Nuclear techniques for investigating migration of pollutants in groundwater

A report on some practical applications for preventing pollution of water resources

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Protecting groundwater resources from pollution has been a high-priority topic in recent years.

Usually purer and cleaner than surface water, groundwater is naturally protected by an excellent filtering system. The system includes soil, clays, and rock particles that remove some soluble species, suspended particles, bacteria, and, to a large extent, viruses.

If such a filtering system is overloaded or bypassed, however, the aquifer itself may become polluted. The same type of pollutants that affect surface water — including domestic and industrial waste, seepage from septic tanks, mine drainage, sanitary landfills, and agricultural chemicals — may have a greater impact and more prolonged effect on groundwater. Pollution in aquifers is retained for hundreds to tens of thousands of years, thus jeopardizing water supplies for future generations.

Aquifers in some areas are less exposed to contamination than in others because geological and hydrochemical conditions limit the risk. The sensitivity of groundwater quality to anthropogenic activities causing pollution is termed "aquifer vulnerability to pollution." A variety of processes, and soil and aquifer properties, affect aquifer vulnerability. The determination of the parameters characterizing these processes is essential for modelling and predicting the migration of pollutants in groundwater systems. Major objectives are to prevent pollution and degradation of groundwater resources, or, if contamination has already occurred, to identify its origin so that remedies can be proposed.

At the IAEA, emphasis has been placed on practical applications of nuclear and isotope techniques that are directly or indirectly related to the solution of groundwater pollution problems. This article reports on how some of these techniques have been used in practice.

Migration of pollutants

An unsaturated (aeration) zone is a thin "skin" of geological bodies through which pollutants start their way from the earth's surface to shallow groundwater aquifers. Special instruments are required to study how fast the groundwater and pollutants move through the soil. A number of investigations have been, or are being, done using such instruments and isotope tracers within some national and international programmes, including those sponsored by the IAEA.

In the Crimean region, for example, investigations were performed in 1985–86 to study salinization of groundwater. In the Chernobyl area, investigations are directed at the migration of radionuclides released as a result of the nuclear power plant accident in 1986.*

In both cases, a number of sites for complex isotope tracer investigations were constructed. At these sites, water containing tritium was injected into the ground to study its redistribution over space and time in the soil. This was done in order to measure the velocity of the groundwater's movement in the unsaturated zone, and to evaluate the residence time (or age) of the groundwater and its recharge rate to shallow aquifers. Simultaneously at the same sites, pollutants labelled with isotopes were injected and their movements were observed.

In the Crimea, results obtained near Djankoy show that salt is transported upwards. This is due to high evapotranspiration and the low depth of the water

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* Both of these studies were performed at the All Union Scientific Research Institute of Hydrogeology and Engineering Geology in Moscow by the researchers V.T. Dubinchuk, Yu. A. Tsapenko, A.V. Borodin, and A.V. Gladkov.

Figure 1. The triple tracer experiment in Crimea.

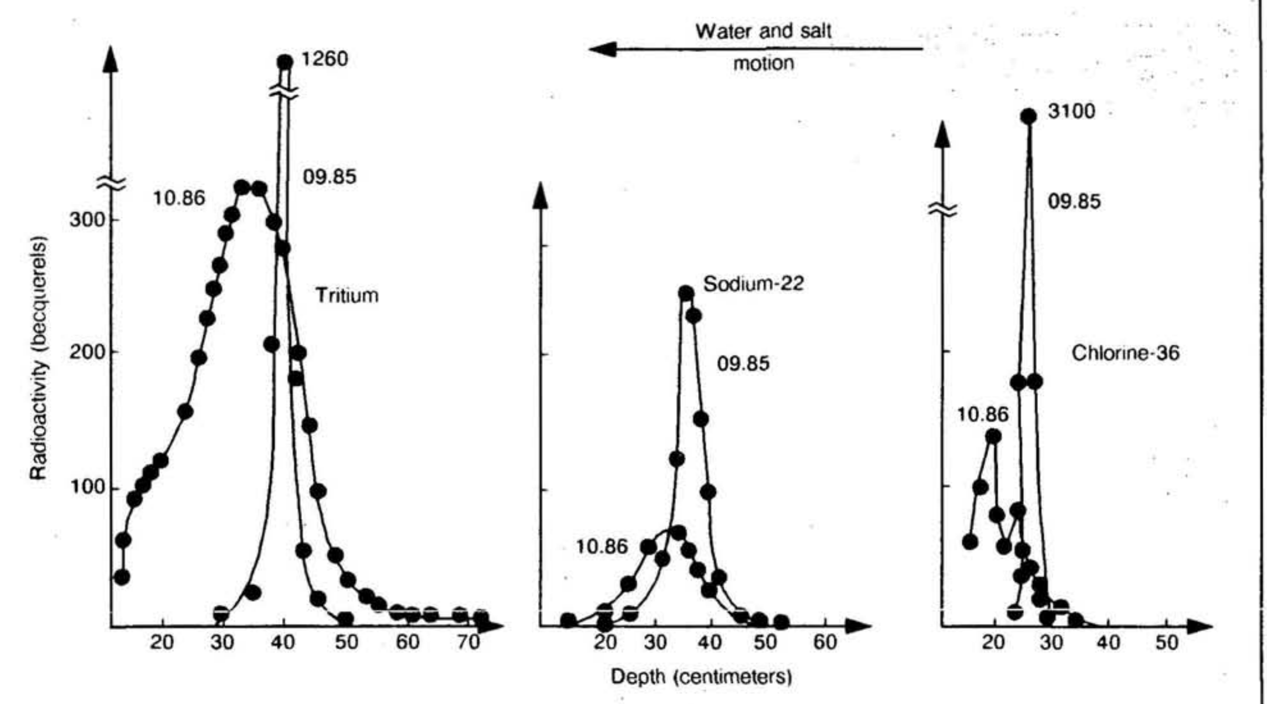
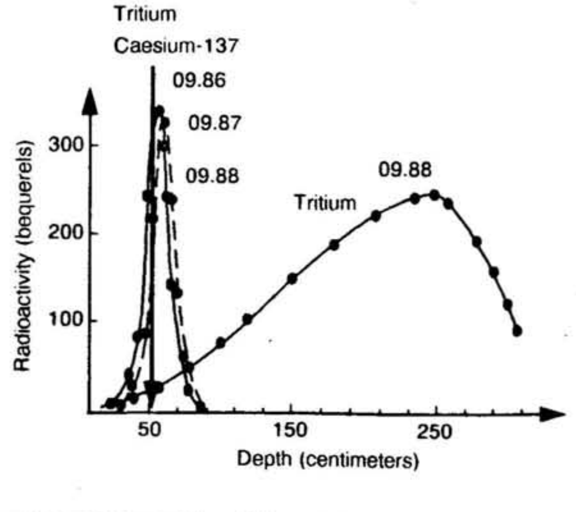


table. *(See Figure 1.)* The experiment was performed using the triple-tracer method (a tritiated water solution of sodium chloride labelled with radioactive sodium-22 and chlorine-36).

However, in other sites that are near a very thick aeration zone, similar experiments demonstrated a downward movement of salt infiltration. The investigation provided very useful data on the *in situ* transport of water, as well as on sodium and chlorine ions. Such data allow a better understanding of mechanisms and dynamics of soil salinization, which is particularly important from an agricultural point of view in this region.

In the Chernobyl area, an experiment using double tracers (tritiated water and a solution of caesium/chloride labelled with caesium-137) was performed. Injections were made simultaneously at several definite depths to obtain prognostic information. *(See Figure 2.)* The results showed, in explicit form, a retardation and dispersion of caesium in relation to water that, in turn, allows a better assessment of the risk of groundwater contamination by this radionuclide.

Figure 2. The double tracer experiment near Chernobyl.



Intrusion of polluted waters

A typical problem in water management is the intrusion into groundwater bodies of polluted waters from natural and engineered lakes, streams, or reservoirs.

In Nicaragua, near Managua, studies of intrusion are being carried out within the framework of an IAEA technical co-operation project. Investigations are based on the combined application of environmental isotopes (both radioactive and stable ones) and artificial tracers.

In Managua, Lake Asososca is one of four main pumping stations, supplying 60% of the water for the local population. Its water is heavily polluted. *(See Figure 3.)*

Lake Asososca, which is a crater of an old volcano, has a maximum depth of about 90 metres and is about

Nuclear and isotope techniques

A variety of nuclear and isotope techniques can contribute to answering specific questions about the flow of groundwater and the migration of pollutants.

Among nuclear tools, nuclear well logging plays an important role. It is based on the utilization of nuclear radiation (gamma, neutron, etc.) to determine physico-chemical properties of soils and rocks *in situ*. Gamma-logging is used to evaluate the natural emission of gamma rays from soils and, thereafter, to determine the soil's clay and carbonate content, and its permeability. Gamma-gamma logging is based on the scattering of gamma radiation emitted by an artificial source. Measurements of the radiation scattered along a well gives information about bulk density of soils. Neutron logging provides data on the water content in soil. A combination of gamma-gamma and neutron logging allows evaluation of a number of characteristics, including porosity (in saturated soils); soil matrix density; and water velocity and diffusivity (in unsaturated soils). Sometimes it facilitates estimations of fractures in consolidated rocks.

Among isotope techniques, tracers can be injected into a given water system either naturally or by a scientist to track the movement of water and its components.

The use of tracers has a long history. Simple ones, such as tree leaves, chips, and chopped straw, have been used to observe water direction and velocity as well as interconnections between streams and reservoirs. Later, different natural and man-made pigments and dyes were used, and they are still applied for this purpose.

Impressive development of tracer techniques was achieved at the end of the 19th and beginning of the 20th centuries following the invention of conductometry and colorimetry. They allowed instrumental measurements of salts and dyes as water tracers in the field. The hydrological tracer experiments at the Danube-Rhine area, in Crimea and Dinar karst, and in other regions are examples of the use of salts and dyes by this generation of hydrologists.

But the real height of development in tracer techniques was reached with the discovery of isotopes and the introduction of equipment for their measurement.

Isotopes are suitable for studying most sorts of pollution partly because they are available in a wide variety. Sometimes they are a quite unique tool with significant advantages in comparison with other tracers.

Isotope tracing can be done for either water molecules, for pollutant particles, or for both. Migration experiments and observations can be carried out at the microscopic and macroscopic levels, on a local, regional, and even global scale. Water and pollutants can be labelled without any disturbances to the conditions and processes under investigation. Experiments using isotope tracers can be done under extreme conditions — huge dilution, high mineralization, turbidity, turbulence, low and high temperatures — when other tracers are not applicable. Additionally, there are a great variety of natural isotopes in the geosphere and hydrosphere that can be used successfully to trace environmental transport processes.

One important advantage of isotope techniques is that they provide the chance to estimate, qualitatively and quantitatively, what are called "retardation parameters" of geological media with respect to the movement of pollutants. Retardation basically refers to delays in the transport of pollutants relative to water movement; it is a very complex phenomenon involving sorption/desorption, diffusion/dispersion, solution/precipitation, and many other processes.

Different isotopes can be used together as tracers to help evaluate retardation. "Double tracer" techniques may use tritium (radioactive isotope of hydrogen) as a tracer of water movement and another relevant isotope as a tracer of a particular pollutant. The techniques provide valuable information for estimating the effects of retardation.

The best results are obtained by combining laboratory methods (batch, column, and radiography) and field tracer experiments, including artificial and environmental isotope tracers. Batch experiments include observing the redistribution of labelled components between a model solution and a soil/rock sample in hydraulic static conditions. In this way, sorption-desorption isotherms, sorption distribution coefficients, and their dependency on hydrogeochemistry, pollutant, and soil properties, can be studied. It is noteworthy that the major amount of data on radionuclide and metal ion sorption has been obtained recently with batch tracer experiments.

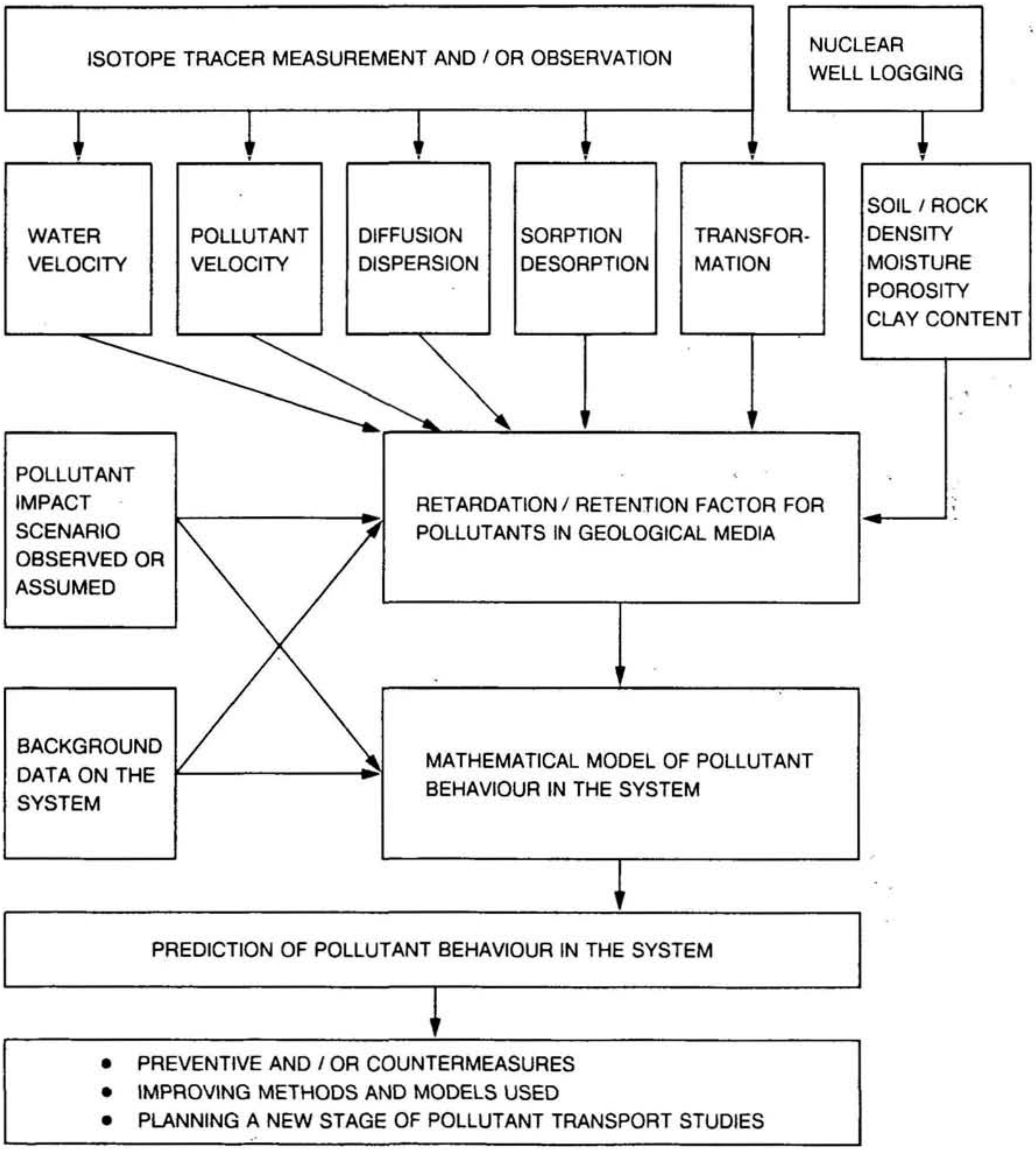
Column techniques are based on the measurement of space-time distributions of labelled components flowing through a column of soil/rock material. This provides simultaneous evaluation of hydraulic transport, sorption/desorption, and diffusion/dispersion parameters in hydraulic, dynamic conditions. Compared to batch techniques, data from column experiments are more closely related to natural systems.

Field experiments are the most preferable techniques. For the evaluation of retardation parameters under field conditions, multitracer experiments are usually done. They use artificial tritium as a tracer of water itself together with reacting solutes (pollutants) labelled by another isotope. Commonly, the tritium concentrations needed for such experiments are on the order of several tens of becquerels per liter (Bq/l).

In general, field experiments using artificial isotopes as tracers can help address a number of local problems:

- Protection of specific areas from which water is pumped for human consumption. The prediction of the pollutant's travel time from the source of the aquifer is the main objective.
- Investigation and prediction of groundwater contamination in areas surrounding waste disposal sites and big industrial complexes;
- Evaluation of water dynamics in artificial recharge systems;
- Study and forecast of the movement and behaviour of salt and agricultural chemicals in soil and groundwater.

Figure 5. Strategy for applying tracer techniques in pollution studies.



one kilometre in diameter. Its water comes from the groundwater flow which is discharged at Lake Managua. Water tables at all four pumping stations are below the water level of Lake Managua because of intensive pumping over the past decades. This fact would make it possible for polluted water to intrude from Lake Managua into groundwater and Lake Asososca.

To check this possibility, an investigation was undertaken to measure the velocity of groundwater flow and water characteristics. Studies included analyses of environmental isotopes (tritium, deuterium, and oxygen-18) and the chemical composition of water, as well as artificial tracer experiments.

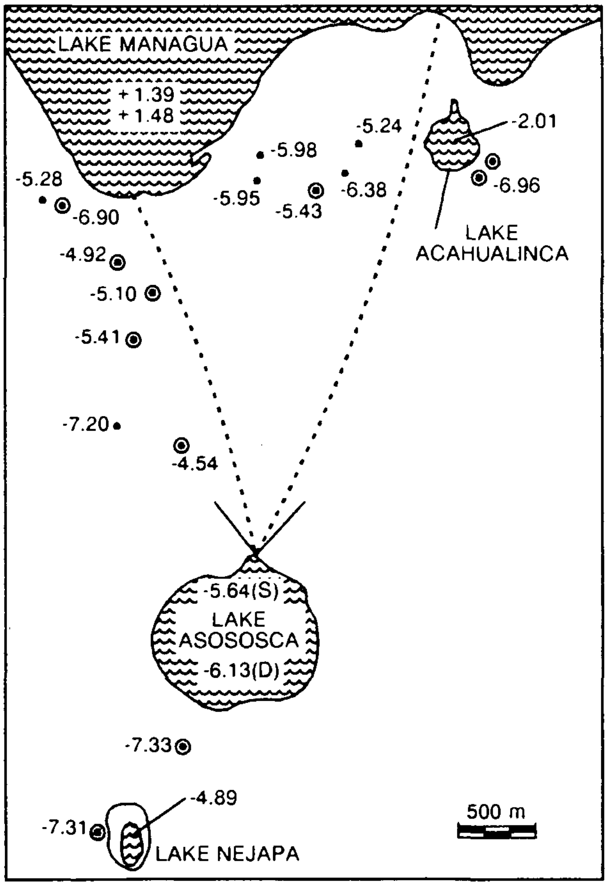
Results indicated that no intrusion of polluted water was occurring at a detectable amount. Studies showed a high difference in isotopic composition between the water from Lake Managua and the groundwater. Moreover, it was found that water being pumped from Lake Asososca is relatively old. This determination could be made because the water has no measurable amount of tritium. (Because tritium was released into the atmosphere from weapons tests during 1952–62, it can be used to date water resources and to identify modern recharge through infiltration of rain water.) Lake Asososca's water also has the same isotopic composition as the deep groundwater. On the basis of the isotopic information, it can be said that any potential intrusion of polluted water from Lake Managua due to changes in hydrodynamic conditions will be detected a long time in advance.

Artificial recharge of waste water

In Tunisia, a project was implemented for studying the artificial recharge of waste water in the Nabeul-Hammamed alluvial coastal aquifer. This aquifer is practically exhausted and useless due to intensive pumping and salinization. The recharged water was labelled with tritium to investigate the aquifer's capacity to purify itself over a long period of time (14 months) and to study mixing processes between older and younger water resources.

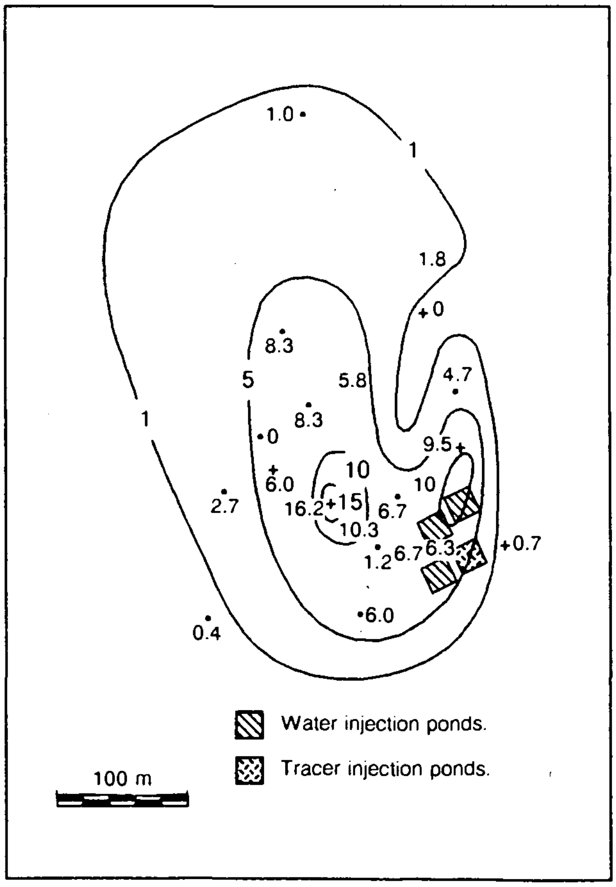
In the studies, about 32 000 cubic metres of waste water were labelled with an average concentration of

Figure 3. Isotope study of pollution intrusion in Nicaragua.



Note: Data represent oxygen-18 values in per mille deviations from the Vienna-SMOW standard.

Figure 4. Isotope study of waste water recharge in Tunisia (tritium isolines).



Note: Tritium isolines are in μ Ci/m^3.

1.2 kilobecquerel per liter (kBq/l) through the continuous injection of the tracer over 42 days. The space distribution of the labelled water 394 days after the experiment started shows a dynamic water movement. *(See Figure 4.)* The information will allow better planning of a future large-scale project to artificially recharge the waste water with the aim of using it for irrigation purposes. Citrus fruits, rice, and vegetables are the main crops in this area. It was concluded that the waste water has to be mixed with other, less mineralized water to make it possible to use the recharged water for irrigation.

Some advantages and problems

Isotope tracers provide a window into what happens with labelled pollutants (as well as with any other water component that is labelled) when they come in contact with solid soil/rock matrices or weighted, bedloaded, or suspended materials. The tracing can be made at every stage of motion, transformation, and interaction of the pollutant in geological media.

Moreover, isotope tracers can be used in extremely low concentrations from the chemical point of view, and therefore do not disturb any physico-chemical and thermodynamic conditions of the system under study. They are measurable in such small amounts that conventional tools are not able to detect them.

Finally, tracer experiments may be carried out in a non-destructive way. For example, diffusion and sorption can be observed in laboratory column experiments without destroying soil and rock samples. In the field, similar tests are easily done using gamma tracers and gamma-logging.

Isotope tracer techniques have some problems and limitations, however. As a result, in the area of pollution migration studies, scientific emphasis is now shifting from the development and improvement of particular isotope tracer techniques to their complex utilization in combination with traditional methods. This is happening in order to provide the information that is needed to make relevant prognoses and propose countermeasures.

The further development of nuclear techniques in this area should be directed to the elaboration of system algorithms for transferring laboratory and field data into environmental systems. It is necessary to develop more adequate interpretative and prognostic models, and to establish databases on typical characteristics of pollutant migration under typical geological and thermodynamical conditions.

It is often stated that a general drawback to the application of radioactive isotopes is the requirement for an environmental impact statement when researchers apply for a license to use them. This requirement can give rise to misunderstandings among the public and even decision-makers. In most cases, artificial isotopes in hydrological and hydrogeological investigations are only applied in low concentrations, which are not dangerous to workers, the public, and the environment when well-known and well-understood safety precautions are followed. The use of environmental isotopes that are naturally radioactive does not present any special safety problem.

To assist researchers and governmental authorities, the IAEA is now preparing a guidebook on the safe handling of artificial radioactive tracers in hydrology.

Other IAEA activities

Other IAEA activities in the area of pollution migration cover a number of projects involving isotope hydrology and geochemistry studies of surface water and groundwater.

Additionally, a number of expert advisory groups have been convened to review the usefulness, reliability, and applicability of nuclear techniques in groundwater assessment. Specific topics addressed include isotope tracer techniques in hydrology, vulnerability of groundwater aquifers, rare and noble gas isotopes in the study of atmosphere and hydrosphere processes, and computer modelling in isotope hydrology.

Several co-ordinated research programmes (CRPs) on these topics are being carried out. They include a CRP on modelling of the transport of isotopes in hydrogeological systems, and a CRP on the elaboration and evaluation of nuclear techniques in pollutant transport studies. The latter CRP involves scientific groups from Australia, Brazil, France, Czechoslovakia, Denmark, Finland, Germany, United States, and USSR.

Also being supported are several regional technical co-operation projects in the field of hydrology and hydrogeology in Latin America, the Middle East, Africa, and Asia. The IAEA also regularly holds training courses to educate specialists from different countries about methodologies and practices of isotope techniques, including those applications and methods most useful for solving problems of water protection.

In March 1991, the IAEA is convening an International Symposium on Isotope Hydrology that will include discussion of the use of isotope techniques in groundwater pollution/protection studies. The meeting is the eighth symposium on isotope hydrology since 1963.