Radioactivity and earth sciences: Understanding the natural environment

The earth's natural radioactivity is helping scientists learn more about geological processes and global climate changes

by Kazimierz Rozanski and Klaus Froehlich

Virtually all natural substances contain radionuclides to a greater or lesser degree. Such natural radioactivity — discovered by French physicist Henri Becquerel a century ago — today is being put to practical uses in many fields of science and industry.

In the geological sciences, the transfer of natural radionuclides according to their physical and chemical properties is being studied to trace the evolution of the crust and mantle of the Earth, to follow processes associated with the hydrological cycle, and to account for some aspects of the composition of the atmosphere. The radioactivity of rocks, minerals, water, and organic matter is widely used in dating geological and archeological materials as well as groundwater.

Energy liberated during radioactive decay is now considered to be one of the main sources of heat within the Earth. The driving force for lithospheric plate movements, mountain orogenesis, and vulcanism may have an origin in the presence of radionuclides. Even on local scale, the presence of radionuclides may give rise to areas of high heat flow in crustal rocks. This in turn causes convective circulation of groundwater which can lead to the formation of hydrothermal systems and associated mineralization.

The discovery and subsequent utilization of fission and fusion reactions have added new elements to the pool of radioactive materials present in the environment. Numerous radionuclides have been added to the earth's ecosystem mainly through atmospheric nuclear bomb tests, and through emissions from the nuclear industry, among others. Tracking their transport through different compartments of the global ecosystem provides new insights into the dynamics of the atmosphere and the hydrological cycle. *(See box.)*

Mr. Rozanski is a staff member in the IAEA Istotope Hydrology Section, Division of Physical and Chemical Sciences, and Mr. Froehlich is Head of the Section.

The earth's natural clock

Perhaps the most characteristic and successful application of radioactivity in earth sciences is its use as a natural "clock" measuring successively various processes occuring on the Earth. The versatility of this clock is remarkable: it can work effectively over more than 15 orders of magnitude quantifying processes ranging from minutes to billions of years. To effectively date any material of geologic or biologic origin, two basic requirements need to be fulfilled: (1) possible sources and sinks of the radionuclide(s) in use need to be carefully assessed; and (2) the past physical status of the system to be dated has to be known or assessed (open or closed system).

A variety of radionuclides is used for geological and archeological dating: radionuclides produced during stellar nucleosynthesis (primordial radionuclides), radionuclides of the natural decay series, radionuclides produced by natural nuclear reactions in the atmosphere (cosmogenic radionuclides) and lithosphere (*in-situ* produced radionuclides), and radionuclides produced in artificial nuclear reactions (anthropogenic radionuclides).

How does the radioactive clock function? It relies on the fact that radioactive decay is independent of the physical and chemical conditions and changes in the environment. The rate of decay for a particular radionuclide is governed by the half-life of the decay. This can be defined as the time taken for a given number of atoms of that radionuclide to decay to half that number. To enable dating to be carried out accurately, half-lives that are of the same order as the age of the material to be dated are required. Fortunately, natural radionuclides have half-lives which range from less than one second to more than 10^{10} years. *(See table next page.)* Consequently, a very wide range of dating is possible, including estimates of the age of the Earth and the solar system. Some of the dating methods are based on the relationship between the radionu-

Major applications of natural and anthropogenic radionuclides in earth sciences

Studies of Atmosphere

- dispersion, transport, and mixing processes on local, regional, and global scale (tritium, krypton-85, radon-222, carbon-14)
- transport of water vapour (tritium)
- stratosphere-troposphere exchange (tritium, carbon-14, krypton-85, beryllium-7, beryllium-10)
- sources and sinks of CO_2 and CH_4 (tritium, carbon-14)
- atmospheric deposition (chlorine-36, beryllium-7, beryllium-10, strontium-90, caesium-137)

Studies of Hydrosphere

Continental hydrosphere

- replenishment of groundwater resources (tritium, chlorine-36)
- dispersion studies in surface waters (tritium)
- aeration studies (krypton-85)
- interaction between surface and groundwaters (tritium, radon-222, carbon-14)
- groundwater dating (tritium, carbon-14, krypton-85, argon-39, chlorine-36, krypton-81)
- rock-water interactions (uranium-238, uranium-234, radium-226, radium-228)
- sedimentation rates in lakes and reservoirs (caesium-137, lead-210)
- radioactive waste disposal (chlorine-36, iodine-129)

Ocean

- circulation and mixing processes (tritium, carbon-14, krypton-85)
- age of water masses (tritium, carbon-14, argon-39, krypton-85)
- transfer of anthropogenic CO_2 into the ocean (carbon-14)
- dating of oceanic sediments (carbon-14, potassium-40)
- variations of sea level in the past (carbon-14, uranium-234, thorium-230)

Studies of Lithosphere

- dating of rocks and minerals (potassium-40, argon-39, rubidium-87, lutetium-176, hafnium-174, samarium-147, neodymium-143, rhenium-187, radionuclides of uranium and thorium decay series)
- dating of carbonate deposits (carbon-14, uranium-234, thorium-230)
- dating of lacustrine sediments (caesium-137, lead-210, carbon-14, uranium-234, thorium-230)
- surface exposure dating (beryllium-10, carbon-14, aluminum-26, chlorine-36)
- soil erosion (caesium-137, lead-210, beryllium-10)
- mineral exploration (radionuclides of uranium and thorium decay series)
- earthquake monitoring (radon-222)
- paleoseismicity and volcanic eruptions (chlorine-36, aluminum-26, beryllium-10)

clide and its decay product, which in such cases is usually a stable daughter nuclide.

Until the early 20th century, the opinions about the age of the Earth differed considerably: the estimates ranged from some tens of millions of years to several tenths of billions of years. The debate was resolved in 1929 when Rutherford used

Natural and anthropogenic radionuclides frequently used in earth sciences

Nuclide	Half-life (years)	Origin *
Tritium	12.43	N+A
Beryllium-7	9.7×10^{-2}	N
Beryllium-10	1.6×10^{6}	N
Carbon-14	5730	N+A
Silicon-32	140	N
Chlorine-36	3.01×10^{5}	N+A
Argon-39	269	N
Krypton-85	10.76	A
Krypton-81	2.1×10^{5}	N
Iodine-129	1.57×10^{7}	N+A
Potassium-40	1.31×10^{9}	N
Rubidium-87	4.88×10^{10}	N
Caesium-137	30.17	A
Radionuclides of uranium and thorium decay series		
Uranium-238	4.47×10^{9}	N
Uranium-235	7.13×10^{8}	N
Uranium-234	2.48×10^{5}	N
Protactinium-231	3.43×10^{4}	N
Thorium-230	7.52×10^{4}	N
Radium-226	1602	N
Radium-228	5.75	N
Radon-222	1.05×10^{-2}	N
Radon-220	1.76×10^{-6}	N
Lead-210	22.3	N

* N — Natural (decay of primordial radionuclides produced in stellar nucleosynthesis, interactions of cosmic rays with the atmosphere and/or with the earth's crust); A — anthropogenic (atmospheric and underground nuclear explosions, nuclear industry, watch industry, hospitals, etc.), N+A — significant contributions from both natural and anthropogenic sources.

the newly discovered radioactivity for measuring ages of rocks and soon obtained an age of billions of years using the U-He method. The first precise determination of the age of the Earth and meteorites was performed in the early 1950s and was based on lead isotopes, the stable end-products of the respective radioactive decay series. It gave the age of the Earth as 4.55 billion years — a value that remains generally accepted today for the age of the Earth and the solar system.

Detecting natural radioactivity

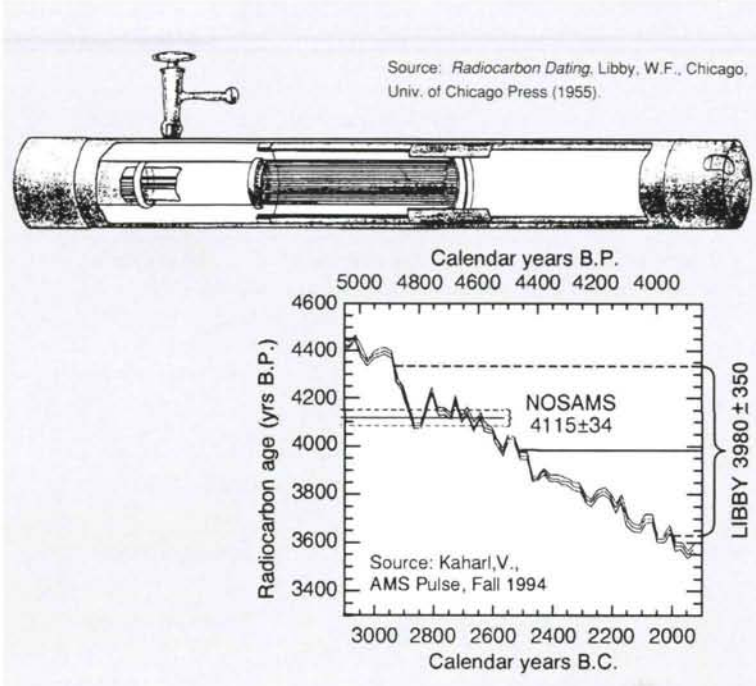
Natural radionuclides were first detected through the ionizing radiation they emit during their decay. This "decay-counting" technique has been progressively developed since the dis-

The radiocarbon clock

A multi-wire wall counter used by Willard Libby to measure the carbon-14 content in archeological artefacts *(right)*. Libby won the Nobel Peace Prize in 1960 for his work in developing the method of radiocarbon dating. ***Bottom:*** Example of modern carbon-14 AMS equipment, a commercially available tandetron accelerator dedicated for radiocarbon analysis. The first sample dated by Libby in 1948 was a piece of *acacia* wood from furniture found in Pharoah Zoser's tomb at Saqqara, Egypt. Libby needed about 20 grams of this precious archeological artefact to perform the analysis. Based on astronomical events, hieroglyphics and other historical records, various Egyptologists have consistently dated the reign of King Zoser to about 2600 BC. As shown in the graph, Libby dated the wood to 2030 plus or minus 350 years BC or 3980 plus or minus 350 years BP (Before Present). In 1992, the same piece of *acacia* wood from Pharaoh Zoser's tomb was dated as a first sample in the newly established National Ocean Sciences Accelerator Mass Spectrometry Facility at Woods Hole Oceanographic Institution, Woods Hole, Massachusetts, USA. This time only about *10 milligrams* of wood was sufficient to perform the analysis. The AMS radiocarbon age of the Zoser wood was 4115 plus or minus 34 years BP, i.e. within one sigma error of the first Libby analysis.

To convert radiocarbon years to calendar years, corrections are required which account for fluctuations of radiocarbon content in atmospheric carbon dioxide in the past. Such corrections are based on measurements of radiocarbon concentrations in a series of tree rings dated by dendrochonological techniques. The tree ring calibrated age for the AMS analysis of the Zoser wood yields two solutions with a 95% degree of confidence: 2877 to 2800 BC and 2780 to 2580 BC. This result is consistent with archeological estimates. However, a marked step in the calibration curve during the time of Zoser's reign precludes a high precision calibrated age estimate for this particular material.

(Photo credit: Prof. P.M. Grootes, Christian Albrechts University, Kiel, Germany)

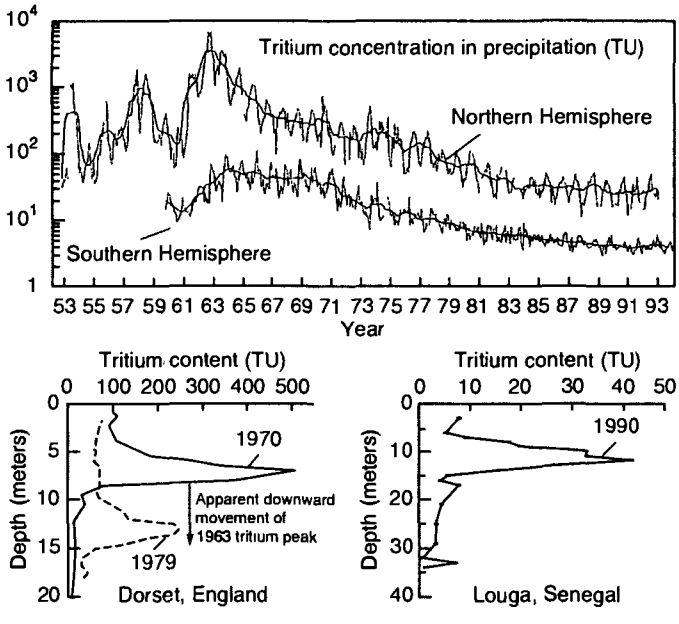


covery of natural radioactivity a century ago. Decay-counting has become a well established field and a wide variety of materials (gases, liquids, semiconductors, etc.) are being used to detect different types of ionizing radiation emitted by various radionuclides. Since the activities of natural radionuclides are usually very low, sophisticated signal processing and background reduction techniques are often employed to increase the sensitivity of such detectors. For the detection of short-lived radioisotopes, with half-lives of less than about one year, the sensitivity of advanced decay-counting techniques is appropriate. Primordial radioisotopes with half-lives greater than about 10^9 years are relatively abundant naturally (since they have not decayed completely over the life of the solar system). The decay-counting is often replaced here by measuring the build-up of stable decay products by conventional mass spectrometry. Radioisotopes with half-lives in the intermediate range of 10^3 to 10^8 years are difficult to measure using the decay-counting techniques since only a small fraction of the atoms decay over a reasonable counting period. For instance, if carbon-14 concentration in organic samples is measured using a gas

Tritium content from atmospheric nuclear bomb tests

Shown in the upper graph below is the increase of tritium content in precipitation as a result of atmospheric nuclear bomb tests. The tritium content of precipitation is being monitored on the global scale by the IAEA/WMO Global Network Isotopes in Precipitation (GNIP). Data of two long-term stations are presented here covering the northern and southern hemispheres.

Lower panel: Penetration of the bomb-tritium peak in the unsaturated zone of the aquifer located in a temperate climatic zone (Dorset, England) and in a semi-arid region (Louga, Senegal). For the Louga profile, obtained in Senegal in the framework of an IAEA regional technical co-operation project, the average replenishment of this aquifer over the past three decades was estimated to be around 22 mm/yr. Such low recharge rates are very difficult to measure using classical hydrological methods.



proportional or liquid scintillation counter, on the average only one atom out of each 10^6 atoms of carbon-14 present in the sample decays and contributes to the measured signal. Therefore, relatively large samples of the analyzed material are required.

Particle accelerators, such as those built for research in nuclear physics, can also be used together with magnetic and electrostatic mass analyzers to measure radioisotopes at very low concentrations. The work in this direction started in the late 1970s. Today, several long-lived radioisotopes which were very difficult to measure by decay-counting (beryllium-10, carbon-14, aluminum-26, chlorine-36, calcium-41, iodine-129) can routinely be measured in small natural samples having isotopic abundances in the range 10^{-12} to 10^{-15} and as few as 10^5 atoms. With this new analytical technique, called Accelerator Mass Spectrometry (AMS), it has been possible to reduce the sample size by several orders of magnitude. For instance, the amount of carbon required for radiocarbon dating could be reduced from a few grams (decay counting) to some tens of micrograms (AMS). Also, the counting period can be reduced substantially. Over the last decade, research applications of AMS have been concentrated in earth sciences (climatology, cosmochemistry, geochronology, geomorphology, hydrology, glaciology, minerals exploration, sedimentology) and in anthropology and archeology (radiocarbon dating). *(See box, previous page.)* Over the past few years, AMS has also become an important analytical tool for the materials and biological sciences.

Dating of groundwater

Radioactive isotopes have found numerous applications in hydrology. Tritium and carbon-14 are extensively used as "dating tools". The atmospheric tests of nuclear fusion bombs carried out during the 1950s and early 1960s released large amounts of artificial tritium into the atmosphere and consequently the hydrosphere. The observation of this transient tritium pulse in the water cycle led to numerous hydrological studies on a global, regional, and local scale. This "bomb-tritium" became a powerful indicator of young water in groundwater systems and a useful tracer in determining the rate of replenishment of aquifers, particularly in semi-arid and arid regions. *(See graphs, this page.)*

In spite of the complexity of the carbon geochemistry in groundwater systems, both natural and anthropogenic carbon-14 became a widely used used tool in assessing groundwater ages in the range between a few thousand and a few tens of thousands of years. For instance, radiocarbon made it possible for the first time to assess the age of large groundwater reserves under the Sahara. *(See graph, next page.)*

The AMS technique also has enabled the applications of some new radioisotopes in hydrology, such as chlorine-36 and iodine-129. With chlorine-36, groundwater ages in large sedimentary basins such as the Great Artesian Basin in Australia were estimated to be in the order of up to one million years. Measurements of iodine-129 in deep formations of water associated with oil deposits help to clarify the origin and age of these waters.

Quantifying erosion and sedimentation

The process of erosion involves different time scales: from relatively fast, often anthropo-

genically triggered processes of soil erosion, to relatively slow processes of weathering of rock surfaces. Soil erosion represents a serious problem in many parts of the world because of its impact on sustainable agricultural development.

A number of radioisotopes, both natural and anthropogenic, can in principle be used to assess soil erosion rates, depending on the time scale involved. Among them, caesium-137 and lead-210 by far have been the most commonly used.

The IAEA is addressing this problem through the ongoing Co-ordinated Research Programme (CRP) "Soil Erosion and Sedimentation Assessment Studies by Environmental Radionuclides and their Applications to Soil Conservation Measures" with the participation of research institutes from ten Member States. The programme is aimed at further development of isotope-based methodologies for evaluating the soil erosion rates in different climatic settings, for measuring sediment yields from river basins, and for evaluating siltation rates in lakes and reservoirs.

The past decade has seen development of several new methods for quantitative age-determinations of geomorphic surfaces, triggered by the introduction of the AMS technique. These methods are based on the accumulation of cosmogenic radionuclides (beryllium-10, carbon-14, aluminum-26, chlorine-36, and calcium-41) in rocks exposed at the earth's surface. The radionuclides in question are produced in the interactions of cosmic rays with atoms in minerals by high-energy spallation, neutron-capture reactions and muon-induced nuclear disintegrations. The effective dating limits of the radionuclides produced *in situ* are from a few thousands to several millions of years.

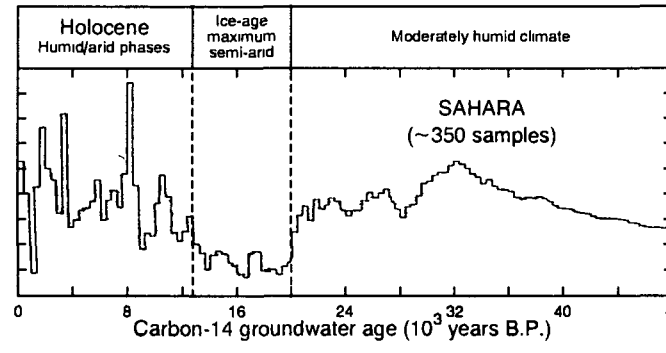
Chronology of past climate variations

Radioisotopes remain the major tool for providing the chronological framework for reconstructions of past climatic and environmental changes. A broad suite of isotopes has been used for this purpose, depending on the time scale of the processes being studied and the nature of samples to be dated. Among them, radiocarbon remains the most often used; hundreds of thousands of radiocarbon analyses have been performed over the last four decades in carbon containing materials from oceanic and lacustrine sediments, tree rings, ocean water, groundwater, atmospheric carbon dioxide.

Additionally, the uranium-thorium dating method is becoming more frequently used. This is particularly the case for its modern version based on the detection of uranium and thorium isotopes in the analysed sample using Thermal Ionization Mass Spectrometry (TIMS) instead of decay counting. This method is capable of dating geological materials (carbonates, sediments) back to about 350,000 years. Of particular importance for high-resolution reconstructions of climatic changes during the last deglaciation are efforts to reconstruct past changes of carbon-14 content in the atmosphere, linked with variations in the production rate of this radioisotope and changes in the circulation of the ocean.

Frequency distribution of radiocarbon ages in groundwater samples collected in the Sahara

The fact that large groundwater reserves are laying beneath the largest desert in the world has been known since the beginning of this century. However, only with the onset of radiocarbon dating has it been possible to estimate the age of these waters. It is evident from the graph (based on data from 350 samples collected in Algeria, Libya, Egypt, and the Southern Sahara) that aquifers in Northern Africa where recharged predominantly during both the pluvial periods of the Holocene and during the interstadials of the last glacial period. The well-marked minimum in the distribution curve ranging from about 12 to 20 thousands of years BP reflects the period of maximum aridity in Northern Africa. The estimation of groundwater age has a direct impact on exploitation of groundwater resouces: the lack of tritium and low concentrations of radiocarbon suggest that the given aquifer is not being replenished at present.



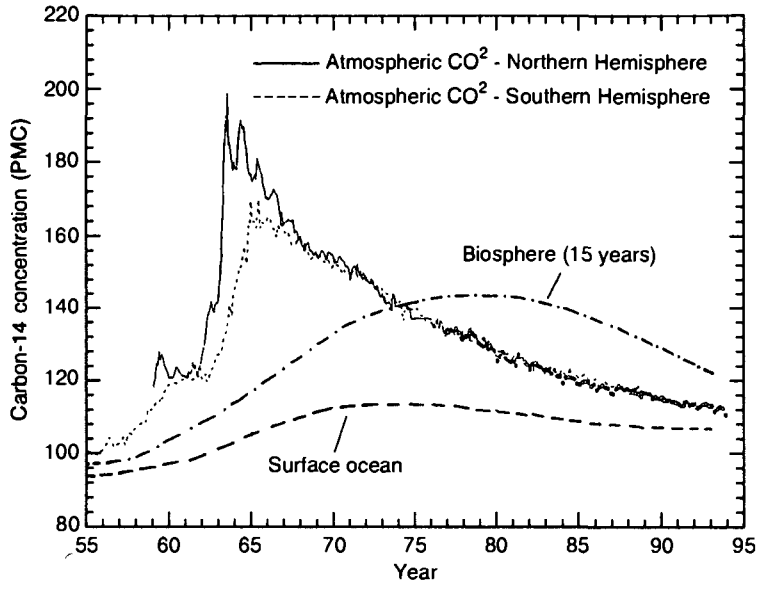
Source. Sonntag et. al, Radiocarbon, 22 (1987), 871-879

Search for the "missing sink"

The nuclear bomb tests in the atmosphere left a distinct "pulse" of carbon-14 concentration in atmospheric carbon dioxide. *(See graph, next page.)* This pulse can serve as a tracer for the global carbon cycle, in an analogous way as bomb-produced tritium is being used in studies of the global water cycle. The atmospheric reservoir of bomb carbon-14 is now being gradually emptied and the excess of carbon-14 activity is going through the biosphere and the ocean carbonate system. By watching the time evolution of carbon-14 in corresponding reservoirs (atmosphere, biosphere, ocean) one can learn more

Changes of carbon-14 concentration in atmospheric CO_2 due to nuclear bomb tests

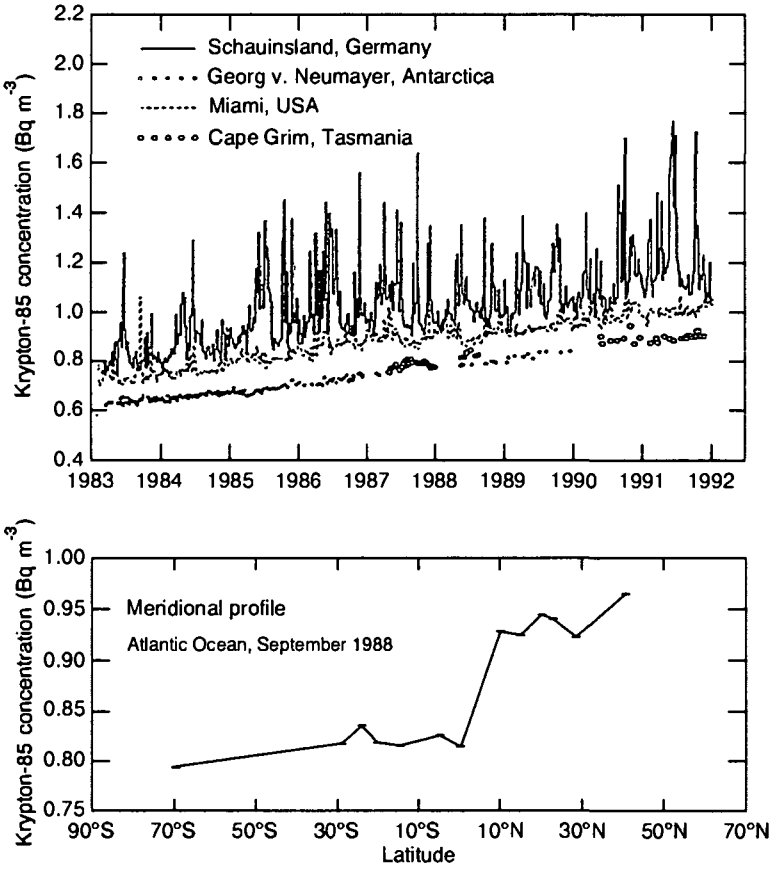
Long-term observations of carbon-14 are performed at several stations of both hemispheres (Schauinsland, Germany; Izaña, Tenerifa, Spain; Cape Grim, Tasmania; Merida, Venezuela; Neumayer Station, Antarctica). Shortly after the atmospheric test ban treaty in 1962, the carbon-14 level in the northern hemisphere was twice as high as the natural level defined here as 100%. The carbon-14 decline after 1963 is caused by strong uptake of bomb carbon-14 by the oceans and the biosphere. The broken line shows the calculated response of the biosphere to the increase of carbon-14 in atmospheric carbon dioxide, assuming that the biosphere is a well-mixed reservoir with turnover time of carbon equal to 15 years. The terrestrial biosphere is probably the most complex reservoir within the global carbon cycle. Atmospheric carbon-14 observations help to assess the nature and dynamics of this reservoir.



Source Hessheimer et al , Nature, 370 (1994), 201-203, Levin, I Thesis B, University of Heidelberg, (1994)

Krypton-85 concentration in the troposphere

Krypton-85 (half-life 10.76 years) is a fission product and is being released into the atmosphere during reprocessing of spent nuclear fuel. Krypton-85 concentrations in the southern hemisphere are systematically lower due to the fact that major reprocessing plants are located in the northern hemisphere and part of the krypton-85 released at mid-northern latitudes decays before it has a chance to be transported to the southern hemisphere. ***Lower panel:*** Meridional profiles of krypton-85 concentrations in the troposphere are used to calibrate physical parameters of the global models of atmospheric circulation. As inert gas, krypton-85 is also an excellent tracer for young groundwaters.



Source: Weiss et al. STI/PUB/859, IAEA, (1992), W Weiss, Fort. Strahl. Umwelt. Radioec. (1993)

about the rates of carbon transfer among these reservoirs, in particular among the atmosphere and the ocean.

A quantitative understanding of the global carbon cycle is of the utmost importance in view of the fact that carbon dioxide is the main greenhouse gas contributing about 50% to the postulated global warming.

The central problem in balancing the global carbon cycle is the "missing sink" for the CO_2 released every year into the atmosphere by combustion of fossil fuels amounting to about six gigatons. From atmospheric observations of CO_2 it is known that approximately 50% of this amount, i.e. about three gigatons, stays in the atmosphere. On the other hand, the available coupled ocean-atmosphere general circulation models predict that the world ocean is capable of absorbing only about two gigatons per year. The present imbalance amounts therefore to about one gigaton. This "missing sink" is in fact twice as high due to the biospheric sources associated with changes in land use, estimated to be in the order of one gigaton per year.

The results of atmospheric $^{14}CO_2$ observations offer an attractive tool to constrain the global carbon cycle. In fact, in a recently published study, based on the analyses of tropospheric carbon-14 concentrations, it has been suggested that the oceans take up about 25% less anthropogenic CO_2 than had previously been believed. Thus, the search for an additional sink of carbon, not accounted for in the present global carbon budget, needs to be continued.

Improving atmospheric transport models

Krypton-85 is being released into the atmosphere during reprocessing of spent nuclear fuel. The major reprocessing plants operate in the Russian Federation, North America, and Europe and are located in the latitudinal band 30°N to 50°N. Current atmospheric krypton-85 levels (around one Bq/m^3) can be easily measured using the decay-counting technique. Because krypton-85 is chemically inert and the only removal process of importance is radioactive decay, it is considered a very powerful atmospheric tracer.

A quantitative understanding of global atmospheric circulation is indispensable for meaningful estimates of the global balances of atmospheric pollutants and their climatic implications. In particular, two aspects of this circulation have to be described correctly: (1) the large-scale exchange of air between the northern and southern hemispheres, and (2) the intensity of vertical mixing. Because of the complexity of the processes involved, the application of numerical models is indispensable in this field. The most advanced among them, called general circulation models (GCMs), are used also for predictions of climatic consequences of emissions of greenhouse gases. The global distribution of krypton-85 can be used to adjust the model parameters related to the long-range transport and mixing between the northern and southern hemisphere. *(See graph, previous page.)* Other processes of importance, such as convective mixing within the troposphere, particularly in tropical regions and over continental areas of the northern hemisphere, cannot be parameterized by atmospheric krypton-85 distribution because the time scales involved are much shorter. Other tracers such as radon-222 can be used for this purpose.

Perspectives

The impact of the discovery of radioactivity on the earth sciences cannot be overemphasized. Natural radioisotopes have long been used as an important (and often the only) source of information regarding the chronology of geological processes, the history of meteorites and cosmic rays, human evolution, and the dynamics of biological systems. Anthropogenic radioisotopes, although viewed in the public's perception as a threat to the human environment, have turned out to be excellent global tracers. They allow us to better understand the water cycle and to learn more about the biogeochemical cycles of important life-supporting elements such as carbon, nitrogen, or sulfur.

As we approach the coming century, issues related to global environmental change are rising higher on the agendas of many international organizations, including the UN system. Already, the hydro-climatic evolution of the earth over the short-term, and humanity's influence on climate, stands as a major challenge and will necessarily become one of the most important tasks for scientists of the next century.

The quantification of the possible response of the earth's climate to anthropohenic stress is crucial, particularly for the inter-tropical zone. This region, occupied mostly by developing countries, is submitted to extreme hydroclimatic variations, such as droughts and floods. Consequently, predictions of such events over the short-term are a major requirement for the region's sustainable development. Scientific techniques based on the use of both radioactive and stable isotopes have an important role to play in accounting for the mechanisms and processes shaping the continuing hydro-climatic evolution of the globe. ❐