Isotope hydrology at IAEA: history and activities

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Abstract The International Atomic Energy Agency (IAEA) was established in 1958 as a new United Nations special organization, with isotope hydrology as part of its research and technical cooperation programme, in conformity with the mission to encourage the Peaceful Applications of Atomic Energy. Shortly afterwards the Isotope Hydrology group at the IAEA was consolidated into the Section of Isotope Hydrology. Since that time the Section has promoted and supported the development and practical applications of nuclear and isotope techniques in hydrology and related earth sciences, including oceanology and climatology. The paper highlights specific contributions of the Agency to these developments such as the establishment and operation of the Global Network for Isotopes in Precipitation (GNIP), the provision of isotope standards and reference materials, the organization of coordinated research programmes, and the support of technical co-operation projects in developing countries. The collaboration between the IAEA and other UN organizations, such as UNESCO, in hydrology and climatology is also underlined.

Key words climatology; environmental isotopes; hydrology; International Atomic Energy Agency (IAEA); radioactive isotopes; stable isotopes; United Nations Educational, Scientific and Cultural Organization (UNESCO)

THE BEGINNING

Isotope hydrology is the discipline within the earth sciences that studies the hydrological systems using techniques based on the determination of isotopic abundances and their changes in water and the environment. This scientific discipline was born in the fifties, after the development of mass spectrometers capable of measuring small isotope ratio variations and gas proportional counters capable of detecting the occurrence of radioactive isotopes at very low concentration, and the consequent publication of a number of studies and applications related to natural waters.

Since 1958, that is shortly after the establishment of the International Atomic Energy Agency (IAEA) as a new United Nations organization, isotope hydrology was included in its research and technical cooperation programme, in conformity with the mission to encourage the Peaceful Applications of Atomic Energy. The Isotope Hydrology group at the IAEA, initially led by Erik Eriksson, was then consolidated into the Section of Isotope Hydrology, headed by Bryan R. Payne until his retirement in 1987. Since then, the Section of Isotope Hydrology of IAEA has been led by
The Isotope Hydrology Section's strategy was set up in two advisory panels convened in the early 1960s, which brought together isotope experts and their counterparts of the hydrological community. As stated in the report of the first panel:

"the panel considered that there were two possible approaches to the development of isotope techniques aimed at the solution of hydrological problems. The first might be termed an isotope technique development approach having no particular application in mind. The second approach would be to consider particular projects and then to try and find the isotope techniques which could be applied to their solution. The disadvantage of the first approach is that the isotope specialists are not always aware of the real hydrological problems, and on the other hand, consideration of only what might be called the project approach would handicap application owing to the prevailing lack of standard working techniques. The panel strongly recommended that both lines of attack should be simultaneously followed, resulting in mutual incentive to the two main groups of workers". 

This dual role was then further emphasized as the work progressed.

The main focus of isotope hydrology in the fifties and early sixties was on the introduction of short-lived radioisotopes (readily available from the nuclear industry) in groundwater and surface waters, in order to obtain local parameters of hydrological systems such as mixing characteristics, transit times, storativity, porosity, transmissivity, discharge rate, etc. The new technology was rather well received by field hydrologists and applied in different parts of the world. However, the increasing difficulty of obtaining permission for the introduction of even small amounts of radioactive isotopes into hydrological systems used for water supply, as well as the limited time and space scale in which they could be employed, resulted in the gradual curtailment of the use of artificial tracers, and their replacement by isotopes of natural or anthropogenic origin distributed in the environment by natural processes. These isotopes are called environmental isotopes.

ENVIRONMENTAL ISOTOPES

The first of these isotopes to come into focus was tritium, the radioactive isotope of hydrogen with a half-life of 12.3 years. It is not only produced naturally in the atmosphere by the secondary cosmic radiation via the reaction $^{14}\text{N} (\text{n}, ^3\text{H}) ^{12}\text{C}$, but has also been released in large amounts (about $7.5 \times 10^5$ TBq per Megaton equivalent of fusion energy) by the thermonuclear explosions. Indeed the tritium released into the environment by the atmospheric thermonuclear tests performed in the years 1961 and 1962—just before their suspension from 1 January 1963 by the Limited Test Ban Treaty between the UK, USA and the former USSR—was $1.8 \times 10^8$ TBq. This amount corresponds to about 140 times the natural inventory, and to 1300 times that naturally produced by cosmic rays during the same period. In those years, and for many years to follow, the thermonuclear tritium completely overshadowed the natural tritium signal in precipitation. During the four decades elapsed after the occurrence of its maximum value (1963), the concentration of thermonuclear tritium in atmospheric waters has been decreasing almost regularly, reaching during the 1990s values equal, or very
close to, the natural, pre-bomb level. This trend was only slightly disturbed by the relatively smaller French and Chinese tests which took place from 1967 to 1980 and which introduced additional tritium pulses into the atmosphere.

The tritium injection by the atmospheric thermonuclear tests inadvertently resulted in the largest tracing experiment ever attempted of the whole hydrological cycle. The introduction and mixing of the excess (bomb-) tritium into the surface and groundwater bodies, promised hitherto unrealized opportunities for the study of the dynamics of these systems. This "tracer experiment" complemented the dating of hydrological systems by the natural cosmic-ray produced tritium as was suggested by W. Libby and co-workers in Chicago just prior to the bomb pulse. The latter obviously terminated the possibility of a straightforward dating approach based on cosmogenic tritium.

THE GLOBAL NETWORK OF ISOTOPES IN PRECIPITATION (GNIP)

The global aspect of this "tracer experiment" necessitated as a prerequisite the documentation of the tritium distribution in the atmosphere and surface waters (oceans, lakes, etc.), focusing the attention on the water cycle as a whole, especially the atmosphere–surface–groundwater continuum. Since from the very beginning an important task of the IAEA had been the monitoring of the radioactive fallout of nuclear explosions, it was only natural that the Isotope Hydrology Section was charged with the duty of measuring the tritium content in the environment. Under the assistance and advice of other scientists—among which were Robert Brown (Canada), Willi Dansgaard (Denmark), Joel R. Gat (Israel), Karl-Otto Münich (Germany), Edgar Picciotto (Belgium), Ezio Tongiorgi (Italy)—about 150 stations out of the network of the World Meteorological Organization were selected and monthly composite samples of precipitation were collected for tritium analysis. This network was the nucleus for GNIP (Global Network of Isotopes in Precipitation).

Soon thereafter it was decided to also determine the stable isotope composition of precipitation, i.e. the $^2\text{H}/^1\text{H}$ and $^{18}\text{O}/^{16}\text{O}$ ratios, which would help to understand the general atmospheric circulation patterns as well as in applying isotope techniques in water resources assessment. Furthermore, an intensive measurement programme of tritium in rivers, lakes and groundwater, at selected sites representative of different climate zones, was initiated, yielding some invaluable information on the dynamic aspects of the water cycle, such as the residence time of water in the different hydrological reservoirs, the hydrograph separation, the exchange rate and mechanisms between the stratosphere (where most thermonuclear tritium was injected) and troposphere and between the northern and southern hemisphere, etc. By monitoring the concentration and movement of tritium and other isotopes in water molecules, hydrologists were able to determine the rate at which water moves through the hydrological cycle: from clouds to the earth surface, to rivers, underground reservoirs, aquifers or glaciers; to the ocean; and back again to clouds. In this way hydrologists could determine the origin of water and its age, the rate of precipitation and evaporation or infiltration into the ground.

Similarly, the stable isotope data on precipitation were an invaluable source of information on the global circulation patterns of atmospheric vapour and on the
connection between isotopic composition and meteorological conditions and major climatic characteristics. The isotopic composition of atmospheric precipitation defines the isotopic input to the majority of the natural archives used for reconstruction of the climate in the past (paleo-climate).

Thus, the establishment of the GNIP has to be considered a very fruitful scientific operation, which only an international body such as IAEA could have undertaken. No wonder that the scientific communities dealing with (paleo-) climate and global circulation modelling are making increasing use of the GNIP database. Several international programmes related to paleo-climatology have commended the relevance of the GNIP database as a primary reference for recent isotope data and as a means to validate paleo-climatic reconstruction based on isotope records. In this context it should also be noted that, starting in 1998, the GNIP database was supplemented by a database compiling isotope and related data from water resources projects worldwide. The database is known as ISOHIS. The GNIP and ISOHIS data were recently merged in a common GNIP/ISOHIS database accessible through the Internet under http://isohis.iaea.org.

THE IAEA ISOTOPE HYDROLOGY LABORATORY

As the number of isotopic analyses required for the GNIP network and field projects for water resources assessment was large for any research laboratory, IAEA decided to set up its own Isotope Hydrology Laboratory for measuring tritium and carbon-14, which some years later was upgraded to include stable isotope measurements ($^{2}$H/$^{1}$H, $^{13}$C/$^{12}$C and $^{18}$O/$^{16}$O ratios). During the years, the equipment of the Isotope Hydrology Laboratory was gradually increased; nowadays it includes four gas proportional and five scintillation counters for low-level $\beta$-counting (tritium and carbon-14), five isotope ratio mass spectrometers for stable isotopes, a full water chemistry laboratory, and gas chromatographs for chlorofluorocarbons (CFCs) determination in the atmosphere and natural waters. Recently, a mass spectrometer dedicated to measurements of $^{3}$He was acquired, which will enable combined $^{3}$H and $^{3}$He analyses and thus the application of the $^{3}$H/$^{3}$He method to groundwater and surface water studies. Whenever possible, the analytical systems are run automatically and are able to process a large number of samples. For instance in recent years the samples processed for research and technical cooperation field projects were: 1200 for $^{3}$H; 3000 each for $^{2}$H/$^{1}$H and $^{18}$O/$^{16}$O; 300 for $^{13}$C/$^{12}$C; 150 for $^{14}$C (carbon-14); 150 for water chemistry; and up to 100 for the CFCs.

The IAEA laboratory provides assistance to member states in establishing and operating isotope hydrology laboratories, for which it serves as reference laboratory. In addition, the laboratory provides assistance in the preparation and distribution of reference materials for isotope measurements and organizes inter-comparison and inter-calibration exercises among laboratories. In this respect, an early highlight was set by the first Advisory Group Meeting on Stable Isotope Reference Materials in 1966, which recommended the production of two water reference materials to improve the inter-laboratory comparability of measurements of hydrogen and oxygen stable isotope ratios in water samples. This led to the preparation of VSMOW (Vienna
Standard Mean Ocean Water) and VSLAP (Vienna Standard Light Antarctic Precipitation) by Professor Harmon Craig of the Scripps Institution of Oceanography, La-Jolla California, USA. In subsequent meetings the preparation of several (35) other reference materials was suggested and undertaken, materials which were calibrated through the cooperation of selected laboratories. Towards the end of the nineties, further efforts were made to improve the analytical quality of the Isotope Hydrology Laboratory and its partners worldwide and to improve the precision of the stable isotope and tritium measurements according to the new challenges by applications to climate and groundwater replenishment studies.

FIELD PROJECTS IN ISOTOPE HYDROLOGY

Having established its own isotope hydrology laboratory, the IAEA started to be involved in field projects to assess the water resources in developing countries which were financially supported by the Technical Co-operation programme of IAEA and other UN organizations such as the FAO, UNESCO, UN, and others. In fact, it soon became clear that isotope techniques can provide information on the origin, history and dynamics of water bodies which often is inaccessible by other means. In particular, isotope techniques are capable of evaluating groundwater age, that is the time elapsed since the rain or surface water infiltration recharged the aquifers, by using the radioactive isotopes tritium and carbon-14.

With carbon-14, the age is obtained by determining the decrease of the carbon-14 concentration in the dissolved inorganic carbon due to radioactive decay. Carbon-14 has a half-life of 5730 years and thus enables determination of groundwater ages of up to 30 000 years. Carbon-14 is particularly useful in arid and semiarid regions where the recharge is low (or even negligible in modern years) and where little is known about the replenishment and flow regime of groundwater. The IAEA has provided technical assistance to many member states in arid regions.

During the five decades of the existence of Isotope Hydrology, the practical applications in hydrology have increased continuously and were extended to tackle many of the categories and aspects of water problems, including: water resources assessment in arid and semiarid countries, the origin of water resources contamination, geothermal resources assessment, environmental investigations related to climate change and its impact on the water cycle, the reconstruction of palaeo-climatic conditions, the use of artificial isotopes for measuring river discharge and sediment transport, etc.

RECENT DEVELOPMENTS AND FUTURE PROSPECTS

Some early work that followed the discovery of the stable isotopes of hydrogen and oxygen had already established the depletion of the heavy isotopes of meteoric waters relative to oceans and, in contrast, their enrichment in surface waters exposed to evaporation. However, it was not until the stable isotope composition of precipitation was determined within the framework of the GNIP programme, establishing the time
and space patterns of its variability, that the potential of stable isotope methodology for studying the water cycle became fully evident. Questions such as the interrelations between surface and sub-surface waters, the geographic and temporal origin of groundwater recharge, and the water balance of whole hydrological systems, especially in wetlands and evaporation-prone areas, could be addressed. Thus, the emphasis of isotope hydrology shifted from detailed and local problems to larger scales, such as that of the complete watershed, and beyond to the continental scale. This development required, however, a widening of the horizons to encompass the total environment and integrating engineering, geography, meteorology, and agricultural science in the programme of hydrological science. The IAEA isotope hydrology programme was thus thrust into the position of spearheading the transition to this new concept of hydrology. The environmental isotope applications rely on an in-depth understanding of the water cycle, in both detail and its totality and complexity. This has put great challenges at the door of the Hydrology Section, in that it had to play a dual role of initiating very basic research, as well as continuing its important function of furthering practical applications, especially in developing countries.

As a consequence, in recent years the practical problems of water resources assessment and management shifted increasingly from the supply of adequate amounts of water towards water quality protection from pollution and overexploitation. This development enhanced the importance of following up the movement of particular parcels of water in hydrological systems, on the one hand, and of identifying the origin and the geochemical behaviour of contaminants, on the other hand. Isotopes other than those of hydrogen, oxygen and carbon are used to trace contaminants, in particular those of sulphur, nitrogen, boron, chlorine and noble gases. Indeed the link with the geochemistry of water resources was strengthened, including the water–rock interaction processes.

In the 1980s concern about global warming and its impact on the world’s climate began to increase. One product of this concern was growing interest in establishing models of the global atmospheric circulation of water, water vapour and other greenhouse gases, as well as of ancient climates (palaeo-climatology) as a potential key to predict future changes. Since the isotopic ratios of precipitation are closely related to climatic parameters and prevailing atmospheric circulation patterns, it is evident that proxy materials recording the isotopic composition of past precipitation, namely ice-cores, palaeo-groundwater, lake and sea sediments, cave deposits and tree rings, can be used for palaeo-climatic reconstructions. Radioactive environmental isotopes, in particular radiocarbon, have been used to provide the time-scale of these climate changes. Thus, isotope hydrology is playing a pivotal role in palaeo-climological research. This explains the participation of IAEA in international research programmes such as: PALAEAUX-Management of Coastal Aquifers in Europe, and GASPAL-Continental Isotope Indicators of Palaeo-climate, both financed by the European Union, and ISOMAP, a special programme under the IGBP core-project PAGES (Past Global Changes). Moreover, permanent cooperation is established with scientists working in oceanography, atmospheric global circulation modelling, ecology (plant water balance studies), etc.

Another field of application of isotope techniques is the study of the greenhouse effect. Gases such as water vapour, carbon dioxide and methane, absorb part of the
infrared radiation released from the Earth’s atmosphere into outer space and transform its energy into kinetic energy, so causing a rise of the mean air temperature. The carbon isotopic ratio variations of atmospheric carbon dioxide and methane allow better quantification of the contribution of the various sources of these greenhouse gases and better understanding of their atmospheric cycling at the global scale. This provides a basis for improving the prediction of climate changes. The isotopic variations to be detected are small and require long-term and high-accuracy measurements. The IAEA promotes collaboration among specialized laboratories and provides support in refining the methods and assuring the quality of the measurements. In addition, since 1993 IAEA has regularly organized symposia on the use of isotopes in climate research, with an emphasis on studies related to hydroclimatic changes and their impact on catchment and surface water systems.

Recognizing the concern about “water: a looming crisis”, which is rapidly growing—especially in developing countries—the IAEA Isotope Hydrology programme continues to place emphasis on integrating isotopes in technical co-operation projects on water resources assessment and development in member states. The programme’s objectives have been defined as follows:

“to assist, through integration of isotope techniques in applied research and practical applications, in solving practical problems related to sustainable and efficient management of water resources and to promote research and field studies related to human induced changes and hydroclimatic influences on the water cycle and its interaction with other geospheric systems”.

IAEA’s Board of Governors assigns high priority to this Isotope Hydrology programme.

The IAEA has from the beginning of its Isotope Hydrology programme collaborated with several international organisations. In particular, IAEA contributes to UNESCO’s International Hydrological Programme (IHP), for which it has recently supervised the preparation of a textbook on Isotope Hydrology. Through a Memorandum of Understanding signed in 1998, the co-operation between IAEA and the World Meteorological Organization in operating the Global Network of Isotopes in Precipitation (GNIP) has been strengthened. Thus, this remains a high-priority, permanent programme of the IAEA, assuring the continuous updating of a database which is readily accessible to hydrologists, climatologists, geochemists, and environmental scientists.

To fully integrate isotope techniques into national/regional water sector programmes it was deemed necessary to increase the awareness of the communities of scientists, field hydrologists, and managers, at the national and international levels. To this aim, an International Programme of Isotopes in the Hydrological Cycle (IPIHC) was initiated in 1998 in cooperation with UNESCO and WMO. The programme includes the objective of integrating isotope hydrology in: (1) national and regional water sector programmes; (2) syllabi of universities (as part of geological, hydrological and climatological sciences); and (3) climatological research (with an emphasis on climate change and its impact on drinking water availability). In April 2000 a joint UNESCO–IAEA Planning Group discussed further details and stressed the need for such a long-term inter-agency programme, which is now called the Joint International Isotopes in Hydrology Programme (JIIPP) and is included in the International Hydrological Programme of UNESCO Phase VI.
REFERENCES

This paper is based on recent publications by the IAEA addressing the development of its isotope hydrology programme: