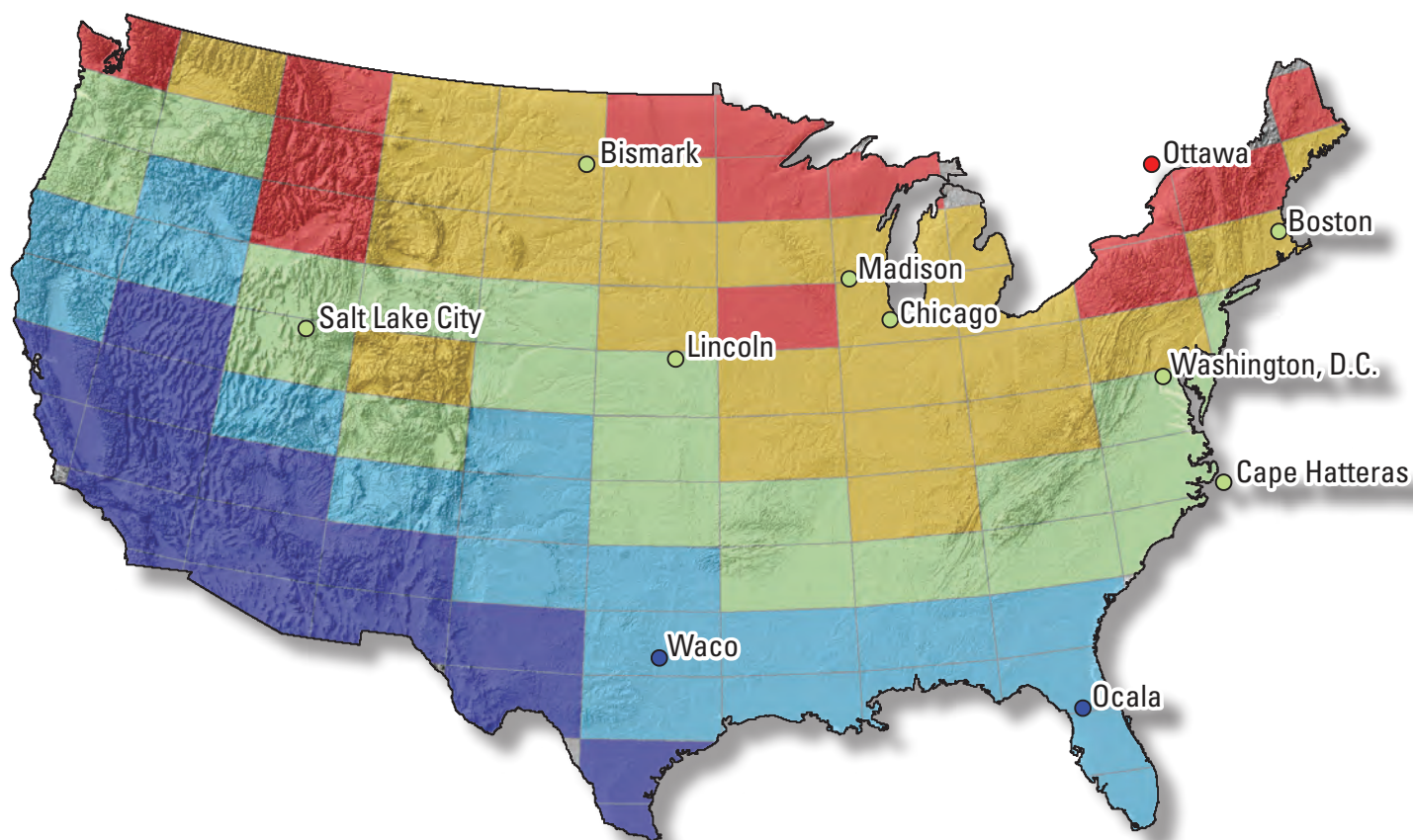


**National Water-Quality Program**

# **Tritium Deposition in Precipitation in the United States, 1953–2012**



Scientific Investigation Report 2018–5086

**Cover image:** Map showing estimated tritium deposition for the continental United States from 1953 to 1983.

# **Tritium Deposition in Precipitation in the United States, 1953–2012**

By Robert L. Michel, Bryant C. Jurgens, and Megan B. Young

National Water-Quality Program

Scientific Investigations Report 2018–5086

**U.S. Department of the Interior**  
**U.S. Geological Survey**

**U.S. Department of the Interior**

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**U.S. Geological Survey**

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## Conversion Factors

U.S. customary units to International System of Units

Multiply	By	To obtain
Length		
foot (ft)	0.3048	meter (m)
Mass		
pound, avoirdupois (lb)	0.4536	kilogram (kg)
Radioactivity		
picocurie per liter (pCi/L))	0.037	becquerel per liter (Bq/L)
Tritium in tritium units (TU)	3.19	Tritium in picocuries per liter

## Datum

Horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).

## Supplemental Information

Activities for radioactive constituents in water are given in picocuries per liter (pCi/L).

## Abbreviations

Big Meadows station	Big Meadows, Virginia, station
IAEA	International Atomic Energy Agency
Lincoln station	Lincoln, Nebraska, station
Ocala station	Ocala, Florida, station
Ottawa	Ottawa, Canada, station
Vienna station	Vienna, Austria, station
USGS	U.S. Geological Survey
Washington station	Washington, D.C., station

## Acknowledgments

This effort is the result of decades of work by many laboratories throughout the world. Particular mention has to be made of the foresight of H.E. Suess (University of California), F. Pearson (U.S. Geological Survey), H.G. Ostlund (University of Miami), R.M. Brown (Chalk River Nuclear Laboratory), and the International Atomic Energy Agency in recognizing the long-term importance of tritium concentrations in precipitation and setting up the early monitoring networks to provide this information. The authors also thank the large number of individuals who collected and carried out the technical analyses of the samples during the past 50 years. Among the agencies contributing to the support of this work during this period has been the U.S. Geological Survey, National Science Foundation, National Oceanic and Atmospheric Administration, Atomic Energy Commission, International Atomic Energy Agency, and World Meteorological Organization.

# Tritium Deposition in Precipitation in the United States, 1953–2012

By Robert L. Michel, Bryant C. Jurgens, and Megan B. Young

## Abstract

Tritium is a radioactive isotope of hydrogen (half-life is equal to 12.32 years). Since it is part of the water molecule, tritium can be used to track and date groundwater and surface water when the history of tritium in precipitation and recharge is known. To facilitate that effort, tritium concentrations in precipitation were reconstructed from measurements and correlations for 10 precipitation stations in the continental United States. Using these data, and other data sets and correlations, estimates of tritium concentrations in precipitation from 1953 through 2012 and total tritium deposition from 1953 through 1983 were derived for 2 degrees latitude by 5 degrees longitude quadrangles of the continental United States. For August 1953 through December 1987 correlations were derived from the data of the Ottawa, Canada, station; for estimates after 1987, the Vienna, Austria, station was used. For quadrangles where no precipitation station records are available from measurements or correlations, concentrations were interpolated. The International Atomic Energy Agency has proposed that a correlation with the Vienna, Austria, station, which has been in operation since 1960, be used instead of the Ottawa, Canada, station for the period after 1987. Linear correlations calculated for all stations with the Vienna data are reported. Correlations also have been calculated for each latitude-longitude quadrangle using the estimated concentrations and the measured Vienna data for 1960–87. Because the Vienna, Austria, station is on a different continent, and not subject to the same seasonal weather patterns that affect North American stations, a correlation was obtained for each month individually. This reflects the fact that the North American stations were strongly affected by the input of low-tritium moisture from the south during the summer. The correlated station values were then used with measured Vienna data to obtain estimates of tritium concentrations in precipitation for the period 1988–2012 at each precipitation station and latitude-longitude quadrangles. This approach has two major advantages: (1) it blends the Ottawa correlation with the Vienna correlation to cover the period 1953 to present, and (2) it yields correlation coefficients for each quadrangle that can be used in future years when more data become available from

the Vienna, Austria, station. The data, the estimated tritium concentrations derived from the correlations, and the correlation coefficients are provided as spreadsheets and associated comma delimited files in a data release that accompanies this report.

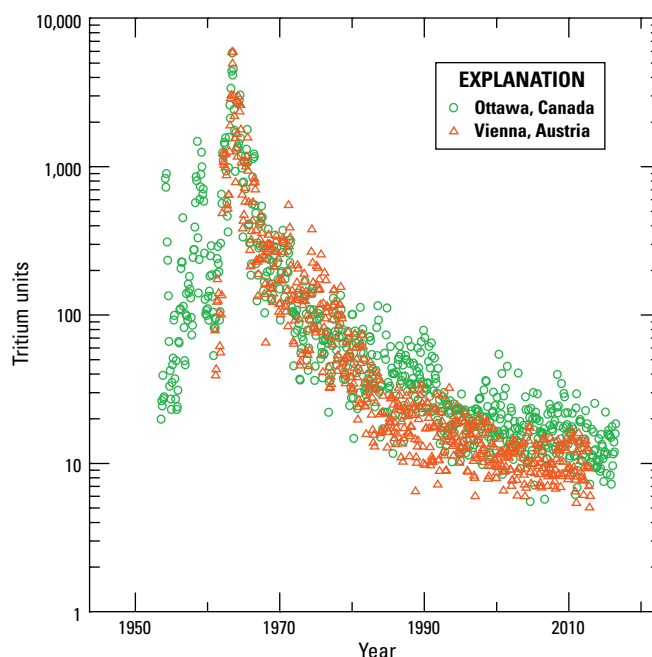
## Introduction

Tritium has been one of the most useful tracers in oceanography and hydrology for more than 50 years (Suess, 1969). Being a radioactive isotope of hydrogen (half-life is equal to 12.32 years), it is ideal for tracing the movement of water through the environment at decadal timescales. It participates in the same reactions as water with only slight variations caused by fractionation (Bigeleisen, 1962). These variations are small and can be ignored in almost all hydrologic systems. The radioactive decay constant is known, and concentrations are unaffected by mineral exchange, gas loss, oxidation-reduction reactions, or processes that frequently affect other tracers. In situ tritium production is rare and can generally be ignored (Macklin and Banta, 1954; Andrews and Kay, 1982). Thus, the concentration of tritium in any system is simply a function of the input, mixing with other waters, and the fraction lost through decay since it entered the system. Tritium has been used to study timescales for surface-water processes and interactions between surface water and groundwater. Tritium balances can be used as a proxy for water balances in many of these studies. One difficulty in using tritium as a tracer in hydrology is the variation in its concentration in precipitation, temporally and geographically. Accurate interpretation of tritium concentrations requires accounting for these variations in local input functions. The purpose of this report is to provide a methodological framework for estimating spatially and temporally varying tritium-input functions for the contiguous United States. Estimated tritium-input functions for precipitation stations and 2 degrees latitude by 5 degrees longitude quadrangles for the United States are provided in tables in the accompanying data release (Jurgens, 2018).

## Background

The radioactive nature of tritium was known in the 1930s, but measurement techniques were inadequate to detect its presence in the environment at that time (Alvarez and Cornog, 1939). Using electrolysis of water to enrich the tritium concentration, the discovery of natural tritium in the environment roughly coincided with the beginning of the nuclear age (Grosse and others, 1951; Giletti and others, 1958). Before nuclear testing, tritium found in the environment resulted from accretion from the sun or cosmic-ray spallation, the latter of which was thought to be the dominant process (Wilson and Fergusson, 1960; Nir and others, 1966; Teegarden, 1967). The cosmic-ray production probably kept a steady-state burden of tritium of 3–4 kilograms (kg) on the Earth's surface (Lal and Peters, 1967). A few measurements of tritium were taken before the beginning of major atmospheric testing, the natural concentrations of tritium in precipitation at that time are not well known. From the few measurements available, it is assumed that tritium concentrations in precipitation were about 1–2 tritium units (TU) near the coast and less than 10 TU in the mid-continental areas (Kaufman and Libby, 1954; von Buttlar and Libby, 1955; Thatcher, 1962).

Tritium produced by atmospheric testing of nuclear weapons resulted in a large increase in the tritium burden of the Earth (Carter and Moghissi, 1977). Most of this tritium was quickly converted to water and entered the hydrosphere. Production by fusion testing was much greater than what was produced by fission testing (Miskel, 1973), and several hundred kg of tritium were produced by the time of the Nuclear Test Ban Treaty in 1963 (Michel, 1976). The earliest time series of measurements of tritium concentrations in precipitation began in Ottawa, Canada, in mid-1953 (Brown and Grummitt, 1956; Brown, 1961). Data from this station are available with almost no interruption from August 1953 to 2016 (fig. 1). Beginning in the late 1950s and early 1960s major atmospheric nuclear testing increased, and many other monthly stations were established, either by individual laboratories, or through a network established by the International Atomic Energy Agency (IAEA) and the World Meteorological Organization to monitor isotopes in precipitation. Measurements from these stations demonstrated a large increase in tritium concentrations in precipitation following major nuclear test series, with highest concentrations following the large tests of 1962 (Eriksson, 1965). Concentrations in precipitation reached 10,000 TU in inland areas of Canada during this period and were routinely above 1,000 TU for large areas of the North American continent (International Atomic Energy Agency, 2017). Tritium concentrations in fresh surface waters and ocean waters also increased dramatically following these tests, reaching concentrations of more than 40 TU in the North Pacific Ocean and more than 1,000 TU in some continental rivers (Eriksson, 1963; Stewart, 1966; Dockins and others, 1967; Stewart and Farnsworth, 1968; Ostlund and Fine, 1979).



**Figure 1.** Tritium in precipitation at Ottawa, Canada, 1953–2016, and Vienna, Austria, 1961–2012 (International Atomic Energy Agency, 2017).

## Prior Tritium Source Functions

Little or no data are available on tritium concentrations in precipitation for many areas and periods during the nuclear era for the continental United States. Data from the period before 1960 are particularly limited. Because the bomb-tritium pulse is a transient event, it is now not possible to take measurements to cover these deficiencies, as it might be with steady-state tracers or stable isotopes. Estimates of historical tritium concentrations in precipitation have been obtained by measuring wine samples or other archived waters (Roether, 1967; Allison and Hughes, 1977; Michel and Kraemer, 1995). There are probably few of these types of water available to overcome the gaps in space and time. There are locations where historical tritium concentrations can be derived from ice cores (Miller and others, 1965; Merlivat and others, 1977; Naftz and others, 1996). However, for most of the areas in the United States, this approach is not possible. One option to derive estimates of tritium concentrations in precipitation where data are not available is through correlations obtained with stations that were operating for a long time and the use of interpolation for regions which were not well covered.

For the Northern Hemisphere, the Ottawa, Canada, station (Ottawa station) can be used for correlations because it was in operation almost continuously starting in August 1953 and the data were of high quality (International Atomic Energy Agency, 1981). Correlations among the Northern Hemisphere stations and the Ottawa station can be made at monthly and



yearly time steps using linear regression of measured tritium concentrations and log, base-10, concentrations:

$$C_{i,j} = a_{i,j}C_{ott,j} + b_{i,j} \quad (1a)$$

$$\text{Log}10(C_{i,j}) = a_{i,j}\text{Log}10(C_{ott,j}) + b_{i,j} \quad (1b)$$

where

- $C_{i,j}$  is the concentration at the station of interest,  $i$ , of month  $j$ ;
- $C_{ott,j}$  is the concentration at the Ottawa station of month  $j$ ;
- $a_{i,j}$  is the slope of the correlation regression at the station of interest,  $i$ , of month  $j$ ; and
- $b_{i,j}$  is the intercept of the correlation regression at the station of interest,  $i$ , of month  $j$ .

Correlations are normally done using a log transformation because tritium in the atmosphere changed by four orders of magnitude between 1953 and 1983. For the Southern Hemisphere, similar correlations can be done with the Pretoria, South Africa, station, which started collection of tritium data in precipitation in the mid-1950s. A later publication changed the stations used to Vienna, Austria, for the Northern Hemisphere and Kaitoke, New Zealand, for the Southern Hemisphere (International Atomic Energy Agency, 1992). Kaitoke was more suitable because Pretoria has seasonal lapses in rainfall and was discontinued for long periods of time. Kaitoke has operated continuously since 1960. The Ottawa station did not have large gaps in tritium measurements, but those measurements could have been influenced by a local nuclear power station that made the station unsuitable for correlations starting in the late 1980s. The Vienna, Austria, station (Vienna station), which has tritium in precipitation from 1961 to 2012, was substituted (fig. 1).

Other efforts have been made to fill in the tritium data gaps in precipitation. Some authors have developed local-source functions for limited areas (Rabinowitz and others, 1977; Simpkins, 1995). For the United States, Michel (1989) used the Ottawa correlation (International Atomic Energy Agency, 1981) to estimate tritium deposition in 2- by 5-degree quadrangles on a yearly basis for the United States from 1953 to 1983. The rationale for the correlations and using the 2- by 5-degree grid is discussed in Michel (1989). There are long-term monitoring programs in some countries that include precipitation and surface-water concentrations over time (Rank and others, 1998; Schurch and others, 2003). Other efforts have provided global input functions for tritium, generally focusing on the world oceans (Weiss and Roether, 1980; Doney and others, 1992). In general, input functions for the oceans are smoother than functions over land surfaces. The tritium/helium-3 pair also has been used to overcome the source-function problem in many cases. It is particularly useful in groundwater studies (Solomon and others, 1992; Carmi and Gat, 1994; Larsen and others, 2003), but also can be of value in studies of subsurface waters in lakes (Peeters

and others, 2000; Hofer and others, 2002). However, issues related to gas retention can raise concerns with the use of this type of data, so a source function for tritium is still valuable (Schlosser and others, 1988).

Estimates of tritium concentrations in precipitation that take into account the temporal, geographical, and seasonal variations that influence the concentrations in the continental United States are needed in a wide range of hydrologic studies (Michel, 2005). Most modeling approaches for groundwater and surface water issues depend on an understanding of the tritium-input function (Przewlocki and Yurtsever, 1974; Zuber, 1986; Michel, 2004). Because recharge generally occurs on a seasonal basis, it is sometimes necessary to know what the variations in tritium concentrations are on a monthly basis (Foster and Smith-Carrington, 1980). Correlations with existing databases can be used to overcome these deficiencies and establish a basis to estimate tritium concentrations in future years when more data become available from the IAEA.

## Methods

Tritium data used in this report are primarily from five sources: (1) U.S. Geological Survey (USGS) Tritium Laboratory, that was initially in Washington, D.C.; relocated to Reston, Va., in 1972; and then relocated to Menlo Park, Calif., in 1994; (2) La Jolla Tritium Laboratory, Scripps Institution of Oceanography, University of California, San Diego, Calif.; (3) Miami Tritium Laboratory, Rosenstiel School of Marine and Atmospheric Sciences, University of Miami, Miami, Fla.; (4) Chalk River Nuclear Laboratory, Ontario, Canada; and (5) International Atomic Energy Agency Tritium Laboratory, Vienna, Austria. All of these laboratories perform high-quality tritium analyses and have participated in the inter-comparison studies conducted by the IAEA over the past several decades. The results of these analyses of tritium in precipitation done on a monthly basis, along with analytical uncertainties, are available from the IAEA Global Network of Isotopes in Precipitation (GNIP) website (International Atomic Energy Agency, 2017).

Two approaches were used to construct source functions for the continental United States. One approach used the long-term collection stations in the continental United States, and used correlations to fill in months and years when no data were collected. The stations chosen for this approach are listed in Jurgens (2018, table 1) and are USGS stations where tritium in precipitation was analyzed and reported to the IAEA (International Atomic Energy Agency, 2017). Some USGS stations that reported tritium in precipitation values to the IAEA were not used; data from Albuquerque, N. Mex., and St. Louis, Mo., were not used because samples were composites of 3 months of precipitation; data from Portland, Oreg., were not used because samples were composites of non-monthly periods; and data were omitted from Menlo Park, Calif., which suffered from occasional tritium contamination from a local source.

Correlations of the form  $y = ax + b$  were made using the Ottawa station for the period before 1987, and the Vienna station for the period 1987–2012 as recommended by the International Atomic Energy Agency (1992). For the period before 1987, the Ottawa station was used for correlations with United States stations because of the close proximity to the United States and the availability of monthly concentrations throughout the 1950s (International Atomic Energy Agency, 1992). Tritium concentrations were estimated for every month except February 1954 because no measurements were available for the Ottawa station. Concentrations were changing rapidly during this period and it was not possible to make an estimate. After 1987, the Vienna station was used and there were data gaps for some months between 2003 and 2012. Correlations with the Vienna station were done for each month spanning the entire tritium record at each Northern Hemisphere station to account for the differences in weather patterns in the United States and Europe. Thus, separate correlation coefficients of  $a$  and  $b$  were calculated for each month for each station when using the Vienna correlation.

A second effort was done to develop tritium source functions for locations where long-term stations are not available. In this effort, the reconstructed station data done in the first approach was spatially interpolated to a grid covering the continental United States. Estimates of tritium concentrations in the continental United States from August 1953 to December 2012 were computed for quadrangles using the same latitude-longitude pattern as in Michel (1989). The 48 contiguous states are divided into 96, 2- by 5-degree grid cells. Long-term tritium precipitation stations operated in about 25 percent of these boxes during the 1960s, which is the period when concentrations changed most rapidly. There also are a series of stations operated by the Canadian government on the United States–Canada border that were used to help estimate concentrations in the northern United States. Monthly correlations with Ottawa data were used to estimate tritium concentrations for periods when data were not available at the long-term precipitation stations. This was primarily the period prior to 1961, before most stations were established, and after about 1980 when many stations ceased.

For estimates after 1987, the Vienna station was used for correlations. For a given 2- by 5-degree quadrangle, the correlation was carried out by using a regression of the estimated concentrations from the quadrangle with data from the Vienna station for 1961–87. Similar to previous correlations (International Atomic Energy Agency, 1981, 1992; Michel, 1989), a linear regression was used with the natural log:

$$\ln(C_{i,j}) = a_{i,j} \ln(C_{vie,j}) + b_{i,j} \quad (2)$$

where

$C_{i,j}$  is the concentration at the station of interest,  $i$ , of month  $j$ ;

$C_{vie,j}$  is the concentration at the Vienna station of month  $j$ ;

$a_{i,j}$  is the slope of the correlation regression at the station of interest  $i$ , of month  $j$ ; and

$b_{i,j}$  is the intercept of the correlation regression at the station of interest,  $i$ , of month  $j$ .

The regression coefficients,  $a$  and  $b$ , were determined with Vienna data measured between 1987 and 2002 to estimate tritium concentrations for each quadrangle. These correlations were extended to estimate concentrations in precipitation for the 2002–12 period. The last data available from the IAEA website for Vienna are from December 2012, but these correlations can be extended further if more data become available.

The estimated concentrations for tritium in precipitation in tritium units (TU) have been compiled in three tables for ease of use. These tables are available as spreadsheet files (.xlsx format) and text files (.csv format) in the data release that accompanies this report (Jurgens, 2018). Reconstructed tritium concentrations in precipitation for ten monitoring stations in the United States for the period 1953–2012 are reported in Jurgens (2018, table 1). Estimated tritium values were determined from correlations with Ottawa, Canada (before 1987), or Vienna, Austria (after 1987), and are identified by italic text in the table. Estimated tritium concentrations in precipitation for ninety-six, 2- by 5-degree quadrangles covering the continental United States in monthly increments for the period 1953–2012 are reported in Jurgens (2018, table 2). The slope, intercept, and  $R^2$  values for monthly correlations between the tritium concentration in precipitation at the Vienna station and the 10 precipitation stations and 96 quadrangles in the continental United States are reported in Jurgens (2018, table 3). These correlation coefficients can be used with equation 2 to extend the estimated monthly tritium concentrations for the stations and quadrangles from 2013 to future years as new tritium concentrations from the Vienna station become available. There were a few months when data were not collected at Vienna, and an estimate was derived from concentrations the month prior to and following the missing month. All tritium concentrations in the three tables are given in TU.

Tritium deposition for the continental United States was calculated by multiplying the estimated tritium concentration with the average precipitation value (in meters [m]; PRISM Climate Group, 2018) for every month from 1953 to 1983 in each 2- by 5-degree quadrangle. The sum of these values in a quadrangle is the total tritium deposition that primarily resulted from atmospheric nuclear weapons testing that occurred from 1953 to 1980.

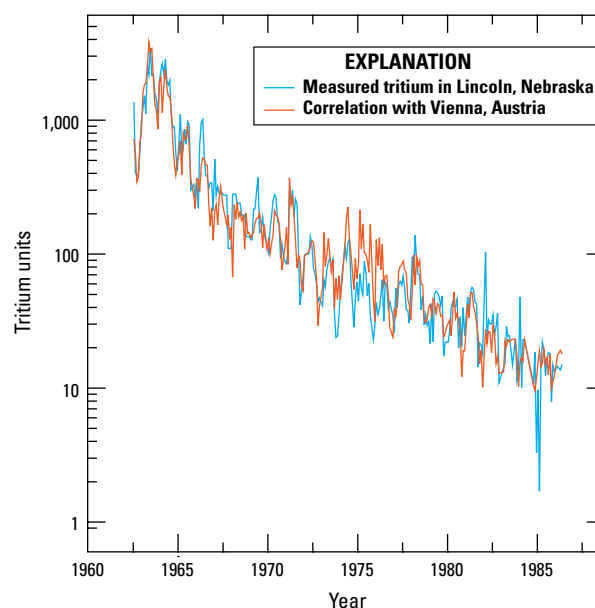
## Tritium Correlations

The estimated tritium concentrations are only meant to be used for periods and locations when no measured data are available (Jurgens, 2018, table 2). They should not be considered a substitute for periods when historical data are available or as a replacement for measurements of tritium in precipitation. Figure 2 is a plot of actual measured data (International Atomic Energy Agency, 2017) versus concentrations reconstructed from the monthly correlations with the Vienna data for the Lincoln, Nebr., station (Lincoln station; Jurgens, 2018, table 1). The reconstruction of the Lincoln station data from Vienna station data showed that estimated tritium agreed well ( $R^2=0.87$ ) with long-term and short-term tritium trends overall, but correlations also can tend to overestimate or underestimate measured values at a particular month or year.

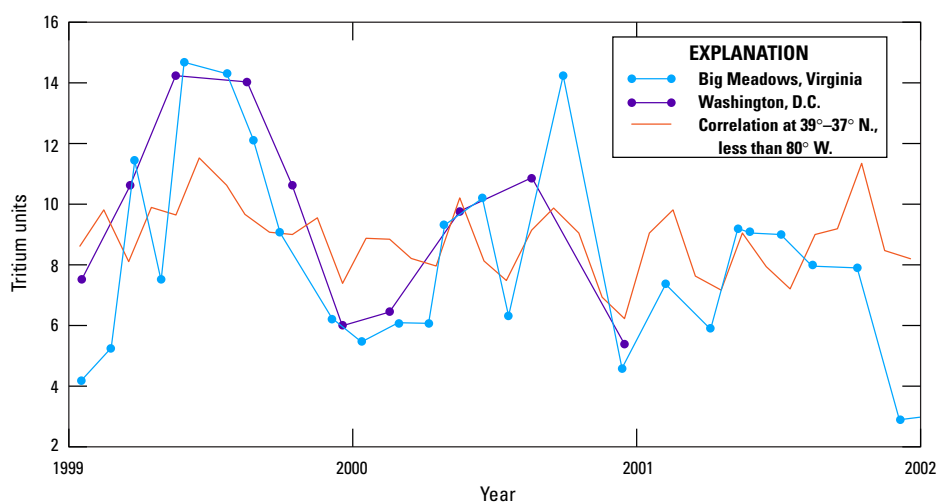
During the period of high-tritium concentrations, and particularly during periods of nuclear testing in the 1960s, tritium concentrations in precipitation were primarily controlled by the changing global burden of tritium and the location and timing of nuclear tests. Thus, tritium concentrations in precipitation were impacted in all parts of the country at the same time. This global control of concentrations has resulted in high values (more than 0.8) for the  $R^2$  of the correlation for all stations and latitude-longitude quadrangles (Jurgens, 2018, table 3). There has been no major atmospheric testing since the 1980s, and the atmospheric burden has decreased through decay and deposition with most of the tritium now in the Northern Hemisphere oceans.

The 2- by 5-degree grid is coarse, and there can be large variations in tritium concentrations in precipitation within a quadrangle. This is particularly true for coastal areas and areas where mountain ranges are crossed. Stable isotopes of water can vary over small distances in such areas, and many of the same variations are in tritium concentrations (Yurtsever and Gat, 1981).

Few stations with tritium data are available that can be compared to concentration estimates from the correlation model. In the quadrangle formed at lat  $39^{\circ}$ – $37^{\circ}$  N. and long  $<80^{\circ}$  W. to the eastern border of United States, two stations are available—Big Meadows, Va. (Big Meadows station; lat  $38^{\circ}52'$  N., long  $78^{\circ}21'$  W.), and Washington, D.C. (Washington station; Jurgens, 2018, table 1; lat  $38^{\circ}51'$  N., long  $77^{\circ}2'$  W.). The Washington station (International Atomic Energy Agency, 2017) had data from 1963 to 2001, whereas the Big Meadows station had data beginning in 1999, so only a 2-year period was available for comparison (fig. 3). The measurements at the two stations compare well to each other, with maxima in the spring of each year. The estimated tritium from the correlation model also compares well to the data from



**Figure 2.** Measured and reconstructed tritium concentrations in precipitation at Lincoln, Nebraska, 1962–86 (International Atomic Energy Agency, 2017).



**Figure 3.** Tritium concentrations in precipitation at Big Meadows, Virginia, and National Airport at Washington, D.C. (International Atomic Energy Agency, 2017), compared to computed tritium in precipitation for quadrangle lat  $39^{\circ}$ – $37^{\circ}$  N., long  $<80^{\circ}$  W. (data are from Jurgens, 2018, table 2).

the two stations, although it underestimates the differences between the highest and lowest measured concentrations. However, the average concentration appears to lie well within the variations of the measured data, so the tritium estimates give a good representation of the concentration over time for these stations in recent years.

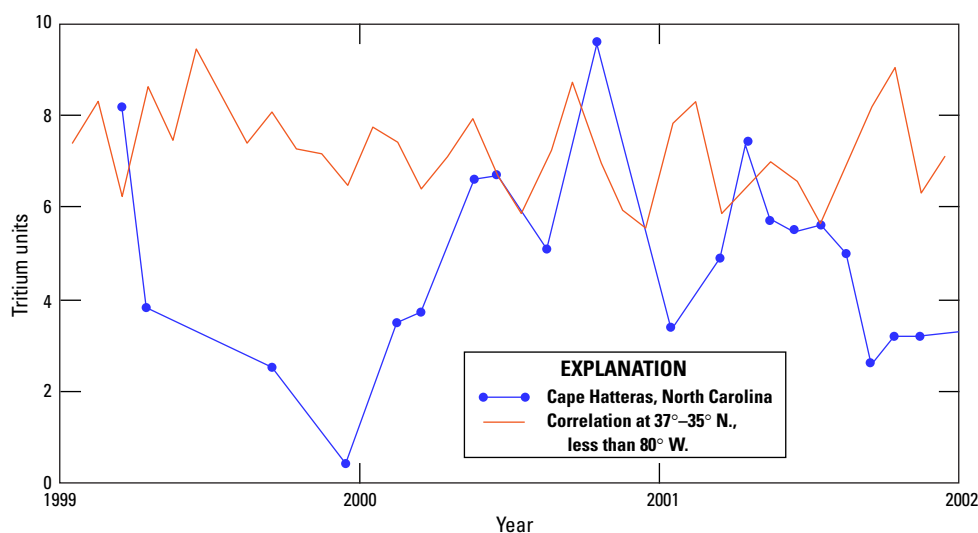
Another station available for comparison is near Cape Hatteras, N.C., where the most recent years of data are plotted versus the correlation model estimate for quadrangle lat-  $37^{\circ}$ – $35^{\circ}$  N., long  $<80^{\circ}$  W. (fig. 4). The correlation model estimate generally lies near, but above the measured data. This discrepancy is not surprising because Cape Hatteras is a coastal location and can receive precipitation that has been affected by water vapor from the ocean, which has a low-tritium concentration—see the “Tritium Deposition” section in this report about the “continental effect”. A more inland location would likely lie closer to the estimated data. Consequently, the correlation model can often reproduce seasonal and yearly tritium patterns but may not capture the full extent of variability of tritium concentrations within a quadrangle, particularly near shorelines where water vapor from the oceans can influence local tritium concentrations. In these cases, tritium values could need adjustment for local climatic and geographic factors. Comparisons of the estimated tritium source functions to measured tritium and other environmental tracers collected from local groundwater and surface water can often reveal these deviations and be used to adjust the correlation model values to the local conditions.

## Tritium Deposition

Estimated tritium deposition for the continental United States is shown in figure 5. In general, tritium concentrations reported in precipitation for mid-continental areas tend to be higher than in coastal areas for the same latitude. This

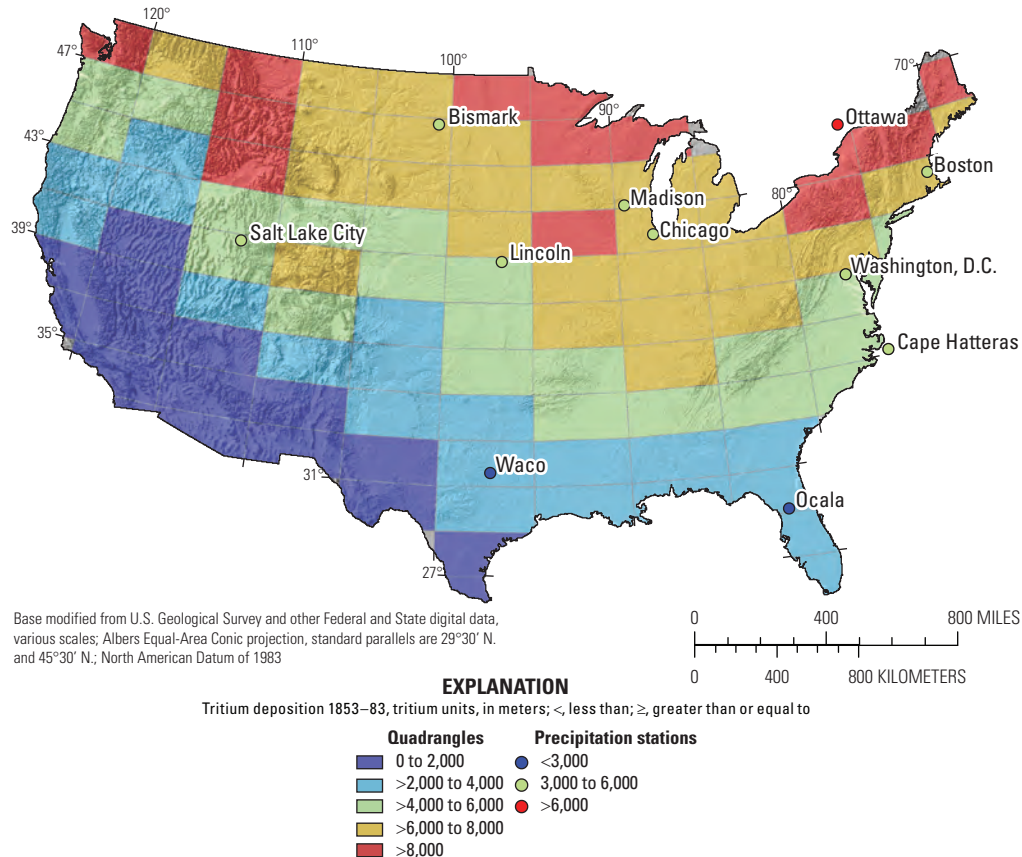
“continental effect” was seen in figure 4 and can be seen in figure 6, which is a plot of the tritium concentrations in precipitation at Ocala, Fla. (Ocala station), and Lincoln, Nebr., during the height of tritium releases to the atmosphere. The Ocala station is a low-latitude station with a strong oceanic influence. The Lincoln station is a mid-continental station, which had some of the highest tritium concentrations in North America. The low-tritium concentrations reported in coastal precipitation are the result of exchange of atmospheric water vapor with ocean surface water, which generally has a much lower tritium concentration (Schell and others, 1970). As a weather system leaves the coast and continues inland, tritium concentrations in precipitation tend to rise. This is caused by the injection of stratospheric air with high-tritium concentrations in water vapor into the troposphere and the exchange of atmospheric water vapor with continental surface-water bodies that can have higher tritium concentrations than those in oceanic waters. As weather systems traverse the continent, tritium concentrations in precipitation tend to increase. Highest concentrations are reported in the mid-continental areas at high northern latitudes (fig. 5).

There also is a strong latitudinal gradient in concentrations caused by the location of nuclear weapons tests and atmospheric circulation patterns (fig. 5). Most of the tests were in the Northern Hemisphere and the largest tests by the Union of Soviet Socialist Republics (USSR) were at high northern latitudes. These tests deposited large amounts of tritium into the troposphere and the stratosphere (Libby, 1961). The tritium in the water in the troposphere tended to be removed rapidly, as water vapor in the atmosphere has a residence time of 10–30 days. Most of this tritium is removed by molecular exchange, primarily to the ocean (Ostlund and Berry, 1970; Simpson, 1970; Suess, 1970). The tritium in the stratosphere can be mixed into the troposphere by two processes in the Northern Hemisphere. During thunderstorms, thunderheads can penetrate the tropopause, resulting in rain bringing down

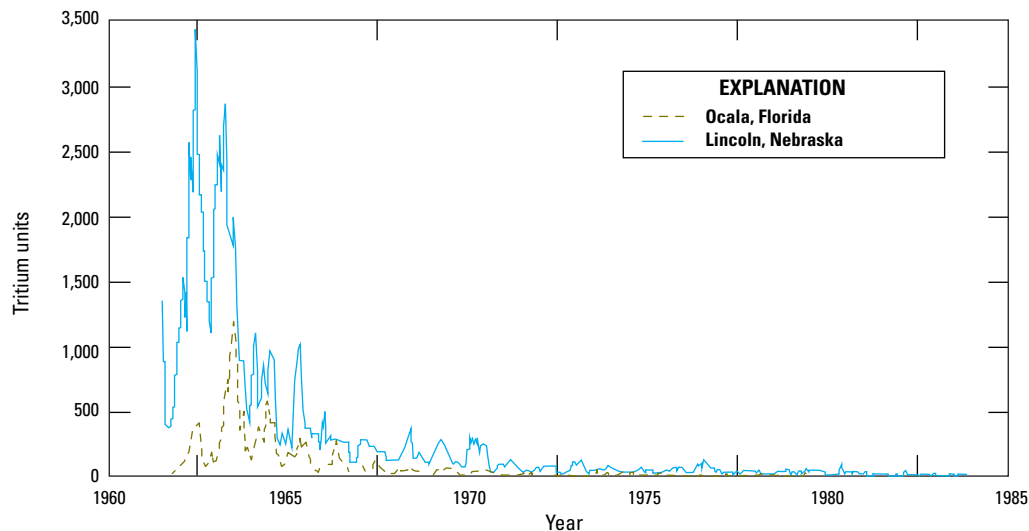


**Figure 4.** Tritium concentrations in precipitation at Cape Hatteras, North Carolina (International Atomic Energy Agency, 2017), compared to computed tritium in precipitation for quadrangle lat  $37^{\circ}$ – $35^{\circ}$  N., long  $<80^{\circ}$  W. (data are from Jurgens, 2018, table 2).





**Figure 5.** Estimated tritium deposition for the continental United States from 1953 to 1983.



**Figure 6.** Tritium concentrations in precipitation at Ocala, Florida, and Lincoln, Nebraska, 1962–84 (International Atomic Energy Agency, 2017).

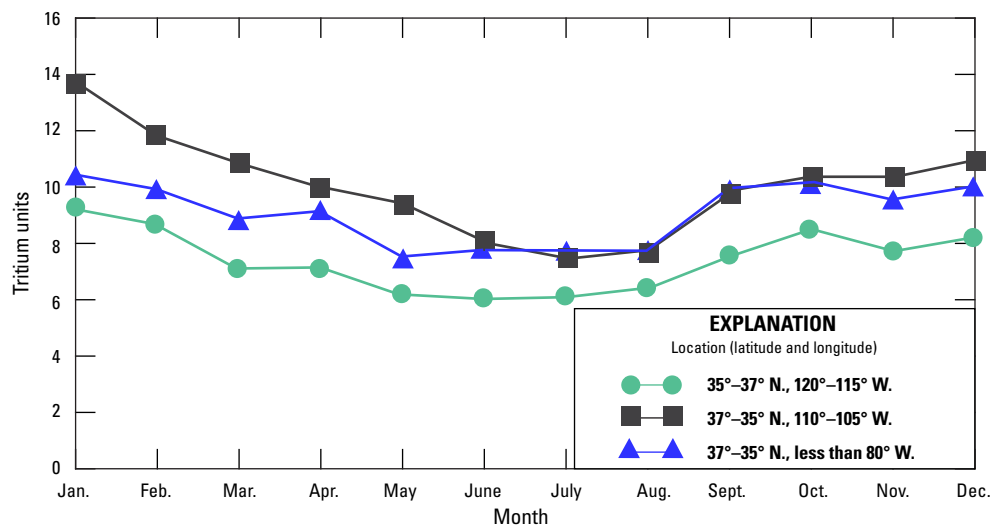
tritium or other nuclear debris from the stratosphere. The other mechanism is the “spring leak,” which occurs in the Northern Hemisphere in the spring of each year. During the spring, the tropopause breaks down between lat 30° N. and 60° N., resulting in the mixing of stratospheric air with higher tritium concentrations and air in the troposphere. As a result, tritium

concentrations in precipitation tend to increase in the spring of each year. This break up only occurs in the Northern Hemisphere because it is brought about by the heating up of the land masses at those latitudes in the spring. This results in the seasonal effect seen in tritium and cosmogenic radioisotope data in the Northern Hemisphere. In the Southern Hemisphere,

no equivalent land masses are at similar southern latitudes, so the breakup of the tropopause does not occur. Penetration of thunderheads into the stratosphere does occur in the Southern Hemisphere, and the stratosphere can come close to the land surface in the high plateaus of Antarctica, but these processes introduce much lower amounts of nuclear debris into the troposphere. Therefore, most nuclear debris introduced into the stratosphere will ultimately be deposited in the 30°–60° N. latitude band. As a result, the largest fraction of nuclear fallout occurred in the Northern Hemisphere, producing a strong north-south gradient across the continental United States (fig. 5).

Local weather patterns also can have a major impact on tritium concentrations (Kigoshi and Yoneda, 1970). Superimposed on the general circulation patterns are seasonal effects caused by changes in weather patterns. In the continental United States, winter storms tend to track from west to east, bringing moisture with high-tritium concentrations across much of the United States. In the summer, a large amount of water vapor is transported north from the Gulf of Mexico and the equatorial Atlantic Ocean. Accordingly, the tritium concentration in water vapor during the summer is much lower (fig. 7).

One of the major factors now controlling tritium concentrations in precipitation at any station is the local weather pattern (Kigoshi and Yoneda, 1970). Although local weather patterns have always had some influence on concentrations, their effect was over-ridden during the height of tritium releases into the atmosphere by global factors such as the location and timing of atmospheric tests and large scale atmospheric patterns like the spring release of stratospheric air into the troposphere. The differences caused by local phenomena have become relatively more important compared to the years during the peak of the bomb transient. This problem is partly offset by using independent monthly correlations, which adjust for general weather patterns in the United States on a long-term basis. The yearly average tritium concentrations at most stations are likely close to the estimates in this paper, and over the long term, the estimated tritium concentrations from the correlation model will probably be close to those estimates. However, the variations caused by local storm tracks can be important on a monthly basis, and errors can be greater for estimated tritium concentrations using Vienna measurements. Thus, it is still important to measure concentrations when possible, and to utilize local historical data when available.



**Figure 7.** Differences in estimated tritium concentration in precipitation using the correlation model coefficients for 37°–35° N. latitude and three longitudes (120°–115° W., 110°–105° W., <80° W.) calculated monthly, assuming a concentration of 20 tritium units in Vienna, Austria. All three longitudes show a relative decrease in tritium concentrations in summer, because of the changes in weather patterns during the year. Highest concentrations are reported at the most inland longitude (110°–105° W.).

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