

# MODELING OF THE CONTAMINATION IN THE ALSATIAN AQUIFER BY CHLORINATED SOLOVENTS

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## ABSTRACT

The Alsatian aquifer in the southern upper Rhine valley in the north-east of France has been contaminated with carbon tetrachloride ( $\text{CCl}_4$ ) on account of a tanker accident happened on 1970. Several years after the accident, abnormal concentrations of  $\text{CCl}_4$ , have been detected in the underground water aquifer. Measured data have been collected from different locations of the aquifer during the period between 1992 and 2004. To match the history and predict the distribution of the contaminant, one requires knowledge of: (i) the behavior of the contaminant source, (ii) the geophysical parameters that affect water flow and contaminant transfer in the aquifer. In this work, we show the mathematical and numerical modeling of the problem. All measured data collected between 1992 and 2004 were used to calibrate our model and simulations are performed for the period from 1970 to 2024. The chemical  $\text{CCl}_4$  is modeled as a tracer in water (that is, single-phase flow). Numerical results were obtained with an efficient, 3D single-phase code “TRACES”, developed at the IMFS. This code combines the mixed-hybrid finite element and discontinuous Galerkin methods to solve both the flow and the transport problems. No information is available about the behavior of the contaminant at the source. To estimate the contaminant concentration at the source, the travel time between the source and measurement-wells is calculated by the method of moments. Another difficulty one had to overcome was because no measured data of the rock and fluid parameters such as hydraulic conductivity, porosity, longitudinal and transversal dispersivities, were available. Several simulations were thus performed to get the parameters that match best the history. We show the sensitivity of the transport processes to the geophysical parameters (permeability, porosity) and longitudinal and transversal dispersivities. The results with the fitted parameters show a good agreement with the measured data.

## 1. INTRODUCTION.

The Alsatian aquifer is located in the north-eastern of France and extends to the border between France and Germany. Alsatian part of the Rhine Valley has a surface length of 160 km and a maximum width of 20 km. This aquifer represents the most important aquifer in Europe. The groundwater reservoir contains about 50 billion  $\text{m}^3$  of water, with an annual renewal of 1.3 billion  $\text{m}^3$ .

In 1970, a tanker truck containing carbon tetrachloride ( $\text{CCl}_4$ ) capsized at the North of Benfeld (a small town about 35 km South of Strasbourg, eastern France). According to a note of SGAL (1971), about 4000 l of carbon tetrachloride, (nearly 1056 gallons) were spread on the area of the accident, infiltrating in the ground or disappearing by evaporation. In 1992, the analyses carried out by BRGM (1971), showed abnormal quantities of  $\text{CCl}_4$  in the supplies of

drinking water located downstream of Erstein (about 60  $\mu\text{g/l}$ ). The Environmental Protection Agency (EPA) has set a limit for carbon tetrachloride in drinking water of 5 parts of carbon tetrachloride per billion parts of water (5 ppb).

The goal of this work is to provide a numerical study to match the reported data of the contaminant and eventually to predict its migration in the aquifer. This requires knowledge of:

- The source behavior at the accident location.
- The fluid properties such as the diffusion and degradation coefficients.
- The rock properties (porosity, permeability) for different layers in heterogeneous medium.
- The boundary conditions of the hydrodynamic problem in the aquifer.

Vigouroux et al. 1983, Hamond et al. 1995, and Beyou 1999 have been carried out several works, to study the hydrodynamic state of the aquifer and to predict the pollution transfer.

We have use a numerical model (TRACES) that combines the mixed-hybrid finite element (MHFE) and discontinuous Galerkin (DG) methods to solve the hydrodynamic state and mass transfer problems.

The following assumption are considered in our model:

- The pollutant  $\text{CCl}_4$  is considered as a tracer in water. We are interested in the dissolved  $\text{CCl}_4$ . A single-phase flow is thus assumed.
- The degradation mechanism is ignored. Since the aquifer is aerobic (dissolved oxygen is more than 1  $\mu\text{g/l}$ ), the degradation of  $\text{CCl}_4$  is not significant (Vannelli et al., 1990).
- The adsorption reaction is also ignored since the distribution coefficient;  $K_d$  of  $\text{CCl}_4$  is negligible (about 0.11 L/Kg, Schafer, 2001).
- Our work concerns the saturated zone so that the volatilization was not included since it is generally only significant in unsaturated zone (vadose).

The source term versus time is defined by estimating the travel time of the contaminant between the source and the measurement-wells. Several simulations were thus performed to get the parameters that match best the history. The permeability was calibrated with several runs carried out by changing randomly the permeability of each zone. Porosity, longitudinal and transversal dispersivities were also fitted to the transport model.

## 2. MODEL STRUCTURE AND DESCRIPTION.

The aquifer domain is discretized using a 3D triangular prismatic grid with 25388 nodes and 45460 elements. The source term is represented by eight mesh elements (each layer 2 mesh elements). The domain is divided into 11 layers and each layer has a depth of about 10 m. The numerical model TRACES is based on steady-state groundwater flow and transient solute transport in 3D heterogeneous media.

In our work, we consider two computational domains; a far field and a near field (see Figure 1). The calculated hydraulic heads in the far field are used to define the boundary conditions of the near field, where the contaminant migration takes place. The computational far field is considered as a 2D domain, where the hydraulic head is computed using a 2D flow model. The calculated hydraulic head at the boundaries of the near field is used as boundary conditions for the flow inside the 3D near field domain.

The types of the boundary conditions are the following:

- Dirichlet boundary conditions, defined by known heads, are imposed at the south and the underlying domain (see Figure 1);
- Neumann boundary conditions, defined by known fluxes are set on the western boundary.
- The eastern boundaries along the Rhine borders are described by an aquifer-river exchange model.

For the 2D model, the studied area is bounded in the south by the Kogenheim city, in the east by the Rhine, in the North by the Strasbourg city, and in the west by the border of the aquifer. The studied surface has an average width of about 25 km and a length of less than 35 km from north to south. The 3D near field, which is in the area of Benfeld and downstream of Erstein, is defined by a cubic domain of width of 6 km, length of 20 km, and depth of 110 km (see Figure 1).

The hydraulic head and the velocity field are described by Darcy's law and mass conservation equations and the mass transfer is described by a diffusion/dispersion-convection-reaction equation. The 2D/3D numerical model TRACES, which is developed at IMFS, combines the mixed-hybrid finite element and discontinuous finite element to solve the hydrodynamic state and mass transfer in the porous media.

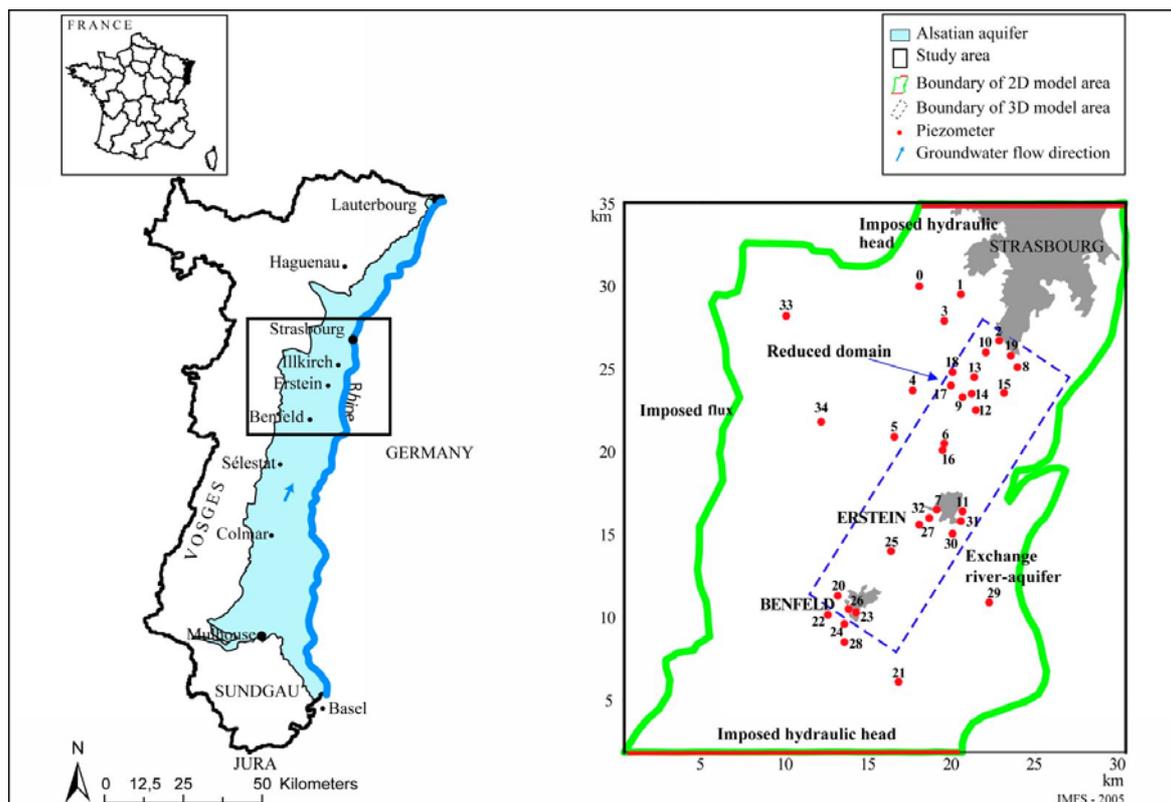


FIGURE 1. Geographic situation of the Alsatian aquifer, Alsace region, France. The map on the right shows the piezometers and the models boundary.

### 3. MODEL APPLICATION.

**3.1 Estimate of the source term.** The three dimensional model is used in order to study the behavior of the contaminant source at different depths. In order to estimate the source behavior we must assumed: 1) the source location, as  $\text{CCl}_4$  is highly volatile. The location selected depends on the highest concentration of measured data, which are near Benfeld. 2) the pollution depth and surface of the infiltration by approximating calculations. As the volume of the pollutant infiltrated, ( $\text{CCl}_4$ ), was  $4 \text{ m}^3$ , the pollution depth assumed was 35 m and the surface of infiltration is 7 to  $37 \text{ m}^3$ .

The technique used to estimate the source function at the first four layers is as follows (see Figure 2).

First, we fixed the concentration at the four layers of the source. Then, we used the code TRACES to calculate the concentration at each piezometer in the domain and for a given time. In order to find the concentration at the source, we matched the calculated concentrations with measured concentrations using this formula:

$$C_s(t-t_c) = C_{init}(t-t_c) \frac{C_{mes}(t)}{C_{cal}(t)} \quad (1)$$

The travel time of the contaminant between the source and the measurement-wells is calculated by method of moment as follows:

$$t_c = \frac{\int_0^{\infty} C_{cal}(t) \cdot t \cdot dt}{\int_0^{\infty} C_{cal}(t) \cdot dt} \quad (2)$$

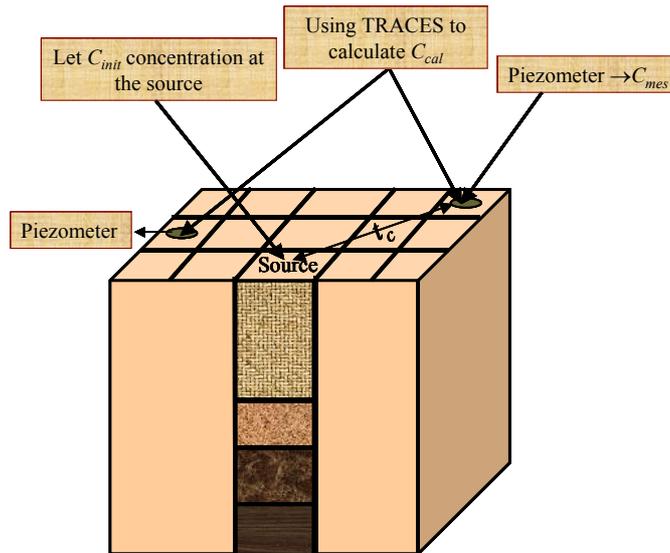


Figure 2. The technique used to estimate the source term.

The approximated concentrations at the source term was very oscillatory, In order to have a smooth behavior of the source function in each layer, the predicted concentrations at the source are approached by using 1) the mean value interpolation and 2) the fit exponential interpolation. The source functions in the four layers are depicted together in Figure (3) below:

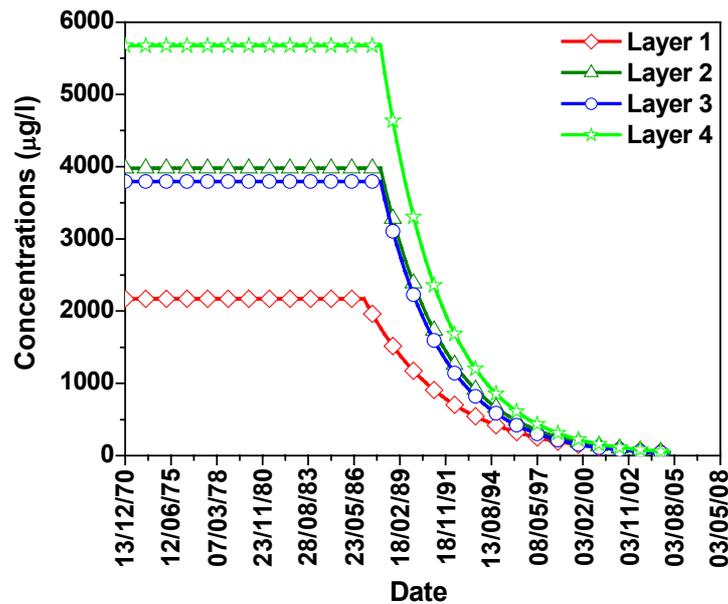


FIGURE 3. The source functions in the four layers.

**3.2 Calibration of the physical parameters.** The source functions presented in Figure (3) are considered in the numerical simulator (TRACES) to have the distribution of predicted concentrations of pollutant for the whole domain and for consequent years. In order to be able to obtain realistic results, we need realistic parameters in the mathematical model (such as hydraulic conductivity, longitudinal and transversal dispersivities, porosity). No measurement of these geophysical parameters is available in the domain. Depending on pervious studies on the aquifer, we calibrated some geophysical parameters like, porosity, and permeability to improve the results of simulation. The longitudinal and transversal dispersivities were fitted in the model by using prior information from similar cases (Gelhar et al., 1992) and geological formations. In order to compare the goodness of fit between the observed and predicted concentrations, we presented the observed and predicted concentrations on a scatter diagram. For the porosity, two values were fitted to transport model. (20% and 10%). The final calibrated values for the porosity was 10%; this porosity gives the smallest standard deviation (Figures 4-5):

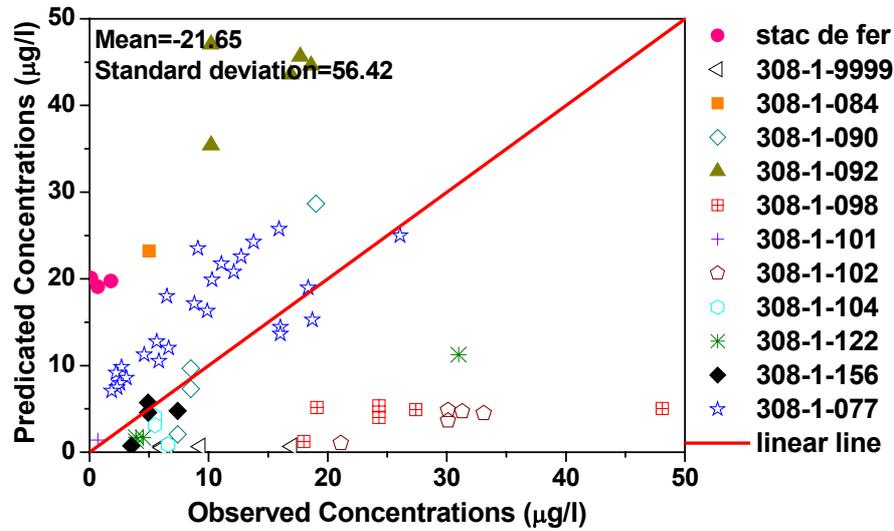


FIGURE 4. Comparing observed and predicted concentrations, (porosity 20%).

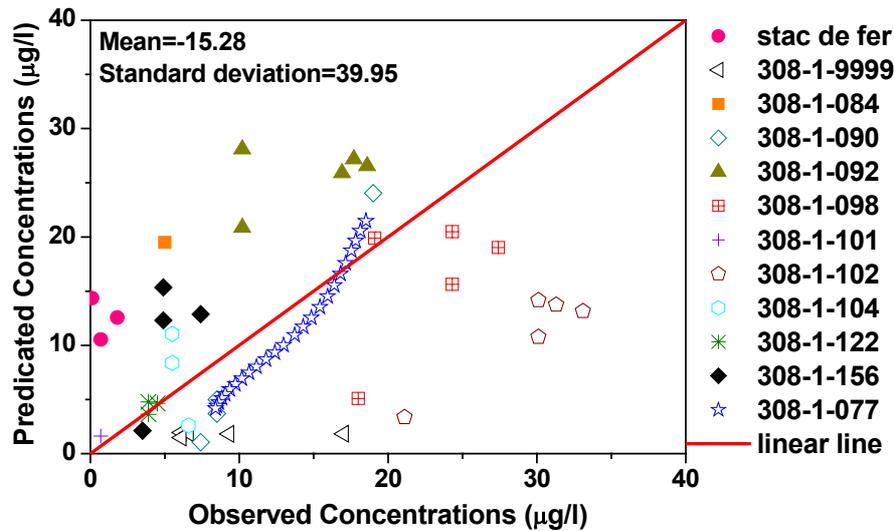


FIGURE 5. Comparing observed and predicted concentrations, (porosity 10%).

The dispersivities coefficients are calibrated with several runs to approach the solution. The longitudinal dispersivity is calibrated with values of 10 m and 20 m while the transversal dispersivity with (0.5, 1, 2 and 3) m. The results show that the reasonable values of the present transport model when both the longitudinal and transversal dispersivities coefficients were 3 m and 20 m respectively. See Figure (6).

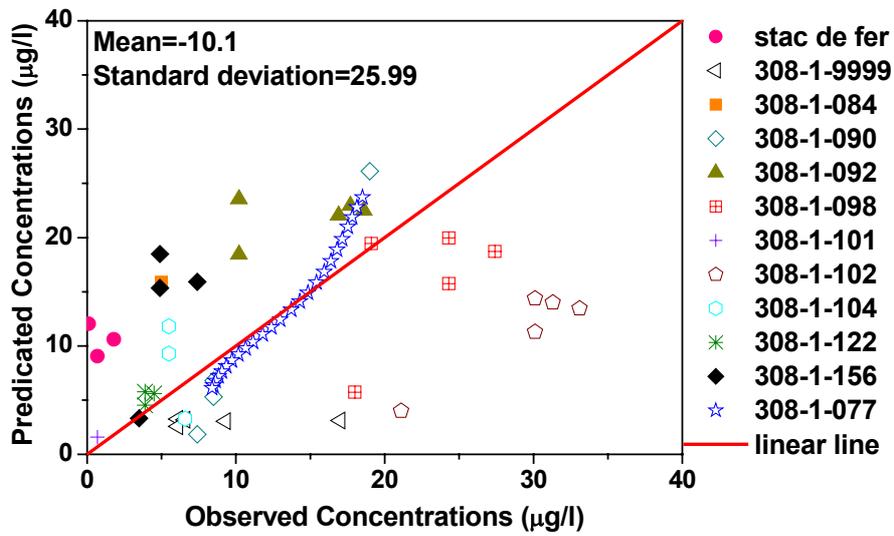


FIGURE 6. Comparing observed and predicted concentration,  $\alpha_t=3$  and  $\alpha_l=20$ .

The permeability of the transport model is calibrated also by changing it randomly for each zone of the domain. This calibration gives smaller standard deviation (see Figure 7).

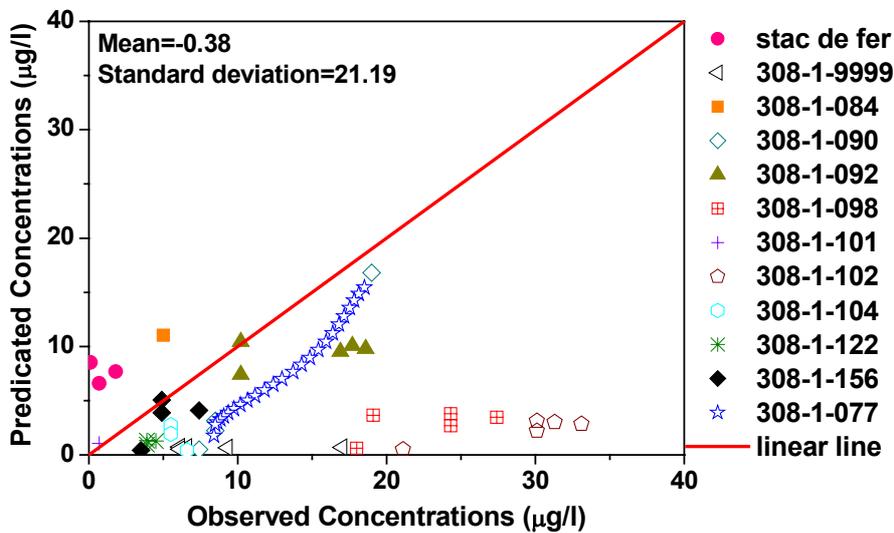


FIGURE 7. Comparing observed and predicted concentration, by calibrating the permeability of each zone.

The source function considered in the numerical model (TRACES) and the fitting parameters leads show also a good agreement between measured and computed concentrations at pumping well, Figure (8).

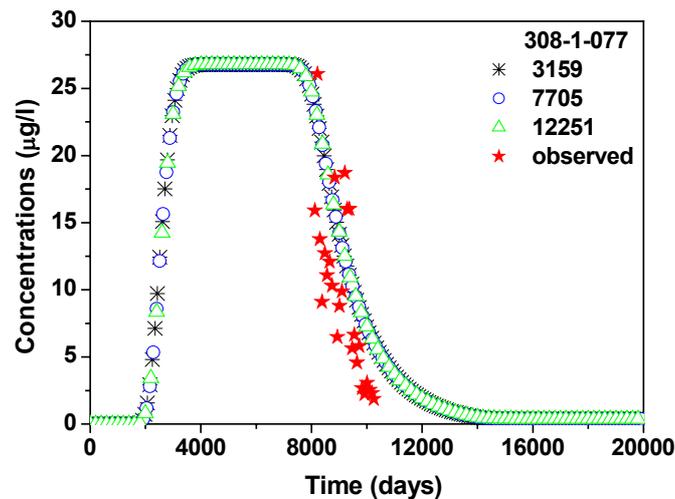


FIGURE 8. Comparison between measured and simulated concentrations in Negrodorf at 5515 m from the source

#### 4. CONCLUSION.

This work essentially concentrated on an accident happened in north-eastern of France on 1970. Our objective, in this work, is to model and simulate the contaminant ( $\text{CCl}_4$ ) transfer in the aquifer. We used a technique to determine the source term at the accident location. The model is calibrated using measured data that are collected during 12 years (1992 to 2004), and simulations are performed for the period from 1970 to 2024. The permeability was calibrated with several runs carried out by changing randomly the permeability of each zone in the domain. Porosity, longitudinal and transversal dispersivities were also fitted to the transport model. We showed the sensitivity of the transport processes to these parameters. The results with the fitted parameters showed a good agreement with the measured data.

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