



Assessment of natural background levels in potentially contaminated coastal aquifers



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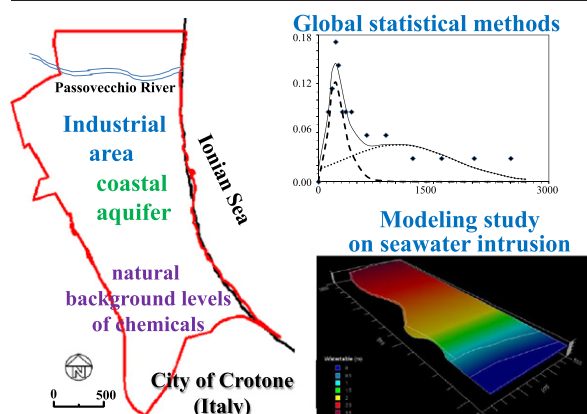
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HIGHLIGHTS

- Component Separation (CS) is applied to study natural background levels (NBL).
- Seawater intrusion could originate dispersive processes acting as external forcing.
- Mn status classified by CS is consistent with natural composition of the soil matrix.
- Sulfate status classified by CS is not consistent with seawater intrusion processes.
- CS interprets natural seawater intrusion as an anthropogenic source.

GRAPHICAL ABSTRACT



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ABSTRACT

The estimation of natural background levels (NBLs) of dissolved concentrations of target chemical species in subsurface reservoirs relies on a proper assessment of the effects of forcing terms driving flow and transport processes taking place within the system and whose dynamics drive background concentration values. We propose coupling methodologies based on (a) global statistical analyses and (b) numerical modeling of system dynamics to distinguish between the impacts of different types of external forcing components influencing background concentration values. We focus on the joint application of a statistical methodology based on Component Separation and experimental/numerical modeling studies of groundwater flow and transport for the NBL estimation of selected chemical species in potentially contaminated coastal aquifers. We consider a site which is located in Calabria, Italy, and constitutes a typical example of a Mediterranean coastal aquifer which has been subject to intense industrial development. Our study is keyed to the characterization of NBLs of manganese and sulfate and is geared to the proper identification of the importance of a natural external forcing (i.e., seawater intrusion) on NBL assessment. Results from the Component Separation statistical approach are complemented by numerical simulations of the advective–dispersive processes that could influence the distribution of chemical species (i.e., sulfate) within the system. Estimated NBLs for manganese are consistent with the geochemical composition of soil samples. While Component Separation ascribes the largest detected sulfate concentrations to anthropogenic sources, our numerical modeling analysis suggests that they are mainly related to the natural process of seawater intrusion. Our results indicate that the use of statistical methodologies in complex groundwater systems should be

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assisted by a detailed characterization of the dynamics of natural (and/or induced) processes to distinguish effective anthropogenic contamination from natural conditions and to define realistic environmental clean-up goals.

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1. Introduction

During the last decades, pressure loads acting on the soil–water environment have caused a remarkable increase in the number of locations where concentrations of one or multiple chemical species exceeding threshold values imposed by regulations are detected. Chemical compounds might also occur at naturally large concentrations for reasons which are unrelated to the effect of anthropogenic activities. The abundance of some species in groundwater might be linked, for example, to the petrographical composition of the aquifer (Hinsby and Condesso de Melo, 2006) or to the site-specific occurrence of organic matter (e.g., vegetal matter) which can enhance the release of chemical species to groundwater (Redman et al., 2002; Molinari et al., 2013). Because of factors such as (a) the strength of water–rock interactions, (b) solute residence times, (c) chemical and biological processes taking place in variably saturated host porous media, (d) interactions with other water bodies, (e) climate impacts on the subsurface environment, and (f) temporal fluctuations of flow field patterns, a given groundwater body is characterized by its own natural chemical signature. The latter may display significant space–time heterogeneity (Edmunds et al., 2003; Wendland et al., 2005; Panno et al., 2006; Edmunds and Shand, 2008; European Commission, 2009). In some cases, the application of technologies aimed at bringing the chemical status of an aquifer to a level of compliance with current regulations might not be feasible and all remediation strategies would prove to be ineffective and economically unaffordable because of unrealistic clean-up goals that might correspond to concentration levels which should be considered as the system natural background level (NBL). Procedures and criteria for a proper interpretation of the actual status of a groundwater body are outlined in the EU Water Framework Directive, WFD (2000/60/EC) and the GroundWater Daughter Directive, GWDD (2006/118/EC). These define the NBL as “the concentration of a substance or the value of an indicator, in a groundwater body, corresponding to no, or only very minor, anthropogenic alterations to undisturbed conditions”. A reliable estimation of the NBL of a target compound is required to (a) define threshold values which are consistent with specific natural features of water bodies; (b) discriminate between effective anthropogenic contamination and natural conditions; and (c) properly identify the chemical status of a groundwater body. Achieving these objectives is a critical aspect especially within regions where detected concentrations attain large values which can in turn lead to the classification of a site as potentially highly contaminated.

The most reliable approach for the estimation of the NBL of given inorganic compounds within a groundwater body is based on the analysis of samples collected at locations where water quality has not been altered by anthropogenic activities (Banks et al., 1995; Lahermo et al., 1995). The difficulty of identifying pristine portions of aquifers in populated areas has contributed to the development of other approaches. Estimating NBLs by means of global statistics of monitored data represents a relatively recent and useful tool for applications involving large scale groundwater bodies (Edmunds et al., 2003; Wendland et al., 2005; Panno et al., 2006; Edmunds and Shand, 2008; Walter, 2008). Klusman (1996) suggested grouping samples collected within target aquifer bodies into uncontaminated and contaminated sets through statistically based arguments. The idea that the empirical frequency distributions of available monitored concentration can be decomposed into a mixture composed by the contributions of natural and/or anthropogenic origin underpins the Component Separation (CS) method (Wendland et al., 2005). Application of this procedure is recommended by the EU research project BRIDGE (2007), Background cRiteria for the

Identification of Groundwater thresholds, in cases where the quality and quantity of data is sufficient for the consistent application of statistical analysis tools. Molinari et al. (2012) applied CS to assess (a) NBLs of chemical compounds in a series of large scale groundwater bodies in the Emilia Romagna Region, Italy and (b) the temporal dynamics of NBL variations. A key conclusion of this study is that, when possible, methodologies based on global statistics should be coupled with improved process understanding through robust experimental campaigns and/or detailed mathematical modeling studies aimed at characterizing the actual chemical–physical processes contributing to background concentration values within the investigated groundwater body. Following these conclusions, here we propose coupling methodologies based on global statistical analyses and conceptual/mathematical modeling of system dynamics to distinguish between the impact of different types of external forcing components influencing background concentration values.

Within the framework of brownfield remediation strategies, such as in the case of the site we consider in our demonstration study, the evaluation of the actual levels of anthropogenic contamination is key to define reliable clean up goals and avoid unnecessary and unrealistic actions under scenarios where a specific natural condition is demonstrated to take place. This issue could be addressed through the application of CS procedures. These are typically associated with an intrinsic inability to ground NBL estimates on the analysis of the key chemical–physical processes taking place in the system. In this context, a critical point is the validation of the estimated quantities through appropriate hydrogeochemical and hydrogeological studies. Coupling CS with process modeling is relevant in all coastal aquifers where the identification of the effects of tides and seawater intrusion can play a significant role for the definition of NBL (Benedini et al., 1983; Troisi et al., 1994; Bear et al., 1999; Werner and Simmons, 2009).

Coastal regions are among the most productive areas in the world. Their beauty and richness have made them popular settlement areas and tourist destinations, important business and industrial zones and transit points. The high density of population and excessive exploitation of natural resources within these areas puts enormous pressure on coastal ecosystems leading to biodiversity loss, habitats destruction, pollution, as well as conflicts between potential uses. Since the well-being of populations and the economic sustainability of a variety of businesses in coastal zones depend on the environmental status of these areas, it is essential to make use of appropriate management tools to enhance the protection of coastal resources and increase the efficiency of their uses (Al-Otaibi et al., 2006; European Commission, 2013).

As mentioned above, this paper illustrates issues related to the reliability of the results stemming from the application of the Component Separation method for NBL estimation in potentially contaminated coastal aquifers. We consider a site which is located in Calabria, Italy, and constitutes a typical example of a Mediterranean coastal aquifer which has been subject to intense industrial development. The site under consideration is part of an industrial area close to the city of Crotone and is described in details in Section 2.1. We focus our study on the characterization of NBLs of manganese (Mn) and sulfate, which are found to be associated with dissolved concentration levels which are significantly larger than threshold values defined by Italian regulations. We examine the way a natural external forcing term, as represented by the seawater intrusion process, acts on groundwater NBLs by complementing the NBL assessment procedure, based on CS, with experimental and modeling studies. To the best of our knowledge, no other studies documented the effect of such a natural external

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