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Examining nitrogen dynamics in the unsaturated zone under an inactive cesspit using chemical tracers and environmental isotopes

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ABSTRACT

This study evaluates the dynamics of nitrogen compounds generated by infiltration of wastewater from an inactive cesspit in the unconfined and sedimentary Adamantina Aquifer in Urânia, Brazil. A monitoring station, consisting of an 11.2 m well (1.8 m in diameter) with an array of 12 tensiometers and 12 suction lysimeters, was installed to monitor the shallow unsaturated zone from 0.5 to 9 m depth. A monitoring well was also installed below the water level to monitor the shallow aquifer. High amounts of ammonium (up to 96 mg/L NH_4^+-N) and nitrate (up to 458 mg/L NO_3^--N) were observed in the unsaturated zone porewater which is comparable to active septic systems effluents. The distribution of NO_3^-, Cl^- and Na^+, typical constituents of sewage effluents, varied seasonally and spatially, which is correlated with changes in infiltration rates between the wet and dry seasons and with hydraulic conductivity variations in interlayered sandy and clayey sediments. A detailed monitoring of porewater geochemistry demonstrated the occurrence of several important reactions affecting nitrogen dynamics in the unsaturated zone: i) oxidation of organic matter, ii) ammonification, iii) nitrification, iv) methanogenesis, v) denitrification and likely, vi) sulfate reduction. The changes in nitrogen compound distribution and $\delta^{15}N_{NO3}$ and $\delta^{18}O_{NO3}$ values in porewater, in association with the N_2O concentration and $\delta^{15}N_{N2O}$ and $\delta^{18}O_{N2O}$ signatures in gas samples, indicate the occurrence of nitrification and denitrification, suggesting the coexistence of reducing and oxidizing microsites in the unsaturated zone. This study indicated that cesspits can generate a significant amount of nitrate even a few years after being inactivated which can represent a potential long-term source of nitrate to groundwater in highly populated areas.

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1. Introduction

Nitrate is the most common pollutant found in groundwater worldwide. Concentrations of NO_3^--N above 10 mg/L may cause methemoglobinemia and some types of cancer (WHO, 2011; USEPA, 2012). A number of studies have focused on evaluating the extent and fate of nitrate contamination in the short- and long-term in the unsaturated zone (Close, 2010; Rudolph et al., 2010). Recent efforts have focused on evaluating the impact of the implementation of Best Management Practices (BMPs), used in agriculture to decrease nitrate contamination to groundwater. These studies have demonstrated that agricultural activities have often created elevated concentrations of nitrate in the unsaturated zone. This large mass of nitrate in the unsaturated zone has long-term implications, even under the most effective BMPs, for achieving the reduction of nitrate concentration in groundwater (Rudolph et al., 2010).

Another important source of nitrate contamination to groundwater are on-site sanitation systems, such as cesspits and septic systems. Cesspits are open shallow holes where the domestic sewage is dumped without any previous treatment. These on-site sanitation systems are frequently used in poor and developing countries in both rural and urban areas, where public sewage systems have not been installed (Foster and Hirata, 1988; Foster et al., 2002; Hirata et al., 2007). In several studies conducted near the study site in Urânia, Brazil, nitrate concentrations above the drinking water standard were found in deep and shallow wells (Cagnon and Hirata, 2004; Gutierrez and Hirata, 2004), as well as in

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the unsaturated zone (Bertolo et al., 2006; Varnier et al., 2007). Until the public sewerage system was constructed in 1970, residents used cesspits for on-site sanitation. Cagnon and Hirata (2004) estimated that there was a high concentration of cesspits (1250 cesspits/km^2) in Urânia, which had a population density of 5000 inhabitants/km^2.

Over the last 50 years, many urban areas in Brazil were developed without prior construction of sewage mains. Most cities have installed sewage systems, but nitrogen contamination from decommissioned cesspits constitutes a long-term source of nitrate in the unsaturated zone, which will continue to contaminate groundwater in the saturated zone over years. Many authors have studied the fate of nitrogen compounds and other nutrients emanating from on-site sanitation systems (Robertson et al., 1991; Harman et al., 1996; Ptacek, 1998; Cagnon and Hirata, 2004; Varnier and Hirata, 2005; Varnier et al., 2007), but only at on-site sanitation systems that were still actively being utilized.

The objective of this study is the evaluation of the transport and fate of nitrogen in the unsaturated zone under a decommissioned cesspit of the Adamantina Aquifer, using chemical and isotopic tools.

2. Research approach

The main reactions controlling dissolved nitrogen species and DOC transformations in the unsaturated zone under the influence of sewage effluents are: i) organic carbon oxidation (reaction 1); ii) ammonification (reaction 2); iii) nitrification (reactions 3 and 4); iv) carbonate dissolution (reaction 5); vi) denitrification (reaction 6); vii) sulfate reduction (reaction 7) and viii) methanogenesis (reaction 8).

$CH_2O + O_2 \rightarrow CO_2 + H_2O$ (reaction 1)

$N_{org} \rightarrow NH_4^+$ (reaction 2)

$NH_4^+ + 1.5O_2 \rightarrow NO_2^- + H_2O + 2H^+$ (reaction 3)

$NO_2^- + 0.5O_2 \rightarrow NO_3^-$ (reaction 4)

$H^+ + CaCO_3 \rightarrow HCO_3^- + Ca^{2+}$ (reaction 5)

$NO_3^- + 5CH_2O + 4H^+ \rightarrow 2N_2 + 5CO_2 + 7H_2O$ (reaction 6)

$SO_4^{-2} + 2CH_2O \rightarrow H_2S + 2HCO_3^-$ (reaction 7)

$CH_2O + H_2O \rightarrow CH_4 + HCO_3^- + H^+$ (reaction 8)

Then, the study was designed to collect data that provide evidence to evaluate the occurrence of reactions 1 to 8 in the water that have been in contact with the inactive cesspit sediments. These include O_2 involved in reactions 1 and 3; DOC involved in reactions 1, 6, 7 and 8; NH_4^+ and NO_3^- involved in reactions 2, 3, 4 and 6. $\delta^{15}N$ and $\delta^{18}O$ measurement in NO_3^- and porewater N_2O gas analyses were used to evaluate to role of nitrification and denitrification processes in nitrogen transformation and attenuation in the unsaturated zone (Aravena and Robertson, 1998; Bol et al., 2003). ^{13}C analyses in CO_2 were performed to evaluate carbon cycling and redox conditions in the unsaturated zone (Lloyd et al., 1987; Aravena et al., 1992; Stevens et al., 1997; Brady and Weil, 2002; Gooddy et al., 2002).

3. Study area

The study was conducted in Urânia, in northwestern São Paulo state, a community with a large number of decommissioned cesspits (Fig. 1). Public water supply is extracted from the Guarani Aquifer System, a confined unit approximately 900 m deep in this area. Overlying the Guarani Aquifer System, the Serra Geral Aquifer System is a confined, fractured, anisotropic, heterogeneous aquifer constituted by basalts and intrusive rocks with a maximum thickness of 150 m.

The uppermost aquifer is the Bauru Aquifer System, locally comprised by the Adamantina Aquifer. It is an unconfined, sedimentary aquifer that is generally assumed to be homogeneous, horizontally isotropic and has a thickness between 60 and 160 m (Almodovar, 2000; Cagnon and Hirata, 2004; Varnier et al., 2005).

The unsaturated zone of the Adamantina Aquifer is composed of heterogeneous, fine to medium-grained sandstone, with some clay content (Bertolo et al., 2006; Varnier et al., 2007). Below 7 m depth, the clay content decreases and the sand percentage increases. The hydraulic conductivity (K), measured in non-deformed sandstone samples, varies from 3.1×10^{-6} to 1.4×10^{-5} m/s (Varnier et al., 2007).

4. Materials and methods

4.1. Monitoring station

In March 2003, a monitoring station was installed 1 m northeast from an inactive cesspit (Fig. 2). The cesspit was unlined, 3 m in depth, and covered with a concrete lid. It received effluents between 1996 and 2002, from the kitchen and bathrooms from a household with four adults.

The monitoring station consists of a hand-excavated well, 11.2 m deep and 1.8 m in diameter, in which 28 concrete rings (1.6 m in outer diameter) were inserted to stabilize the sides of the excavation. Twelve suction lysimeters and 12 tensiometers were installed in one-meter intervals, for sampling porewater and measuring water potentials in the unsaturated zone (Fig. 2). The suction lysimeters were installed oriented toward the southwest side of the monitoring station closest to the location of the inactive cesspit. At

Fig. 1. Location of the study area.

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