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Vapour intrusion from the vadose zone—seven algorithms compared

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Abstract
Background, aim and scope Vapours of volatile organic compounds (VOCs) emanating from contaminated soils may move through the unsaturated zone to the subsurface. VOC in the subsurface can be transported to the indoor air by convective air movement through openings in the foundation and basement. Once they have entered the building, they may cause adverse human health effects. Screening-level algorithms have been developed, which predict indoor air concentrations as a result of soil (vadose zone) contamination. The present study evaluates seven currently used screening-level algorithms, predicting vapour intrusion into buildings as a result of vadose zone contamination, regarding the accuracy of their predictions and their usefulness for screening purpose. Screening aims at identifying contaminated soils that should be further investigated as to the need of remediation and/or the presence of an intolerable human health risk. To be useful in this respect, screening-level algorithms should be sufficiently conservative so that they produce very few false-negative predictions but they should not be overly conservative because they might have insufficient discriminatory power.
Materials and methods For this purpose, a comparison is made between observed and predicted soil air and indoor air concentrations from seven reasonably well-documented sites, where the vadose zone was contaminated with aromatic or chlorinated VOCs. The seven screening-level algorithms considered were: Vlier–Humaan (Be), Johnson and Ettinger model (USA), VolaSoil (NL), CSoil (NL), Risc (UK) and the dilution factor models from Norway and Sweden. Calculations are presented in two scatter plots (soil air and indoor air), each containing the predictions versus the observations. Differences between predicted and observed VOCs concentrations were evaluated on the basis of three statistical criteria to establish their accurateness and the usefulness for screening purposes. Results from the applied criteria are presented in a table and figures.
Results It was found that the screening-level algorithms investigated tended to overestimate soil air concentrations more than indoor air concentrations. Differences between predictions and observations were up to three orders of magnitude. The algorithms with the highest accuracy for predicting the soil air concentration are in ascending order the Johnson and Ettinger model (JEM), Vlier–Humaan and VolaSoil algorithms. For the indoor air, it is concluded that all algorithms have a tendency to overestimate the predicted indoor air concentrations, except for the JEM and Vlier–Humaan algorithms, which produced frequent underestimations.
Discussion Several earlier studies have investigated the accuracy of some of the screening-level algorithms for

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vapour intrusion and the results presented in the present study agree with the findings. However, the present study presents the accuracy of vapour intrusion algorithms via three statistical criteria that allow their ranking. The present study also determines the suitability of screening-level algorithms as screening tool. It is found that algorithms may rank differently as to accuracy and suitability as a screening tool.

Conclusions The algorithms with the highest accuracy for predicting the indoor air concentration are the JEM and Vlier–Humaan algorithms. The most suitable algorithms to serve for screening purposes are CSoil, VolaSoil and Risc, since they are sufficiently conservative, have fewer false-negative predictions and still have sufficient discriminatory power.

Recommendations and perspectives Given the over-predictions and under-predictions of the algorithms considered, a combination of modelling and measurements will often be required to produce multiple lines of evidence for the presence of an intolerable human health risk or the need for remedial actions at a site. Integrated programmes of modelling and field observations can reduce the uncertainty of predicted soil air and indoor air concentrations, and a tiered approach is presented in this study.

Keywords Accuracy · Algorithm · CSoil · DF Norway · DF Sweden · Indoor air · Intrusion · JEM · Model · RISC · Soil air · Soil contamination · Vadose zone · Vapour · Vlier–Humaan · VOC · VolaSoil

1 Background, aim and scope

Contaminants of concern in vapour transport in the unsaturated zone are typically volatile organic compounds (VOCs). Examples of VOCs include chlorinated solvents such as tetrachloroethylene (PCE), trichloroethylene (TCE) and 12-dichloroethene (DCE) and fuel hydrocarbons such as benzene, toluene, ethylbenzene and xylenes. These VOCs can be released into the subsurface environment from improper disposal, accidental spillage or leaking storage tanks. Once in the subsurface, these compounds will be distributed between soil gas, pore water, soil and pure contaminant phases. Organic vapours emanating from contaminated soil or groundwater may move through the unsaturated zone by diffusion or convection due to pressure or density gradients or a combination of these processes (Tillman and Weaver 2006). Vapours in the subsurface can be transported to the indoor air by convective air movement through openings in the foundation and basement. Once VOCs have entered the building, they can cause adverse human health effects. Humans spend 64% to 94% of their time indoors and therefore the indoor air quality is of primary importance for exposure to VOCs (Kaplan et al. 1993; Fugler and Adomait 1997).

Researchers have suggested to use screening-level algorithms with site-specific data in order to evaluate them for the vapour intrusion pathway (Fitzpatrick and Fitzgerald 2002; Johnson et al. 2002; Hers et al. 2003; Huijsmans and Wezenbeek 1995; Evans et al. 2002; Tillman and Weaver 2006; van Wijnen and Lijzen 2006; Provoost et al. 2009). Screening-level algorithms typically performed phase partitioning calculations to estimate the concentration of a particular contaminant in soil gas from its concentration in another phase (i.e. bulk soil) followed by diffusive and/or convective transport to the zone of influence from the building. The mathematical formulation of each of these components is described below. The concentration of a contaminant in soil gas in contact with contaminated soil (i.e. a bulk soil sample) is by most screening-level algorithms calculated as followed:

$$C_{sg} = \frac{C_{bs}}{\frac{1}{H} \times \left[f_{oc} \times K_{oc} + \frac{1}{\rho_b} \times \left(n_w + n_g \times H \right) \right]} \quad (1)$$

Where C_{sg} is the concentration in the soil gas (μg/L), C_{bs} the concentration in the bulk soil (μg/kg), H the contaminant-specific Henry's Law constant [(μg/L vapour)/(μg/L H_2O)], f_{oc} the fraction of organic carbon (–), K_{oc} the organic carbon partition coefficient (mL/g), ρ_b the soil bulk density (g/mL), n_w the volumetric moisture content [L H_2O/L soil] and n_g the volumetric gas content ($=n_T - n_w$) [L vapour/L soil].

Organic vapours emanating from contaminated soil may move through soil gas in the unsaturated zone by diffusion or soil gas convection due to pressure or density gradients or a combination of these processes. In a typical scenario, organic vapours above a contaminated source in the soil (high concentration) diffuse towards the subsurface (lower concentration). The well-known relation describing the diffusion of a compound across a unit of cross-sectional area is Fick's First Law (Little et al. 1992; Nazaroff 1992; Jury et al. 1983).

$$J_g = -D_{eff} \times \frac{\partial C_{sa}}{\partial z} \quad (2)$$

Where J_g is the mass flux [g/m^2 s], D_{eff} is the effective diffusion coefficient of the compound in the gas phase [m^2/s], ∂C_{sa} is the concentration of the compound in the soil gas [g/m^3] and ∂z is the distance over which diffusion occurs. In porous media, the D_{eff} depends on the porosity and water-filled porosity of the medium, as formulated by Millington and Quirk (1961).

$$D_{eff} = D_a \frac{\theta_a^{10/3}}{\theta_T^2} + \frac{D_w}{H} \frac{\theta_w^{10/3}}{\theta_T^2} \quad (3)$$

where D_a is the free-air diffusion coefficient [L^2/T], D_w the aqueous diffusion coefficient [L^2/T], θ_a the soil air-filled porosity [volume vapour/total volume], θ_T the soil total porosity [volume pores/total volume], θ_w the soil water-filled porosity [volume water/total volume] and H the dimensionless Henry's Law Constant [molar concentration in gas/molar concentration in water].

From these two equations, it is apparent that the rate of molecular diffusion in the gas phase depends upon the concentration gradient and the effective diffusion coefficient of the compound of interest. The flow of soil gas in the subsurface may be caused by gas-pressure gradients and can be described via Darcy's law:

$$u_v = -\frac{k_v}{\mu} \cdot \nabla P \tag{4}$$

where u_v is the average vapour phase velocity (cm/s), k_v the soil air permeability (cm^2), μ the air viscosity (g/cm s), P the vapour pressure (g/cm s^2), with the ∇P as vapour pressure gradient (Johnson and Ettinger 1991; Nazaroff 1992; Loureiro and Abriola 1990).

Pressure-driven convection is produced when differences in soil gas pressure form, causing soil gas to flow and carry any vapours present with it. The effects of overlying buildings play a very important role in the subsurface-to-indoor-air pathway. Different building construction techniques may have different impacts on the ability of vapours to enter indoor air space. Buildings with basements may have more surface area through which vapours can move inside, as well as be closer to subsurface sources than slab-on-grade buildings. A single-pour cement foundation may not have the "perimeter crack" often associated with foundations whose footers and floor are poured separately, but may still become cracked along stress lines. Building under-pressurisation relative to soil gas pressure can be caused by temperature differences between indoor and outdoor air (i.e. stack effects), wind or barometric pressure cycles. The under-pressurisation of buildings relative to subsurface pressure may cause contaminated soil gas to flow into indoor air spaces, increasing exposure and potential human health effects. Figure 1 shows a simplified example of a contaminant release in the subsurface with the contaminant being present in the solid phase with contaminated soil gas that move through the vadose zone to the subsurface and the close proximity of the foundation and basement floor. Vapours in the subsurface can be transported to the indoor air by convective air movement through openings in the foundation and basement where they can cause adverse human health effects.

A common use of vapour intrusion algorithms is to screen out sites, or individual buildings at sites, that are deemed to require further investigation. Screening-level algorithms should be sufficiently conservative so they produce a minimum of false-negative predictions (prediction is lower than observation). Screening-level algorithms should also not be overly conservative because they might have insufficient discriminative power. There has been little evaluation of the false-negative (or type II) error produced by the algorithms at field sites, with the possible exception of the widely studied Johnson and Ettinger model (JEM) (Fitzpatrick and Fitzgerald 2002; Johnson et al. 2002;Hers et al. 2003; Abreu and Johnson 2005).

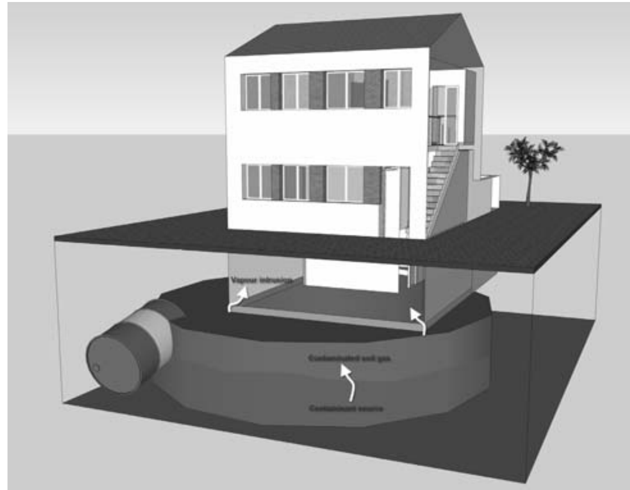


Fig. 1 Overview vapour intrusion from the contaminated source

This paper investigates the accuracy of seven vapour intrusion screening-level algorithms as a result of contamination in the vadose zone. Accuracy of screening-level algorithms is, in this context, related to the difference between predicted versus observed air concentrations. For this purpose, a comparison is made between observed and predicted soil air and indoor air concentrations from seven reasonably well-documented sites, where the vadose zone was contaminated with aromatic or chlorinated VOCs. The results presented in this paper can contribute to understanding of the suitability of screening-level algorithms for regulatory purposes and the possible occurrence of false-negative errors.

2 Materials and methods

2.1 Algorithms

The screening-level algorithms that are selected for this comparison are the newest version of the algorithms CSoil (2008) and VolaSoil (1.9) from the Netherlands, Vlier–Humaan (2.1) from the Flemish region (Belgium), the Johnson and Ettinger model (JEM) (3.1) from the USA, Risc (4.03) from the UK and the dilution factor algorithm from respectively Sweden (1996) and Norway (1999).

These algorithms are frequently used by various countries within Europe for site-specific health risk assessment and/or the derivation of soil screening levels. A short description of these algorithms and further references are given in Provoost et al. (2009) and below an overview table with key references is presented (Table 1).

2.2 Field observations and sites

Spatial and temporal field observations from seven fairly well-documented sites, which are contaminated with chlorinated or aromatic VOCs, are collected to investigate the accuracy of the selected screening-level model algorithms. Contaminants of concern are benzene, toluene, ethylbenzene and xylenes (BTEX), tetrachloroethylene (PCE), trichloroethylene (TCE) and 12-dichloroethene (DCE). Table 2 provides a high-level summary table of the site data and for further details the reader is referred to the references.

Each observed vadose zone concentrations of the contaminants of concern under or near the building serves as an input for the algorithms to predict the soil air and indoor air concentrations. For each site, soil- and building-related properties plus (synoptically) measured soil, soil air and indoor air concentrations are recorded in a data matrix. The site characterisation includes spatial and temporal data and data obtained from this effort are believed to provide useful order-of-magnitude findings and context (Fitzpatrick and Fitzgerald 2002). The soil properties that are collected per site are the soil type, bulk density, total porosity, pore water and air-filled porosity, soil air permeability, soil temperature, organic matter content and organic carbon content. The collected building characteristics are thickness of the floor, length, width and height of the building, volume of the building and compartments (basement and indoor space), indoor air exchange rate and pressure differences. Other factors such as the building type and its quality, proximity of the sampling point to the building and the type of surface pavement are taken into consideration to arrive at the predicted soil and indoor air concentration. Observed indoor air concentrations are included in the data matrix only if elevated above the observed ambient air concentration.

Each algorithm includes a standard set of physical–chemical parameters for contaminants that frequently occur in soils. An analysis of the physical–chemical

Table 1 Overview of mathematical models

Descriptor per model	CSoil	Vlier–Humaan	JEM	VolaSoil	Risc	DF Sweden	DF Norway
Compartment/floor							
Slab-on-grade		●	●	●	●		
Concrete basement		●	●	●	●		
Crawl space	●	●		●		●	●
Transport							
Diffusive	●	●	●	●	●		
Diffusive plus convective			●	●	●		
Attenuation factor/empirical						●	●
Source							
Groundwater		●	●	●	●	●	●
Vadose zone	●	●	●	●	●	●	●
Soil gas				●			
Application							
Site-specific assessments		●	●	●	●		
Derivation of screening levels	●	●	●			●	●
Main (original) reference(s)	a	b	c	d	e	f	g

JEM Johnson and Ettinger model, *DF* dilution factor, ● applies for model

[a] Rikken et al. 2001; Brand et al. 2007

[b] Jury et al. 1983, 1990

[c] Johnson and Ettinger 1991, 1997; Johnson et al. 1998

[d] Waitz et al. 1996; van Wijnen and Lijzen 2006; Bakker et al. 2008

[e] BP 2001

[f] Naturvårdsverket 1996

[g] SFT 1995, 1999

Table 2 Overview of sites

Descriptor per site	Site 1	Site 2	Site 3	Site 4	Site 5	Site 6	Site 7
Geographical							
City	Ukkel	Mortsel	Hemiksem	Catterton	Alameda Naval	Vilvoorde	Borstbeek
Country	Belgium	Belgium	Belgium	UK	USA	Belgium	Belgium
Soil type							
Sandy loam	•						
Loamy sand		•					
Sand			•	•	•		•
Loam						•	
COC							
BTEX	•		•	•	•	•	•
PCE, TCE, DCE		•				•	
Observed conc. range (mg/kg dm)	0.05–65	0.5–1,600	0.05–1.1	450–5,500	0.54–1.4	0.05–470	2.1–390
Depth COC (m gl)	0.8–1.7	0.7–2.3	0.6–0.9	0.2–1.4	0.2–0.7	1.2–3.0	1.8
Building							
Building type	sog	sog	sog	sog	sog	Basement	sog
Volume (m³)	83	6,000	85	124	136	143	488
Air exchange rate (L/h)	0.3	2	0.1	0.4, 4.8, 14	2.1	0.05	0.05
Main reference(s)	[a]	[a]	[a]	[b]	[c]	[d]	[e]

Observed conc. range=indicative range between minimum and maximum soil concentrations for all COC. Depth COC=subsurface zone where contaminant was detected in the solid phase

COC contaminant of concern; *gl* ground level; *sog* slab-on-grade; *BTEX* benzene, toluene, ethylbenzene and xylenes; *PCE* tetrachloroethylene; *TCE* trichloroethylene; *DCE* 12-dichloroethene

[a] Van Geert et al. 2004

[b] Hers et al. 2002

[c] Fischer et al. 1996

[d] Mava 2002

[e] Soresma 2004

parameter values reveals that the Henry constant, vapour pressure, solubility and diffusion coefficient in water and in air differed (considerably) between the various algorithms. To exclude variation in soil air or indoor air as a result of this variation, a default physical–chemical parameter set was selected for each contaminants of concern from Provoost et al. (2004) and used in each algorithm prediction.

Ventilation rates are not measured and therefore estimated in view of the quality of the building and the observed frequency that doors are opened and closed. Likewise, the indoor air exchange rates and pressure differences between the soil and the building were harmonised between algorithms to decrease the effect of different default values between the screening-level algorithms.

The data matrix, for the seven sites, results in around a hundred observed soil and indoor air concentrations. Each of the observed air concentrations was predicted by using seven different algorithms. Results are presented in two figures that compares all observed versus predicted soil air or indoor air concentrations.

2.3 Accuracy and suitability

2.3.1 Accuracy

Accuracy is defined as the algorithms' ability to predict air concentrations that are in agreement with the observed air concentrations. Thus, the closer the predicted concentration is associated with the observed concentration, the lower the values for the three criteria are, and therefore the higher the algorithms' accuracy. The accuracy for screening-level algorithms is objectified by using three general accepted statistical criteria as described by Loague and Green (1991). These criteria can be used for inter-algorithm comparison and provides a ranking of the seven algorithms towards their accuracy. Accuracy as such does not necessarily express a ranking of suitability for regulatory purposes.

The first criterion is built on the maximum relative error (ME).

$$\mathrm{ME} = \frac{\max_{i=1}^{n}[abs(O_i - P_i)]}{O} \quad (5)$$

where O is the observed concentration and P is the predicted concentration. ME represents the maximum difference that is recorded for all pairs of observed and predicted concentrations. The lower the ME value, the smaller the maximum difference between O and P, and the better the accuracy of the algorithm.

The second criterion is related to the root mean squared error (RMSE):

$$\mathrm{RMSE} = 100\frac{\sqrt{\frac{\sum_{i=1}^{n}(O_i - P_i)^2}{n}}}{\bar{O}} \tag{6}$$

where O is the observed concentration, P the predicted concentration and n the number of cases. The RMSE indicates the average deviation (difference) from all pairs of O and P. The lower the RMSE is, the smaller the difference between O and P, hence the better the accuracy of the particular algorithm.

The third criterion can be described by the coefficient of residual mass (CRM).

$$\mathrm{CRM} = -\frac{\left[\sum_{i=1}^{n} O_i - \sum_{i=1}^{n} P_i\right]}{\sum_{i=1}^{n} O_i} \tag{7}$$

where O is the observed concentration and P is the predicted concentration. The CRM indicates whether algorithm predictions have an overall tendency to over- or underestimate observations. If the CRM value is negative (−), the predicted concentration overestimates the observed concentration which is an indication of the conservatism of the algorithm, and vice versa for a positive (+) value. The closer the CRM is to zero, the smaller the overprediction or underprediction and thus the better the accuracy of the particular algorithm to predict a concentration that is close to the observed concentration.

Values from different algorithms have a range over several orders of magnitude so a logarithmic scale is applied, which requires positive values. Therefore, CRM results in the chart will be presented as absolute positive values; however, the table will provide for each algorithm the actual values for the CRM.

2.3.2 Suitability

Screening-level algorithms should have a certain degree of conservatism so they produce a minimum of false-negative predictions (prediction is lower than observation), but still have sufficient discriminative power. The conservatism should compensate for the uncertainties related to the simplification of the vapour intrusion process in an algorithm. Therefore, screening-level algorithms should neither be overly conservative because they might have insufficient discriminative power nor have any conservatism at all, since they might underestimate the risk. Thus, it is reasonably expected for a screening-level algorithm that the difference between all observed and predicted concentrations should be within one order of magnitude (Johnson et al. 2002) to be considered as sufficiently suitable for regulatory purposes (Evans et al. 2002).

3 Results

3.1 Soil air

Figure 2 plots all observed versus predicted soil air concentrations, and reveals that all seven algorithms have a tendency to overestimate the soil air concentrations. The dashed line represents the points for which the observed concentration equals the predicted concentration. The difference between observed and predicted soil air concentrations is frequently less than one order of magnitude, but may be up to four orders of magnitude. The scattergram does not reveal which algorithm is most accurate in predicting the soil air concentration. Therefore, the three criteria (ME, RMSE and CRM) were applied to all observed and predicted soil air concentrations in the data matrix and result in Table 3. The soil air criteria values from Table 3 are presented in ascending order in Fig. 3. Table 3 reveals that the algorithms that have a high accuracy in predicting soil air concentrations are the JEM and Vlier–Humaan algorithms. VolaSoil, CSoil and Risc are somewhat less accurate and more conservative, while the DF algorithms from Sweden and Norway very frequently overpredict the soil air, thus are not accurate and include a high degree of conservatism.

Table 3 shows a negative CRM for all algorithms, meaning that the predicted concentrations are in general higher than the observed soil air concentrations. Vlier–Humaan has a higher ME and RMSE value than the JEM algorithm, which indicates that the maximum and average difference between observations and predictions is higher for Vlier–Humaan than for the JEM algorithm.

3.2 Indoor air

Figure 4 plots all observed versus predicted indoor air concentrations, and shows that all screening-level algorithms frequently underpredict and overpredict the indoor air concentration. Predictions for low indoor air concentrations (<1 μg/m^3) are included since several countries issue tolerable concentrations in air that are just above this range, like for example the 1.3 μg/m^3 for benzene for Sweden and Norway. Furthermore, indoor air concentra-

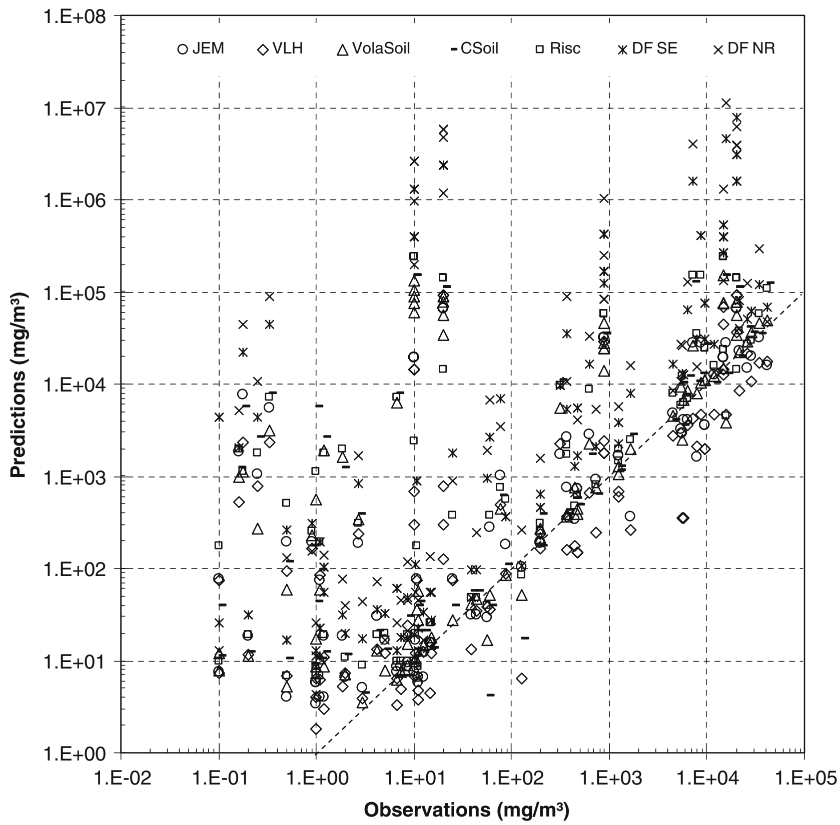


Fig. 2 Scattergram for observed versus predicted soil air concentrations (mg/m³). *VLH* Vlier–Humaan, *JEM* Johnson and Ettinger model, *DF* dilution factor, *SE* Sweden, *NR* Norway. *Striated line*: prediction equals the observation

tions were included in the data matrix if they were elevated above the ambient background concentration or detection limit. All algorithms overpredict the indoor air concentration for the lower concentration range (0.01–1 μg/m^3), thus the algorithms have sufficient conservatism to prevent false-negative predictions for this low concentration range. More variation occurs—ranging from 1.5 underprediction up to four orders-of-magnitude overprediction—for the medium to high range concentrations (>1–1,000 μg/m^3). The tolerable concentrations in air for toluene from different EU countries range from 40 to 7,500 μg/m^3 and for PCE from 680 to 5,000 μg/m^3 (Provoost et al. 2008). This requires a high accuracy over an almost two orders-of-magnitude concentration range. Figure 5, in which the calculated values for the three criteria (ME, RMSE and CRM) are presented in ascending order, shows that the JEM and Vlier–Humaan algorithms have the highest accuracy for predicting the indoor air concentration. Besides, it shows that in ascending order CSoil, VolaSoil, Risc, DF Sweden and DF Norway are less accurate, hence more conservative in their predictions.

Table 3 provides the three criteria values per algorithm for the indoor air and are visualised in Fig. 5. Table 3 shows a negative CRM value for most of the algorithms, meaning that the predicted indoor air concentrations are in general higher than the observed concentrations, except for JEM and Vlier–Humaan that have a positive CRM value. It could therefore be argued that these models are not always sufficiently conservative to prevent false-negative predictions. In other words, the algorithms could

Table 3 Criteria values for soil air and indoor air

Criteria	VLH	JEM	VolaSoil	CSoil	Risc	DF SE	DF NR
Soil air							
CRM	−1.2	−1.9	−3.5	−6.7	−8.0	−98	−166
ME	21	15	30	34	54	1762	2549
RMSE	481	466	780	1,314	1,729	26,903	44,393
Indoor air							
CRM	0.2	0.6	−2.6	−8.4	−9.6	−162	−345
ME	19	17	97	119	144	3,141	8,819
RMSE	233	306	1,508	2,506	2,544	46,192	134,071

VLH Vlier–Humaan, *JEM* Johnson and Ettinger model, *DF* dilution factor, *SE* Sweden, *NR* Norway, *ME* maximum relative error (ME), *RMSE* root mean squared error, *CRM* coefficient of residual mass

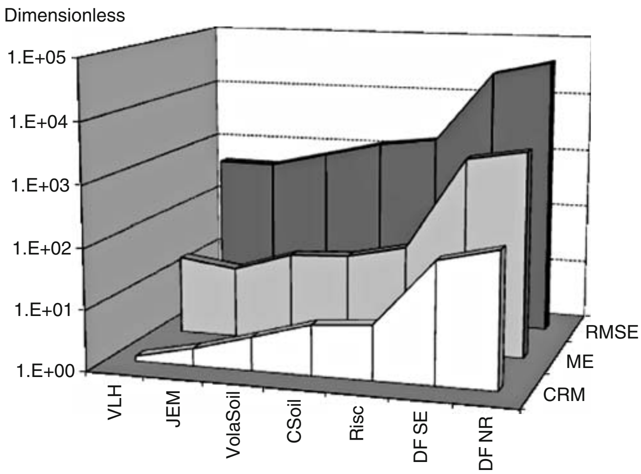


Fig. 3 Criteria values for soil air. *VLH* Vlier–Humaan, *JEM* Johnson and Ettinger model, *DF* dilution factor, *SE* Sweden, *NR* Norway, *ME* maximum relative error (ME), *RMSE* root mean squared error, *CRM* coefficient of residual mass

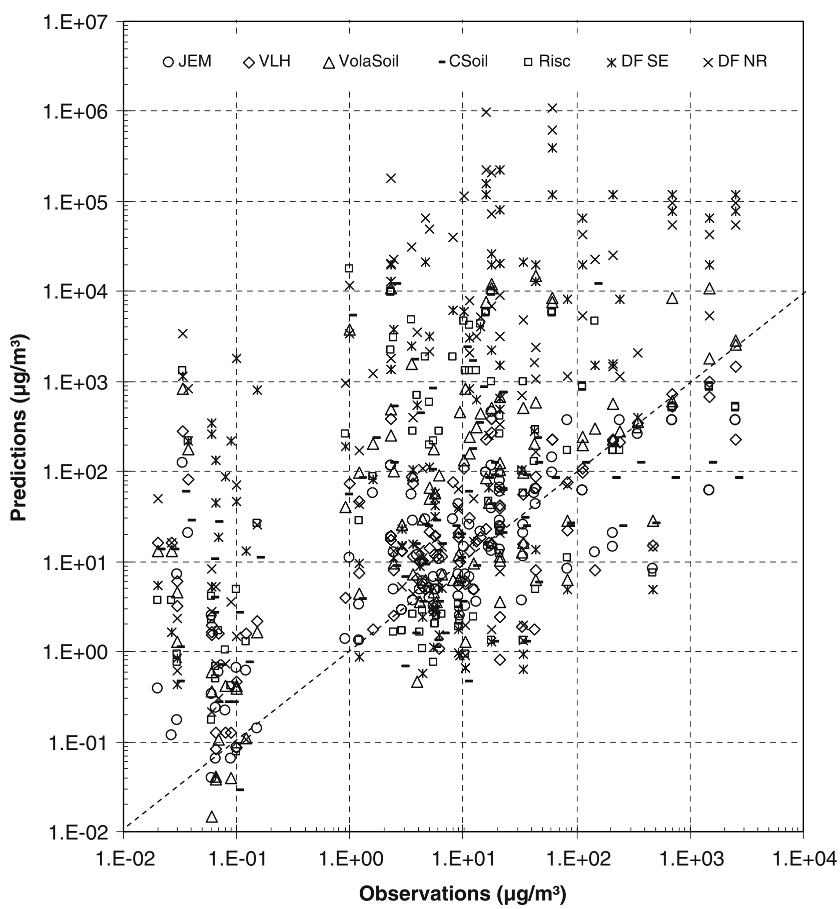
predict an indoor air concentration that is lower than what would be observed via indoor air measurements. However, the RMSE and ME values suggest that the JEM and Vlier–Humaan algorithms should not produce a lot of false-negative errors for indoor air concentrations. The algorithms CSoil, VolaSoil and Risc produce less false-negative predictions than Vlier–Humaan and JEM, but still have sufficient discriminatory power. The dilution factor algorithms from Sweden and Norway are not so accurate in their indoor air predictions and considered least suitable for regulatory purposes because of their overconservatism.

4 Conclusions

Comparison of predicted and observed soil air concentration from seven sites showed that all screening-level algorithms have a tendency to overestimate the soil air concentration. The algorithms with the highest accuracy for predicting the soil air concentration are the JEM, Vlier–Humaan and VolaSoil algorithms.

From a comparison with observed indoor air concentrations, it is concluded that all algorithms have a tendency to overestimate the predicted indoor air concentrations in relation to observations, except for JEM and Vlier–Humaan. These algorithms show a positive CRM, indicating an overall tendency to slightly underestimate observations. Based on the three criteria (ME, RMSE and CRM), the

Fig. 4 Scattergram for observed versus predicted indoor air concentrations (μg/m^3). *VLH* Vlier–Humaan, *JEM* Johnson and Ettinger model, *DF* dilution factor, *SE* Sweden, *NR* Norway. *Striated line*: prediction equals the observation



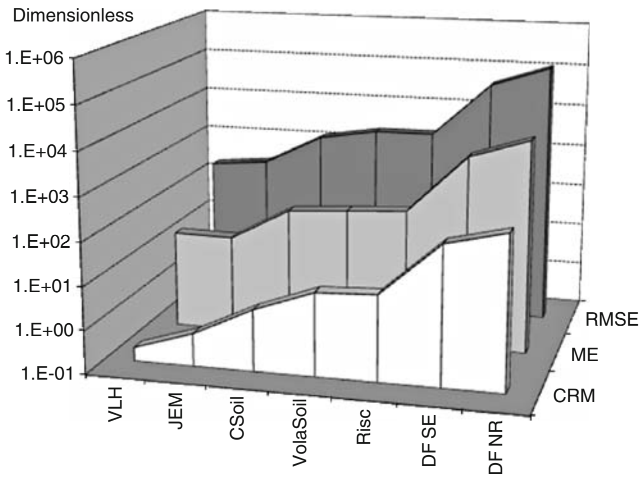


Fig. 5 Criteria values for indoor air. *VLH* Vlier–Humaan, *JEM* Johnson and Ettinger model, *DF* dilution factor, *SE* Sweden, *NR* Norway, *ME* maximum relative error (ME), *RMSE* root mean squared error, *CRM* coefficient of residual mass

algorithms with the highest accuracy for predicting the indoor air concentration are the JEM and Vlier–Humaan algorithms. However, the most suitable algorithms to serve for screening purposes are CSoil, VolaSoil and Risc, since they are sufficiently conservative, have fewer false-negative predictions than the Vlier–Humaan and JEM algorithms and still have sufficient discriminatory power. Therefore, it is found that algorithms may rank differently as to accuracy and suitability as screening tool.

The present study shows that the screening-level algorithms appear to be overall sufficiently conservative for regulatory purposes, but that they differed in their accuracy and therefore ability to exclude false-negative predictions. The algorithms that are closest to satisfy the purpose of screening are CSoil, VolaSoil and Risc.

5 Discussion

The results from the present study are in line with earlier testing of accurateness based on comparing predictions with observations (Johnson et al. 2002; Tillman and Weaver 2006; Hers et al. 2002, 2003; Huijsmans and Wezenbeek 1995; Van Wijnen and Lijzen 2006; Evans et al. 2002; Provoost et al. 2009). Their results focussed however on one or several screening-level algorithms and were therefore not able to rank the algorithm according to their accuracy, except in Provoost et al. (2009) where algorithms were ranked for vapour intrusion as a result of groundwater contamination.

It should be noted that another ranking of algorithms may be obtained for vapour intrusion from the vadose zone if data from other sites or other contaminants are considered, especially when other transport processes (e.g. degradation), sources (e.g. groundwater) or building types (e.g. crawl space) apply. In Provoost et al. (2009), a different ranking was obtained for vapour intrusion

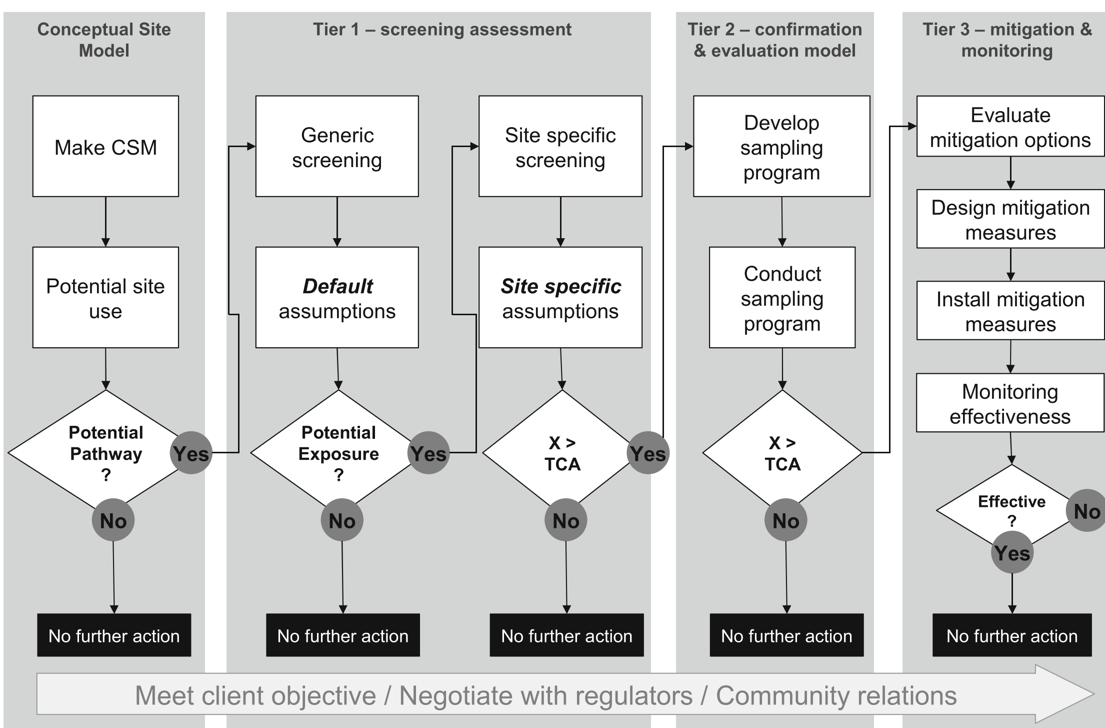


Fig. 6 Flow sheet for multiple lines of evidence. *CSM* conceptual site model, *TCA* tolerable concentration in air

from groundwater by using the same screening-level algorithms.

Screening-level algorithms tend to overestimate the soil air concentrations (see Fig. 3), but this overprediction did not reflect in the data from the indoor air concentration (see Fig. 4). Differences between soil air and indoor air predictions and observations are up to four orders of magnitude and can be partially related to different parameters in or excluded in a given algorithm and the mathematical concept used, for example, the inclusion or exclusion of temperature correction for the Henry constant in the mathematical concept or the inclusion or exclusion of convection as a transport process. It is not clear why all algorithms tend to overpredict the soil air concentration, and the concept of soil/air equilibrium partitioning of organic VOC in a homogenous porous soil should be revised and alternatives suggested (Goss and Schwarzenbach 2001; Goss 2004).

Several studies like those of Hers et al. (2002, 2003), Evans et al. 2002, Ririe et al. 2002, Devaull (2007) and Hohener et al. (2006) have investigated and suggested that biodegradation of VOCs in the soil air might be a factor that contributes to the differences between predictions and observations, but further research is needed to predict for what contaminants and in what soil types this occurs.

6 Recommendations and perspectives

Given the over-predictions and under-predictions of the screening-level algorithms considered, a combination of modelling and measurements will mostly be required to produce multiple lines of evidence for deciding if an intolerable human health risk occurs or remedial actions at a site are needed. Integrated programmes of modelling and field observations can reduce the uncertainty related to predictions and observations (measurements). This is of importance since the present study, and that of Provoost et al. (2009), have demonstrated that the most accurate algorithms can produce false-negative errors. Figure 6 presents an adapted practical approach from BBL (2006) for creating multiple lines of evidence.

The approach starts with the consideration if vapour intrusion is a potential route of exposure by examining the conceptual site model. If the use of the site prevents exposure, vapour intrusion needs no further investigation. However, if a vapour intrusion is a potential pathway, a tier-1 screening assessment can be conducted.

Tier 1 starts with a generic screening prediction of the indoor air with conservative default algorithm assumptions followed by a more site-specific screening that includes site-specific parameter values for predicting the indoor air concentration. It is important that the algorithm used in the generic screening does not produce false-negative predictions. The more site-specific screening allows the use of more accurate algorithms to predict the indoor air concentration and exceedance of the tolerable concentration in the air. Vadose zone and groundwater of soil air concentrations are mostly the source of the vapour intrusion, and sufficient special and temporal variation should be included in this tier.

Tier 2 confirms or rejects the exceedance of the tolerable concentration in air by conducting a sampling programme in the soil air and/or indoor air. The sampling programme should include sufficient spatial and temporal variation since indoor air concentrations may vary substantially over time.

Tier 3 includes mitigations and monitoring activities that prevent the exposure to contaminants of concern. It is important to monitor the effectiveness of mitigation measures since soil air migration might change over time and more information about mitigation can be found in EPA (1993), Eklund et al. (2007), Folkes (2002) and ITRC (2007).

For the derivation of a soil screening value or preliminary remediation goal, a more conservative model algorithm or an accurate algorithm with a more conservative parameter set should be used. Many factors affect the final soil screening values and more details are given in Provoost et al. (2008).

Parameters from the different algorithms have a wide range of values, for example, soil air permeability or indoor air exchange rates. A probabilistic approach with a sensitivity analysis could determine which parameters contribute most to the variation in predicted air concentrations and therefore reveal parameters whose values need to be reviewed or adapted to arrive at a reasonable safe level of conservatism.

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