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Nanosized iron based permeable reactive barriers for nitrate removal – Systematic review

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ABSTRACT

It is unquestionable that an effective decision concerning the usage of a certain environmental clean-up technology should be conveniently supported. Significant amount of scientific work focussing on the reduction of nitrate concentration in drinking water by both metallic iron and nanomaterials and their usage in permeable reactive barriers has been worldwide published over the last two decades.

This work aims to present in a systematic review of the most relevant research done on the removal of nitrate from groundwater using nanosized iron based permeable reactive barriers.

The research was based on scientific papers published between 2004 and June 2014. It was performed using 16 combinations of keywords in 34 databases, according to PRISMA statement guidelines. Independent reviewers validated the selection criteria.

From the 4161 records filtered, 45 met the selection criteria and were selected to be included in this review.

This study's outcomes show that the permeable reactive barriers are, indeed, a suitable technology for denitrification and with good performance record but the long-term impact of the use of nanosized zero valent iron in this remediation process, in both on the environment and on the human health, is far to be conveniently known. As a consequence, further work is required on this matter, so that nanosized iron based permeable reactive barriers for the removal of nitrate from drinking water can be genuinely considered an eco-efficient technology.

1. Introduction

In soil, the essential nutrients to plant growth are mainly given by nitrates, which in their turn are naturally provided by both soil bacteria, that convert the atmospheric nitrogen into nitrate, and by man. Most significant anthropogenic activities related to nitrate soil supplies are the drainage of human waste (septic systems and sewage sludge) in water flows without convenient operation or maintenance, the usage of recycled domestic wastewater in agriculture irrigation systems and the usage of farmland nitrogenous fertilizers (Galloway et al., 2003, 2004; Galloway, 2005; Wakida and Lernerb, 2005, 2006; Almasri, 2007).

Although nitrate NO_3^- is a relatively stable element in soil that is rarely combined with other compounds or bind to soil particles, it is also a very soluble ion that can easily move with water that percolates through the soil profile ending, eventually, in an underlying water body. In addition, being leachable to nitrite (NO_2^-).

Up to certain limits, nitrate present in drinking water has a toxic effect in human health due to the natural reduction to nitrites by gastric enzymes. In the form of nitrite, and up to certain limits, it can even cause more serious problems as it combines with haemoglobin in the human blood to form methaemoglobin, which is incapable of binding the oxygen. In addition, there is a clear link between nitrite and the formation of nitrosamines, a group of carcinogens produced from the reaction of nitrite with amines,

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amides, and other nitrogenous compounds that induce damages in liver and other organs over the long run of adult life. For these reasons, in many parts of the world and since decades, nitrate concentrations levels in drinking water supplies, both in surface waters and in groundwater, present a serious motive of concern due to their potential threats to human health (Leeuwen et al., 1999; UNEP, 2007; Fowler et al., 2003; Sebiloa et al., 2013).

According to the most updated World Health Organization (WHO) guidelines a limit of 50 mg l^{-1} should be considered for nitrate ion concentration (or 11 mg l^{-1} as nitrate-nitrogen) to protect against the formation of methaemoglobinaemia in bottle-fed infants (short-term exposure), a limit of 3 mg l^{-1} as nitrite ion (or 0.9 mg l^{-1} as nitrite-nitrogen) to protect against the formation of methaemoglobinaemia in bottle-fed infants (short-term exposure) and a limit of 1 for the sum of the ratios of the concentrations of combined nitrate plus nitrite (WHO, 2011).

The most conventional water treatment technologies applied to nitrate removal from water include chemical process (such as chemical nitrate reduction and catalytic denitrification), physical processes (such as reverse osmosis), chemical–physical processes (such as ion exchange) and biological denitrification processes. These processes have pros and cons and are well documented: chemical reduction is often not viable at large scale due to the cost of operation, reverse osmosis and ion exchange processes eventually generate secondary wastes and biological denitrification tend to be a time-consuming process and requires constant supply of organic substrates. On the other hand, some clogging of the ground by biomass and gas formation tend to occur and the assurance of optimum, and safety conditions in a biological process are difficult to maintain due to the unavoidable contamination of soil and water by dead-bacteria (Kapoor and Viraraghavan, 1997).

Permeable reactive barriers (PRBs) have been used since the early nineties as a cost-effective in-situ technology for nitrate removal from water (USEPA, 2002; Puls, 2006). Typically, a PRB is constructed perpendicular to the groundwater flow and below the water table in order that the natural hydraulic gradient transport the contaminant trough the reactive media within the PRB turns it able to degrade (transform) or immobilize them as the water flows trough it. From the geotechnical point of view, installation of a PRB does not cause increased subsoil softening (USEPA, 2002).

Among different materials that can be considered as reactive media in PRBs, zero-valent iron (ZVI) and biologic walls have been prevalent (USEPA, 1998, 2002; Puls, 2006). However, and taking in consideration the previous statements, chemical nitrate reduction process using ZVI is considered to be more effective due to the fact that this process is faster, simpler and more efficient for the complete removal of nitrate and, most important, it does not require maintenance nor it produces waste (Huang et al., 1998; USEPA, 1998).

In the beginning of the twenty-first century, industrial advances related to nanotechnology led the chemical nitrate reduction process using ZVI particles to be optimized (Zhang, 2003). The main advantages of nanosized zero-valent iron particles (NZVI) are due to the fact that they possess larger surface areas and thus a higher surface reactivity than the ones at macro-scale, which eventually improve the efficiency of the removal process in conditions of high concentration levels of nitrate (USEPA, 2007). So far, the use of this new-generation material in groundwater denitrification PRBs seams to be a success (USEPA, 1998).

It is well known that a capable selection of the best management option for controlling groundwater nitrate contamination should be always sustained on the most up-to-date information available. Taking also into account both the high abundance of information available covering the past decade on the use of nanosized iron based permeable reactive barriers (NZVI-PRBs) in denitrification of groundwater sources and the fast progress that is unquestionably is attached to nanotechnology industry, the objective of this paper is to conduct a systematic review on the use of NZVI-PRBs in nitrate reduction in water for human consumption.

2. Material and methods

2.1. Search strategy

The review methodology employed followed the PRISMA statement reporting guidelines (Liberati et al., 2009). A thorough search process for relevant articles was conducted, between April and July 2014, in 34 databases: ACM Digital Library, ACS Journals, AHA Journals, AHA Journals, AIP Journals, AMA Journals, Annual Reviews, ASME Digital Library, BioMed Central Journals, Cambridge Journals Online, CE Database (ASCE), Directory of Open Access Journals (DOAJ), Emerald Fulltext, Geological Society of America (GSA), Highwire Press, IEEE Xplore, Informaworld (Taylor and Francis), Ingenta, IOP Journals, MetaPress, nature.com, Oxford Journals, Political Science: A SAGE Full-Text Collection, Project Muse, Royal Society of Chemistry, SAGE Journals Online, SciELO – Scientific Electronic Library Online, Science Magazine, ScienceDirect, Scitation, SIAM, Sociology: A SAGE Full-Text Collection, SpringerLink, The Chronicle of Higher Education, Wiley Online Library. The above mentioned search process was structured in the following four stages in order to capture all relevant papers:

- Stage 1: Definition of an initial set of pertinent terms to carry out the initial set of individual searches for relevant studies. The following key words were employed: nanoparticle, permeable reactive barrier, denitrification;
- Stage 2: Usage of synonyms of the initial set of keywords to carry out a second set of individual searches for relevant studies. The following synonyms of the first set of key words were employed: denitrification barrier, denitrification wall, nitrate removal;
- Stage 3: From the papers selected from stages 1 and 2 of the searches, further pertinent terms were identified to perform a third set of individual searches for relevant papers. At this stage the following addition key words were employed: nanomaterial, nitrogen removal;
- Stage 4: finally, a search to capture additional relevant papers was conducted from the references of the selected papers identified during the previous three stages.

In every search, a combination of two terms at a time, linked with the Boolean operator “AND”, was employed contemplating papers published from 2004 onwards and written in English. It was admitted a possible bias inherent to the fact that non-English language papers were excluded from the search process.

2.2. Exclusion criteria

For the first three stages of the search process, the documents yielded by each individual search with a given pair of terms were compiled in an excel data-base, considering the type of document (e.g. article, encyclopedia, etc.), document title, author, publication, year of publishing, abstract, and database from which the document was retrieved. Afterwards the listed documents were analysed one by one and the duplicates were identified for exclusion from the in-depth analysis; 1199 duplicates were identified. Then the listed documents were once again analysed and those without author name, title or date were identified for exclusion, respectively 216, 0 and 0. The papers were then screened by title to identify those that were not related to the subject. At this step of the process