Evaluating Contaminant Transport in the Vadose Zone to Support Remedy Decisions at Aqueous-Waste Disposal Sites – 16195

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ABSTRACT

Inorganic contaminants in aqueous waste disposed at land surface have a potential to contaminate groundwater. However, contaminant concentrations are attenuated during transport through the vadose zone. Quantifying contaminant attenuation and the resulting temporal concentration profile of in underlying groundwater is important for assessing the need for, and type of, remediation in the vadose zone and groundwater. An approach was developed for evaluating vadose zone transport and attenuation of aqueous wastes containing inorganic (non-volatile) contaminants that were disposed at land surface (i.e., directly to the ground in cribs, trenches, tile fields, etc.) and their effect on underlying groundwater. The approach provides a structured method to estimate transport of contaminants through the vadose zone and the resulting temporal profile of groundwater contaminant concentrations.

A series of numerical model simulations was applied to investigate how different factors affect contaminant transport in the vadose zone and the conditions for which individual factors are important. This analysis demonstrated that, while water flow and contaminant transport can be complex during aqueous-waste disposal, conditions will change over time after disposal ceases and fewer factors will control the contaminant transport rate and resulting temporal profile of groundwater contaminant concentrations. Data verifying that these conditions have been reached can be provided by a few key metrics such as the vertical profile of moisture content, the temporal profile of recharge at the site, and basic information about waste disposal and subsurface properties. Under these conditions, and with information about specific contaminant transport parameters (e.g., for partitioning), estimates of contaminant transport can be made and used to support remedy decisions.

INTRODUCTION

In many cases, inorganic contaminants in aqueous waste solutions disposed of at the land surface must migrate through the vadose zone before entering groundwater. Because of contaminant transport through the vadose zone, the temporal profile of concentrations entering the groundwater is different from the temporal profile for aqueous waste disposal. Vadose zone transport mechanisms tend to decrease contaminant concentrations and limit the rate of contaminant movement. In these ways, contaminant concentrations are attenuated during transport through the vadose zone. The temporal profile of contaminant discharge into underlying groundwater defines the source characteristics with respect to predicting the resulting groundwater plume. At sites where inorganic contaminants in aqueous waste solutions have been disposed of at the land surface and a groundwater plume is still evolving or has not yet emerged, methods to evaluate

vadose zone contaminant transport are needed to estimate the future temporal profile of contaminant discharge to the groundwater. These estimates can be used to support remedy decisions for the vadose zone and groundwater.

A recent publication [1] provides an approach for evaluating vadose zone transport and attenuation of aqueous wastes containing inorganic (non-volatile) contaminants that were disposed of at the land surface (i.e., directly to the ground in cribs, trenches, tile fields, etc.) and the resultant effect on underlying groundwater. In summary, the approach provides a structured method for estimating transport of contaminants through the vadose zone and the temporal profile of groundwater contaminant concentrations in the resulting source area. The approach builds on vadose zone contaminant transport concepts [2] by providing a quantitative analysis useful for estimating future contaminant mass discharge from the vadose zone to the groundwater. Multi-dimensional numerical model simulations were conducted for a range of waste disposal conditions and results were interpreted to provide insight into vadose zone transport behavior and define appropriate transport analysis approaches.

DESCRIPTION

Contaminant transport through the vadose zone beneath aqueous waste disposal sites is affected by two types of attenuation processes: 1) attenuation caused by unsaturated flow and 2) attenuation caused by biogeochemical reactions and/or physical/chemical interaction with sediments. Mixing processes with the groundwater are also important for estimating contaminant concentrations in groundwater resulting from vadose zone contaminant flux. Truex et al. [1] conducted a series of simulations to evaluate the attenuation caused by unsaturated flow. The simulation matrix also included evaluating the effect of contaminant retardation as part of evaluating attenuation caused by physical/chemical interaction with sediments.

The water mode of the STOMP simulator [3] was used to simulate vadose zone aqueous phase flow and contaminant transport [1]. A cylindrical model was constructed for the vadose zone with the same lateral discretization, independent of the source size. For all simulations, the upper and lower 5 m of the domain used the same grid z-direction discretization (0.1 m). In between these two zones, 0.25-m grid blocks were used. Several grid refinement iterations were conducted until no changes were observed in the water and contaminant fluxes across the water table. The simulations included waste disposal scenarios in homogeneous sand and sandy loam and systems with silt layers in sand and sandy loam.

Results of simulations were interpreted in terms of identifying characteristics of vadose zone contaminant transport. In addition, simulation results were used for comparison to analytical solutions describing vadose zone contaminant transport under some types of conditions.

DISCUSSION

The vadose zone transport evaluation conducted by Truex et al. [1] revealed that transport behavior could be grouped into two categories. In one category, the disposed volume is not sufficient to cause short-term (i.e., during waste disposal) contaminant impact to the groundwater. The contaminant arrives later and the mass discharge is characterized by a nearly symmetrical single-peak elution curve (Fig. 1). For a contaminant with this transport behavior, the disposal site is termed a Category I site. The disposal volumes associated with this category are relative to the vadose zone size and do not have to be small in absolute terms. For instance, Truex et al. [4] identify Category I sites that had disposal of several million liters of wastewater over a 2 to 3 month period into a 100-m-thick vadose zone.
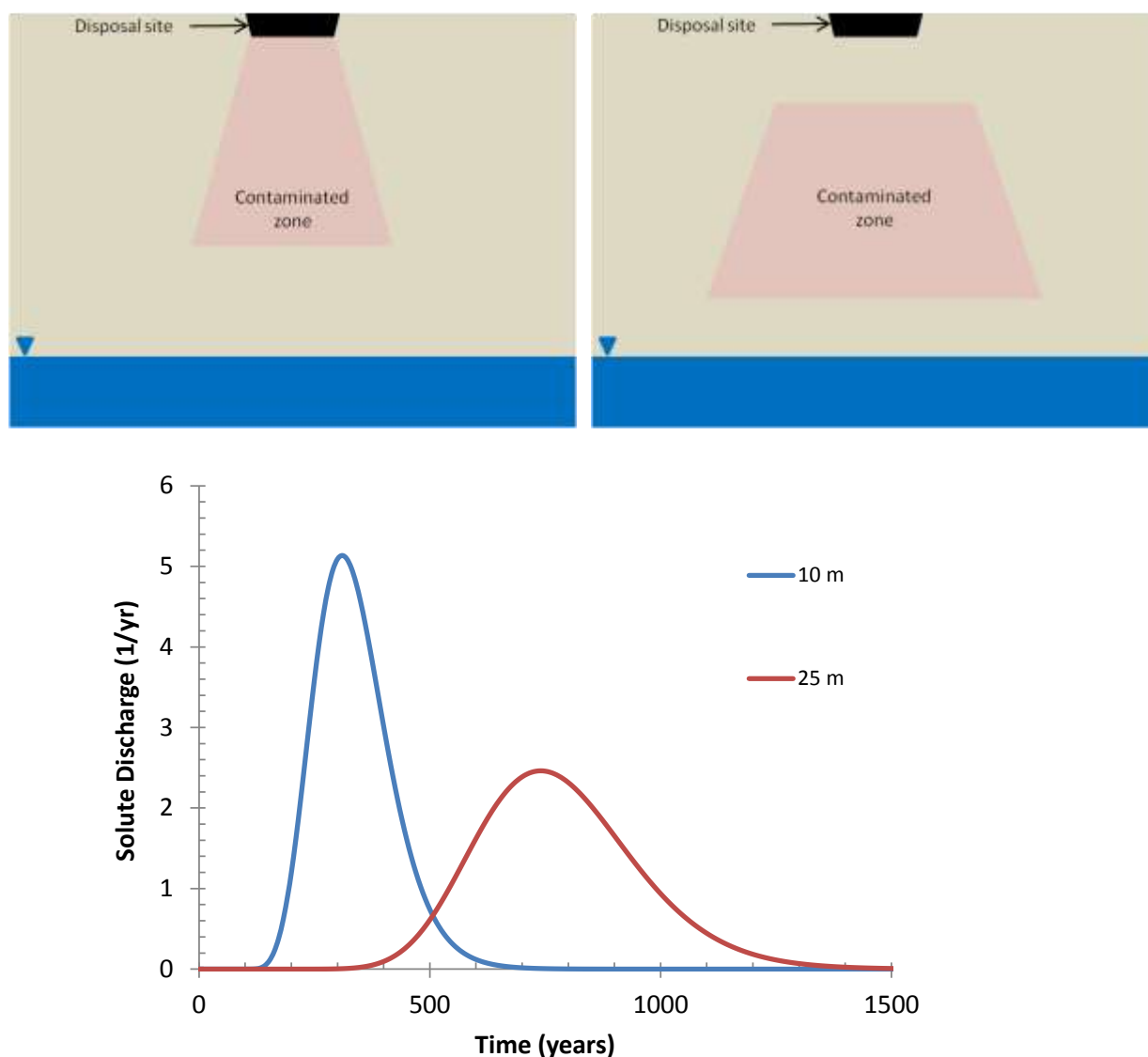
Fig. 1. Contaminant distribution during disposal (top left) and at the time of the transport evaluation (top right) for Category I, where the disposed volume

was not sufficient to cause short-term impact on the groundwater. In the bottom figure, example plots of solute discharge into groundwater over time after waste disposal has ended for this scenario are shown for two vadose zone thicknesses (10 and 25 m) and the same disposal volume.

When the disposed volume is sufficient to cause short-term impacts to groundwater, part of the contaminant mass is discharged into the groundwater quickly after (or during) disposal (Fig. 2). During this period some contaminant also migrates laterally in the vadose zone due to advection and hydrodynamic dispersion. This process results in some contaminant mass in the vadose zone that will be discharged into the groundwater after the main discharge subsides. Elution curves for this type of disposal are characterized by an initial peak, representing the immediate impact to groundwater, and a second peak representing the impact of contaminant not transported by the initial pulse of waste fluid. For a contaminant with this discharge behavior, the disposal site is termed a Category II site.
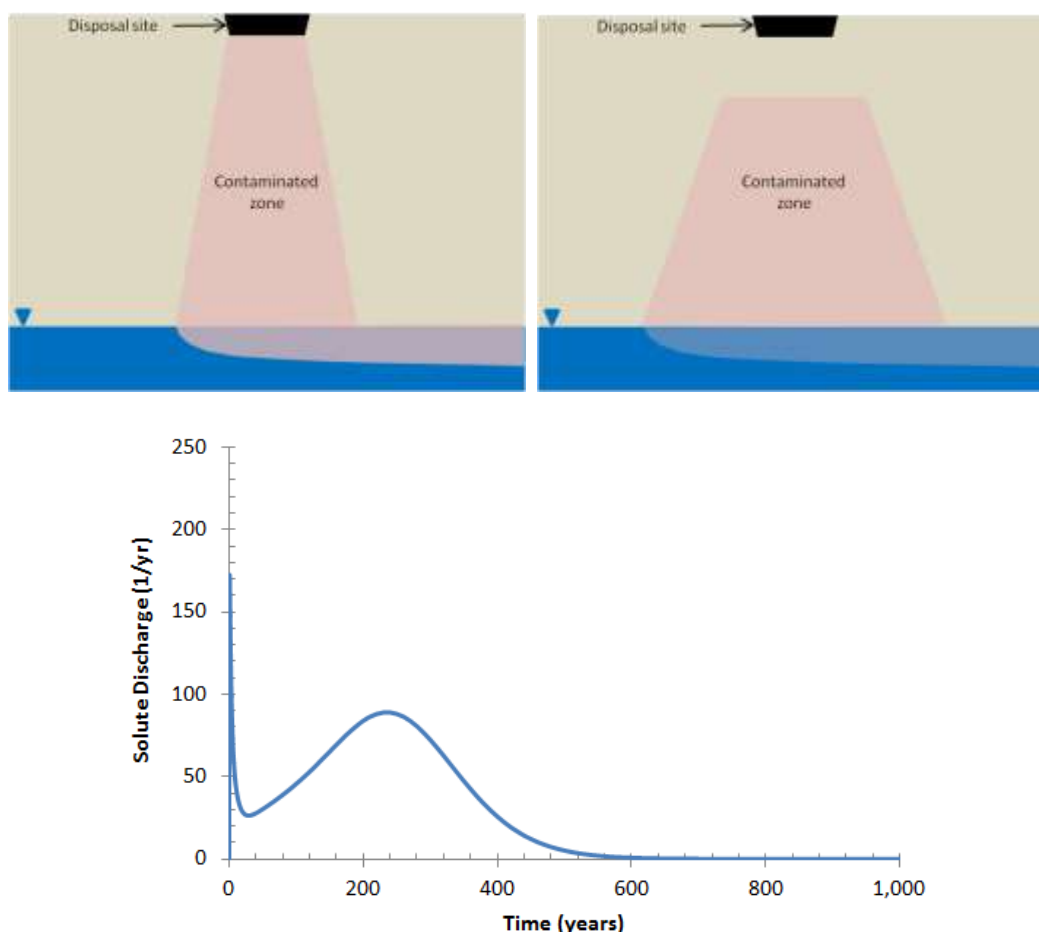
Fig. 2.   Contaminant distribution during disposal (top left) and at the time of the transport evaluation (top right) for Category II contaminant discharge, where the disposed volume was sufficient to cause a short-term impact on the groundwater. In the bottom figure, an example plot of solute discharge into groundwater for this scenario is shown.

Some sites may have contaminants with both Category I and Category II waste disposal characteristics. This situation might arise when multiple waste disposals occurred with different volumes. Another scenario when both categories are represented may be when a sorbing and a nonsorbing contaminant are in the same high-volume waste disposal; in such a case the sorbing contaminant may lead to a Category I mass discharge and the nonsorbing contaminant may result in a Category II mass discharge. However, in some instances, the observed mass discharge behavior may be characterized by an asymmetrical single-peak elution curve, indicating a condition in between Category I and II.

There are several implications of the above results. For Category I sites, vadose zone transport after disposal ends is controlled by the recharge rate and a simplified analysis of transport and the resulting groundwater contaminant concentration profile using analytical or streamlined numerical simulation approaches described by Truex et al. [1] can be applied. For Category I sites, detailed knowledge of the subsurface hydraulic properties and their distribution is not needed to conduct these analyses. Because only a relatively small volume of water is added to the vadose zone for a Category I site, the dynamics of unsaturated water flow are minimal and the vadose zone flow quickly reverts to a recharge-controlled system, for which contaminant transport can be reasonably estimated with simplified approaches (unless there are complicated biogeochemical reaction dynamics to consider).

For Category II sites, the implications of the evaluation are related to the characteristic shape of the contaminant (solute) discharge curve (Fig. 2). The initial discharge into the groundwater occurs during or shortly after aqueous waste disposal. Thus, Category II sites will be associated with a near-term groundwater plume. Under some conditions, the initial contaminant discharge to groundwater will appear as a peak that diminishes in the near term as shown in Fig. 2, but is followed by a longer-term second peak in contaminant discharge. Thus, a site may observe a diminishing source term for a plume in the near term. However, for a Category II site, the source will not continue to decline to zero. Thus, decisions about remedy approaches must consider the magnitude and duration of the second contaminant discharge peak.

Analysis of the contaminant discharge and associated groundwater contaminant concentration profile is needed to determine whether the second peak from a Category II site may result in a plume that can be addressed through natural attenuation or if additional actions are needed. The dynamics of unsaturated flow causing the first peak at a Category II site may be complex and affected significantly by the distribution of subsurface hydraulic properties. These vadose zone transport dynamics would be manifested in the observed existing groundwater plume dynamics. However, the dynamics of vadose zone transport will subside over time so that a Category II site relaxes to a condition with recharge controlling contaminant transport. Thus, estimates of future contaminant flux to groundwater and the resulting groundwater contaminant concentration profile may, for many situations, use the analytical or streamlined numerical simulation approaches

described by Truex et al. [1] to provide reasonable estimates in support of remedy decisions associated with the second peak of a Category II site.

Contaminants with higher retardation factors will be more likely to result in a Category I site response because the initial advection due to even a large volume of aqueous waste discharge may be insufficient to transport contaminants to the groundwater during or shortly after disposal. Higher contaminant retardation works to extend the timeframe for peak arrival and lower the resulting groundwater contaminant concentration relative to contaminants with lower retardation factors. Retardation-based effects can be readily incorporated into the analytical or streamlined numerical simulation approaches described by Truex et al. [1] to estimate future impact to groundwater. However, there may be other complexities in contaminant transport parameters that need to be considered for some contaminants. For instance, the disposal chemistry may interact with contaminants, causing reactions that sequester some of the disposed contaminant mass, or may include co-contaminants that increase the fluid ionic strength and lead to reduced contaminant retardation [5, 6]. Thus, each site should consider the biogeochemical factors that may be important in selecting appropriate contaminant transport parameters to couple with the analyses approaches presented by Truex et al. [1].

CONCLUSIONS

Several aspects of the approach summarized herein, and presented in more detail by Truex et al. [1], offer new perspectives for evaluation of vadose zone contamination.

- Contaminant transport behavior can be categorized based on a set of site and waste disposal parameters, using groundwater plume data to corroborate the selection of a category. There are distinct characteristics of the temporal profile of groundwater contamination for each category.

    - Some sites are characterized by contaminant discharge from the vadose zone to the groundwater resulting in a single peak of groundwater contamination that, in most cases, will occur in the future. Remedy decisions for these sites will need to consider whether the contaminant discharge from the vadose zone will cause a plume of concern and, if remediation is needed, whether a near-term vadose zone remedy or a later (longer-term) groundwater remedy would be more effective.

    - Other sites—those with large waste disposal volumes compared to the thickness of the vadose zone—are characterized by contaminant discharge from the vadose zone to the groundwater resulting in two peaks in groundwater contamination: one peak associated with an existing/near-term plume, and one peak that will occur in the future. Remedy decisions for these sites must consider that, even though groundwater concentrations in the source area are diminishing in the near term (i.e., the decline of the first peak), the contaminant mass discharge from the vadose zone will not decline to zero and will rise again much later as part

of the second peak. Thus, near-term remedy decisions must consider the nature and extent of the existing or near-term plume and how a combined vadose zone/groundwater remedy or a groundwater-only remedy can be applied to reach remedial action objectives. In addition, remedy decisions for these sites will need to consider the continuing vadose zone source that will occur and, potentially, will be of a higher magnitude than the current source.

- For sites that are expected to exhibit a single peak in groundwater contamination, algebraic equations can be used to estimate the arrival time of the peak groundwater contaminant concentration beneath the vadose zone source and its concentration relative to the disposed concentration. Tabulated values presented by Truex et al. [1] for the duration of the contaminant mass discharge from the vadose zone to the groundwater (between the initial arrival and final elution of 1% of the peak concentration value) can be used to further estimate the characteristics of the vadose zone source area.

- While nuances in site properties and waste disposal details will affect contaminant transport in the vadose zone, the technical basis for the evaluation approach herein suggests that reasonable estimates of vadose zone transport can be made. That is, transport of contaminants in the vadose zone is predictable under most conditions that are relevant to predicting the future contaminant mass discharge into groundwater from an aqueous waste disposal. In addition, a relatively small set of site and waste disposal parameters are needed to make estimates that will represent the bulk behavior of contaminant mass discharge into the groundwater.

REFERENCES

1. M. J. TRUEX, M. OOSTROM, and G. D. TARTAKOVSKY, *Evaluating Transport and Attenuation of Inorganic Contaminants in the Vadose Zone for Aqueous Waste Disposal Sites*, PNNL-24731, Pacific Northwest National Laboratory, Richland, WA (2015).
2. M. J. TRUEX and K. C. CARROLL, *Remedy Evaluation Framework for Inorganic, Non-Volatile Contaminants in the Deep Vadose Zone*, PNNL-21815, Pacific Northwest National Laboratory, Richland, WA (2015).
3. M. D. WHITE and M. OOSTROM, *STOMP: Subsurface Transport Over Multiple Phases, Version 4.0, User's Guide*, PNNL-15782, Pacific Northwest National Laboratory, Richland, WA (2006).
4. M. J. TRUEX, M. OOSTROM, G. V. LAST, C. E. STRICKLAND, and G. D. TARTAKOVSKY, *Evaluating Contaminant Flux from the Vadose Zone to the Groundwater in the Hanford Central Plateau: SX Tank Farms Case Study*, PNNL-23737, Pacific Northwest National Laboratory, Richland, WA (2015).
5. M. J. TRUEX, J. E. SZECSODY, N. QAFOKU, and J. R. SERNE, *Conceptual Model of Uranium in the Vadose Zone for Acidic and Alkaline Wastes Discharged at the Hanford Site Central Plateau*, PNNL-23666, Pacific Northwest National Laboratory, Richland, WA (2014).

6. J. E. SZECSODY, M. J. TRUEX, N. QAFOKU, D. M. WELLMAN, T. RESCH, and L. ZHONG, "Influence of acidic and alkaline waste solution properties on uranium migration in subsurface sediments," *J. Contam. Hydrol.* 151:155-175 (2013), dx.doi.org/10.1016/j.jconhyd.2013.05.009

ACKNOWLEDGEMENTS