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The Effect of Artificial Recharge on Hydrochemistry: A Comparison of Two Fluvial Gravel Pit Lakes with Different Post-Excavation Uses in The Netherlands

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Abstract: Gravel pit lakes form when gravel deposits are excavated below the water table. We studied two fluvial gravel pit lakes called De Lange Vlieter (DLV Lake) and the Boschmolen Plas (BP Lake), in the Meuse River valley (The Netherlands). Water from the Meuse River is pumped only into the DLV Lake that is used for drinking water production. The mean values, the linear trends and seasonal patterns of time series data (2003–2014), of temperature, pH, nitrate, phosphate and sulphate were compared using one-way tests of variance and tests of differences. The effects of river water infiltration on DLV Lake are (1) a change in lake water temperature; (2) an increase in nitrate concentration (3) an increase in phosphate concentration and (4) a decrease in sulphate concentration. The effects of the air blowers in DLV Lake are (1) mixing of lake water; (2) decreasing pH in spring and summer (3) water oxygenation. Linear regression analysis shows an initially increasing nitrate concentration in DLV Lake that can be explained by the input of nitrate rich Meuse river water. Instead decreasing nitrate and phosphate concentrations in BP Lake and Meuse River reflect a diminished use of fertilizers. The gravel pit lake water temperature does not reflect climatic changes but the use of DLV Lake for artificial recharge has an impact on the seasonal and long-term trends in hydrochemistry. This poses a challenge to lake managers to find the right balance between reduction of eutrophication and accumulation of nutrients and sulphate.

Keywords: gravel pit lake; river; artificial recharge; water quality; nutrients; time series data; seasonal variation; drinking water production; Meuse River; The Netherlands

1. Introduction

Gravel and coarse sand are excavated to fulfill the need for construction materials. The Netherlands do not have a very large gravel production in absolute terms but if the production of gravel is divided over the surface area of the country, it has the largest production per square km in the world (Figure 1). The excavation of so much gravel caused the formation of more than 500 gravel pit lakes. Most of these lakes are used for recreational purposes if they are used at all. However, one particular fluvial gravel pit lake along the river Meuse near the town of Heel, is used by the company NV WML Waterleiding Maatschappij Limburg for storage and infiltration of river water used in the production of drinking water. In densely populated and low-lying countries such as The Netherlands (499 persons/km^2), there may not be enough space for natural recharge to supply all needs for drinking water. This is because salt-water intrusion threatens coastal aquifers [1] and efficient drainage systems cause upwelling of salt-water in low lying areas [2] and drought in higher parts that threaten the ecosystems of natural areas and complicate agricultural development [3]. As a result, Artificial Recharge (AR) of river water into coastal dunes [4] and river bank filtration [5] has been common practice for more than 50 years, but using gravel pit lakes for AR and production of drinking water is relatively new.

After excavation, gravel pit lakes offer space for ecosystem services thus adding different types of habitats in an often urban or agricultural setting. Ecological communities consisting of phytoplankton, zooplankton, macrophytes, fish and birds can flourish in these lakes as in natural lakes (see [6] for an overview of gravel pit lake ecology). Another service of gravel pit lakes is the denitrification of groundwater flowing into the lakes via uptake of nutrients by algae or bacterial processes and the release of nitrogen (N_2) [7–10]. Groundwater, however, may also bring dissolved (toxic) metals into the lakes that subsequently accumulate in the bottom sediments [11]. Variations in hydrochemistry of lakes and rivers include seasonal fluctuations that may be related to biological processes such as growth of phytoplankton during spring or biochemical process as mineralization of carbon and sulphide during times of intense precipitation or human activities such as fertilization of the agricultural lands in the spring time. On top of these seasonal variations, there is considerable year-to-year variability [12]. In this paper, we present a comparison of the hydrochemistry of two flow-through gravel pit lakes with one another and with Meuse River water. One of the studied gravel pit lakes is used for recreation and the other one is used for AR and drinking water production. The hydrochemical comparison is carried out with the scopes of (1) determining the similarities and differences in water temperature pH, nitrate (NO_3^-), phosphate (PO_4^{3-}) and sulphate (SO_4^{2-}) among the three types of surface water with respect to range of values, (mean) values, outliers and seasonal patterns; (2) determining whether there is a significant increase or decrease in mean values over time of temperature, pH, NO_3 PO_4 and SO_4; (3) to deduce the effect of AR with river water and the operation of air blowers on lake water and (4) to determine if the effects of AR can be distinguished from the effects of a change in land use or climate. Throughout this paper we add the correct charges on the ions the first time they are mentioned but then are left out for simplicity.

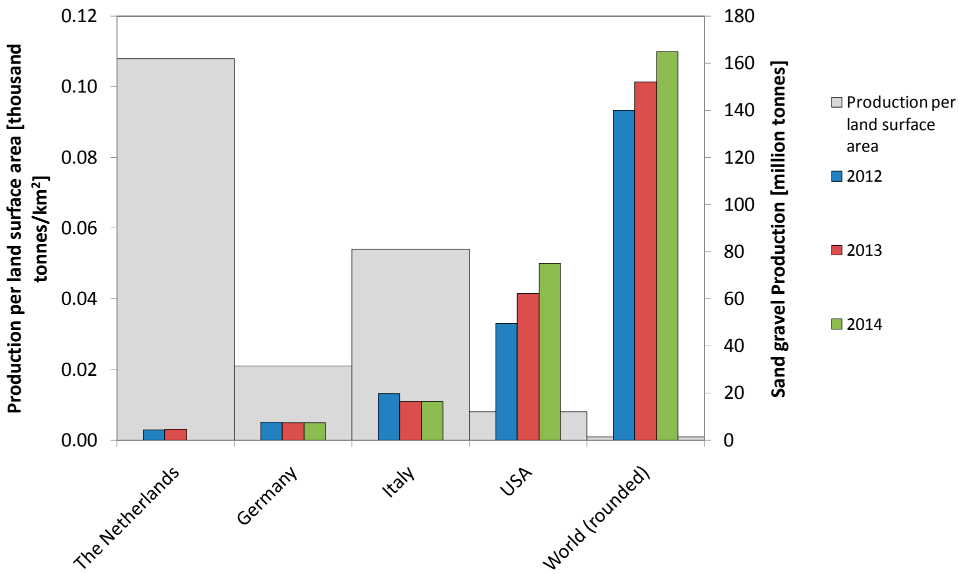


Figure 1. Gravel production of selected countries around the world. Although production in absolute values is not high in The Netherlands (color histogram bars), in relation to its small surface area it is (gray histogram bars). Source [13–15].

2. Study Area

The two gravel pit lakes of this study are located along the Meuse River that has its origin in France, flows through Belgium and merges with a branch of the river Rhine before it flows into the

Dutch North Sea. The gravel pit lakes of concern are De Lange Vlieter (DLV Lake) and Boschmolen Plas (BP Lake) and they lie close to the city of Roermond (Figure 2). The DLV Lake is one of about 70 gravel pit lakes excavated in fluvial deposits along a 22-km-long stretch of the Meuse River between Maaseik (Northeast Belgium) and Asselt (South Netherlands). The DLV and BP Lakes are isolated from the Meuse River and other surface waters. Groundwater flows into the lakes on their western sides and lake water flows out downstream into the aquifer eastward. The excavation started in the 1970s and ended in 1996 [16]. The DLV Lake is used by the drinking water company NV WML Waterleiding Maatschappij Limburg for AR with Meuse River water since the beginning of 2000 and drinking water production started in 2002. DLV and BP Lakes have a similar area (123 and 104 ha respectively; Table 1) and depth (max = 35 m and 30 m), but the water retention time in BP Lake (4.5 years) is longer than in DLV Lake (1.5 years).

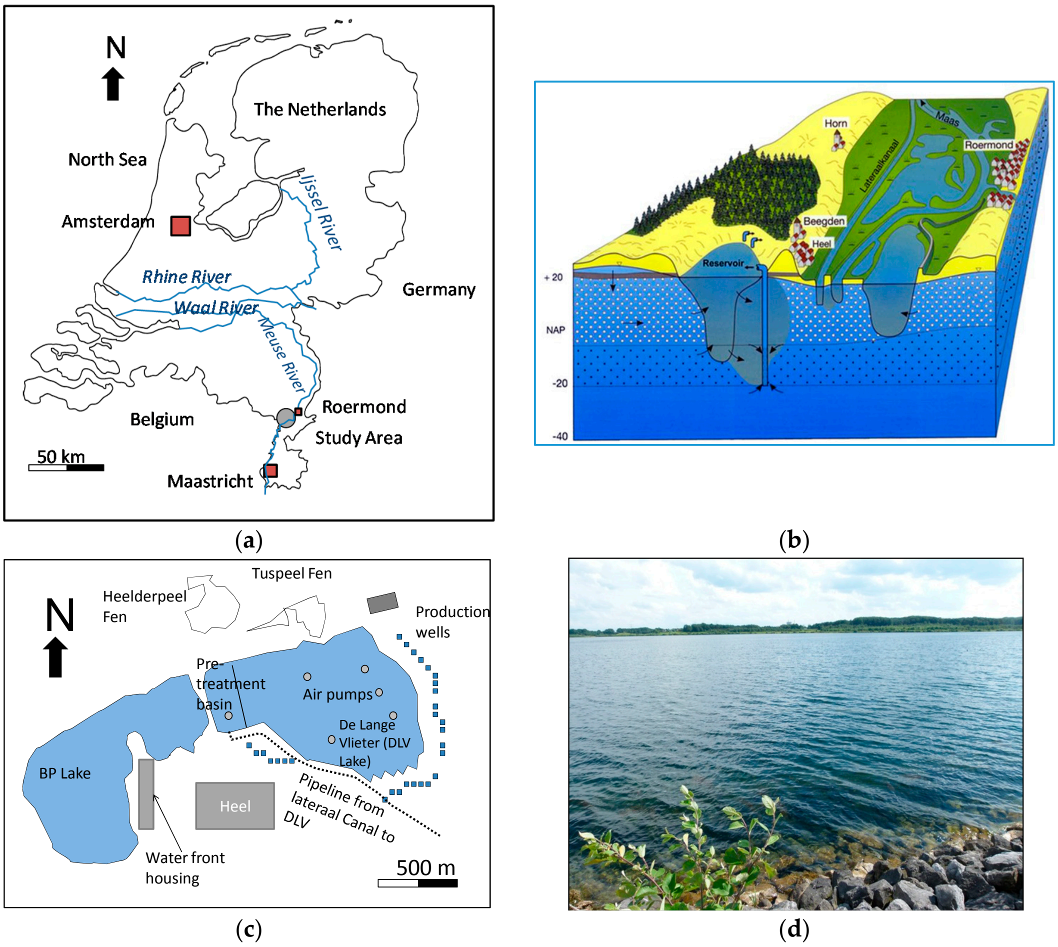


Figure 2. (**a**) Location of study area within the Netherlands; (**b**) 3D artist's sketch of DLV Lake and Meuse River; (**c**) Map of study area with DLV and BP Lakes, location of air blowers and extraction wells and (**d**) Photograph of DLV Lake.

Water from the Meuse River is lifted 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 (Figure 2b,c). Six air blowers in the main part of the lake (since 2002) and three in the settling basin (since 2004) prevent thermal stratification and algal blooms from April until October of each year.

There are 29 water production wells on three sides around the DLV Lake at a distance of 75 to 180 m from the lakeshore (Figure 2). These wells pull the water from the DLV Lake through the soil around the lake into the pipe system that leads to the water purification plant.

Table 1. Properties of the DLV and BP gravel pit lakes.

	De Lange Vlieter (DLV) Lake	Boschmolen Plas (BP) Lake
Surface area	123 ha	104 ha
Maximum depth	35 m	25–30 m
Water residence time *	1.5 years	4.5 years
Current use	Artificial Recharge and drinking water production	Recreation: water front housing
Summer Stratification	No	Yes
Air blowers	9	0

Note: * Water residence time estimated as ratio to lake volume and outflow.

The water level in the lake is maintained constant as much as possible at 20.85 m ASL (meters above sea level) in wintertime and at 21.05 m ASL in summertime to preserve the relatively shallow water table of the natural areas near the lake. Since the construction of the Lateraal Canal in 1972, the water table subsided up to 2 m over a distance of 1 km from the north shore of DLV Lake and up to 6 km north of the lake the water table subsidence was measureable. More information about these (and other) gravel pit lakes can be found in [6,11].

3. Sampling, Chemical Analysis and Statistical Analysis

Lake water at the surface of the DLV and the BP Lakes and of the Meuse River water was sampled bi-weekly or monthly by the WML drinking water company from 2003 to 2014. The Meuse River water was sampled in the channel (Lateraal Canal) from which water is pumped into the DLV Lake. All water samples were filtered through a 0.45 μm filter just after collection. Each sub-sample for analysis of cations and PO_4 was acidified with suprapure 65% HNO_3 (0.7 mL/100 mL). The water was stored in pre-cleaned HDPE plastic bottles. pH, and temperature were measured with a multi-parameter probe. All water samples were analyzed by AcquaLab Zuid. The water temperature depth profiles in the DLV and Boschmolen Lakes were logged by lowering a Schlumberger™ Diver multi-probe from a boat.

The resulting hydrochemical data was described in terms of range of observed values, mean value, standard deviation, range and timing of summer maximum and range and timing of winter minimum values. Where mean values are reported in the text they are accompanied with the standard deviation. One-way tests of variance ANOVA [17] and an analysis of differences (REQW [18]) between the categories (i.e., type of surface water) with a confidence interval of 95% was performed with XLSTAT of Microsoft Office Excel™. This was done to evaluate if the hydrochemistry of the three types of surface water (DLV Lake, BP Lake and Meuse River) are significantly different with respect to one another. The REQW method is a so-called step down procedure, a modification of Tukey's procedure which is considered a very reliable method for comparisons [18]. The missing data were ignored. The number of outliers (extreme values with residuals outside of (−1.96, 1.96)) was counted for each time series and a linear regression over all the time series was performed and tested for significance. Only significant regression lines ($p < 0.05$) are shown in the graphs. The tests were carried out for all five parameters (temperature, pH, NO_3, PO_4 and SO_4).

4. Results

4.1. Temperature with Depth Profiles

The water temperature profile measured in the summer of 2012 in the DLV Lake was constant with depth at 19.3 °C except in the first 2–3 m below the water surface where it reached up to 27 °C during the summer and in the deeper part of the lake where below a 30–40 m depth it dropped to 11–13 °C (Figure 3).

The water in the BP Lake had a temperature stratification typical for the summer period of deep lakes in a temperate climate. The water had a temperature of 20.4 °C down to a depth of 8 m below which there was a sharp decrease in temperature down to 20 m depth (thermocline). Below this depth, the temperature was constant at 5.4 °C.

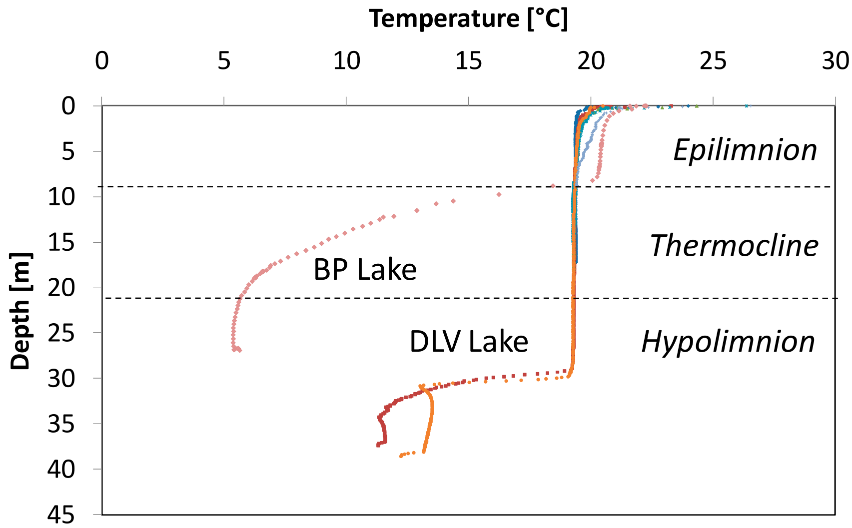


Figure 3. Temperature versus depth profiles at five different locations in DLV Lake and in one location in BP Lake, measured in August 2012.

4.2. Seasonal Variations in BP Lake

The pH and temperature of the BP Lake had seasonal highs and lows where maximum values occurred in summer and minimum values occurred in winter. Peaks in the NO_3 coincided with minimum temperature values in winter (Figure 4). Until summer 2009, the timing of peaks in Total Phosphorus (PO_4) values coincided with peaks in temperature. After that, there was a strong decrease in PO_4 and there were no more clear seasonal maximum values. See Tables 2 and 3 for range of values, mean, standard deviation, outliers, range of summer maximum and winter minimum, timing of maximum and minimum, number of samples and linear regression parameters.

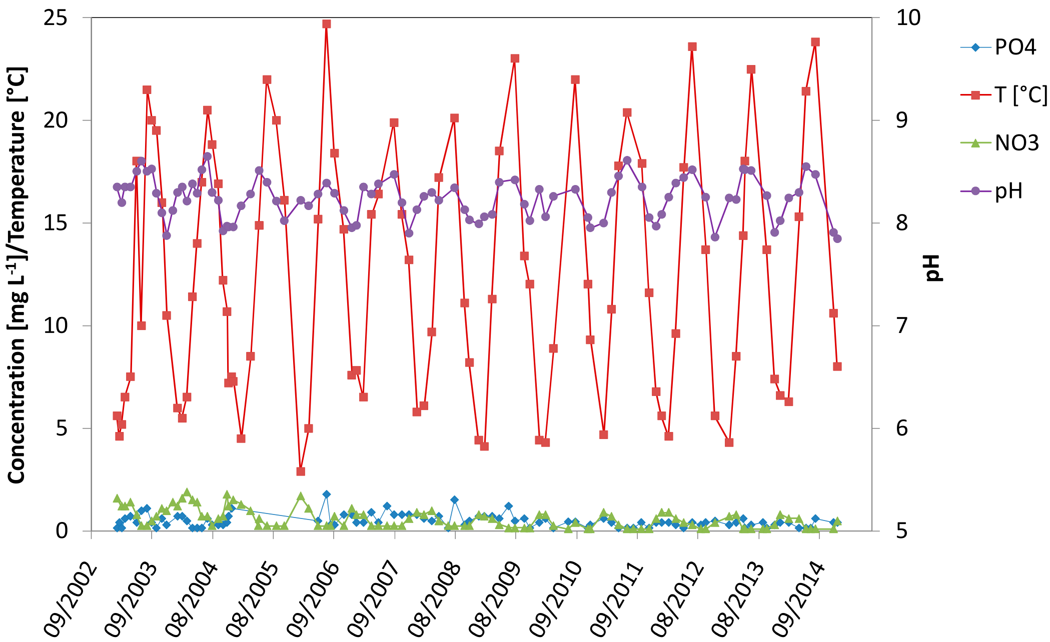


Figure 4. Temperature, pH, NO_3 and PO_4 data of BP Lake from 2003 to 2014. Maximum values of temperature, pH and PO_4 typically occur in summer while maximum NO_3 vales are observed in winter.

Table 2. Statistics for temperature, pH, NO_3 PO_4 and SO_4 concentration in gravel pit lake with artificial recharge (DLV Lake), groundwater-fed only (BP Lake) and River Meuse water. 'n.a.' stands for 'not available'.

Temperature (T)	**DLV Lake (Artificial Recharge)**	**BP Lake (Groundwater Fed)**	**Meuse River**
Range in T (°C)	2.7–23.2	2.9–24.7	3.2–25.0
Mean T and (standard deviation) (°C)	13.3 (5.5)	12.3 (6.0)	14.0 (5.7)
Range summer maximum T (°C)	19.9–23.2	19.9–24.7	20.0–25.0
Range winter minimum T (°C)	2.7–7.6	2.9–6.5	3.2–9.1
Timing maximum T	15/7–31/8	15/7–31/8	15/7–31/8
Timing minimum T	30/1–15/3	30/1–15/3	30/1–15/3
Number of samples *n*	215	94	153
Linear regression R^2	0.001	0.002	0.00001
Linear regression *p*	0.69	0.71	0.84
Linear regression trend over time	No significant trend	No significant trend	No significant trend
Acidity pH	**DLV Lake (Artificial Recharge)**	**BP Lake (Groundwater Fed)**	**Meuse River**
Range in pH	8.0–8.7	7.9–8.7	7.4–8.1
Mean pH (standard deviation)	8.2 (0.1)	8.2 (0.2)	7.7 (0.1)
Range summer maximum pH	8.1–8.7	8.3–8.7	n.a.
Range winter minimum pH	(7.7) 8.0–8.2	7.9–8.0	n.a.
Timing maximum pH	15/3–31/7	1/6–31/8	n.a.
Timing minimum pH			n.a.
Number of samples *n*	47	91	152
Linear regression R^2	0.044	0.007	0.014
Linear regression *p*	0.16	0.428	0.150
Linear regression trend over time	No significant trend	No significant trend	No significant trend
Nitrate Concentration (NO_3)	**DLV Lake (Artificial Recharge)**	**BP Lake (Groundwater Fed)**	**Meuse River**
Range in (NO_3) ($mg{\cdot}L^{-1}$)	1.8–12.8	0.1–1.9	2.8–19.9
Mean (NO_3) ($mg{\cdot}L^{-1}$) (standard deviation)	6.2 (1.2)	0.6 (0.5)	14.8 (2.0)
Range summer maximum (NO_3) ($mg{\cdot}L^{-1}$)	3.1–21.3	0.0–1.9	
Range winter minimum (NO_3) ($mg{\cdot}L^{-1}$)	1.8–5.6	<0.25	
Timing maximum (NO_3) ($mg{\cdot}L^{-1}$)	End of January–end o March	End of November–end of March	December–April
Timing minimum (NO_3) ($mg{\cdot}L^{-1}$)	August–September	July–August	July–September
Number of samples *n*	234	101	251
Linear regression R^2	0.341	0.267	0.055
Linear regression *p*	<0.00001	<0.00001	0.00002
Linear regression trend over time (all data)	Positive	Negative	Negative
Linear regression period 1 R^2	0.706		
Linear regression period 1 *p*	<0.0001		
Linear regression period 1 trend over time	positive		
Linear regression period 1 R^2	0.0008		
Linear regression period 1 *p*	0.332		
Linear regression period 2 trend over time	No significant trend		
Total Phosphate Concentration (PO_4)	**DLV Lake (Artificial Recharge)**	**BP Lake (Groundwater Fed)**	**Meuse River**
Range in (PO_4) ($mg{\cdot}L^{-1}$)	<0.015–0.72	<0.0150–0.18	<0.015–2.2
Mean (PO_4) ($mg{\cdot}L^{-1}$) (standard deviation)	0.09 (0.06)	0.05 (0.03)	0.77 (0.28)
Range summer maximum (PO_4) ($mg{\cdot}L^{-1}$)	n.a.	n.a.	n.a.
Range winter minimum (PO_4) ($mg{\cdot}L^{-1}$)	n.a.	n.a.	n.a.
Timing maximum (PO_4) ($mg{\cdot}L^{-1}$)	All year	All year	All year
Timing minimum (PO_4) ($mg{\cdot}L^{-1}$)	All year	All year	All year
Linear regression R^2	0.09	0.254	0.159
Linear regression *p*	0.072	<0.0001	0.0001
Linear regression trend over time (all data)	No significant trend	Negative	Negative
Number of samples *n*	355	94	137

Table 2. *Cont.*

Sulphate Concentration (SO_4)	DLV Lake (Artificial Recharge)	BP Lake (Groundwater Fed)	Meuse River
Range (SO_4) of complete monitoring period ($mg \cdot L^{-1}$)	51.0–67.0	68.0–88.0	25.0–80.0
Mean (SO_4) of complete monitoring period ($mg \cdot L^{-1}$) (standard deviation)	58.6 (4.3)	76.4 (3.5)	47.6 (13.1)
Range summer maximum (SO_4) ($mg \cdot L^{-1}$)	n.a.	n.a.	55–80
Range winter minimum (SO_4) ($mg \cdot L^{-1}$)	n.a.	n.a.	25–37
Timing maximum (SO_4) ($mg \cdot L^{-1}$)	n.a.	n.a.	End August beginning November
Timing minimum (SO_4) ($mg \cdot L^{-1}$)	n.a.	n.a.	January–March
Number of samples n	47	94	152
Linear regression R^2	0.56	0.22	0.032
Linear regression p	<0.00001	0.150	0.027
Linear regression trend over time (all data)	Negative	No significant trend	Negative
Linear regression period 1 R^2		0.621	
Linear regression period 1 p		<0.00001	
Linear regression period 1 trend over time		positive	
Linear regression period 1 R^2		0.631	
Linear regression period 1 p		<0.00001	
Linear regression period 2 trend over time		negative	

Table 3. Results of the one-way ANOVA, testing whether the mean value of time series of temperature, pH, NO_3, PO_4 and SO_4 for each type of surface water (DLV Lake, BP Lake or Meuse River) is significantly different from an overall mean value. R^2 are the standard residuals, F is the Fisher F-value, p is the significance. Tests of difference for the time series for all three types of surface water.

	One Way ANOVA Analysis				REGWQ: Analysis of the Differences		
Parameters	R^2	F	p-**Value**	**Number of Outliers (St. Residuals Outside (−1.96, 1.96)**	**Significant Difference between BP Lake vs. Meuse River**	**Significant Difference between BP Lake vs. DLV Lake**	**Significant Difference between DLV Lake vs. Meuse River**
Temperature	0.009	3.1	0.047	0	Yes	No	No
pH	0.737	401.7	<0.0001	10 (0 DLV Lake, 8 BP Lake, 2 Meuse River)	Yes	No	Yes
NO_3	0.927	3728.6	<0.0001	28 (8 DLV Lake, 0 BP Lake, 20 Meuse River)	Yes	Yes	Yes
PO_4	0.844	1574.1	<0.0001	27 (1 DLV Lake, 0 BP Lake 26 Meuse River)	Yes	Yes	Yes
SO_4	0.633	250.6	<0.0001	0 DLV Lake, 0 BP, 21 Meuse River	Yes	Yes	Yes

4.3. Seasonal Variations in DLV Lake

The water temperature in the DLV Lake ranged from 2.7 to 23.2 °C (Table 2) and had a seasonal maximum in summer and a minimum in winter. The pH of DLV Lake ranged from 7.7 to 8.7 and initially did not show a distinct seasonal pattern (Figure 5) but from 2010 onward, there was a more systematic pattern where the pH reached a maximum in early spring and then decreased over the summer and increased again in winter. The decrease coincided with the installation and operation of air blowers in the lake. The nitrate concentration reflects a seasonal pattern with maximum values of up to 21.3 $mg \cdot L^{-1}$ reached by the end of winter and minimum values of minimum 1.8 $mg \cdot L^{-1}$ by the end of summer.

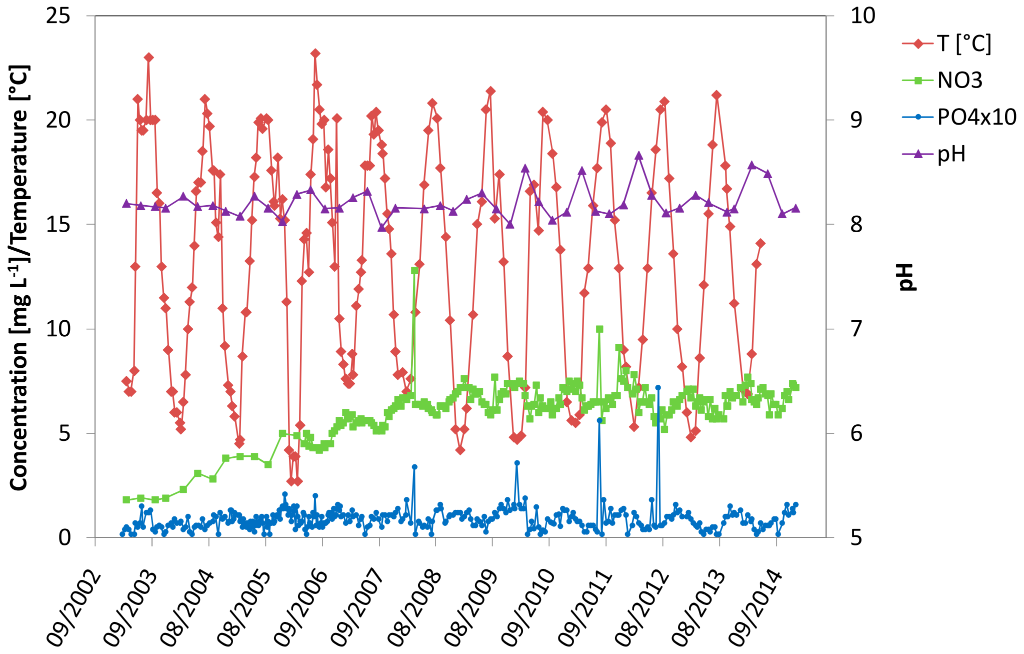


Figure 5. Temperature, pH, NO_3 and PO_4 data of DLV Lake from 2003 to 2014. Maximum values of temperature and pH typically occurred in summer whereas maximum NO_3 and PO_4 vales were observed in winter.

4.4. Comparison of BP Lake, DLV Lake and Meuse River

The ANOVA and tests of difference indicated that the mean values of the NO_3, PO_4 and SO_4 concentrations over the period 2003–2014 are significantly different for all three types of surface water. Meuse River water contained, on average, the most NO_3, PO_4 and the least SO_4 and showed more outliers (Table 2). DLV Lake contained on average more NO_3, PO_4 but less SO_4 than BP Lake. There was only a significant difference in temperature between BP Lake and Meuse River and with respect to pH there was only a significant difference between BP Lake and the Meuse River, or between DLV Lake and Meuse River, but not between the two lakes (Figures 6 and 7 and Table 3).

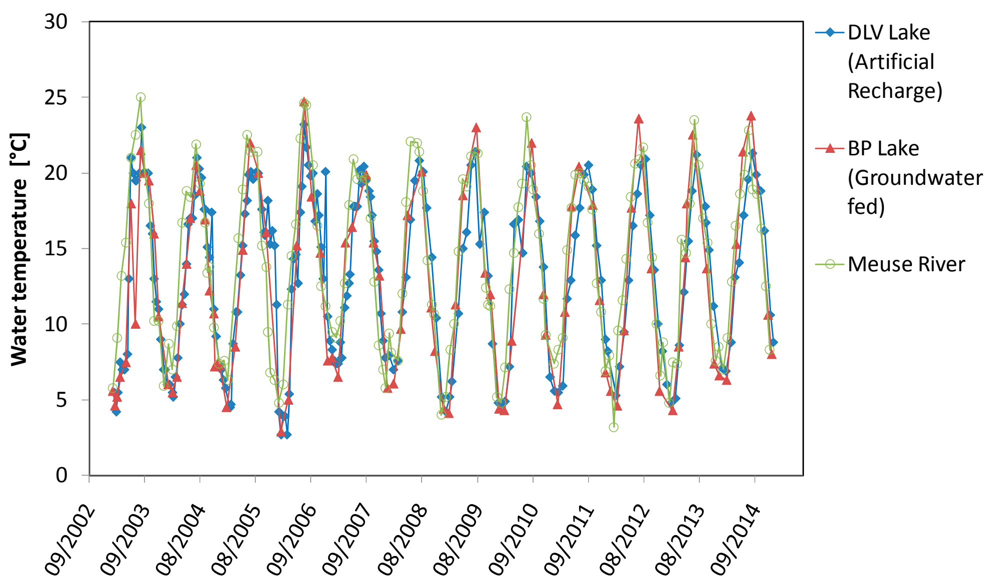


Figure 6. Temperature of gravel pit lake with artificial recharge (DLV Lake), groundwater fed only (BP Lake) and River Meuse water from 2002 to 2014.

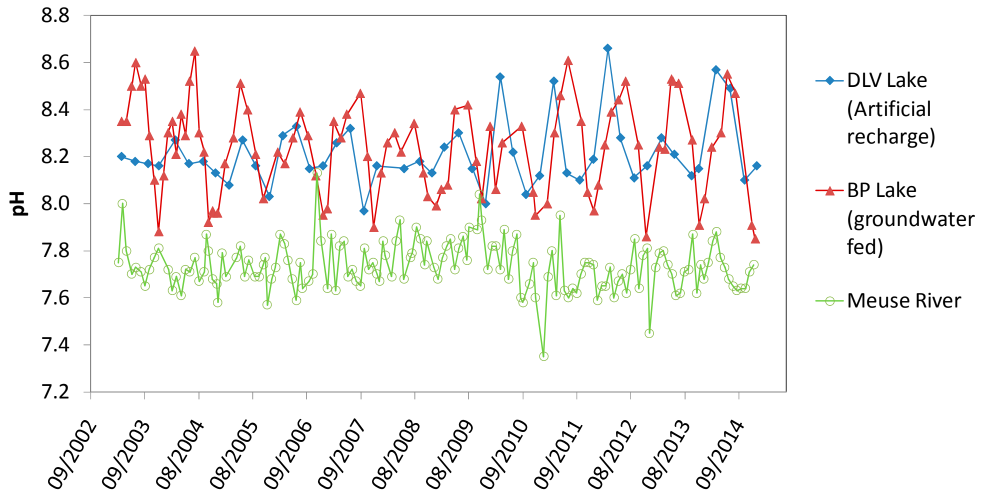
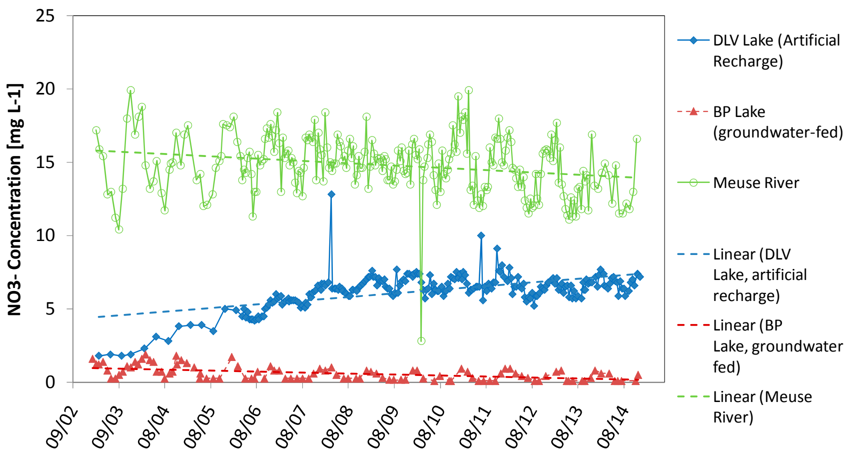


Figure 7. pH of gravel pit lake with artificial recharge (DLV Lake), groundwater fed only (BP Lake) and River Meuse water from 2001 to 2014.

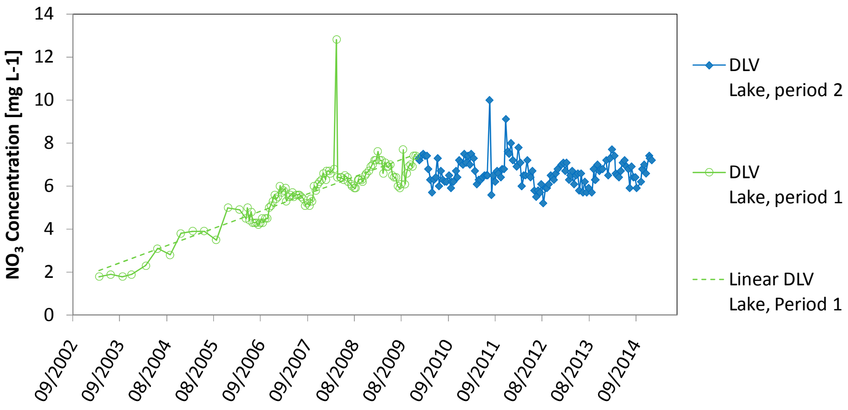
4.5. Linear Trend Analysis of Time Series

Neither the temperature nor the pH time series showed a significant increase or decrease of mean value over the time period considered in any of the three surface water types (Figures 6 and 7 and Table 2). The NO_3 concentration instead showed a significant increase over time in DLV Lake while it decreased in BP Lake and Meuse River water. The mean PO_4 concentration did not show a significant trend over time in DLV Lake but instead did show a significant decrease over time in BP Lake and Meuse River. The SO_4 concentration showed a significant decrease over time in DLV Lake and Meuse River water but no significant trend over the complete monitoring period in BP Lake. However, after we divided the data into two time periods, we found a significant increase until May 2009 and a significant decrease after that. The significant linear regression lines are indicated in Figures 8–10 and the R^2 and p-values for all are listed in Table 2.



(**a**)

Figure 8. *Cont.*



(**b**)

Figure 8. (**a**) NO_3 concentration of gravel pit lake with artificial recharge (DLV Lake), groundwater fed only (BP Lake) and Meuse River from 2002 to 2016; (**b**) NO_3 concentration of gravel pit lake with artificial recharge (DLV Lake) and linear regression trend line over the period 2002–2009 and 2010–2014.

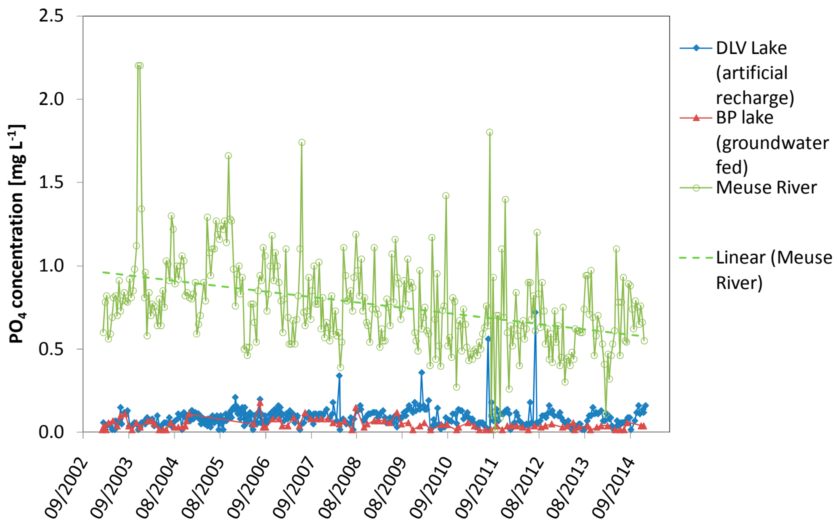
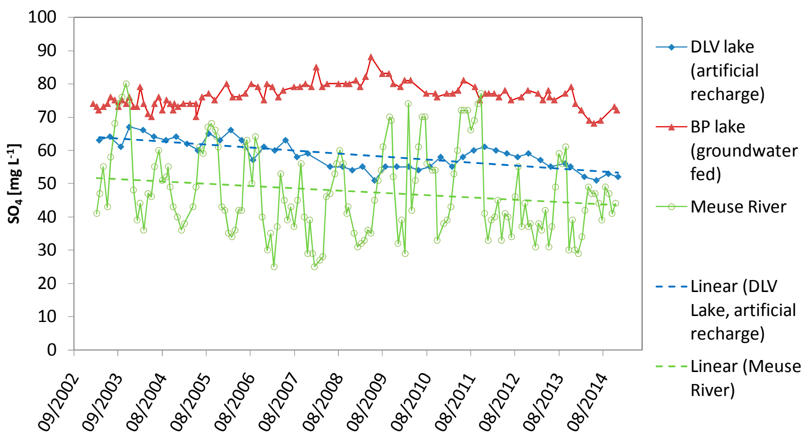
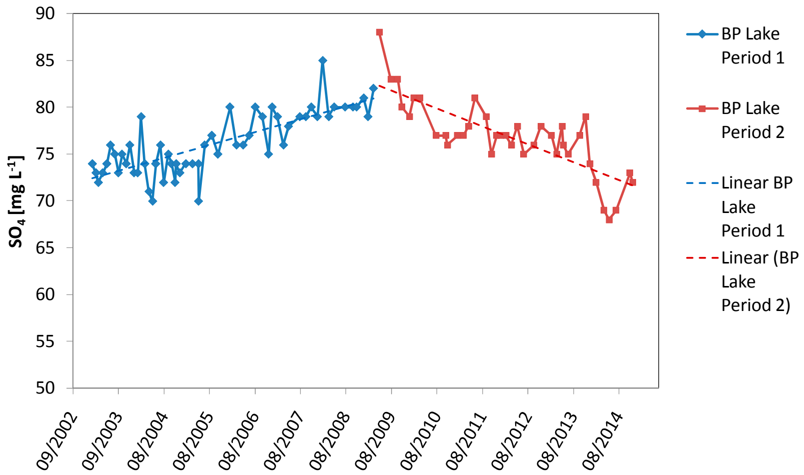


Figure 9. Total PO_4 concentration of gravel pit lake with artificial recharge (DLV Lake), groundwater fed only (BP Lake) and Meuse River from 2002 to 2014.



(**a**)

Figure 10. *Cont.*



(**b**)

Figure 10. (**a**) SO_4 concentration of gravel pit lake with artificial recharge (DLV Lake), groundwater fed only (BP Lake) and Meuse River from 2002 to 2014; (**b**) SO_4 concentration in BP Lake, divided into two periods with linear regression lines.

5. Discussion

5.1. Temperature

The mean temperature of BP Lake (12.3 ± 6.0 °C) was significantly lower than the temperature of Meuse River (14.0 ± 5.7 °C; Tables 2 and 3). The lower temperature is explained by the fact that BP Lake is fed mostly by groundwater. The mean water temperature of DLV Lake (13.3 ± 5.5 °C) was not significantly different from BP Lake or Meuse River, reflecting the input of both groundwater and River Meuse water. There were no significant trends over time in the temperature data presented here. Others have found that mean water temperatures of the Meuse River and other large north European rivers have increased from 1920 to 2000 and a decreasing temperature trend from 2000 onwards is recognized in the Meuse River [19]. The time period of our current data set could be not long enough to see the effect of increasing mean air temperatures on mean surface water temperatures or other characteristics of the time series data are affected by climate change, e.g., monthly mean values or seasonal mean values (see for example [20]).

5.2. pH

The mean pH of BP Lake (8.2 ± 0.1) water and DLV Lake (8.2 ± 0.2) are significantly higher than the mean pH of Meuse River water of 7.7 ± 0.1. There is no significant difference in mean pH between the two lakes. The values reported here confirm that the pH of gravel pit lakes studied so far is higher than that of typical natural lakes [6]. A likely explanation is the leaching of calcium rich sediments by groundwater upstream from the lakes [6]. The number of outliers (extreme values with residuals outside of (−1.96, 1.96)) was highest (8) for BP Lake and reflects the large seasonal fluctuations. These are common features in any lake and are related to variations in the CO_2(aq) and the lake food web caused by periods of rapid growth and uptake of CO_2(aq) by phytoplankton [21–23]. The seasonal peaks and lows of pH in DLV Lake are less distinct partly because Meuse River water with a highly variable pH and less distinct seasonal maximum and minimum values, is added to DLV Lake. The air blowers in the DLV Lake that operate from April to October explain the reduction of pH values in summer. The air blowers prevent stratification and warming up of the top layer, so that the rates of growth of phytoplankton are reduced. Therefore, the maximum pH from 2010 onwards (after the air blowers were put into action), is reached at the end of March after which the air blowers start working and the pH decreases. The peak pH in the BP Lake occurs at the end of summer when the productivity of the phytoplankton starts to decrease as the lake water begins to cool down. There are no significant linear trends over the monitoring period for pH in any of the data series.

5.3. NO_3

The mean NO_3 concentration of DLV Lake (6.2 ± 1.2 mg·L^{-1}) was significantly higher than that of BP Lake (0.6 ± 0.5 mg·L^{-1}) but lower than that of Meuse River water (14.8 ± 2.0 mg·L^{-1}). The AR of DLV Lake must have added large quantities of NO_3 because groundwater flowing into the lake contains very little NO_3 [11]. The number of outliers of NO_3 data is highest for the Meuse (20) and for DLV Lake (8) while they are zero for BP Lake (Table 3). There are a few extreme values, for example a very high NO_3 concentration of 12.8 mg·L^{-1} measured on 15 April 2008 in DLV or a very low concentration of 2.8 mg·L^{-1} in Meuse water on 30 March 2010. It is not known whether these values are real or due to sampling or laboratory errors. The hydrochemistry of Meuse river water is more subject to variation and extreme values as it receives groundwater and effluent water from three different countries and various industrial areas and is sensitive to the amount of precipitation in the watershed. Some, but not all of the variation in Meuse River water was transported to DLV Lake water especially at the beginning of the monitoring period: all outliers were early in the monitoring period (2003–2004). The linear trend over time is significant and negative for BP Lake and Meuse River. This confirms the generally observed decreasing NO_3 concentration in ground- and surface water with the enforcement of a more modest use of fertilizers by the EU recognized in most European watersheds [24–26]. However, in the DLV Lake the NO_3 concentration increased until 2010, because lake water was replaced more and more by river water. After 2009 there is no significant trend (Table 2) so it is unclear whether the NO_3 concentration in DLV Lake is still increasing or decreasing, or stable. Regardless, the change is not as rapid as it was before and it appears that phytoplankton activity and degassing currently compensate for additional input of NO_3. The seasonal fluctuations in NO_3 concentration for both lakes and the Meuse River can be related to agricultural practices and phytoplankton activity. Farmland fertilization usually occurs in late winter/early spring and that shows up as peaks of NO_3 concentration. The annual minimum concentration occurs in late summer when algae have consumed the NO_3 over the summer period.

5.4. PO_4

The mean PO_4 concentration was significantly different in all three waters: lowest in BP Lake (0.05 ± 0.03 mg·L^{-1}), almost twice as high in DLV Lake (0.09 ± 0.06 mg·L^{-1}) and the highest in Meuse River (0.77 ± 0.28 mg·L^{-1}). Most outliers (26) were in Meuse River water and only one in DLV Lake. Because of the high variability in the DLV Lake and Meuse River water, it is difficult to distinguish a seasonal maximum or minimum. The AR adds large amounts of PO_4 to DLV Lake but this does not lead to very high mean and increasing concentrations as it does for NO_3 (compare Figures 8 and 9). At least three processes play a role in P-cycling in lakes: (1) P is consumed by phytoplankton; (2) phosphor (P) may precipitate in lake sediments together with calcium (Ca^{2+}), metals (i.e., iron Fe^{2+}), and other (trace) elements such as nickel (Ni^{2+}) and arsenic (As^{2+}) [6,27]; (3) Internal loading of P by release from lake sediments [27–32]. Of these (1) and (2) are the most likely explanations for the disappearance of P from the water column in DLV Lake. Although the mean concentration of PO_4 in DLV Lake is much smaller than that of the Meuse River water, it is still higher by a factor of almost 2 than that of BP Lake. This is probably because aeration of DLV Lake water partly prevents algal growth that could eliminate more PO_4 from the water column. Previous studies on DLV Lake sediments showed that P in sediments is bound to Fe and Ca or present as organic P [11,28] but the risk for internal loading of DLV Lake with P is thought to be small because there is a large amount of dissolved Fe available that can bind P [28].

The trend of PO_4 concentration in BP Lake and Meuse River water over the monitoring period is significant and negative in agreement with observations that reflect a reduced use of fertilizers [26]. The PO_4 concentration in the DLV Lake does not show any significant trend over the monitoring period.

5.5. SO_4

The mean SO_4 concentration was higher in the lakes (DLV Lake 58.6 $\pm$ 4.3 mg·L^{-1}; BP Lake 76.4 $\pm$ 3.5 mg·L^{-1}) than in the Meuse River (47.6 $\pm$ 13.1 mg·L^{-1}) and all outlier values occurred in the Meuse River (Table 3). The artificially added Meuse River water dilutes the SO_4 concentration in DLV Lake. The trend over the whole period is negative for DLV Lake and Meuse River but is not significant for BP Lake. Instead if we divide the BP Lake data into two periods, there is a significant increase in SO_4 until 2009, after which there is a negative trend. Seasonal fluctuations of SO_4 concentration in the Meuse river water are very clear with highs in late fall and lows in early spring. They are possibly the result of the use of fertilizers, and pH regulators (Ammonium Sulphate) [33,34] but can also result from mineralization of S from the soil humus layer in the catchment, particular in periods with intense precipitation [35] or dissolution of evaporites [36]. Since the peak concentration of SO_4 in Meuse river water typically occurs in the fall, it is probably related to intense rainfall and biochemical processes in the soil rather than to fertilization [35]. The mean SO_4 concentration in DLV Lake and the Meuse River has a negative trend over the whole period. The SO_4 concentration in the BP Lake was higher than of DLV Lake and Meuse River water and initially (until about 2009) showed a slightly increasing trend. The fact that the SO_4 concentration in the BP Lake was higher than that of DLV Lake and Meuse River indicates that SO_4 was carried to the lake by groundwater. This, in fact, was measured within monitoring wells upstream from the gravel pit lakes and interpreted as the result of redox reactions in the soil [11,37].

5.6. Effect of Artificial Recharge with River Water and of Air Blowers and Implications for Drinking Water Quality

The AR with Meuse River water and aeration of DLV Lake is needed to produce large volumes of good quality drinking water. The effects of adding Meuse river water to DLV Lake water on the hydrochemistry are: (1) a change in lake water temperature; (2) an increase in mean NO_3 concentration; (3) an increase in mean PO_4 concentration; and (4) a decrease in mean SO_4 concentration of DLV Lake. The effects of air blowers in DLV Lake are: (1) internal mixing of the water which eliminates the typical summer lake stratification; (2) decreasing pH values from March through October; and (3) adding oxygen (air) to the water.

The addition of Meuse River water has a concentrating effect for NO_3 and PO_4, whereas it dilutes the concentration of SO_4. These and the other changes have secondary impacts on chemical cycling and ecological functioning of the lake. Some of these secondary effects are recognized. For example, adding dissolved oxygen to the lake water is beneficial for the precipitation of Fe, Aluminium (Al), Manganese (Mn), As, and other trace elements that flow in with groundwater [6,11]. Other effects still need to be studied. For example, what is the precise effect on an artificially reduced pH in summer on the ecological communities (besides algae) and chemical cycles? Almost all chemical processes and life forms are influenced by pH so an unnatural reduction in pH, changing the yearly cycle, must change those processes and the ecology to a certain extent.

The mean water temperatures have not significantly increased over the monitoring period, but they are likely to increase in the future as is observed for other lakes and this would affect the ecology [38,39]. Among others, the period of phytoplankton growth could lengthen. To maintain water quality in the DLV Lake, the air blowers would have to operate over longer periods to prevent stratification and eutrophication. However, if the air blowers operate longer this will mean that a smaller portion of the PO_4 and NO_3 coming in with river water will be consumed or broken down by algae and so their concentrations would increase in the DLV Lake. Previous studies on DLV Lake have shown that zooplankton and in particular mussels, have kept algal biomass low until 2010. The composition of algae and macrophytes communities, on the other hand, has indicated that the amount of available PO_4 in the water and lake sediments is increasing [40,41]. If PO_4 and NO_3 concentrations increase due to extended periods of operation of air blowers, this will have repercussions for the whole ecology of the lake as well as the hydrochemistry and chemistry of the lake sediments.

Climatic changes will eventually also have an effect on gravel pit lake hydrochemistry [6]. The more so because of the complicated feedback mechanisms at work. For example, a future decrease in mean annual precipitation could limit the influx of nutrients with groundwater into surface water [42]. This would cause a decrease in phytoplankton growth and less eutrophication, but could negatively affect the fixation of metals and potentially toxic trace elements in lake sediments [6,11,41]. On the other hand, intense rainstorms concentrated over shorter periods of time could cause the sudden input of PO_4, NO_3 and SO_4 towards lakes and rivers. These hydrological extremes can abruptly alter the properties of lake ecosystems [7].

The changing hydrochemistry of DLV Lake could have consequences for the water treatment process that is needed to make the water suitable for consumption. Water from DLV Lake passes through the lake bank downstream and through tens of meters of soil before reaching the pumping well and the treatment plant. This helps to attenuate strong seasonal variations in water quality, for example in (toxic) algae concentration due to biodegradation processes in the soil. However, bank filtration causes clogging related to redox reactions caused by the mixing of (sub)oxic lake water and anoxic groundwater [43]. Arsenic concentration is often a problem in bank filtration [44] but currently it is not a problem in DLV Lake [11]. However as nutrient loading from the watershed is decreasing over time as the decreasing NO_3 and PO_4 concentrations of BP Lake and Meuse River indicate, arsenic or other metal concentrations in water of DLV Lake could increase, as they are released by the lake sediments [7,45]. In that case additional water treatment could be needed [46]. Other metals (e.g., manganese) could become a problem by accumulating over time [11] needing special attention during treatment and transport of drinking water [47]. On the other hand the water treatment process itself may affect water quality, adding disinfection by products [48] or even affecting the bacterial antibiotic resistance [49]. Therefore prevention of eutrophication and diminishing surface water quality should have a preference over additional treatment [50].

The data presented in this paper shows that AR and operation of air blowers change the hydrochemistry of a lake. Therefore, the management of a lake used for AR and drinking water production is challenged to find the right balance between reduction of eutrophication and building up of nutrients and other chemical components in lake water.

6. Conclusions

We compared the means, the linear trends and seasonal patterns of temperature, pH, NO_3, PO_4 and SO_4 from 2003 to 2014 of a groundwater-fed gravel pit lake (BP Lake) with that of an artificially recharged gravel pit lake (DLV Lake) and Meuse River water (the water used for AR). The ANOVA and tests of difference indicate that the mean values of the NO_3, PO_4 and SO_4 concentrations over the period 2003–2014 are significantly different for all three types of surface water. Meuse River water contains the most NO_3, PO_4 and the least SO_4 and shows more outliers. DLV Lake contains on average more NO_3, PO_4 but less SO_4 than BP Lake. BP Lake (but not DLV Lake) is significantly colder than Meuse River. With respect to pH there is a significant difference between BP Lake (pH = 8.2) and the Meuse River (pH = 7.7), or between DLV Lake (pH = 8.2) and Meuse river, but not between the two lakes. We find that while BP Lake has a significantly lower temperature than Meuse River water, which is explained by ground water inflow, this difference is lost in DLV Lake. During summer, the pH in DLV Lake is lower than in BP Lake due to operation of the air blowers that limit algal growth. Linear regression analysis of the time series shows that NO_3 and PO_4 concentrations have decreased in BP Lake and Meuse River reflecting the diminished use of fertilizers. This trend is visible in DLV Lake only after 2010 as, at the beginning of the monitoring period (2003–2009), the addition of river water has increased NO_3 concentration. SO_4 concentration, on the other hand, has decreased in Meuse River and DLV Lake over the complete monitoring period whereas in BP Lake it increased until 2009 and decreased afterwards. The operation of the air blowers reduce the number of outliers in pH of

DLV Lake compared to BP Lake and the mixing of groundwater with river water in DLV Lake reduces the number of extreme values compared to Meuse River water of NO_3 and PO_4 and SO_4. A large part of the PO_4 added to DLV Lake with river water disappears from the water column by phytoplankton consumption and precipitation on the lake bottom but the concentration is still higher than that of BP Lake. The air blowers that prevent stratification and phytoplankton growth during the spring in the DLV Lake also prevent an increase in pH during spring-summer and the consumption of NO_3 and PO_4 by algae. In these two particular gravel pit lakes, the temperature does not reflect climatic changes but the use of the DLV Lake for AR has an impact on the seasonal and long-term trends in hydrochemistry. Management of a lake used for AR and drinking water production is challenged to find the right balance between control of eutrophication and building up of nutrients, SO_4 and metals in lake water and sediments.

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