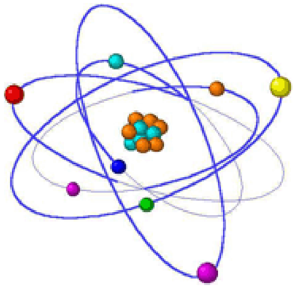
Radioisotope and Radiation Applications (FS2013)



Radionuclides to Protect the Environment (Week 7b)

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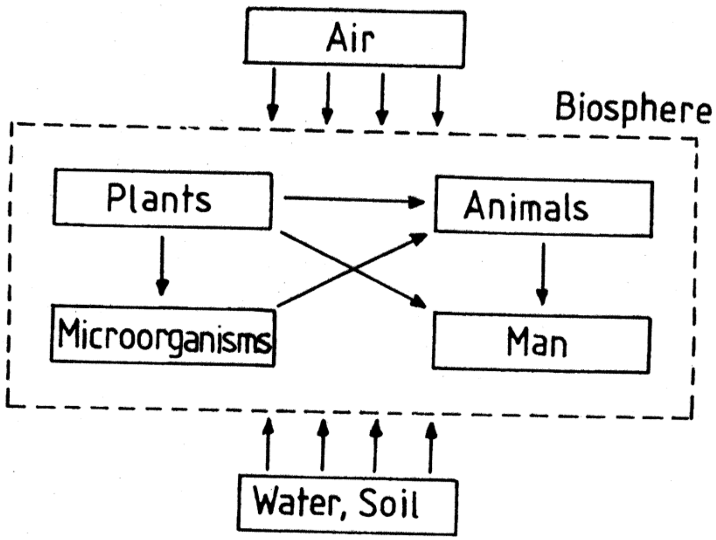
19.11. 2013

Outline

- Introduction
- Applications Overview
- Environmental Applications of man-made Radioisotopes
  - River Flow Measurements
  - Studies of Contaminants-Dispersion
  - Case Study: Sewage Dispersion
  - Sediment and Sand Tracing
- Environmental Applications of natural Radioisotopes
  - Erosion Studies
  - Groundwater Studies
  - Oceanography
- Nuclear Waste Disposal

Introduction

- Radionuclides are also applied to assess the impact of human activities on the environment. Here two methods may be distinguished:
  - **The modeling approach:** Numerical models are widespread in environmental science to reach detailed understanding of processes in the oceans, the atmosphere, coastal and terrestrial ecosystems and to make predictions. **Investigations with radiotracers are a powerful tool for model validation.**
  - **The archival approach:** Use evidence from the past to understand the present and to predict the future. The approach involves two steps:
    1. Systematic dating of material that accumulated layer by layer over long periods of time (cores extracted from sediments, ice sheets, old growth trees, …). From the location in the cores and the dates of the samples the average growth rate can be calculated.
    2. Measurement and interpretation of the indicators of past environmental change found in the samples.
- **Radioecology** is a branch of ecology that studies the behaviour of radionuclides in ecosystems comprising air, water, soil, microorganisms, plants, animals and man.



Applications Overview (1)

No	Environmental Sector	Investigation	Environmental Impact	Radioisotope technique	Comments
1.1	*Environmental science and engineering* Contaminant dispersion	Evaluation of ocean sewage outfalls	Potential impact on public health, on the recreational uses of beaches, on fisheries, etc.	Artificial radiotracers (tritium (HTO), ^{198}Au or ^{99m}Tc) used to study the dynamics of the processes, and to validate numerical models	**discussed later**
1.2	Dynamics of inland sewage ponds	Verification of the engineering design specification and seeking causes for non-compliance	Potential for impacting on the local river systems or groundwater	Tritium and ^{99m}Tc tracers used to measure residence time distributions and identify 'short circuiting'	See Section 8.3.8 Essential that the residence times of the sewage ponds are sufficient to ensure complete aerobic oxidation of pathogens
1.3	Ecological impact of contaminant dispersion	Quantifying the distribution through ecosystems of specific heavy metals, organic toxicants or nutrients	The degradation of ecosystems must be avoided if sustainable development is to be achieved	A wide range of radionuclides are available e.g. ^{65}Zn, ^{64}Cu and ^{54}Mn used to study uptake of heavy metals, ^{32}P for phosphates, etc.	
1.4	Discharges of air pollutants from industrial stacks	Validation of stack dispersion models	Degradation of air quality in the vicinity of the stack	Chemical tracers detectable with high sensitivity are normally used	

Applications Overview (2)

2.1	*Coastal and hydraulic engineering* Applications to dredging	Optimisation of location of dumping sites for dredge spoil	If not optimally located, the dredge spoil might for example migrate into a shipping channel, or contaminate the coastline	Dredge spoil labelled with e.g. ^{198}Au, ^{46}Sc and tracked	
2.2	Harbour and port development	Measurements of the migration of sand and sediment to e.g. optimise the alignment of dredged shipping channels or sea walls	Failure to optimise alignment will give rise to a need for ongoing maintenance, dredging and other negative impacts	Sand and sediment tracing (^{198}Au, ^{192}Ir, etc.) over periods from 1 week to 1 year used to validate transport codes	**discussed later**
2.3	Hydraulics	Gauging of rivers		Tracer dilution methods for calibration or where gauging stations are not available	**discussed later**
3	*Groundwater*	Efficiency of recharge to groundwater basins; the dynamics of groundwater flow	Over-exploitation or pollution of a groundwater basin can have impacts which persist for tens to hundreds of years or longer	Cosmogenic isotopes tritium, ^{14}C used to age samples and hence define recharge areas and migration rates; stable isotopes D/H and ^{18}O/^{16}O to study groundwater source and evidence for mixing.	Isotopic methods are of greatest value where extensive hydraulic data are unavailable

Applications Overview (3)

No	Environmental Sector	Investigation	Environmental Impact	Radioisotope technique	Comments
4.1	*Sedimentation and erosion* Sedimentation	Rate of accumulation of sediments in reservoirs and estuaries	Loss of capacity of reservoirs Impact on harbour development and on the maintenance of shipping channels	Sediments are cored; the cores sectioned with depth and the sections dated	The dating technique chosen depends on the time scale: ^{137}Cs up to decades; ^{210}Pb to 100 y; ^{14}C to 20 000 y; ^{230}Th/ ^{234}U to 250 000 y
4.2	Erosion	Rate of erosion and accumulation of soils in catchments	Impact of land use change on soil erosion and productivity	^{137}Cs profiles over catchments allow quantitative estimates of soil erosion and accumulation	Such data can be used to calibrate empirical equations designed to predict erosion rates over geographical regions
5.1	*Oceanography*	Transport of solutes and particulates in the water column		Profiles of 'bomb' isotopes, analysis of U, Th and Ra isotopes in the water column; ^{230}Th /^{234}U in sea bed	Applications widespread, e.g. impact of rivers on coastal zone, transport of nutrients, development of coral reefs, etc.
5.2	Ocean currents	Validation of ocean current models	Global ocean currents directly impact climate	^{14}C used to measure the residence times water bodies	Residence time is the time elapsed since the ocean water sample was at the surface
6.1	*Global climate change* Atmospheric transport	Validation of atmospheric transport models	Improved prediction of postulated 'green-house' effects	Radon used to establish whether air mass passed over land	Improved prediction of postulated 'Green-house' effects
6.2	Glaciology	Ice cores dated and provide an archival record of global temperatures and the levels of 'green-house' gases		Stable isotopes D/H ^{18}O /^{16}O (temperature and dating) and ^{14}C techniques correlated with the analyses of gases from ice cores	Accelerator Mass Spectrometry (AMS) essential as small samples normally only available Information on past climate also from sediment cores
7.1	*Nuclear waste disposal* Natural analogues of waste repositories	Validation of performance assessment models over long time scales	Possible impact of the leakage of radionuclides on future generations	Uranium ore bodies as analogues of spent reactor fuel in waste repositories	Uranium series nuclides, e.g. ^{234}U/^{238}U, ^{230}Th/ ^{234}U, ^{226}Ra/^{230}Th isotope ratios in host rock and groundwater near a deposit used to validate aspects of radionuclide transport models

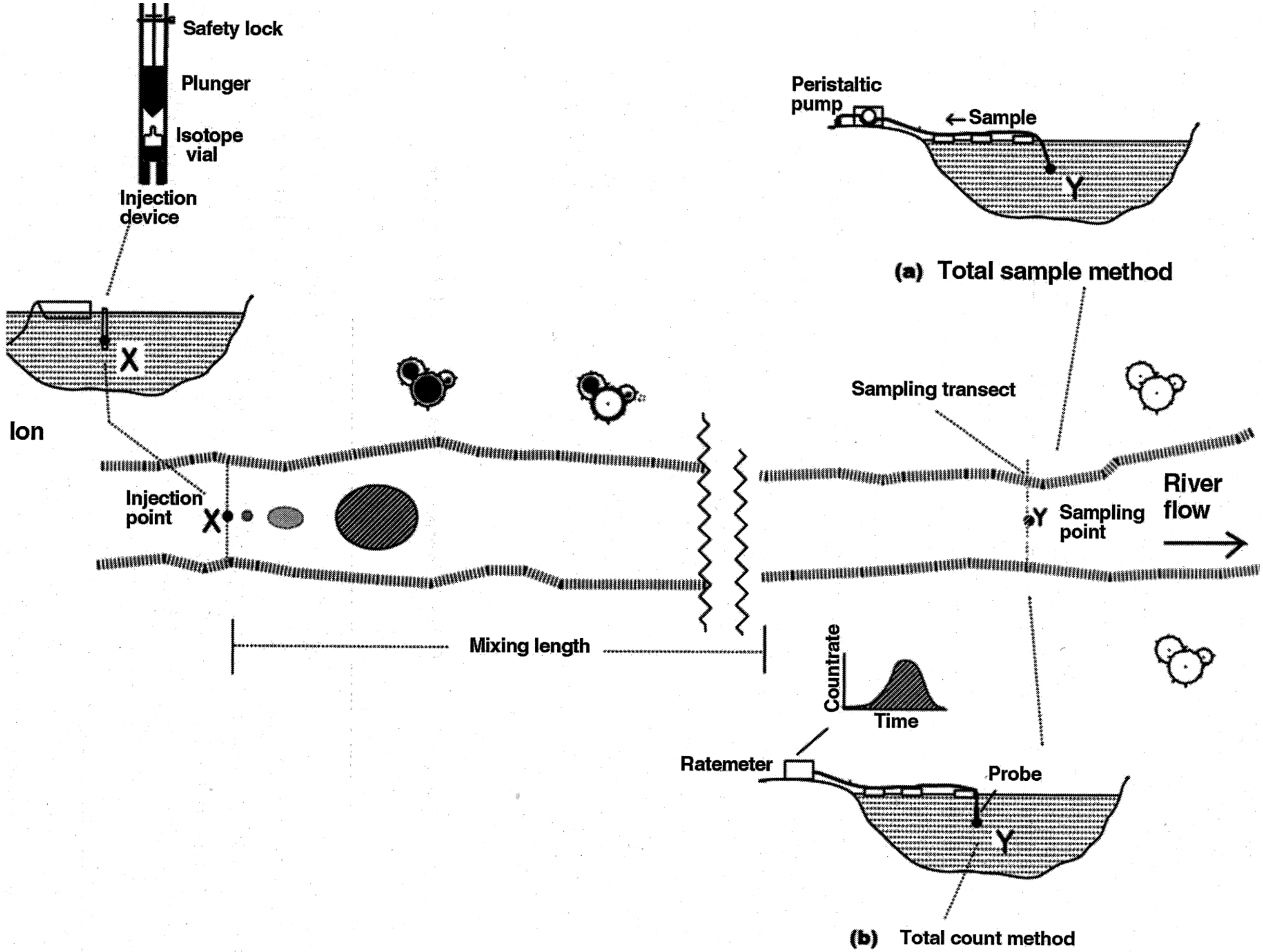
Man-made versus Environmental Radioisotopes

- Applications of man-made and environmental radioisotopes are generally complementary:
  - **Man-made radionuclides** are used to label a particular component of a complex system and are injected at a well-chosen location and according to a precise protocol. Thus very detailed information is obtained on the behaviour of an environmental system.
  - **Environmental radioisotopes** are widely distributed by natural processes and are used to obtain information over a regional scale. The data reflect the cumulative effect of environmental processes (e.g., erosion, siltation, groundwater movement).
- Naturally occurring radionuclides (or environmental radionuclides) may be classified into two [three] subgroups according to their source:
  - **Cosmogenic radionuclides** are generated in the upper troposphere (0-10km) and lower stratosphere (10-30km) by the impact of cosmic radiation.
  - **Primordial isotopes** have been formed before the earth came into existence and have been decaying since then.
  - [**Fall-out products** resulting from past nuclear testing in the atmosphere.]

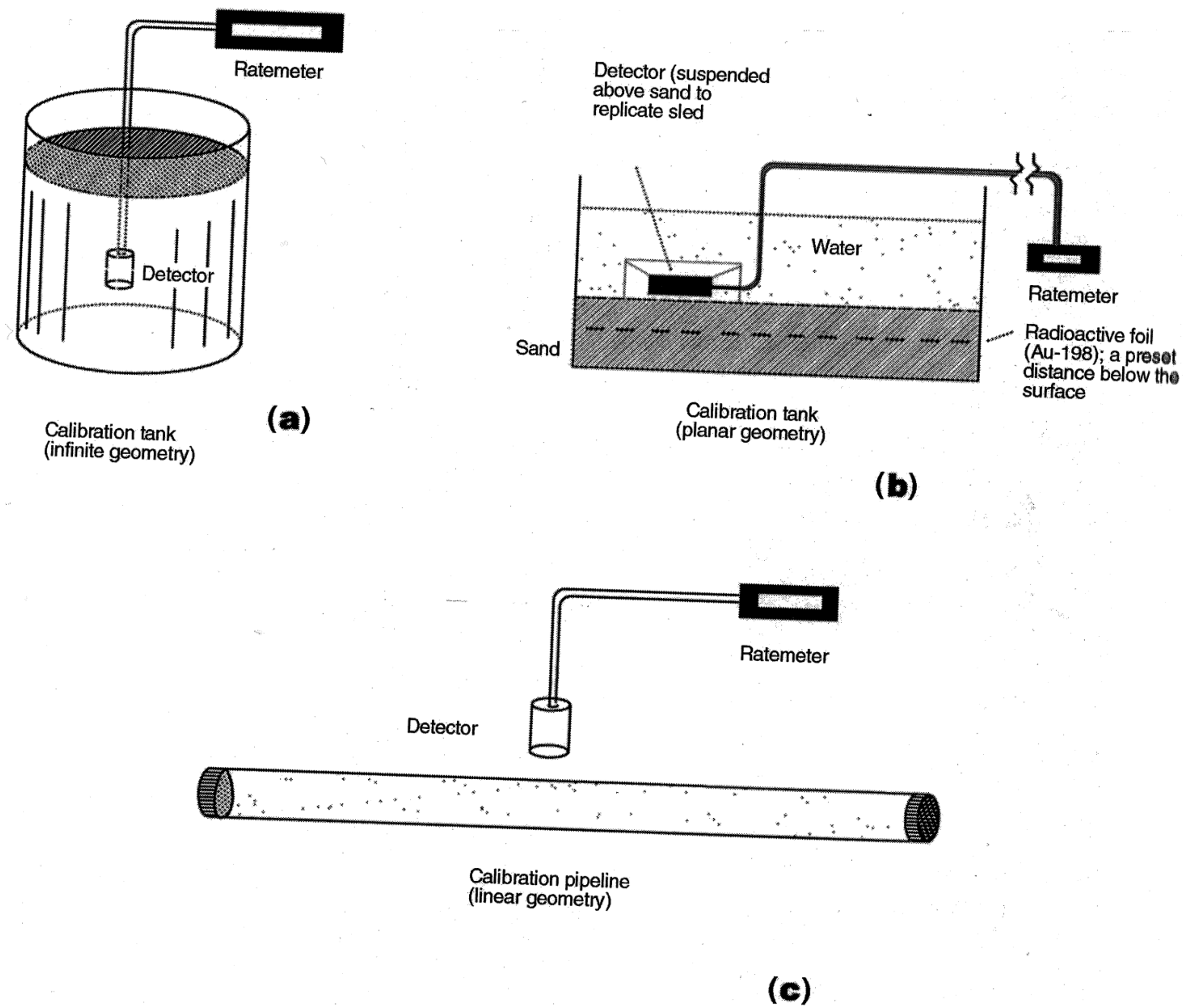
River Flow Measurements (1): Techniques

- The methods that have been applied extensively to the gauging of rivers and streams are the **total sample** and the **total count method**. Both have been discussed in the lecture of week 5a (flow rate measurements using radiotracers).
- In both methods the **isotope is injected as an instantaneous pulse** and the **probe is taken after complete mixing has been achieved** (⇔results are independent of the specific location of the sampling point or detector; concept of the mixing length).
- Total sample method:
  - Tritium as HTO is the perfect tracer for water: easy to transport in sealed glass vials, requires minimum shielding.
  - Bottle-breaking devices for remote handling have been designed to avoid inhalation of T.
  - The flow rate Q [liters/s] of the river is given as **Q= qA/a** (A=activity of tracer, q=sampling rate [liter/s], a=tracer activity in the sample, cp. "week 5a" and corresponding exercises).
- Total count method:
  - The isotopes ^{82}Br or ^{99m}Tc are often used, γ-s can be counted in real time!
  - Flow rate **Q = AF / N** (A[Bq] = total injected activity, N = total # of counts, F [cps per Bq/l] = calibration factor relating A to N).

River Flow Measurements (2): Principle

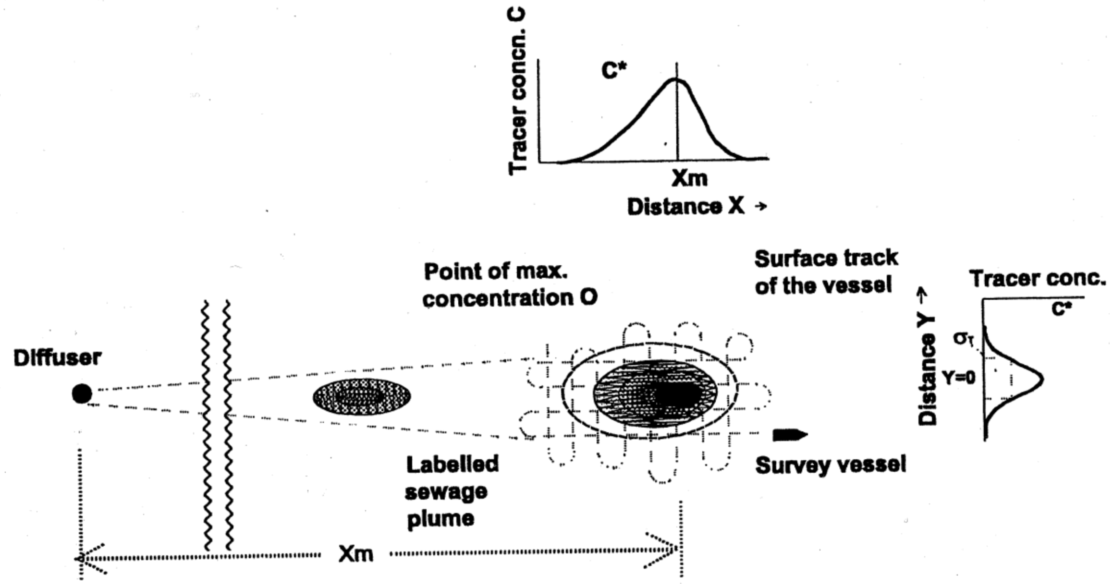


River Flow Measurements (3): Calibration

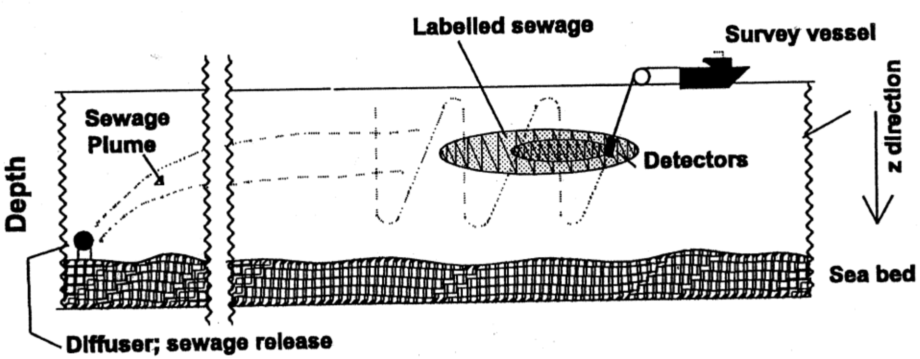


- The accuracy of river flow measurements depends on the accuracy of the calibration of the detection equipment.
- In calibration experiments the counting geometry found in the field is reproduced in the laboratory.
- On the left three limiting cases for calibration geometries are shown:
  - a) quasi-infinite geometry
  - b) quasi-planar geometry
  - c) quasi-linear geometry

Studies of Contaminan – Dispersions (1)



(a)



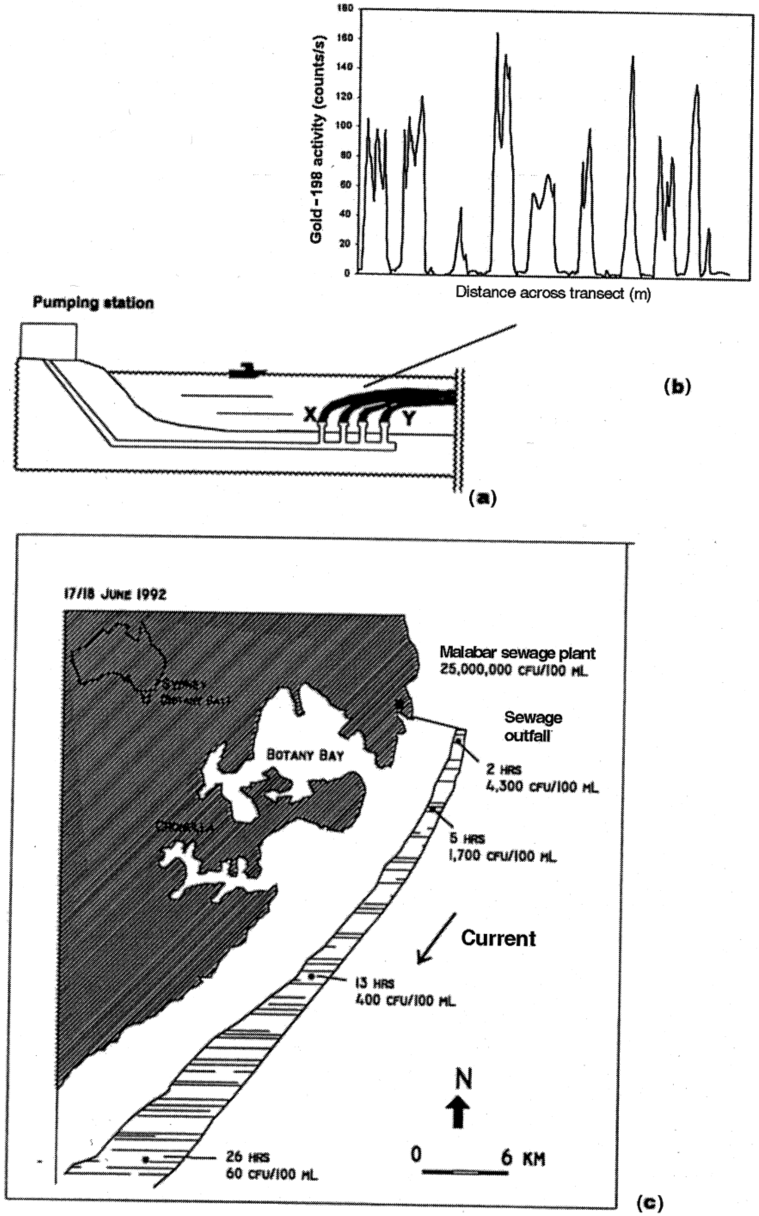
(b)

- ❑ The pollution of river systems and the coastal zone by industrial effluents and sewage is of major concern (disposal of effluents ⇔ public health).
- ❑ Radiotracer techniques can offer powerful tools for validating predictive mathematical models.
- ❑ Transport of contaminants can be studied by injecting a pulse of a tracer mimicking the pollutant and observing its subsequent dispersion.
- ❑ There are two components to the transport of the tracer:
  - **advective transport** due to the bulk movement of the water
  - **dispersion and dilution** due to the turbulent structure of the water
- ❑ The efficiency of dispersion is often quantified in terms of dispersion coefficients, which can directly be estimated by tracer techniques.

Studies of Contaminant-Dispersions (2): Analytical Treatment

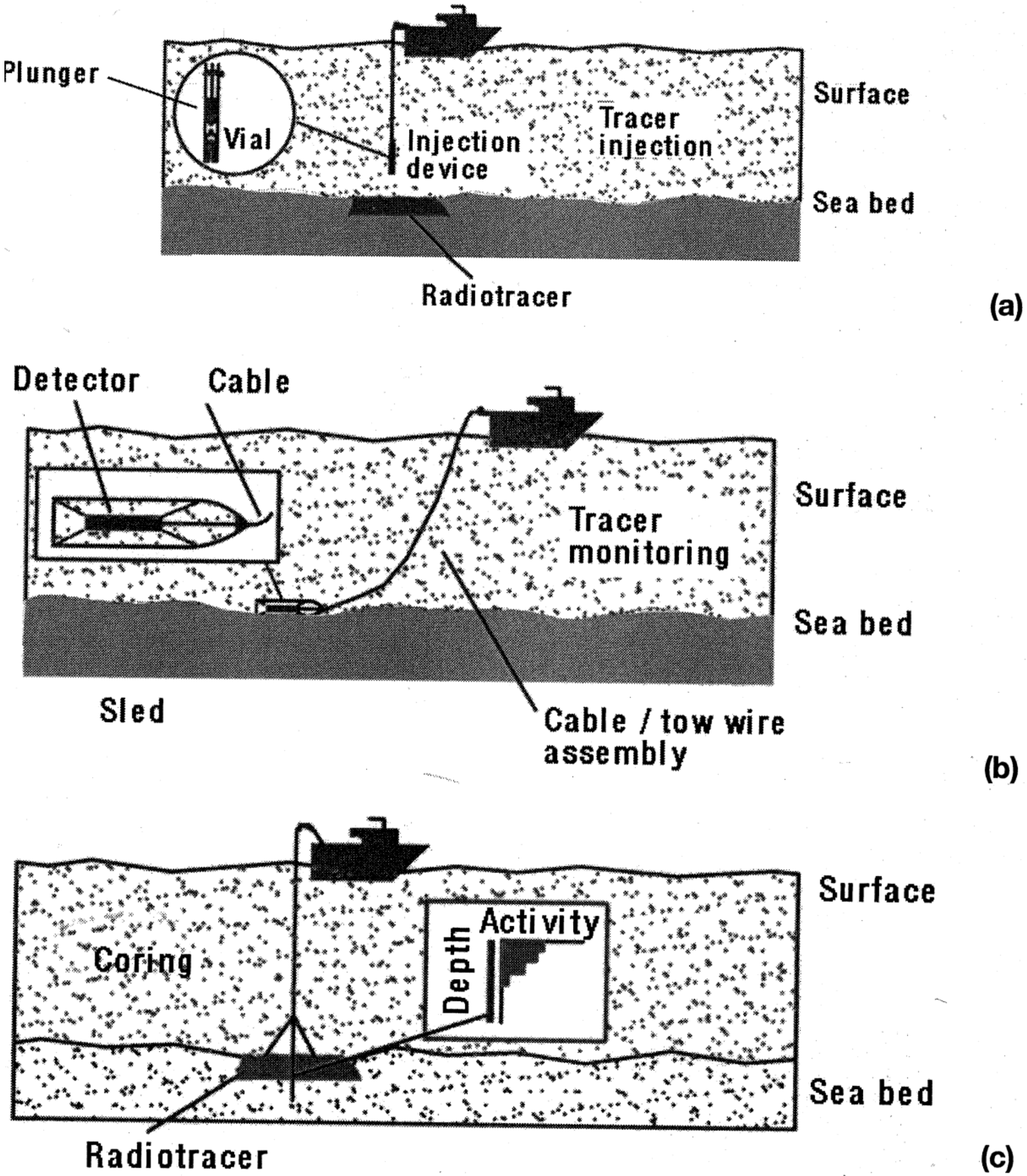
- Neglecting the effects of boundaries, the concentration profile of a tracer following a pulse injection resembles a Gaussian distribution:
  - The profile in the transverse (or y) direction may be expressed as: **$C(y,t)=C_y{}^* \exp[-(y-y_m)^2 / 4D_T t]$** ($D_T$=transverse dispersion coefficient, variance $\sigma^2=2D_T t$)
  - The profile in the longitudinal (or x) direction may be expressed as: **$C(x,t)=C_x{}^* \exp[-(x-u_L t)^2 / 4D_L t]$** ($u_L$=average flow rate, D_L=longitudinal dispersion coefficient)
- **In many investigations advection and diffusion may be considered separately,** because at any point within the bulk flow:
  - the instantaneous velocity u of a fluid particle may be written as: u=<u>+u'
  - the instantaneous concentration c of a contaminant may be written as: c=<c>+c'
  - the instantaneous rate of transport of the contaminant through a cross section at a particular point in time is the product uc
  - hence the mass flux Φ through a unit cross section averaged over a longer time interval is the average value of this product **Φ=<uc>=<u><c>+ <u'c'>**,
  - i.e., the flux is the sum of an **advection term <u><c>** and a **diffusion term <u'c'>**.

Case Study: Sewage Dispersion



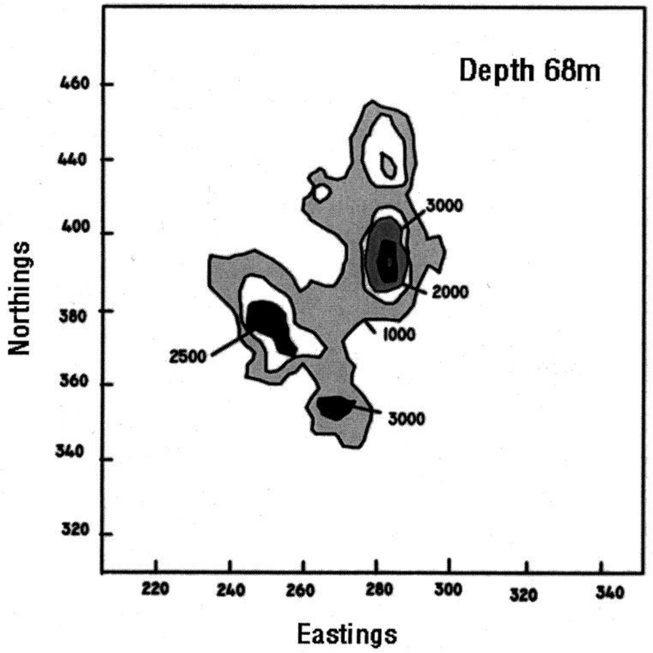
- Many major cities around the world are located on the coastal zone, and their sewage is frequently disposed of through deep ocean outfalls following pre-treatment.
  a) The effluent is pumped a substantial distance offshore and released at intervals through a series of diffuser heads.
  b) By the response of the radiation detectors tracking close to the diffusers the performance of individual diffusers can be distinguished. As radiotracers ^{198}Au and HTO are commonly used.
  c) Results of a radiotracer study of the dispersion of sewage from a deep ocean outfall near Sydney, Australia. Observations were made for 26 hours. Levels of bacteria in the sea water are shown as the empirical measure colony forming units (CFU).
- Treated sewage is a complex mixture of dissolved aqueous contaminants, suspended particulates and dispersed greases. With radionuclides these different components can be tagged independently and studied separately.

Sediment & Sand Tracing (1)

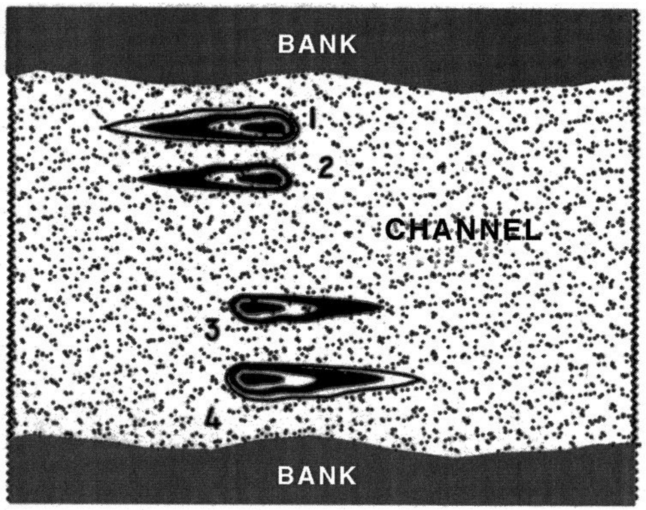


- Offshore migration of sand and sediment is often important for port and harbor development.
- Radioisotopes applied in a study are chosen according to the runtime of the investigation, e.g.:
  - ^{198}Au ($T_{½}$=2.7d)
  - ^{51}Cr ($T_{½}$=27.7d)
  - ^{192}Ir ($T_{½}$=73.8d)
  - ^{46}Sc ($T_{½}$=83.8d)
  - ^{110m}Ag ($T_{½}$=250d)
- In order to label the sand or sediment:
  - the radiotracer is adsorbed onto the surface of the sand,
  - or incorporated into a glass, then ground and sieved to match the particle size distribution of the target material.
- Illustration (on the left) of tracer-application-techniques to sediment migration:
  a) Injection of a tracer using the bottle breaker technique.
  b) Tracking of the labelled sediment (may last from days to many months) using calibrated detectors mounted on a sled.
  c) The depth distribution of the tracer is obtained by coring.

Sediment & Sand Tracing (2): Case Studies



(d)



(e)

d) Long-term investigation of sand migration at a depth of 68 m with the aim to examine the effect of storms on the movement of sand at depth:

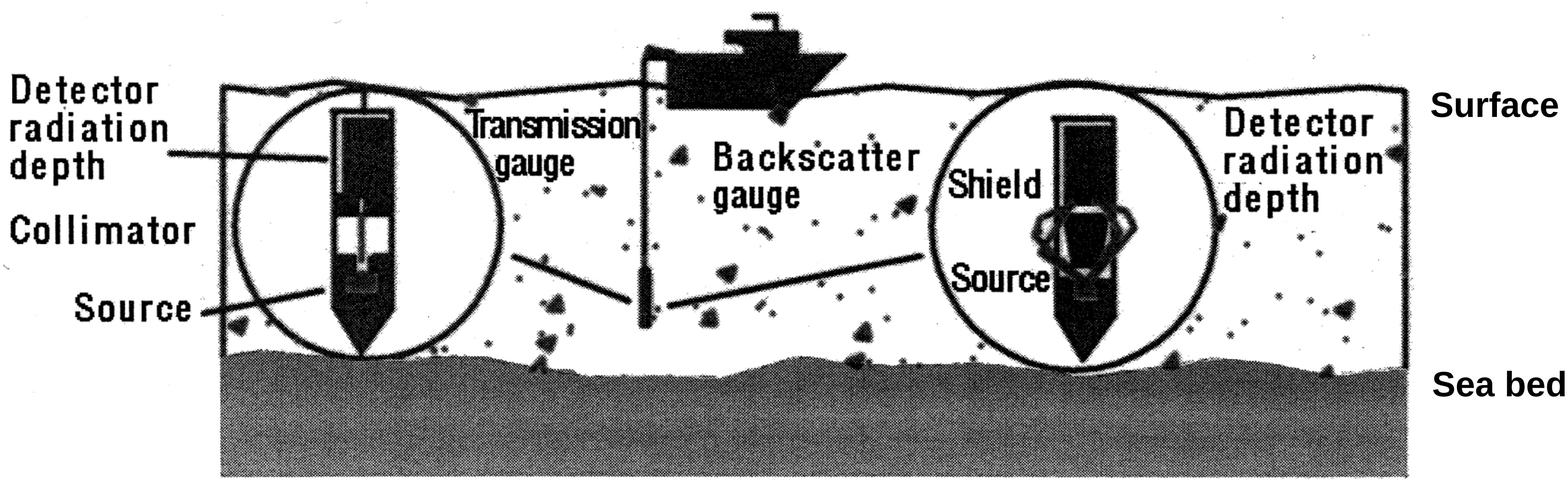
- Glass beads with the same particle size distribution as the sand were labelled with ^{192}Ir.
- The tracer was released at three sites by detonating the vials.
- Result: Despite some significant storm activity only little movement of the sand was observed over a period of 3 months.

e) Results of a typical bed load study (coastal engineering) to investigate the dynamics of sand transport in a channel connecting a coastal lake system to the sea:

- Radiotracer was injected at four locations across the channel.
- Opposite migration directions were found in the northern and southern part of the channel due to the current patterns during ebb and flood tides.

Sediment & Sand Tracing (3): Nucleonic Sediment Gauges

- Nucleonic gauges have been developed for accurate measurements in real time of the levels of suspended sediments in rivers and estuaries with the goal to understand the stability of dredge spoil grounds and the impact on estuaries of sediments transported by rivers to the coastal zone.
- Suspended sediment gauges are designed as (cp. lecture week 5a, “gauges”):
  - gamma ray transmission gauges, sensitive to the small volume between source and detector
  - gamma ray backscatter gauges, sensitive to a much larger volume, but prone to effects from boundaries
- Both devices can measure sediment loadings equivalent to bulk density changes of less than 1%.

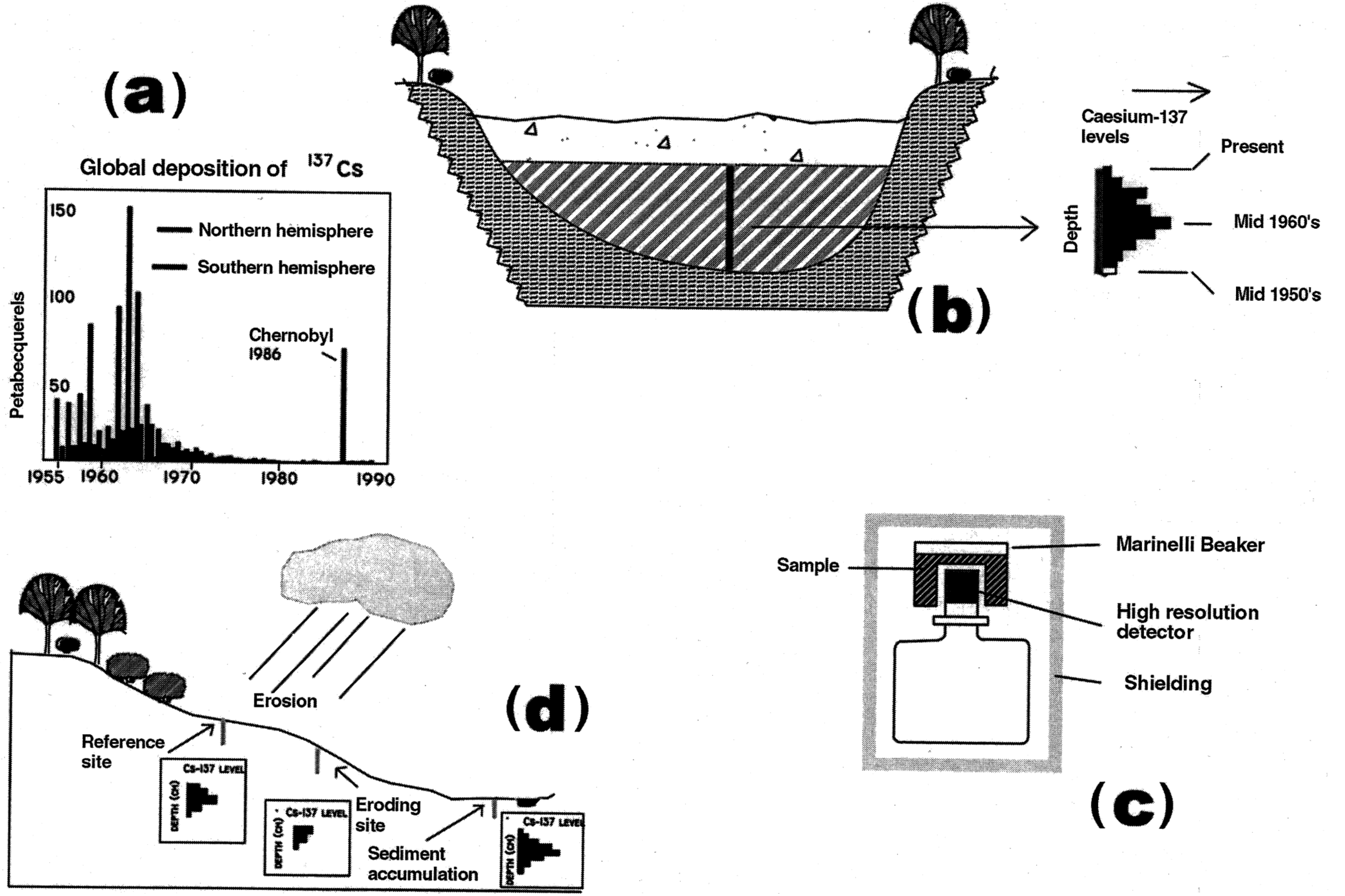


Nucleonic sediment gauges

Erosion Studies (1)

- The measurement, and in particular the prediction, of the rate of erosion of fertile soils is a key to the development of strategies for the long-term sustainability of agriculture.
- By use of the environmental isotope ^{137}Cs ($T_{1/2}$=30.1 y) erosion rates over a whole catchment area may be estimated.
- ^{137}Cs was injected into the upper troposphere and lower stratosphere during atmospheric nuclear tests (~1950−1963). Together with other fall-out products it was distributed world-wide by global atmospheric circulation and finally diffused to the surface of the earth. As ^{137}Cs is strongly adsorbed onto the clay fraction of the soils, it acts as a natural radiotracer for erosion and accumulation processes.
- Experimentally the **^{137}Cs profile** is measured by:
  - collecting and sectioning a core obtained from the soil in the region of interest
  - placing each dried and weighted section of the core in a Marinelli beaker over a high-resolution detector
  - measuring the count rate in the 662keV gamma ray peak
  - calculating the specific activity of the ^{137}Cs by comparison with results obtained from a calibrated standard
- For instance: a study of a vineyard revealed that for each bottle of wine produced, approximately one bottle of fertile soil was lost to the cultivated area.

Erosion Studies (2): Application of ^{137}Cs



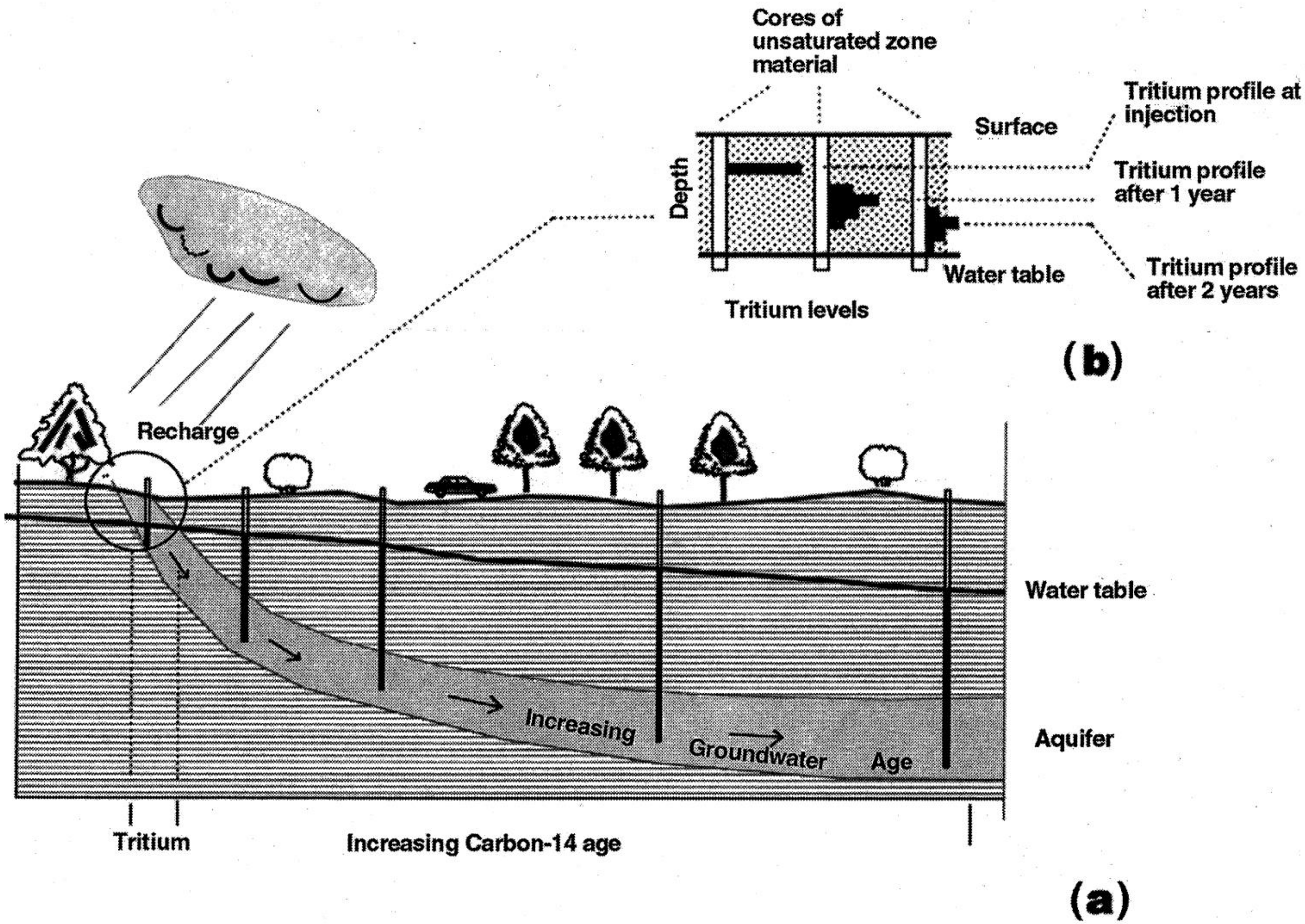
Groundwater Studies (1)

- In addition to adequate resources of fertile soil also **clean drinking water is essential for public health**.
- Environmental radioisotopes are used to investigate underground water resources. Applications have been directed towards:
  - identifying the sources of recharge water, i.e., the sources of surface water which seep underground to replenish the basin,
  - estimating the extent of mixing of underground water from different sources,
  - calculating the age of groundwater samples , i.e., the time which has elapsed since the water percolated underground,
  - deducing the direction and rate of groundwater flow,
  - understanding the processes leading to degradation in groundwater quality.
- The **data** obtained in groundwater studies are often **used to validate groundwater hydraulic models** which link the known water levels to the properties of the water bearing strata.
- For the **dating of underground** water tritium ($T_{½}$=12.3y) , ^{14}C ($T_{½}$=5730y, carbon is a solute in groundwater) and ^{36}Cl ($T_{½}=3 \cdot 10^5$y) may be used.

Groundwater Studies (2): Two Examples

a) Schematic diagram showing underground water percolating through an aquifer and sampled at various locations for age dating with (e.g.) ^{14}C. By such studies recharge areas can be identified and well managed.

b) Use of tritium to study the rate of recharge of infiltrating rainwater to an aquifer. Tritium is injected at a constant depth and cores are collected at a suitable mesh of given times.

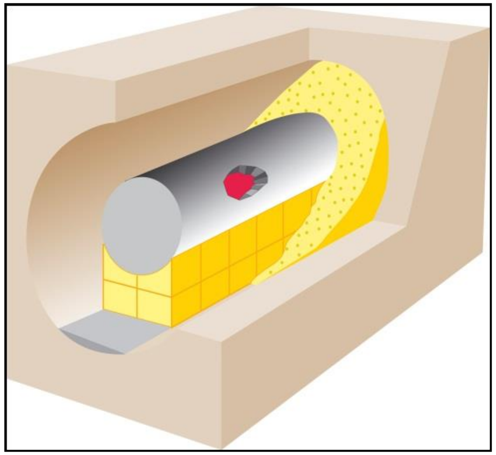


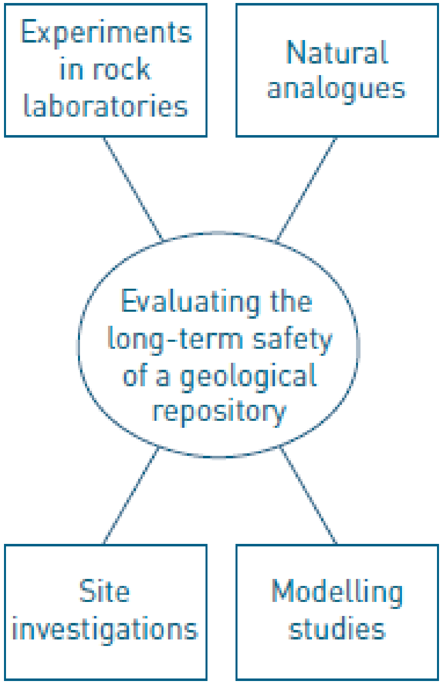
Oceanography

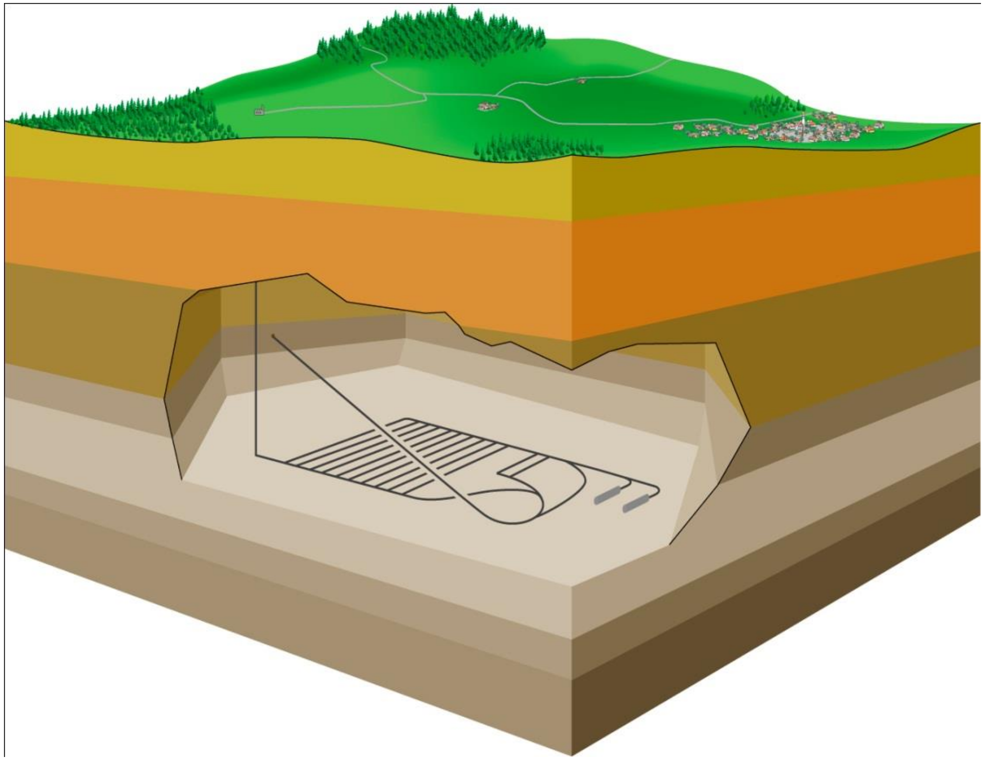
- Environmental isotope techniques are well suited to investigate the dynamics of large and complex systems and have contributed significantly to the understanding of many characteristics of the oceans. Two classes of studies may be distinguished by their targets:
  - Processes within the coastal zone due to the impact of river systems leading to the release of contaminants derived from agriculture, industrial development or urbanization.
  - Circulation patterns within the deep oceans and their impact on weather and climate change and on fisheries.
- There are two principal groups of radionuclides in the oceans:
  - Tritium and ^{14}C which, respectively, enter the ocean in rainfall and by exchange with the $^{14}CO_2$ of the atmosphere. Both exhibit a 'bomb pulse'.
  - Thorium and Uranium and their daughters which enter the coastal zone dissolved in river water or associated with river sediments.

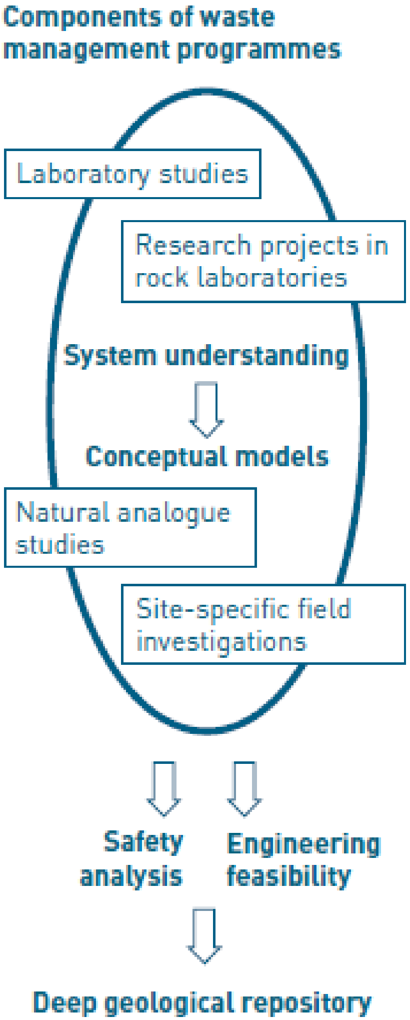
Nuclear Waste Disposal (1)

- Nuclear power plants produce high and low level radioactive waste (actinides, fission products, activation products). The latter are also generated in research, industry and medicine.
- Radioactive waste has to be isolated from the biosphere for ten thousands of years.
- **The solution favored worldwide is deep geological disposal**.

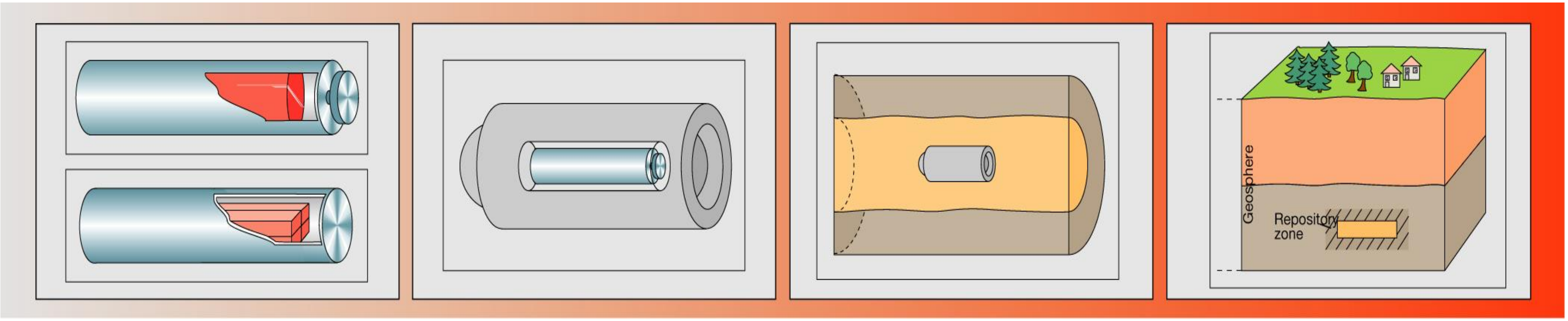








Nuclear Waste Disposal (2): Safety Barrier System



Glass matrix (in steel mould)

- *Low corrosion rate of glass*
- *High resistance to radiation damage*
- *Homogeneous radionuclide distribution*

or
Spent fuel elements

- *Low UO2 dissolution rate*
- *High radiological / thermal stability of UO2-matrix*

Steel canister

- *Completely isolates waste for > 1000 years*
- *Corrosion products act as a chemical buffer*
- *Corrosion products take up radionuclides*

Bentonite backfill

- *Long resaturation time*
- *Low solute transfer rates (diffusion)*
- *Retardation of radionuclide transport (sorption)*
- *Chemical buffer*
- *Low radionuclide solubility in leachate*
- *Colloid filter*
- *Plasticity (self-healing following physical disturbance)*

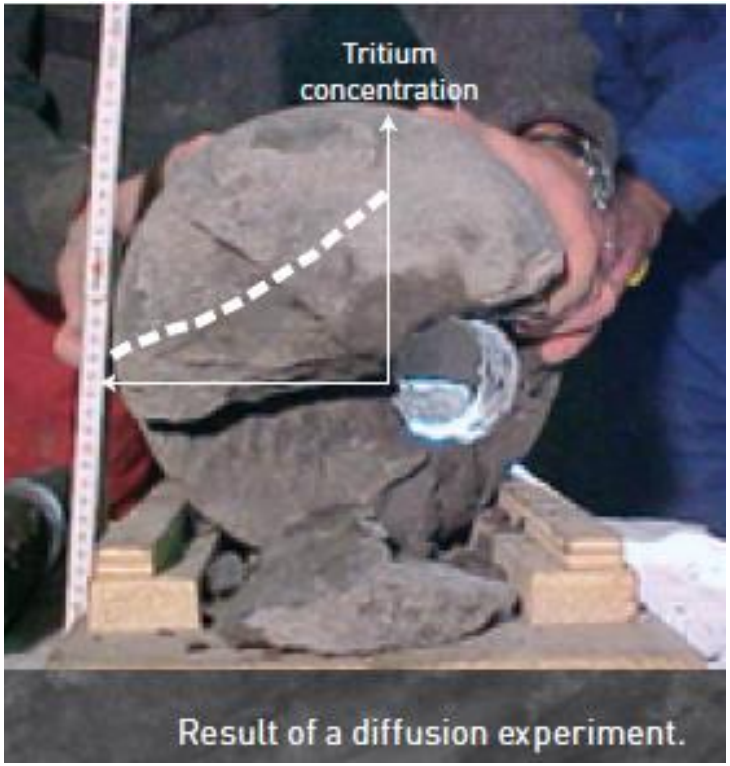
Geological barriers Repository zone:

- *Low water flux*
- *Favourable geochemistry*
- *Mechanical stability*

Geosphere:

- *Retardation of radionuclides (sorption, matrix diffusion)*
- *Reduction of radionuclide concentration (dilution, radio-active decay)*
- *Physical protection of the engineered barriers (e.g. from glacial erosion)*

Nuclear Waste Disposal (3): Examples



- The geosphere around a repository is an important component of the multi-barrier-system.
- Long-term predictions of the radionuclides retardation in the geosphere are based on numerical models, which can be validated with radiotracer methods.
- In the Mont Terri Rock Laboratory Nagra performs research on safe geological disposal. E.g., in the DI-A experiment the diffusion of radionuclides in Opalinus Clay is investigated. An isolated test interval in a small borehole is saturated with water and a defined volume of radiotracer is then added. After more than one year, the small borehole is over-cored and the new, larger-diameter drill core is used to investigate how far the tracer penetrated into the rock (photograph on the left). Non-retarded radionuclides such as T are seen to mitigate faster than strongly retarded nuclides such as Cs, Co, which will be present in a repository for high-level waste.

- An approach to the difficult problem of validating transport codes for long-term predictions is the use of **natural analogues**. A natural analogue is a feature in the environment exhibiting processes which are similar in essential respects to those being modeled.
  - The Oklo uranium deposit in the country of Gabon in Central Africa contains several sites where chain reactions in uranium occurred naturally and repeatedly. Fission products, Pu and other transuranic elements were produced, and from an analysis of their distribution within the host rock, it could be concluded that they were bound over very long periods of time. Similar studies have been performed for other uranium deposits.
  - There are also natural analogues for the materials of the engineered barriers (boro-silicate glass, iron, copper).

Literature / WWW-references

- G.C. Lowenthal, P.L. Airey, *"Practical Applications of Radioactivity and Nuclear Reactions"*, Cambridge University Press (2001) Chapter 9
- K.H. Lieser, *"Nuclear and Radiochemistry"*, WILEY-VCH (2^{nd} edition, 2001) Chapters 21, 19
- Home page of the "National Cooperative for the Disposal of Radioactive Waste" (Nagra): http://www.nagra.ch
- Home page of the *"Laboratory for Radiochemistry and Environmental Chemistry"* at PSI: http://lch.web.psi.ch

Concluding Remarks:

Greek bust of Janus,
Vatican Museums.

- Applications of radiation and radioisotopes in medicine, industry and research are numerous, manifold and (sometimes) complex.
- Each application has advantages and disadvantages in comparison to other methods.
- The goal in applying radiation and radioisotopes methodologies/techniques must be to **minimize the harmful effects** and to **maximize the benefits** of its use.
- Especially: One should always be aware of the **consequences** for the patient/object/system under treatment/monitoring/investigation **if radiation and radioisotopes would NOT be applied !**