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Use of Carbon-14 and CFCs to Study Gas Movement and Reactions in the Unsaturated Zone

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Gasses in the unsaturated zone were measured at two sites above the High Plains Aquifer in the central United States. The measurements included carbon-14 and carbon-14 ratios in carbon dioxide, a series of chloroflourocarbons (CFC-11, CFC-12 and CFC-113), and nitrous oxide and carbon dioxide concentrations. One of the sites is a field that has been undergoing irrigation for the past 50 years (UMA) and primarily grows corn. The other location is rangeland that has not been farmed or irrigated in the past century (IMP). Depth to water table is about 28 m at the rangeland site and presently about 47 m at the irrigated site. The depth to water table has increased about 15 m since the initiation of irrigation in the late 1950s at UMA. Tritium profiles in the UNZ show a prominent peak from nuclear weapons testing at about 12 m at IMP and about 25 m at UMA. The tritium peak at UMA is spread over a larger depth range and is not as pronounced, suggesting that irrigation has increased recharge. Carbon dioxide concentrations showed no consistent trends at IMP, being about 3500 ppmv. At UMA concentrations are 6000-7000 ppmv throughout most of the profile below the root zone, but decrease to about 3500 ppmv at the deepest sample. The difference in the two carbon dioxide profiles may be due to the fact that much more carbon dioxide is produced in the root zone at the irrigated site. It is likely that the enhanced production has not yet fully equilibrated throughout the profile. Carbon-13 shows no trends with depth with ratios ranging between -11.5 o/oo and -14.8 o/oo at the two sites. CFCs show a decrease with depth at both sites, most of which is likely due to the atmospheric increase of CFCs until the mid to late 1990s. CFC concentrations near the

ground water interface are similar to atmospheric concentrations during the late 1980s at the IMP site and the late 1970s at the UMA site. The CFC data demonstrate that the non-reactive UNZ gasses can equilibrate with the atmosphere on a timescale of a couple of decades. Production of carbon dioxide within the SUIR (Saturated/Unsaturated Interface Region) would result in an increasing trend in carbon dioxide concentrations with depth which is not seen. There is little evidence from nitrous oxide data that biochemical reactions are producing gasses in the UNZ below the surface layers.

Carbon-14 concentrations decrease with increasing depth much faster than would be anticipated from gas phase diffusion alone. Atmospheric carbon-14 concentrations did increase during the bomb transient, but the increase would not result in the profiles found. It is an indication that chemical processes must be important in controlling carbon-14 concentrations in the UNZ. Options could include production of CO_2 by microbial processes in the UNZ, exchange with carbonate in groundwater, or reactions with low ^{14}C carbonate within the UNZ. Exchange of atmospheric carbon dioxide occurs on a decadal scale so it is possible that the groundwater could have an impact on the carbon-14 concentrations in the UNZ. The carbon-13 values also indicate that exchange is a possible control on the isotopic content of the carbon dioxide near the water table. A combination of exchange with groundwater and carbonates within the UNZ may explain the ^{14}C distributions in these profiles. The combination of CFCs and ^{14}C in CO_2 makes it possible to calculate these rates by accounting for gas diffusive processes independent of reactive processes.