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Prediction and Remediation of Groundwater Pollution in a Dynamic and Complex Hydrologic Environment of an Illegal Waste Dumping Site

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Abstract: The characteristics of groundwater pollution caused by illegal waste dumping and methods for predicting and remediating it are still poorly understood. Serious 1,4-dioxane groundwater pollution—which has multiple sources—has been occurring at an illegal waste dumping site in the Tohoku region of Japan. So far, anti-pollution countermeasures have been taken including the installation of an impermeable wall and the excavation of soils and waste as well as the monitoring of contamination concentrations. The objective of this numerical study was to clarify the possibility of predicting pollutant transport in such dynamic and complex hydrologic environments, and to investigate the characteristics of pollutant transport under both naturally occurring and artificially induced groundwater flow (i.e., pumping for remediation). We first tried to reproduce the changes in 1,4-dioxane concentrations in groundwater observed in monitoring wells using a quasi-3D flow and transport simulation considering the multiple sources and spatiotemporal changes in hydrologic conditions. Consequently, we were able to reproduce the long-term trends of concentration changes in each monitoring well. With the predicted pollutant distribution, we conducted simulations for remediation such as pollutant removal using pumping wells. The results of the prediction and remediation simulations revealed the highly complex nature of 1,4-dioxane transport in the dumping site under both naturally occurring and artificially induced groundwater flows. The present study suggests possibilities for the prediction and remediation of pollution at illegal waste dumping sites, but further extensive studies are encouraged for better prediction and remediation.

Keywords: 1,4-dioxane; dynamic; pollution; dumping site; transport phenomena

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1. Introduction

Many areas around the world still rely heavily on groundwater for daily water consumption. Thus, the maintenance of a suitable groundwater quality is crucial [1–5]. During the 2000s, however, various human activities significantly impacted groundwater quality and availability through various forms of pollution [4], with landfills, mines, and industrial plants being some of the main sources of groundwater pollutants [6,7]. The most common pollutants include heavy metals, non-aqueous phase liquids (NAPLs), and volatile organic compounds (VOCs) [8,9]. These pollutants are hazardous chemicals that can potentially negatively impact human health. Unfortunately, pollution from illegal waste dumping sites also occurs, causing serious health and environmental problems [10,11]. Previous studies have demonstrated that the respiratory exposure of VOCs via the inhalation route is associated with the risk of specific diseases [12]. However, the characteristics of pollution caused by illegal waste dumping as well as ways of predicting and addressing the problem are still poorly understood. This limited understanding is due to the dynamic and highly complex nature of pollution caused by illegal waste dumping, and the complexity originates from multiple pollution sources and the artificially induced pollution-prevention changes (e.g., excavation and pumping) in hydrologic environments.

At illegal waste dumping sites, pollutants leak from the waste to the groundwater over a long period of time. At the time of discovery, the distribution of contamination may be widespread, requiring countermeasures such as monitoring and remediation, which generally require considerable time and expense [13–15]. Consequently, it would be desirable to understand the characteristics of pollutant transport and to predict it in dynamic and complex hydrologic environments in illegal waste dumping sites [16]. For this purpose, simulations of pollutant transport in the presence of groundwater flow require consideration of the characteristic history of the illegal waste dumping site in addition to the conventionally considered subsurface flow and transport properties [17,18]. However, such simulations have not been conducted yet.

In the prefecture of the Tohoku region, Japan, an illegal dumping site was discovered in 1990, with various countermeasures having been implemented to date (2021) due to the existence of 1,4-dioxane groundwater pollution—a regulated substance (groundwater standard: $\leq$0.05 mg/L) [19–23]. At this site, multiple pollutant sources have been expected based on data from monitoring wells, and multiple countermeasures (e.g., the installation of impermeable walls) that potentially impact the hydrologic environment have been implemented. 1,4-Dioxane is highly miscible with water, and its biodegradation and adsorption to soil may be neglected [24–26], making this site suitable for fundamental studies on pollutant transport in the dynamic and complex hydrologic environments of illegal waste dumping sites. In this context, the objective of this study was to clarify the possibility of numerically predicting pollutant transport in the dynamic and complex hydrologic environments at this site in Japan, and to investigate the characteristics of pollutant transport under both naturally occurring and artificially induced groundwater flow using various countermeasures including remediation with pumping.

2. Materials and Methods

2.1. Site Information

History of Site Modification and Monitoring 1,4-dioxane

The illegal waste dumping site is located in the Tohoku region of Japan, Figure 1 shows a map of the site. The site covers an area of approximately 0.16 square kilometers and has illegally collected and buried industrial waste for decades. Approximately 270,000 m^3 of waste has been dumped at this site including incinerator ash, sludge, and refuse-derived fuel materials, which has caused the spread of serious contaminants in both the soil and groundwater.

Table 1 shows the history of the illegal waste dumping. A company started illegally dumping waste in the 1990s, and when the local government in one of Japan's prefectures discovered this, they immediately conducted a site survey. The prefectural government conducted an initial field survey in 2000, and for 10 years (from 2004 to 2014), it conducted additional surveys, observed groundwater contamination, and cleaned up the buried waste. To prevent groundwater from the waste burial site in one prefecture from contaminating the neighboring prefecture, a barrier wall (i.e., impermeable wall) was installed along the prefectural border (between 2005 and 2007). In 2009, the municipality implemented a groundwater treatment plant, installing pumping wells along the barrier wall in 2010. Moreover, the municipality monitored and documented 1,4-dioxane contamination from 2013 to 2020, as 1,4-dioxane had been detected in monitoring wells installed on the site.

This study focused on an area of 500 $\times$ 500 m for this site, as shown in Figure 1. For this area, the elevation and groundwater levels were obtained from a Geological Information System (GIS) and site investigation data [27]. There is a groundwater divide in this area, which divides groundwater flow into two major directions. The groundwater level was used to determine the flow potential in the governing equations for subsurface flows as described below.

Table 1. History of site modifications (well installation and monitoring period).

Year	Events
1990	Started illegally dumping industrial waste
2000	Started removing waste
2005–2007	Installed impermeable wall
2009	Implemented a groundwater treatment plant
2010	Installed pumping well
2013	Started monitoring of 1,4-dioxine (Under survey (2021))
2014	Finished removing waste

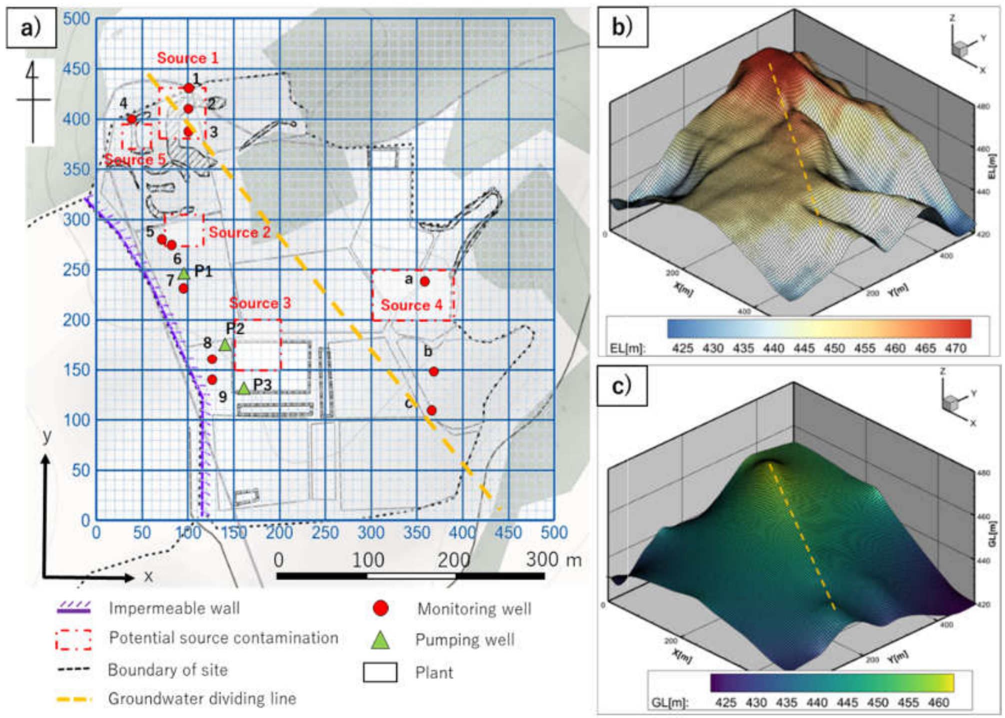

Figure 1. Map of an illegal waste dumping site. The blue grid means the calculated area. (**a**) Site map and simulation area. (**b**) Distribution of elevation level (EL) from a GIS. (**c**) Distribution of the groundwater level (GL). The EL and GL is the distance from the sea surface.

2.2. Flow and Transport Simulation

2.2.1. Governing Equation

In this study, the aquifer was assumed to be a single layer with a thickness of 5 m because it was difficult to obtain a detailed geological cross section of the aquifer. The flow in the aquifer was modeled as a 2D two-phase flow of water and non-aqueous phase liquid (NAPL) in the x-y coordinates, where NAPL is 1,4-dioxane [27]. However, the flow potential of the model takes into account the groundwater level (i.e., difference between land surface elevation and depth to water). As a result, the model is a quasi 3D model. The governing equations for the flows for NAPL and groundwater with/without dissolved NAPL and the advection-diffusion equation for NAPL in water are respectively represented by Equations (1)–(3), as follows.

$$\frac{\partial}{\partial x}\left(\frac{Kk_{ro}}{\mu_o}\rho_o\frac{\partial}{\partial x}\Phi_o\right)+\frac{\partial}{\partial y}\left(\frac{Kk_{ro}}{\mu_o}\rho_o\frac{\partial}{\partial y}\Phi_o\right)=\frac{\partial(\varnothing\rho_o S_o)}{\partial t} \quad (1)$$

$$\frac{\partial}{\partial x}\left(\frac{Kk_{rw}}{\mu_w}\rho_w\frac{\partial}{\partial x}\Phi_w\right)+\frac{\partial}{\partial y}\left(\frac{Kk_{rw}}{\mu_w}\rho_w\frac{\partial}{\partial y}\Phi_w\right)=\frac{\partial(\varnothing\rho_w S_w)}{\partial t} \quad (2)$$

$$\begin{aligned} \frac{\partial}{\partial x}\left(x_{ocw,k}\frac{Kk_{rw}}{\mu_w}\rho_w\frac{\partial}{\partial x}\Phi_w\right) \quad & +\frac{\partial}{\partial y}\left(x_{ocw,k}\frac{Kk_{rw}}{\mu_w}\rho_w\frac{\partial}{\partial y}\Phi_w\right) \\ & +\frac{\partial}{\partial x}\left[D_{ocw,k}\frac{\partial}{\partial x}\left(x_{ocw,k}\varphi\rho_w S_w\right)\right] \\ & +\frac{\partial}{\partial y}\left[D_{ocw,k}\frac{\partial}{\partial y}\left(x_{ocw,k}\varphi\rho_w S_w\right)\right] = \frac{\partial\left(x_{ocw,k}\varphi\rho_w S_w\right)}{\partial t} \end{aligned} \quad (3)$$

where K is the absolute permeability (m^2); k_{ro} is the relative permeability for NAPL (fraction) [28–30]; k_{rw} is the relative permeability for water (fraction) [28–30]; μ_o is the NAPL viscosity (Pa·s); μ_w is the water viscosity (Pa·s); ρ_o is the molar density of NAPL (mol/m^3); ρ_w is the molar density of water (mol/m^3); Φ_o is the flow potential of NAPL (Pa); Φ_w is the flow potential of water (Pa); S_o is the NAPL saturation (fraction); S_w is the water saturation (fraction); φ is the porosity (fraction); $D_{ocw,k}$ is the diffusion coefficient (m^2/s); $x_{ocw,k}$ is the concertation of NAPL dissolved in water (fraction); and t is time (s). The parameter values used in this study are listed in Table 2. The parameter values for the groundwater layer were set by assuming a clay soil, and the parameter values for 1,4-dioxane were acquired from a database [31–33]. Note that the biodegradation and adsorption to soil for 1,4-dioxane were neglected based on the chemical properties of 1,4-dioxane [34].

Table 2. Basic 1,4-dioxane information and geotechnical information from the site.

Symbol	Parameter	Unit	Input Values	Reference
K	Permeability	m^2	1.4×10^{-12}	[35]
φ	Porosity	-	0.3	
μ_0	Viscosity of 1,4-dioxane	Pa·s	1.31	
μ_w	Viscosity of water	Pa·s	1.138	
ρ_0	Molar density of 1,4-dioxane	$kmol/m^3$	18.36	
ρ_w	Molar density of water	$kmol/m^3$	17.83	
$D_{ocw,k}$	Molecular diffusion coefficient	m/s	1.0×10^{-9}	

These governing equations were solved using the finite difference method by applying the implicit pressure explicit saturation solution method [36] for implicit solutions for pressure and explicit solutions for saturation and concentration. To solve the governing equations, the 500 × 500 m area shown in Figure 1 was divided into a 5 × 5 m grid, and a hydrologically opened boundary condition was applied for each 500 m side and a constant flow-rate boundary for each pumping well. The water level was kept constant at the hydrologically opened boundaries. No detailed geological data were available for the sites targeted in this study. Therefore, the layer was assumed to be a single layer with the parameters used in Table 2.

2.2.2. Prediction and Remediation Simulations

The prediction simulation computed the evolution of the groundwater flow and the concentration of 1,4-dioxane from their initial conditions based on the history of the illegal waste dumping site (Table 1). The initial groundwater flow was determined based on the distribution of the groundwater level (GL) (Figure 1). The initial concentrations of 1,4-dioxane for the assumed contamination source areas 1–5, shown in Figure 1, were obtained by applying a constant annual input rate of 1,4-dioxane from the waste/contaminated soils for each grid within each source area, whereas the initial concentration was set to zero for the other locations. The input rate was set to zero to simulate the removal of waste/contaminated soils after excavation. The total input volume of 1,4-dioxane for each area is listed in Table 3, which indicates that Source 1 is where the largest amount of 1,4-dioxane was discarded. To simulate the permeability changes due to the installation of the impermeable wall, the absolute permeability at the corresponding location after the wall installation was changed to 1.00×10^{-15} m^2. Additionally, when monitoring wells were added, the corresponding grids were changed to a constant flow-rate boundary at a prescribed discharge rate (pumping rate). More specifically, the pumping rates were 10 m^3/day for pumping well 1, 31 m^3/day for pumping well 2, and 23 m^3/day for pumping well 3, respectively [37].

Table 3. Assumed sources of contamination at the site.

Name	Pollution Area [m^2]	Amount of Source [kg]
Source 1	2500	600
Source 2	1500	20
Source 3	2500	30
Source 4	4500	15
Source 5	400	30

These values were assumed based on fitting by the model.

In the remediation simulation, the 40-year evolution of groundwater flow and the concentrations of 1,4-dioxane from the final conditions in the prediction simulation were computed. We conducted two types of remediation simulations considering passive and active treatments with pumping [37,38]. The concentration of 1,4-dioxane in the studied area was expected to decrease during the passive and active treatments due to dilution and removal, respectively, by naturally occurring and pumping-induced groundwater. In the remediation simulation with active treatment, in addition to preexisting pumping wells 1–3, the monitoring wells were used as new pumping wells at a constant pumping rate of 250 m^3/day. Table 4 summarizes the pumping rates of each well.

Table 4. Pump specifications by active treatment.

Name	Symbol in Figure 1	Pumping Rate [m^3/day]	Reference
Monitoring well 1	1	250	[19]
Monitoring well 2	2	250	
Monitoring well 3	3	250	
Monitoring well 4	4	250	
Monitoring well 5	5	250	
Monitoring well 6	6	250	
Monitoring well 7	7	250	
Monitoring well 8	8	250	
Monitoring well 9	9	250	
Monitoring well a	a	250	
Monitoring well b	b	250	
Monitoring well c	c	250	
Pumping well 1	P1	10	[35]
Pumping well 2	P2	31	
Pumping well 3	P3	23	

3. Results and Discussion

3.1. Reproduction of Concentration of 1,4-Dioxane Groundwater Pollution in Monitoring Well and Distribution Prediction

A comparison between the reproduced 1,4-dioxane concentrations in each monitoring well using a model that considered groundwater flow (Figure 1) and the field monitoring data is shown in Figure 2. Between 2013 and 2017 (4 years), the reproduced data and monitoring data for all monitoring wells generally exhibited good fitting results. In 2013, both reproduced data and observed data showed relatively higher 1,4-dioxane concentrations in monitoring wells 1, 2, and 3 for Source 1, and monitoring well 6 for Source 2 than in the other wells. Significant decreases in 1,4-dioxane concentration were observed from 2014, when the excavation was completed. The good fitting results suggest that predicting the 1,4-dioxane concentrations by considering groundwater flow is possible due to its water-soluble characteristics.

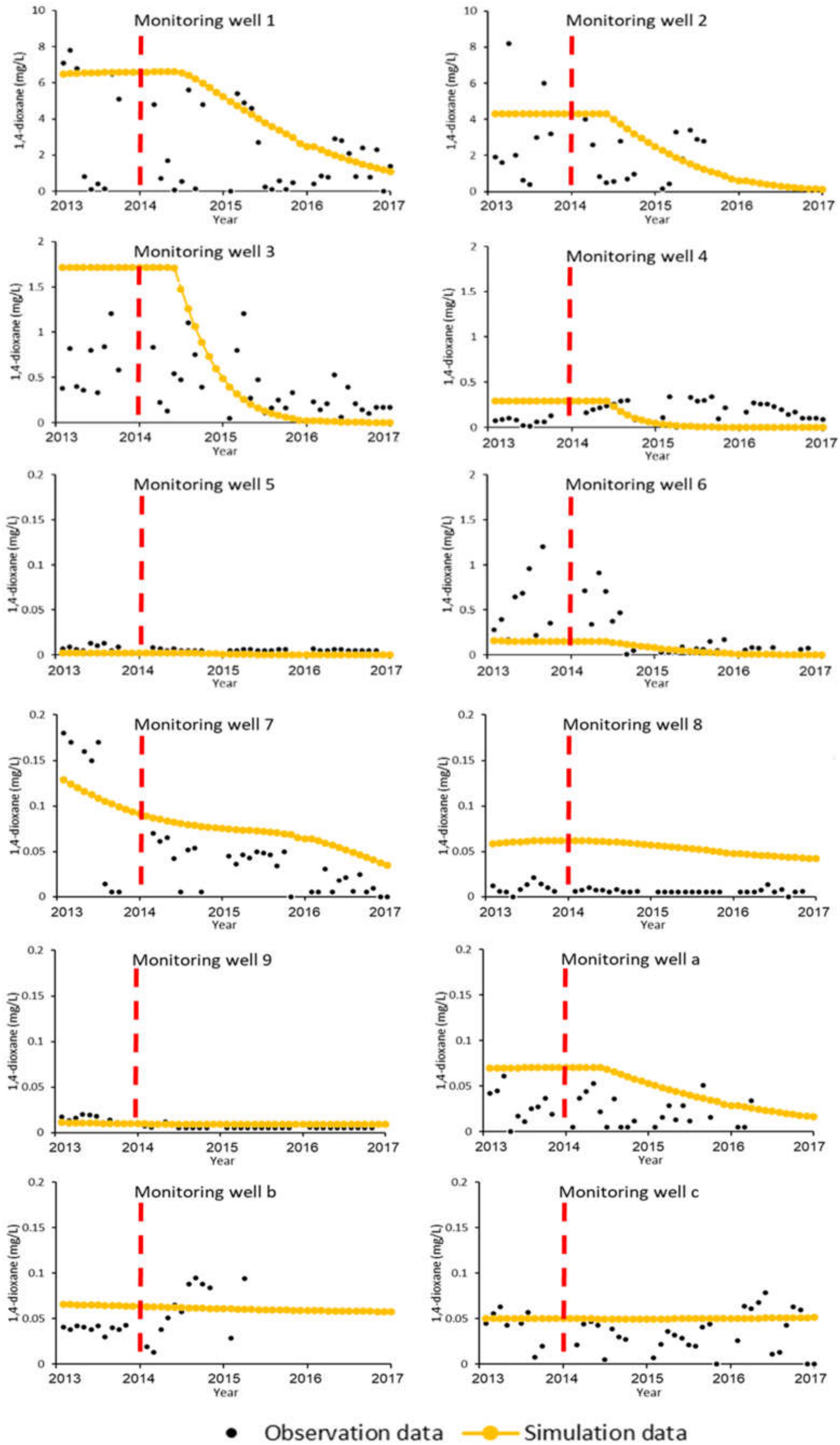

Figure 2. Comparison between the simulation results and observed data of 1,4-dioxane [26]. Red line means the excavation removal completed at the site (2014).

It should be noted that in terms of short periods of time such as each year, the reproduced 1,4-dioxane concentration did not fit well with the monitoring data. All monitoring wells, especially monitoring wells 1, 2, and 3, demonstrated highly fluctuating

1,4-dioxane concentrations each year. These fluctuations may be related to the complex geological structure of the site and dynamic environment such as seasonal rainfall or artificial activities, for example, pumping wells, impermeable walls, and excavation work, which may have increased the complexity of the groundwater environments. However, the difference between the monitoring data and reproduced data over short periods does not influence their consistency in long-term trends; therefore, this model can be used to predict 1,4-dioxane concentrations and distribution.

Using the reproduced 1,4-dioxane concentrations shown in Figure 2, the distribution of 1,4-dioxane in the groundwater of the study area over 25 years (1995–2020) is shown in Figure 3. The GL changes over time are also illustrated in the figure (light blue). In 1995 and 2000, the flow of groundwater containing pollutants was generally divided into two directions depending on the groundwater divide (northwest to southeast) of this dumping site area.

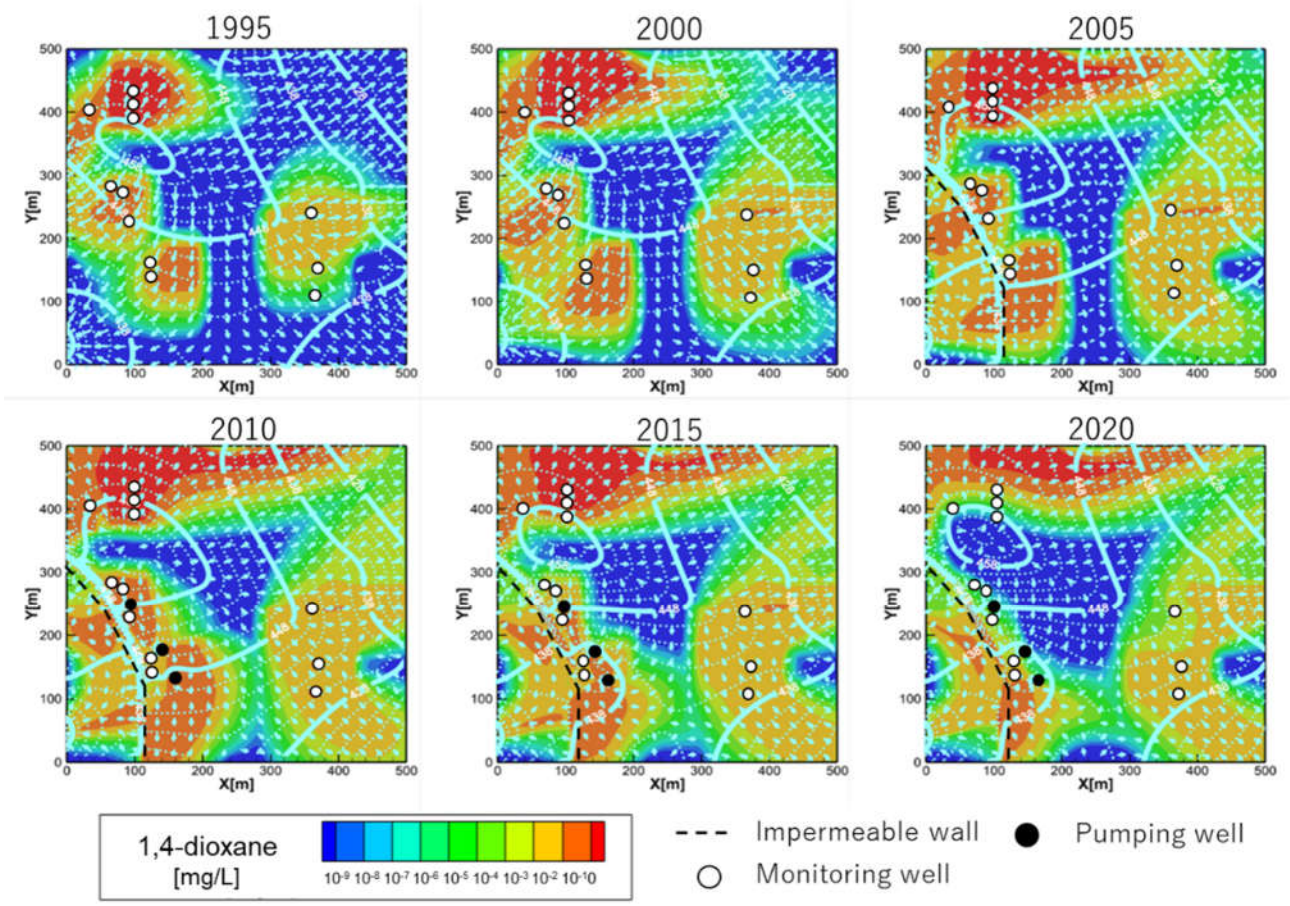

Figure 3. Distribution of groundwater pollution of 1,4-dioxane in the site over time (1995–2020). Contour lines indicate groundwater levels and vectors indicate flow directions. The color of the legend indicates the concentration of 1,4-dioxane.

From 2005, an impermeable wall was constructed along the border between the two prefectures to stop the movement of groundwater as well as the pollutants within it. As a result, the groundwater contours shown in Figure 4 overlapped along the impermeable wall, and the flow direction near this area changed from north to south. As expected, 1,4-dioxane was trapped on the east side of the impermeable wall to some extent.

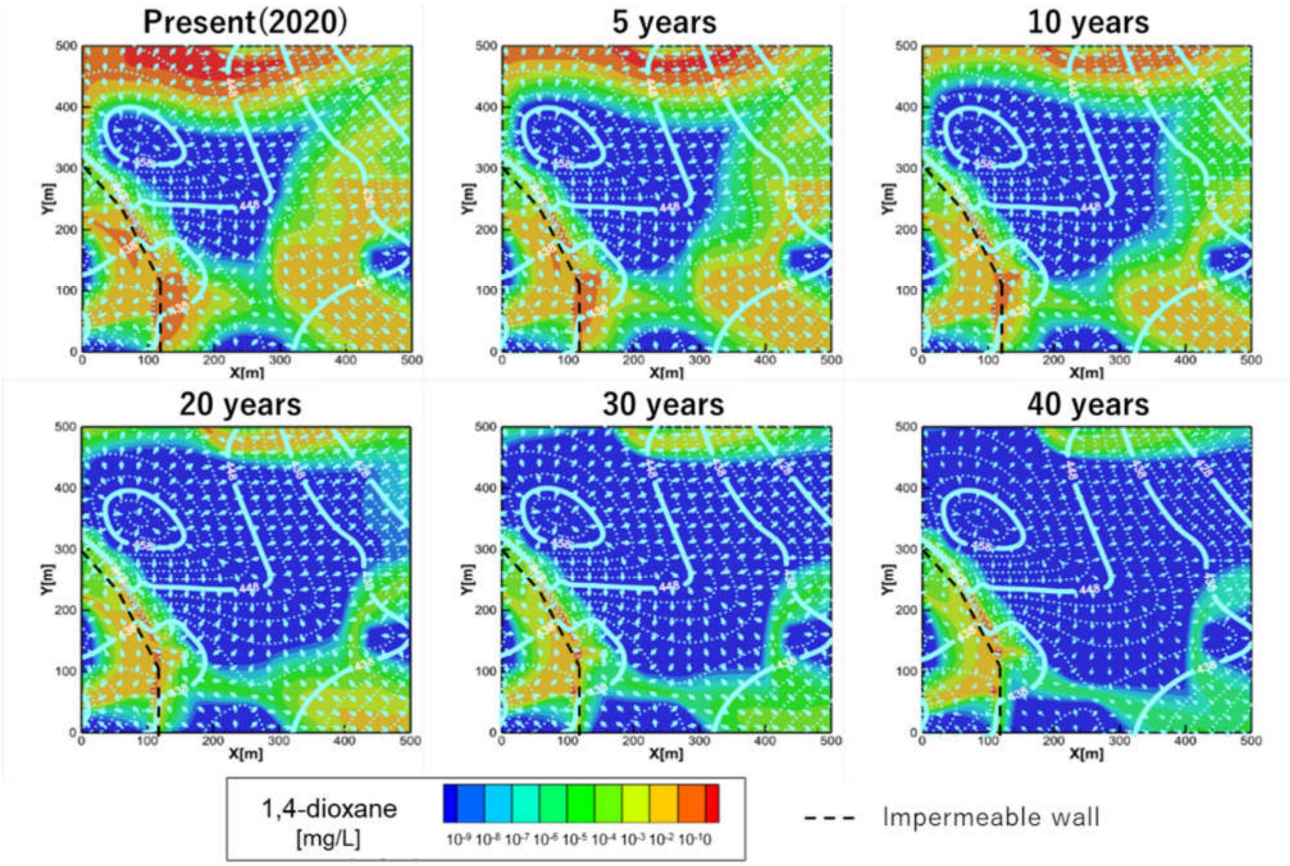

Figure 4. 1,4-Dioxane concentrations in groundwater by passive treatment over time. Contour lines indicate groundwater levels and vectors indicate flow directions. The color of the legend indicates the concentration of 1,4-dioxane.

In 2010, three pumping wells were installed near the impermeable wall at the waste dumping site for groundwater treatment. This action again increased the complexity of groundwater flow and groundwater levels. The groundwater near the pumping wells started to flow toward the wells, and the groundwater levels near the pumping wells declined. These changes imply accelerated groundwater flow and the transportation of pollutants in the groundwater. Interestingly, an obvious accumulation of 1,4-dioxane, along with the impermeable wall, was observed as a result of these artificial activities, that is, the impermeable wall and pumping well installments.

3.2. Evaluation of Different Methods for Groundwater Treatment

Based on the simulated current distribution of 1,4-dioxane in groundwater at the study site, two possible treatment methods, that is, passive and active treatments, were proposed and evaluated.

Passive treatment refers to the natural attenuation of 1,4-dioxane in groundwater. The simulation results of the distribution of 1,4-dioxane in the study site under natural attenuation over the next 40 years (2020–2060) are shown in Figure 4. The hydraulic environment of this site is relatively stable because no artificial activities have been conducted. Figure 4 suggests that as time passes, 1,4-dioxane moves with the groundwater flows, its concentration gradually decreasing. After 10 years of natural attenuation, most areas of the dumping site have 1,4-dioxane concentrations $\leq 10^{-3}$ mg/L, with very few areas in the range of 10^{-3}–10^{-1} mg/L. The concentration of 1,4-dioxane is relatively high near the impermeable wall, even after 40 years of natural attenuation, which may be attributed to the stagnation of groundwater flow near this area.

With the active treatment method, groundwater pumping, followed by treatment technologies, is applied. It is recommended to pump water through the monitoring wells, which are more widely installed than the current pumping wells—monitoring wells 7, 8, and 9 are close to the impermeable wall, which has an accumulation od 1,4-dioxane. A constant pumping rate of 250 m^3/day is suggested.

The distribution of 1,4-dioxane under active treatment is shown in Figure 5. The groundwater levels and groundwater directions near the monitoring wells and the impermeable wall were significantly influenced by groundwater pumping actions. These changes make the groundwater more dynamic and the groundwater environment becomes more complex, which also influences the movement of 1,4-dioxane. The northern areas with relatively high 1,4-dioxane concentrations (10^{-2}–10^{-1} mg/L) were quickly remediated within five years, with the 1,4-dioxane concentration decreasing to $<10^{-3}$ mg/L. After 30 years of active treatment, the 1,4-dioxane concentration in the groundwater in most areas of the site was $\leq 10^{-3}$ mg/L, and did not accumulate near the impermeable wall.

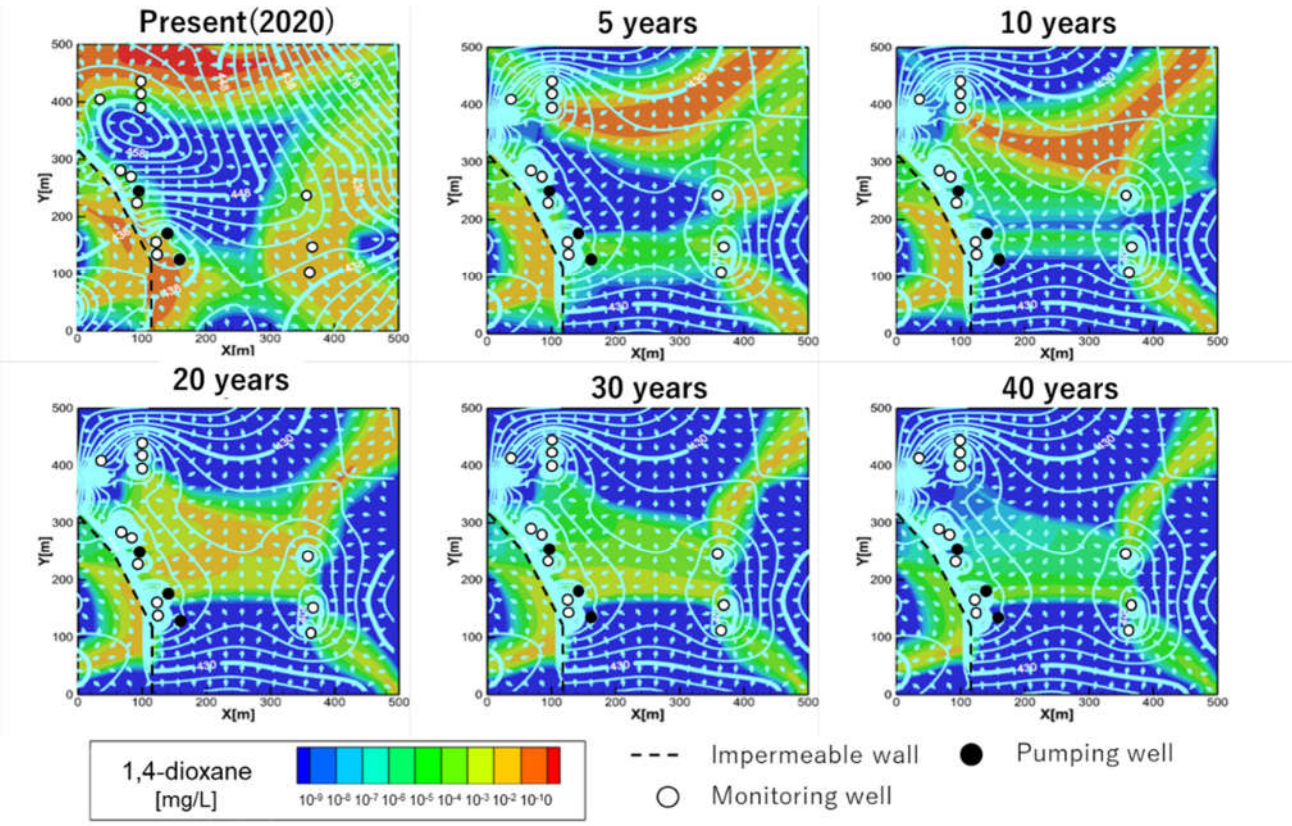

Figure 5. 1,4-Dioxane concentrations in groundwater by active treatment over time. Contour lines indicate groundwater levels and vectors indicate flow directions. The color of the legend indicates the concentration of 1,4-dioxane.

Comparing the simulation results of 1,4-dioxane distribution in the groundwater shown in Figures 4 and 5, a wider 1,4-dioxane distribution may occur in the case of the active treatment (e.g., after 30 years of treatment). It has been proposed that the pumping actions during active treatment may result in a more dynamic groundwater flow than during passive treatment, which promotes 1,4-dioxane transport to areas where it would otherwise be difficult to reach. This phenomenon was unanticipated and would influence the groundwater treatment efficiency; therefore, when designing pumping well locations, both the natural hydraulic environment and artificially induced groundwater flows should be taken into consideration. Finally, we suggest further research to study better prediction models and remediation strategies for groundwater pollution based on the findings of the present study.

4. Conclusions

The objective of this study was to clarify the possibility of numerically predicting pollutant transport in a dynamic and complex hydrologic environment and to investigate the characteristics of pollutant transport under both naturally and artificially induced groundwater flows. We attempted to reproduce the changes in 1,4-dioxane concentrations in groundwater observed in the monitoring wells using a quasi-3D flow and transport simulation that considered the multiple sources and spatiotemporal changes in hydraulic conditions.

The reproduced data and monitoring data (over a period of five years) for all monitoring wells generally exhibited good fitting results, suggesting that it is possible to predict the 1,4-dioxane concentration by considering groundwater flow. From the distribution of 1,4-dioxane, an obvious accumulation of 1,4-dioxane along with the impermeable wall was observed in these artificial activities, impermeable walls, and pumping well installments.

Passive treatment suggested that 1,4-dioxane moves with groundwater flows, and its concentration gradually decreases over time. The concentration of 1,4-dioxane was relatively high near the impermeable wall. With active treatment, the 1,4-dioxane concentration in the groundwater in most areas was $\leq 10^{-3}$ mg/L, and no longer accumulated near the impermeable wall. It is understood that the pumping actions during active treatment may result in a more dynamic groundwater flow than during passive treatment, promoting 1,4-dioxane transport to areas where it would otherwise be difficult to reach.

The study suggests the potential for the prediction and remediation of pollution at illegal waste dumping sites. However, further extensive studies on the complex geological structure of illegal waste dumping sites, dynamic seasonal rainfall, and groundwater changes are encouraged. In order to evaluate such complex environments, it is essential to improve prediction accuracy using more advanced models [39] and to analyze detailed field data.

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