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An Assessment of the Effectiveness of Riverbank Filtration in a Sewage Plant Effluent-Impacted River Using a Full-Scale Horizontal Well

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Citation: Noh, J.-H.; So, S.-H.; Park, J.-W.; Kim, S.-Y.; Song, K.-G.; Choi, J.; Kim, G.-B.; Son, H.; Kim, H.; Maeng, S.-K. An Assessment of the Effectiveness of Riverbank Filtration in a Sewage Plant Effluent-Impacted River Using a Full-Scale Horizontal Well. *Water* **2022**, *14*, 1873. https://doi.org/10.3390/w14121873

Academic Editor: Andrea G. Capodaglio

Received: 30 April 2022
Accepted: 7 June 2022
Published: 10 June 2022

Abstract: From 2014 to 2020, a full-scale horizontal well was operated to investigate the performance of full-scale riverbank filtration (RBF) in the Nakdong River in Korea, which is significantly impacted by the effluents from sewage treatment plants. In this study, an individual lateral full-scale horizontal collector well was investigated for the first time in Korea, and its performance was determined based on the turbidity and levels of iron, total nitrogen, dissolved organic matter, and four selected trace organic contaminants (TrOCs) (tebuconazole, hexaconazole, iprobenfos, and isoprothiolane) in the RBF and Nakdong River. The turbidity of the river was high with an average of 10.8 NTU, while that of the riverbank filtrate was 0.5 NTU or less on average. The average dissolved organic carbon (DOC) concentrations were 2.5 mg/L in the river water and 1.4 mg/L in the riverbank filtrate, which indicated a 44% reduction in DOC content during the RBF. Out of the 10 laterals, 8 laterals exhibited similar levels of iron, manganese, total nitrogen, DOC, and total hardness, electrical conductivity, and turbidity. The characteristics of the remaining two laterals were different. Because the groundwater inflow was relatively low (<10%), the laterals were contaminated by agricultural land use before the installation of the RBF. This is the first study to report changes in water quality according to individual laterals in a river affected by wastewater effluents. The filtration unit exhibited more than 90% removal rates for tebuconazole and hexaconazole. However, the removal rate for iprobenfos was approximately 77%, while that for isoprothiolane was 46%. The four selected TrOCs in this study were not detected in the groundwater. We found that some organic micropollutants were effectively removed by the RBF.

Keywords: riverbank filtration; dissolved organic matter; Nakdong river; trace organic contaminants

1. Introduction

Countries that rely on surface water for drinking have difficulty securing high-quality water resources due to excess nutrients, climate change, industrialization, and urbanization [1]. Among the trace organic contaminants (TrOCs) that have recently become an issue, refractory organic substances are not effectively removed from sewage treatment plants, and these substances can affect aquatic environments [2]. TrOCs exposed to aquatic environments can influence tap water quality directly or indirectly; therefore, further research is required [3]. Advanced treatment methods, such as advanced oxidation, granular activated carbon, and membrane filtration, are effective ways to remove TrOCs for drinking water production. Some advanced drinking water treatment processes are not sustainable, owing to their high energy consumption and unwanted transformation products. No single water treatment process can eliminate all contaminants, thus multibarrier water treatment is essential [4,5]. Water bodies affected by industrial wastewater effluent or sewage treatment plants have a high risk of contamination by TrOCs and chemical accidents. Therefore, eco-friendly, chemical-free, and sustainable water treatment techniques, such as riverbank filtration (RBF), are effective methods for multibarrier water treatment.

RBF has been used in Europe for more than 100 years, particularly in the Rhine, Donau, and Elbe rivers [6]. Some wells in Germany have been operating for more than a century [7]. Countries including Germany, Austria, Hungary, and the Netherlands have been successful in improving drinking water sources via RBF, artificial recharge, or both. RBF has been used for many years to improve drinking water quality and provide protection against water accidents, such as chemical spills. The RBF is an intake system for water that has traveled in an aquifer for a substantial amount of time, depending on the location of the production wells. It consists of river water flowing through the aquifer with some groundwater and can remove contaminants during soil passage [8]. By pumping surface water through vertical wells or horizontal collection wells in aquifers around rivers or lakes, it induces river and lake water into alluvial aquifer formations to improve the water quality. By maintaining a steady water temperature in the winter, the RBF prevents freezing in the supply pipe network. Furthermore, the use of coagulants and the cost of sludge treatment can be reduced by reducing the amount of suspended matter [9]. RBF provides high-quality filtrate via sorption, biodegradation, and groundwater mixing as the raw water passes through the aquifers. In an RBF, a portion of dissolved organic matter (DOM) is removed by the complex biofilm layer present on the upper part of the filtration layer (i.e., schmutzdecke), and biodegradation plays key role in improving the water quality [10,11]. RBF is a natural water treatment technology for the reduction in TrOCs levels, such as pharmaceutically active compounds [12]. However, a disadvantage of RBF is that the concentration of iron in the riverbank filtrate is usually high, and the produced sludge requires additional treatment at water treatment facilities, in case of further treatment applied. Moreover, local farmers in South Korea are concerned about the possible changes in groundwater levels and quality that could be caused by RBF. The contaminant removal efficiency of RBF is affected by various factors, such as the raw water quality, the soil characteristics of the aquifer, the distance between the river and intake well, and the dilution of the groundwater [13,14]. The RBF are functional in the United States and Europe; however, academic research and field studies in other countries are limited. Consequently, the process for importing these RBF is not well-established; hence, studies in other countries are required.

Natural organic matter (NOM) in aquatic environments combines with heavy metals and hydrophobic compounds, increasing the mobility of contaminants in aquatic environments and decreasing bioavailability [15]. It is also essential to understand the behavior of DOM in RBF because natural organic matter is a known precursor of disinfection by-products [14]. To understand the behavior of natural organic matter in an RBF, it is necessary to investigate the characteristics of the DOM in the riverbank filtrate. In many previous studies on RBF, the behavior of DOM has been investigated using column studies, but limited field studies have been done. Moreover, only a few reports have been published on the effects of TrOCs in filtrates of full-scale RBF in Asian countries.

Various water pollution sources threaten several cities downstream of the Nakdong River (Busan, South Korea). Few studies have investigated RBF, especially in areas highly affected by sewage treatment plant effluents, such as the Nakdong River. According to the Ministry of Environment, 1117 public sewage treatment plants and 58 public industrial wastewater treatment plants have been operating in the Nakdong River watershed as of 2019 [16]. Many industrial wastewater treatment facilities discharge effluents containing substantial amount of pollutants, such as TrOCs, into the Nakdong River. Currently, three drinking water treatment plants are using RBF as a pretreatment technology to provide water from the Nakdong River. The city of Changwon, which is on the Nakdong River in South Korea, has been providing 80,000 m^3/day of drinking water since 2006, using RBF with vertical and horizontal collector wells. This system was the first RBF site to be installed to supply drinking water in South Korea. Moreover, using the RBF of the Nakdong River, it is currently providing 127,000 m^3/day to Gimhae and 20,000 m^3/day to Haman-gun. However, limited research has been conducted to improve the water quality of RBF facilities.

In this study, we investigated the performance of an RBF affected by wastewater effluent and examined the characteristics of DOM using a full-scale horizontal collector well. This study also provides insights for water utilities interested in implementing RBF. This is the first time in Korea that water quality parameters have been compared to determine the significance of lateral location in a horizontal collector.

2. Materials and Methods

2.1. Full-Scale Horizontal Collector Well

This field study on a horizontal collector well will be used to design the remaining 9 horizontal collector wells that supply drinking water to the city of Busan. Before the installation of all 10 horizontal collector wells, which provide 280,000 m^3/day, a horizontal well was constructed and used to determine the performance of RBF. This study was conducted from October 2014 to September 2020, focusing on RBF via a horizontal collector well installed in Changnyeong-gun (Kyeongsangnam-do, South Korea), located downstream of the Nakdong River. Several observation wells had been installed and monitored before and after the collection well operation. The horizontal collector and monitoring wells selected for this study are shown in Figure 1. Since the operation of the horizontal collector well began in May 2015, the quality of the river water, riverbank filtrate, and 8 monitoring wells (OMW 1–8) were analyzed. All 8 monitoring wells were constructed to a depth of approximately 40 m and located in the horizontal and vertical directions of the stream flow (Figure 1). The RBF used in this study consisted of 10 laterals, which were installed at elevations of −20.5, −22, and −23 m from the surface for 3 laterals in the upper layer, 3 laterals in the middle layer, and 4 laterals in the lower layers. The diameter of the collecting pipe was 300 mm, and the total flow rate of the facility was 25,000 m^3/day.

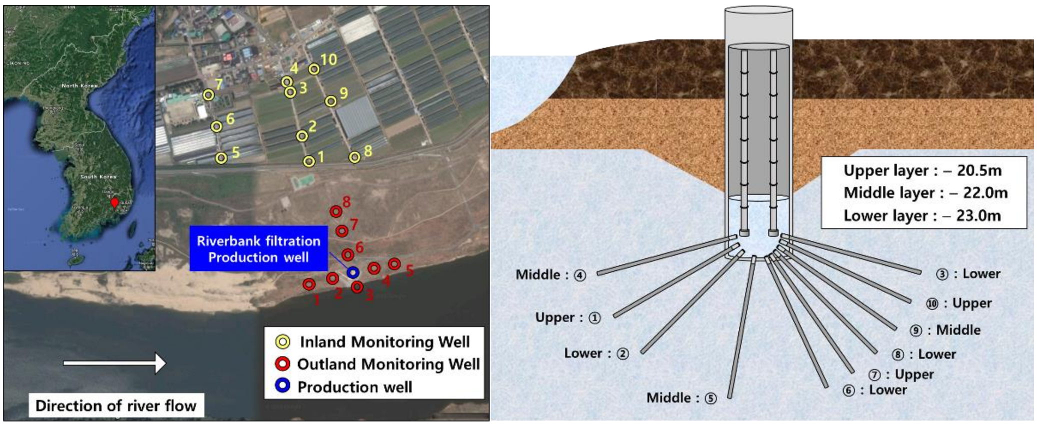

Figure 1. A horizontal collector well with 10 inland monitoring wells (IMW) and 8 outland monitoring wells (OMW) (Nakdong River, Korea).

2.2. Nitrogen and Phosphorus Content

Standard methods [17] were used to analyze the total nitrogen and ammonia (NH_4-N) content in the water. Chloride ions were analyzed using ion chromatography (ICS-900, Thermo Fisher Scientific Inc., Waltham, MA, USA).

2.3. Characteristics of Dissolved Organic Matter (DOM)

DOC level and ultraviolet absorbance at 254 nm (UV254) of the water samples were characterized; each sample was filtered using a 0.45 µm filter (Whatman, Kent, UK). The specific UV Absorbance (SUVA) value was obtained by dividing the UV254 value with the DOC value and multiplying the obtained value by 100. For the characterization of the DOM, the fluorescence excitation–emission matrix (F-EEM) was measured using an RF-5301 spectrofluorometer (Shimadzu, Japan). Four peak components were selected: T1 (tryptophan-like, ex/em: 220–240 nm/em: 330–360 nm), T2 (tyrosine-like, ex/em: 270–280 nm/em: 330–360 nm), A (fulvic-like, ex/em: 230–260 nm/400–450 nm), and C (humic-like, ex/em: 300–340 nm/400–450 nm) [5]. The organic matter was characterized according to molecular weight, and liquid chromatography-organic carbon detection (LC-OCD) analysis was performed using Model 8 (DOC Labor, Karlsruhe, Germany); the DOM fractions were separated into biopolymers (proteins and polysaccharides, 20 kDa or higher), humic substances (350–1000 Da), building blocks (300–500 Da), low-molecular-weight (LMW) (350 or lower) neutrals, and acids according to the retention time based on separation and molecular size [18].

2.4. Trace Organic Contaminants (TrOCs)

The organic micropollutants were analyzed using an online sample concentration method, which involved column switching as a sample pretreatment method. An Equan Max (Thermo Fisher Scientific Inc., Waltham, MA, USA) UPLC model was used for the analysis, which was equipped with a Hypersil Gold aQ (20 mm, 12 µm) concentration column and a Hypersil Gold C18 (50 mm, 1.9 µm) analytical column. The sample injection volume was set to 1 mL, and all the samples used for calibration and analysis were filtered using a 0.2 µm filter (Whatman, Kent, UK). Mass spectrometry was performed using the Orbitrap Exactive mass spectrometer (Thermo Fisher Scientific Inc., Bremen, Germany) model that used the high-resolution full scan method. The resolution and mass accuracy of the equipment were measured under the conditions of 50,000 and 5 ppm, respectively. After analyzing the equipment, a Quan Browser (version 2.1) was used for the qualitative and quantitative analyses.

3. Results and Discussion

3.1. Groundwater and Riverbank Filtration

To understand the contaminant removal efficiency of an RBF, it is necessary to determine the effects of groundwater dilution. In this study, the effects of groundwater were estimated using the levels of chloride and magnesium ions, electrical conductivity, and total dissolved solids of the ground water, and the mixing ratio of the river water and groundwater were calculated based on these values (Figure 2). The average groundwater inflow rates calculated based on the levels of chloride and magnesium ions were approximately 12% and 13%, respectively. Therefore, the ratio of river water in the riverbank filtrate was estimated to be 88%, which was relatively high. Additionally, the groundwater ratio in the riverbank filtrate was 12%, as per electrical conductivity, and 20%, as per the total dissolved solids, indicating that the groundwater ratios as per different parameters were within 20%. The average groundwater ratio in riverbank filtrate was approximately 14.2%, and the river water ratio in the riverbank filtrate was approximately 85.8%. From the previous study on RBF, the groundwater ratio in the riverbank filtrate was approximately 20–40% [19]. The reason behind the high ratio of river water in the riverbank filtrate was that the horizontal collector well was located close to the river and the 10 lateral wells were located below the river.

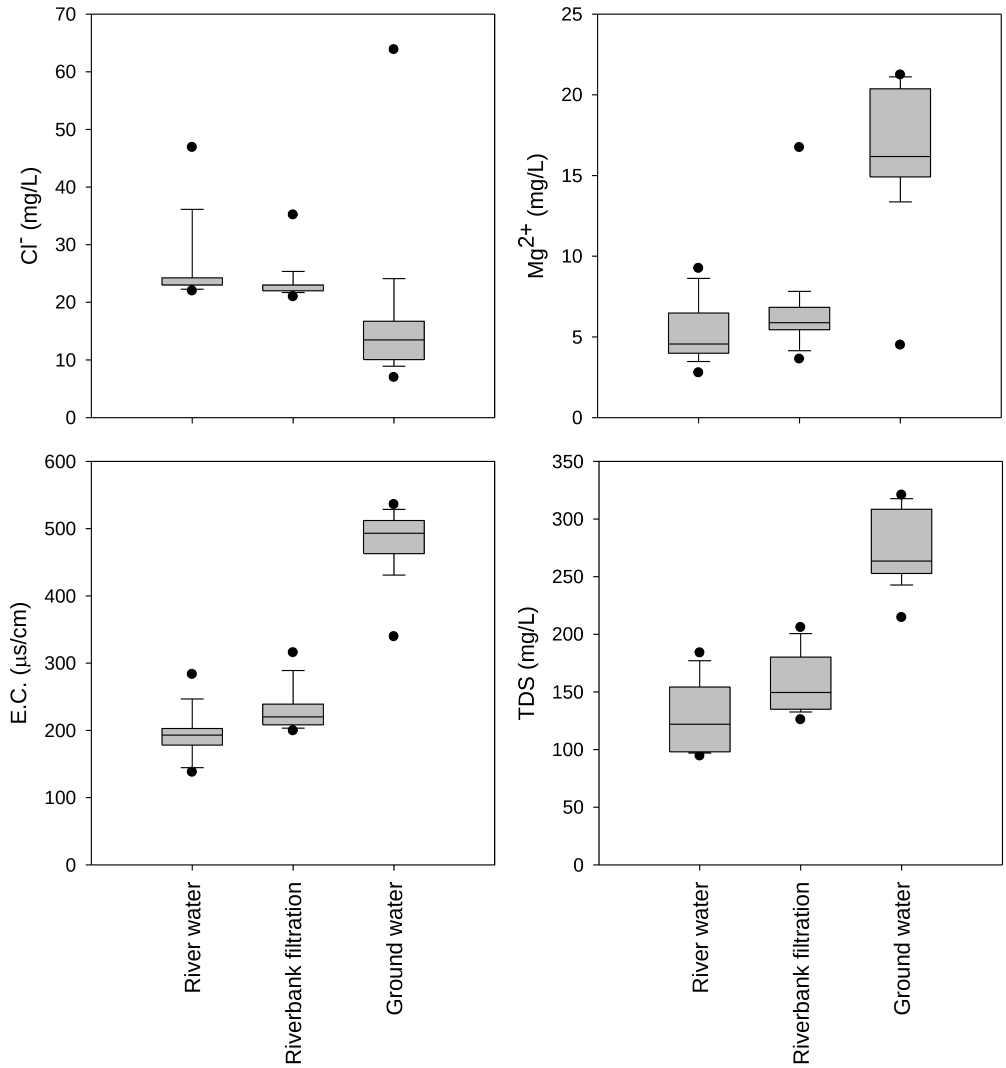

Figure 2. Concentration of Cl^- and Mg^{2+}, electrical conductivity (E.C.), and total dissolved solids (TDS) of the river water, riverbank filtrate, and groundwater (OMW 8G) (n = 20–32).

To confirm the effect of the RBF on the original groundwater quality, the change in the water quality of the groundwater observation well near the RBF facility was assessed (Figure 3). The concentration range of chloride ions was approximately 6–26 mg/L, which varied over time; however, a trend in the concentration was not observed before and after the operation of the RBF facility (Figure 3a). The electrical conductivity was not affected by the RBF, and no significant difference in the concentrations of magnesium and total dissolved solids in the groundwater of the collecting well during the operation period were observed (Figure 3b). Therefore, groundwater accounted for approximately 14% of the riverbank filtrate, and its effect on the original groundwater was not significant. The effect of riverbank filtrate on the quality of groundwater must be observed over a long period.

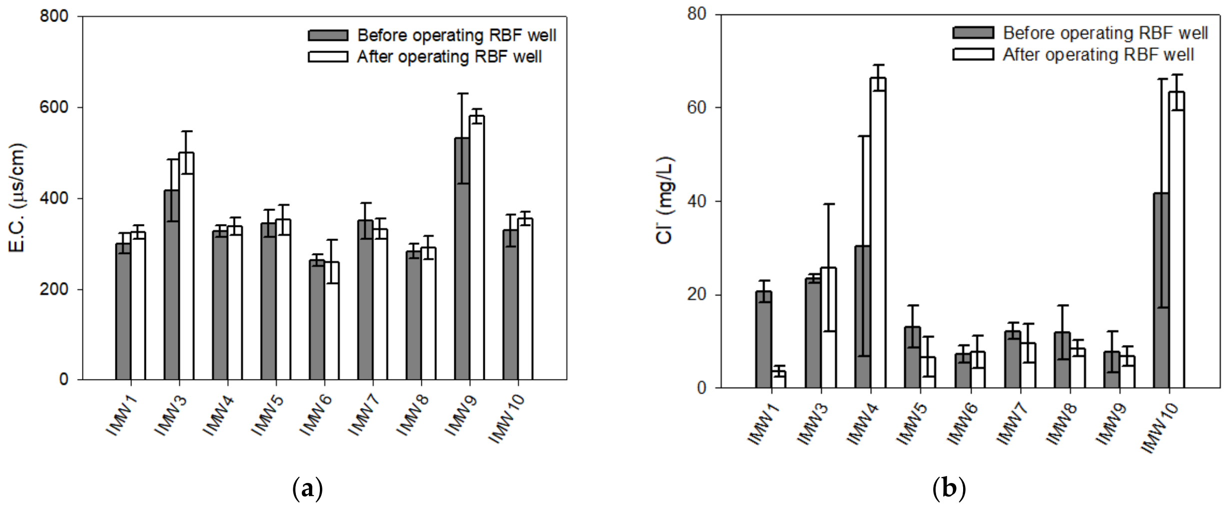

Figure 3. Concentration of chloride ions (Cl^{-}) (**a**) and electrical conductivity (E.C.). (**b**) in inland monitoring wells (IMWs) before and after the operation of a horizontal collector well.

3.2. Performance of a Horizontal Collector (Temperature, Turbidity, and Concentrations of Fe, Mn, and DOC)

While the temperature of the riverbank filtrate varied from 10 °C to 25 °C, the temperature of the river water ranged from 10 °C to 30 °C. The temperature changes in the riverbank filtrate were smaller because the aquifer temperature differed from that of the river. A small change in water temperature compared with that of river water was also confirmed in the monitoring wells, where no significant change in water temperature was observed. For the well OMW 8G, located 160 m from the horizontal well, which represented the groundwater, the temperature change was between 5 °C and 15 °C. The water temperatures of the monitoring wells parallel to the river were between 15 °C and 28 °C. Moreover, no significant difference in water temperature was observed even in the monitoring wells installed with sand and gravel layers. Therefore, the effect of temperature according to the location was greater than that of depth. In winter, the temperature of the riverbank filtrate did not change significantly, which could act as an advantage because the coagulants in the drinking water treatment plant can be easily controlled, and pipe freezing can be prevented.

The turbidity of the river water was higher than that of the riverbank filtrate, with average turbidity of river water and riverbank filtrate being 10.8 and 0.5 NTU, respectively. Additionally, even when the river water turbidity reached 100 NTU or higher, the turbidity of the riverbank filtrate remained fairly stable (e.g., 1 NTU or less). The change in turbidity in the horizontal collector well is presented in detail in Figure 4 and Section 3.3. The average concentration of iron in the river water was approximately 0.5 mg/L, and the concentration of manganese was exceptionally low, with an average of approximately 0.1 mg/L. However, the average iron concentration of the riverbank filtrate was 9.3 mg/L, and the average manganese concentration was 1.4 mg/L, which significantly increased during soil passage. An increase in iron and manganese concentrations in the riverbank filtrate was observed due to reducing conditions resulting from the biodegradation of organic matter, which requires electron acceptors (i.e., oxygen) [20]. Moreover, the iron content in the soil of this site was relatively high compared with that in other regions (data not shown). The concentrations of DOC in the river water and riverbank filtrate were 1.4 mg/L and 2.5 mg/L, respectively, with an organic matter reduction rate of 44%. The reduction in the DOM content due to RBF is known to occur predominantly by biodegradation and adsorption. Dissolved oxygen (DO) levels during RBF decreased significantly from the river, 9.9 mg/L to RBF > 1 mg/L, indicating that biodegradation played an important role in pollutant removal. The DOC of the OMW 3 and OMW 6 wells was similar to that of the riverbank filtrate. OMW 7 and OMW 8G had average DOC concentrations from 4 to 6 mg/L, which were higher than that of the river water (2.8 mg/L). The monitoring wells OMW 7 and 8G represented the groundwater instead of the river water. Because OMG 8G well was located near agricultural land, the risk of increase in DOC level existed.

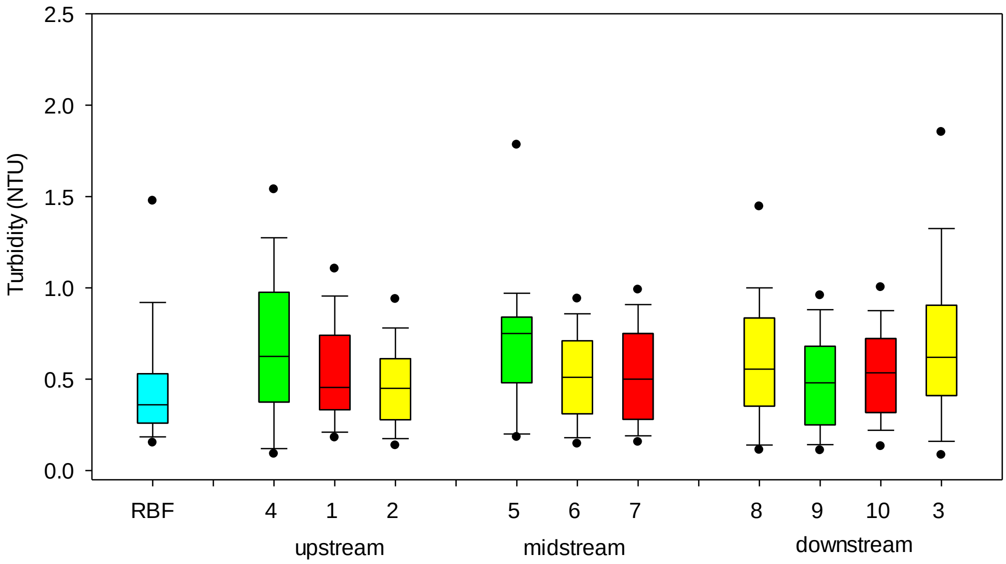

Figure 4. Turbidity changes in a collector well and 10 laterals.

3.3. Performance of Laterals (Temperature, Turbidity, and Concentrations of Fe, Mn, and DOC)

We collected samples with a 1 L bailer from 10 laterals below the water table in monitoring wells to observe how the water quality differed according to the location of each lateral in the horizontal collector well. The temperature changes were not significant, and the turbidity results are shown in Figure 4, which includes the production well and 10 laterals. The turbidity of the river water was high at approximately 11 NTU. The riverbank filtrate had an average turbidity of only 0.5 NTU, which was significantly lower than that of river water, and was within the range of 1 NTU or less for each lateral.

Figure 5 presents the concentrations of iron, manganese, and total nitrogen, total hardness, electrical conductivity, and DOC for each lateral. The iron and manganese concentrations in the individual laterals varied based on their location. The iron concentration was high in lateral #4, and the manganese concentrations were high in laterals #9, #10, and #3. In the riverbank filtrate, an increase in the concentration of iron and manganese was observed, which could be due to reducing conditions; however, the concentrations increased in the lateral direction, where the concentration was measured only in a specific location; this observation could be due to the geological characteristics of the area and not the river water. Lateral #4, which had high levels of iron, exhibited DOC and nitrogen levels different from those in the river water and other laterals. In addition to the effects of upstream and middle streams, the effect of fertilizers accumulated on the land could affect the riverbank filtrate from lateral #4 because that specific location was used as an agricultural land.

In general, when the hardness of the riverbank filtrate is high, the inflow rate of groundwater is expected to be high; however, in this study, the hardness of the riverbank filtrate was not significantly different from that of river water. Owing to the geological characteristics of the target area and the location of the horizontal collector well, the riverbank filtrate consisted of a significant proportion of surface water. The total nitrogen levels in laterals #3 and #4 were slightly higher than those of iron, manganese, and hardness detected in other laterals. The total nitrogen levels in laterals #5, #6, and #7, located close to the river, were lower than those in other pipes, and the ammonia nitrogen levels were more than twice those found in other laterals. The total nitrogen levels in laterals #5 and #6 were also lower than those in other laterals by approximately 60–88%.

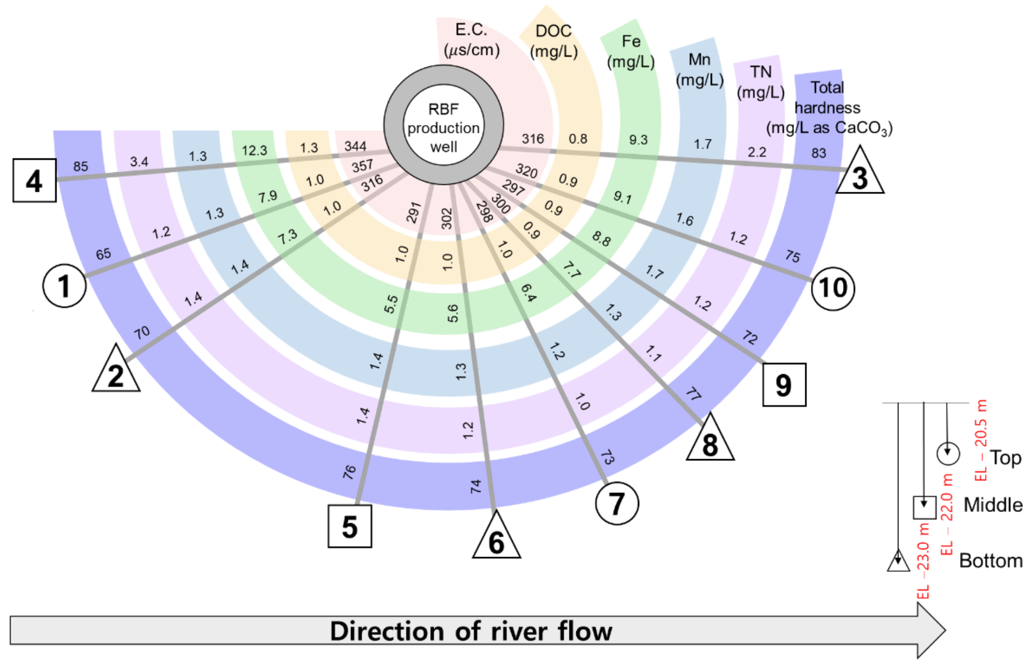

Figure 5. The water quality parameters of lateral collecting pipes (n = 41–192).

After RBF, it was confirmed that the average DOC concentration was reduced by 43.6%. All laterals, except for lateral #4, showed similar DOC removal rates. The total nitrogen level detected in all the laterals except for #3 and #4 appeared to be similar. This could be the result of agricultural activity in the regions close to laterals #3 and #4. The water quality of the laterals connected to the horizontal collector well in this study varied slightly according to the location of the laterals (Figure 5). To improve the water quality of the riverbank filtrate, reduction in the flow rates of laterals #3 and #4 was considered. As a result of this study, the change in the water quality characteristics of the laterals in the RBF facility was examined with respect to the depth of the laterals. It is expected that these results will be useful for water utilities where RBF facilities are installed.

3.4. Characteristics of Dissolved Organic Matter (DOM) (LC-OCD and F-EEM)

LC-OCD was used to analyze the characteristics of organic matter according to its molecular weight in the riverbank filtrate (Figure 6). In the groundwater (OMW 8G), humic substances comprised 64% of the DOM, whereas biopolymers comprised 1% compared to those in the river water. In the groundwater, humic substances made up the majority of the DOM, whereas biopolymers were relatively less abundant in comparison to their abundance in the river water. The biodegradable organic matter content in the groundwater was low. The biopolymers in the river water in this study were measured with an OCD detector, as less amount was detected with a UVD detector. During RBF, biopolymers (97%), humic substances (70%), and building blocks (30%) were removed from the river water, indicating that organic substances with relatively large molecular weights were removed preferentially. Furthermore, because biopolymers such as proteins and polysaccharides can be removed using biological filtration methods [21,22], the hydrophilic biopolymers in the river water can be also removed using biological filtration in RBF, and similar findings have been reported in previous studies on RBF [10,20,23].

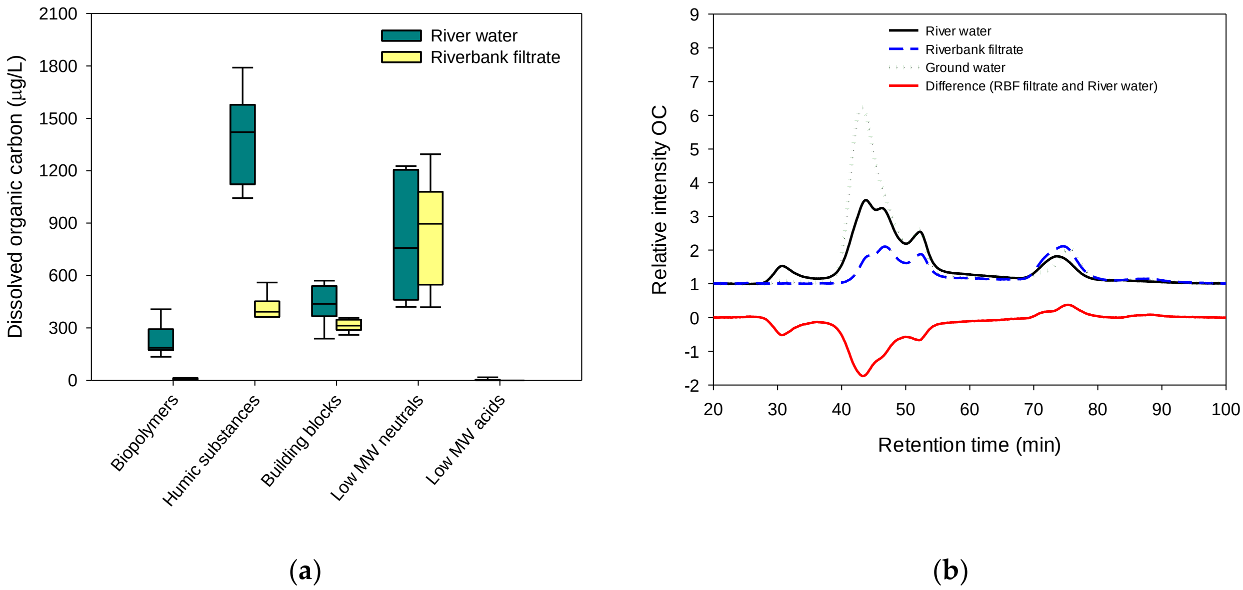

Figure 6. Changes in dissolved organic matter fractions determined by LC−OCD (biopolymers, humic substances, building blocks, low molecular weight (MW) neutrals, and low MW acids) (**a**) (n = 9), and (**b**) LC−OCD chromatograms of the river water, riverbank filtrate, and groundwater (OMW 8G) (n = 7).

The fluorescence characteristics of the DOM were used to compare the results of the F-EEM analyses (Figure 7). The fluorescence intensity in the river water was characterized by four peak regions, which were divided into T1 and T2 regions, with tryptophan- and protein-like characteristics, respectively, and A and C regions, with humic-like characteristics [5,24,25]. The fluorescence intensity of the river water was high in T1 (1182 ± 546), T2 (748 ± 377), A (639 ± 150), and C (418 ± 128), whereas those of the groundwater were high in A (2784 ± 576), C (1760 ± 387), T1 (965 ± 201), and T2 (593 ± 133). Compared with the groundwater, the river water contained more humic-like substances, and protein-like substances were also prevalent. According to the F-EEM analysis of the riverbank filtrate, its fluorescence intensity was weaker than that of the river water and groundwater, and fluorescence was detected near T1 and T2, suggesting that it was affected by the river water rather than the groundwater with relatively high A and C fluorescence. Similar reductions in the fluorescence intensity were observed for the four peak regions by approximately 36 to 42%, and the result was consistent with the DOC removal rate of approximately 42%. Although the results of the F-EEM analyses were difficult to compare quantitatively with DOC removal rate, it was possible to understand the characteristics of DOM in the RBF. This finding was also consistent with the LC-OCD results, which identified biopolymers in the river water and high levels of humic substances in the groundwater.

3.5. Trace Organic Contaminants (TrOCs) (Tebuconazole, Hexaconazole, Iprobenfos, and Isoprothiolane)

The removal rates of the RBF for the selected TrOCs (tebuconazole, hexaconazole, iprobenfos, and isoprothiolane) were investigated. The removal rates were 90% for tebuconazole and hexaconazole, 77% for iprobenfos, and 46% for isoprothiolane. Because none of the substances were detected in the groundwater, and the dilution rate of the groundwater was not significant, a significant portion of the selected TrOCs was removed from the riverbank filtrate. The physicochemical properties of the TrOCs (i.e., pesticides) selected in this study indicate that they were hydrophobic and were removed by soil adsorption (Table 1). When filtered through an RBF, even if these TrOCs are easy to remove, their removal can increase the effectiveness of activated carbon in drinking water treatment plants. Consequently, RBF removes pollutants through biodegradation and adsorption, which makes it an effective method for treating water to remove TrOCs.

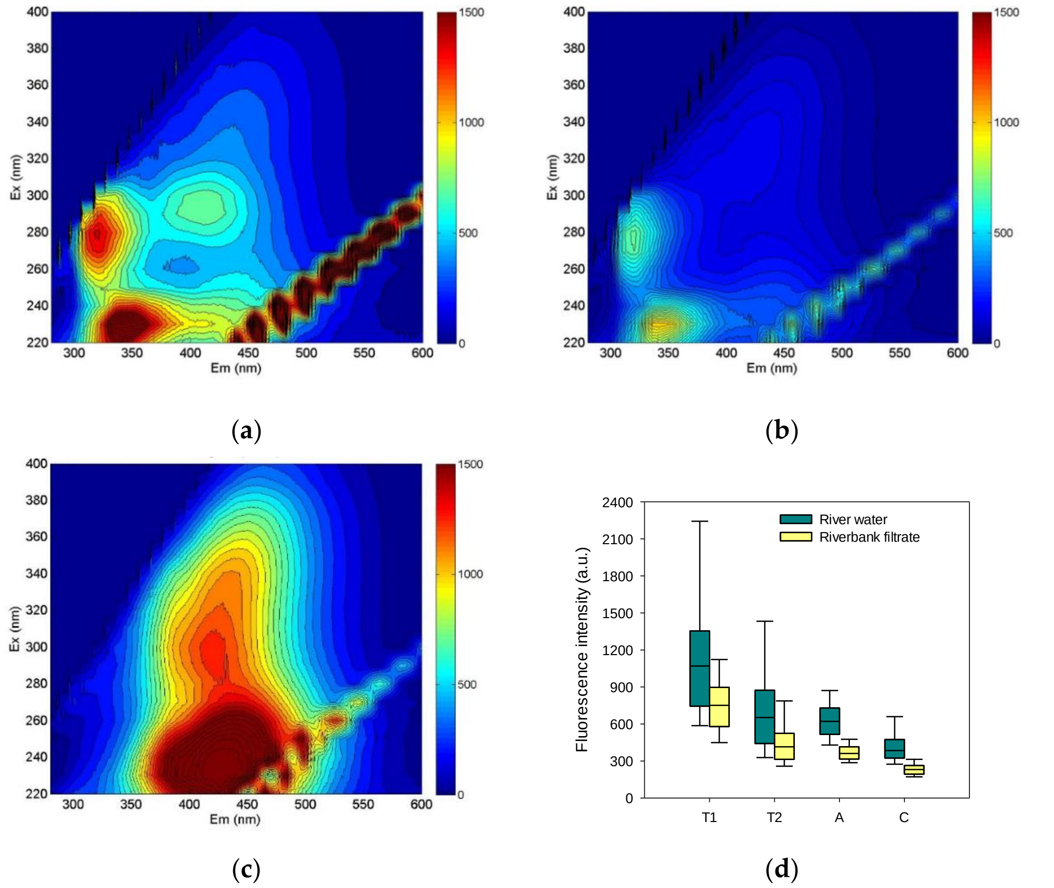

Figure 7. Fluorescence excitation–emission matrix (F-EEM) spectroscopy contour plots of river water (**a**), riverbank filtrate (**b**), groundwater (OMW 8G) (**c**), and changes in the fluorescence intensities in selected regions (T1, T2. A, C peaks) in the river water and riverbank filtrate (**d**) (n = 12).

Table 1. Physicochemical properties of selected trace organic contaminants.

Compound	MW (g/mol)	CAS# [1]	pK a [2]	log Kow [3] (pH = 8)	logD [4] (pH = 7.4)	Classification @ pH = 8 [5]	Use
Hexaconazole	314.21	79983-71-4	2.3	3.66	3.63	Hydrophobic–Ionic	Fungicide
Iprobenfos	288.34	26087-47-8	−8.2	3.57	3.21	Hydrophobic–Neutral	Fungicide
Isoprothiolane	290.40	50512-35-1	−7	2.79	3.44	Hydrophobic–Neutral	Fungicide
Tebuconazole	307.82	107534-96-3	2.3	3.89	3.74	Hydrophobic–Ionic	Fungicide

[1] Chemical abstracts service registry numbers. [2] pKa calculated from SPARC. Available online: http://www.archemcalc.com/sparc.html (accessed on 4 May 2021). [3] log Kow value reported for neutral molecule form; based upon U.S. environmental protection agency, 2005, log Kow calculation, EPI Suite KOWWIN Program. Available online: https://www.epa.gov/tsca-screening-tools/epi-suitetm-estimation-program-interface (accessed on 9 June 2022). [4] Chemspider search and share chemistry. Available online: http://www.chemspider.com (accessed on 10 April 2021). [5] For acidic pharmaceuticals: hydrophobic, log D > 1; hydrophilic, log D < 1 at pH 7.4; for neutral pharmaceuticals: hydrophobic, log Kow > 2; hydrophilic, log Kow < 2.

4. Conclusions

LC-OCD and F-EEM analyses of water from deep observation wells confirmed that the groundwater contained humic-like DOM of soil origin. Water quality analyses of the 10 laterals installed in the collector well of RBF revealed that the water quality trends of 8 laterals were similar. The water quality of laterals #3 and # 4 was poor compared with that of the other laterals. It is necessary to examine various possibilities regarding the water quality of laterals #3 and #4, such as whether the aquifer characteristics differ from those of other areas or whether agricultural activity was carried out in the area. To improve the quality of the riverbank filtrate, it is necessary to reduce the flow rates of lateral #3 and

#4. Based on these results, it can be concluded that controlling the lateral valve has the potential to change the water quality of the final riverbank filtrate. This study demonstrates that, in the future, in the horizontal collection well of the riverbank filtrate, it is necessary to conduct a detailed geological investigation into the water quality changes according to the location of the collection pipe and the target area.

Author Contributions: Conceptualization, S.-K.M. and K.-G.S.; methodology, S.-H.S.; formal analysis, J.C., H.S. and H.K.; investigation, J.-H.N. and J.-W.P.; resources, G.-B.K.; writing—original draft preparation, J.-H.N. and S.-K.M.; writing—review and editing, S.-K.M. and S.-Y.K.; funding acquisition, G.-B.K. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Korea Ministry of the Environment (MOE) and the Korea Environmental Industry and Technology Institute (KEITI) as "Demand Responsive Water Supply Service Program (#146523)".

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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