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Analysis of air mass trajectories to explain observed variability of tritium in precipitation at the Southern Sierra Critical Zone Observatory, California, USA

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ABSTRACT

Understanding the behavior of tritium, a radioactive isotope of hydrogen, in the environment is important to evaluate the exposure risk of anthropogenic releases, and for its application as a tracer in hydrology and oceanography. To understand and predict the variability of tritium in precipitation, HYSPLIT air mass trajectories were analyzed for 16 aggregate precipitation samples collected over a 2 year period at irregular intervals at a research site located at 2000 m elevation in the southern Sierra Nevada (California, USA). Attributing the variation in tritium to specific source areas confirms the hypothesis that higher latitude or inland sources bring higher tritium levels in precipitation than precipitation originating in the lower latitude Pacific Ocean. In this case, the source of precipitation accounts for 79% of the variation observed in tritium concentrations. Air mass trajectory analysis is a promising tool to improve the predictions of tritium in precipitation at unmonitored locations and thoroughly understand the processes controlling transport of tritium in the environment.

1. Introduction

Understanding the behavior of tritium, a radioactive isotope of hydrogen, in the environment is important to evaluate the exposure risk of accidental and routine releases from nuclear power, medical and industrial activities. In addition, tritium is an important tracer for hydrology and oceanography (Kaufman and Libby, 1954). Processes in the hydrological cycle can be thoroughly understood by studying tritium concentrations in precipitation, vegetation, surface water and groundwater. Its 12.32 year half-life (Lucas and Unterweger, 2000) enables age dating parcels of water in the environment and assessing flow velocities and storage volumes. Its application as a hydrological tracer started shortly after the release of anthropogenic tritium into the atmosphere during above-ground nuclear testing (Kaufman and Libby, 1954; Nir, 1964; Roether, 1967). The application of tritium as a hydrological tracer relies on accurate estimates of initial concentrations in precipitation (Michel et al., 2015).

Current levels of tritium in the environment are a combination of natural cosmogenic production in the upper atmosphere, depending on solar activity and geomagnetic field intensity, and anthropogenic sources of tritium from nuclear industry at the Earth's surface. Many studies have addressed the effects of anthropogenic sources on elevated tritium levels in terrestrial and aquatic environments (Jean-Baptiste et al., 2007; Thompson et al., 2015), vegetation and agricultural products (Korolevych et al., 2014; Le Goff et al., 2016; Vichot et al., 2008), surface water and aquatic organisms (Ciffroy et al., 2006; Fiévet et al., 2013; Janovics et al., 2014; Péron et al., 2016) and humans (Yoon et al., 2013). These studies have incorporated detailed physical atmosphere-land exchange processes or empirical exchange coefficients to understand and predict the behavior of tritium in the environment (Jeffers and Parker, 2014; Lamego Simões Filho et al., 2013; Maro et al., 2017). Elevated tritium levels in precipitation caused by anthropogenic releases are expected to be limited to the near vicinity of the facility (Atanassov and Galeriu, 2011; Köllo et al., 2011), although elevated tritium in precipitation after the Fukushima incident was observed at distances up to 220 km for five weeks (Matsumoto et al., 2013).

Tritium in precipitation is monitored by a volunteer network of stations around the globe and reported to the online data base of the Global Network of Isotopes in Precipitation (GNIP), maintained by the IAEA (International Atomic Energy Agency, 2016). Studies of the spatial distribution of tritium in precipitation find poleward increases in tritium related to the latitudinal dependence of cosmogenic production rates caused by geomagnetic modulation, with a stronger shielding effect at the geomagnetic equator (Masarik and Beer, 2009; Weiss and

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Roether, 1980) and inland increases related to recycling of precipitation and less dilution by oceanic water vapor (Tadros et al., 2014). Increased mixing between tropospheric and stratospheric air masses in spring (the spring leak of the tropopause) results in seasonal variation of tritium in precipitation. Locations that receive limited annual precipitation from distinct moisture sources exhibit variation in tritium from event to event and are expected to have inter-annual variability due to short-term climate variability. In addition, varying proportions of convective and stratiform precipitation result in variations in tritium in precipitation, correlated to shifts in stable isotope (^{2}H, ^{18}O) signatures (Aggarwal et al., 2016).

Global statistical models have aggregated GNIP data to predict present and reconstruct historical concentrations of tritium in annual precipitation (Doney et al., 1992; Newman et al., 2013; Weiss and Roether, 1980; Wu, 2013; Zhang et al., 2011), whereby seasonal fluctuations are approximated by a sinusoid function. These models perform well in study areas with high precipitation rates and nearby active GNIP stations but prediction errors increase with distance to reliable records and in drier climates with distinct sources of precipitation. A number of studies investigated the variability of tritium in precipitation at regional (Eastoe et al., 2011; Harms et al., 2016) or continental (Michel, 1989; Tadros et al., 2014) scales, finding general trends with latitude and distance to coast. Incidental anthropogenic releases of tritium have been detected in precipitation samples in urban areas in California and Arizona (US) (Eastoe and Osborn, 2017).

An atmospheric general circulation model incorporating the transport of tritium was demonstrated to reproduce global patterns of natural cosmogenic tritium in precipitation (Cauquoin et al., 2015) as well as the general shape of the temporal evolution of tritium in precipitation after above-ground nuclear testing (Cauquoin et al., 2016). Differences between modeled and observed tritium levels yield valuable insight into model performance and the residence time of tritium in the stratosphere in the model, but limit its capability as a predictive tool.

Global atmospheric circulation patterns have also been analyzed to track precipitation to distinct source areas to understand the variability of stable isotope (^{2}H, ^{18}O) signatures in precipitation for improving hydrological and paleoclimate interpretation. Air mass trajectories can easily be analyzed using NOAA's online available HYSPLIT Atmospheric Transport and Dispersion Modeling System (Stein et al., 2015). Distinct source areas explain stable isotope variation in precipitation in the Eastern US (Sjostrom and Welker, 2009), India (Breitenbach et al., 2010), the Tibetan Plateau (Bershaw et al., 2012; Yu et al., 2015), Australia (Crawford et al., 2013; Guan et al., 2013), Saudi Arabia (Michelsen et al., 2015) and China (Wu et al., 2015). Air mass trajectories have been analyzed the interpret peak tritium levels in Australian precipitation as a result of nearby nuclear tests in 1968 (Tadros et al., 2014). HYSPLIT has functionality for calculating pollutant deposition forward in time for a number of preconfigured species (including tritium) originating from a distinct source. In this study, we focus on the origin of tritium at ambient levels from diffuse sources, backward in time.

HYSPLIT is capable of calculating air mass trajectories but the transport of water and associated processes (evapotranspiration, precipitation, recycling) are not simulated. Previously mentioned studies have been limited to qualitative or semi-quantitative analyses of water sources. Oceanic and terrestrial moisture sources that contribute to North American monsoon (Hu and Dominguez, 2015) were studied using the modified analytical dynamic recycling model (Dominguez et al., 2006) which explicitly simulates the transport of water in the atmosphere. These studies show the potential for air mass trajectory analysis to improve the prediction and reconstruction of tritium levels in precipitation at locations that are difficult to predict using statistical models.

In this study we address the question of how much of the observed variability in tritium levels in precipitation at the Southern Sierra Critical Zone Observatory (SS-CZO, California, USA) is explained by the source areas of the water contributing to precipitation events. We hypothesize that precipitation events originating in the low-latitude Pacific Ocean (referred to in California as Pineapple Express storms) bring lower tritium levels than storms originating at higher latitudes (Pinecone Express) or inland. We investigate the improvement of predictions of tritium in precipitation for events, monthly and annual aggregates if the precipitation source area is considered. To address these questions, we compare measured concentrations of tritium in 16 aggregate precipitation samples collected at the SS-CZO over a two-year period with results of HYSPLIT particle tracking analyses of air masses, combined with a post-processing procedure considering air water content data along the trajectory to delineate water source areas.

2. Methods

2.1. Study site and sample collection

Precipitation samples were collected at the Southern Sierra Critical Zone Observatory (Anderson et al., 2008) (SS-CZO, 37.055N, 119.205W, elevation 2000 m) in the Kings River watershed in the California Sierra Nevada (US) as part of a multi-isotope tracer study into the residence times of water in the Providence Creek catchment. The SS-CZO has a sub-alpine Mediterranean climate with an annual precipitation (mean ± standard deviation) of 1306 ± 539 mm (Safeeq and Hunsaker, 2016), over 80% of which falls during fall and winter. The elevation of the site is at the rain/snow transition, receiving approximately equal proportions of precipitations as rain or snow, strongly variable between years. Water years (defined as the period between October 1st of the previous year and September 30th) 2012 to 2015 resulted in the severe drought in California (He et al., 2017).

Precipitation samples were collected in water year 2015 (1 Oct 2014–30 Sep 2015), the last year of a 4-year drought receiving only 558 mm precipitation, and water year 2016 (1 Oct 2015–30 Sep 2016) which received near average precipitation (1273 mm). The variation in precipitation amounts result in a strong vulnerability for California water supply, which relies for 60% on runoff from the Sierra Nevada. A few intense precipitation events (atmospheric rivers) strongly determine the precipitation total for a year.

Sixteen samples (Table 1) were collected in large 60 L plastic bins, left out in the forest for several weeks to collect enough precipitation for analyzing multiple cosmogenic isotopes (sulfur-35, sodium-22, tritium). A 500 mL subsample was analyzed for tritium by helium-3 ingrowth and noble gas mass spectrometry (Surano et al., 1992).

2.2. HYSPLIT

The National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory's (ARL) Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) (Draxler and Hess, 1998; Stein et al., 2015) is capable of computing air parcel trajectories through atmospheric circulation patterns described in publicly available meteorological data files. The air parcel is represented by an infinitely small particle. The movement of the particle is calculated by integrating the horizontal and vertical atmospheric flow velocities in time, either forward or backward. In this study, we used the global reanalysis data set from the National Centers for Environmental Prediction and National Center for Atmospheric Research (NCEP/NCAR) (Kalnay et al., 1996). The NCEP/NCAR reanalysis data set comprises of global wind speed, air pressure, temperature and humidity data at 2.5° horizontal resolution, 17 pressure levels ("vertical layers": 1000 - 10 mbar) and 6 h time steps for every month since 1948. The reanalysis data set incorporates observed meteorology with global circulation model results and provides the estimated precipitation rate at 6 h intervals.

To study the origin of precipitation at the SS-CZO, particles were tracked backward through the global atmospheric circulation for 10 days. Particles were released at the SS-CZO location on every hour with