

ENVIRONMENTAL TRITIUM IN HYDROLOGY: PRESENT STATE (1992)

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ABSTRACT. Tritium measurements for groundwater dating have become an essential part of hydrological investigations. Aside from two basic assumptions, that ^3H -free groundwater contains only precipitation water from the "pre-bomb period" and is, thus, >40 yr old, and that ^3H -rich water contains much precipitation water from the 1960s, ^3H dating of water has been based primarily on models of the long-term trend in the ^3H content of hydrological systems. The gradual decrease in the concentration of bomb ^3H in the environment diminishes the importance of such calculations. The significance of local and regional environmental contamination by ^3H increases. In the future, more emphasis must be placed on reliable and frequent sampling in a closely spaced network of sampling stations. Progress in measurement and sampling techniques in hydrology has opened new avenues of investigation. Examples include studies of single hydrological events and of water movement in the unsaturated zone. The detection limits of the new generation of liquid scintillation counters are applicable to hydrological research after the decline of the anthropogenic portion of environmental ^3H .

INTRODUCTION

The hydrological application of isotope measurements is based mainly upon variations in the concentrations of ^2H , ^3H and ^{18}O , along with ^{13}C and ^{14}C in dissolved carbon, in natural waters. The most important objectives of isotopic studies are determining 1) hydrological and hydrogeological relations, 2) the residence times of groundwaters and 3) estimating possible pollutant transfer. Evaluating ^3H and ^{14}C measurements and seasonal variations of isotopic ratios provides insight into the age structure of groundwaters. The youngest water components, with the shortest transit times, play the major role in environmental studies. For questions concerning water resources management, long-term storage is of foremost interest.

ENVIRONMENTAL ^3H LEVEL AND WATER DATING

Two basic assumptions are made: 1) that ^3H -free (*i.e.*, below normal detection limits) groundwater contains only precipitation water from the "pre-bomb period" and is, thus, >40 yr old; and 2) ^3H -rich (up to ~100 TU) water contains much precipitation water from the 1960s. The use of ^3H dating of groundwaters has been based primarily on models of the long-term trend of ^3H content in hydrological systems (*e.g.*, Moser & Rauert 1980). Such models can, in principle, produce an age distribution of waters at the output of the hydrological system. The usual method is to calculate output functions from known input variations, assuming different flow models and pulse responses. The model providing the best fit of measured and calculated output data is then adapted to describe the flow system. Several models, such as piston-flow, dispersion, exponential (completely mixed reservoir) and mixed cells, have been used to describe aquifer behavior (Moser & Rauert 1980).

The input function for these models is the long-term trend of the ^3H content in precipitation (Fig. 1). Since the maximum of bomb ^3H in the early 1960s, the ^3H content of precipitation has decreased, and during the last few years, has approached natural levels. However, most hydrological systems, *e.g.*, karstic aquifers, still contain significant amounts of bomb ^3H (Fig. 2) which can be applied to dating.

As the concentration of bomb ^3H in the environment gradually decreases, such model calculations lose their significance because of the small difference in the ^3H content in waters of varying ages (Fig. 3). In the near future, this method of dating waters will be reserved for hydrological systems, for which ^3H data from previous years are available, as well as for systems that still contain large

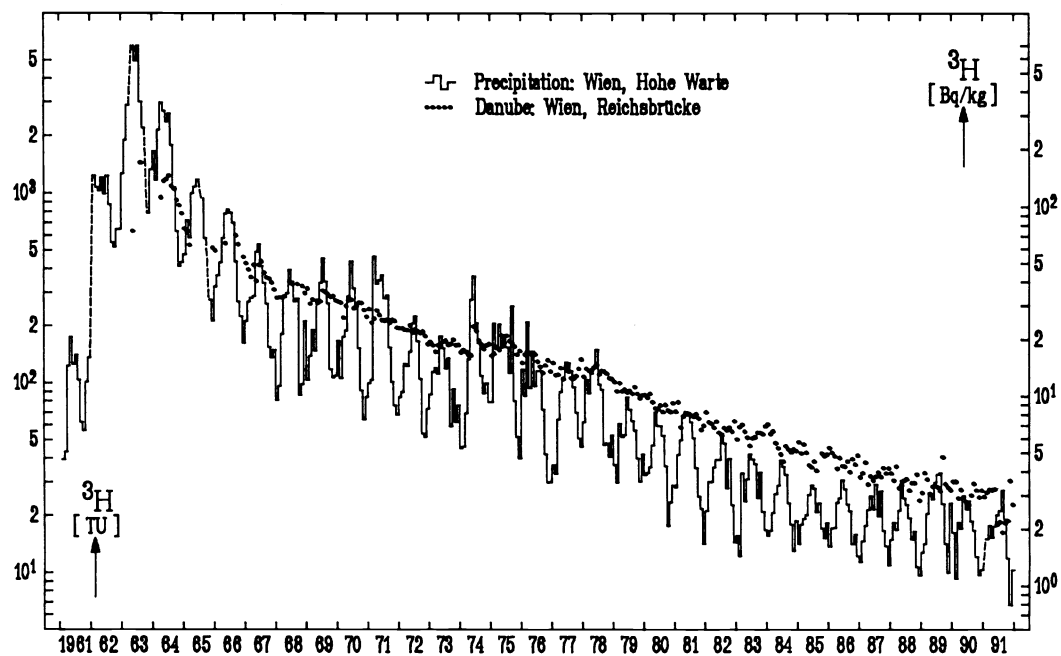


Fig. 1. Long-term trend in the ^3H concentration in precipitation (monthly mean) and in surface water (monthly grab samples) in central Europe (1 TU = 0.118 Bq kg $^{-1}$)

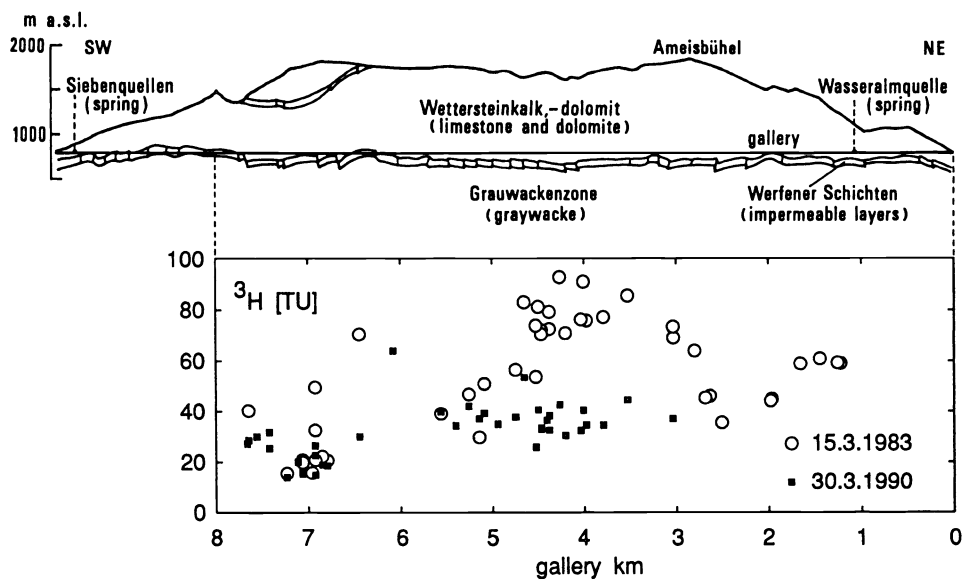


Fig. 2. Cross-section through the Schneealpe (karst massif in the eastern Alps). Variations in the ^3H content of the karstic aquifer indicate a wide-ranging age distribution of the waters in the karst massif. While the spring waters have mean residence times of 2–4 yr, some gallery waters are >50 yr old.

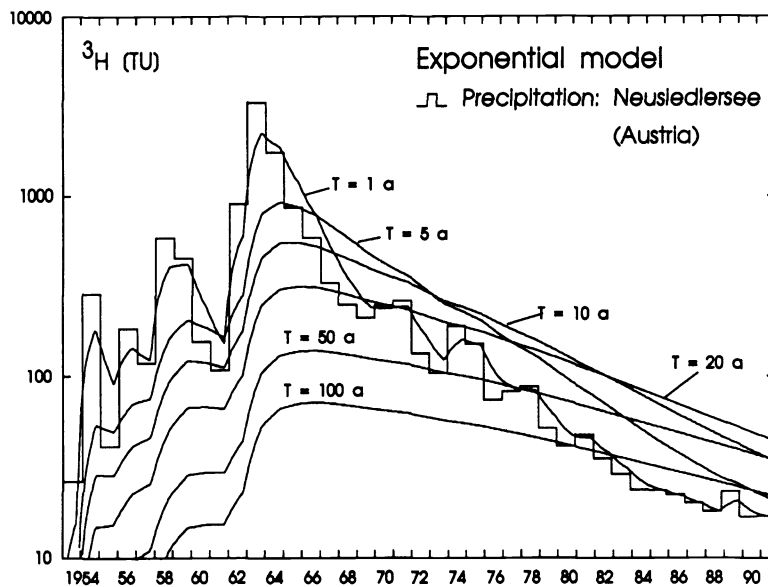


Fig. 3. Estimate of the mean residence time of groundwaters based upon exponential model calculations; expected trends in ^3H concentration at the output of a hydrological system with full mixing, dependent upon the mean residence time. The input function is the trend in the ^3H concentration in precipitation (T = mean residence time).

amounts of water from the 1960s and 1970s. However, even here, results are becoming less and less exact. For residence times up to several years, seasonal variations in isotopic ratios in precipitation can be used as an alternative input signal, as far as there is evidence of a suitable output signal in the hydrological system. Further, the portion of youngest water can be estimated through observation of the effect single events have on the isotopic ratios in hydrological systems.

The absence of ^3H in groundwater as an indicator of water age is becoming more and more important. Such waters originate from precipitation from the "pre-bomb period". As bomb ^3H decreases, the influence of local ^3H releases increases. Thus, a general description of the behavior of environmental ^3H , which could be made during the bomb ^3H period, is no longer possible. In the future, even more emphasis must be placed on a reliable, closely spaced basic network for sampling of precipitation, surface water and groundwater to provide accurate input data for age estimation.

LOCAL AND REGIONAL ^3H CONTAMINATION

Most cases of environmental ^3H contamination are harmless, from the standpoint of radiation protection. The concentrations are far below the maximum permissible value for drinking water. However, the contamination does cause an increase in environmental ^3H concentrations, which are several orders of magnitude more than normal values, and which must be considered in isotope hydrology. ^3H that has been released into the environment may also be used as a tracer. In addition to industrial emissions, such as those from nuclear power plants and the watch industry, as well as accidental releases, other possible sources include consumer products and medical wastes. Whereas the causes of higher ^3H concentrations in surface and groundwaters can be identified relatively easily, the sources of local and regional increases in the ^3H level in precipitation can be determined only rarely (Rank, Rajner & Lust 1992).

Following are several examples of environmental ^3H contamination, as detected during the operation of the Austrian basic ^3H sampling network. Until 1985, the ^3H concentration of the river March, on the Austrian-Slovakian border, correlated with the general decrease of ^3H in the environment (Fig. 4). Since 1985, the values for the river March, and differences between individual measurements, have been greater. The source of the additional ^3H is evidently the Dukovany nuclear power plant, located in the March watershed. The first nuclear plant commenced operation in February 1985, about the same time that increased levels of ^3H first appeared. The second unit followed in January 1986; the third in November 1986; the fourth in June 1987 (440 MW_e each). A rough estimate of the average excess ^3H from local releases in the river March amounts to $\sim 3 \text{ Bq kg}^{-1}$. For a year, the total ^3H activity attains a magnitude of 10^{13} Bq , or 300 Ci. In the river Danube, below the mouth of the river March, average excess ^3H equals 0.15 Bq kg^{-1} . The ^3H levels in the river March are within 1‰ of the maximum permissible concentration in drinking water.

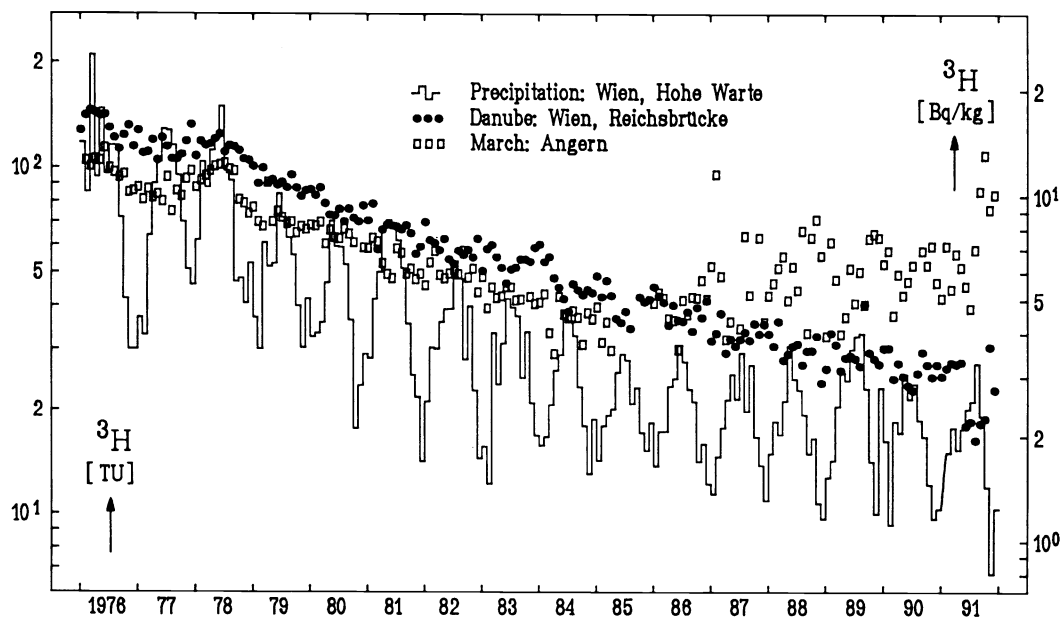


Fig. 4. Development of ^3H content in the rivers March and Danube (monthly grab samples) and in precipitation (monthly average samples) since 1976. Since the Dukovany nuclear power plant began operating in 1985/1986, the ^3H content of the river March shows larger and greatly varying values.

At the Patscherkofel mountain station (2245 m asl) in the eastern Alps near Innsbruck, the ^3H content of precipitation since the late 1970s has increased from the expected trend (Rank, Rajner & Lust 1992). On the average, the values are considerably higher, scattered, and do not follow normal seasonal variations, with a maximum in late spring and a minimum in late fall (Fig. 5). This normal seasonal pattern is represented in the values collected in Vienna, Hohe Warte station (203 m asl), which can be considered “clean”, that is, not influenced by local ^3H releases.

The irregularities in the ^3H content measured at Patscherkofel station could be caused either by local or regional releases of ^3H . A local source has not been found. Indeed, local releases would probably result in higher values for residential and industrial areas in the neighboring Inn valley.

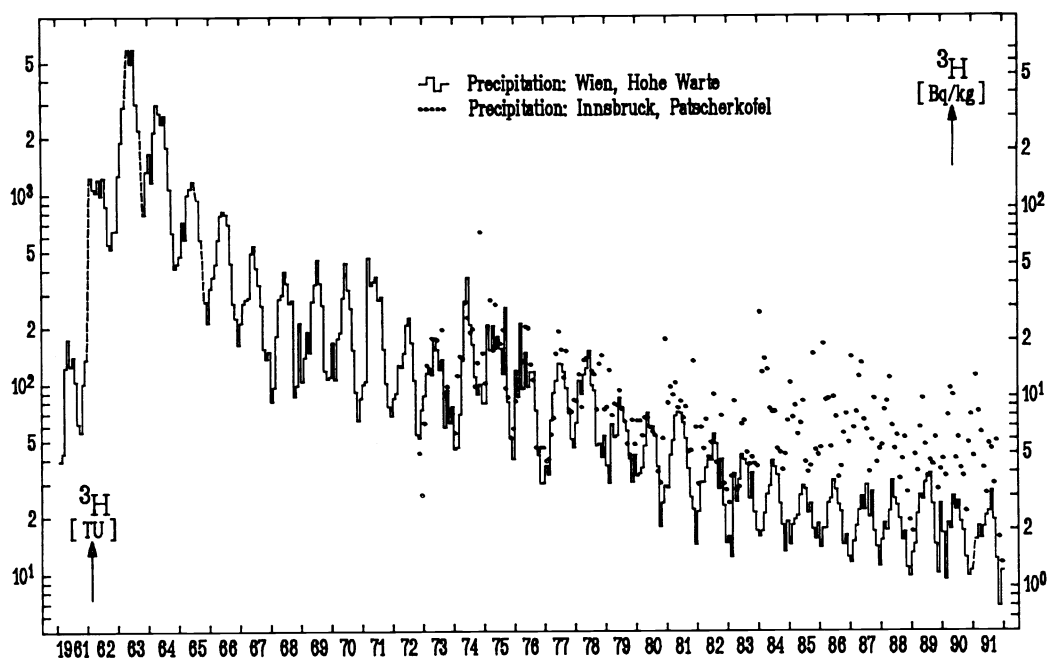


Fig. 5. Long-term ^3H concentration in monthly precipitation at Wien-Hohe-Warte and Innsbruck-Patscherkofel. Since the late 1970s, Patscherkofel values have varied greatly from the expected trend.

Surprisingly, in the valley, no similar rise in the ^3H content of precipitation could be observed. A possible regional source is ^3H release from (nuclear) industry in Western Europe, due to prevailing westerly winds. It is possible that ^3H -contaminated moisture is transported in atmospheric layers to the Alps and rains down on the summit regions. Apparently, orographic effects, as well as differing meteorological conditions, play important roles in the concentration of ^3H in precipitation. Despite extensive investigations, the source of the excess ^3H has not yet been determined.

Observations confirm increased ^3H concentrations in leachate from several domestic landfill projects. For example, ^3H concentrations up to and above 3000 TU (400 Bq liter⁻¹) were found in the leachate of the Breitenau Experimental Landfill, a research landfill for domestic waste in Lower Austria (Fig. 6; Rank *et al.* 1992). The apparent cause is a ^3H source that is brought in with the

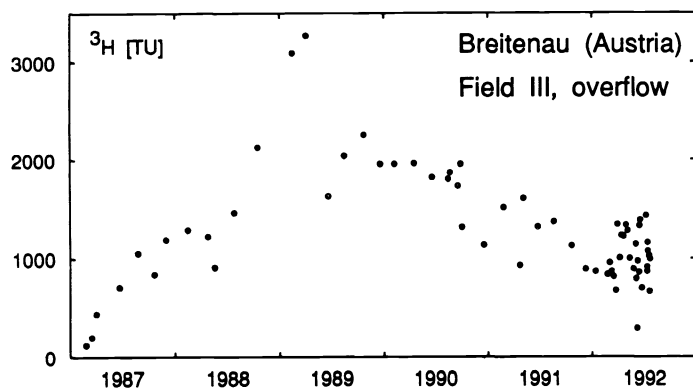


Fig. 6. Breitenau Experimental Landfill (Lower Austria): ^3H concentration in leachate from domestic waste

domestic waste, and which has not yet been identified. From the concentrations measured, one can estimate that the ^3H activity of a single wrist watch with a ^3H light source ($\sim 10^{10}$ Bq) could cause, theoretically, the contamination of all water in the landfill ($\sim 30,000$ m³). Because ^3H has been used in consumer products for many years, one must expect increases in ^3H contamination of landfill waters in the future. From the viewpoint of isotope hydrology, such ^3H contamination of leachate can be useful in the search for landfill leaks.

NEW APPLICATIONS

The investigation of single hydrological events, such as heavy rainfalls, melting snow and floods, has been made possible by automatic samplers and the availability of isotope measurement techniques. The large number of samples is due to the short sampling intervals and the fact that samples must be collected "on suspicion", because neither the point in time, the length of time, nor the isotopic significance of an event can be foreseen. In addition to purely quantitative considerations (measurement of precipitation depths, water levels and discharges), the goal of such investigations is to obtain data on the movement of individual water molecules, transit times and storage in hydrological systems.

A prerequisite for successful isotopic investigations of single events is to determine variations in the isotopic ratios of the input, which is primarily precipitation (Rank 1990). The deviation of the ^3H concentration of single precipitation events from the average seasonal trend depends on the source and development of moist air masses. The more the isotopic ratios in precipitation water differ from the mean value of the hydrological system under investigation, the more suitable is a single hydrological event, with an adequate amount of precipitation, for isotope-hydrological investigations. The results of lysimeter measurements from Lindau/Black Forest, Germany (Fig. 7) illustrate the occurrence of a single event with heavy precipitation and very low ^3H content at the end of 1990. This isotopically traced precipitation water reached the bottom of the lysimeter ten weeks later.

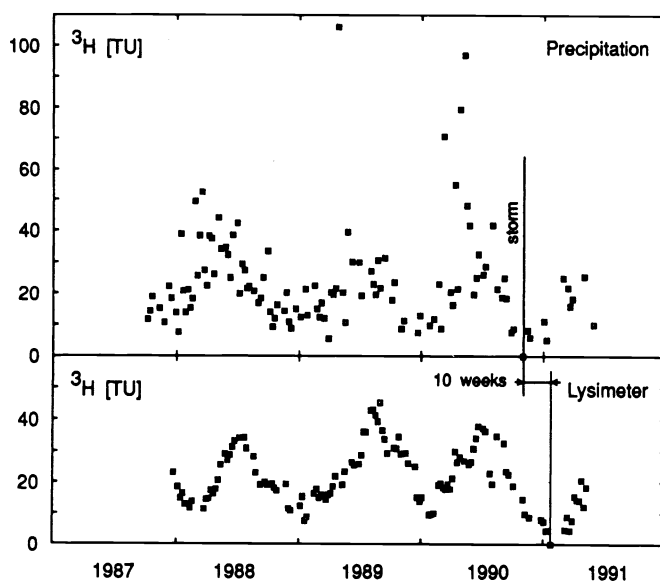


Fig. 7. Lindau/Black Forest (Germany) test area: ^3H concentration in precipitation and lysimeter seepage water (weekly samples) (Rank 1992)

Questions on groundwater contamination caused by seepage of toxic agents into the ground, or the assessment of landfill sites, demand methods through which the movement of individual water molecules in the unsaturated zone can be followed. Only isotopic methods (^2H , ^3H , ^{18}O) can fulfill this requirement. Seasonal variations and isotopically significant single events (heavy precipitation, snow melt) provide signals for short-term processes. The most important input signal for long-term seepage in fine-grained material is provided by the long-term trend of ^3H in precipitation. Because they are components of water molecules, these isotopes are ideal tracers.

In the course of the lysimeter investigations illustrated in Figure 7, seasonal variations in the ^3H content of precipitation were used to calculate the transit time of the precipitation water through the lysimeter. The seasonal variations in the ^3H content of the seepage water are easily recognizable; their maxima are, in comparison with those of precipitation, delayed by approximately ten weeks. Due to the manner of presentation (weekly values) the near-sinusoidal seasonal trend in precipitation does not emerge very clearly. Single high values are, however, connected with low amounts of precipitation and play no consequential role.

Artificial tracer experiments cannot replace these investigations (single events, unsaturated zone). Their results are valid only for the conditions in the hydrological system that prevailed during the experiment and only for that part of the system affected by the tracing.

CONCLUSIONS

The following conditions are essential for a wide-ranging application of ^3H measurements in hydrology:

1. A large number of measurements with reasonable accuracy at reasonable costs. The new generation of LS counters can fulfill this condition. An electrolytical enrichment unit is required for ^3H concentrations <5 TU. This enables an adequately sensitive measurement of the ^3H concentrations of water in the natural cycle (~ 0 – 20 TU) after the anthropogenic portion of the environmental ^3H has diminished.
2. A closely-spaced basic sampling network to enable exact recording of ^3H input data, as well as recognition of local or regional ^3H anomalies (contaminations). In addition to precipitation and surface waters, selected groundwaters and landfill waters should also be studied.
3. Combining ^3H values with other parameters, for example, stable isotopes or chemicals which make the ^3H data more meaningful. Above all, in case of an ambiguous dating result with ^3H , the correct residence time can be determined. Such decisions are becoming more important, as time progresses from the 1960s maximum concentration of ^3H in precipitation.
4. Adaptation of the entire system of water-dating methods to the decline of bomb ^3H , for example, development of routine methods for dating by ^3H input that is, on average, a long-term constant.
5. Introduction of new applications of environmental ^3H measurements in hydrology, as made possible by advanced measurement and sampling techniques, for example, investigating single hydrological events and water movement in the unsaturated zone.

Environmental ^3H measurements have become an essential part of hydrological investigations. Since bomb ^3H has diminished, isotope hydrologists are again tending to more intensive use of ^3H as an artificial tracer. However, care must be taken to keep the effect on environmental ^3H levels as small as possible (IAEA 1991).

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