ANN-based approach for the estimation aquifer pollutant source behaviour

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ABSTRACT

The problem of identifying an unknown pollution source in polluted aquifers, based on known contaminant concentrations measurement, is part of the broader group of issues, called inverse problems. This paper investigates the feasibility of solving the groundwater pollution inverse problem by using artificial neural networks (ANNs). The approach consists first in training an ANN to solve the direct problem, where the pollutant concentration in a set of monitoring wells is calculated for a known pollutant source. Successively, the trained ANN is frozen and it is used to solve the inverse problem, where the pollutant source is calculated which corresponds to a set of concentrations in the monitoring wells. The approach has been applied for a real case which deals with the contamination of the Rhine aquifer by carbon tetrachloride (CCl_4) due to a tanker accident. The obtained results are compared with the solution obtained with a different approach retrieved from literature. The results show the suitability of ANNs-based methods for solving inverse non-linear problems.

Key words | ANNs inversion, groundwater pollution source identification, inverse problems

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INTRODUCTION

Groundwater is an important source for the production of drinking water. Consequently, the protection of groundwater resources quality appears of extreme importance for life support systems. Nevertheless, groundwater is exposed to man-made pollution that might prevent its use for drinking as well as for other domestic, industrial and agricultural purposes. When groundwater is polluted, the restoration of quality and removal of pollutants is a very slow, hence long, and sometimes practically impossible task.

In the field of groundwater resources contaminations, it should be highlighted that in some cases pollution might result from contaminations whose origins differ in time and place from the point where the contaminations were witnessed. To tackle such situations, it is necessary to develop specific techniques for identifying the behaviour of unknown contaminant sources from both spatial and temporal points

of view. Getting to know the initial conditions of pollution is consistent with the implementation of the European Union Directive 2004/35/EC. This Directive, based on the 'polluter-pays' principle, concerns the environmental liability in relation to the prevention and compensation of environmental damages. The application of the Directive 2004/35/EC requires the development of novel methodologies, such as that proposed in this work, for the identification of unknown pollution sources in contaminated aquifers. The problem of identifying an unknown pollution source in contaminated aquifers, based on known contaminant concentrations measurement, is part of the wide group of issues called inverse problems. During the last decade, several studies have been dedicated to the development of different methods for solving inverse problems, however works using artificial neural networks (ANNs) are less

popular. Among these latest, in Zio (1997), an ANN is trained to identify the value of the dispersion coefficient in a simple analytic contaminant transport model, in Fanni et al. (2002), an ANN is used to locate a pollutant source, **Q1** in Rajanayaka et al. (2002), a hybrid approach based on a combination of two types of ANN models is applied to esti- **Q2** mate hydrogeological parameters, in Scintu (2004), ANNs are trained to predict the coordinates of the pollutant source and the time the pollution occurred, in Singh & Datta (2007), ANN is used to identify unknown pollution sources with partially missing concentration observation data, in Foddis et al. (2013), ANNs are used for locating the source of a contamination event in time and space.

In this work, an ANN-based inverse problem solving method is presented for the identification of unknown contaminant sources and their characteristics. Compared to the state of art, the originality of the developed method lies in the approach used for solving the inverse problem, which consists of inverting the ANN model instead of solving the equations that described the physical system. In particular, different ANNs are trained to associate the pollution source features with the contaminant concentration measurements acquired in the monitoring wells. After this phase, the trained ANN is inverted by fixing the measurements at the wells and determining the corresponding pollutant source. This approach might be used for defining contaminant punctual sources to face groundwater quality problems. In particular, location and fluxes over time can be defined. The procedure described in this paper has been validated during a real case which tackles an actual contamination which occurred in the Rhine aquifer (France) (Vigouroux et al. 1983).

METHOD DESCRIPTION

Multi-layer perceptron network model

An ANN is a distributed, adaptive, generally non-linear learning machine, built from many different artificial neurons (AN) that interact with each other through weighted connections. The ANN is symbolized like a graph where patterns are represented in terms of numerical values attached to the ANs, the nodes of the graph. In particular, multi-layer perceptron (MLP) ANNs are logically arranged in two or more layers (Figure 1), and the numerical data conceptually flow forward from the input to the output layers. This is the structure of ANN used to model the system under study.

Such a neural model is trained by using a set of input–output pairs of patterns. These patterns are created by means of a flux and transport contaminant modelling software, which calculates both the source of contaminant and the set of measurements in the monitoring wells. These data are converted into patterns which represent the input and the output of the ANN, respectively. Thanks to the weighted links, the ANN associate an output pattern to any input pattern. The training procedure consists of adapting the weights so that the ANN performs a desired association between the input and output patterns of a given training set of examples. In this work, the desired output values (target) represent the measured values so that the trained ANN is able to associate a given source of pollutant with the corresponding measurements in the monitoring wells. The Levenberg–Marquardt algorithm has been used to train the ANN.
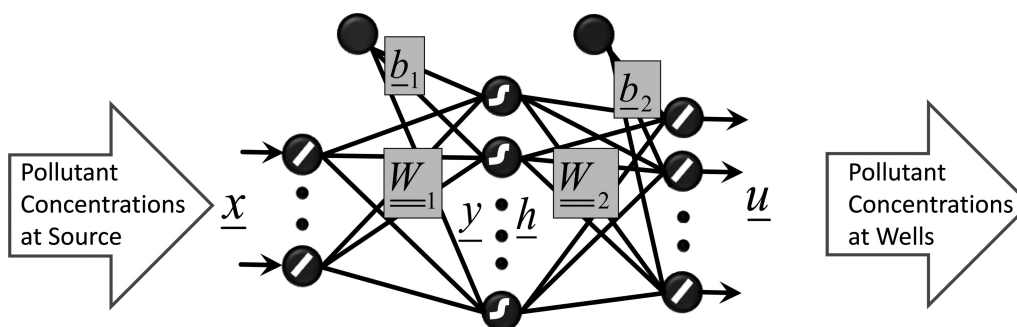

Figure 1 │ MLP structure.

The trained MLP realizes a relationship between input and output patterns described by the following algebraic equations system:

$$
\begin{array}{ll}
\text{Input layer} & \left\{ \begin{array}{l} \underline{\underline{W}}_1 \cdot \underline{x} + \underline{b}_1 = \underline{y} \\ \underline{h} = \sigma\,(y) \\ \underline{\underline{W}}_2 \cdot \underline{h} + \underline{b}_2 = \underline{u} \end{array} \right.
\end{array}
\tag{1}
$$

where (see Figure 1): $\underline{x}$ is the input of the network, $\underline{\underline{W}}_1$ is the weights matrix of the input layer, $\underline{b}_1$ is the bias vector of the input layer, $\underline{y}$ and $\underline{h}$ are the input and the output of the hidden layer, respectively, $\sigma(\cdot)$ is the hidden neurons logistic activation function, $\underline{u}$ is the output of the network, $\underline{\underline{W}}_2$ is the weights matrix of the output layer, and $\underline{b}_2$ is the bias vector of the output layer.

Solving the system Equations (1) for the input $\underline{x}$, given an assigned value of the output $\underline{u}$, means solving an inverse problem of determining the characteristics of the pollutant source. In the following two sections, the adopted methods to extract features from data and to solve the system (1) are described.

Feature extraction

Data which describe groundwater flow and contaminant transport are characterized by huge volume and strong redundancy of information, so that in general it is possible and also mandatory to pre-process the data by reducing the volume without a meaningful loss of information. This operation is referred to as feature extraction and it allows condensing the information contained in the original data into a strongly reduced dimension of data. Feature extraction techniques have been chosen in compliance with two important requirements. At first, a high rate of compression of the information has to be ensured in order to guarantee a significant correspondence between the back transformed data and the original values. Secondly, the procedure can be easily backward applied to allow the reconstruction of the initial data set with the least possible error.

The adopted feature extraction procedure consists of: (1) calculating the two-dimensional discrete fast Fourier transform (2D-FFT) for constructing both the input and output matrices; (2) reducing the matrices by a fixed threshold of energy content; (3) reducing the matrix size by means of the principal component analysis (PCA) method.

The training set is constituted by input/output matrices pairs, calculated by a simulator, where the former one reports the time evolution of pollutant source at four different depths, while the latter one is the time evolution of measured pollutant concentration in correspondence of monitoring wells. Therefore, the former matrix has four rows, while the latter one has as many rows as the number of monitoring wells. The number of columns is equal to the number of time samples. Both input and output matrices have to be converted into a pattern, and this pair of patterns will represent a specific case or example. By performing a suitable number of simulations, we finally obtain an input and an output matrix having the same number of columns, each one corresponding to an example.

The feature extraction procedure is first performed on the two matrices which, respectively, describe source and monitoring wells for each example. The two-dimensional Fourier Transform (Mersereau & Dudgeon 1975) of the two **Q3** matrices is calculated, switching to the frequency domain. Here, each element represents a spectral component of the signal, whose relevance in the signal is measured by its amplitude. Therefore we can define a preliminary threshold of amplitude in order to eliminate the components which are less relevant. The selected components have to be the same for all the examples, therefore all the components which overcome the threshold in at least one example will be kept. The same set of components will have to be kept for any future example. For each example, the selected components of both input and output matrices are rearranged in two column vectors, therefore at the end of this stage we have two matrices, input and output, respectively, for the entire training set, and for both, the number of columns is equal to the number of examples. The dimension of these two matrices can be further reduced, taking into account the redundancy of information due to the correlation existing among rows of the same matrix. The most efficient way to linearly eliminate such redundancy is the PCA (Henebry & Rieck 1996). This analysis consists of rotating the reference **Q4** axis in such a way that the variance of points distribution along some axis (principal) is maximum, so that the residual variance along the remaining axis can be neglected. The quantity of information which is lost with the neglected axis can be easily estimated, and in the real cases the PCA allows strong reduction of the dimension of patterns with

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very little loss of information. The dropped matrices are the input and output matrices that will be used to train the ANN. The training of the ANN is performed by using the Leven-berg–Marquardt algorithm. The quality of the training affects the reliability of the successive inversion of the ANN, therefore all the standard expedients have to be adopted to guarantee suitable approximation level and the generalization capability of the trained ANN.

Inversion procedure

Once the training of the MLP ANN is completed it is used to solve the inverse problem by exploiting the MLP inversion method described in Fanni & Montisci (2003) and Carcangiu **Q5** *et al.* (2007). It consists of determining the input pattern which corresponds to a given output pattern, which in our case means identifying the unknown pollution source on the basis of a set of measurements acquired in the monitoring wells. As in any identification problem, it is fundamental to guarantee the uniqueness of the solution. This implies that the two linear equations systems in (1) have to be determined or over-determined, then the number of input neurons has to be less or equal to the number of hidden neurons, and this one has to be less or equal to the number of output neurons. Secondly, for the same reason, the first and the second connections weights matrices $\underline{\underline{W}}_2$ and $\underline{\underline{W}}_1$ have to be full-rank. The parameter that can affect such features of the ANN is, generally speaking, the approximation degree of the representation of both input and output data. This means that by increasing the precision used to describe the input and output data, we could find out that the weights matrices are not full-rank. For this reason a trial and error procedure should be adopted in order to seek the best approximation level which guarantees the invertibility of the MLP ANN.

Inverting the MLP ANN means solving the system (1), for the unknowns $\underline{h}$, $\underline{y}$ and $\underline{x}$, while the output $\underline{u}$ is fixed. If the constraints on the rank of $\underline{\underline{W}}_1$ and $\underline{\underline{W}}_2$ are fulfilled, solving the system (1) is quite simple as the three unknown patterns $\underline{h}$, $\underline{y}$ and $\underline{x}$ can be determined one after the other univocally.

On the basis of the third equation in (1), starting from the output $\underline{u}$, the vector $\underline{h}$ can be determined. To guarantee the uniqueness of the solution, the number of rows (number of output neurons) must be higher than the number of

columns (number of hidden neurons). In this case, the equations system results are over-determined and the uniqueness is ensured by assuming the solution which corresponds to the minimum mean squared error. Such a solution can be found by solving the following modified equations system, which has a squared coefficients matrix.

$$\underline{\underline{W}}_2^T \underline{\underline{W}}_2 \underline{h} = \underline{\underline{W}}_2^T (\underline{u} - \underline{b}_2) \tag{2}$$

where the mark ^{T} represents the transposition operator. If the number of hidden neurons is equal to the number of output neurons, the weights matrix is squared, and the solution corresponding to the minimum sum squared error can be directly calculated as:

$$\underline{h} = \underline{\underline{W}}_2^{-1} \cdot (\underline{u} - \underline{b}_2) \tag{3}$$

The second equation in (1) states a bi-univocal relationship between $\underline{y}$ and $\underline{h}$, therefore the vector $\underline{y}$ is:

$$\underline{y} = \sigma^{-1}(\underline{h}) \tag{4}$$

Finally, provided that the matrix $\underline{\underline{W}}_1$ is full rank, the input pattern $\underline{x}$ can be calculated as:

$$\underline{x} = \left(\underline{\underline{W}}_1^T \cdot \underline{\underline{W}}_1\right)^{-1} \cdot \underline{\underline{W}}_1^T (\underline{y} - \underline{b}_1) \tag{5}$$

Also in this case, if the number of input neurons is equal to the number of hidden neurons, the system can be solved by directly inverting the matrix $\underline{\underline{W}}_1$.

The pattern $\underline{x}$ describes the unknown source of pollutant. To generate the time evolution of the source, the transforms that have been applied for the feature extraction have to be back applied to the obtained pattern $\underline{x}$.

APPLICATION OF THE METHOD TO A CASE STUDY

The performance of the proposed methodology has been evaluated by defining the behaviour in time and space of the Alsatian aquifer unknown pollution source.

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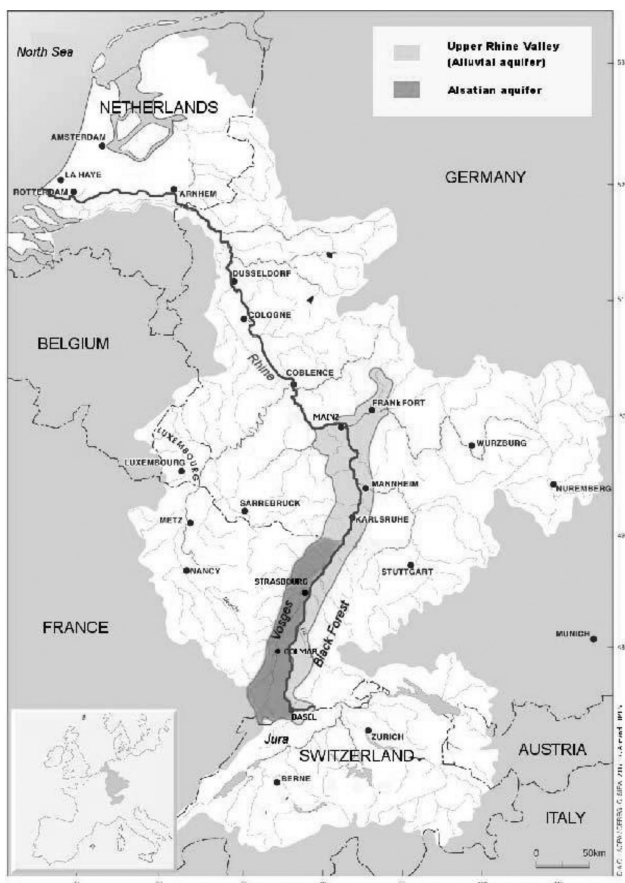## General description of the Upper Rhine Graben and Alsatian aquifer

The Alsatian aquifer is located in the southern part of the Upper Rhine valley. The Upper Rhine Graben is a segment of the European Cenozoic rift system that developed in the north-western forelands of the Alps. It extends over 300 km from Basel (Switzerland) in the south to Frankfurt (Germany) in the north, with an average width of approximately 40 km. As shown in Figure 2 it is flanked in the south by the Vosges and Black Forest (Schwarzwald) mountains, to the west and the east, respectively (Bertrand *et al.* 2006).

The Alsatian aquifer surface is over 3,000 km^2 and contains a volume of alluvial about 250 billion m^3. It represents one of the largest fresh water reserves in Europe. The groundwater reservoir contains about 50 billion m^3 of water, with an annual renewal of 1.3 billion m^3. This large aquifer is of vital importance since it supplies 75% of the drinking water requirements, 50% of the industrial water needs and 90% of the irrigation water needs in Alsace. The Alsatian part of the Rhine aquifer has a surface length of 160 km and a maximum width of 20 km (Hamond 1995; Bertrand *et al.* 2006).

Alsatian aquifer is an extensive alluvial aquifer with a layered structure composed of a random superposition of different alluviums (clay, sand fine to rough, gravels, coarse etc.). This permeable alluvial has a thickness of a few meters at the Vosgean edge, and 150–200 m in the centre of the Rhine plain (Hamond 1995).

The groundwater reservoir is part of a complex hydrosystem which includes frequent exchanges between the rivers and the aquifer which vary with the seasons and it is highly exposed to contamination from rivers (Stengera & Willingerb 1998).

History of the pollution by CCl4 in the aquifer

In 1970, a tanker truck containing carbon tetrachloride (CCl_4) property of a Dutch company overturned in the north of Benfeld (a small town located about 35 km south of Strasbourg). According to a note from Service Géologiqued'Alsace-Lorraine of 21st December 1971 about 4,000 litres of CCl_4 were spread in the area of the accident, infiltrating into the ground or disappearing by evaporation (Hamond 1995). In 1991, the analyses carried out by Bureaude Recherche Géologique et Minière showed abnormal quantities of CCl_4 in the supplies of drinking water (about 60 μg/L). These quantities exceeded the safe limits recommended by the World Health Organization (2 μg/L). This high level of CCl_4 concentration has caused a serious problem in the region by contaminating the main sources of drinking water in the Alsace Region (France) (Aswed 2008).

The exact amount of chemical infiltrated in the subsoil is unknown, as is the behaviour of the pollution source along the time. The assessment of such characteristics constitutes the main issue for the individuation and remediation of the accident effects and it is the objective of this study.

Q7 **Figure 2** | The Rhine Valley and the Alsatian aquifer.

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ANN to study the Alsatian aquifer

The behaviour of the Alsatian aquifer unknown contaminant source is determined based on the contaminant concentration measured in the monitoring wells dislocated in the study area.

In a first phase, the ANN was trained to solve the direct problem by using a set of patterns created by means of a flux and transport contaminant modelling software, where the patterns describe both the source of contaminant and the set of measurements in the monitoring wells,. The data set of pattern has been constructed through a coherent number of hydrogeological scenarios, based on a 3D model of the domain developed by Aswed (2008). The input patterns were made of the pollution source features in terms of the injection rates in the four hydrogeological layers. The output patterns were contaminant concentration observation data at 45 monitoring wells. Sources and monitoring wells are related by a bi-univocal relationship, meaning that any specific profile of monitoring wells corresponds to one specific contaminant source behaviour.

In a second phase, the trained network was inverted in order to identify the source of contaminant corresponding to a set of data measured in correspondence of monitoring wells.

Groundwater flow and contaminant transport numerical model

Q6 The CCl_4 Alsatian aquifer pollution has been the subject of several studies (Vigouroux *et al.* 1983; Beyou 1999; Aswed 2008). The 3D flux and contaminant transport numerical model used for constructing the patterns was calibrated using measured data of CCl_4 concentration that were collected over 12 years (1992–2004) and simulations were performed from 1970 to 2024.

The patterns have been simulated using the non-commercial numerical software Transport or RadioActiver Elements in the Subsurface (TRACES) that combines the mixed-hybrid finite elements and discontinuous finite elements to solve the hydrodynamic state and mass transfer in the porous media (Hoteit & Ackerer 2003). The contaminated zone is enclosed within a 3D domain of 6 km width, 20 km length, and about 110 m depth. This aquifer domain

is discretized using a 3D triangular prismatic grid with 25,388 nodes and 45,460 elements. According to the estimated geometry of the cross sections (the landfill site was divided into eight zones by soil type) the domain is discretized into 10 layers. The contaminant source is locate in the first four layers (the thicknesses of the layers are, respectively, 16 m, 4 m, 5 m, and 5 m from top to the bottom). The volume of contaminated aquifer is about 230–1,300 m^3. The surface of contaminant infiltration is about 7–37 m^2 (Aswed 2008).

Using TRACES, various states of pollution sources have been constructed adjusting the source characteristics in terms of injection rates over the vertical section. One hundred and four examples were constructed with the same duration of activity of about 31 years (11,520 days) and were located in the same positions in the domain (accident site in Benfeld). So for each of the 104 sources, a different evolution of the contaminant concentration in the time for the four layers in the numerical domain has been considered. The total time of simulation is about 54 years (20,000 days).

ANN pattern construction: elaboration and features extraction

The examples patterns obtained with TRACES consist of 208 matrices of contaminant concentrations:

- 104 matrices corresponding to the pollution sources features. These had dimensions [11,520 × 4]; 11,520 represents the time (days) and 4 represents the layers in the source location;
- 104 matrices corresponding to contaminant concentration in monitoring wells. These had dimensions of [4,000 × 45]; 4,000 represents the time (one value each 5 days) and 45 are the monitoring wells in the domain.

The 104 input matrices and the 104 output matrices have been reorganized to form two matrices which describe the whole training set: one for the input and one for the output.

The two groups of 104 input and output matrices were considered separately (Figure 3).

For the first step, the 2D-FFT has been calculated for each matrix, using Matlab. For each case we obtained two

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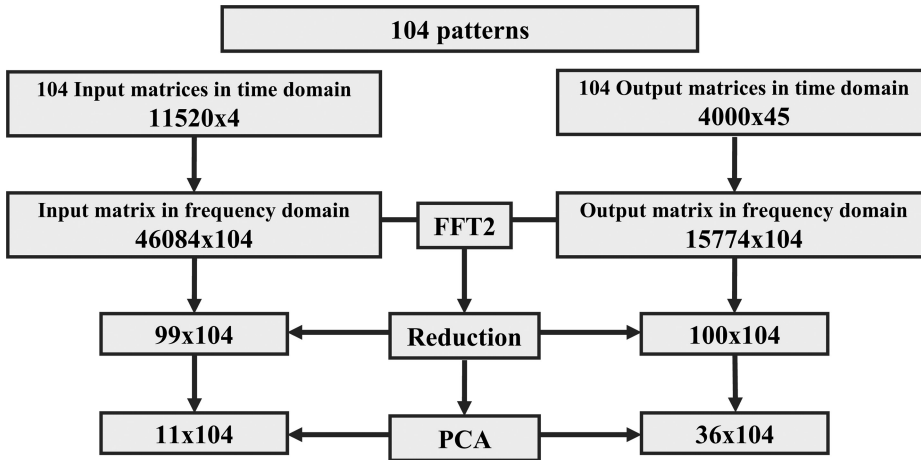
Figure 3 | Matrices reduction schema.

vectors: one for the input that has the dimension of 46,084 elements and one for the output that has the dimension of 15,774 elements. Vectors, corresponding to the different cases, have been joined to form a matrix where each column corresponds to one example, so at the end of this step the dimensions of the two matrices are $[46{,}084 \times 104]$ for the input matrix and $[15{,}774 \times 104]$ for the output matrix, respectively.

In the second step, from each of these matrices the negligible components have been removed. A component is considered negligible if is its amplitude does not overcome a prefixed threshold in any of the example. The value of the threshold is determined by seeking a crossover between the level of approximation of the acquired data and the dimension of the input and output matrices. For applying the PCA, the number of remaining values is pushed to have a number of rows less or equal to the number of the examples. As a result of this second step, the input matrix had the dimension $[99 \times 104]$ and the output matrix had the dimension $[100 \times 104]$.

In the third step, the PCA transform has been applied to further reduce the volume of data. The degree of compression that can be obtained depends on the fraction of information, evaluated as variance of the distribution that could be lost. Such a fraction has been determined by means of a trial and error procedure which provided a value of 2×10^{-3} for the input and 10^{-7} for the output. Consequently, the input matrix had dimension $[11 \times 104]$ and the output matrix dimension $[36 \times 104]$.

ANN training and inversion

One 11-11-36 MLP has been trained with the Levenberg–Marquardt algorithm by presenting the pairs of input (pollution sources scenarios) and target patterns (contaminant concentration for the 45 monitoring wells). Special attention has been paid to train the ANN in such a way that it is able to generalize the information contained in the training set. To this end, during the training phase, the error was evaluated even in the validation set, and the training was interrupted as soon as the error in the validation set started to rise (cross validation stop criterion). A third set (test set) of examples has been put aside in order to check on a totally independent set the generalization capability of the trained ANN. The 104 examples have been so subdivided: 74 in the training set, 19 in the validation set and 11 in the test set.

Once the training phase was completed, the trained network was inverted to identify the contaminant source. To this end, an output pattern was generated on the basis of the contaminant concentrations measured in the 45 monitoring wells and the procedure described above, under the section Inversion procedure, has been applied. In the present case, the number of hidden neurons is lower than the number of output neurons, so the system is over-determined. By applying Equation (2), the solution corresponding to the minimum sum squared error for the vector $\underline{h}$ was found. The second equation in (1) states a bi-univocal relation between $\underline{y}$ and $\underline{h}$, then the constant term of the third equation in (1)

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was determined. Finally, by solving this equation for $\underline{x}$, the solution of the inverse problem was determined.

RESULTS AND DISCUSSION

On the basis of the known contaminant concentration data in monitoring wells, the pollution sources injection rates in the cross section were identified. To evaluate the performance of the method, the re-constructed pollution source reported in Aswed (2008) was assumed as source of the simulation and the corresponding value of the wells data were calculated with the simulator. The four diagrams in Figure 4 show the comparison between the behaviour by

Aswed (blue) and the diagram obtained by inverting the ANN (green) for the four layers. The constant concentration at the early period could indicate that in the early stage of the process an accumulation in the surface layer continued to release contaminant at a constant rate for 10 years.

As can be noted, the calculated behaviour of the source satisfactorily fits the trend reported in Aswed (2008). The approximation is better in the deeper layers (layer 4) where the concentration is higher due to the deposition of the contaminant during the time. The ripple and the boundary peaks do not correspond to a real behaviour of the source, but they are a consequence of the cut of frequencies performed in the feature extraction procedure. Standard signal filtering procedures, like Kalman filters, could be
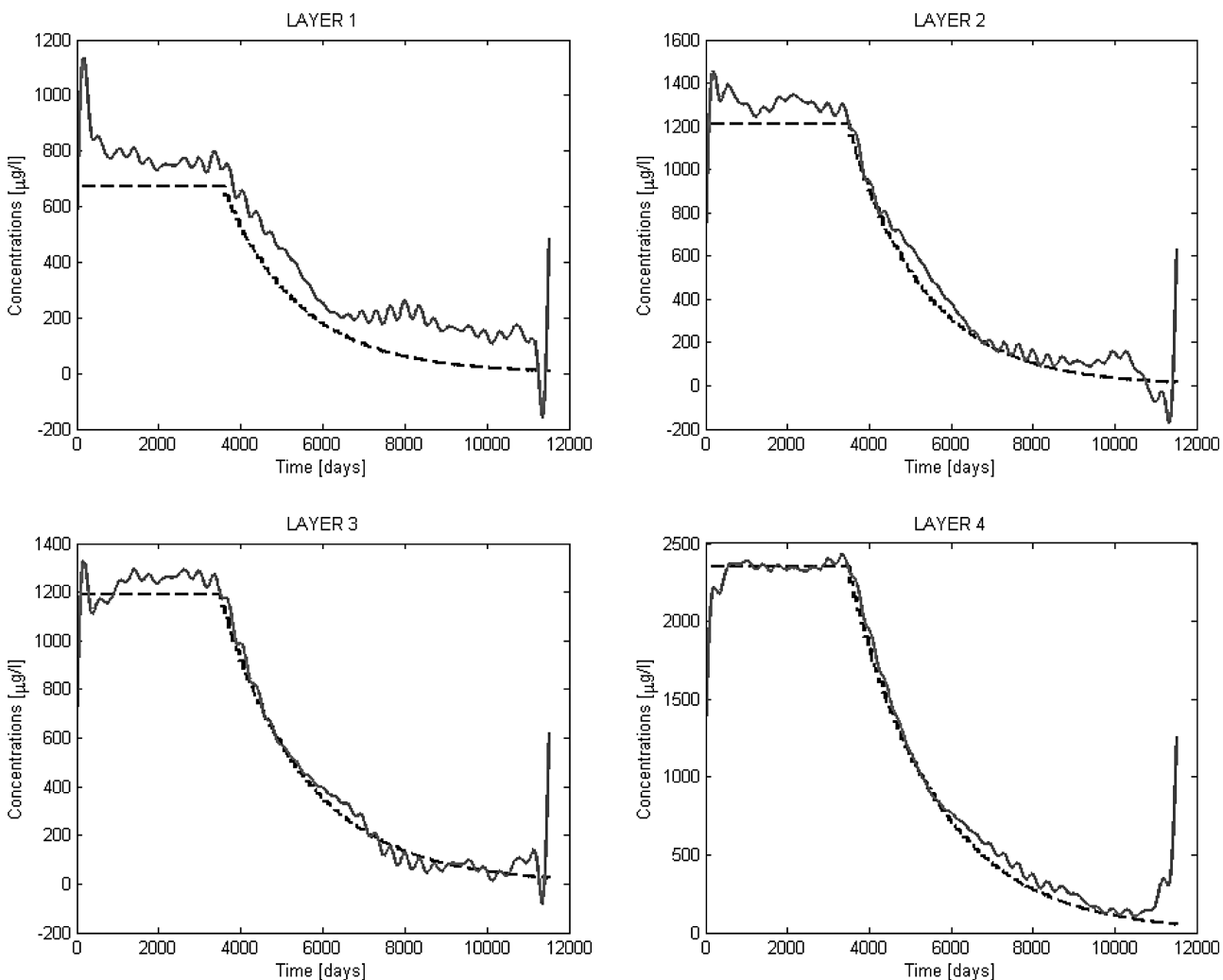
Q8 **Figure 4** | Simulated (dashed black) and the inverted (continued grey) source trend at layer 1 (16 m), layer 2 (20 m), layer 3 (25 m) and layer 4 (30 m), respectively (Foddis 2011).

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applied in order to improve the appearance of the signal, both filtering the ripple and automatically cutting the peaks by fixing a bound of the profile derivative. Nevertheless, this kind of post-processing of the results appears to be useless for an expert assessment.

CONCLUSION

This work mainly focuses on an ANN-based methodology for solving the inverse problem in aquifer pollution. A crucial aspect of the procedure is represented by the training of the ANN, which has to guarantee a satisfactory approximation of the physical system, but at the same time it has to fulfil several requirements in terms of number of neurons and rank of the connections weights matrices. The trained ANN is analytically inverted to solve the inverse problem, which consists of determining the profile of the pollutant source on the basis of a set of measures in monitoring wells. This procedure has been tested on a real case: an accident, which took place in the North-East of France in 1970, gave rise to a contamination by CCl_4 in the Alsatian aquifer. Various numerical model scenarios of the CCl_4 contamination in Alsatian aquifer have been generated to train the ANN to solve the direct problem of associating the pollution source characteristics with the measures acquired in the monitoring wells. The inverse problem is solved by fixing the output pattern and analytically calculating the corresponding input. Finally, the obtained results are compared with the pollution source behaviour re-constructed with another method retrieved from the literature. The obtained results show the suitability of the method, which can provide simple and reasonable solutions to various problems in hydrogeology, especially for this kind of non-linear problems.

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Author Queries

Q1   Rajanayaka *et al.* (2002) is not listed in reference list. Please add it to the list or delete the citation.

Q2   Scintu (2004) is not listed in reference list. Please add it to the list or delete the citation.

Q3   Mersereau (1975) has been changed to Mersereau & Dudgeon (1975) as per the reference list.

Q4   Henebry *et al.* (1996) has been changed to Henebry & Rieck (1996) as per the reference list.

Q5   Fanni *et al.* (2003) has been changed to Fanni & Montisci (2003) as per the reference list.

Q6   Beyou (1999) is not listed in reference list. Please add it to the list or delete the citation.

Q7   In supplied Figure 2 is not sufficient print quality. Please resupply as a high resolution file (300 dpi or above) with sharp lines and text.

Q8   In supplied Figure 4 is not sufficient print quality. Please resupply as a high resolution file (300 dpi or above) with sharp lines and text.