



Delayed nitrate dispersion within a coastal aquifer provides constraints on land-use evolution and nitrate contamination in the past

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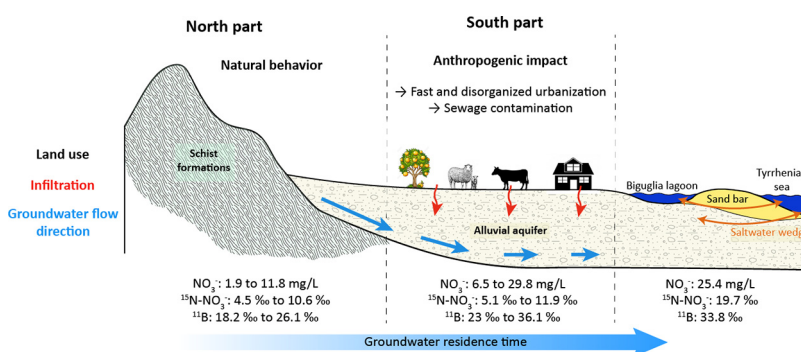
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HIGHLIGHTS

- High nitrate contents are measured in an alluvial aquifer supplying a coastal lagoon.
- $\delta^{15}\text{N-NO}_3^-$ and $\delta^{11}\text{B}$ signatures identify sewage as the main nitrate source today.
- Increase in NO_3^- with groundwater age displays the archiving capacity of groundwater.
- Highest NO_3^- concentrations can be linked to agricultural activities in the past.
- Legacy pollution that accumulated in the aquifer is a threat for the lagoon.

GRAPHICAL ABSTRACT



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ABSTRACT

Identifying sources of anthropogenic pollution, and assessing the fate and residence time of pollutants in aquifers is important for the management of groundwater resources, and the ecological health of groundwater dependent ecosystems. This study investigates anthropogenic contamination in the shallow alluvial aquifer of the Marana-Casinca, hydraulically connected to the Biguglia lagoon (Corsica, France). A multi-tracer approach, combining geochemical and environmental isotopic data ($\delta^{18}\text{O-H}_2\text{O}$, $\delta^2\text{H-H}_2\text{O}$, ^3H , $\delta^{15}\text{N-NO}_3^-$, $\delta^{18}\text{O-NO}_3^-$, $\delta^{11}\text{B}$), and groundwater residence-time tracers (^3H and CFCs) was carried out in 2016, and integrated with a study of land use evolution in the catchment during the last century. Groundwater NO_3^- concentrations, ranged between 2 mg/L and up to 30 mg/L, displaying the degradation of groundwater quality induced by anthropogenic activities (agricultural activities). Comparatively high $\delta^{15}\text{N-NO}_3^-$ values (up to 19.7‰) in combination with $\delta^{11}\text{B}$ values that were significantly lower (between 23‰ and 26‰) than the seawater background are indicative of sewage contamination. The ongoing deterioration of groundwater quality can be attributed to the uncontrolled urbanization development all over the alluvial plain, with numerous sewage leakages from the sanitation network and private sewage systems. Integration of contaminant and water-residence time data revealed a progressive accumulation of pollutants with time in the groundwater, particularly in areas with major anthropogenic pressure and slow dynamic groundwater flow. Our approach provides time-dependent insight into nitrogen pollution in the studied

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aquifer over the past decades, revealing a systematic change in the dominant NO_3^- source, from agricultural to sewage contamination. Yet, today's low groundwater quality is to large parts due to legacy pollution from land-use practices several decades ago, underlining the poor self-remediating capacity of this hydrosystem. Our results can be taken as warning that groundwater pollution that happened in the recent past, or today, may have dire impacts on the quality of groundwater-dependent ecosystems in the future.

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1. Introduction

Coastal lagoons provide a large spectrum of socio-economic benefits for humans, such as food provision, recreation and ecotourism, water regulation, coastal protection or pollution bioremediation (Farber and Costanza, 1987; Spurgeon, 1999; Perez-Ruzafa and Marcos, 2012). However, due to their location, these unique ecosystems rely on the fragile equilibrium between freshwater and marine water inflows, and are particularly susceptible to natural hazards (climate change, sea-level rise, coastal erosion) (Ketabchi et al., 2016; Klassen and Allen, 2017). This vulnerability is accentuated by the increasing anthropogenic pressure exerted on them (Mahlknecht et al., 2017). Indeed, a large part of the world's population is concentrated in coastal territories, causing an ever-growing urbanization (Barragán and de Andrés, 2015) and exposure to human activities, like agriculture, industry or mass tourism. Such activities are often associated with pollution (sewage leakages, fertilizers) that directly impact the quality of surface and ground water (Wang et al., 2016; Senthilkumar et al., 2017; Shuler et al., 2017). Groundwater plays an important role in the water supply of coastal wetlands (Cartwright and Gilfedder, 2015; Krogulec, 2016; Paz et al., 2017) and could be an important vector of anthropogenic pollution. Hence, groundwater degradation by nitrate contamination, for example, can contribute to the eutrophication of groundwater dependent ecosystems. A better understanding of pollution sources in aquifers, and the hydraulic connectivity between groundwater and groundwater dependant ecosystems is essential to ensure the protection and conservation of these ecosystems.

The isotopic signatures of dissolved nitrogen species ($\delta^{15}\text{N}-\text{NO}_3^-$ and $\delta^{18}\text{O}-\text{NO}_3^-$) can provide valuable constraints on the sources and fate of fixed (i.e., bioavailable) nitrogen (N) in groundwater. Past studies have shown that common pollutant sources of nitrate in groundwater include soil-N, manure, synthetic fertilizer and sewage effluents (Widory et al., 2013; Gooddy et al., 2014; Zhang et al., 2014; Grimmeisen et al., 2017; Vystavna et al., 2017a). To some extent, these nitrate sources have distinct isotopic signatures, which can be used diagnostically to semi-quantify the relative importance of single N sources (Aravena et al., 1993; Yakovlev et al., 2015; Grimmeisen et al., 2017; Vystavna et al., 2017b). However, N does not necessarily behave conservatively, and processes such as denitrification lead to isotope fractionation altering the primary isotopic signatures (Fukada et al., 2004; McMahon and Böhlke, 2006). At the same time, such processes can act to alleviate fixed nitrate contamination by transforming it into harmless forms of N (i.e., N_2 in the case of denitrification and anaerobic ammonium oxidation) (Jetten et al., 2001; Yenigün and Demirel, 2013).

Indirect tracers of NO_3^- sources that co-migrate with dissolved nitrate, such as boron (B), may be employed to gain additional information on groundwater pollution. In contrast to N isotopes, the B isotopic composition ($\delta^{11}\text{B}$) is not affected by biogeochemical transformation processes in aquifers. It can therefore be helpful to discriminate the NO_3^- sources where denitrification may alter primary source signatures. On the other hand, the B isotopic composition can be affected by adsorption-desorption processes on clays, organic matter or aluminium and iron oxide surfaces (Palmer et al., 1987; Bassett, 1990; Yingkai and Lan, 2001; Lemarchand et al., 2002). The great value of B is that its isotopic signature is characteristic for human and animal waste (Vengosh et al., 1994; Widory et al., 2005). Hence B isotope ratios have successfully been used to trace wastewater leakage/injections to the

groundwater in various studies (Xue et al., 2009; Cary et al., 2013; Guinoiseau et al., 2018), or, in combination with NO_3^- isotopic signatures, to identify sewage contamination in groundwater, and to discriminate animal manure and fertilizer sources (Widory et al., 2004, 2005; Seiler, 2005; Xue et al., 2009).

Although correlated isotopic signatures of N and B permit the semi-quantitative identification of anthropogenic sources of pollution, they do not provide information on the timing and temporal evolution of the contamination. Yet, NO_3^- infiltration can be a very lasting process, which depends on the watershed and aquifer properties. The time-lag between the emission of a pollutant and the contamination actually determined in groundwater can vary greatly (Han et al., 2015; Vero et al., 2017). Thus, detrimental impacts of past land use often leave footprints that may only be discovered several years or decades later. Identifying current NO_3^- sources is not sufficient for understanding actual groundwater NO_3^- concentrations. It is also necessary to evaluate the contribution of legacy NO_3^- related to past activities in the catchment. Integrated information on past land-use, the groundwater age (or time since infiltration), and the concentration of nitrate allows insight into the evolution of pollutant sources in the past, which is crucial for assessing a groundwater system's resilience towards contamination (Provitolo and Reghezza-Zitt, 2015). Groundwater residence time is an essential parameter for predicting future pollutant levels, necessary to establish a sustainable water resource management (Re et al., 2014; Han et al., 2015). To this end, tracers like ^3H , CFCs and SF_6 are commonly used to gain constraints on the groundwater age (Carreira et al., 2013; Kralik et al., 2014; Prada et al., 2015; Caschetto et al., 2016). Because of their long residence time, porous aquifers are particularly prone to the detrimental impacts of anthropogenic contamination. Indeed, continuous anthropogenic pollution that infiltrated an aquifer for several decades is likely to have accumulated to high levels in the groundwater, and the measured pollution today is the result of current and past anthropogenic activities.

Our goal was to combine geochemical (major ions), environmental isotope ($\delta^{18}\text{O}-\text{NO}_3^-$, $\delta^2\text{H}-\text{H}_2\text{O}$, ^3H , $\delta^{15}\text{N}-\text{NO}_3^-$, $\delta^{18}\text{O}-\text{NO}_3^-$, $\delta^{11}\text{B}$), groundwater residence time tracer (^3H and CFCs) data, as well as information on land use evolution for the last century, in order to 1.) identify pollution sources in an alluvial plain aquifer in Northern Corsica (France), and 2.) assess the temporal relationships (i.e., time lag) between the actual pollution and the groundwater contamination as observed today. Understanding the dynamics and pathways of pollution within an aquifer is essential for predicting the impact on coastal groundwater dependent ecosystems, as well as their vulnerability to land-use change. The studied site in particular, and groundwater dependent ecosystems in general, are essential components of the watershed, which are often susceptible to anthropogenic pollution. They represent the terminal sink for all sorts of dissolved and particulate matter, including harmful substances that may have been released to the environment long ago in the past.

2. Study area

2.1. Geography, geomorphology and land use

The Biguglia lagoon is a shallow (maximum depth of 1.8 m), brackish coastal lagoon located in the northern part of the island of Corsica

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