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Overview of tritium records from precipitation and surface waters in Germany

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Abstract

Tritium is one of the most important environmental tracers in isotope hydrology for understanding the dynamics of groundwater and connected surface water and has been used in a wide range of applications at different scales. A key requirement for using tritium as a tracer is the knowledge of its spatial and temporal distribution in different water types. As a fundamental input, quantity long-term time series of tritium in precipitation are of particular importance. In this paper, the authors present an overview of tritium data sets of the Federal Institute of Hydrology (BfG), the Helmholtz Zentrum München (HMGU) and the German Weather Service (DWD). Since the 1970s, all three institutions have monitored the tritium concentration at 53 surface water and 37 precipitation stations on a monthly basis. The primary purpose of the data set was to provide baseline information for different water types all over Germany as an integral part of the German radiation protection monitoring system. Additionally, as geochemically inert tracer, tritium provides a unique tool to different user groups in a wide range of research questions and applications.

1 INTRODUCTION

Radiation protection regulation requires the monitoring of tritium concentration in precipitation and in major streams and coastal waters of Germany used as federal water ways. Since the 1970s, the BfG collects samples and measures the tritium concentration in rivers, the North Sea, and the Baltic Sea as well as in precipitation. The monitoring includes 17 precipitation collection stations and 53 surface water collection stations. This tritium monitoring program of the BfG has been complemented by tritium analyses in precipitation by the HMGU and the DWD. Thus 20 precipitation stations have been added in 1997 (HMGU) and 2005 (DWD). In total, long-term time series of tritium in precipitation at 37 locations and in surface water at 53 locations are available (Figure 1). They provide a temporally and spatially unique high-resolution data set, to be used for regulatory and scientific purposes, advancing our understanding in hydrology by using tritium as an environmental tracer.

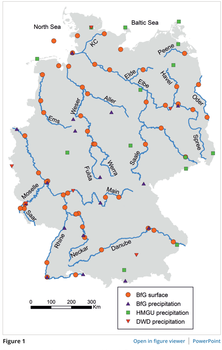

Figure 1 Open in figure viewer | PowerPoint

Precipitation and surface water sampling stations in Germany. KC, Kiel Canal

2 METHODS

2.1 Collection of precipitation samples

The precipitation is collected as a monthly pooled sample. Each collection station of the BfG consists of an approximately 0.6-m^2 large collection pan from where the water flows through a hose into a 50-L container connected to an overflow. At each BfG station, there is a heater installed for preventing the water from freezing. At the beginning of each month, a subsample of 2.5 L is drawn from the container and sent to the BfG Environmental Radioactivity Laboratory (BfG ERL) for further analysis while the remaining sample is discarded. In addition, water is collected as monthly pooled samples at stations from the DWD, stored at the stations and sent (a) to the isotope laboratory of the DWD once a month and (b) to the isotope laboratory of the Institute of Groundwater Ecology, HMGU, once per year. Some of the monthly-pooled samples were combined to get seasonal samples because too little precipitation was available for analysis (Arkona, Norderney).

At the Schleswig and Trier stations, parallel sampling and measurements for quality assurance were carried out.

2.2 Collection of surface water samples

Surface water is sampled as daily pooled samples (40 stations) and monthly pooled samples (12 stations) by automated water samplers (MAXX GmbH, Germany). Only at the sampling site Helgoland are grab samples taken once each month, due to technical reasons. For daily pooled samples, 50-ml subsamples are collected every 30 min (~2.4 L of water per day). All samples are sent to the BfG ERL, where all of the daily subsamples are pooled to a monthly mixed sample. An aliquot (300 ml) is finally used to determine the tritium concentration.

2.3 Tritium analysis

The samples were distilled and electrolytically enriched, and the tritium concentration was detected by liquid scintillation counters. Results are reported in tritium units (TU) with a 2-sigma analytical uncertainty (Table 1). As a quality control measure, all three institutions have regularly participated in tritium intercomparison studies offered by the IAEA and other official institutions.

Table 1. Additional information about analytical and technical settings

	BfG	DWD	HMGU
Detection limit [TU]	1.1	1.1	0.7
Activity of the working calibration standard [TU]	220,000	83,275	1,696
Primary standard provider	ptb[a]	ptb[a]	ptb[a]
Enrichment factor	12–15	10–15	10–15
Instrumentation	Perkin Elmer TriCarb	Perkin Elmer TriCarb	Quantulus 1,220
LSC cocktail	Ultima Gold LLT	Ultima Gold LLT	Ultima Gold LLT

[a] Physikalisch-Technische Bundesanstalt (National Metrology Institute of Germany).

3 IMPORTANCE OF THE LONG-TERM TRITIUM DATA SET

3.1 Tritium origin and decay

Tritium as an environmental tracer has been used since the early 1960s, when tritium was released to the atmosphere by thermonuclear bomb tests (Carlston, Thatcher, & Rhodehamel, 1960). Once released, the tritium concentration in precipitation fell exponentially through washout from the atmosphere and radioactive decay. Within the last two decades, the tritium precipitation values in the northern hemisphere reached a nearly constant level, with seasonal variability (Figure 2a,b). Natural tritium is formed mainly in the atmosphere due to spallation reactions of cosmic radiation with atmospheric components. The resulting tritium is oxidized to water (HTO) and enters via precipitation the surface waters and thus the hydrological cycle of the earth. With a half-life of 12.32 years (Lucas & Unterweger, 2000), tritium is an excellent environmental isotope tracer and widely used in hydrology (e.g., Michel et al., 2015); in particular, it is often used in hydrogeology for age dating and characterization of flow paths (e.g., Gleeson, Befus, Jasechko, Luijendijk, & Cardenas, 2016; Maloszewski, Stichler, Zuber, & Rank, 2002; Wan et al., 2019). Mostly, these investigations are only possible when long-term tritium concentrations in precipitation are available (Table 2).

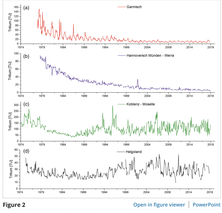

Figure 2 Open in figure viewer | PowerPoint

Long-term tritium concentrations in different water types. (a) Garmisch—precipitation; (b) Hannoversch Münden—surface water; (c) Koblenz/Moselle—surface water; (d) Helgoland—North Sea

Table 2. Overview about tritium records in different surface waters

Waters	Number of stations	Impacted by controlled tritium discharges	Longest record until 12/2018 (years)
Aller	1		40.4
Baltic Sea	1		19.6
Danube	4	x	40.5
Elbe	6	x	44.8
Elde	1		22.1
Ems	3	x	44.9
Fulda	1		40.4
Havel	2		22.1
Main	3		40.5
Moselle	3	x	44.9
Neckar	4	x	40.5
North Sea-Baltic Sea-Canal	1		39.5
North Sea	3	x	44.8

Note: Significance of x. x means that the surface waters are influenced by controlled tritium dicharges.

The gross of tritium concentration of surface waters (e.g., rivers, lakes, and oceans) reflects the tritium from precipitation. However, nuclear power plants or nuclear fuel reprocessing plants may directly impact the tritium concentrations of surface water (Figure 2c,d) due to controlled tritium discharges and deliver the opportunity for tracer studies from point sources (Mundschenk & Krause, 1991).

3.2 Applications

Tritium is considered to be geochemically inert, and it is part of the water molecule. Therefore, it is particularly suited as an environmental tracer to study hydrological processes and to determine water fluxes that occur on a timescale of less than 100 years (e.g., Koeniger, Schwientek, Uhlenbrook, Leibundgut, & Krause, 2008; Koeniger, Wittmann, Leibundgut, & Krause, 2005; Morgenstern, Stewart, & Stenger, 2010). In this context, the presented data set provides the input and baseline to advance the understanding of runoff processes in the main catchments in Germany, to investigate time or space variances of hydrological processes and to estimate timescales of transport. To improve the estimation of timescales of hydrological processes, the data set can be combined with stable oxygen and hydrogen isotope time series (Reckerth, Stichler, Schmidt, & Stumpp, 2017; Stumpp, Klaus, & Stichler, 2014), which were also measured at some of the stations used for the tritium monitoring program. Further applications of the data set include, for example, the improved understanding of regional hydrological processes, the validation of hydrological models, or the testing of new hydrological concepts. Additionally, the data contribute to the global databases of the Global Network for Isotopes in Precipitation (GNIP) and the Global Network for Isotopes in Rivers (GNIR) managed by the International Atomic Energy Agency (IAEA, https://nucleus.iaea.org/wiser/).

4 DATA

4.1 Contributors and ownership of the data

Samples are collected by the BfG, together with the Waterways and Shipping Administration (WSV) and the German Weather Service (DWD). The samples were analysed in the three laboratories, and each institution retains full ownership of their own tritium data.

4.2 Data access

The data set is available at: ftp://geftp.marbau@ftp.bafg.de.

In total, four data files are available: (a) BfG surface water, (b) BfG precipitation, (c) HMGU precipitation, and (d) DWD precipitation.

For each station, data from the beginning of the activity until December 2018 or, if a station was shut down, until the end of operation are reported. The data sets of the BfG and the DWD are updated annually; due to the closure of the isotope laboratory, HMGU will no longer provide new updates. In addition, a list with the exact location of all stations is available. As previously mentioned, some of the data can be assessed through the WISER database of the International Atomic Energy Agency (IAEA) (https://nucleus.iaea.org/wiser/).

ACKNOWLEDGEMENTS

The data set would not exist without the support of numerous people from several different institutions. Special thanks go to the staff of the WSV and DWD who continuously run and maintain the sampling stations. We are grateful to the recent and former staff of the BfG, Department G4, and the HMGU, Institute of Groundwater Ecology. In particular, we like to thank Walter Krause, Werner Speer, and Tanja Lüllwitz for delivering the vision and the continuous action at BfG, as well as Petra Seibel for the analyses at HMGU.

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