5. TRITIUM IN THE HYDROLOGIC CYCLE

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1. Introduction

Tritium, the radioactive isotope of hydrogen of mass three and a half-life of 12.32 years, is one of the most important transient tracers used in hydrological research. After the discovery of natural carbon-14 in the late 1940s, it was suspected that tritium and other radionuclides might also be produced by cosmic-ray spallation. Early studies of heavy water produced by electrolysis indicated that natural tritium was present in the environment at a level of about $1 \times 10^{-18}$ moles of tritium to 1 mole of hydrogen (Grosse et al., 1951). This led to the designation of this ratio as the now familiar tritium unit (1 tritium atom/$10^{18}$ atoms of hydrogen). Although the major natural source of tritium on the earth is the spallation of atmospheric molecules by cosmic rays, a small amount is also produced by natural fission and reactions with thermal neutrons in the lithosphere (Lehmann et al., 1993). However, this production is rarely of importance in hydrologic studies. The natural production by cosmic rays results in a steady-state inventory on the Earth’s surface of about 3.5–4.5 kilograms of tritium (Lal and Peters, 1967; O’Brien et al., 1992), most of which is present as part of the water molecule (HTO). As part of the water molecule, tritium follows the pathway through the hydrologic cycle almost exactly, with only small perturbations due to fractionation effects during phase changes (Bigeleisen, 1962). These fractionation effects are usually small and can be ignored relative to measurement uncertainties and the larger change resulting from radioactive decay. Consequently, information on time scales for hydrologic processes can be obtained from tritium measurements (Buttlar and Libby, 1955).

Tritium is one of the nuclides produced during nuclear weapon testing. Beginning in 1953, the atmospheric testing of fusion weapons dramatically increased the inventory of tritium on the Earth’s surface, with the highest increase resulting from the nuclear tests of 1962 (Eriksson, 1965; Carter and Moghissi, 1977). These atmospheric bursts increased the tritium inventory on the Earth’s surface by more than two orders of magnitude, with most of the tritium finally ending up in the world oceans (Miskel, 1973; Michel, 1976). Tritium concentrations in rain prior to nuclear testing were of the order of a few tritium units, with highest concentrations occurring in mid-continental areas. Tritium concentrations in
precipitation peaked in the northern hemisphere during 1963, rising to a few thousand tritium units in most mid-continental locations (IAEA, 1981, 1992). Smaller increases were seen in the tritium concentrations of oceanic and continental surface waters in the mid-1960s (Stewart, 1966; Dockins et al., 1967). After deposition on the continents, tritium entered into the vadose zone, surface water and groundwater. The value of this tritium transient for studying hydrologic processes was quickly recognized (IAEA, 1962; Suess, 1969), and for the past five decades tritium has been widely used to obtain time scales for physical mixing processes in oceanographic and hydrologic systems. In hydrology, tritium has proved useful in studies on the movement of water through the unsaturated zone, on time scales for physical processes occurring in surface waters and groundwaters, and interactions between groundwater and surface water systems. Frequently, tritium has been able to provide information on time scales that are not easily obtained from other methods or to furnish a baseline against which results from other methods could be compared. Below, we discuss some of the approaches that have been used in studies of hydrologic processes with tritium data.

2. Tritium in the atmosphere

Initially, most tritium is produced in the atmosphere, where it enters the hydrologic cycle. Tritiated water within the troposphere can be deposited quickly in the ocean and other surface bodies of water, either by direct precipitation or by molecular exchange across the air–water interface. It is generally thought that molecular exchange is the dominant mechanism for removal of tritium from the atmosphere and deposition into oceans and lakes (Ostlund and Berry, 1970). Pre-bomb tritium concentrations are not well known, although some estimates are available which suggest they were on the order of 2–8 tritium units across the continental USA (Thatcher, 1962). Tritium concentrations in precipitation rose rapidly following the atmospheric bomb tests and were found to have a strong dependence on location and season (Libby, 1962; Gat et al., 1962; Eriksson, 1965). Tritium concentrations inland tended to be much higher than those at coastal or mid-ocean locations due to the dilution of atmospheric tritium concentrations by the low concentrations found in ocean waters. There is also a major seasonal difference caused by the interaction of the stratosphere and troposphere. During the northern hemisphere Spring of each year, the warming of the land masses causes an instability in the tropopause between 30° and 60°N. This break-up results in the mixing of stratospheric air into the troposphere. The stratosphere is the main repository for atmospheric tritium, and since water vapour in the stratosphere cannot exchange directly with water vapour in the ocean, the tritium concentrations in that reservoir remain higher than those in the troposphere. As a result, there is an increase in tritium concentrations in the troposphere of the northern hemisphere each year, frequently referred to as the “Spring Leak”. This atmospheric pattern, combined with the fact that most of the major nuclear tests occurred in the northern hemisphere, has resulted in a strong latitudinal gradient in tritium concentrations within the atmosphere. Tritium concentrations are higher in the northern hemisphere with the maximum tritium concentrations occurring at about 50° to 60°N. Tritium