

A REGIONAL NETWORK FOR INVESTIGATING ATMOSPHERIC TRITIUM

GYÖRGY UCHRIN, KRISTÓF KOZÁK, ESZTER CSABA

Institute of Isotopes of the Hungarian Academy of Sciences, P. O. Box 77
H-1525 Budapest, Hungary

DIETER RANK and VINZENZ RAJNER

Bundesversuchs- und Forschungsanstalt Arsenal, Faradaygasse 3, A-1030, Vienna, Austria

ABSTRACT. The huge amount of ^3H released to the environment during nuclear bomb testing in the early 1960s is decreasing, but anthropogenic sources such as the nuclear fuel cycle, fusion test experiments and military and industrial use of ^3H inject the isotope into the atmosphere. A regional HT/HTO network for investigating atmospheric ^3H was established five years ago by six institutions from Austria, Croatia, Slovenia, Poland and Hungary. A unified differential HT/HTO sampler was developed, which is in use at five laboratories. Regular weekly sampling of HTO began four years ago, and has been extended to HT collection at some laboratories. We discuss this project, its technical basis and some results of regular HT and HTO sampling at different laboratories.

INTRODUCTION

The present environmental ^3H level is governed by three main sources: 1) naturally produced ^3H ; 2) ^3H from nuclear explosions and nuclear facilities; and 3) consumer products. Natural ^3H production has been insignificant in the past few decades; its inventory is estimated at 1.3×10^{18} Bq or 3.6 kg (Craig & Lal 1961). However, during atmospheric nuclear tests from October 1952 through 1962, $\sim 2.2 \times 10^{20}$ Bq or 600 kg of ^3H were injected into the atmosphere (Michel 1976).

The average global ^3H discharge to the atmosphere by nuclear power reactors (NPRs) during the years 1983–1985 was around 5300 TBq yr⁻¹, and the estimate for 1988 was 8500 TBq or 24 kg (Rozanski, Gonfiantini & Araguas-Araguas 1991). Fission-produced ^3H is retained in the fuel until it is reprocessed. The estimated ^3H release rate from nuclear fuel reprocessing plants is equal to 407 TBq GW_e⁻¹ yr⁻¹ (GW = gigawatt) and the ratio of liquid to airborne discharges varies between 1.5 and 24% (Luykx & Fraser 1986). Experimental fusion facilities use large amounts of ^3H . The discharge during normal operation of fusion power reactors is expected to be relatively low, ~ 800 TBq GW_e⁻¹ yr⁻¹ in the gaseous form ($\sim 60\%$ as HTO) and ~ 50 TBq GW_e⁻¹ yr⁻¹ as liquid effluents (Rocco & Kirchmann 1986). Some consumer products, such as self-luminous light sources, contain considerable ^3H . About 1 kg ^3H per year is used in consumer products; most is released into the environment after disposal (Wehner 1979; Krejci & Zeller 1979).

The distribution of ^3H in the geosphere and environment is affected by many factors. The ^3H content in northern hemisphere precipitation reached a maximum in 1963, then decreased exponentially until the end of 1967. The concentration of elemental ^3H in atmospheric hydrogen was observed to be 4000 TU (Faltings & Harteck 1950). The concentration reached a maximum of $\sim 2 \times 10^6$ TU in 1973–1975, then dropped to $\sim 5 \times 10^5$ TU (0.023 Bq m⁻³) (Brown 1989).

Technogenic emissions of ^3H will probably play an increasingly important role in controlling spatial and temporal variations of ^3H in the global atmosphere in the future. Table 1 summarizes the concentration ranges of environmental ^3H in TU (Brown 1989).

The regular measurement of ^3H started with its determination in precipitation in mid-1953, in Ottawa, Canada. The IAEA and WHO established jointly the global network of stations in 1960, with the aim of collecting monthly precipitation samples and determining their isotopic composi-

TABLE 1. Ranges of Environmental Tritium (1987)

Source	Reservoir	Northern hemisphere (TU)	Southern hemisphere (TU)
Precipitation	Continental	30–80	5–20
	Coastal	2–20	0.5–2
	Surface water	10–80	5–20
	Sub-surface water	3–300	0–50
Oceans	Mixed layer	0.5–2	0.1–0.5
Glaciers		0–200	0–50
Atmosphere	HTO (troposphere)	1–100	
	HT	ca. 10^6	
	CH ₃ T	ca. 2×10^4	

tions. At present, 80 IAEA/WHO network stations are in operation, and another 82 stations belonging to national organizations contribute data to the IAEA.

³H monitoring in atmospheric water vapor started at Miami, Florida, USA, in 1968; additional stations in Alaska, Hawaii, New Zealand and West Germany also provided samples for analysis (Östlund & Mason 1985). Similar projects were undertaken for shorter periods in the USA (Ehhalt 1971), East Germany (Levis, Fröhlich & Hebert 1987) and Japan (Takashima 1985). Faltings and Harteck (1950) pioneered monitoring for elemental ³H (HT) in the atmosphere. The Miami group also extended their sampling for HT (Östlund & Mason 1985). ³H is a global radiocontaminant that may play an increasing role as fusion experiments continue; thus, investigation of its environmental behavior and significance are important.

INVESTIGATING TRITIUM IN THE ATMOSPHERE

The establishment of an HT/HTO network for atmospheric studies was proposed during Hungarian bilateral scientific cooperation with ³H laboratories of Austria, Poland and Yugoslavia. A round-table discussion was organized in Budapest, Hungary, on 22–23 October 1987 to discuss the basic objectives, requirements and possibilities of a coordinated project of atmospheric ³H monitoring. Participants started regular weekly sampling of HTO in January 1988, using their own sampling and measuring methods. Results were discussed at the second meeting on HT and HTO Monitoring of the Atmosphere, Vienna, 27–28 October 1988. It was agreed that:

1. A post-monitoring system should be introduced
2. Sampling should be carried out for ³H in elemental ³H, HT and in tritiated water vapor, HTO (³H in methane form may be considered in the future)
3. Sampling should be performed weekly
4. Local and global ³H contamination in the air should be followed with a sensitivity of 10 TU for ³H in HTO form.

The main goal of the project is to establish and to operate a regional (or global) network for HT and HTO monitoring of the atmosphere. Other goals include:

1. Standardize sampling by constructing a differential HT/HTO sampling device, based on a molecular sieve absorber
2. Incorporate HT and HTO data collection from the beginning of 1990, together with ³H measurement in precipitation
3. Establish a database for HT and HTO in the atmosphere and use the database to:

- a. Monitor the local and global transport of ^3H in the air
- b. Localize ^3H sources
- c. Evaluate radiological impact of normal and accidental releases
- d. Collect data for studies aimed at converting elemental ^3H into tritiated water under natural conditions
- e. Evaluate atmospheric circulation using ^3H data.

Table 2 lists current and probable participants in the study.

TABLE 2. Atmospheric ^3H Network Participants

Institutions

Bundesversuchs- und Forschungsanstalt Arsenal Vienna, Austria
 Institute of Isotopes, HAS, Budapest, Hungary
 Institute of Physics and Nuclear Technology, University of Mining and Metallurgy, Krakow, Poland
 Ruder Boskovic Institute, Zagreb, Croatia
 Jozef Stefan Institute, Ljubljana, Slovenia

Future Participants

Boris Kidric Institute, Vinca, Serbia
 Comenius University, Department of Nuclear Physics, Bratislava, Slovakia
 ENEA - Casaccia, Rome, Italy
 Institute of Physics, Georgian Academy of Science, Tbilisi, Georgia
 Institute of Radiation Dosimetry, Prague, Czech Republic
 Institute of Geology of the Estonian Academy of Science, Tallinn, Estonia
 Institute of Physics, Lithuanian Academy of Science, Vilnius, Lithuania
 Sofia University, Faculty of Chemistry, Sofia, Bulgaria
 Water Problem Institute, Moscow, Russia

METHODS AND RESULTS

A differential HT/HTO sampler using a molecular sieve has been constructed. The principle of the sampler was similar to that of Östlund and Mason (1974). Experiments were performed to establish adsorption capacity of a molecular sieve of different type and origin, memory effect of molecular sieves, HT-to-HTO conversion rate on the water vapor adsorption column, efficiency of desorption, adsorption of elemental hydrogen, influence of different parameters affecting the adsorption and desorption, such as flow-rate of sampling, geometry of adsorption column, temperature and duration of desorption. A prototype differential sampler (Fig. 1) was designed, constructed and tested. Table 3 gives the main features of the sampler.

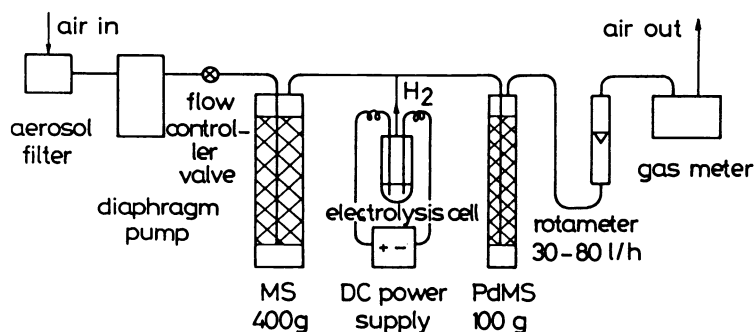


Fig. 1. Layout of the HT/HTO differential molecular sieve-based sampler

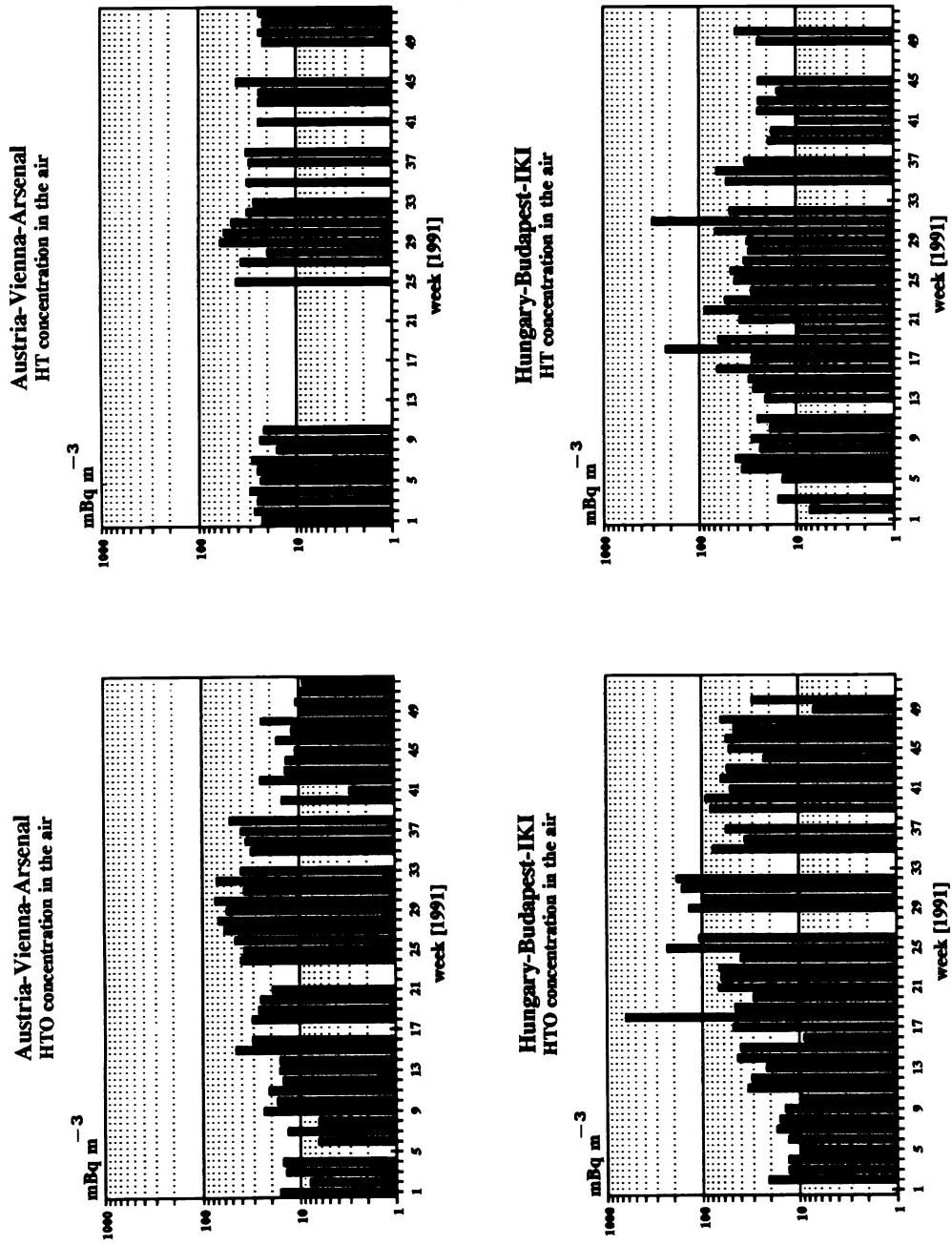


Fig. 2. HT and HTO concentrations in Vienna and Budapest. Sampling sites: Bundesversuchs- und Forschungsanstalt Arsenal Geotechnisches Institut and Main Meteorological Station, Pestfürinc, Budapest

TABLE 3. Main Features of Molecular Sieve-Based HT/HTO Differential Sampler

Sampling period	One week
Flow-rate	30–80 l/h
HTO absorber	Molecular sieve 4A
Mass	350 g
HT converter	Pd on molecular sieve
Mass	100 g
Carrier gas	H ₂ from electrolytic cell
Concentration	0.1–0.3%
Current in electrolysis	0.2 A
HTO absorption	
Max.capacity	73 g
Efficiency	99.9%
Memory	700 mg
T recovery	99%
HT conversion	
Temperature	Ambient
Efficiency	98 ± 1%
Sample recovery	
Vacuum desorption	At 50 Pa
Temperature	500°C
Duration	3 h
Technical data	
Power supply	220 V
Built-in safety system to avoid explosion	
Weight	8 kg
Size	250 × 400 × 500 m

Such differential HT/HTO samplers are now in use at the Bundesversuchs- und Forschungsanstalt Arsenal Geotechnisches Institute, Vienna, Austria, at Jozef Stefan Institute, Ljubljana, Slovenia, at ENEA-CRE, Casaccia, Italy, at Boris Kidric Institute, Vinca, Serbia and at the Institute of Isotopes, Budapest, Hungary. Participating laboratories work mostly with low-level ^3H , and use liquid scintillation counting (LSC) or low-level gas proportional techniques. At the present ^3H concentration, the sensitivities of different techniques allow to ^3H measurement without enrichment.

Figure 2 shows the results of regular ^3H sampling of the atmosphere in Vienna and Budapest; HTO concentration is *ca.* 20 mBq m⁻³ and 30–40 mBq m⁻³, respectively. The HT concentration values are similar at both locations, but slightly lower than HTO concentrations. Higher concentrations in Hungary might be due to local emissions. This proposition is supported by the time-variation spectra of concentrations. High spikes in the Budapest data contrast with rather smooth variations measured in Vienna. Data collection and compilation continue.

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