to reach the insignificat low value of 6×10^{-13} mg/l at 10000 years. Also considering the maximum value of k, the contaminant will not be detected at the control point before 3700 years and is supposed to reach the concentration of 2.4×10^{-7} mg/l at the end of the simulation; this maximum value is far lower than the treshold limit for groundwater for Nichel, highlighting a tolerable value of risk.

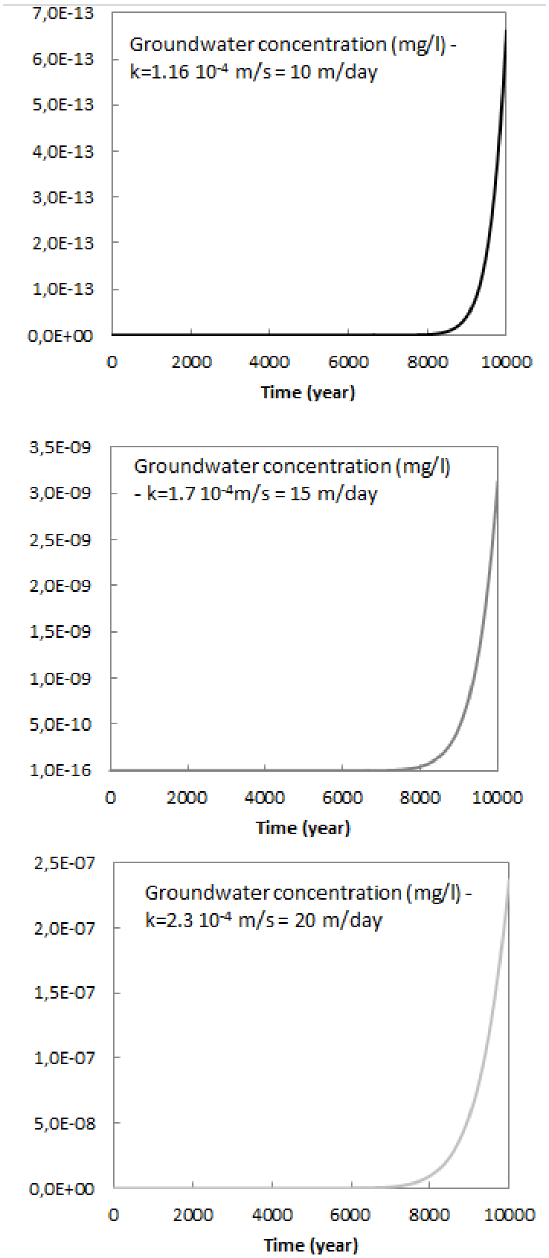

Figure 5. Trends of concentration of Nickel in groundwater at control point with time, varying k.

This extremely slow migration is due to the high value of soil-water partition coefficient, k_d, of Nickel that varies as a function of pH and, for the pH measured during characterization (pH=7.9- LCL 95% of the Mean), k_d is equal to 1400 ml/g, causing great adsorption on soil particles along the migration pathway.

Comparing results from the two types of model (Figure 6), the stationary model gives an output concentration at the control point of 1.58×10^{-2} mg/l, 5 orders of magnitude higher than maximum results of concentration given by $RISC_5$ but lower than the CTC for groundwater too.

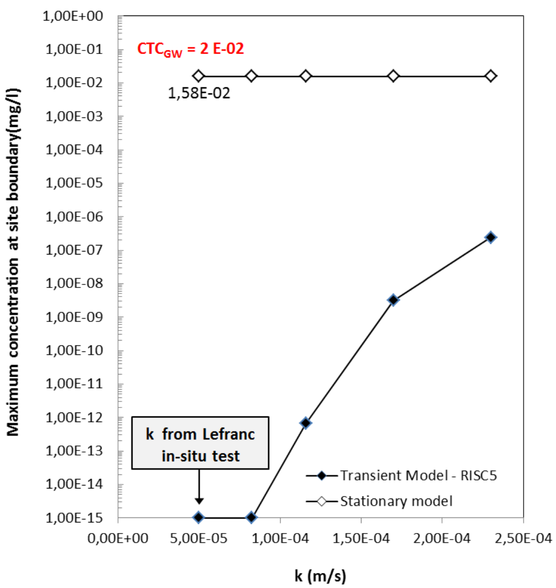

Figure 6. Comparison between maximum values of Nickel concentration of transient model and results of stationary model at control point

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

4.2 Backward assessment

The clean up levels derived are lower than the measured concentrations, as obviously expected, given the tolerable values of the calculated risks and hazard indexes.

5 CONCLUSIONS

The present paper presents a case study of characterization by geotechnical investigation and risk assessment of a potentially contaminated site. The careful characterization activities allow to demonstrate that the groundwater contamination by chlorinated solvents is incoming in the site from upstream. This hypothesis is an important key to properly set the site conceptual model and to include/exclude receptors in the assessment. Given the difficulties in measuring permeability values with in-situ Lefranc test, also a sensitivity analysis of migration in groundwater of Nickel to permeability values of the aquifer is performed to verify the absence of untolerable risk also for high values of hydraulic conductivity.

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