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# VAPOR EMISSIONS FROM CONTAMINATED SOILS INTO BUILDINGS: COMPARISON BETWEEN PREDICTIONS FROM TRANSPORT MODELS AND FIELD MEASUREMENTS

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## 1. Summary

Soil vapor migration into house, with subsequent inhalation, is often the main exposure pathway to humans at sites contaminated with Volatile Organic Compounds (VOCs). In the case of VOCs contamination, quantification of indoor gas concentrations is therefore essential while assessing risks for human health.

Two approaches are commonly used for quantification of indoor concentrations: indoor gas measurement or transfer modelization from the source (soil, soil vapor phase, groundwater).

Model development is relatively well advanced but measurements for model calibration and "validation" hardly exist in the literature. Furthermore, predictions of indoor gas concentrations from different models may vary by several orders of magnitude, depending on the application. Therefore, the validity of the risk calculation obtained through models, and hence of the site management, remains highly uncertain.

The research project presented here aims at providing such "validation" data. Long-term goals of the research are improvement of modelization and also development of tools for site-related model-selection.

The program consisted in comparing modelization and experimental measurements on a test site. The site was a former factory with a concrete slab, contaminated with chlorinated solvents (trichloroethene and perchloroethylene) in unsaturated soils. Measurements concerned contaminant concentrations and fluxes in different media and at different transfer stages, but also key model parameters. The equations of Johnson & Ettinger and VOLASOIL models were used.

Air concentrations measured at various times show significant variations, and also differ from model prediction by one or two orders of magnitude. Despite thorough parameter measurements, uncertainty on input values, related to site heterogeneity, induced high uncertainty in the modelization. The preliminary results presented here show intrinsic limitation of some measurements and hence of model validation, but also the need for data on more sites, including very important site instrumentation which would allow to document the impact of site-heterogeneity.

## 2. Introduction

Soil vapor migration into house, with subsequent inhalation, is often the main exposure pathway to humans at sites contaminated with Volatile Organic Compounds (VOCs). In the case of VOCs contamination, quantification of indoor air concentrations is therefore essential while assessing risks for human health.

Two approaches are commonly used to produce indoor air concentrations:

- repeated indoor air measurement;
- transfer modelization from the source (soil, soil vapor phase, groundwater), from concentrations measured in soil, in groundwater or in soil gas.

Soil-vapor modeling is often required to predict the risk on such sites: modelization can limit the uncertainty in measurements due to the high time-variability of the soil gas to indoor air transfer, and also allows predictions for future situations. Several soil vapor intrusion models have been developed to predict indoor air concentrations. The most frequently used in France, Europe and probably in the world, are:

- the VOLASOIL model from the Dutch RIVM (Waitz *et al*, 1996), which is included in the Dutch computer model RISC-HUMAN;
- the model developed by Johnson & Ettinger (1991), proposed in Johnson & Ettinger computer model from the US EPA (1997), which is included in many American or British commercial computer models, such as RBCA, RISC BP,....;
- empirical "rules of thumb" on dilution factors between soil air and atmospheric air (Rippen, 2002).

The "rules of thumb" model is hardly adaptable for a precise site management, and of limited accuracy considering the main factors which may impact on the transfer.

The physical models VOLASOIL and Johnson & Ettinger are generally used as "black boxes", the site-suitability and validity of which remains unknown. Actually, conceptual validation of the equations can be found, either directly in the official model presentation for VOLASOIL (Waitz *et al*, 1996), or in the background literature for Johnson & Ettinger and some steps of VOLASOIL.

On the contrary, one common criticism of several models is the lack of experimental validation. Few or no studies for certain models, or only for some compounds (pesticides, benzene) were carried out in order to evaluate the operational use for each model, and compare models predictions with field or laboratory data (Evans & al. 2000; Olson *et al*. 2001).

The Johnson & Ettinger parameters were calibrated with actual empirical results from radon (Nazaroff, 1988), whereas the calibration of the VOLASOIL model is not documented in Waitz *et al* (1996).

So far as we know, validation through further comparison between models predictions and field measurements is not reported for neither VOLASOIL (Lijzen, 2001) nor Johnson & Ettinger. Furthermore, predictions of indoor air concentrations from different models may vary by several orders of magnitude, depending on the application. Therefore, the validity of the risk calculation obtained through those models, and hence of the site management, remains highly uncertain.

That is why INERIS launched a long term research project, co-funded by the MEDD (Ministère de l'Ecologie et du Développement Durable) and ADEME (Agence de l'Environnement et de la Maîtrise de l'Energie), which aims to document the empirical validity of those models in different relevant situations.

Long-term goals of the research are improvement of modelization and also development of tools for site-related model-selection.

The following procedure was used to conduct the comparison-study between transport models predictions and field measurements:

- review of literature and analysis of transport models in terms of their modeling conditions, parameters sensitivity..., and of case studies;
- field studies on contaminated sites;
- comparison of calculated indoor air concentrations with field measurements using different models and site input parameters;
- evaluation and interpretation based on models concepts and input parameters.

This paper reports the first field study realized and its preliminary results, and draws some consequence for the whole program.

### **3. Experimental methods**

#### **3.1. EXPERIMENTAL SITE**

The study reported here was set up in a part of a former factory located in urban zone. Former investigations had shown a main pollution by chlorinated solvents (trichloroethylene - TCE, tetrachloroethylene - PCE) in soil and soil air (2-8 m deep), and in groundwater (about 25 m). The study area corresponds to a building with a concrete slab, surrounded by old buildings, asphalt or concrete roads and bare ground.

The investigation realized for the study aimed at measuring:

- the contaminant transfers between media (soil, soil air, flux at the surface, indoor air);
- the model key parameters: source depth, soil and concrete characteristics, ventilation and other characteristics of the building.

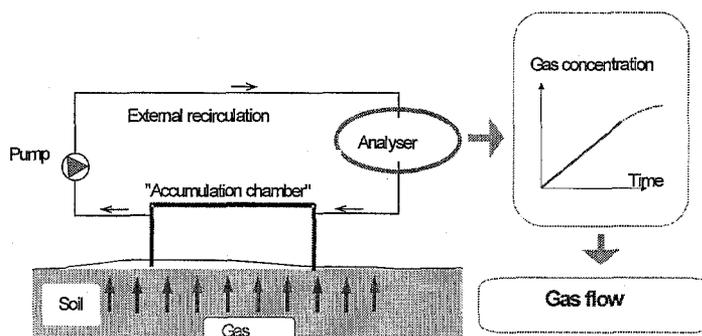
### 3.2. MEASUREMENTS

#### Flux measurements at the soil surface

Two measurement materials developed by INERIS were chosen to estimate flow rates of chlorinated solvents.

*The "accumulation chamber":*

This system (Pokryszka & al. 1999) is based upon a principle of accumulation chamber connected with an analyzer, in this case a FID (Figure 1). Gases emitted from the site within an elementary surface (50 cm x 50 cm) are pumped, sent to the analyzer and reinjected into the chamber, creating a recirculating flow. In that way the atmosphere enclosed in the chamber is enriched with contaminant compounds. The local flow can be easily deduced from the measurement of the atmosphere enrichment as a function of time. The flow of contaminant gas is expressed in relative total flow, in this case, PCE equivalent.



**Figure 1 : Flux chamber diagram** Figure 1: "Accumulation chamber" diagram

This measurement system is easy to operate. One measurement can be performed every 5 to 10 minutes (therefore 30 to 60 measurements per day), which ensures a rather dense sampling grid in the study area. Due to the low precision of the FID, for low measured concentrations (as in the case of the site), this tool provides only indicative and relative results.

*The "dynamic scanning chamber":*

The "dynamic scanning-chamber" (Pokryszka & al. 1995) displayed in Figure 2 is a chamber connected to an inert gas supply system and an exit with measurement points. The system gathers contaminant gas emitted from soil within the inert gas flow. The mixed gas sampled on charcoal are analyzed. Therefore the measured parameters (concentrations and outgoing air-flows) allow to quantify the flow of emitted vapors outgoing from the chamber, and thus to deduce the flow released through the concrete slab.

Because of the size of the chamber (250 cm x 100 cm x 15 cm) and its time of operating system, and also those of gas pumping required for the analysis of this case, the portability is not real. Only between 1 and 3 points per day may be measured.

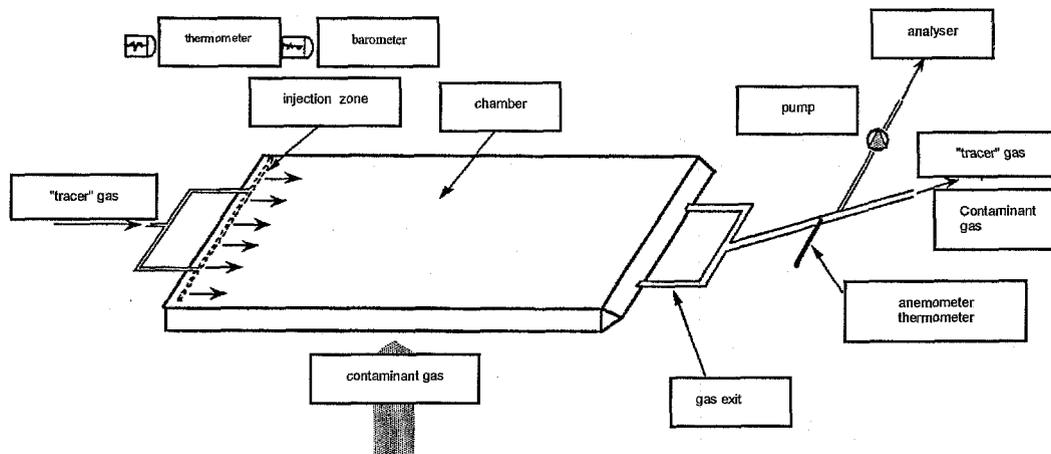


Figure 2: "Dynamic scanning chamber" diagram

### Ventilation rate measurement

A tracer-gas, in this case SF6, was injected into the indoor air. Air samples were collected at two heights (1 m and 4 m) at different times and analyzed by chromatography. The air exchange rate is then calculated using the following formula:

$$\ln(C/C_0) = -x(t-t_0),$$

where C is the tracer-gas concentration at time t, C0 is the tracer gas concentration at time t0, x is the air exchange rate in the buildings.

The results of the field survey of November 2002 are given in Figure 3.

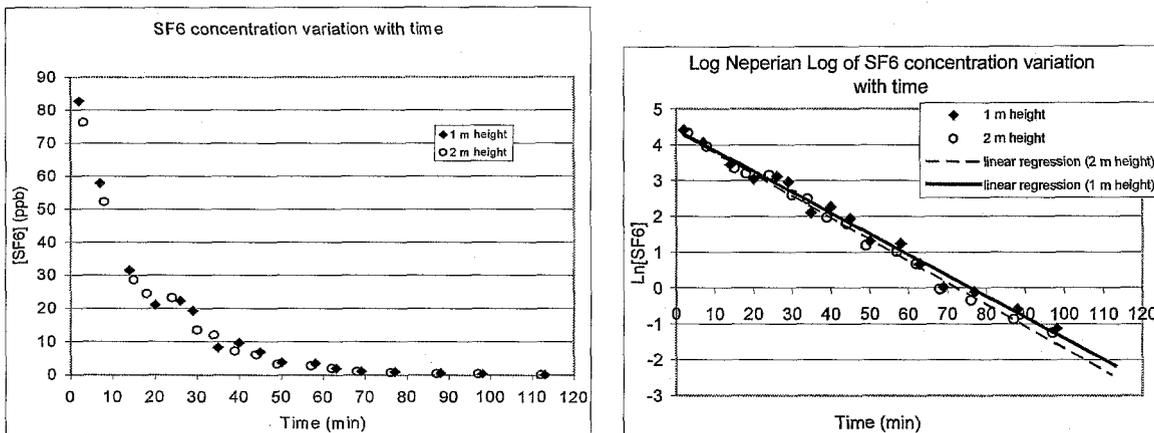


Figure 3: Air exchange rate

### Sampling, soil gas wells, concentrations measurements

Soil was sampled at different depths using techniques which limit vapor loss: quick sampling from an hollow shape boring into brown-glass, gas-proof glasses,...

Soil gas wells were built in the bore-holes: they consist of:

- a 1 1/4 " full-HDPE tube surrounded by clay at no-sampling depths;
- a 1 1/4 " filter-HDPE tube surrounded by gravel at sampling depth;
- a HDPE gas-proof sealing with sampling valve.

Indoor air or soil air concentrations were pumped through charcoal at a rate (< 1 l/ min) and with a volume in the recommended constructor range. The charcoal was then analyzed.

### Atmospheric / soil pressure difference

The atmospheric / soil pressure difference was measured using a differential pressure measuring device, with a detection limit and a precision of 1 Pa.

### Other measurements

Other measurements (soil and concrete water-permeability, grain size distribution,...) were performed according to relevant standards.

### 3.3. FIELD INVESTIGATIONS

Investigations performed:

In March 2001, nine soil gas wells have been installed at various depths in the unsaturated zone in a selected area in and outside of the building. According to existing data on lithology and contamination, soil air samples were collected at various depths between 0,4-1,3 m and 6,8-8 m.

In order to consider the potential seasonal variations of the vapor emissions, field investigations were performed at different seasons (April, July, September, November).

Samples were collected to analyze the following parameters :

- organic compounds in the soil : benzene, toluene, ethylbenzene, xylenes (BTEX) and volatile halogenated organic compounds (VHOC) such as trichloroethylene, tetrachloroethylene, cis and trans dichloroethylene, vinylchloride;
- physico-chemical parameters of the soils : organic carbon fraction, water permeability, grain size distribution...;
- physical parameters of the concrete slab;
- organic compounds in the soil air, indoor and outdoor air (also with the "dynamic scanning chamber"), with sampling on charcoal.

About sixty measurements of soil-to-air fluxes with the "accumulation chamber" were carried out at two field investigations (April and July). Firstly according to a regular grid, and secondly following the results, at points where the highest fluxes had been recorded.

The "dynamic scanning chamber" points were located on the points of highest fluxes as measured by the "accumulation chamber" (4 points).

The pressure difference between soil air and indoor air was measured twice (July, September), and the air exchange rate of the building once (November).

The summary of the contaminant measures for trichloroethylene (TCE) and tetrachloroethylene (PCE)<sup>1</sup>, taking account March to September data, are presented in Table 1:

Media	Unit	TCE			PCE		
		April	July	Sept	April	July	Sept
<b>Soil</b>	µg/kg	1 - 8			< 1 <sup>(*)</sup> - 15		
Surface flux, "accumulation chamber"	µg/m <sup>2</sup> /s eq. PCE	< 0,35 - 1 (FID, ppm levels)					
<b>Surface flux,</b> "dynamic scanning chamber"	µg/m <sup>2</sup> /s	< E-4 - 0,006			0,4 E-4 - 0,025		
<b>Soil-gas</b>	mg/m <sup>3</sup>	0,15 - 38			0,73 - 1900		
<b>Indoor air</b>	µg/m <sup>3</sup>	<1 <sup>(*)</sup> - 1	<1 <sup>(*)</sup> - 15	nd	0,4 - 2	2 - 40	nd
<b>Background air</b>	µg/m <sup>3</sup>	<1 <sup>(*)</sup>	1	nd	0,3 - 0,7	<0,3 <sup>(*)</sup>	nd

(\*): below quantification limit; nd : not detected<sup>2</sup>

Table 1: Summary of the contaminant measures for TCE and PCE

<sup>1</sup> Other components proved not to be relevant, due to very low concentrations. Results for non-contaminant parameters are presented below with the parametrisation of the modelization.

<sup>2</sup> Due to sampling or analyse: results are below detection limit (<0,04 µg/m<sup>3</sup> TCE and <0,08 µg/m<sup>3</sup> PCE on all points) are discarded because they do not appear compatible with usual background concentrations (>0,1 µg/m<sup>3</sup>).

Those investigation results show significant time-variations in soil air and indoor air concentrations and in fluxes. No coherence between those time-variations could be observed considering:

- soil gas concentrations, fluxes, and indoor air concentrations: the highest concentrations in indoor air (July) were neither linked to highest soil air concentrations, nor to the highest fluxes with the "dynamic scanning chamber";
- spatial repartition for Surface flux, "accumulation chamber";
- soil gas spatial repartition in one campaign compared to the two others.

Possible causes for those time-variations and uncertainty sources include:

- meteorological conditions;
- major excavation works 50 m from the building;
- analytical results? But measures on soil gas samples which were repeated within a day showed less than 30% variation.

On the contrary, a relatively good spatial coherence was observed between soil concentrations, soil gas concentrations (in two campaigns out of three), Surface flux obtained with the "dynamic chamber" (one campaign with enough points), and indoor air concentrations (one campaign with enough points)<sup>3</sup>.

#### 4. Modelization

Simulations have been conducted with the equations of The Johnson & Ettinger (Johnson & Ettinger, 1991 ; with infinite source equations) and VOLASOIL (Waitz *et al*, 1996) models. The main characteristics of those two models are summarized in table 2.

Reference	Model type	Source pollution	Vapor transport	Building foundation
Johnson & Ettinger (Johnson & Ettinger, 1991)	Steady-state Semi-analytical	Finite or infinite	Diffusion and convection	Basement or constructed slab-on-grade
VOLASOIL (Waitz <i>et al</i> , 1996)	Steady-state Semi-analytical	Infinite	Diffusion and convection	Crawl space

Table 2: Main model characteristics

The VOLASOIL model was designed and explained for the case of a crawl space and a one-layer soil over the contamination source which is not relevant to the situation of the experimental site. Therefore, starting from the basic diffusion and convection equations, we developed the VOLASOIL equations for a multi-layer soil, which allowed us to apply it for a concrete layer over a soil layer.

Main parameters used in modeling and input values are presented in Table 3. Site specific values were used when available.

<sup>3</sup> No spatial coherence was observed on fluxes between the two chamber tools, but this can not be commented, considering the solely indicative role of the accumulation chamber in the case of the low FID values measured.

			PARAMETRISATION	
<b>SOIL</b>			Filling: gravel, sand, silt	
Organic carbon fraction		Measured		
Dry bulk density	1,7	From standard values of similar soils, with filling of gravel gaps by sand and coherence with measured water content		
Total porosity	0,25			
Water content	0,1			
Water-filled porosity	0,19			
Air-filled porosity	0,06			
Gas permeability	E-12 m <sup>2</sup>	~ water permeability of 8E-6 m/s: from measured water permeability (4E-8 m/s) and standard values for similar soils (5E-5 m/s)		
<b>BUILDING</b>				
Slab characteristics regarding diffusion	Standard values (HESP model, wet slab), (8 times less diffusive than dry concrete)			
Slab characteristics regarding convection	Standard values from each model			
Length, width, height	26 - 20 - 6,5 m	Measured		
Floor thickness	0,1 m	Average measured		
Soil-building pressure difference	1 Pa <sup>4</sup>	Measured (July, September): < 1 Pa		
Air exchange rate	3,6 h <sup>-1</sup>	Measured (December)		
<b>SOURCE</b>				
Depth	2,5 m <sup>5</sup>	Minimal depth of maximal concentrations in soil gas or in soil		
Soil concentrations	4,8 µg/kg TCE, 5,2 µg/kg PCE		Average March	
Soil gas concentrations	6,1 mg/m <sup>3</sup> TCE, 43 mg/m <sup>3</sup> PCE		Average July	

Table 3: Main modeling parameters

Based on site observations, the soil layer above the source was considered homogeneous. Actually, the site observations did not prove precise enough for the needed characterization of the diffusion and convection parameters. For all parameters, the overall coherence of obtained values was checked. That led to the revision of all parameter values for the upper soil layer: before revision for example, the input values would have induced a negative air-filled porosity.

The soil / building pressure difference was so low (< 1 Pa) that it could not be measured. This can be due to the lack of heating of the building (unused) and to the general soil coverage with concrete or asphalt slab and with buildings, which both are in contradiction with the assumptions for typical pressure differences (4 Pa).

Therefore, the uncertainty of the integrated diffusion (effective diffusivity) and convection parameter (soil gas permeability, pressure difference) appears very high (x or / 20 or more), with a corresponding impact on model predictions. More precisely, intermediate results from the modelization show that in this case, the VOCs transport mechanism is:

- mainly diffusion with VOLASOIL (60-80%), limited by air-filled porosity;
- mainly convection with Johnson and Ettinger (99%), limited by soil gas permeability.

<sup>4</sup> Further simulations with 0,1 Pa, not reported here, show a reduction of the building air concentration (factor between 1,6 to 5, depending of the models) with the permeability reported here.

<sup>5</sup> A further simulation with 1 m, not reported here, shows an increasing of the building air concentration (factor between 1 to 2, depending of the models).

In Table 4, the main modelization results, starting from soil or soil gas concentrations, are summarized and compared to measurement results.

	TCE					PCE				
	Measured (average, <5m)			Calculated with		Measured (average, <5m)			Calculated with	
				VOLASOIL	J&E				VOLASOIL	J&E
<i>Modelization starting from soil concentrations</i>										
Soil ( $\mu\text{g}/\text{kg}$ )	4,8						5,2			
	April	July	Sept			April	July	Sept		
Soil-gas ( $\text{mg}/\text{m}^3$ )	2,1	6,1	14	3,3		24	43	421	2,9	
Surface flux ( $\mu\text{g}/\text{m}^2/\text{s}$ ) (Dynamic chamber)	<E-4 n=3	~E-3 n=2	<b>6E-3</b> n=1	9,2 E-5	9,1 E-6	~E-4 n=3	6E-3 n=2	<b>2E-2</b> n=1	1,6 E-4	1,3 E-5
Building air ( $\mu\text{g}/\text{m}^3$ )	<1 <sup>(*)</sup>	5,6	nd	0,014	0,001	1	17	nd	0,024	0,002
<i>Modelization starting from soil-gas concentrations (one synchronized campaign : July)</i>										
Soil-gas ( $\text{mg}/\text{m}^3$ )	6,1						43			
Surface flux ( $\mu\text{g}/\text{m}^2/\text{s}$ )	~E-3 n=2		1,7 E-4		1,8 E-5	6E-3 n=2		2,3 E-3		2,1E-4
Building air ( $\mu\text{g}/\text{m}^3$ )	5,6			0,03	0,003	17		0,36		0,034

(\*): below quantification limit; nd : not detected

Tableau 4: Modelization results

The results show important differences between predicted and observed values in this case:

Soil / soil-gas repartition: Measured values are equal to 1 to 150 times the predicted values. Possible reasons could include uncertainty on soil concentrations or source depth.

Indoor air and surface flux: Measured values are equal to:

- $\leq 10$  to 10.000 times the predicted values starting from soil concentrations;
- $\leq 50$  to 1.000 times the predicted values starting from soil-gas concentrations.

Modelization with VOLASOIL equations appears more conservative than with J&E equations in this case.

Possible reasons could include uncertainty on concentrations (soils, soil-gas,...), and on soil and slab parameters:

- with VOLASOIL, an air-filled porosity of 0,21 instead of 0,06 leads to fluxes and indoor concentrations 15 times higher;
- with Johnson and Ettinger, an air permeability of  $5\text{E}-15 \text{ m}^2$  instead of  $\text{E}-12 \text{ m}^2$  leads to fluxes and indoor concentrations 75 times lower.

Possible reasons could also include validity problems in the models, but possibilities for accurate interpretation in that direction are strictly limited due to the high possible impact of other uncertainties.

## 5. Conclusions

In this experiment, important difference between predicted and measured values were observed. But possibilities for accurate interpretation in term of model validity are strictly limited due to the high possible impact of other uncertainties which still remain in spite of intensive site-instrumentation.

The time variability of field results and the impacts of remaining uncertainties on some input parameters on model results demonstrate the importance of precise measurements and quantification of the main site parameters. Precision on some input parameters (diffusion parameters, soil gas permeability) can be improved by more measures, but may still be intrinsically limited in regard to the needs of the modelization (depending of the site characteristics).

Documenting the empirical validity of models is always a difficult and time-consuming work. This is especially true for soil-atmosphere vapor transfer models, where an accurate quantification of all the main site parameters is hardly reachable, due to soil heterogeneity and limits of the measurement methods. Important instrumentation, several field measurements on each site and on different sites, are required.

More field data are needed, in particular different building designs, constructed materials, soils.... Sites both adequate for characterization ("simple", homogeneous soil instead of filling) and representative of typical model conditions (heating of the building,...) should be looked for, which may prove difficult: used (heated) buildings on sites contaminated with VOCs are quite seldom.

That is why gathering all works on the subject would be necessary with the collaboration of all concerned private and public entities.

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