MONITORING RADIONUCLIDE CONTAMINATION IN THE UNSATURATED ZONE—LESSONS LEARNED AT THE AMARGOSA DESERT RESEARCH SITE, NYE COUNTY, NEVADA

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Abstract—Contaminant-transport processes are being investigated at the U.S. Geological Survey's Amargosa Desert Research Site (ADRS), adjacent to the Nation's first commercial disposal facility for low-level radioactive waste. Gases containing tritium and radiocarbon are migrating through a 110-m thick unsaturated zone from unlined trenches that received waste from 1962 to 1992. Results relevant to long-term monitoring of radionuclides are summarized as follows. Contaminant plumes have unexpected histories and spatial configurations due to uncertainties in the: (1) geologic framework, (2) biochemical reactions involving waste components, (3) interactions between plume components and unsaturated-zone materials, (4) disposal practices, and (5) physical transport processes. Information on plume dynamics depends on *ex-situ* wet-chemical techniques because *in-situ* sensors for the radionuclides of interest do not exist. As at other radioactive-waste disposal facilities, radionuclides at the ADRS are mixed with varying amounts of volatile organic compounds (VOCs). Carbon-dioxide and VOC anomalies provide proxies for radioactive contamination. Contaminants in the unsaturated zone migrate along preferential pathways. Effective monitoring thus requires accurate geologic characterization. Direct-current electrical-resistivity imaging successfully mapped geologic units controlling preferential transport at the ADRS. Direct sampling of water from the unsaturated zone is complex and time consuming. Sampling plant water is an efficient alternative for mapping shallow tritium contamination.

INTRODUCTION

In 1976 the U.S. Geological Survey (USGS) began studies of water movement in the unsaturated zone at a site in the Amargosa Desert near a disposal facility for low-level radioactive waste (LLRW). In 1997 the site became part of the USGS's Toxic Substances Hydrology Program after unexpectedly high levels of tritium (^{3}H) were discovered in unsaturated-zone gas samples from the site (*1*). Ongoing studies are investigating the fate and transport of radionuclides and volatile organic compounds (VOCs) migrating from the facility, which accepted radioactive waste from 1962 to 1992. According to Nevada State records, 382 radioisotopes—comprising about 0.6 million curies of activity—were disposed in unlined trenches during the facility's 30-year operation. Tritium and radiocarbon (^{14}C) are the principal radionuclides moving through the unsaturated zone (*2, 3*). Both form gases and are biologically cycled; both are isotopes of elements essential to life. Tritium comprised ~60% of disposed activity. Calculations taking tritium's relatively short half-life (12.3 years) into account indicate that about half of disposed tritium had decayed by 1992. Radiocarbon (^{14}C) comprised about 1.5% of disposed activity. The relatively long half-life of ^{14}C (5,730 years) means that less than 0.2% of disposed activity had decayed by 1992. Predictive models of varying complexity have failed to account for observed spatial and temporal patterns of tritium transport in the 110-m deep unsaturated zone (*4, 5*). Direct measurements are therefore required to understand transport processes and to assess facility performance. The goal of research at the ADRS is to develop an improved understanding of transport processes in arid environments. Methods development supports this goal. The present paper summarizes research results applicable to monitoring radionuclides in the unsaturated zone. These results are particularly pertinent to arid and semi-arid regions, where much of the Nation's radioactive waste is interred. Additional information about the ADRS is available at http://nevada.usgs.gov/adrs/.

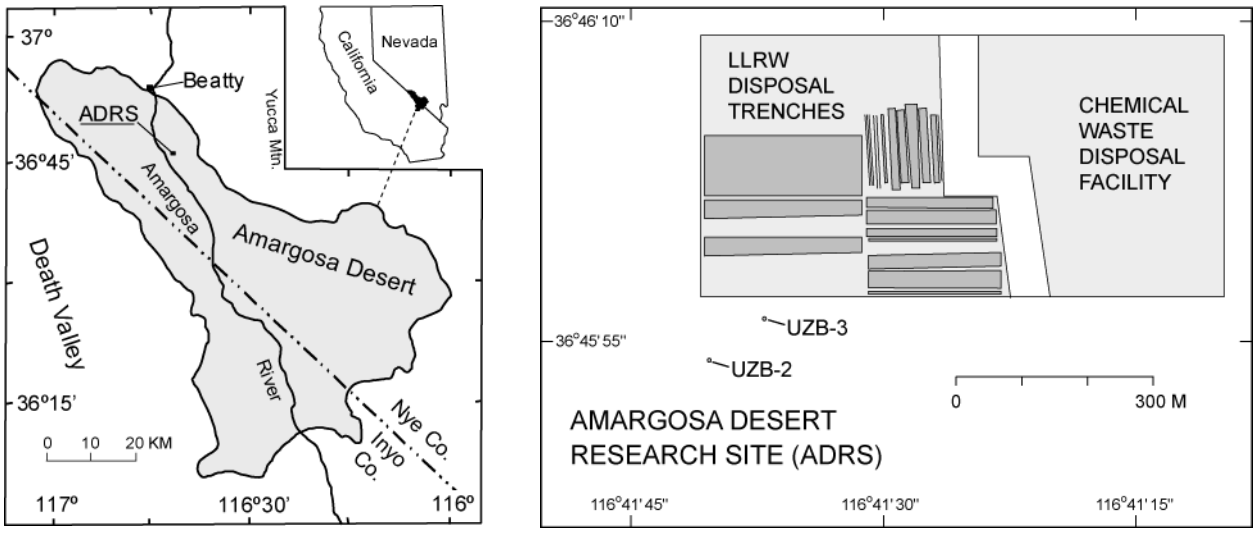

Fig. 1. Location of the Amargosa Desert Research Site (ADRS) and sampling boreholes UZB-2 and 3.

METHODS

Methods for sampling and analyzing unsaturated-zone and plant fluids for radionuclides and VOCs are described elsewhere (*3, 4, 6-8*). Unsaturated-zone gas samples are obtained from two deep boreholes, UZB-2 and UZB-3 (Fig. 1), and from a surrounding grid of shallow vapor probes (pipes driven 0.5 and 1.5-m deep). A larger grid of plant-sampling locations wraps around the south and west sides of the disposal facility. Deep boreholes have multiple screened intervals set in gravel layers 0.6-m thick. Gravel layers are sandwiched between transitional sand and sand-bentonite mixtures; intervening intervals are grouted with bentonite (*6*). Sample tubes lead from screens in the gravel layers to aboveground shelters.

VOCs are sampled by adsorption onto resin columns in the field, followed by desorption, separation by gas chromatography, and identification by mass spectroscopy in the laboratory (*7*). Tritium and radiocarbon emit only low-energy beta radiation and require collection and concentration by wet-chemical techniques in order to be detected. Water vapor (containing ^{3}H) is condensed from unsaturated-zone gases in a dry-ice trap in the field (Fig. 2), followed by electrolytic enrichment and scintillation counting in the laboratory (*6*). Carbon dioxide (containing ^{14}C) is collected in sodium-hydroxide solutions in the field (Fig. 3), followed by precipitation as barium carbonate and liquid scintillation counting in the laboratory (*3*).

Fig. 2. Sampling UZB3 for tritium.

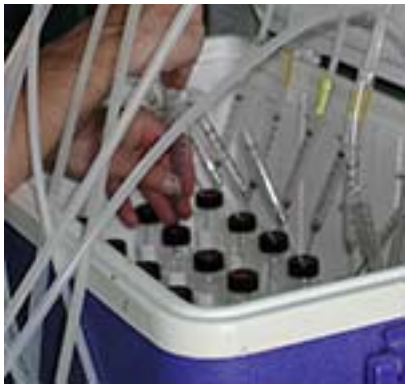

Fig. 3. Sampling UZB2 for radiocarbon.

Fig. 4. DC resistivity measurements.

Several geophysical techniques have been applied to characterize the subsurface at the ADRS, including ground-penetrating radar, surface-based seismic and electromagnetic-induction surveys, and borehole-based natural-gamma logging. The most useful in relation to observed patterns of contaminant transport is direct-current (DC) electrical-resistivity imaging, a surface-based technique employing automated,

inverse-Schlumberger-array soundings (*9,10*). Electrodes are driven into the ground in a line and connected to a common cable by electronic switches (Fig. 4). A programmable controller sends current through the ground using sequential pairs of central electrodes of varying spacings. Voltages measured at non-current pairs of electrodes yield values of apparent resistivity, assuming a uniform half space. Numerical inversion creates a distribution of actual resistivities (the resistivity image) corresponding to modeled apparent resistivities that best match the measured apparent resistivities.

RESULTS AND DISCUSSION

Gradients of water content, matric potential, temperature, and carbon dioxide are directed upward beneath undisturbed native vegetation adjacent to the LLRW disposal facility (Fig. 5.). The upward gradient of matric potential (second panel) exceeds the downward gradient of gravitational potential, indicating an upward (albeit slow) movement of water through the unsaturated zone from the water table under current climatic conditions (*11*). Heat and CO_2 are moving upwards along geothermal and concentration gradients as well. The accumulation of large quantities of atmospherically deposited chloride just beneath the root zone (fifth panel) supports the interpretation of upward water flow, showing that the lack of deep percolation (and ground-water recharge) extends back in time thousands of years (*12, 13, 14*).

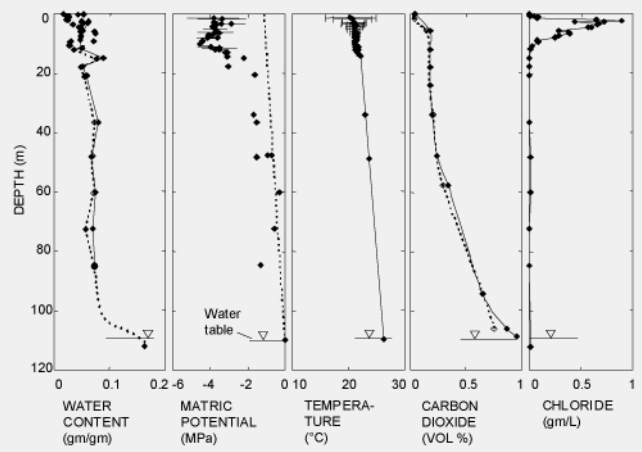

Fig. 5. Profiles at (and near) UZB-2 reveal upward-directed gradients and shallow chloride accumulation (*11, 12, 14*). The dotted line in the second panel represents hydrostatic equilibrium.

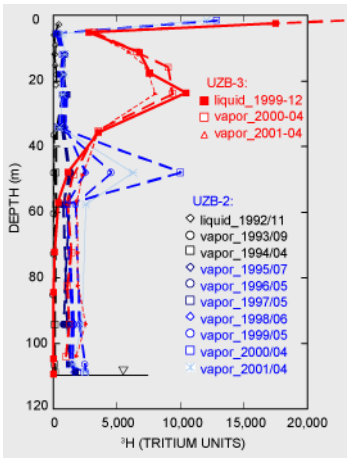

Fig. 6. Tritium (^3H) in unsaturated-zone water at UZB-2 and 3. Liquid samples obtained per Prudic et al. (*6*).

Superimposed on the upward directed natural flow field, contaminants containing tritium and radiocarbon are migrating laterally away from the disposal area along individual concentration gradients (Figs. 6 and 7). This transcurrent movement of contaminants involves exceedingly low masses of gases.

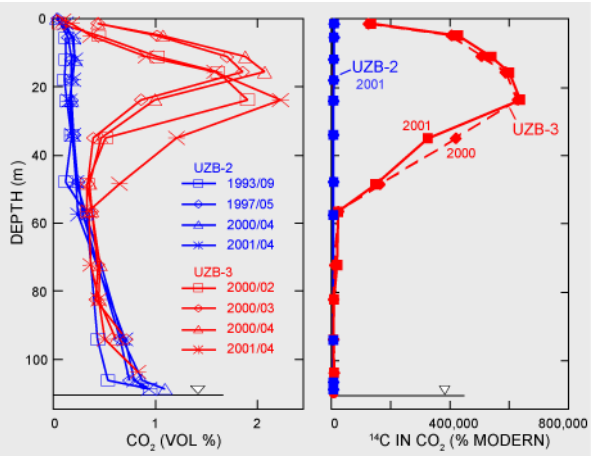

Fig. 7. CO_2 and ^{14}C in unsaturated-zone gases at UZB-2 and 3.

Water sampled from UZB-2 showed sharp increases in tritium between 1992 and 2001 that were not associated with CO_2 anomalies (Figs. 6, 7). In contrast, the occurrence of ^{14}C contamination is highly correlated with CO_2 anomalies. Sensors based on infrared light absorption can readily detect CO_2 anomalies of this magnitude.

The association of CO_2 with radiocarbon suggests that microbial metabolism of radioactive organic compounds is the dominant process mobilizing ^{14}C. Analysis of stable carbon (^{13}C) in CO_2 supports this interpretation. Microbial respiration

produces CO_2 depleted in ^{13}C. The upper part (5–24 m) of the UZB-3 profile is depleted by about 15 permil compared to that of a control borehole, ~3 km to the south of the waste-disposal facility (*3*).

Rapid transport of tritium is inconsistent with predictions of process-based numerical models (*4, 5*). These models, all of which fail to reasonably account for observations, assume transport of ^{3}H as water vapor (H_2O_{gas}) in isotopic equilibrium with pre-existing (uncontaminated) unsaturated-zone water. Various possibilities for resolving the discrepancy are being investigated. Isotopic disequilibrium is one possibility. Another is that ^{3}H transport occurs as methane (CH_4). Methanogenesis is expected in anoxic portions of the waste pile; indeed, detectable levels of methane (CH_4) have been measured in both UZB-2 (up to ~ 9 ppm) and UZB-3 (up to ~18 ppm). However, observed methane anomalies are much broader than the observed tritium peaks at UZB-2 and 3. This poor correlation probably reflects vagaries of waste emplacement and indicates that methane is a less reliable surrogate for tritium than CO_2 is for radiocarbon. Generation of tritiated methane would require tritiated water contacting organic waste in the presence of anoxic conditions (produced, for example, by corroding steel drums). Non-tritiated water would produce non-tritiated methane.

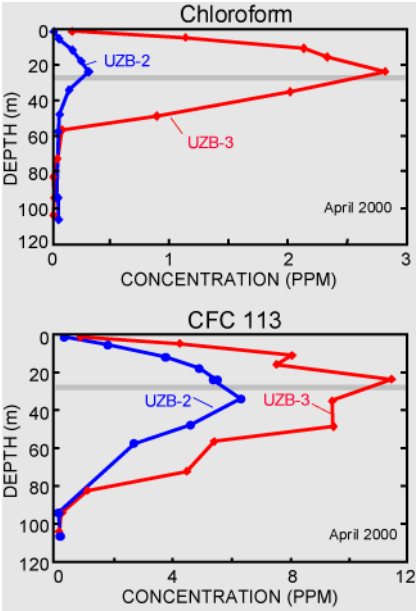

Fig. 8. Chloroform and CFC-113 in unsaturated-zone gases at UZB-2 and 3.

Volatile-organic-compound (VOC) anomalies also indicate radioactive contamination (Fig. 8). Unsaturated-zone gases at the ADRS were analyzed for 88 different VOCs. Fifteen compounds were present at greater than parts-per-million (ppm) levels. Molecular structure, by controlling biochemical reactions and physical interactions with unsaturated-zone materials, controls transport. For example, chlorofluorocarbons like CFC-113 show faster horizontal and vertical migration than chloroform, despite having significantly higher molecular weights (Fig. 8).

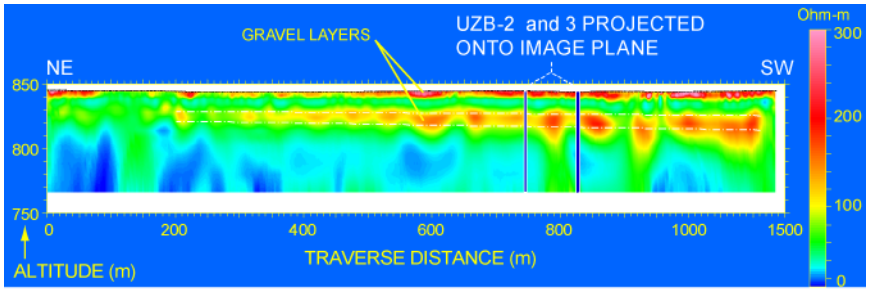

Fig. 9. A vertical cross-section made by direct-current electrical resistivity (DC-resistivity) imaging near UZB-2 and 3 shows gravel layers that thicken toward the center of the basin, providing preferential pathways for contaminant migration.

Chlorofluorocarbons are less soluble in water than chloroform. Thus CFCs partition to a lesser extent than chloroform into unsaturated-zone water. In addition, CFCs are chemically stable in unsaturated-zone environments. These properties, together with the fact that CFCs are common in low-level radioactive waste, make CFCs a good early-warning indicator of radionuclide contamination.

Radionuclides migrate preferentially in the unsaturated zone (Figs. 6–8). Sediments at the ADRS consist mainly of sub-horizontal alluvial sands and gravels. Areally extensive coarse gravel deposits exist at depths of ~1-2 meters (immediately below the root zone), and at ~23-25 meters. DC-resistivity images show these deposits as zones of high resistance (Fig. 9). DC-resistivity imaging can help guide placement of monitoring points.

Movement of tritium through the shallow unsaturated zone can be efficiently mapped by analyzing plant water from creosote bushes (*Larrea tridentata (DC) Colville*) carpeting the desert floor (Figs. 4, 10). Creosote bushes, highly adapted to xeric conditions, form a near monoculture near the LLRW disposal facility. A method for rapidly extracting and accurately analyzing leaf water was developed at the ADRS (*8*). The method is five times faster than direct sampling of soil-water vapor and avoids installation of

unsaturated-zone vapor probes. Although the aboveground portions (canopies) of creosote bushes are sparse, their roots fairly completely fill the soil volume above the shallow gravel layer. Creosote bushes are highly effective at scavenging soil moisture; indeed, extraction of water by roots is a main factor contributing to the overall upward moisture gradient in the unsaturated zone (*13, 14*; Fig. 5). Creosote-bush samples may be better than traditional samples obtained from driven soil-gas probes with respect to obtaining volume-integrated samples. In any case, the tritium distribution inferred from plant water is in good agreement with that inferred from directly sampled soil-water vapor, demonstrating the largely untapped potential of biological monitoring (Fig. 10).

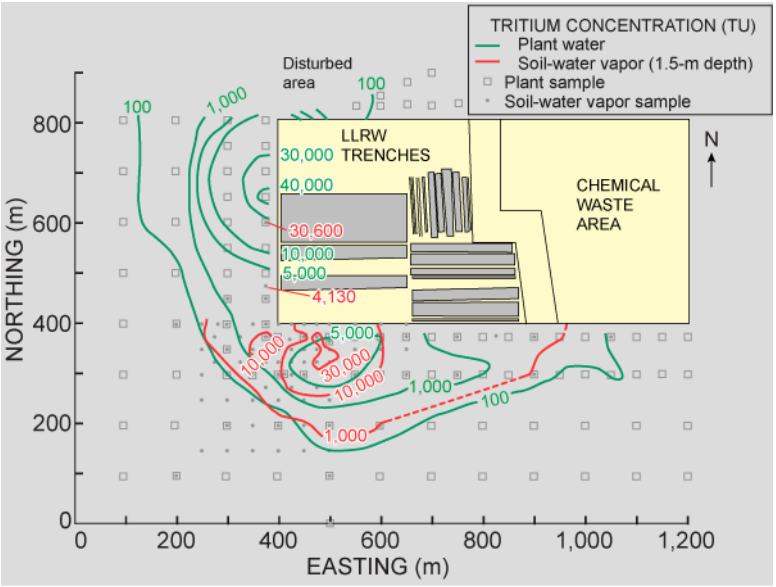

Fig. 10. Comparison of plant-water tritium with soil-water tritium sampled from 1.5 m.

CONCLUSIONS

Radionuclides consisting primarily of tritium and radiocarbon are migrating from a low-level radioactive-waste disposal area adjacent to the Amargosa Desert Research Site (ADRS). The water table is 110-m deep, thus, transport through the unsaturated zone is the primary research interest. ADRS results are relevant to long-term performance monitoring of contaminated sites in arid environments. Geologic structure strongly controls contaminant transport, as shown by preferential migration through laterally extensive gravel deposits. These deposits are readily mapped by direct-current electrical-resistivity imaging, a non-invasive surface-based geophysical technique. Plant-water sampling of creosote bushes provides a fast and accurate alternative to direct pore-gas sampling for tritium contamination in the shallow unsaturated zone. Available *in-situ* sensors cannot detect radionuclides of interest; however, carbon-dioxide anomalies provide a readily detected indicator of radiocarbon contamination. Chlorofluorocarbon anomalies provide a more general early-warning sign of radionuclide migration.

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