DETERMINING THE PLUME SOURCE DISCHARGE RATE, TRAILING EDGE, AND NATURAL ATTENUATION TIMEFRAME: THE F-AREA SAVANNAH RIVER SITE

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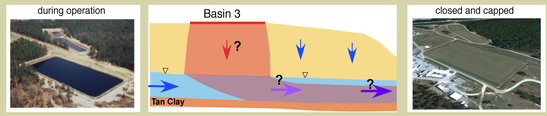

ABSTRACT

Understanding residual drainage from the source zone is essential for predicting attenuation of plumes at contaminated sites such as the Savannah River Site (SRS). This understanding is needed to predict remediation timeframes. At the SRS F-Area, large contaminant plumes resulted from decades of radioactive waste disposal into seepage basins. The objective of this work was to predict groundwater plume attenuation through characterizing its current status and developing a mass balance model for source zone drainage and dilution. Analyses of the sediments reveal the trailing plume edges for tritium and nitrate (Wan et al., 2012). Our mass balance model calculations for the plume in the SRS F-Area indicate that early stages of post-closure waste drainage occurred with high water fluxes (≈ 0.5 m y^{-1}), and declined over the 20 years since basin closure to low rates of a few cm y^{-1} (Tokunaga et al., in press). While the magnitude of late stage vadose zone drainage is low, its impact is large because of the high concentrations of contaminants it continues to supply to groundwater. The model predictions compared well with measured tritium and nitrate concentrations from downstream monitoring wells. The methodology presented here requires only groundwater monitoring data and a few well-constrained input quantities. Thus, this approach can be useful for gaining better understanding of contaminant dissipation at other sites as well.

We also conducted isotopic studies of U, Sr and nitrate to understand the plume's history. The isotopic analyses give no indication of biotic denitrification within the plume, although a surface water sample suggests denitrification occurs between the seep line and the stream. Both chemical and systematic isotopic vertical variations of U and Sr isotopes were identified in the upper aquifer. The data suggest that the lower zone represents the effect of the original contaminant fluids, while the upper zone mainly represents recharge of water contaminated by infiltration post-closure through the contaminated vadose zone. The U isotopic compositions of groundwaters suggest the migration rate of U from up-gradient portions of the plume of 100 m y^{-1}, slightly lower than the velocity (124 m y^{-1}) used in our models of basin drainage and plume attenuation.

HYPOTHESES

- A mass balance and Basin 3 vadose zone drainage analysis can be developed to reliably predict plume attenuation.
- The current plume is largely explained by Basin 3 drainage rates and the hydrogeochemical behavior of contaminants in the SRS environment.
- Isotopic signatures of contaminants can be interpreted to reveal plume evolution and fate.

APPROACHES

- Mass balance analyses of contaminants, based on historical monitoring data, groundwater flow, and net rainfall infiltration.
- Spatially distributed sediment sampling along plume path.
- Geochemical, mineralogical, and isotopic analyses of sediments.

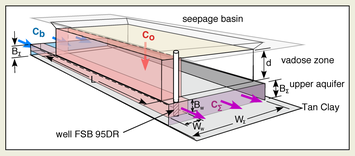

Fig. 1. Mass balance model: The time-dependent concentrations of contaminants at downstream well FSB 95DR reflect drainage of the contaminated vadose zone and dilution by upstream groundwater.

Mass balance model calculations: The time-dependent drainage rate from the contaminated vadose zone is

$$J_o(t) \approx J_{o0}(t-\Delta t) = \frac{B_w}{L} n v_p \left[\frac{C_{\Sigma,i}(t) - C_{b,i}}{C_{o,i} - C_{\Sigma,i}(t)} \right]$$

where C_o, C_b, and $C_\Sigma(t)$ are source zone, background, and downstream tracer concentrations, B_w is the downstream well screen interval (6.1 m), L is the basin length (219 m), n is the Upper Aquifer effective porosity (0.25), and v_p is the aquifer velocity (124 m y^{-1}).

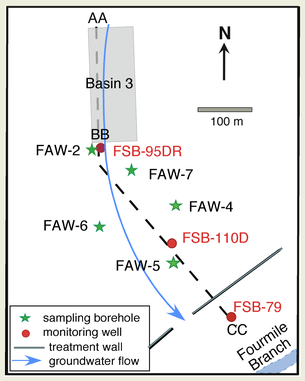

Fig. 2. Map of F Area seepage basin 3 and plume region, showing locations of long-term monitoring wells and boreholes for sediment sampling.

RESULTS

Modeled drainage rates from the contaminated vadose zone:

Using time trends in groundwater tritium, nitrate, and specific conductance immediately downstream of Basin 3 (Fig. 3) along with their source zone and background values (Table 1), we calculated vadose zone drainage rates (Fig. 4), and cumulative drainage (Fig. 5). Vadose zone drainage remains important because the source zone is large, and because its contaminants occur at levels much higher than regulatory goals.

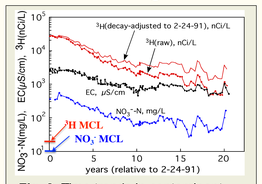

Fig. 3. Time trends in contaminant concentrations measured in the well immediately downstream of Basin 3 (FSB-95DR).

Table 1.	^3H	nitrate	EC
	nCi/L	mM	µS/cm
background $C_{o,i}$	2	0.02	5
Basin 3 $C_{o,i}$	85,800	87	10,050

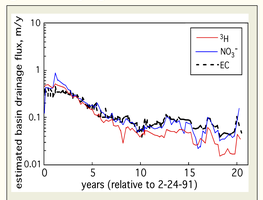

Fig. 4. Calculations of the time-dependent Basin 3 drainage fluxes into the Upper Aquifer, using decay-corrected tritium, nitrate, and specific conductance data as input in the mass balance model. Note that good agreement was obtained using the different tracers.

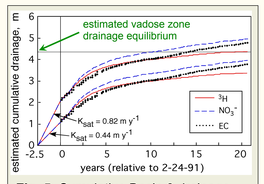

Fig. 5. Cumulative Basin 3 drainage fluxes into the Upper Aquifer, based on model calculations and early stage saturated drainage based on hydraulic conductivities (Phifer et al., 2006). Note (1) good agreement in slopes at "year zero", and (2) long-term convergence toward predicted drainage limit.

Contaminant distributions within the current plume:

In order to obtain a more complete picture of contaminant distributions beyond well FSB-95DR, 5 boreholes were drilled along the plume path and sampled with depth in the Upper Aquifer in order to develop transect maps of various contaminants (Fig. 6). The emergence of the trailing plume edge is evident from these transect maps. Rainfall recharge lowers contaminant concentrations in upper-most regions. Comparing Fig. 6d. and 6e shows pore water U(VI) distribution reflects pH-dependent sorption (Dong et al., 2012).

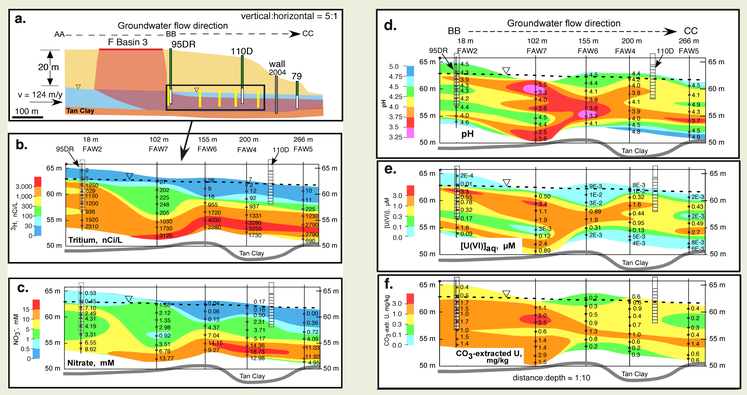

Fig. 6. F-Area Upper Aquifer plume contaminant distributions. (a.) Overview of transect in relation to Basin 3, monitoring wells, and funnel-gate treatment wall. (b.) tritium distribution. (c.) nitrate distribution. (d.) pH distribution. (e.) pore water U(VI) distribution. (f.) Carbonate-extracted sediment U(VI) distribution.

Predicting concentrations of contaminants within the plume:

Before the transport of reactive contaminants can be predicted, an understanding of the primary hydrogeological factors controlling flow is required. In the F Area plume, the key hydrogeologic factors are the background Upper Aquifer flow field, net rainfall infiltration (recharge), and Basin 3 drainage rates. The first 2 factors have been determined by previous SRS researchers. Our recent determination of drainage rates from Basin 3 now permits development of the basic plume flow model. By adding net rainfall infiltration to our drainage model, predictions of plume dilution (natural attenuation) can be made, and tested through comparisons with existing data (Fig. 7).

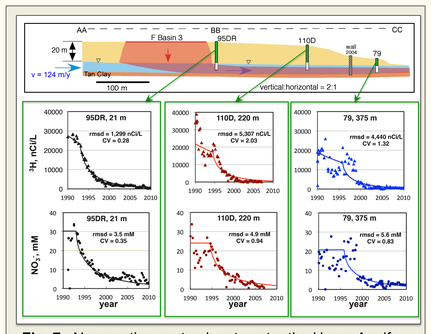

Fig. 7. Nonreactive contaminants enter the Upper Aquifer at diminishing drainage rates (Tokunaga et al., in press). Their concentrations are diluted by mixing with upstream uncontaminated groundwater, and with net rainfall infiltration. Dilution from rainfall infiltration increases with distance downstream. Our model was calibrated only for tritium at well FDB-95DR, and used to predict tritium and nitrate concentrations at locations much further downstream (220 m and 375 m). Predictions (curves) are in generally good agreement with data from monitoring wells (Wan et al., 2012).

RESULTS

Isotopic Studies:

In order to delineate the history of contamination and the current mobility and fate of contaminants in F-Area groundwater, we analyzed the isotopic compositions of U , Sr, Nd, H_2O (d18O, dD) and nitrate ($\delta^{15}N$, $\delta^{18}O$) within the contaminant plume. This data can be used to trace U transport within the plume, evaluate chemical changes of nitrate, and track plume/sediment chemical interaction. We have analyzed a suite of groundwater samples from monitoring wells, depth discrete samples from wells near the center and edge of the plume, as well as pore-water samples extracted from aquifer sediment cores to map out the isotopic variation within the plume. Below are selected results.

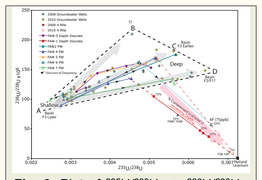

Fig. 8. Plot of $^{235}U/^{238}U$ vs. $^{236}U/^{238}U$. Four different contaminant end-members (A, B, C, D) are indicated by the U isotopic variation of groundwater/porewater samples. A, & C represent late and early, respectively, contamination from Basin F3, while D likely represents Basin F2/F1. Certain other samples indicate dilution by natural U sourced from sediment/porewater (FAW1 and FAW6E). Shown are patterns with depth for depth discrete groundwater and porewater samples.

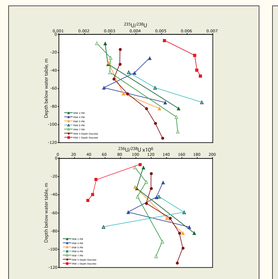

Fig. 9. Depth profiles of $^{235}U/^{238}U$ (top) and $^{236}U/^{238}U$ (bottom). The vertical variation in U isotopes implies the introduction of different contaminant compositions over time. The profiles also reveal different contaminant sources depending on location (e.g. deeper portion of FAW-7 influenced by Basins F2/F1).

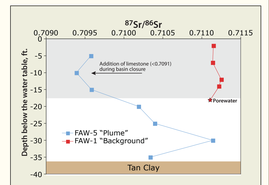

Fig. 10. Comparing $^{87}Sr/^{86}Sr$ in FAW5 and FAW1 suggests both the influence of the introduction of limestone (low $^{87}Sr/^{86}Sr$) during early remediation (upper zone, grey), and fluid/sediment interaction (lower zone, white). Upper and lower zones were defined on the basis of vertical chemical variations observed in FAW-5. The relatively low 87Sr/86Sr seen in the upper zone serves as both a time and source marker indicating the accompanying contaminant U migrated from the vadose zone since closure.

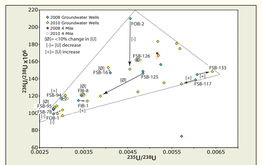

Fig. 11. Changes in U isotopic composition between 2008 and 2010 for sampled wells. For U concentration [+] indicates an increase, [-] a decrease, and [Ø] no change between 2008 and 2010. The observed U isotopic shifts for FOB 2 and FSB125 suggests an approximate groundwater flow rate of 100 m/yr (if no U retardation).

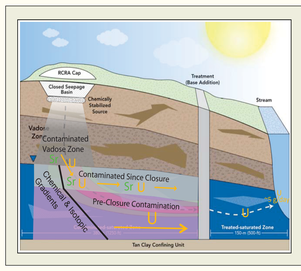

Fig. 12. Cartoon showing the over arching inferences from the isotopic study. Since closure, contaminants (e.g. U) have migrated from the contaminated sub-basin vadose zone to the upper 5 m of the aquifer. Recent migration rate of U within the aquifer appears to be on the order of 100 m/yr. Currently, with base addition, based on U isotopic analysis of surface water, about 5 g/day enters Fourmile Branch.

SUMMARY

- Our seepage basin drainage model demonstrates and explains the continued importance of contaminant discharge from the vadose zone.
- Current plume contaminant profiles reflect diminished source zone drainage and pH-dependent U(VI) sorption.
- Dilution of source zone drainage by background groundwater and net infiltration adequately predicts tritium and nitrate plumes. Our approach can help constrain much more complex models of plumes.
- Sr and U isotopic results indicate migration of U to the top 5 m of the aquifer since basin closure.
- Changes in groundwater U isotopic compositions over two years suggests the migration U from up-gradient portions of the plume at the rate of ~100 m/yr. U is contributed to Fourmile Branch at rate of ~ 5 g/day.
- The deepest porewater sample from FAW-6 has U isotopic composition consistent with a significant fraction (2/3 rds of the 75 ppb total U) of natural U extracted from the sediment.

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