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How can the role of leachate on nitrate concentration and groundwater quality be clarified? An approach for landfills in operation (Southern Italy)

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ABSTRACT

Where the unique natural water resource is groundwater, the attention and the susceptibility of local communities and authorities to groundwater quality degradation risks can be so high to determine relevant problems to waste management, especially for landfills in operation or to be realised. A multi-methodological approach was suggested with the purpose to clarify the role of landfill leakage on groundwater quality degradation.

The selected study area (SSA) hosts some landfills in a narrow portion of a wide and deep coastal karstic aquifer, for these characteristics to be considered a case of high hydrogeological complexity and vulnerability. News concerning nitrate and secondly iron groundwater concentration anomalously high caused concern in the population and strong local opposition to landfills.

The multi-methodological approach includes: the hydrogeological site characterization; the chemical study and the multi-isotope characterization of groundwater and leachate; the land use analysis and the estimation of nitrogen contributions deriving from fertilizers; the mineralogical study of groundwater suspended particles to define the role of natural soil substances.

The hydrogeological site characterisation highlighted the local peculiarities of the aquifer. The chemical study was used to define geochemical features, groundwater and leachate characteristics and their macroscopic mixing.

The environmental isotopes of hydrogen, carbon, nitrogen, and oxygen were used to investigate the groundwater origin, the most relevant geochemical reactions, the existence of groundwater-leachate mixing, and the sources of anthropogenic NO_3^-. The land use analysis highlighted quantity and type of used fertilizers permitting to compare these with groundwater in terms of isotopic signature. The mineralogical study demonstrated the role of suspend natural particles due the presence of *terre rosse* (red or residual soils) in groundwater.

The approach confirmed that there are not the groundwater quality degradation effects of landfills, contributing to reassure population and institutions, simplifying the waste management.

1. Introduction

Municipal or urban landfill leachate consist generally of many different organic and inorganic compounds that are either dissolved or suspended in the wastewater (Mendoza et al., 2017). According to Mor et al. (2006), areas close to landfills have a greater possibility of groundwater contamination because of the potential pollution source of leachate which also contain heavy metals, including iron, and nitrogenous solutes, including nitrate (Mendoza et al., 2017). Anomalous high groundwater concentrations of these substances close to landfills can alarm population, highly increasing opposition to landfills in operation and/or to landfill extensions.

Nitrogen-containing compounds are found in the organic substance, where nitrogen is present, for example, in the amino acids

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that make up the proteins. In contrast, lithospheric minerals containing nitrogen are generally very soluble, and therefore rare (Appelo and Postma, 1996). Nitrogen is an important component of natural waters, where it is present dissolved in different valences such as +5 in NO_3^-, +3 in NO_2^-, 0 in N_2 and −3 in NH_4^+ (Appelo and Postma, 1996). The concentration of dissolved molecular nitrogen in water, in equilibrium at 25 °C with the atmosphere, is 5×10^{-4} M (14 mg/L). The natural groundwater nitrate concentration is generally low; concentrations greater than 1 mg/L can be generally due to anthropogenic activities (Dubrovsky et al., 2010).

High-nitrate concentrations in groundwater are a worldwide problem (Burow et al., 2010; Strebel et al., 1989; Thorburn et al., 2003).

Ingestion of high nitrate rate can cause many problems to humans; the World Health Organization (WHO) states that the NO_3^- concentration for drinking water should be less than 50 mg/L (World Health Organization, 2011).

Potential sources of nitrate for groundwater include mineral fertilizers, septic waste, animal manure, and landfill leachate (Heaton, 1986).

As all crops are not able to use all the added mineral fertilizer, the use of nitrogenous fertilizer creates risk of increasing percolation and leaching of nitrates to groundwater, which are very relevant in the case, almost frequent, of over fertilisation (Bijay-Singh et al., 1995). Many studies have indicated a high correlation between agriculture and nitrate concentrations in groundwater (Heaton, 1986; Sheikhy Narany et al., 2017).

Iron is a metal that can be considered almost common in natural soils and rocks. Groundwater can transport iron as dissolved or suspended particles. The extent to which iron dissolves in groundwater depends on dissolved oxygen and pH of water and not by natural or anthropogenic iron sources (Appelo and Postma, 1996); the suspended particles can be due the natural presence of residual soils (Moresi and Mongelli, 1988).

A low concentration of iron can be considered positive, as it is an essential nutrient for human, if natural (World Health Organization, 2011).

The aim of this paper is to clarify the existence or the absence of groundwater pollution by landfill leachate mixing and, in the latter case, to explain the source of groundwater quality degradation mainly focusing on the role of nitrate and secondly of iron. For this purpose, a multi-methodological approach was proposed. A selected study area (SSA), was considered with the purpose to testify the existence or excluding leachate effects on groundwater quality. The SSA is a narrow portion of a wide and deep coastal karstic aquifer, for these characteristics to be considered a case of high complexity and vulnerability. Five landfills have operated from 1975, one after the other, using increasing safety and technological devices to reduce risks due to leachate leakages. Mainly nitrate and secondly iron groundwater concentration were considered anomalously fearing the population, worried about the potential effects of landfills. For this reason, some landfills were seized by public authorities, hypothesizing a huge role of these in the groundwater quality degradation.

The approach considers each potential source of nitrate, considering the type of local land use: mineral fertilizers, septic waste, animal manure and landfill leachate.

The approach includes the use of nitrogen, oxygen and tritium isotopes as tracers for evaluating contamination of the landfill.

The use of $\delta^{15}N$ is based on the distinct isotopic composition that characterizes nitrate of different origin (Aravena et al., 1993). For the commercial fertilizers, typical $\delta^{15}N$ values range from −2‰ to +4‰, for soil organic nitrogen nitrate the values range from +3‰ to +8‰ and +10‰ to +20‰ for human and animal waste nitrate (Freyer and Aly, 1974; Kreitler, 1975; Gormly and Spalding, 1979; Heaton et al., 2012). In the case of ^{18}O, synthetic fertilizers are characterized by enriched ^{18}O values (+18‰ to +22‰), since the source of oxygen for these chemicals is atmospheric oxygen whose value is ^{18}O = +23.5‰ (Amberger and Schmidt, 1987). This method was successfully used for the resolution of different groundwater nitrate sources in Australia, identifying a leachate derived source down gradient from landfill (Moreau and Minard, 2014).

The method based on the determination of the stable isotopes ^{2}H and ^{18}O, useful to support the hydrogeological conceptualisation (Mook, 2000), is able to highlight leachate contamination of groundwater, i.e. due the enrichment of ^{2}H due to extensive methanogenesis (Hackley et al., 1996).

Tritium (^{3}H) is a radioactive isotope of hydrogen, occurring in very low quantity in the natural waters (Tazioli, 2011). Tritium levels in municipal solid waste landfill leachate can be several orders of magnitude greater than background groundwater levels due to the presence in the waste of some items containing high levels of tritium, such as luminescence paints and watches (Robinson and Gronow, 1996; Hackley et al., 1996; Tazioli, 2011) as verified in municipal landfills throughout the world (Fritz et al., 1994; Robinson and Gronow, 1996; Hughes et al., 2011; Raco et al., 2013). Because of this, tritium may be a sensitive indicator of leachate impacts on groundwater samples (Kerfoot et al., 2003).

Other isotope used for prevention and control of groundwater pollution is the $\delta^{13}C$ of total dissolved carbon. In general, the range of $\delta^{13}C_{DIC}$ in the groundwater may be very wide, the most common values range from −25‰ to −10‰, (Boutton, 1991).

Under reduction conditions (similar to those present in landfill environments), $\delta^{13}C$ is highly enriched ($\delta^{13}C$ > +10‰) (Wimmer et al., 2013). The positive value of $\delta^{13}C$ is due to the production of methane under anaerobic conditions (Wimmer et al., 2013). Focusing on leachate-impacted groundwater, $\delta^{13}C$ of major carbon compounds highlights attenuation due to biogeochemical reaction from dilution as a result of advective dispersion (Mohammadzadeh and Clark, 2011).

2. Selected study area

The SSA is located in the municipality of Conversano, close to Bari, the main town in the Apulia region (Fig. 1). The SSA elevation ranges from 100 to 175 m a.s.l., with a mean land surface slope of 1.3% from west to east (seaward). The SSA is a rural area. Agriculture is the main activity, namely mainly due to vineyards cultivation, orchards, olive trees, and arable crops.

The study area includes 5 solid urban waste landfills, distinguished from I to V in chronological order of realisation, which are located roughly at 5 km northwest from the town of Conversano (Fig. 1b and c).

Landfill I collected wastes from 1975 to 1982, for about a year in 1989 and for about 100 days in 1996, less than 10 years as total. This landfill was not sealed by capping. The leachate tapping system was not realized.

The Landfill II operated from 1993 and was closed in 1996; the landfill post-mortem was terminated. It was equipped with a definitive capping realized with HDPE geomembrane and other typical urban waste tools, to nullify leachate leakage risks. The landfill II top now hosts a photovoltaic system. The leachate tapping system is now completely unuseful and unusable.

Roughly 1,465,000 m^3 of wastes were disposed from 1996 to 2011 in the landfill III. At the present, the top of the III has been realised by a capping of HDPE geomembrane.

The Progetto Ambiente Bacino BA/5 landfill system is made up of two sectors or tanks, landfills IV and V (Fig. 1c). Roughly