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IMPROVING MODELING OF IODINE-129 GROUNDWATER CONTAMINATION PLUMES USING THE SYSTEM ASSESSMENT CAPABILITY

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ABSTRACT

Years of production of radioactive materials at the Hanford Site in southeastern Washington State has resulted in contamination of surface, subsurface, and surface water environments. Cleanup of the site has been aided by various tools, including computer software used to predict contaminant migration in the future and estimate subsequent impacts. The System Assessment Capability (SAC) is a total systems tool designed to simulate the movement of contaminants from all waste sites at Hanford through the vadose zone, the unconfined aquifer, and the Columbia River. Except for iodine-129, most of the contaminants modeled by SAC have acceptably matched field measurements. The two most likely reasons for the inconsistency between the measured field data and SAC modeled predictions are an underestimated inventory and an overestimated sorption value (K_d). Field data tend to be point measurements taken from near the surface of the unconfined aquifer. Thus, the depth of the iodine-129 contamination plume on the site is not well characterized. Geostatistical analyses of the measured data were conducted to determine the mass of iodine-129 for four assumed plume depths within the unconfined aquifer. Several simulations for two different K_d 's using the initial SAC inventory were run to determine the effect of an overestimated sorption value on SAC modeled predictions. The initial SAC inventory was then increased for the two different K_d 's to determine the influence of an underestimated inventory on SAC modeled predictions. It was found that evidence for both an underestimated inventory and for an overestimated sorption value for iodine-129 exist. These results suggest that the K_d for iodine-129 should be reevaluated and that a more complete inventory must be generated in order to more accurately model iodine-129 groundwater contamination plumes that match available field data.

INTRODUCTION

BACKGROUND

The Hanford Site was established in 1944 by the United States Government to produce weapons-grade plutonium. Approximately 2,600 toxic and radioactive waste disposal sites were created at Hanford as a result of plutonium manufacturing processes [1]. Radionuclides and other chemicals associated with the production of radioactive materials have been released into the surface, sub-surface, and surface water environments. The need to understand the total impact of all wastes on the Hanford Site has been the motivation for the development of

various modeling tools to aid in the complex cleanup process of the site. The System Assessment Capability (SAC) [1] is a total systems tool that has been developed to advance the understanding of the environmental transport of contaminants and their impacts on the Hanford Site and surrounding areas.

GEOLOGIC SETTING

The Hanford Site (Figure 1) occupies an area of 1,517 square kilometers (586 square miles) north of the city of Richland in southeastern Washington State. It lies within the Pasco Basin, which is a structural basin formed by the deformation of the underlying Columbia River Basalt Group

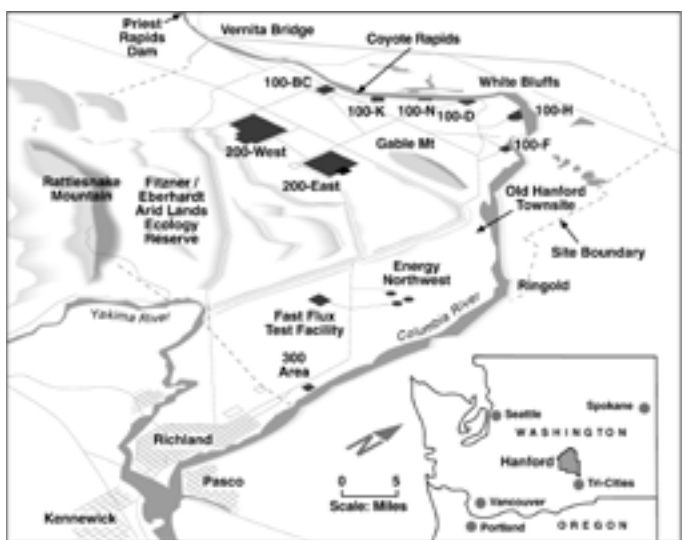


Figure 1. The Hanford Site [7].

[2]. Prominent geologic structures of the area consist of basalt ridges, such as Gable Mountain, which is located in the central part of the Hanford Site. The Columbia River flows eastward through the site, just north of Gable Mountain, until it bends south to form the eastern boundary of the site. The Rattlesnake Hills form the southwestern boundary of the site. The Yakima River flows near the southern border of the site before it joins the Columbia River south of Richland. The underlying sediments and soils of the Hanford Site primarily consist of fluvial, lacustrine, and glaciofluvial sediments [3]. These sediments compose the unconfined aquifer system of the Hanford Site [3]. Some sedimentary interbeds and basalt intra-flow zones above the Columbia River Basalt Group compose a series of confined aquifers [2]. Groundwater from both the confined and unconfined aquifer systems generally flows from the higher elevations of the Rattlesnake Hills northeastward toward the Columbia River [1]. Field data confirm that most of the groundwater contamination at the Hanford Site is located in the upper part of the unconfined aquifer system [4].

Contamination of the Hanford Site

Past records of radioactive products and by-products do not provide all of the information needed to conduct a site-wide evaluation of the threat these contaminants pose to humans living in the immediate Hanford area and to the environment surrounding the Hanford Site. These records have led to numerous inventory discrepancies particularly with respect to identifying quantities of contaminants released at various disposal locations. Many of these discrepancies have been remedied by comparing past records to recent vadose zone and groundwater monitoring data. These comparisons have helped to better determine the amount of contamination that has been leaching or injected into the underlying soils and sediments of the site. However, some of the contaminants modeled by SAC do not have complete inventory data nor have they been traced back to their original source locations. Iodine-129 is a

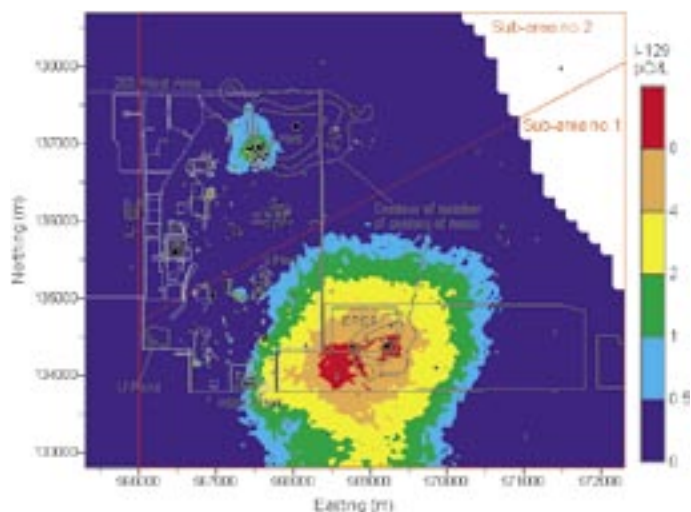


Figure 2. Aerial view of the 200-West REDOX and T-Plant Areas median contamination plume from the geostatistical groundwater data for 2001. Red boundaries in this figure delineate study areas; sub-area 1 has an area of 20.4 square kilometers, while sub-area 2 includes 12.8 square kilometers.

contaminant for which SAC modeled predictions have been inconsistent with field measurements. Groundwater monitoring data through 2001 indicate iodine-129 contamination plumes present around the 200-East and 200-West Areas of the Hanford Site (Figure 1). However, SAC Rev. 0 software did not predict the presence of iodine-129 in the groundwater for any part of the site by 2001. Incomplete inventory coupled with discrepancies in iodine-129 release locations as well as possibly overestimated sorption have resulted in severe under-predictions by SAC for both the total mass and the aerial extent of iodine-129 in the groundwater.

Geostatistical Analysis of Measured Data

Contamination plume field data for iodine-129 are limited and those that do exist tend to be point measurements taken

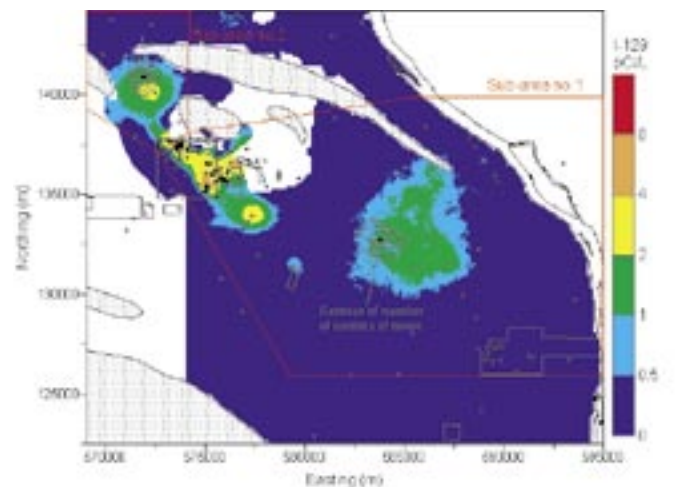


Figure 3. Aerial view of the 200-East Area median contamination plume from the geostatistical groundwater data for 2001. Red boundaries in this figure delineate study areas: sub-area 1 has an area of 207 square kilometers, while sub-area 2 includes 24.9 square kilometers.

were then divided by the VADER data for each area to determine how much more iodine-129 would be needed to generate contamination plumes resembling the measured field data.

Overestimated Sorption Value Approach

The other aspect of our research focused on an overestimated sorption value (K_d) as the possible source for the discrepancy between the observed groundwater contamination plumes in 2001 and the absence of SAC Rev. 0 groundwater contamination plumes. In this study we assume equilibrium between the aqueous and sorbed phases so that retardation R and the sorption coefficient K_d are related by

$$R = 1 + \frac{1-n}{n} \rho_s K_d$$

Where R is retardation (dimensionless), n is total porosity (length³ length⁻³), ρ_s is density of the solid phase (mass length⁻³), and K_d is the distribution coefficient (length³ mass⁻¹).

For this aspect of our research, the original SAC iodine-129 inventory was used, but the retardation for iodine-129 was changed from 6.23 to 1 to allow the maximum amount of contamination to reach the groundwater given the current inventory. The mass of iodine-129 reaching the groundwater for each site was calculated by using the STOMP module. The sites contributing contamination to the unconfined aquifer were then divided into the 200-West REDOX Plant Area, the 200-West T-Plant Area, and the 200-East Area in order to directly compare the SAC modeling predictions to the geostatistical groundwater data.

Assigning a retardation of 1.0 (K_d of zero) means that no iodine-129 contamination is simulated as absorbed onto the underlying soils and sediments of the Hanford Site in our model. Therefore, all of the contamination reaching the groundwater is represented as being in the aqueous phase. For this reason, the

	200-West T-Plant Area	200-West REDOX Plant Area	200-East Area
VADER data	0.307	0.073	2.560
STOMP (retardation = 6.23)	1.400E-04	1.340E-09	1.460E-03
STOMP (retardation = 1.0)	0.058	0.023	0.479

Table 1. Iodine-129 contamination (curies) for SAC modeled data for both the Underestimated Inventory and Overestimated Sorption Value Approaches.

geostatistical groundwater data could be directly compared to the SAC modeling predictions. This comparison was conducted to determine how much more iodine-129 would be needed to generate the observed groundwater contamination plumes for 2001.

RESULTS

Figure 5, Figure 6, and Figure 7 depict the original geostatistical groundwater data for the 200 West T-Plant Area, the 200 West REDOX Plant Area, and the 200 East Area [6]. These data include minimum and maximum contamination activities (curies) for the four assumed plume depths as well as mean, median, and standard deviation for those depths. These data depict the confidence we can hold with respect to the amount of contamination that exists in the groundwater in 2001. The geostatistical data in these figures is only in the aqueous phase. It does not represent the total iodine-129 contamination for each plume because it neglects the sorbed, solid phase.

The total aqueous- and solid-phase minimum and maximum contamination for the 200-West T-Plant Area, the 200-West REDOX Plant Area, and the 200-East Area are represented in Figure 8, Figure 9, and Figure 10, respectively. The total minimum and maximum iodine-129 contamination for each area was generated by multiplying the original geostatistical groundwater data by retardation factor 6.23. The original minimum and maximum contamination from the geostatistical groundwater data is also depicted on these figures to illustrate

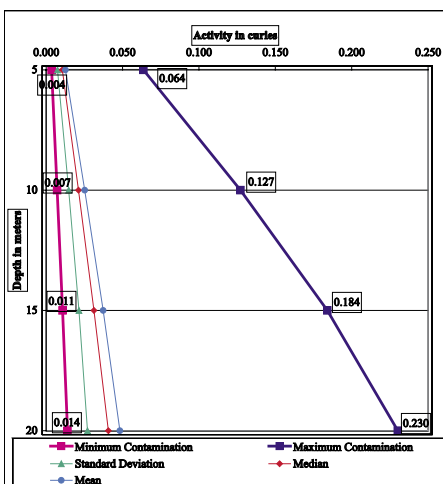


Figure 5. Geostatistical groundwater data for four assumed plume depths within the 200-West T-Plant Area.

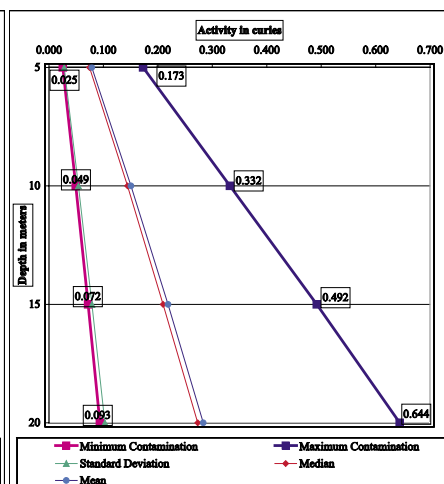


Figure 6. Geostatistical groundwater data for four assumed plume depths within the 200-West REDOX Plant Area.

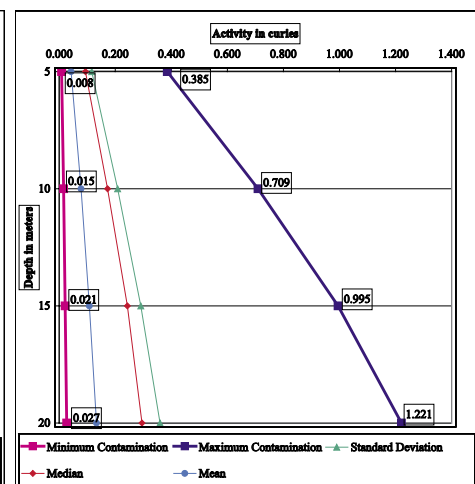


Figure 7. Geostatistical groundwater data for four assumed plume depths within the 200-East Area.

Location	Assumed plume depth			
	5m	10m	15m	20m
200-West T-Plant				
Minimum contamination (retardation = 6.23)	159.4	318.6	476.8	622.4
Maximum contamination (retardation = 6.23)	2835.3	5665.0	8195.9	10249.8
Minimum contamination (retardation = 1.0)	0.1	0.1	0.2	0.2
Maximum contamination (retardation = 1.0)	1.1	2.2	3.2	4.0
200-West REDOX Plant				
Minimum contamination (retardation = 6.23)	1.2E+08	2.3E+08	3.3E+08	4.3E+08
Maximum contamination (retardation = 6.23)	8.0E+08	1.5E+09	2.3E+09	3.0E+09
Minimum contamination (retardation = 1.0)	1.1	2.2	3.2	4.2
Maximum contamination (retardation = 1.0)	7.7	14.8	21.9	28.6
200-East Area				
Minimum contamination (retardation = 6.23)	35.8	66.1	91.4	113.6
Maximum contamination (retardation = 6.23)	1645.6	3028.3	4247.8	5214.5
Minimum contamination (retardation = 1.0)	0.0	0.0	0.0	0.1

Table 2. Inventory upscale numbers for SAC modeled data for both the Underestimated Inventory and Overestimated Sorption Value Approaches.

how sorption influences the amount of iodine-129 contamination in the aqueous and solid phases.

Table 1 contains the SAC modeled data for the 200-West T-Plant Area, the 200-West REDOX Plant Area, and the 200-East Area. The original SAC iodine-129 inventories were used for all of the simulations. The VADER data row in this table shows the quantity of iodine-129 released for transport in the vadose zone, and hence is the maximum mass of iodine-129 that could be in groundwater if all were transported without sorption by the year the geostatistical analysis dealt with (2001). The other two rows show the quantity of iodine-129 predicted to reach the groundwater by 2001 using the STOMP vadose zone flow and transport module for two sorption assumptions. The first was with retardation of 6.23, the nominal value in the SAC data. The second STOMP simulation was made with retardation of 1.0, representing no sorption

of iodine-129 onto solid phase in the vadose zone. The second, zero-sorption case assisted us in determining if the retardation value assigned to iodine-129 in the SAC data is an accurate representation of the sorptive capacity of the underlying soils and sediments of the Hanford Site.

Table 2 compares the inventory upscale factors of the median (retardation = 6.23) and zero-retardation cases for all three areas obtained by dividing the observed mass in groundwater in 2001 from the geostatistical analysis for each of the assumed plume depths for each area by the VADER predicted total mass released to the vadose zone. Note that these factors account for the mass in the sorbed, solid phase. Minimum and maximums are provided, based on the extremes of the geostatistical analysis for iodine-129 mass in groundwater in year 2001. These numbers allow us to gain an understanding of how much additional iodine-129 would be necessary for each area to cause the SAC model to simulate the observed contamination mass in groundwater in 2001 for both the Underestimated Inventory Approach and the Overestimated Sorption Value Approach. Values are given for each of the four assumed plume depths used in the geostatistical analysis. Recall that the estimate of total mass itself from field data depends on the assumed plume depth. So, for example, the factors provided in Table 2 for the 5-meter assumed plume thickness are the factors necessary to upscale inventory of iodine-129 in order to cause the model to predict the mass in groundwater consistent with a 5-meter-thick uniform contamination plume assumption.

DISCUSSION AND CONCLUSION

A critical aspect of our research is determining the cause of the difference between the observed groundwater contamination plumes in 2001 and the fact that SAC Rev. 0 did not predict any groundwater contamination of iodine-129 by that year. The VADER data indicate the maximum amount of iodine-129 contaminants available for SAC to transport through the vadose

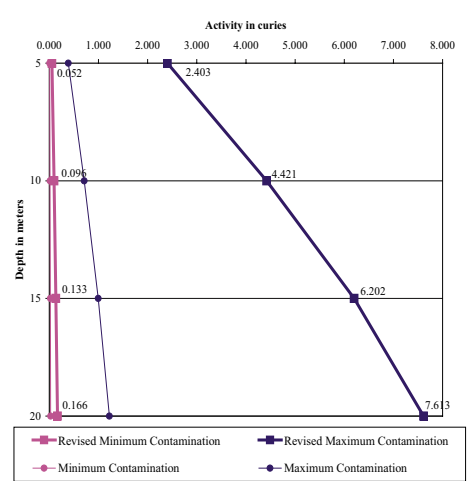
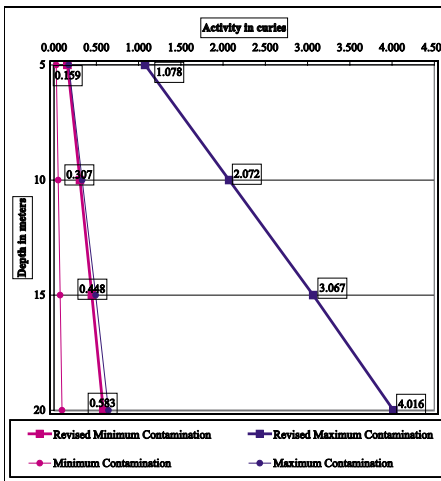
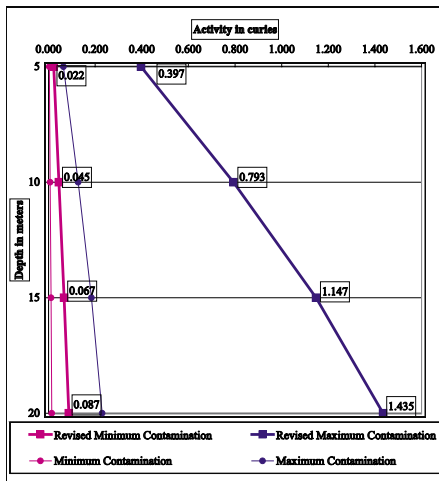


Figure 8. Revised geostatistical groundwater data for four assumed plume depths within the 200-West T-Plant Area.

Figure 9. Revised geostatistical groundwater data for four assumed plume depths within the 200-West REDOX Area.

Figure 10. Revised geostatistical groundwater data for four assumed plume depths within the 200-East Area.

zone and into the groundwater (Table 1). Analysis of this data is helpful for determining if inventory is low, sorption values are high, or if the discrepancy is a combination of both factors. Such analysis is tempered by the awareness that other factors could influence the discrepancy of concern (for example, the simplistic one-dimensional model of vadose zone release sites could lead to systematic under-prediction of iodine release for the most significant iodine release sites); while other potential factors are acknowledged, this study is limited to consideration of inventory and sorption.

200-West T-Plant Area

The 200-West T-Plant Area SAC VADER predictions (Table 1), representing the total quantity of iodine-129 in this area mobilized for transport by 2001, indicate that the inventory is at least adequate to produce the minimum contamination for the groundwater plume for all four assumed plume depths (Figure 8). However, the SAC model is unable to produce the maximum amount of contamination for any of those depths using the quantity of iodine-129 released to the vadose zone (Figure 8). This suggests that the inventory of iodine-129 is adequate to reproduce only the lower end of the range of observed groundwater contamination plume in the 200-West T-Plant Area.

Comparison of groundwater data for retardation 6.23 (Table 1) and the minimum amount of activity for the four plume depths (Figure 8) indicate that a retardation of 6.23 is too large to produce the observed amount of contamination for any of the four assumed plume depths. This strongly suggests that the sorption of iodine-129 is overestimated. The groundwater modeling data for the retardation = 1.0 case (Table 1) indicate that a K_d of 0 will produce the iodine-129 mass predicted assuming either 5 or 10 meter minimum contamination plumes (Figure 8). However, an iodine-129 value for K_d of zero (no sorption) is an inaccurate representation of the sorption abilities of iodine-129 because geochemistry data indicate that some amount of sorption occurs.

200-West REDOX Plant Area

The VADER predictions for the 200-West REDOX Plant Area (Table 1) indicate that there is not enough contamination at the top of the vadose zone for the SAC model to produce the minimum and maximum amount of contamination for any of the four assumed plume depths (Figure 9). This suggests that the inventory for the 200-West REDOX Area is underestimated for sources in this area.

The modeled groundwater contamination data for retardation = 6.23 indicate that there is insufficient contamination reaching the unconfined aquifer to produce the observed mass of iodine-129 in groundwater for any of four assumed plume depths (Figure 9). The STOMP data for $K_d = 0$ further indicates that there is insufficient contamination reaching the groundwater to produce the observed mass for any of the four assumed plume depths (Figure 9). These data suggest that

retardation of 6.23 is also too large to produce the iodine-129 plume in the 200-West REDOX Plant Area. The $K_d = 0$ data also suggest that iodine-129 inventory is underestimated in this area.

200-East Area

The SAC VADER data for the 200-East Area (Table 1) suggest that there is sufficient contamination to produce the minimum observed geostatistical groundwater contamination plume for all four assumed plume depths (Figure 10). The VADER data also indicate that there is adequate inventory to produce the necessary maximum contamination for an assumed plume depth of 5 meters (but not for the other assumed plume depths). These results suggest that the iodine-129 inventory is not significantly underestimated in this area.

The retardation = 6.23 data (Table 1) indicate that there is not enough iodine-129 mass reaching the groundwater in SAC simulations to produce the minimum or maximum amount of contamination for any of the four assumed plume depths (Figure 10). The retardation = 1.0 predictions (Table 1) do produce the amount of contamination needed to generate a the mass consistent with a 5-meter depth groundwater plume, but not the deeper assumed plume depths (Figure 10). These results indicate that the retardation factor of 6.23 overestimates iodine-129 sorption.

FUTURE RESEARCH

The results of this study are being used to carry out forward simulations of iodine-129 migration in the unconfined aquifer for both the median and zero K_d vadose zone release histories. The results of these groundwater simulations, when compared to the field data, are expected to yield improved understanding of the possible factors contributing to the iodine-129 modeling discrepancy (low inventory versus overestimated sorption rates). This knowledge will be used to refine the System Assessment Capability Rev. 1 data and model for the upcoming Composite Analysis scheduled for Fiscal Year 2004.

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