学 位 論 文 内 容 の 要 旨
DISSERTATION ABSTRACT

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Remediation planning of 1,4-dioxane-contaminated groundwater by using numerical simulation
(数値シミュレーションを用いた 1,4-ジオキサンによる地下水汚染の修復計画)

Commonly used as a stabilizer for methyl chloroform, 1,4-dioxane is used extensively and directly in several industrial and commercial processes and in many consumer products. As a result of its widespread use, a large amount of 1,4-dioxane is released to the environment. Once 1,4-dioxane reaches the surface water or groundwater, it migrates as far the water flows because it is highly soluble and persistent in water ambient conditions. World Health Organization (WHO) classified 1,4-dioxane as Group 2B (possibly carcinogenic to human) and in late 2009 1,4-dioxane was added into Japan Environmental Quality Standards (EQS). Elevated 1,4-dioxane concentration has been detected at several contaminated sites in USA and illegal dumping and landfill sites in Japan.

At Kuwana illegal dumping site in Japan, where 30,000 m^3 of hazardous waste was illegally dumped between 1995 and 1996, groundwater was severely contaminated by Volatile Organic Compounds (VOCs), e.g., trichloroethylene and benzene. From 2002 to 2007, VOCs-contaminated groundwater was completely remedied by containing VOCs within the waste layer by implementing vertical slurry walls around the dumped waste, pumping contaminated groundwater inside and outside of walls for VOCs treatment (P&T), and flashing water into the waste layer by using treated water. However, high concentration of 1,4-dioxane was detected in both waste layer and groundwater, especially in deep aquifers inside and outside of the walls after 1,4-dioxane was newly added into EQS. This widely detection suggests that 1,4-dioxane cannot be contained by vertical walls and 1,4-dioxane migrates horizontally through the walls and vertically from the waste layer to the deep aquifer. The objective of remediation is to prevent 1,4-dioxane spreading out of the walls and to treat 1,4-dioxane in groundwater outside of walls within 10 years. For that reason, this study aims to propose a remediation plan for 1,4-dioxane-contaminated groundwater at Kuwana site. The most effective groundwater management tool " numerical simulation " was used to analyze remedial alternatives to achieve the remediation objective. Since the migration of 1,4-dioxane influenced by both vertical and horizontal flow, a 3-dimensional numerical simulation must be precisely developed to estimate the proper combination of amount of waste to be removed and the amount of groundwater to be pumped. Then, the developed numerical model was used to propose a proper remediation plan for Kuwana site considering future risk.

The unique properties of 1,4-dioxane cause its migration in groundwater to have limited influences by sorption, biodegradation, and volatilization. Consequently, 1,4-dioxane migration strongly depends on groundwater flow. For that reason, hydraulic conductivities, the most uncertain parameters and critical to groundwater flow, should be precisely determined. In a conventional modeling approach, groundwater flow is estimated using hydraulic conductivities determined by least-square method in term of observed and calculated groundwater head (calibration), and then the estimated groundwater flow is used for predicting 1,4-dioxane distribution by convective dispersion analysis considering other parameters such as dispersivity, source location and concentration. Although other parameters are properly set, 1,4-dioxane distribution cannot be always precisely predicted because the calibrated groundwater flow model does not perfectly present the real groundwater flow. Thus, the calibrated groundwater flow should be verified to determined appropriate hydraulic conductivities considering 1,4-dioxane distribution. This study proposes a new approach with verification processes of groundwater flow estimation for precisely predicting 1,4-dioxane distribution in groundwater. In this approach, several acceptable sets of hydraulic conductivities in term of groundwater heads were determined by calibration and each set of hydraulic conductivities was verified to match between calculated and observed 1,4-dioxane concentrations. For the case of Kuwana site, five acceptable sets of hydraulic conductivities of the three aquifers were determined by the calibration using observed groundwater heads, and then verified to minimize the variance and average in the observed and calculated 1,4-dioxane concentration. Moreover, the shape comparison of 1,4-dioxane distribution in each aquifer was

also carried out to obtain the best match with the observed field data. Because of adequate field data available at Kuwana site, the efficiency of a new approach comparing to the conventional one was confirmed. As a result, 1,4-dioxane distribution in groundwater was clarified by our approach more precisely than the conventional approach. Therefore, a new groundwater modeling approach that is practical for simulating 1,4-dioxane distribution was proposed by this study.

Afterwards, the above developed model was applied for the remediation planning for 1,4-dioxane-contaminated groundwater at Kuwana site considering waste removal and P&T. The amount of waste to be removed and the pumping locations and rates for P&T were determined by various scenario analysis considering both the depth of waste layer and 3-dimensional 1,4-dioxane distribution in each aquifer. Firstly, the pumping locations were determined based on the groundwater directions and current distribution of 1,4-dioxane in all aquifers, and then the proper combination of pumping rates were calculated so that the minimum average concentration outside of the walls was obtained. As a result, groundwater inside and outside of walls should be pumped with the rate of 50 m^3/d and 10 m^3/d respectively. Afterwards, by combining with the required pumping rates, four remediation plans with different amount of removed waste were considered for the analysis, plan (a): no waste is removed, plan (b): 1/2 of waste (highly 1,4-dioxane-concentrated waste) is removed, plan (c): 2/3 of waste (deeply distributed waste) is removed, and plan (d): all waste is removed. The average 1,4-dioxane concentration obtained from 78 monitoring wells outside of the walls in all aquifers was used to compare the effectiveness of each plan. Within 10 years of calculation period, we assumed that if the average 1,4-dioxane concentration of each aquifer lower than regulation limit in EQS of 0.05 mg/L, and then the remediation objective can be achieved. As a result, all three aquifers in plan (a) and (b) were not remedied. In plan (c), only the third aquifer was not remedied while all aquifers were remedied in plan (d). However, the removal of all the waste for treatment is not feasible because our cost analysis revealed that plan (d) is much more expensive than plan (c). Therefore, we tried to improve the remediation effectiveness of plan (c) considering the technical aspect. Plan (c) was found to be easily improved because the deep waste containing high concentration of 1,4-dioxane was already removed and the only remaining 1,4-dioxane in groundwater should be treated. By observing the concentration in 5 years after remediation, three pumping wells were found to be ineffective because 1,4-dioxane in their locations was already remedied so that they were switched to other locations where 1,4-dioxane was remaining high. As a result, groundwater in all aquifers in plan (c) could be remedied within 10 years. Therefore, this study proposes plan (c) as the most effective and economically feasible remediation plan. However, the remaining one-third of waste might contain 1,4-dioxane with high concentration and serve as the continuous source of 1,4-dioxane and spread out of the walls into the surrounding environment. In order to prevent this future risk, various scenarios with different pumping rates of pumping wells inside of walls were carried out. Accordingly, after the completion of remediation in the proposed plan (c), our simulation suggested that groundwater within the remaining waste must be pumped up for P&T at least 20 m^3/d for preventing future risk.

In conclusion, this study proposed a new modeling approach that suitable and practical for predicting 1,4-dioxane distribution in groundwater. 1,4-Dioxane distribution in groundwater can be predicted more precisely by using the proposed approach. Using the adequate available field data, the effectiveness of our new approach was confirmed. Subsequently, the developed model was used for proposing a feasible remediation plan for 1,4-dioxane-contaminated groundwater at Kuwana site. This study contributes the establishment of a new engineering method for the application of numerical simulation to remediation planning for groundwater at the site which is contaminated by newly regulated 1,4-dioxane.