

IMPLICATIONS OF BOMB ^{36}Cl AND BOMB TRITIUM STUDIES FOR GROUND WATER
RECHARGE AND CONTAMINANT TRANSPORT THROUGH THE
VADOSE ZONE

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The vadose zone (zone between the land surface and the water table) is both the interval through which most ground water recharge moves and the avenue of most movement of contaminants into ground water. Knowledge of how water and solutes move through the vadose zone is thus critical both to ground water management and to aquifer protection. The movement of water and solutes under agricultural fields and humid region soils is fairly well understood. Numerous tracer studies, in the field and in the laboratory, have demonstrated the nature of soil water transport under these conditions.

Unfortunately, the transport of water and contaminants in desert soils is understood far less well. This is in part because there is less economic incentive to investigate water in natural desert soils. It is also due to the nature of the physical processes involved. Water typically moves downward under agricultural soils at the rate of several decimeters per month. In contrast, under desert soils the flow is on the order of centimeters per year. Meaningful tracer experiments would thus take 20 to 40 years, a prohibitively long time.

This difficulty may be overcome through the application of environmental tracers. Environmental tracers are substances that are present in the environment due to natural or anthropogenic phenomena, rather than the efforts of the investigator, and that have desirable properties for tracing the movement of soil water. These properties include chemical stability, lack of sources in the soil, and lack of adsorption on the solid phase. The "classic" soil water tracers for laboratory experiments are tritium (^3H , incorporated in the water molecule as ^3HHO), a tracer for the water itself, and ^{36}Cl (as the $^{36}\text{Cl}^-$ anion), a tracer for conservative solutes (Biggar and Nielsen, 1962; James and Rubin, 1986). The presence of these radioisotopes as environmental tracers would greatly facilitate comparison of field studies with analogous laboratory experiments.

Fortuitously, atmospheric nuclear weapons testing in the 1950s and 1960s has provided us with pulses of atmospheric fallout of tritium and ^{36}Cl . Chlorine-36 was produced by neutron activation of chloride in seawater during the U.S. explosions at Bikini and Enewetok atolls. The fallout of ^{36}Cl and tritium as a function of time is illustrated

in Figure 1. Chlorine-36 fallout is taken from Bentley et al. (1986) and tritium fallout from IAEA (1983).

During the fallout period, the radioisotopes entered the soil surface along with precipitation or as dry fallout. Since that time they have been moving downward along with the soil water. By measuring vertical profiles of the tracers, some basic questions can be answered: (1) How fast does water move down through desert soils? (2) How does the movement of solutes compare to that of the water? (3) How much do solutes spread out (disperse) during flow?

Field Studies

In order to address these questions we performed ^{36}Cl and tritium measurements on samples from vertical auger holes at three sites in central and southern New Mexico. The first site (designated SNWR 1) was in a sandy soil forming an old floodplain of the Rio Salado, on the Sevilleta National Wildlife Refuge 20 km north of Socorro. The second site (SNWR 2) was close to the first, but on a Pleistocene terrace above the river. The soil was a sandy loam at this site, in contrast to the well-sorted fine sand at the first. The third site (NMSUR) also had a sandy loam soil. It was located on the New Mexico State University Ranch north of Las Cruces. The sites are described in greater detail in Mattick et al. (1987).

Water was removed from the soil by azeotropic distillation and analyzed for tritium by direct liquid-scintillation counting. Chloride was leached from the soil with deionized water and analyzed for ^{36}Cl by means of accelerator mass spectrometry (Elmore et al., 1979). Results are shown in Figure 2.

In both cases where tritium and ^{36}Cl were measured together, the result is the opposite of what might have been expected. In spite of the fact that the ^{36}Cl fallout peaked earlier than the tritium, the tritium pulse is found deeper in the soil. This result is particularly anomalous in light of the anion exclusion phenomena which normally causes ^{36}Cl to move faster than tritium even if the two are introduced together (Krupp et al., 1972; Gvirtzman et al., 1986). We tentatively attribute this apparent retardation of the chloride to microscopic-scale vapor-phase transport of the water in the dry desert soils. The tritium moves along with the water molecules in the vapor phase, but the ^{36}Cl must follow much more tortuous paths in the liquid phase and thus moves downward more slowly. The evidence for this hypothesis is discussed in Phillips et al. (1987).

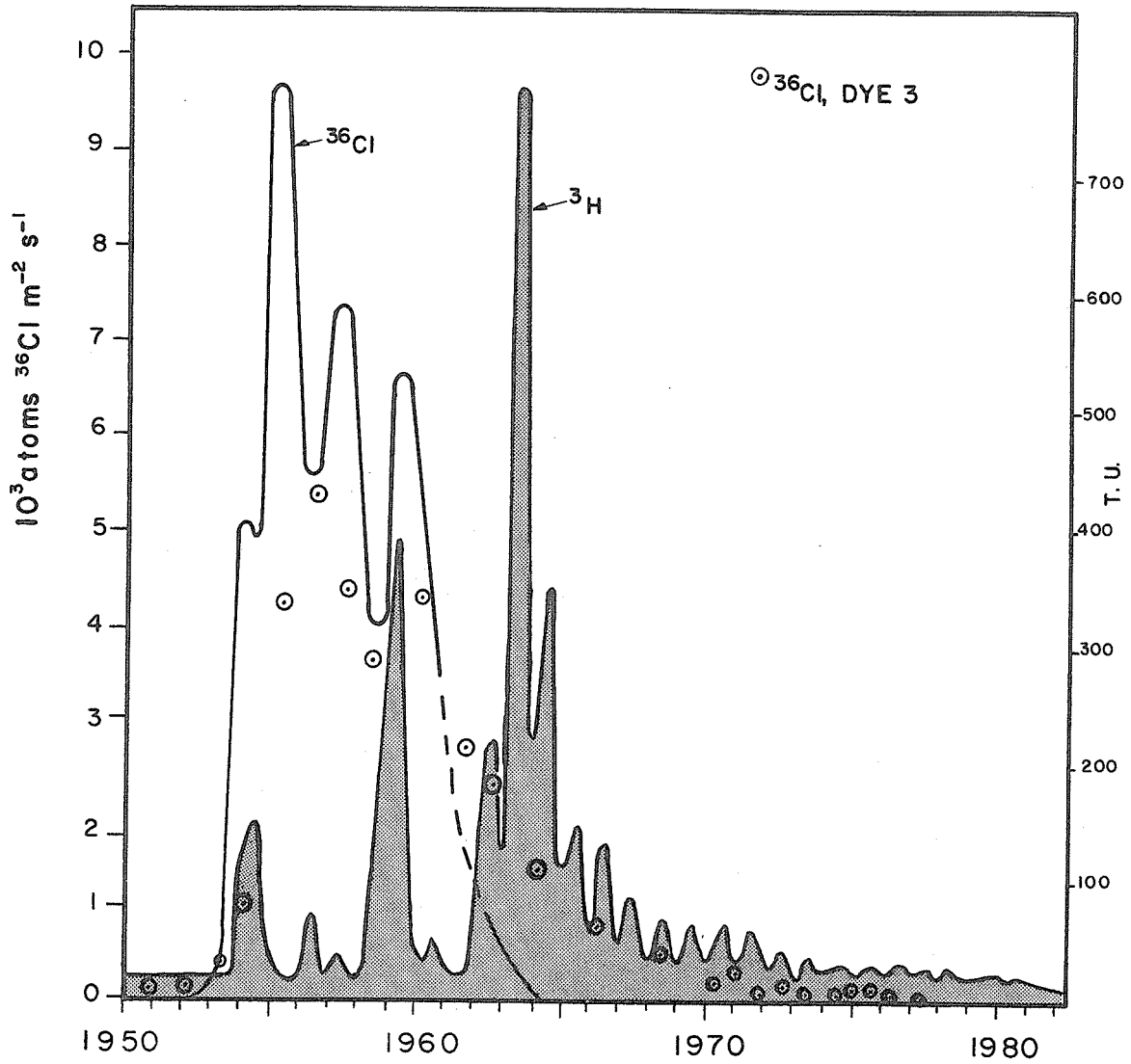


Figure 1. A comparison of average northern hemisphere bomb- ^3H fallout (IAEA, 1983), decay corrected to 1985, with calculated mean global bomb- ^{36}Cl fallout (Bentley et al., 1986).

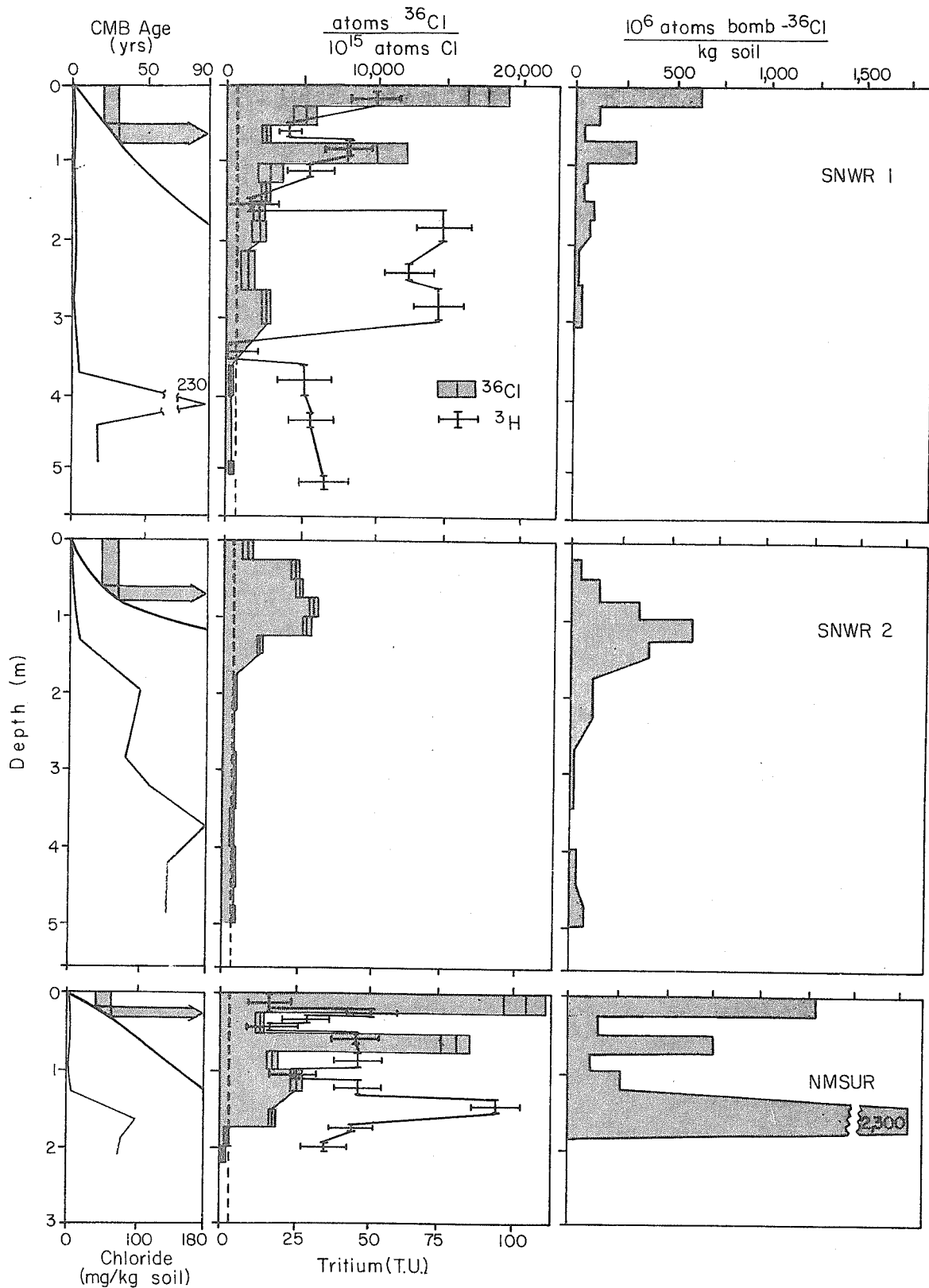


Figure 2. Chloride, chloride mass balance (CMB) age, ^3H , $^{36}\text{Cl}/\text{Cl}$ ratio, and ^{36}Cl content as a function of depth at the SNWR 1, SNWR 2, and NMSUR sites.

The transport of the tritium and ^{36}Cl were simulated using numerical modeling of the advection-dispersion equation (Mattick et al., 1986). The ^{36}Cl profile at the SNWR 2 site could be simulated using an apparent dispersivity of 8.0 cm, and the tritium distribution at SNWR 1 using an apparent dispersivity of 5.3 cm. Much of the tritium dispersion may be due to vapor diffusion. Variable velocities in both space and time are probably another major contributor to the apparent dispersion.

Conclusions

The combination of ^{36}Cl and tritium from nuclear weapons fallout has proved to be a successful environmental tracer for long-term water and solute movement in natural desert soils. The information that they provide is equivalent to that from a 30-year-long artificial tracer experiment. Some of the results from the tritium and ^{36}Cl tracing provide unexpected insights into transport processes in desert soils.

1. The downward velocity of the soil water at SNWR 1 (computed from the penetration depth of the tritium peak) is about 15 cm/yr and at NMSUR it is about 7 cm/yr. This difference could be anticipated, given the more clay rich soil texture at NMSUR. However, the net specific flux to the depth of the tritium peak at the two sites is remarkably similar, about 0.9 cm/yr in both cases. This value is in good agreement with estimates by Stephens and Knowlton (1986) for the SNWR 1 site, based on soil-physics monitoring techniques.
2. Chloride is retarded relative to tritium at both sites, rather than moving faster as in previous laboratory and agricultural field experiments. This is probably due to water movement in the vapor phase (on a microscopic scale) as well as the liquid phase. This property of desert soils may be very beneficial from the viewpoint of retarding the movement of contaminants through the vadose zone to the water table.
3. The amount of dispersion is much larger than in laboratory experiments on the same scale. This is not unexpected, given the much more variable boundary conditions and heterogeneous materials in the natural situation. The greater dispersion of ^{36}Cl than tritium is unexpected, inasmuch as tritium can diffuse in both the vapor and liquid phases whereas ^{36}Cl can move only in the liquid. Again, this may be due to the more tortuous paths that some of the ^{36}Cl must follow in the liquid phase, but which the tritium can avoid by means of vapor transport. The absolute magnitude of the observed dispersion of either tracer is not particularly

large and does not indicate that early arrival of contaminants at the water table due to accelerated dispersive transport is likely to be a major problem.

4. The field results reported here indicate that standard laboratory experiments are of limited value in predicting the movement of water and solutes in natural desert soils. The differences are probably largely due to the much slower water velocities, longer time scales, and greater importance of vapor diffusion at low water contents. Further field investigations in desert soils should yield fundamental insights into the transport processes in this environment.

Acknowledgments

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REFERENCES

- Bentley, H.W., Phillips, F.M., and Davis, S.N. 1986. Chlorine-36 in the terrestrial environment. Handbook of Environmental Isotope Geochemistry. 2B:427-480.
- Biggar, J.W., and Nielson, D.R. 1962. Miscible displacement: II. Behavior of tracers. Soil Sci. Soc. Am. Proc. 26:125-128.
- Elmore, D., Fulton, B.R., Clover, M.R., Marsden, J.R., Gove, H.E., Naylor, H., Purser, K.H., Kilius, L.R., Beukens, R.P., and Litherland, A.E. 1979. Analysis of ³⁶Cl in environmental water samples using an electrostatic accelerator. Nature. 277:22-25.
- Gvirtzman, H., Romen, D., and Margaritz, M. 1986. Anion exclusion during transport through the unsaturated zone. J. Hydrol. 87:267-283.
- IAEA (International Atomic Energy Agency) 1983. Isotope Techniques in the Hydrological Assessment of Potential Sites for the Disposal of High-level Radioactive Wastes. IAEA Tech. Rept. Series No.228. Chapter 7 "Tritium", Vienna.
- James, R.V., and Rubin, J. 1986. Transport of chloride ion in a water-unsaturated soil exhibiting anion exclusion. Soil Sci. Soc. Am. J. 50: 1142-1149.
- Krupp, H.K., Biggar, J.W., and Nielsen, D.R. 1972. Relative flow rates of salt and water in soil. Soil Sci. Soc. Amer. Proc. 36:412-417.

- Mattick, J.L., Duval, T.A., and Phillips, F.M. 1987. Quantification of Groundwater Recharge Rates in New Mexico Using Bomb ^{36}Cl , Bomb ^3H and Chloride as Soil-Water Tracers. New Mexico Water Resources Research Institute, Technical Report No. 220, New Mexico State University, Las Cruces, New Mexico.
- Phillips, F.M., Mattick, J.L., Duval, T.A., Elmore, D., and Kubik, P.W. 1987. Chlorine-36 and tritium from nuclear weapons fallout as tracers for long-term liquid and vapor movement in desert soils. Submitted to Water Resources Research.
- Stephens, D.B., and Knowlton Jr., R. 1986. Soil-water movement and recharge through sand at a semi-arid site in New Mexico. Water Resour. Res. 22: 881-889.