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Artificial road runoff water treatment by a pilot-scale horizontal permeable treatment zone

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ABSTRACT

Runoff water from urbanized areas, including roads and related objects may be a source of micropollution. One of the methods limiting its migration to surface- and groundwater receivers may be the application of horizontal treatment zones enhancing natural attenuation of the soil-water environment. The aim of the presented small-scale field tests was testing the utility of mixtures of zero valent iron – ZVI, activated carbon – AC, zeolite – Z, limestone – LS, and silica spongolite – SS as reactive materials. During the 11-month experiment, runoff water samples were collected from three treatment zones and subjected to physical and chemical analyses (pH, EC, Eh, Cl^-, Fe^{2+}, Cd^{2+}, Cu^{2+}, Ni^{2+}, Pb^{2+}, Zn^{2+}, NH_4^+, NO_2^-, NO_3^-, COD, ΣPAH, PEE). Additionally, the change of the hydraulic conductivity of the mixtures was systematically controlled. During the experiment, the highest mean values of removal efficiency of Cl^- (13%) and COD (82%) were observed for ZVI/AC/Z; Ni^{2+} (94%), Zn^{2+} (99%) and NH_4^+ (82%) for ZVI/AC/LS; and PEE (70%) for ZVI/AC/SS. Cadmium (98%), copper (100%) and lead (100%) were removed with the same intensity by all three systems. ΣPAH were removed with the same intensity at 97% by ZVI/AC/SS and ZVI/AC/LS. However, the contact of runoff water with both mixtures resulted in increased emission of NO_2^- and NO_3^-. The experiment showed also that the hydraulic conductivity of the mixtures decreased to a minimal value of $3.2 \cdot 10^{-5}$ m/s for ZVI/AC/Z.

1. Introduction

Dynamic development of civilization causes increasing degradation of all elements of the environment, including water resources. Runoff from road surfaces and road objects such as motorway service areas are considered as significant linear sources of pollution of water receivers (water reservoirs, rivers, surface- and groundwater being sources of potable water) and air (Mazur et al., 2015; Edwards et al., 2016; Galfi et al., 2016; Laña et al., 2016; Schindler et al., 2017). Typical contaminants of road runoff water are heavy metals (Cd, Cr, Cu, Ni, Pb, Zn, Mn, Pt, Ti and V), organic compounds, nutrients (nitrogen and phosphorus compounds), chlorides and total suspended solids (Brown and Peak, 2006; Davis and Birch, 2010; Fach and Dierkes, 2011; Charters et al., 2016). Part of the mentioned components, i.e. Cr, Cu and Zn are not toxic to plants in lower concentrations, on the contrary, they are indispensable for their development, while others, such as Cd, Pb, Ni and polycyclic aromatic hydrocarbons (PAH) are classified as 'priority substances', i.e. substances that pose serious hazard to the water environment (EU EQS 2007; Hilliges et al., 2013). The main sources of these substances in road runoff water include abrasion of tires and brake blocks, leakage of oil and lubricants, organic bituminous ingredients, fuel combustion, road dust, and remains after winter road maintenance (Novotny et al., 2008).

Acording to Hilliges et al. (2013) and Fu et al. (2014), commonly used objects of the road drainage system, decreasing the inflow of runoff water to receivers, can be subdivided into several groups: 1) filter strips and swales, 2) infiltration systems, 3) storage facilities, and 4) permeable surfaces. Additionally, the main elements of the drainage system, serving to eliminate organic pollution and runoff water suspension solids from roads, are separators and settlers (Kluge and Wessolek, 2012). These devices are to a lesser degree effective for the removal of dissolved heavy metals (not susceptible to biodegradation, accumulating in the environment) and chlorides present in winter (mobile in the soil-water environment). Sustainable Urban Drainage Systems (SUDS), applied for decades in the management of runoff water from urbanized areas, including transportation routes, are studied during the last 15 years with regard to the intensity of micropollution infiltration to the soil-water environment (Radziemska and Fronczyk, 2015; Sas et al., 2015; Koda et al., 2016; Tedoldi et al., 2016). Analysis of the degree of pollution of soils below 120 SUDS, conducted by Tedoldi et al. (2016), has indicated that there is a trend to accumulate heavy metals and organic pollutants in the surface soil horizon from 0.10 to 0.30 m below the surface, but in the case of unfavorable factors, such as large soil permeability, privileged flow pathways and very low

pH, micropollutants may migrate into deeper horizons, including aquifers. The undesirable dispersion of these components may be inhibited by the introduction of selected plants in wetland systems or in soils subject to pollution leaching (Michel, 2012; Radziemska et al., 2017; Schmitt et al., 2015; Vymazal and Březinová, 2015; Li et al., 2016), as well as the application of reactors filled with particular materials on small areas such as parking lots (Birch et al., 2005; Zhou et al., 2003; Fuerhacker et al., 2011).

In ecologically valuable areas in particular, there is a growing need to apply additional protection methods minimizing the inflow of contaminants to receivers. However, the application of typical methods of water treatment, such as chemical precipitation, reduction, osmosis, etc., is very expensive and energy-consuming. An alternative is the support of natural attenuation processes that use mineral and organic sorbents as elements supplementing infiltration systems, such as soakaways, infiltration trenches, infiltration basins, vegetated swales and filter strips, and forming horizontal permeable treatment zones working similarly as permeable reactive barriers (PRB). The theoretical essentials of the PRB method and examples of its application in groundwater treatment in practice have been presented by Starr and Cherry (1994), Powell et al. (1998), Gavaskar et al. (2000), Fiorenza et al. (2000), Finžgar et al. (2005) and Bastiaens et al. (2010). The most commonly used reactive materials filling the treatment zone of PRBs and decreasing pollution migration in the soil-water environment include zero valent iron (Gillham and O'Hannesin, 1994; Feng et al., 2016), activated carbon (Roehl et al., 2005; Bortone et al., 2013), zeolites (Bandura et al., 2015; Bilgin and Sanin, 2014; Vignola et al., 2011) and a number of other natural materials, such as clay, diatomite, limestone, chalcedonite, carbonates, sulfates, iron oxides and hydroxides, aluminum oxides, phosphates, peat and biosorbents (Roehl et al., 2005; Phillips, 2009; ITRC, 2011; Wang et al., 2013; Fronczyk et al., 2014; Fronczyk and Radziemska, 2014, 2016).

This paper presents the results of small-scale field tests focused on three systems treating artificial road runoff water, composed of mixtures of: zero valent iron (ZVI), activated carbon (AC) and silica spongiolite (SS) in system 1; ZVI, AC and zeolite (Z) in system 2; and ZVI, AC and limestone (LS) in system 3. The aim of the research was testing the removal efficiencies of heavy metals, chlorides, nitrogen compounds and PAHs during contact of runoff water with mixtures infilling the analyzed systems, at variable temperature conditions. However, the main purpose of the conducted tests was an attempt to identify the mixture of materials, which removes pollutants from runoff water with the greatest intensity, as well as an understanding of the removal processes occurring in three analyzed systems.

2. Material and methods

2.1. Description of the pilot-scale systems

Three pilot-scale systems of horizontal permeable treatment zones were installed at the campus of the Warsaw University of Life Sciences located in Warsaw, Poland. To prevent the potential infiltration of pollutants to the soil-water environment, horizontal permeable treatment zones were inserted in HDPE containers. To provide a temperature occurring in the field inside the containers, they were installed in the ground. Additionally, the containers were tightly sealed between the measurement cycles, which stopped the inflow of natural precipitation and groundwater. The walls of the containers, 0.4 m in diameter and 0.80 m high, were roughened to prevent the flow of contaminated water downward along the sides. The systems were supplied with piezometers (internal diameter 0.05 m), filtered in the lower part of the quartz gravel, which allowed to collect samples for chemical analyses. The pilot-scale systems were designed to ensure vertical flow through filter media. The scheme of the pilot-scale treatment systems is presented in Fig. 1. At the top and bottom of the containers were placed 0.15 m thick quartz layers (particle diameter of 1.0–5.0 mm).
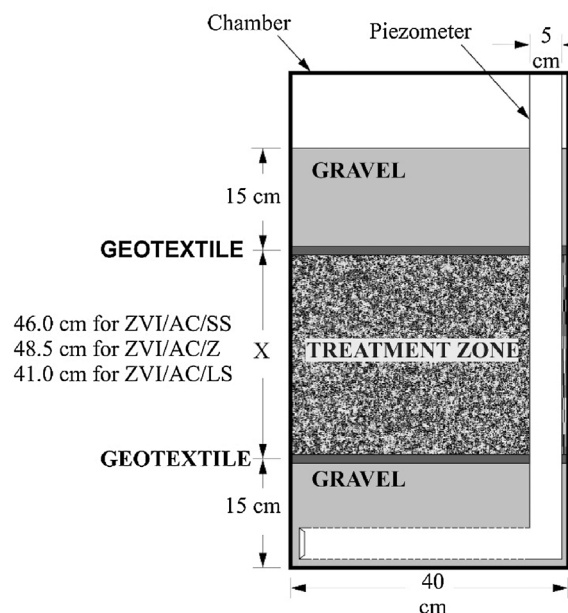

Fig. 1. Schematic diagram of the pilot-scale systems.

Treatment zones with filter media (mixtures) were placed between the quartz layers. Mixtures composing horizontal permeable treatment zones comprised two materials most commonly applied in the PRB technology (Roehl et al., 2005) – zero valent iron (ZVI) and activated carbon (AC). An element differing the systems was the third component of the mixtures: silica spongolite (SS) in system 1, zeolite (Z) in system 2, and limestone (LS) in system 3. For comparison of the pilot-scale systems, the treatment zones were made of mixtures, in which all components were added in a weight proportion of 1:1:1. SEM images and XRD patterns of reactive materials used in the preparation of the mixtures are presented in Fig. 2. The specific surface of the materials was: ZVI – $0.52 \, m^2/g$, activated carbon – $856.05 \, m^2/g$, silica spongolite – m^2/g (Pawluk and Fronczyk, 2015), zeolite – $29.04 \, m^2/g$ (Fronczyk and Garbulewski, 2013), and limestone – $0.30 \, m^2/g$ (Fronczyk et al., 2015).

Investigations presented in the literature (Backstrom et al., 2003; Cekstere et al., 2008; Novotny et al., 2008; Fronczyk et al., 2016) indicate that the chemical composition of runoff water varies with time. Therefore, artificial road runoff water (ARRW) was used in the pilot-scale experiment. ARRW is characterized by a content of heavy metals and organic substances typical of this pollution source. Artificial road runoff water was prepared using NaCl, $CdCl_2 \cdot 5H_2O$, $CuCl_2 \cdot 2H_2O$, $NiSO_4 \cdot 7H_2O$, $Pb(NO_3)_2$, $ZnCl_2$ and diesel in distilled water. The physical and chemical composition of ARRW is presented in Table 1. This solution assures full control of the experiment conditions. It is also worth emphasizing that a stable pollution concentration was applied in the experiment, which results in less favorable work conditions of the treatment zones compared to natural conditions. As noted by Piguet et al. (2008), in the case of road runoff resulting from a single rainfall, the highest concentration of pollution (80%) occurs in the first 30% of rainfall duration, while later the pollution concentration decreases significantly.

2.2. Sampling and analysis of the physical and chemical parameters

The volume of ARRW introduced in each measurement cycle to the systems (16 L) and the frequency of ARRW supply (1 cycle per week) were assumed based on data from the Ursynów SGGW weather station collected in 1960–2009 (Majewski et al., 2010). In the initial phase of the experiment, each system was saturated by free soaking of 50 L of clean distilled water. Next, cyclic measurements were conducted for 11