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Metal accumulation in an artificially recharged gravel pit lake used for drinking water supply

P.N. Mollema [a,b,*], P.J. Stuyfzand [a], M.H.A. Juhász-Holterman [c], P.M.J.A. Van Diepenbeek [c], M. Antonellini [b]

[a] *VU University Amsterdam, Faculty of Earth and Life Sciences, De Boelelaan 1085, 1081 HV Amsterdam, The Netherlands*
[b] *University of Bologna, Laboratory "Renzo Sartori" Ravenna Campus, Via San Alberto 163, 48123 Ravenna, Italy*
[c] *NV WML Waterleiding Maatschappij Limburg, Limburglaan 25, 6229 GA Maastricht, The Netherlands*

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ABSTRACT

Gravel pit lakes offer a variety of uses after excavation has ceased. One of those uses is the storage, infiltration, aquifer passage, and production of drinking water. We have investigated such a gravel pit lake in The Netherlands that is a state of the art drinking water production facility. The gravel pit lake is a flow-through lake, fed naturally by groundwater (~17%) and rainwater (~6%), and artificially with Meuse River water (~77%). The average concentrations of Cd, Cr, Cu, Ni, Pb and Zn in the lake's bottom sediments have increased over a 10 year period. Acidifying redox reactions caused by lowering of the water table and farmland fertilization upstream from the lake explain the mobilization of metals in the soil and subsequent transport with groundwater towards the lake. Dissolved metals (Al, Cd, Cr, Cu, Fe Mn, Ni and Zn) and PO_4^{3-} flow with the groundwater towards the lake, where they interact with oxygen-rich and alkaline water to (co)precipitate as Fe, Mn and Al oxides. The chemistry of the gravel pit lake water is determined by a complex interplay between the input of NO_3^- and PO_4^{3-}, soil composition up stream, biochemical processes in the lake that supply organic material, mixing processes (artificial and natural) that determine the amount of stratification and oxygen, and redox cycling processes in the bottom sediments that influence the concentration of metals and P in the lake water. Gravel pit lakes are young compared to natural lakes and the long term influence of these lakes is still largely unknown. This study confirms that they can form a sink for metals and thus influence the metal budget of a watershed. Compared to other artificial recharge methods, infiltration in a gravel pit lake along a river has certain advantages in that the lake already exists, the transport distance for the water to be infiltrated is short, the pretreatment of the infiltrated water is simple, and the long residence time of the water in the lake and the lake bank filtration attenuates quality variations of the infiltrated river water. For drinking water production or other usage of gravel pit lakes, an integrated monitoring and management of land use, ground- and surface water as well as lake bottom sediments are needed.

1. Introduction

World use of sand and gravel to construct buildings, highways and anything concrete is enormous. According to the USGS (2013), the main producers of sand and gravel extract a total of 140 million metric tons per year. Gravel is produced from natural gravel deposits such as streambeds, river terraces, beach deposits or alluvial fans. Where the gravel is extracted at or below the water table, the pits fill up with groundwater and become an artificial lake. These lakes change the hydrology and hydrochemistry of the nearby surface- and groundwater and soil. Changes in the hydraulic gradient may cause groundwater to flow toward or away or through these gravel pit lakes disturbing the original groundwater flow patterns and hydrologic budget (Gourcy and Brenot, 2011; Mollema et al., 2013; Mollema et al., in prep). The lakes will also affect the hydrogeochemistry of the surrounding groundwater bodies. This may be beneficial as, for example, in the case where phosphates are retained and nitrates are broken down when contaminated groundwater flows through gravel pit lakes (Herzsprung et al., 2010; Muellegger et al., 2013). Gravel pit lakes may also have a negative influence, for example, by allowing the mobilization of soil-bound compounds like arsenic (Marques et al., 2008, 2012; Mollema et al., in prep; Wallis et al., 2010; Weiske et al., 2013) or the growth of poisonous algae in response to accumulation of nutrients such as P and N and availability of light (Downing et al., 2001; Smith, 2011). A gravel pit lake often lacks natural inflow and outflow by streams. This contributes to accumulation of materials coming in with groundwater or from atmospheric deposition on the gravel pit lake bottom, making it a sink for carbon

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* Corresponding author at: University of Bologna, Laboratory "Renzo Sartori" Ravenna Campus, Via San Alberto 163, 48123 Ravenna, Italy. Tel.: +39 0516257958.
E-mail addresses: pmollema@gmail.com (P.N. Mollema), p.j.stuijfzand@vu.nl (P.J. Stuyfzand), M.Juhasz-Holterman@wml.nl (M.H.A. Juhász-Holterman), P.vanDiepenbeek@wml.nl (P.M.J.A. Van Diepenbeek), m.antonellini@unibo.it (M. Antonellini).

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(Muellegger et al., 2013) or metals (this paper). These processes are not instantaneous and may occur over a long time span after the gravel pit lake was originally excavated. Since most gravel pit lakes are very young, scientists have only started to understand their long term impact on the environment. In general, the material making up the porous matrix of an aquifer tends to change the composition of the groundwater downstream, following redox zonations (Appelo and Postma, 2005; Stuyfzand, 1999) and the presence of a gravel pit lake adds a different redox environment. Since gravel pit lakes typically follow a homogeneous sedimentary deposit, there are often many close to one another. These artificial lakes change the landscape. After the excavation activities have stopped, the lakes, in many cases, offer habitat to a large range of native and invasive plant and animal species (e.g. EEA, 2009; Santoul et al, 2009) and can be used as natural areas or for recreational activities such as fishing, swimming and boating. They are, however, also used for development of residential areas on the water as in Gloucester (Wikipedia, 2013), fish farming, heat storage (Novo et al., 2010). This paper is about a gravel pit lake used for or storage and infiltration of river water used for the production of drinking water. In densely populated countries such as The Netherlands (499 persons/km^2) there may not be enough space for natural recharge to supply all the needed (drinking) water, the more so because typically salt water intrusion threatens coastal aquifers in densely populated areas (e.g. Werner et al., 2013), and efficient drainage systems cause upwelling of salt water in low lying areas (e.g. Stuyfzand, 1995) or drought related problems in higher parts (Van Ek et al., 2000). Therefore artificial recharge (AR) of river water in coastal dunes (e.g. Karlsen et al., 2012) and river bank filtration (e.g. De Vet et al., 2010; Stuyfzand et al., 2006) has been common practice for more than 50 years, but using gravel pit lakes for artificial recharge and production of drinking water is relatively new.

The accumulation of metals in water and bottom sediments of freshwater lakes and reservoirs has been of concern with regards to e.g. zink, mercury, lead, cadmium and chromium (e.g. Linnik, 2000; Linnik and Zubenko, 2000; Toivonen and Österholm, 2011). The source of these and other metals may be atmospheric (Belzile et al., 2004) or waste from mining activities (Martin and Pederson, 2002) or AR of contaminated surface water (Stuyfzand et al., 2006). The presence and mobility of trace metals in sandy sediments are linked to various settings in which redox reactions, organic material, low pH and bacteria play an important role (Du Laing et al., 2009 and references in there; Kjøller et al., 2004).

The objectives of this paper are to identify the source and the processes leading to the increasing metal concentrations on the lake bottom of gravel pit lake De Lange Vlieter in the Netherlands, which is used as a water reservoir for drinking water production. Hereto we analyzed the lake bottom sediment and followed groundwater chemistry along a profile upstream and downstream from the gravel pit lake. We hope this will contribute to the evaluation of the use of gravel pit lakes as AR basin in comparison with other types of AR.

2. Methods

2.1. Research area

The gravel pit lakes of this study are located along the Meuse River. The Meuse has its origin in France, flows through Belgium and confluences with a branch of the river Rhine before it flows into the Dutch North Sea. Is total length is about 875 km. The origin of the Meuse is near Pouilly-en-Bassigny on the Langres plateau, north of Dijon in France, in the north-eastern margin of the Paris Basin (Fig. 1a) and close to an important watershed divide. South of this watershed, the rivers flow towards the Mediterranean, east, west and north of here towards the Atlantic and North Sea. The Meuse River cuts for the first 200 km of its course through the Jurrassic Marine Carbonate sediments of the Paris basin. Near Charleville, it enters the geological province of the Ardennes (Fig. 1a) that consists of Paleozoic partly metamorphosed sediments (sandstone, slate, schist, dolomite, conglomerate, limestone) deposited 390–300 my ago, that were pushed up and folded during the Hercynic Orogeny. Near Liege (Belgium), the Meuse leaves the Ardennes and enters a sequence of tertiary sediments that were deposited on top of the Brabant massif, a relic of an old mountain chain formed during the Caledonic orogeny. From Eijsden, in the Netherlands, the river flows further North through fluvial quaternary deposits. There are some gravel or sand pit lakes along the first 200 km of the Meuse course in the Jurassic sediments of France. Most gravel pit lakes, though, are excavated in the Quaternary river deposits that are exposed along the stretch running through Belgium and The Netherlands. The gravel pit lake of concern is called De Lange Vlieter (DLV from now on) and lies close to the city of Roermond (Fig. 1b). DLV is one of about 40 gravel pit lakes excavated along a 22-km-long stretch of the Meuse River between Maaseik (Northeast Belgium) and Asselt (South Netherlands). The excavation started in the 1970s and was terminated in 1996 (Juhasz-Holterman, 2012).

The stratigraphy in our study area consists, of a clay layer of the Brunssum Formation currently at about 100 m depth of the Late-Mioceen en Plioceen (5 Ma years BP). This is the base of the aquifer and is overlain by the Veghel-Sterksel Formation, which consists of very fine to fine sands of Pliocene to Holocene age (2.6 Ma BP; Fig. 1c). This in turn is overlain by the Beegden Formation that consists of gravels and coarse sands deposited in river terraces by the Meuse River during the Weichselian Glacial period (maximum 25,000–13,000 years BP; Berendsen and Stouthamer, 2002). The sediments that now form the river terraces were generated by erosion in the Ardennes and the Paris Basin and subsequently transported into the valley of the Meuse river (Zonneveld, 1974; Zagwijn, 1989) during alternating glacial and interglacial periods in which the river regime changed from meandering to anastoming and back many times (Schaller et al., 2004; Veldkamp and van Dijke, 2000). In colder periods the older deposits were buried by rapid deposition in a braided river regime. In warmer periods the river formed meanders. There was more vegetation and consequently during the infill of channels, in abandoned meanders of the river, or in backswamps, clay layers alternated with peat layers and organic gyttjas formed (Tebbens et al., 1999). These now form layers of a few cm to dm thick of loam, clay, peat and other organic layers within the sand and gravel formation (Dinoloket, 2014). Bulk geochemical variation of Late-glacial and Early Holocene River Meuse sediments can be ascribed primarily to sorting processes during fluvial transport and sedimentation but the post-depositional diagenesis of siderite and vivianite in anoxic gyttja environments affects the contents of Fe_2O_3, MnO and P_2O_5 and is site specific (Tebbens et al., 1998, 1999).

The gravel deposits were buried below a 5–12 m thick layer of fine Aeolian sand with thin layers of loam of the Boxtel Formation (late Pleistocene to early Holocene age; 12,000 years BP) that was removed to reach the gravel below. The gravels and coarse sands of the Beegden Formation represent the best quality construction material and were the scope of the excavation.

Mean annual precipitation over the basin is ca. 950 mm a^{-1}. Air temperatures show marked seasonal variations, and potential evapotranspiration is larger from May to October (76% of annual evapotranspiration) than from November to April (24% of the annual evapotranspiration; Ward et al., 2008). The AR and drinking water production started in 2002. The lake has a surface area of 123 ha and a depth down to 35 m with a volume of 25 million m^3 and a water residence time of about 1.5 years. Water from the Meuse River is pumped up through pipes from the Lateraal Canal that is in open connection with the Meuse River, to the DLV lake and let into a basin separated from the main lake so that suspended particles can settle (Fig. 2a and b). There are in total 29 water production wells on three sides around the DLV at a distance of 50–100 m from the lake bank (Fig. 2b). These wells pull the water from the DLV through the soil of the lake bank. There are six air blowers in