Models of tritium behaviour in hydrological systems

HydroPredict'2012
Predictions for hydrology, ecology and WRM

Vienna 24-27 September 2012

IAEA
International Atomic Energy Agency

L. Araguás, P.K. Aggarwal,
Isotope Hydrology Section, IAEA

B. Newman,
Los Alamos National Lab., USA

Outline

- Overview: Tritium as tracer in hydrology
- Tritium monitoring in precipitation and river water
- Use of lumped parameter models in hydrogeological studies
- Age dating and components of base flow in rivers: implications/predictions for water pollution aspects
- Use of tritium/helium-3 technique for dating “young” groundwater (<100 a)

IAEA

Tracer hydrology - isotopes

- Catchment and groundwater dynamics: characterizing fast and slow flow systems as well as mixing patterns
- Tracer vs. hydrodynamic ages → validation of conceptual/numerical flow models and its use as a forecasting tool
- Qualitative and quantitative approaches with isotopes and dissolved gases

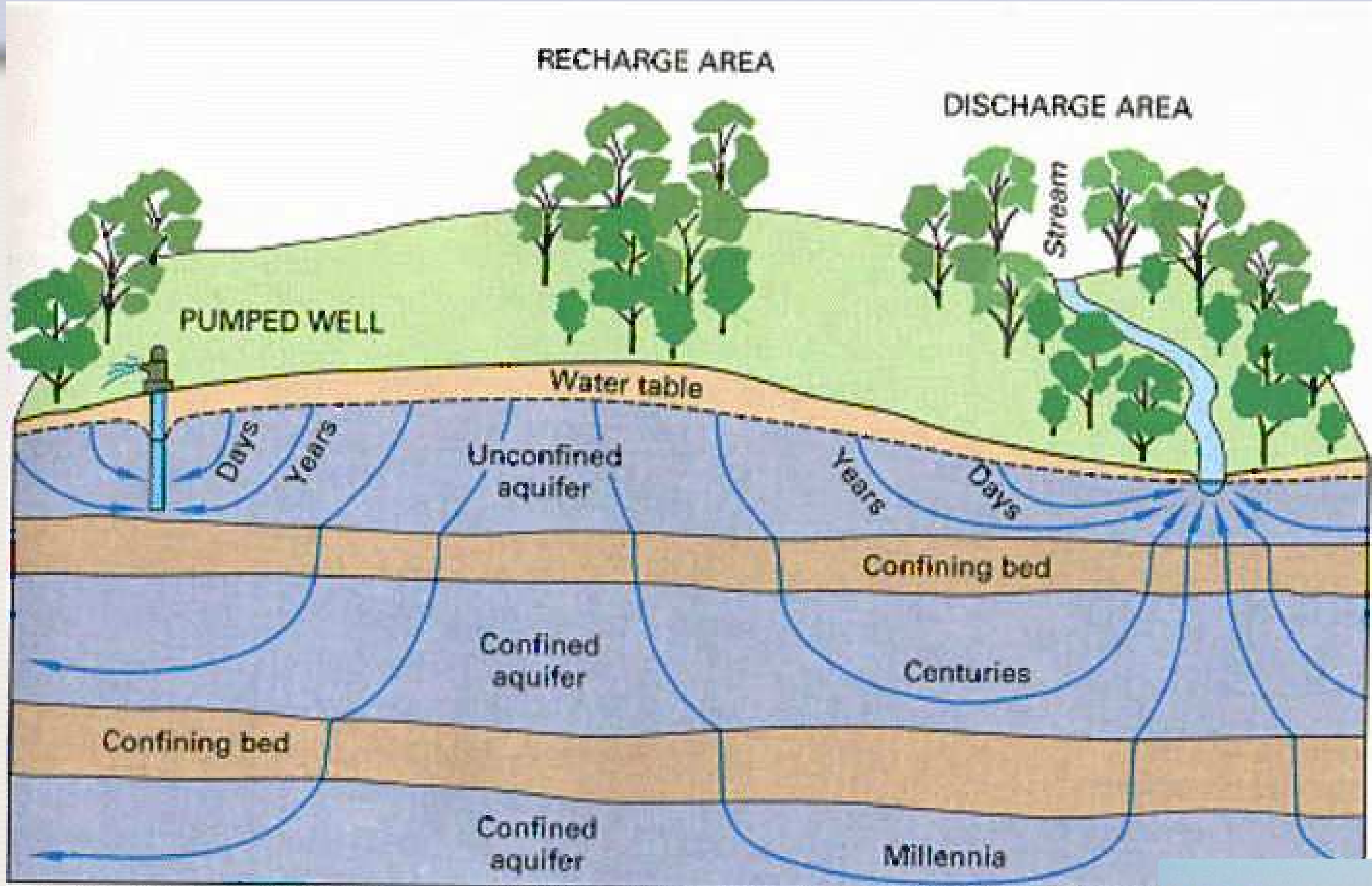
IAEA

Why date groundwater/baseflow?

- Recharge rates (relatively direct compared to water balance)
- Flow model adjustments (recharge, porosity, dispersivity, flow paths)
- Chemical fluxes (contaminants, oxidants, exchangers, etc.)
- Historical records (long-term vs transient)
- Reaction rates (*in situ*)
- Discharge ages and residence times (aquifer volume and flux, watershed mass balance)

IAEA

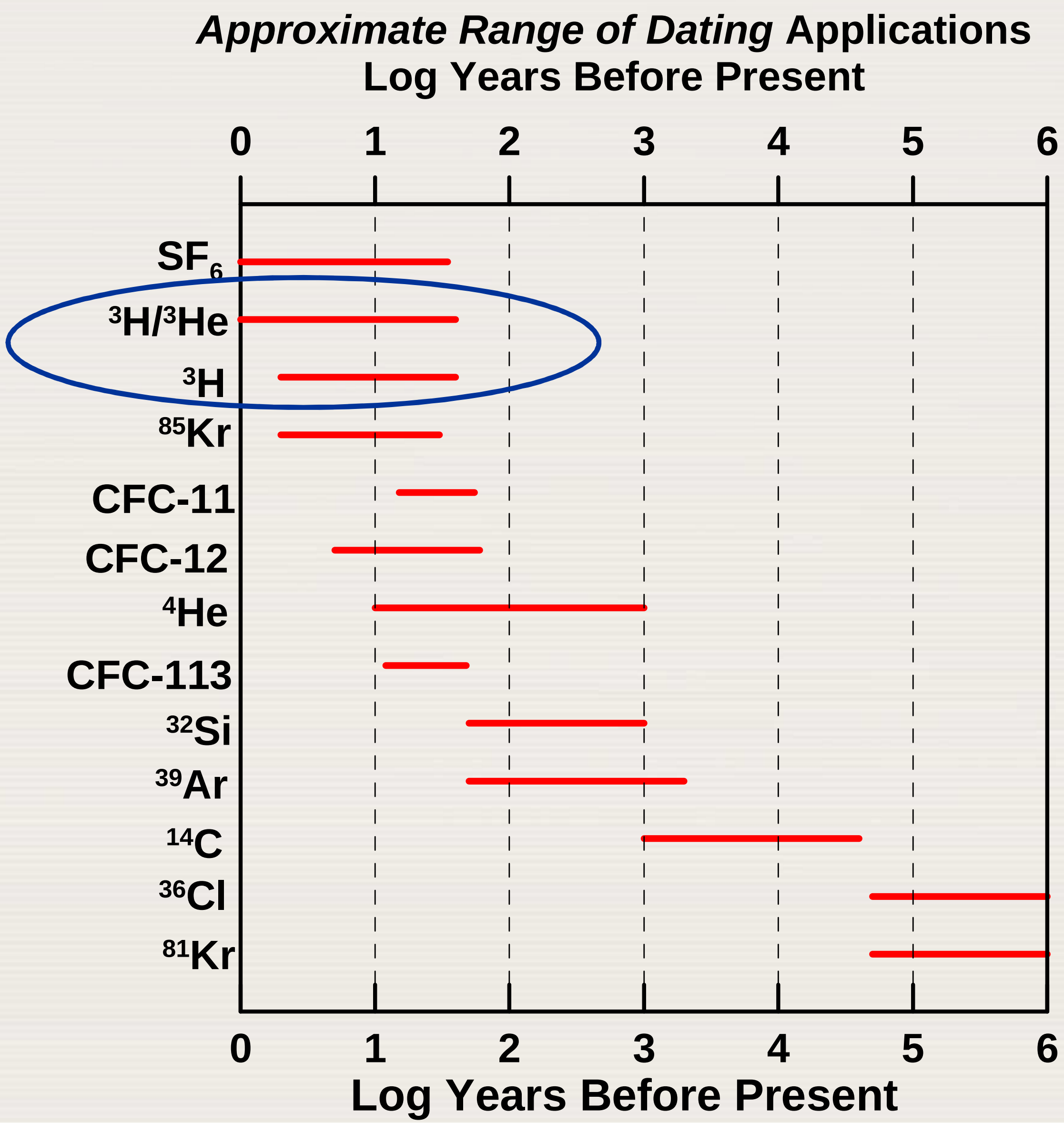
Range of groundwater ages (USGS)



What does it mean if groundwater is old?
Assess vulnerability, mixing (age distribution and MRT, impact on quality)

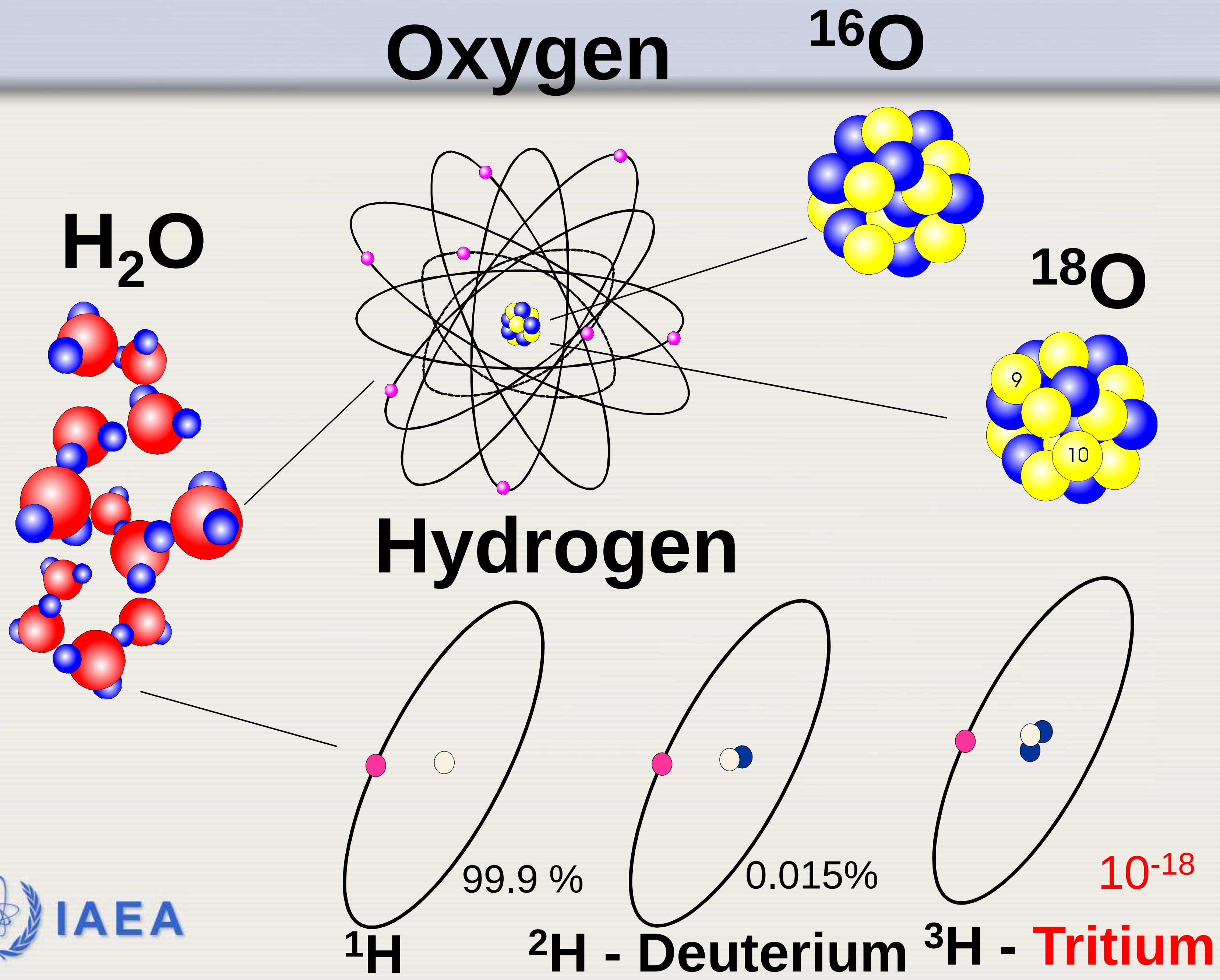
IAEA

Water dating tracers: Range of ages (1a to 1 Ma)



IAEA

USGS


Oxygen
^{16}O
^{18}O
9
10
H_2O
Hydrogen
99.9 %
0.015%
10^{-18}
^{1}H
^{2}H - Deuterium
^{3}H - Tritium

Tritium (^3H)

- **Sources:**

  - Cosmogenic:

    $$^{14}N + ^{1}n \rightarrow ^{3}H + ^{12}C$$

    Production rate: 0.28 at cm^{-2} s^{-1}

  - Anthropogenic:

    (The thermonuclear bomb testing and other uses)

Natural inventory ~ 4 kg

Bomb testing ~ 700 kg injected into the upper atmosphere

IAEA

Tritium (^3H)

- Decay: $^3H \rightarrow {}^3He + \beta^-$ (E_{max} = 18 keV)
- Half-life = 12.32 a (4500 days) → dating ~100 a
- Measurement by liquid scintillation counting after electrolytic enrichment. Content expressed in:

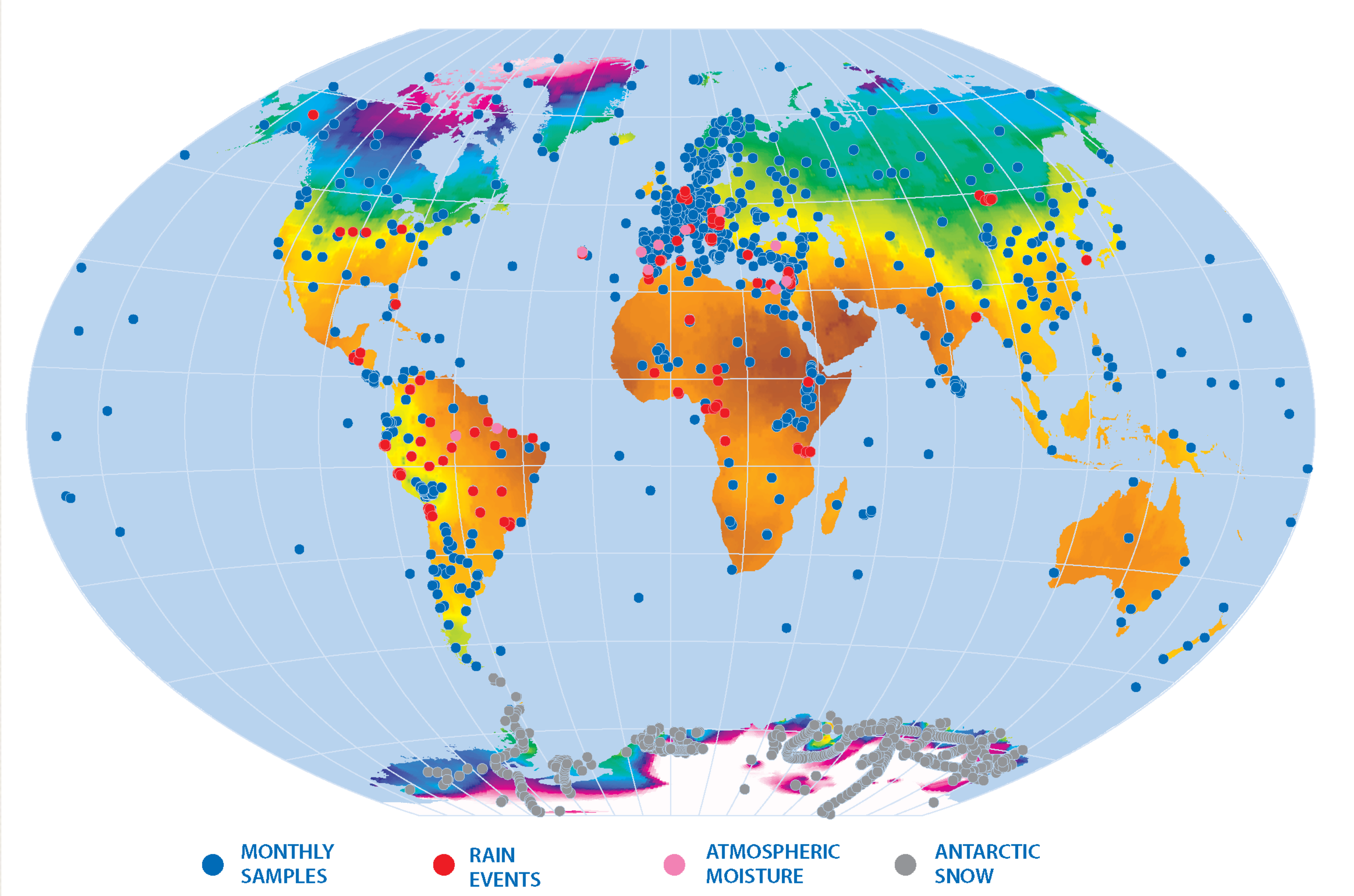
  Tritium Unit (TU) ⇒ tritium ratio

  1TU ⇒ $^3H/^1H = 10^{-18}$

  1TU = 0.119 Bq/kg or 3.2 pCi/liter
- Analytical uncertainty: 0.1 to 0.3 TU. Current levels in precipitation 1 to 15 TU.

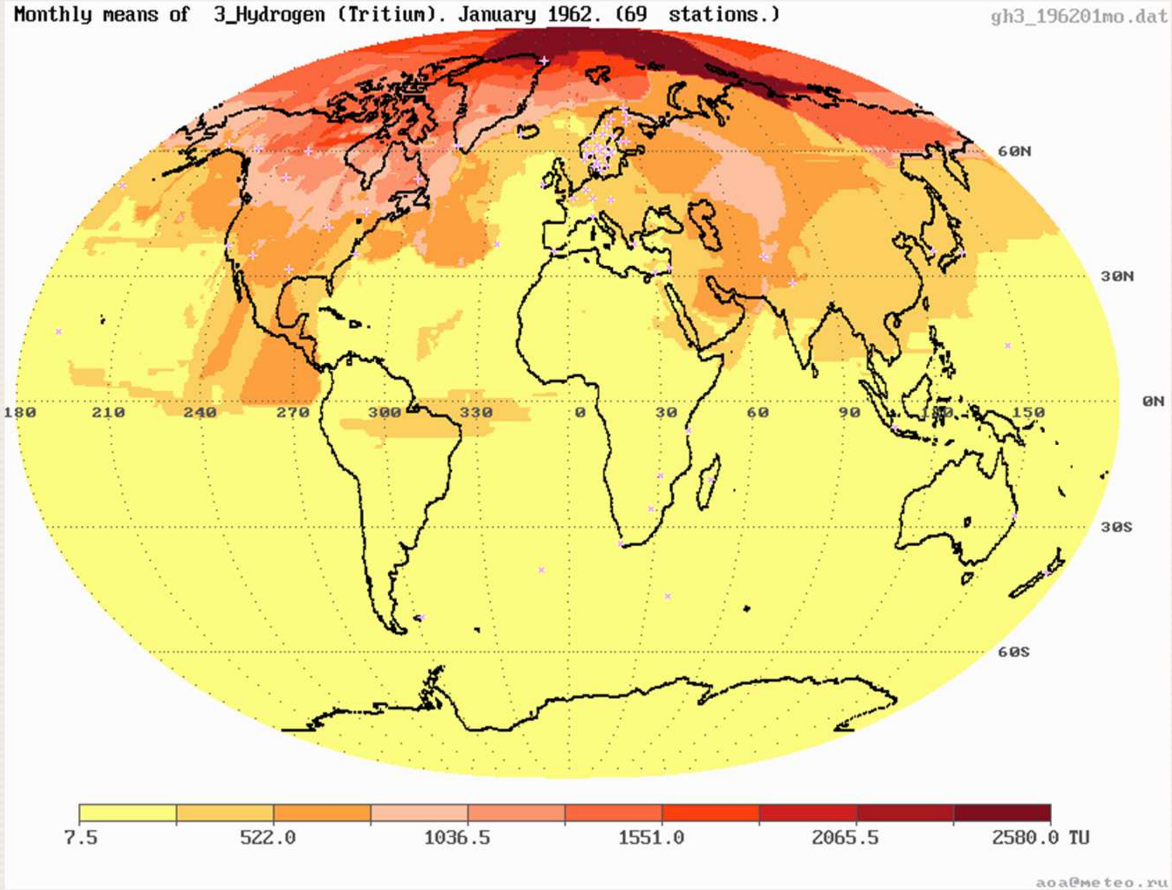
IAEA

Tritium conc in precipitation has been monitored since 1960 by the Global Network of Isotopes in Precipitation



IAEA All GNIP data available in: www.iaea.org/water

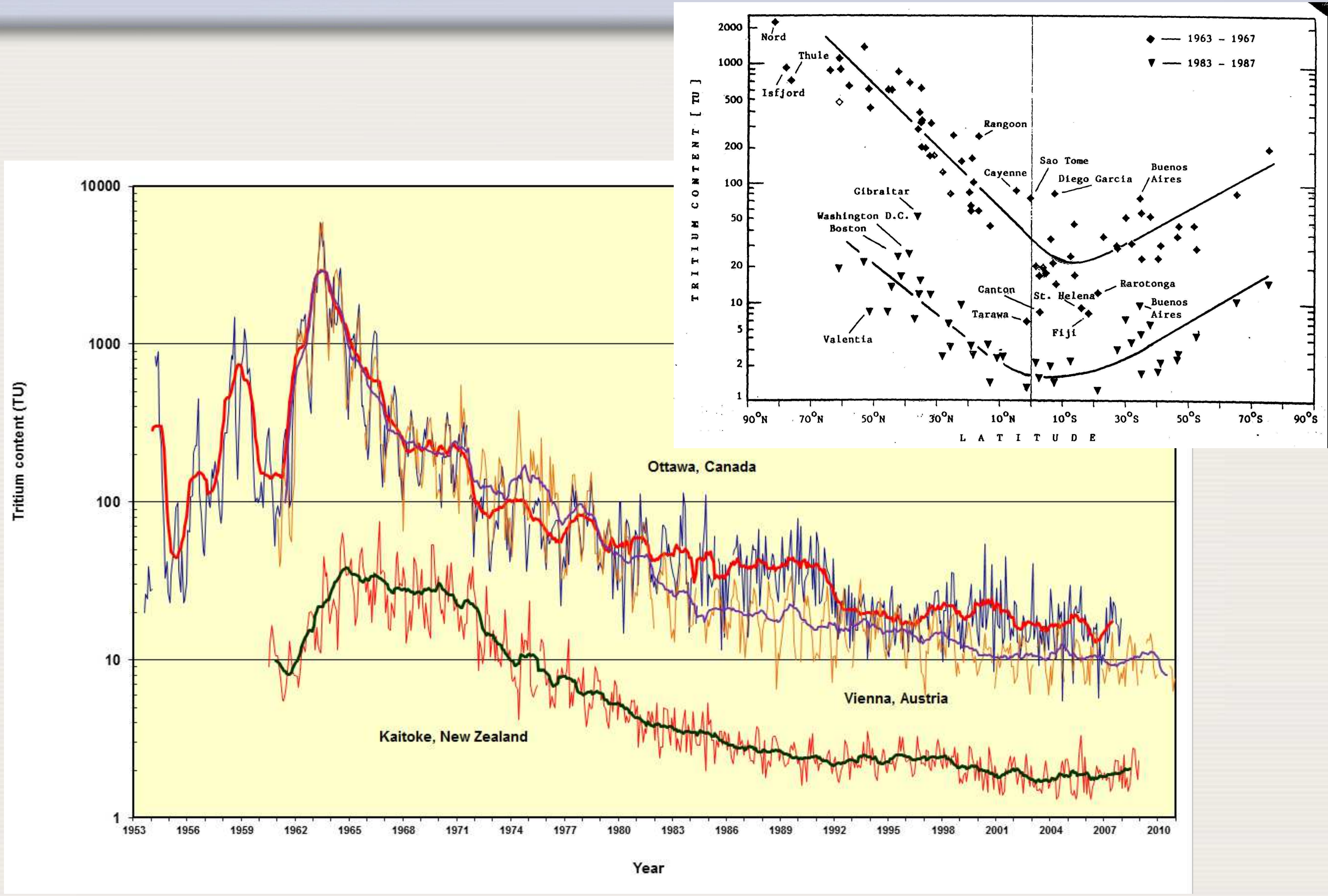
Global tritium distribution in 1960s



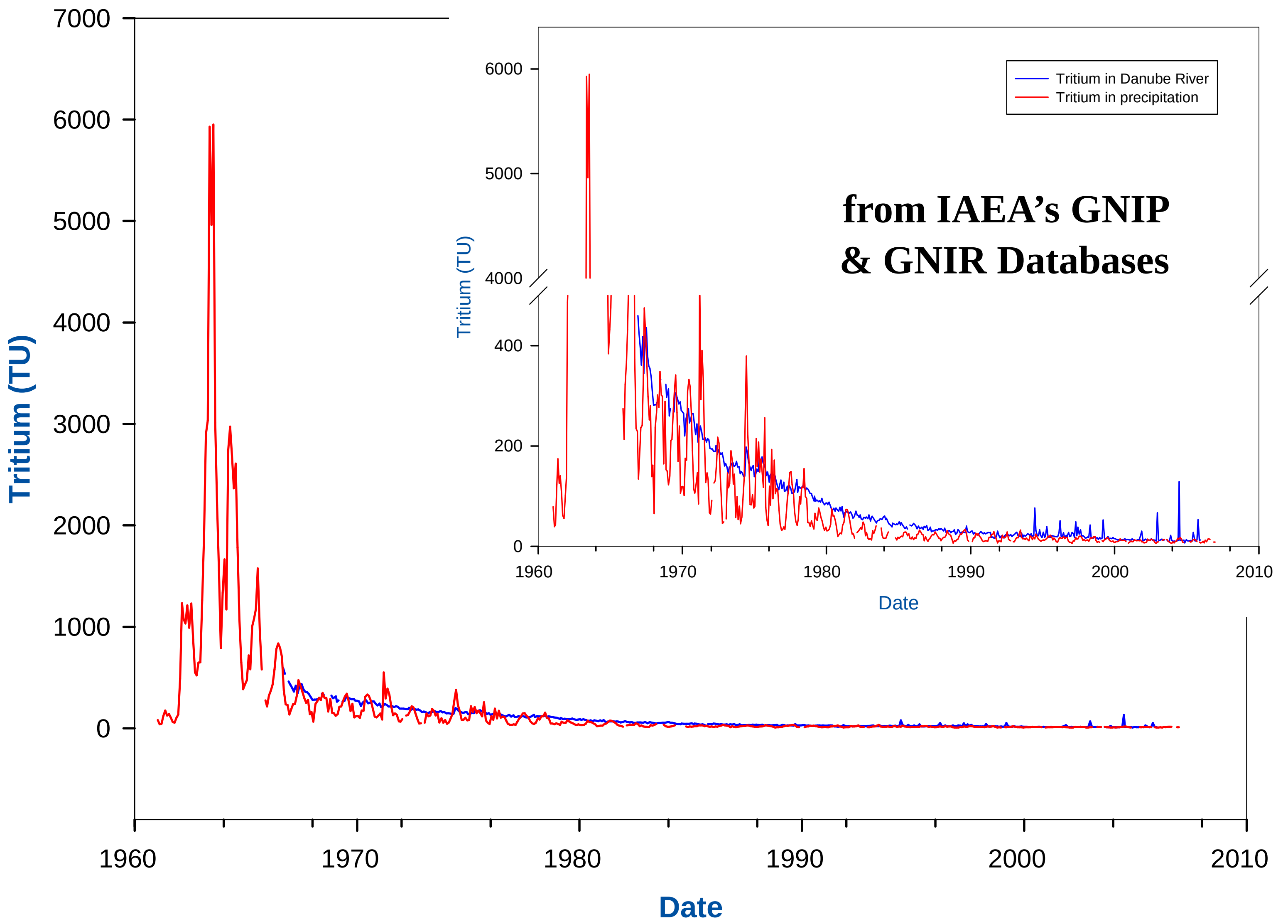
Most nuclear tests conducted in the Northern Hemisphere.
Increase tritium contents up to 3-4 orders of magnitude.

IAEA

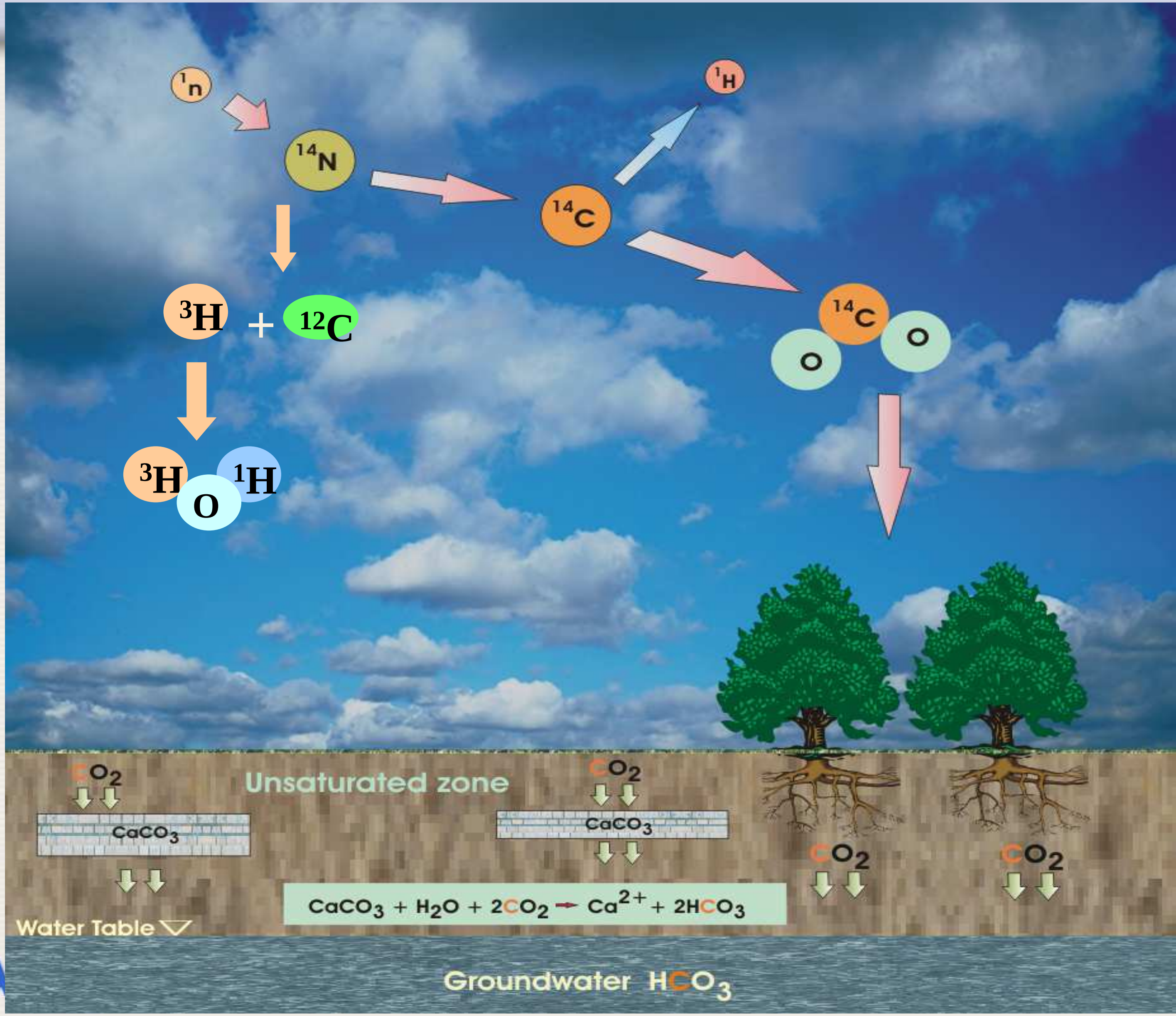
Tritium in precipitation: global picture



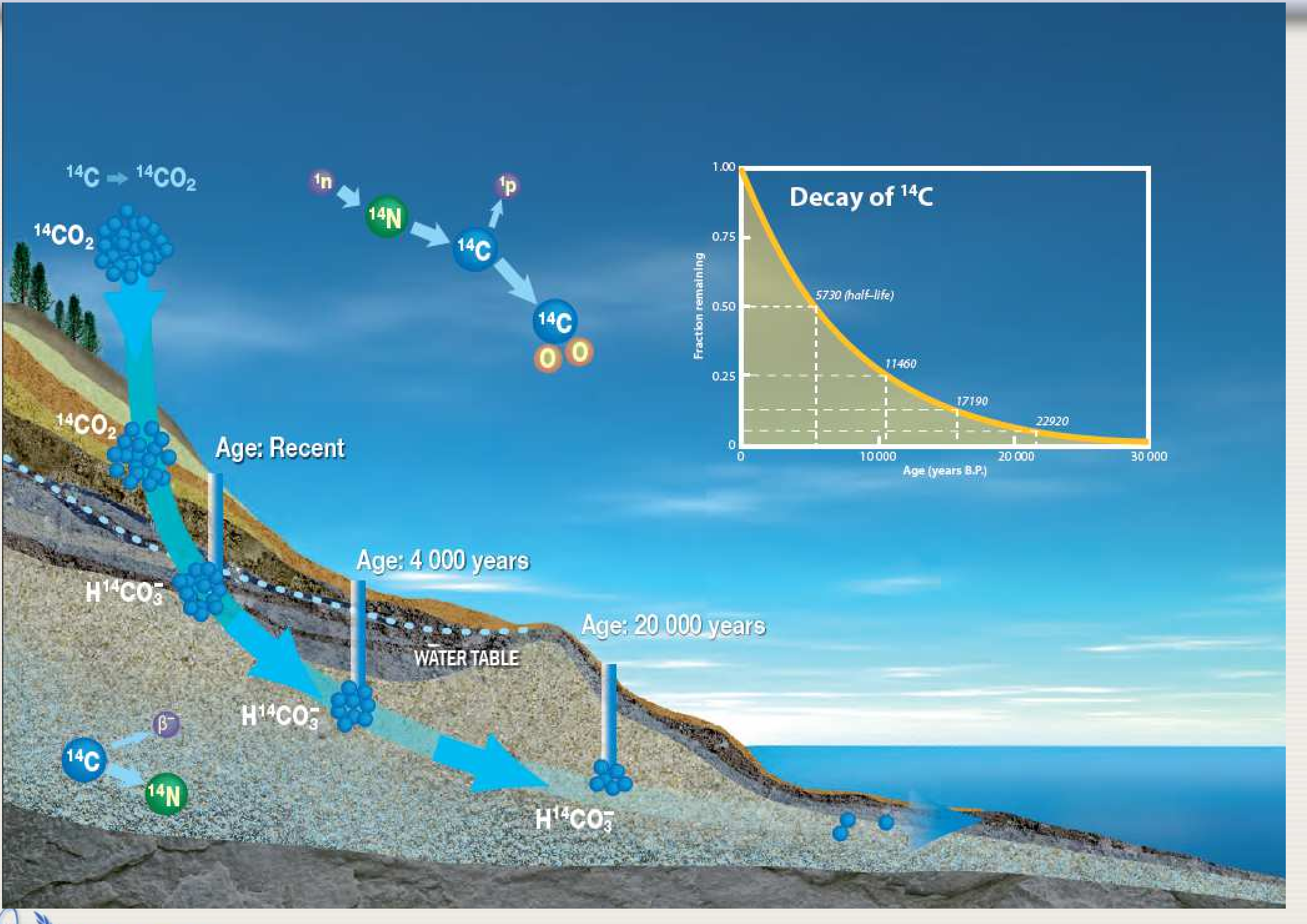
Precipitation & Danube river tritium (Vienna, 1960-2010)



Tritium (^3H) and C-14 in nature

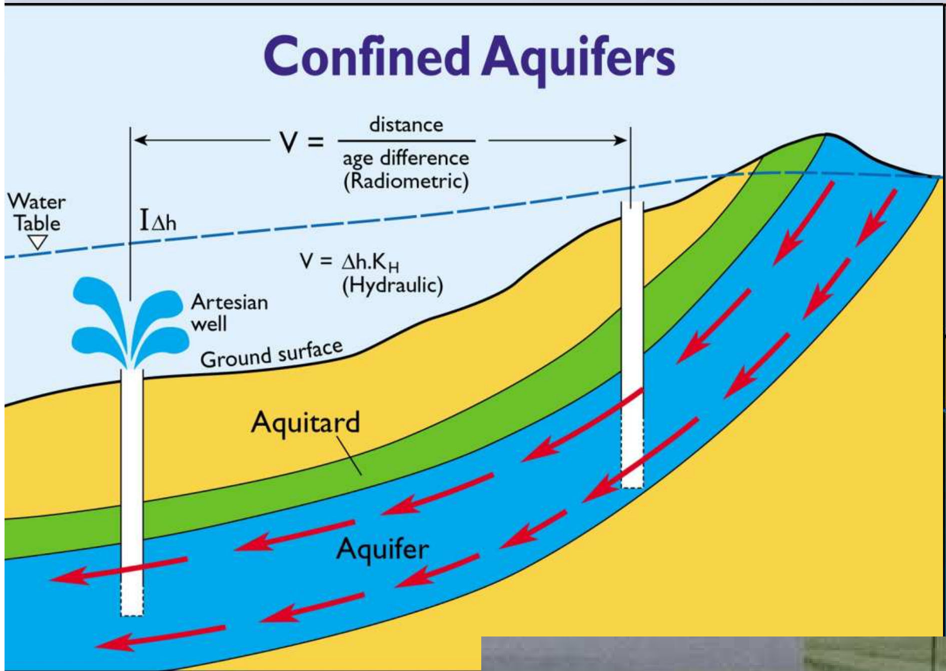


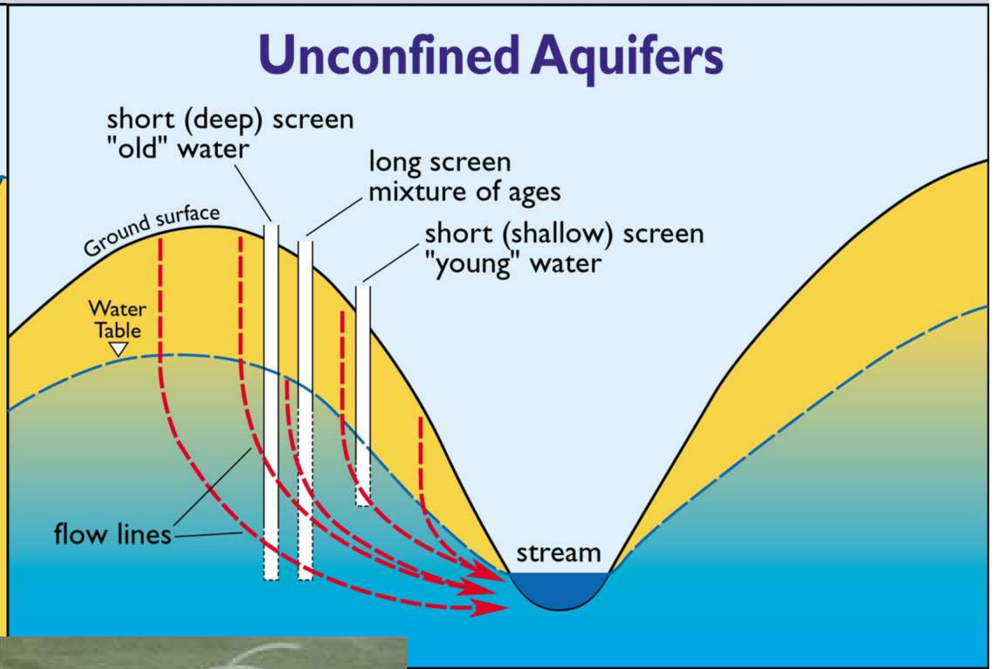
Decay of ^{14}C allows groundwater dating



IAEA

Sampling: confined vs unconfined aquifers

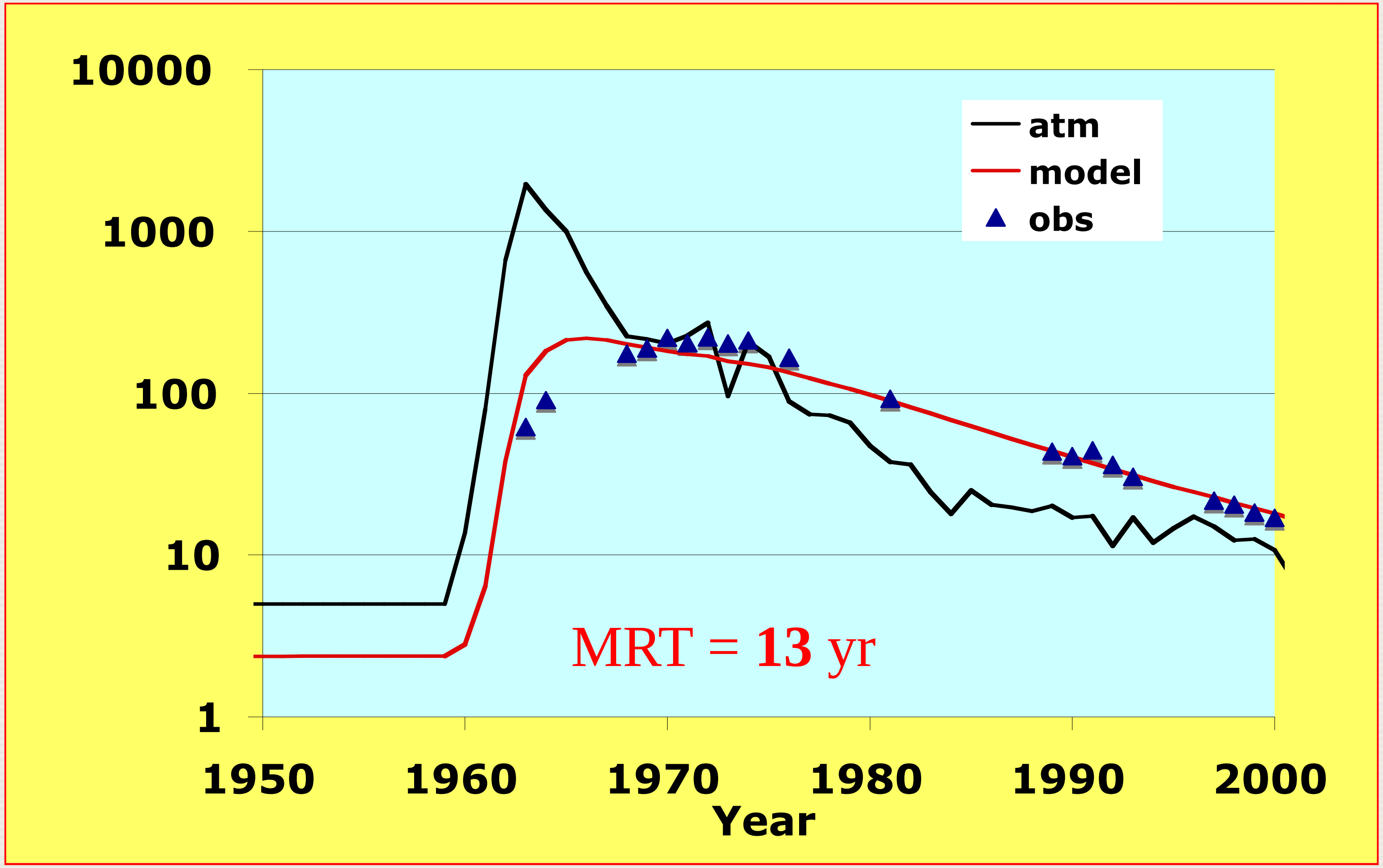




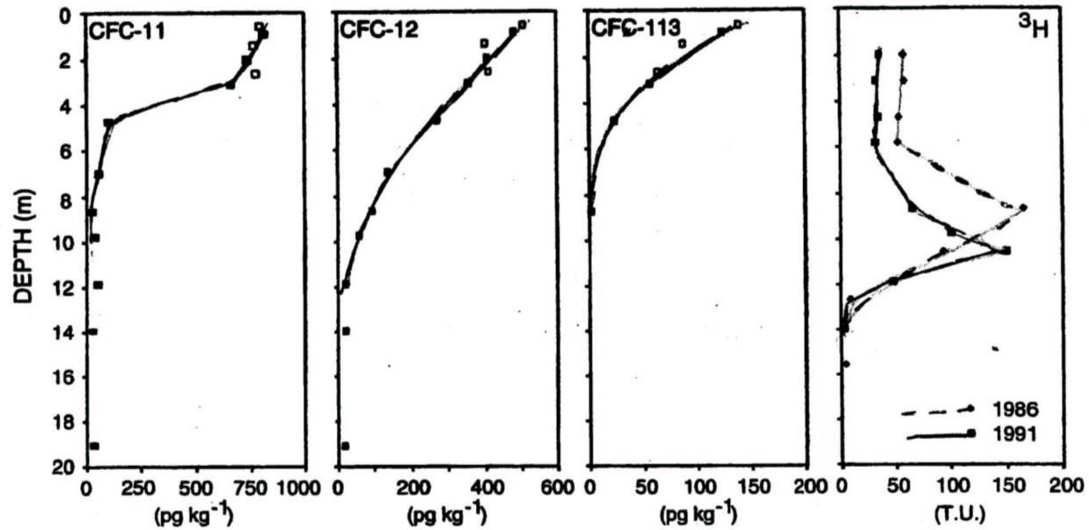
Representative water samples: Importance of adequate protocols for sampling, storage and shipment

IAEA

Single Component Dispersion Model



IAEA



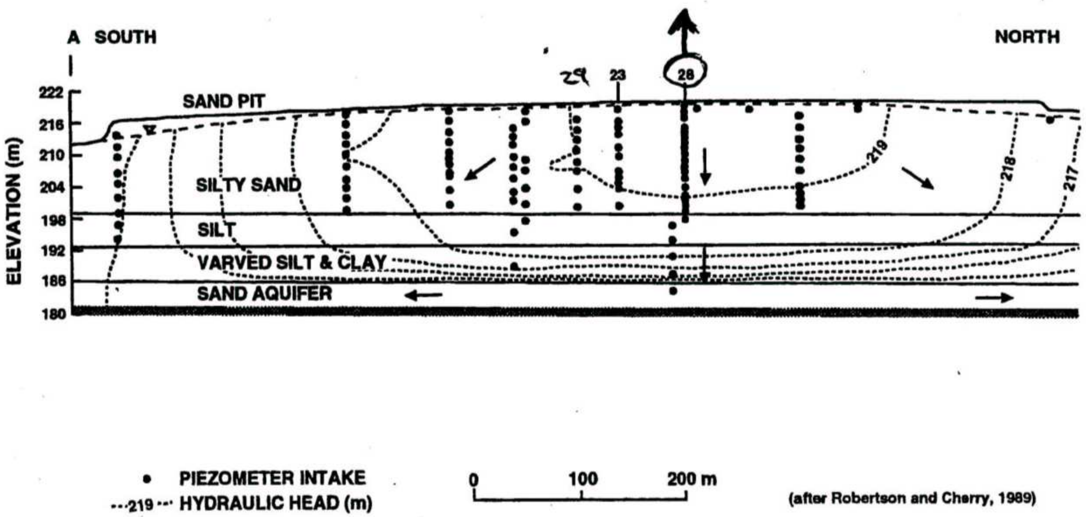


Figure 3. North-south cross section through the Sturgeon Falls surficial aquifer, showing the hydraulic head distribution measured on August 8, 1986. After *Robertson and Cherry* [1989].

Measuring vertical infiltration in a sandy aquifer with CFCs and tritium

Sturgeon Falls, Ontario

Surficial silty sand aquifer

Water table near land surface

IAEA

Cook, P. G., Solomon, D. K., Plummer, L. N., Busenberg, E., and Schiff, S. L., 1995, Chlorofluorocarbons as tracers of groundwater transport processes in a shallow, silty sand aquifer: Water Resources Research, v. 31, p. 425-434.

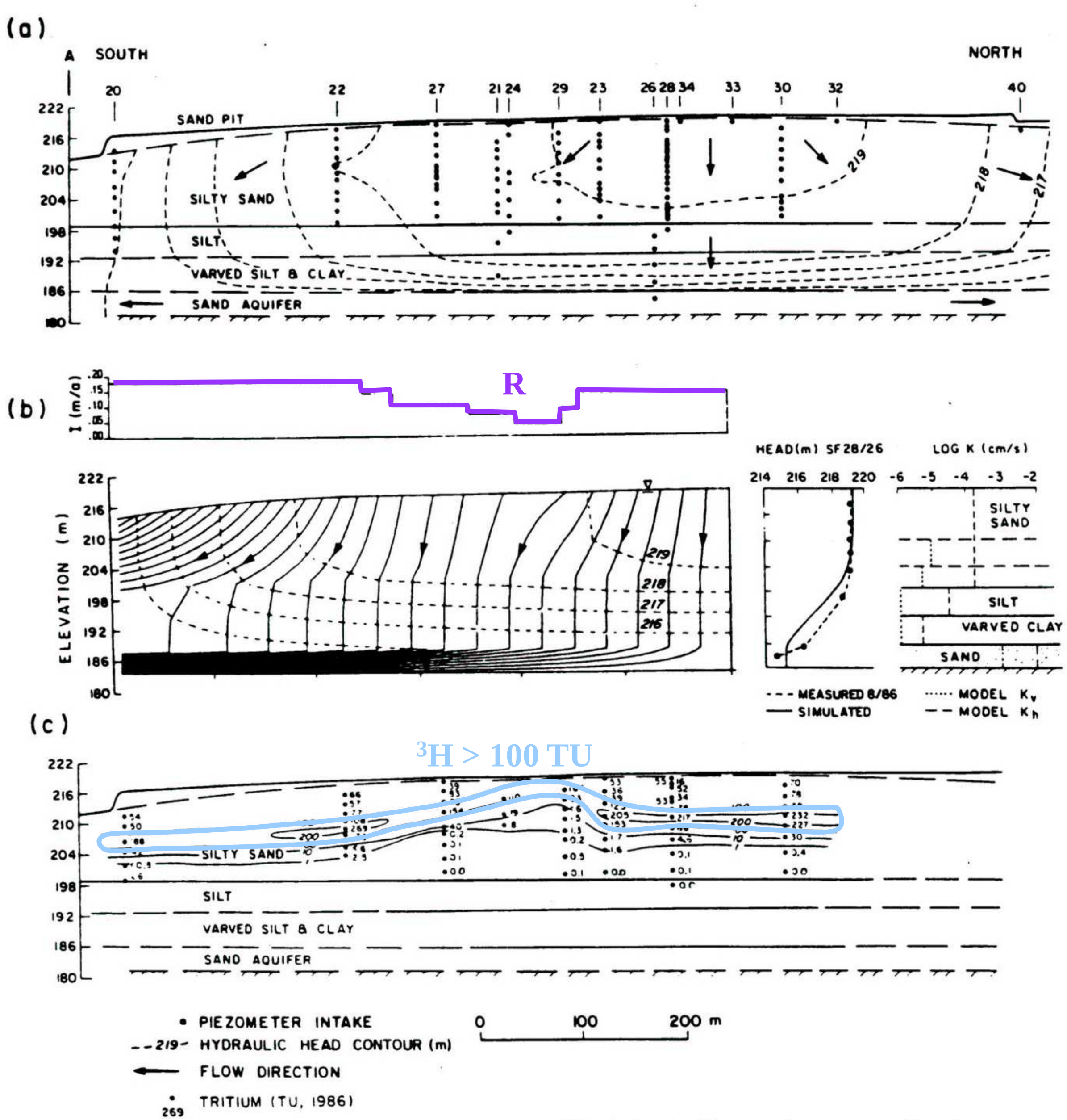


Fig. 3. Section A-A': (*a*) hydraulic head measured, August 1986, (*b*) simulated flow net, showing water table recharge rate and K layering used for simulation, and (*c*) tritium distribution.

Sturgeon Falls, Ontario

2-D flow model with parameters to fit the distribution of ^3H

Recharge varies from about 0.05 to 0.18 m/yr

IAEA

Robertson, W. D., and Cherry, J. A., 1989, Tritium as an indicator of recharge and dispersion in a groundwater system in central Ontario: Water Resources Research, v. 25, p. 1097-1109

Use of tritium for gw dating

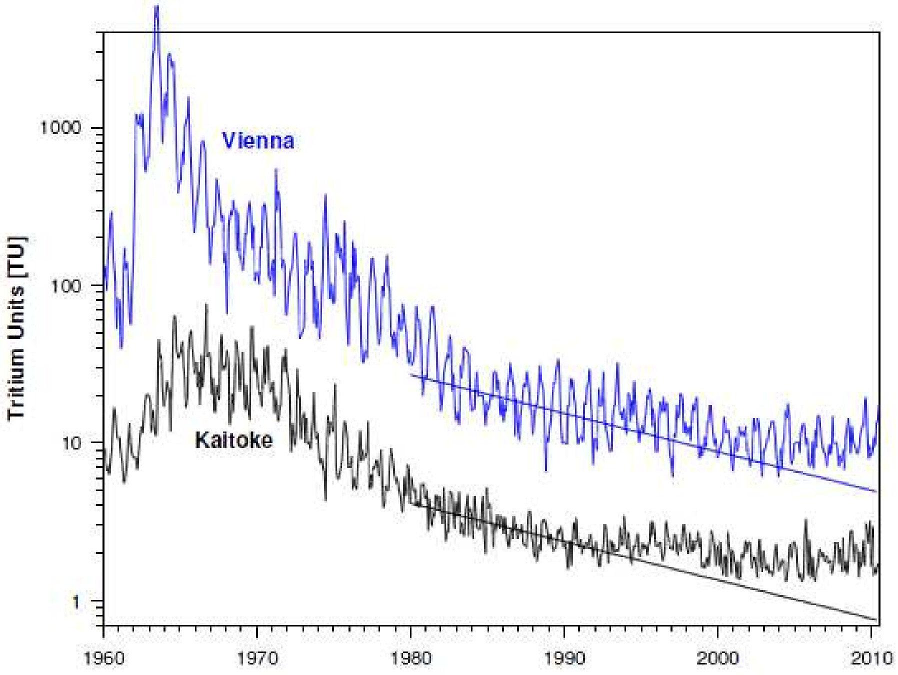


Fig. 1. Tritium in precipitation. The rain record for the Southern Hemisphere is from Kaitoke, New Zealand, and that for the Northern Hemisphere is from Vienna, Austria. Data for Vienna are from Global Network of Isotopes in Precipitation (GNIP), with data for recent years from Manfred Groening, IAEA (personal communication, 2009). One TU corresponds to an atomic ratio of tritium/total hydrogen of 10^{-18}, and a tritium concentration of 0.11919 Bq/kg (Morgenstern and Taylor, 2009).

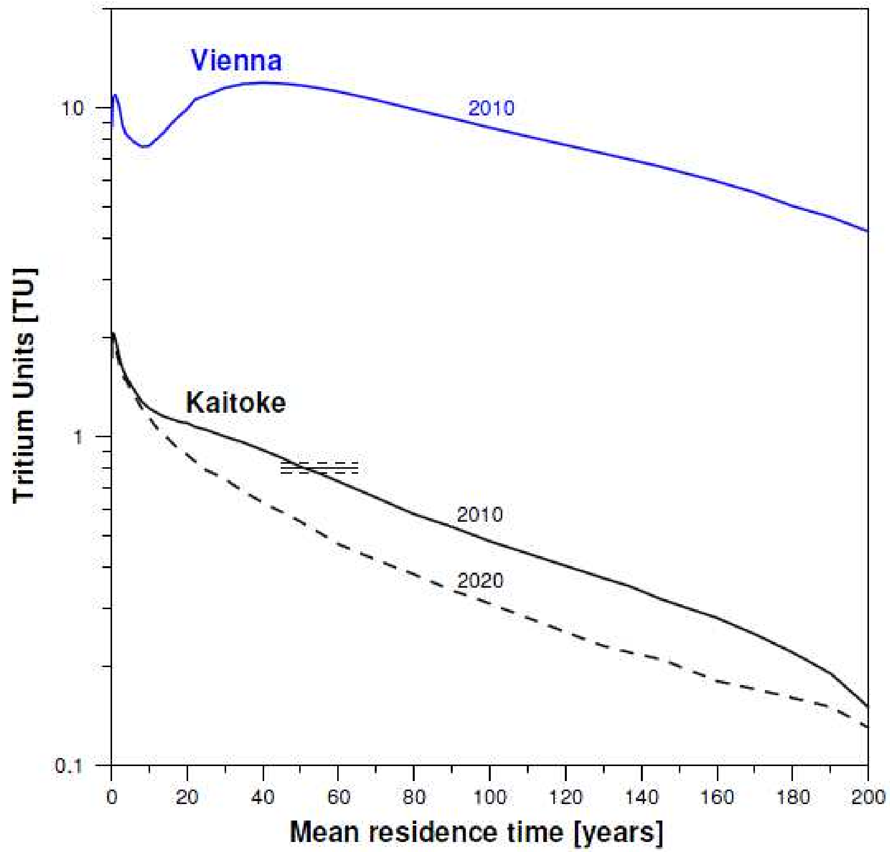
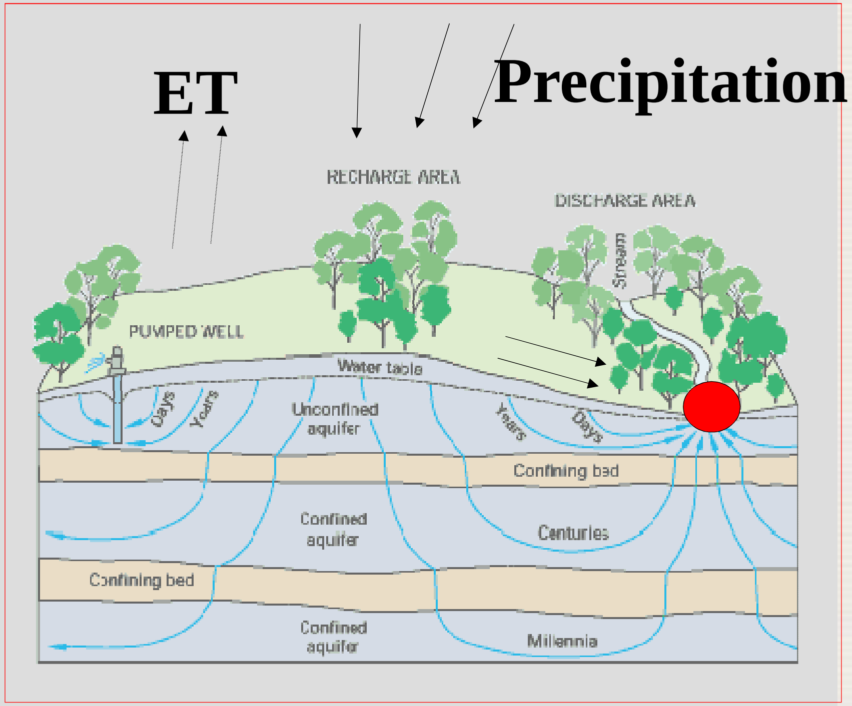


Fig. 2. Tritium output for a typical transfer function of 80% exponential flow within an exponential piston flow model for the Kaitoke (New Zealand) and Vienna (Europe) tritium inputs. Solid lines are current tritium outputs for the year 2010. The predicted output for Kaitoke for 2020 (dashed line) is shown for comparison.

Why do we care about isotopes in streams & rivers?

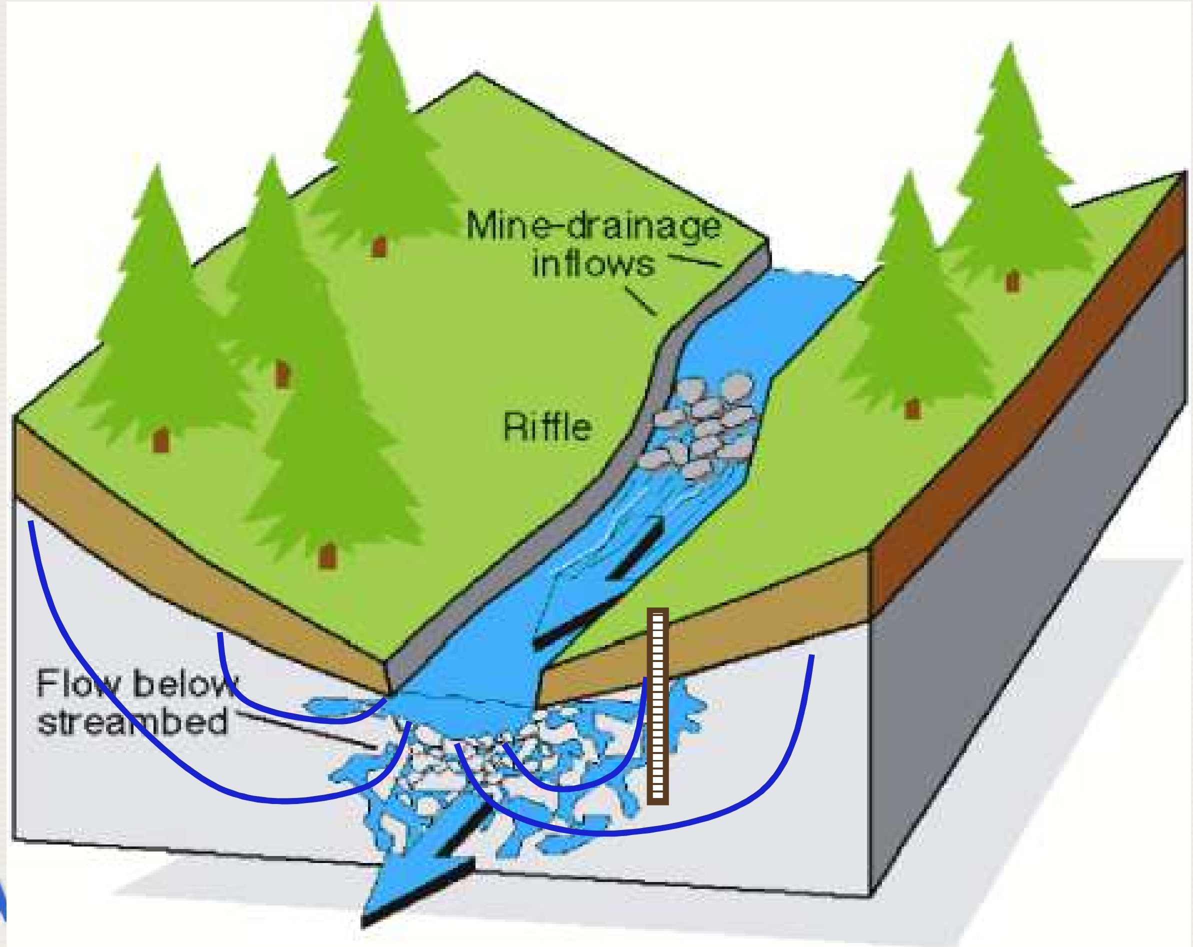
- **Rivers are good integrators of processes affecting basins**
- **EU water framework asks for collection of baseline climate change indicators**
  - **We can use isotopes as baseline indicators!**

- **Surface water/groundwater interactions**
- **Residence times/cycle times**
- **Sources of recharge/precip**
- **Seasonality effects**
- **GNIR as a basis for local to global comparisons**



Concept of Mean Residence Time (MRT) for surface and ground-waters

- Requires a flow weighted sampling of ALL flow paths
- Integrated samples MIGHT be obtained from base flow



IAEA

Understanding nitrate, ^{15}N and groundwater ages

Nitrogen isotopes and groundwater dating provided the key elements for explaining varying nitrate contents and changing redox conditions in an agricultural catchment

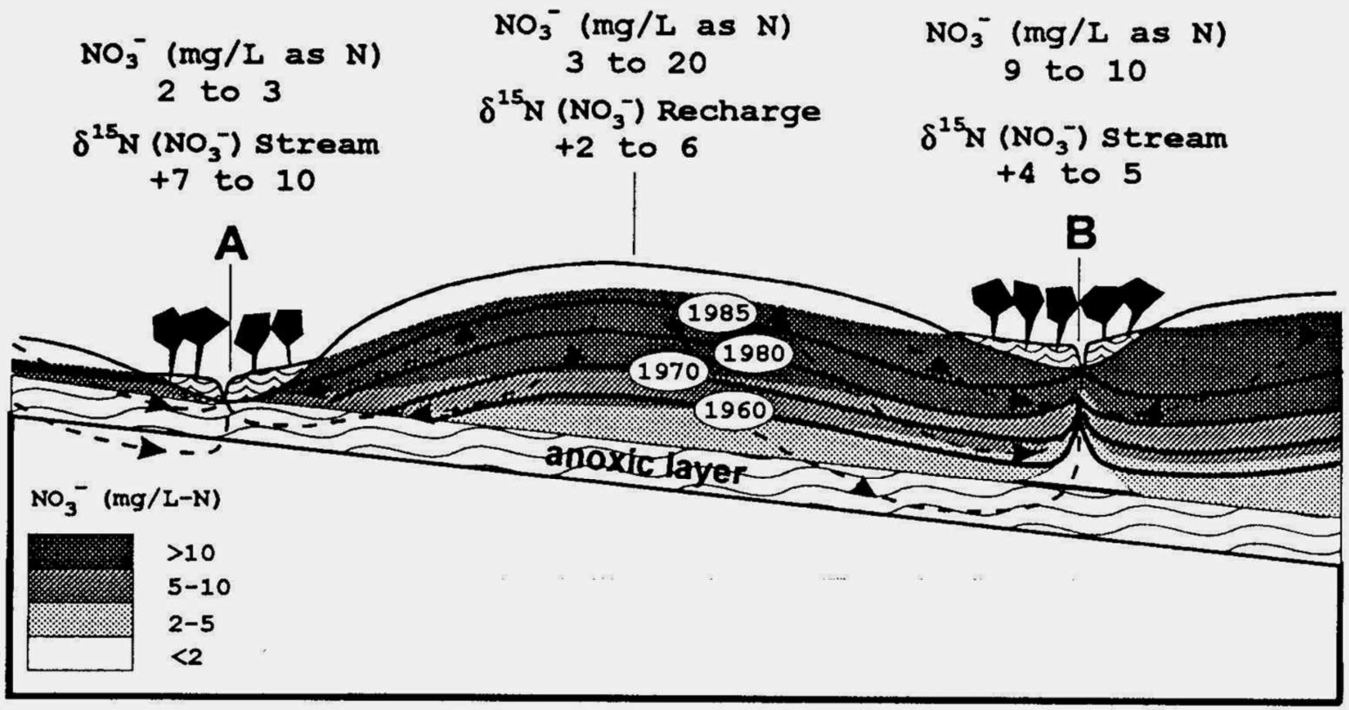
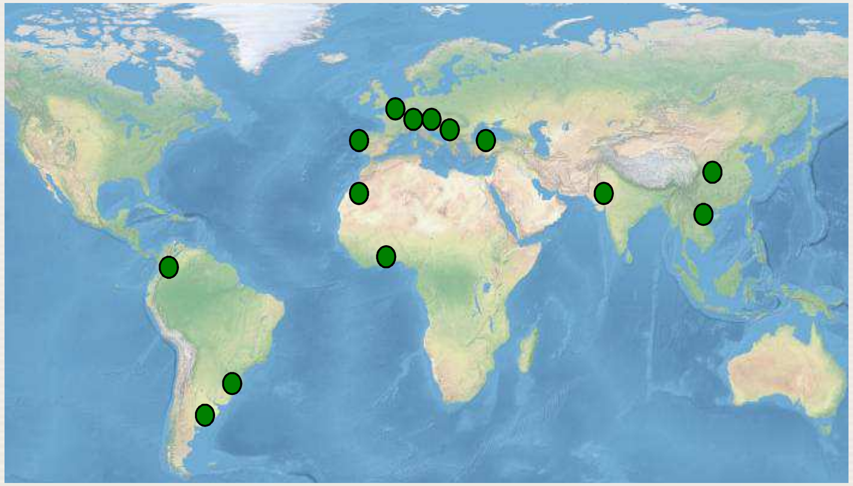
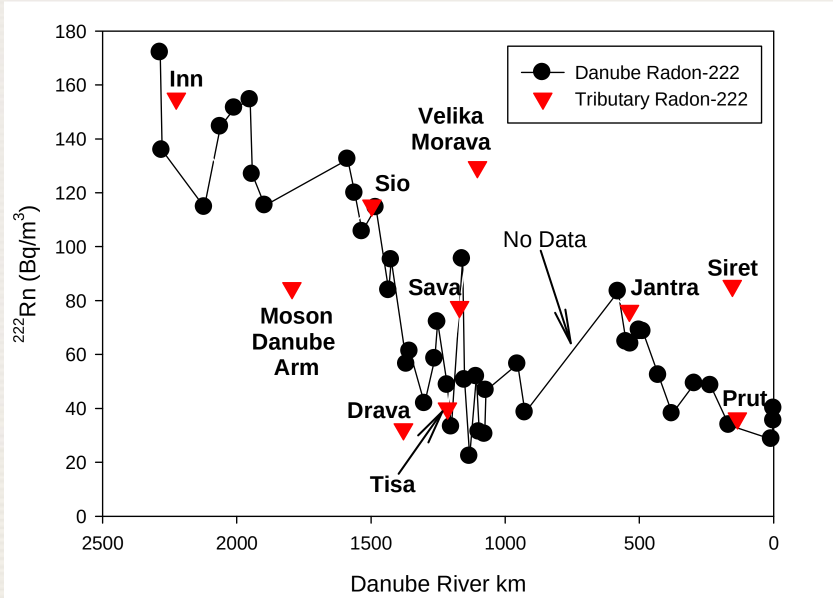


Figure 16.5. Schematic cross-section through an agricultural catchment in the Delmarva Peninsula, Maryland (USA) showing the increase in age of waters (solid lines, based on CFC data) and decrease in nitrate concentration with depth (shaded zones). Flowlines (dashed) to stream A are more shallow than the flowlines to stream B, intersect the riparian zone, and many flowlines are within the anoxic bedrock unit; hence, enchanced denitrification along the flowlines contributing to stream A results in lower nitrate concentrations and higher $\delta^{15}N$ values than in stream B. Modified from Böhlke and Denver (1995).

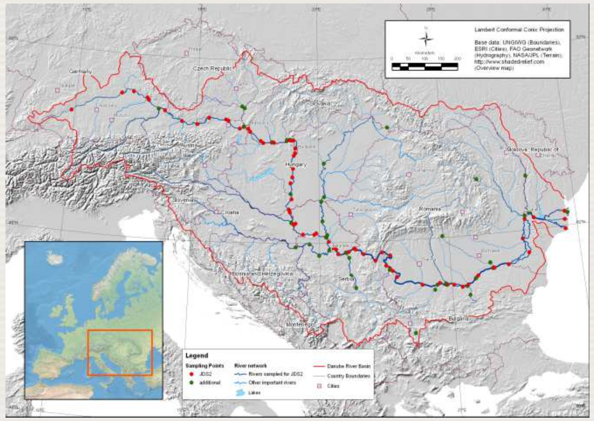
IAEA project on baseflow/shallow groundwater dating





2004-2009: using stable isotopes, tritium, and CFCs (ages: 11- ~50 yrs)

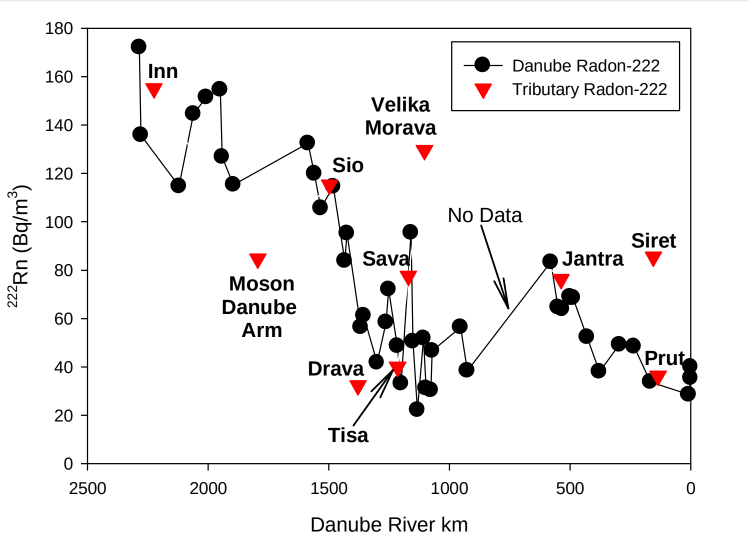
2010-2015: tritium/helium-3 dating
Also to include Rn-222 surveys to locate zones of g/w discharge



IAEA

Groundwater-river interaction: identification of localized discharge in rivers using ^{222}Rn detectors

Recently developed portable Rn-222 analyser is used to identify areas of groundwater discharge in rivers and lakes, facilitating sampling for chemistry/isotopes and optimize monitoring networks



IAEA

Lumped parameter models describing gw flow: Software

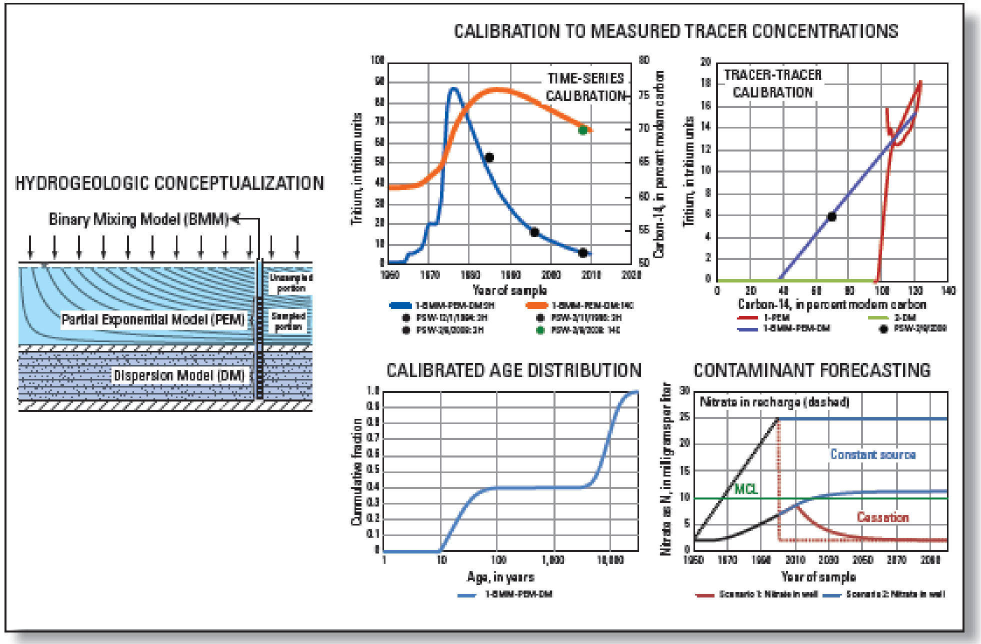
Multis (1995)
FlowPC
Boxmodel
Tracermodel
Lumped
Lumpy
TracerLPM – USGS (2012)

IAEA

USGS
science for a changing world

National Water-Quality Assessment Program
National Research Program

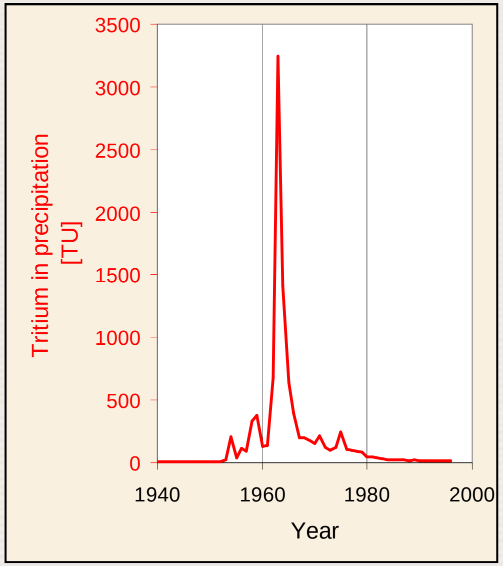
TracerLPM (Version 1): An Excel® Workbook for Interpreting Groundwater Age Distributions from Environmental Tracer Data

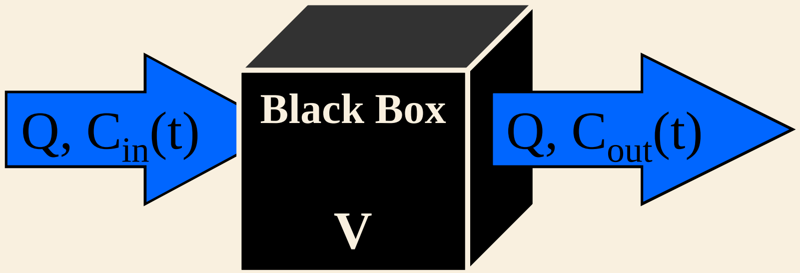


Techniques and Methods 4–F3

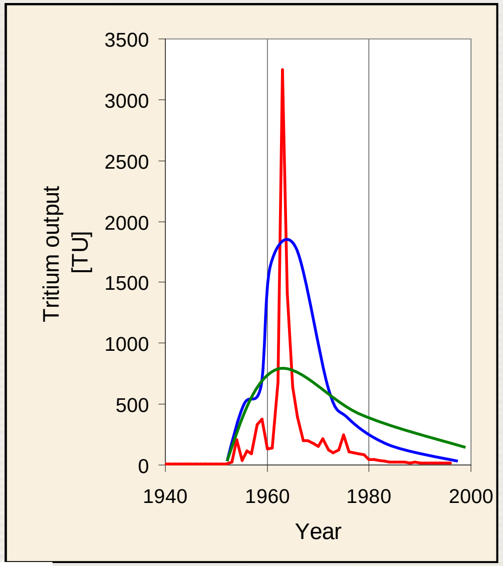
U.S. Department of the Interior
U.S. Geological Survey

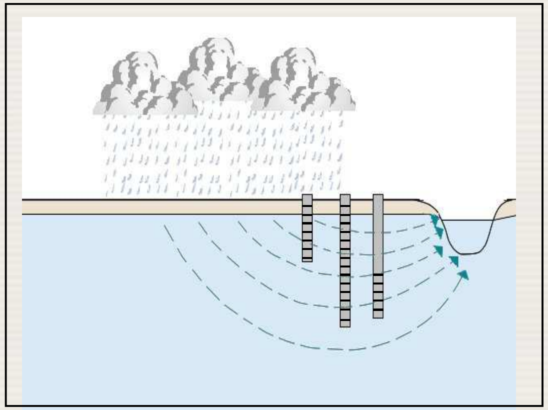
Lumped parameter models describing gw flow

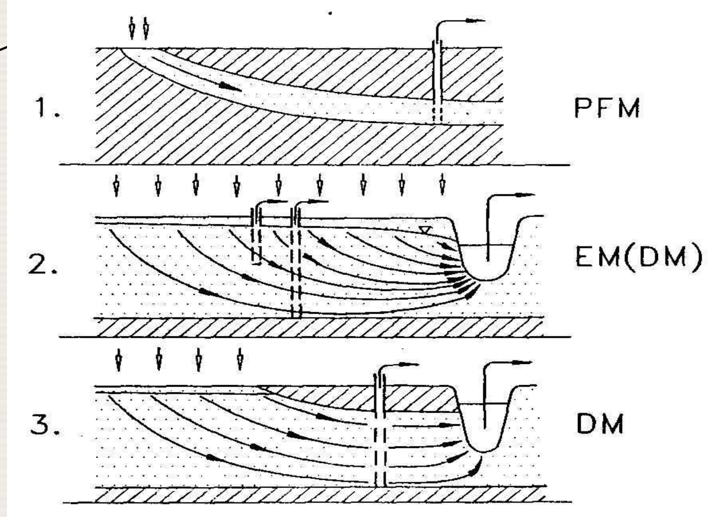




$$C_{out}(t) = \int_{-\infty}^{t} C_{in}(t-t') \cdot g(t') \cdot e^{-\lambda t'} \, dt'$$



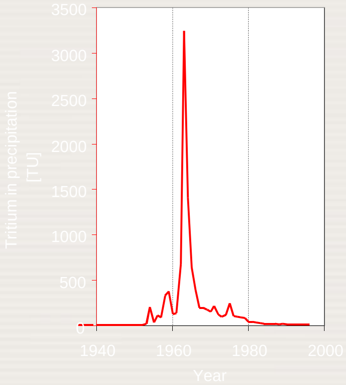


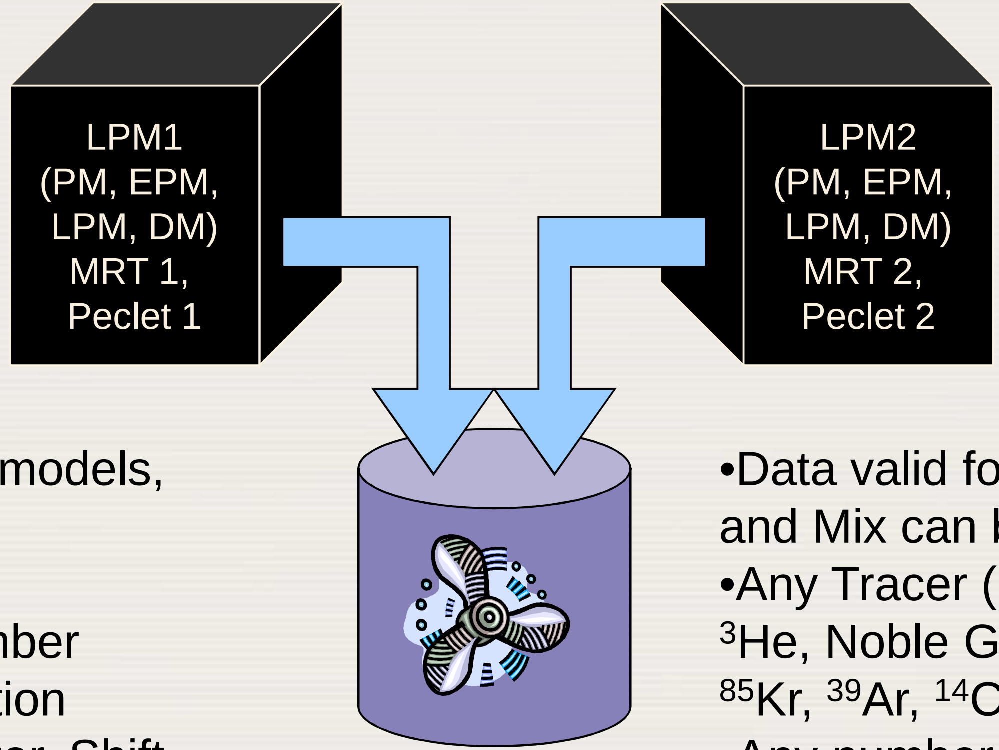


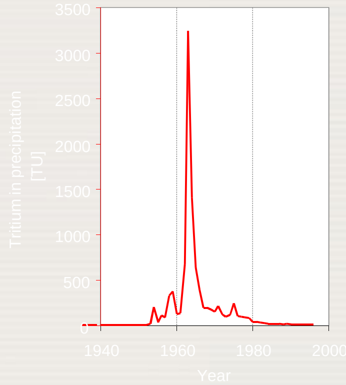
Main flow models:

- **Piston**
- **Exponential**
- **Dispersive**

Lumpy: Two Lumped Parameter Models in Parallel

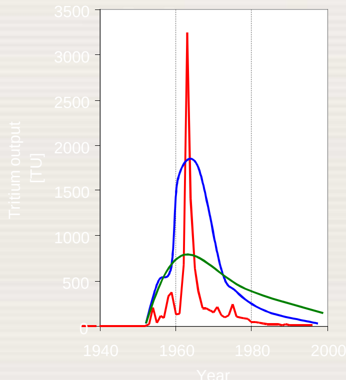




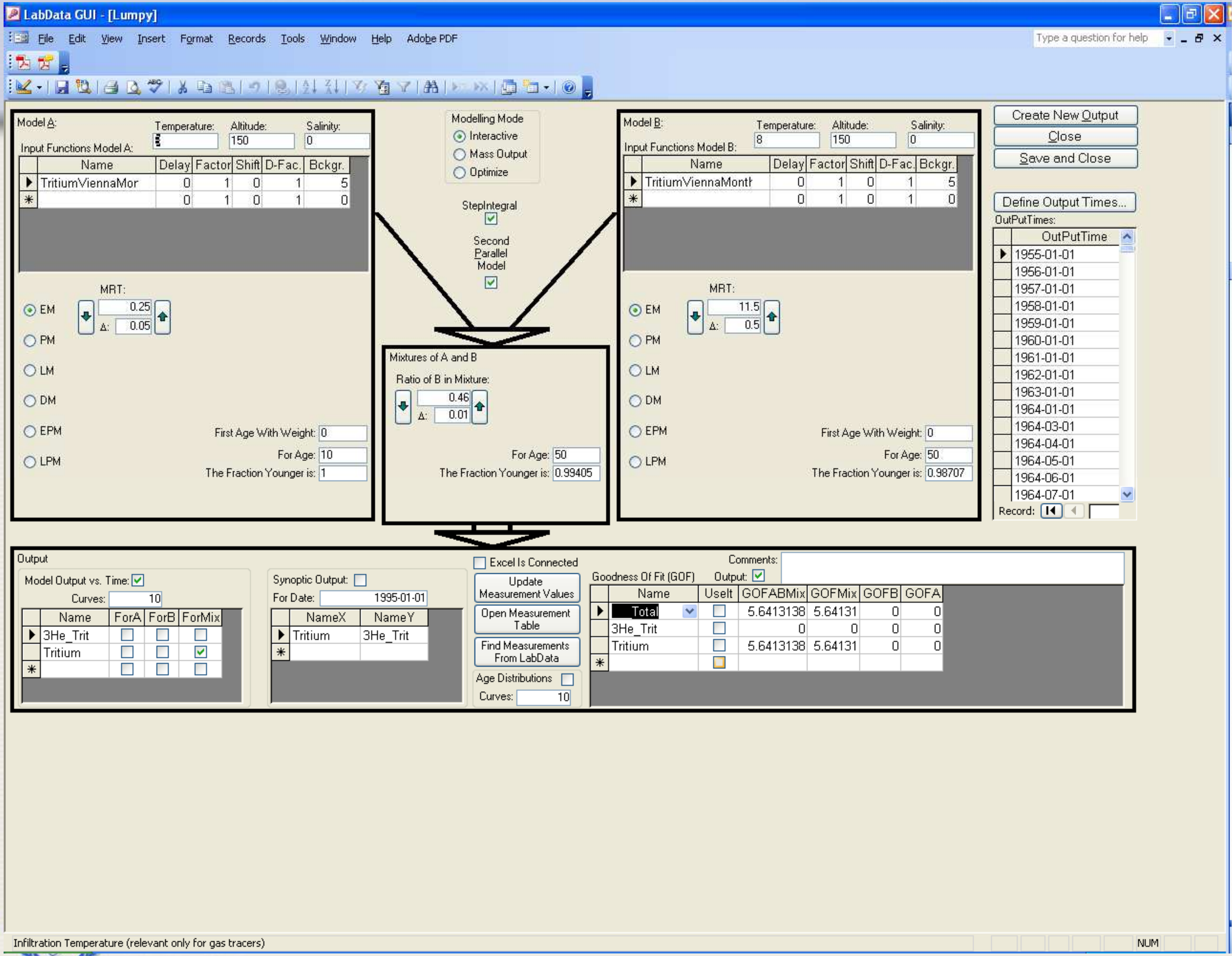


- 2 lumped parameter models,
- Each having:
    - Own MRT
    - Own Peclet Number
    - Own Input Function
    - Own Delay, Factor, Shift
- Are mixed in free proportions

- Data valid for LPM1, LPM2 and Mix can be used to fit.
- Any Tracer (^{18}O, D, Tritium, ^{3}He, Noble Gases, CFC, SF_6, ^{85}Kr, ^{39}Ar, ^{14}C, ^{81}Kr, ^{36}Cl…).
- Any number of measurements.
- Automated best fit selection for 5 parameters (MRT, Peclet, Mix)



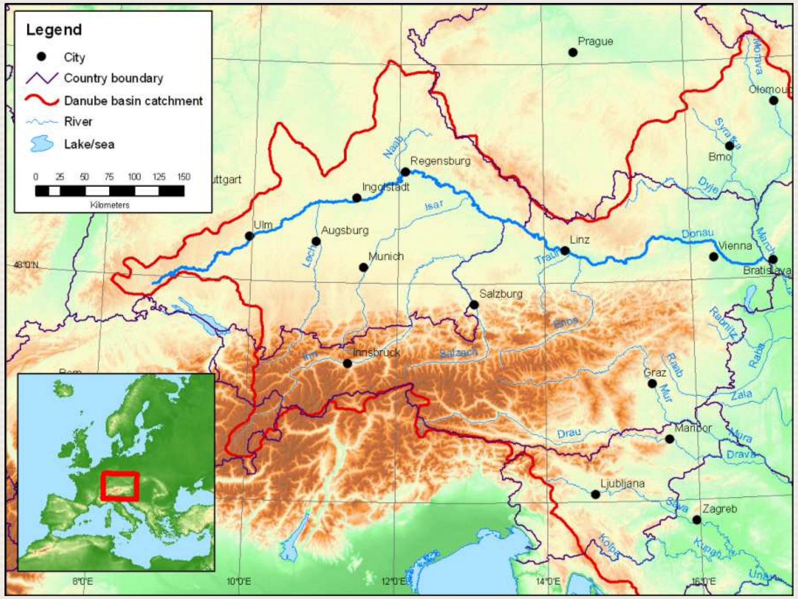
IAEA


LabData GUI - [Lumpy]
File Edit View Insert Format Records Tools Window Help Adobe PDF
Type a question for help
Model A:
Temperature: Altitude: 150 Salinity: 0
Input Functions Model A:
Name Delay Factor Shift D-Fac. Bckgr.
TritiumViennaMor 0 1 0 1 5
0 1 0 1 0
MRT: 0.25
Δ: 0.05
EM
PM
LM
DM
EPM
LPM
First Age With Weight: 0
For Age: 10
The Fraction Younger is: 1
Modelling Mode
Interactive
Mass Output
Optimize
StepIntegral
Second Parallel Model
Mixtures of A and B
Ratio of B in Mixture: 0.46
Δ: 0.01
For Age: 50
The Fraction Younger is: 0.99405
Model B:
Temperature: 8 Altitude: 150 Salinity: 0
Input Functions Model B:
Name Delay Factor Shift D-Fac. Bckgr.
TritiumViennaMonth 0 1 0 1 5
0 1 0 1 0
MRT: 11.5
Δ: 0.5
First Age With Weight: 0
For Age: 50
The Fraction Younger is: 0.98707
Create New Output
Close
Save and Close
Define Output Times...
OutPutTimes:
OutPutTime
1955-01-01
1956-01-01
1957-01-01
1958-01-01
1959-01-01
1960-01-01
1961-01-01
1962-01-01
1963-01-01
1964-01-01
1964-03-01
1964-04-01
1964-05-01
1964-06-01
1964-07-01
Record:
Output
Model Output vs. Time:
Curves: 10
Name ForA ForB ForMix
3He_Trit
Tritium
Synoptic Output:
For Date: 1995-01-01
NameX NameY
Tritium 3He_Trit
Excel Is Connected
Update Measurement Values
Open Measurement Table
Find Measurements From LabData
Age Distributions
Curves: 10
Comments:
Goodness Of Fit (GOF) Output:
Name UseIt GOFABMix GOFMix GOFB GOFA
Total 5.6413138 5.64131 0 0
3He_Trit 0 0 0 0
Tritium 5.6413138 5.64131 0 0
Infiltration Temperature (relevant only for gas tracers)
NUM

Danube basin Residence Times

Basin residence times are a major control on temporal concentrations of nutrients and contaminants

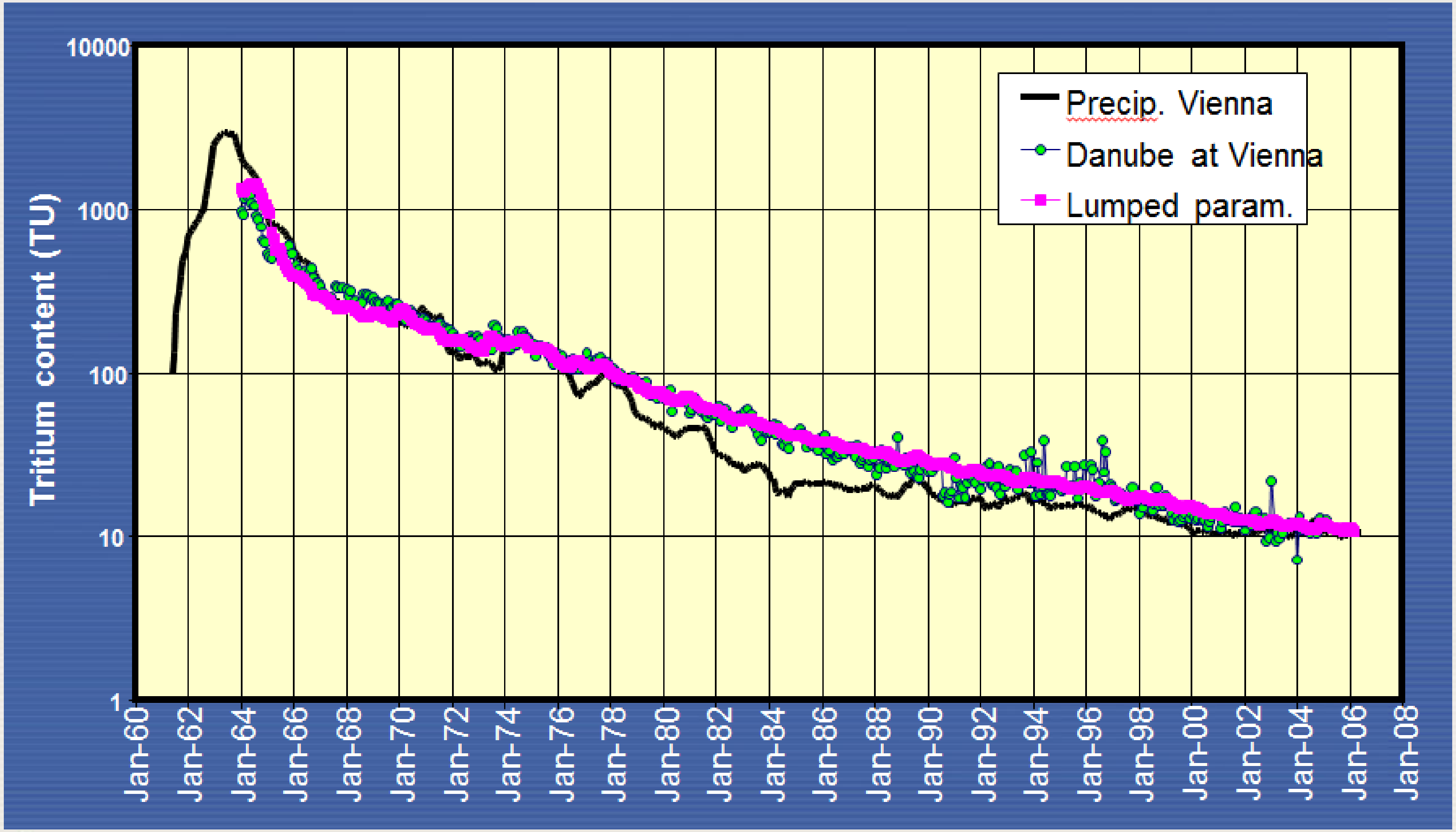
Evaluation of water residence times of the upper Danube using tritium measurements of precipitation and river water



Upper Danube Basin

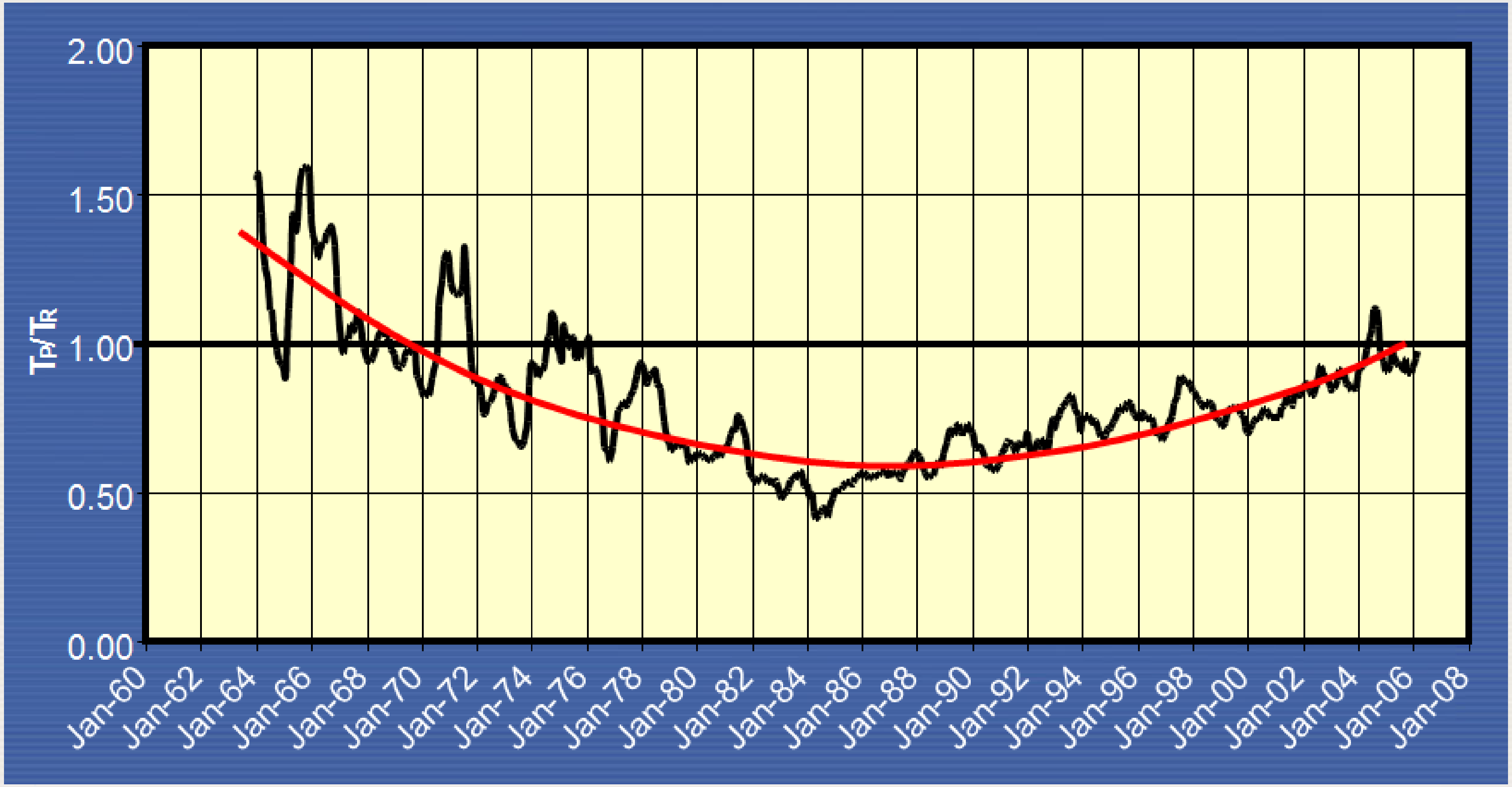
IAEA

Modelled tritium content in Danube (Lumpy)



IAEA

Time evolution of the T_P/T_R ratio in the Upper Danube



River tritium is now at or near pre-bomb levels

IAEA

Model Results

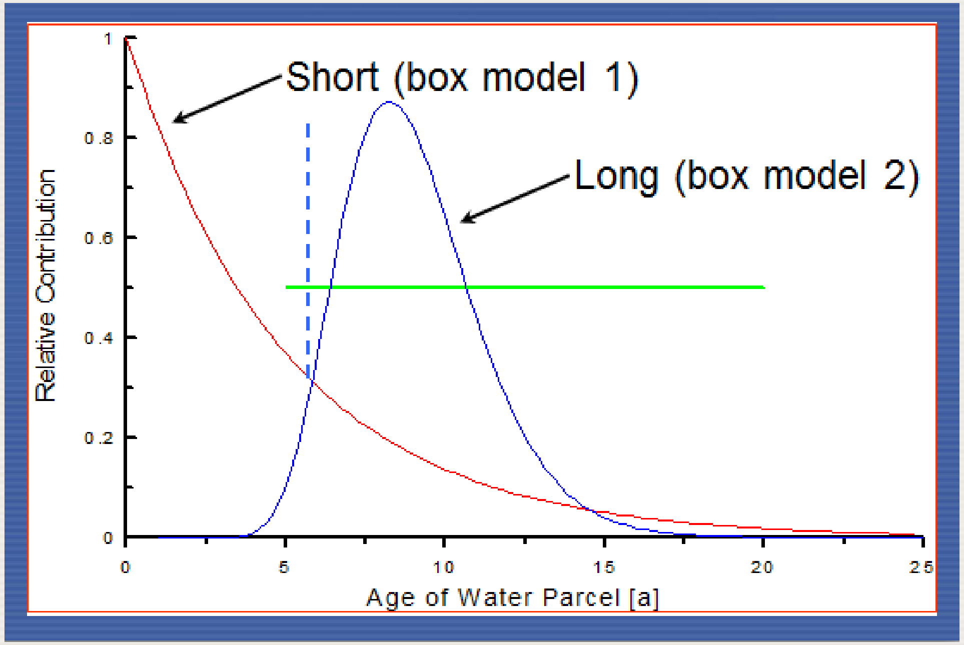
A two-box model provided the best fit.

Box model 1 used piston flow assumption & box model 2 used a gamma assumption

"short" residence time component (mean residence time of 0.75 years) "channel flow"

"long" residence time" component (mean residence time of 15 years) "groundwater flow"

Average combined residence time is about 6 years.



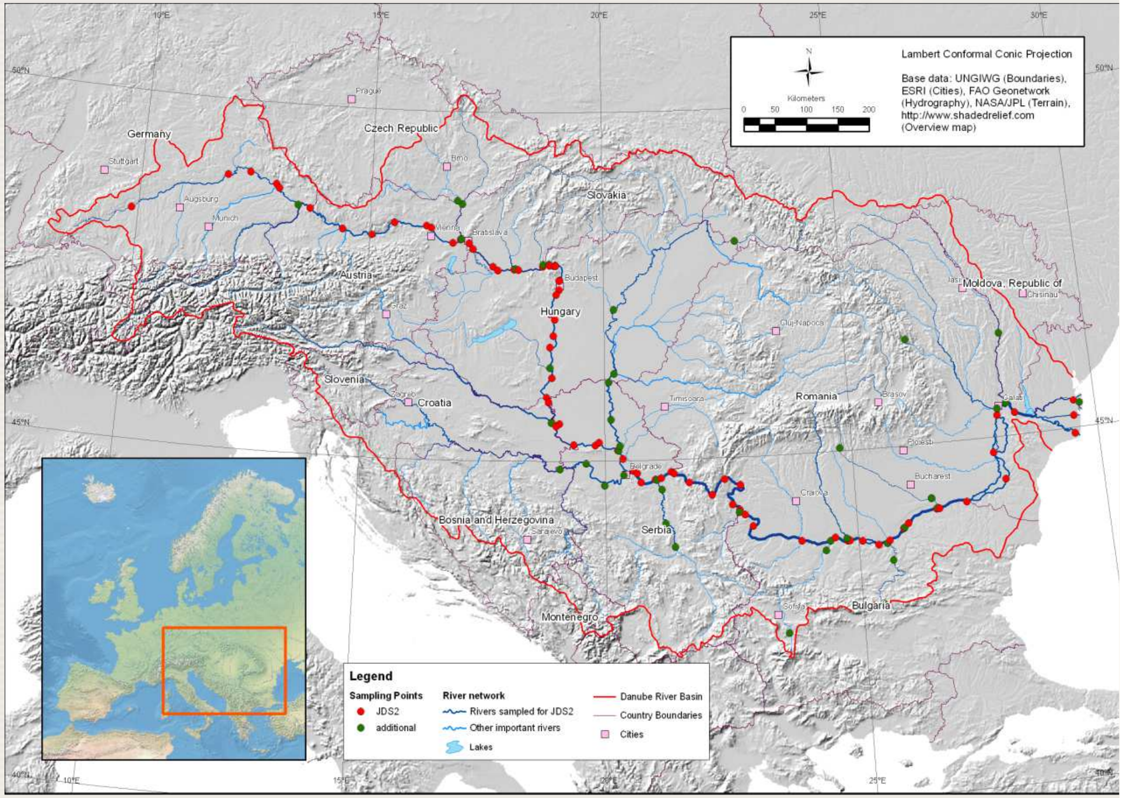
Conclusion - groundwater will moderate concentrations in the basin over the long-term (> 5 years)

IAEA

Danube Survey 2007 – Isotopes, chemistry

Isotopes were used during the 2007 Joint Danube Survey 2, coordinated by the ICPDR to investigate the river hydro-ecology

Source ICPDR, 2009

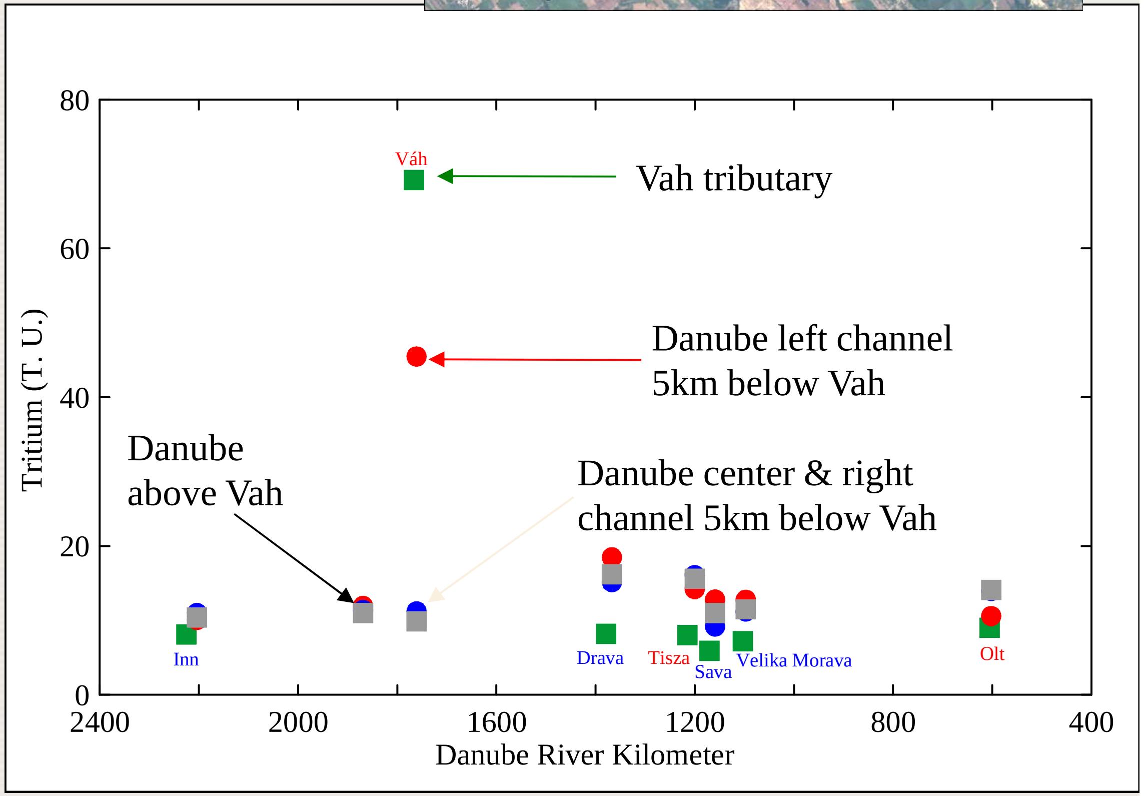


Understanding tributary mixing with tritium

Tritium contents indicate characteristics of river mixing zones below tributary confluences

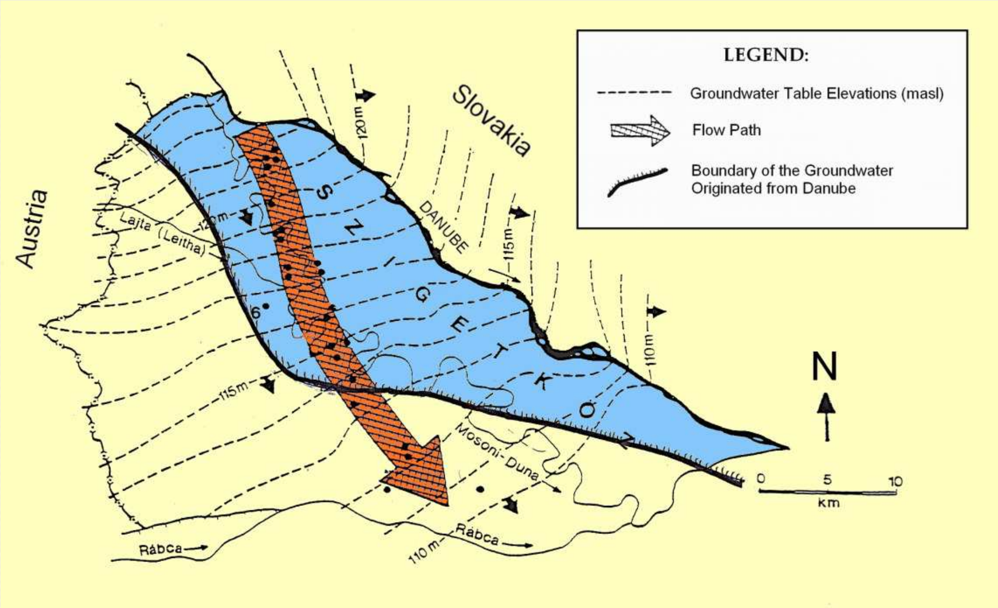
The mixing zone below the Vah extends at least 5 km below the Danube confluence!

Should expect heterogeneity in nutrient and pollutant concentrations at distances well below confluences



IAEA

River bank filtration: Szigetköz, Hungary



The groundwater in the gravel aquifer in Szigetköz is:

- Fresh (less then 50 years old)
- Originates from the Danube
- Horizontal flow velocity is high (up to 500 m/a)

Ref: USGS

Tritium and helium-3 in groundwater

Dating Groundwater Using T/^{3}He

Tritium (radioactive mother) → ^{3}Helium (stable daughter)

$$ {}^3H(t) = {}^3H(0)\cdot e^{-t/\tau} \quad ; \quad {}^3He(t) = {}^3H(0)\cdot(1-e^{-t/\tau}) $$

$$ t = \tau\cdot\ln\left(1+\frac{{}^3He(t)}{{}^3H(t)}\right) $$

- stable ideal tracer T+^{3}He,
- global uniform atmospheric ^{3}He concentration
- independent from tritium input function
- also applicable with tritium contamination

IAEA

T/^{3}He dating: separation of helium components

$$^3He_{meas} = {}^3He_{trit} + {}^3He_{eq} + {}^3He_{exc} + {}^3He_{terr}$$

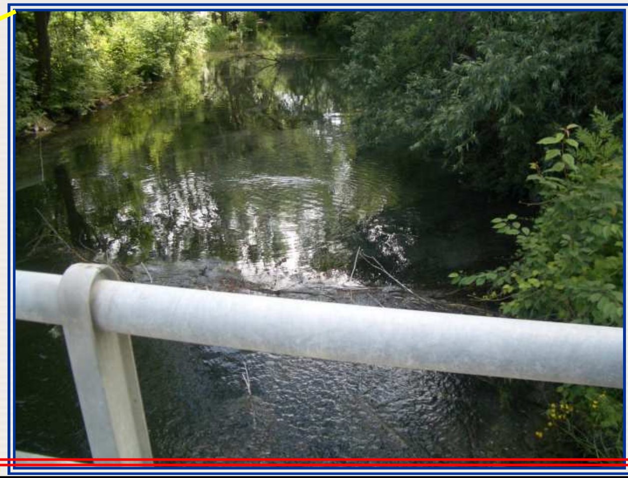
$$^4He_{meas} = {}^4He_{eq} + {}^4He_{exc} + {}^4He_{terr}$$

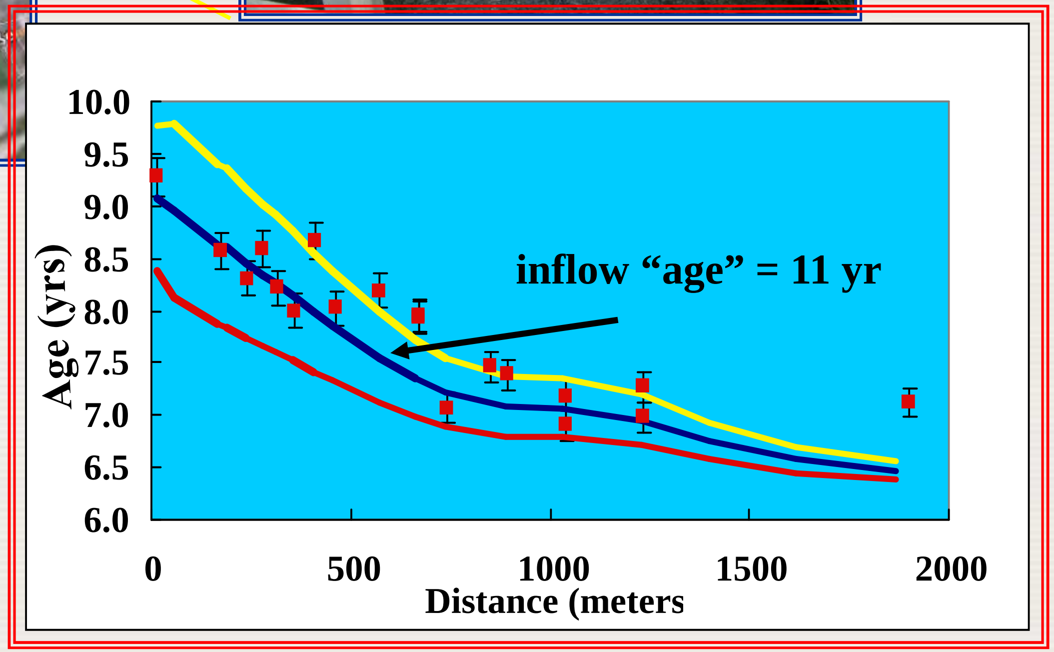
- He_{eq}: Solubility equilibrium, needs infiltration temperature
- He_{exc} Excess air determined via Ne
- He_{terr} separation possible if either crustal He ($^3He/^4He < 10^{-8}$) or mantle He ($^3He/^4He > 10^{-5}$) present, not for both
- typical ^3He in a recent (1a) groundwater:

  80% $^3He_{eq}$, 14% $^3He_{exc}$, 6% $^3He_{trit}$, 0.05% $^3He_{terr}$

IAEA

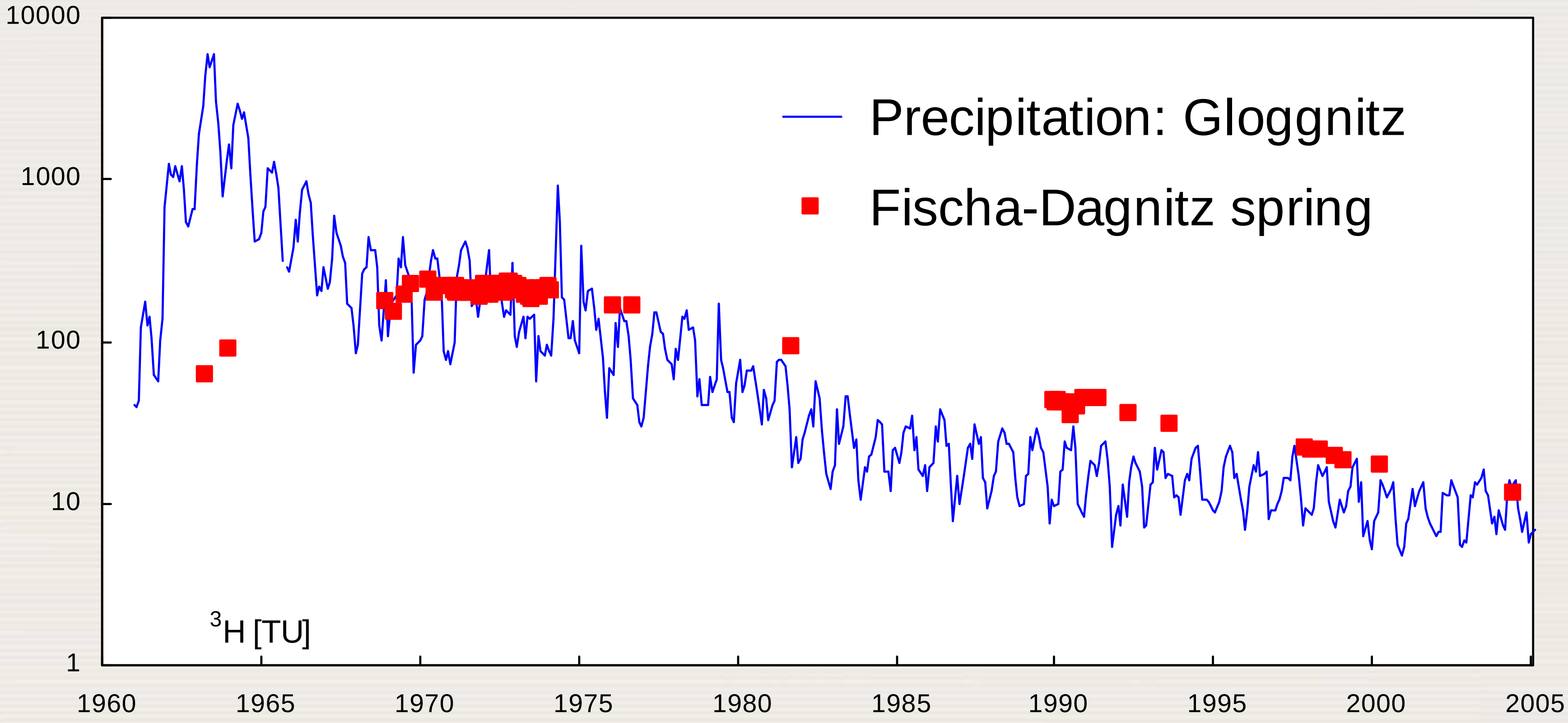
Tritium/helium-3 dating of baseflow
Fischa River, Austria





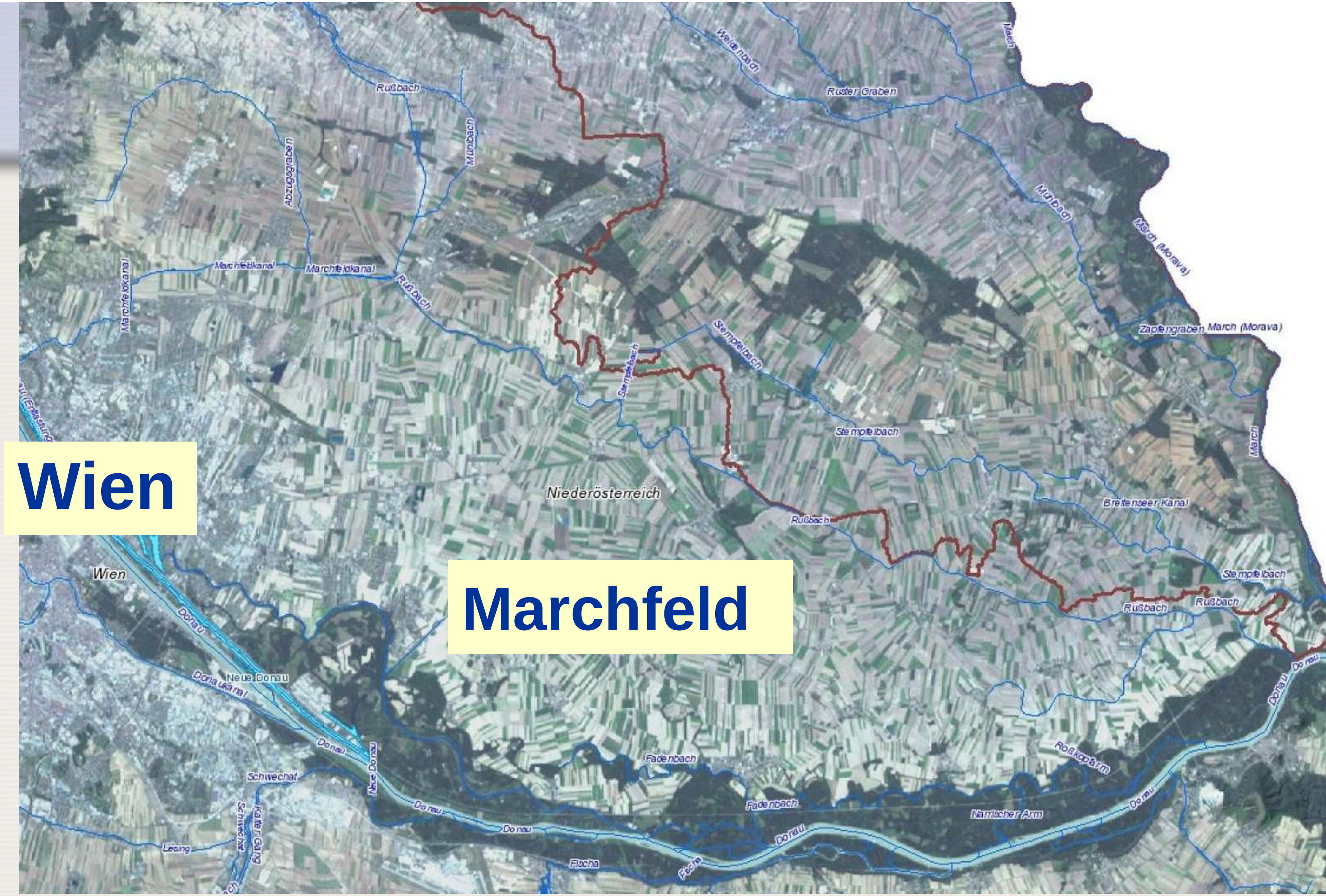
IAEA/K. Solomon, Univ. of Utah

Spring water in the Vienna Basin dated using tritium



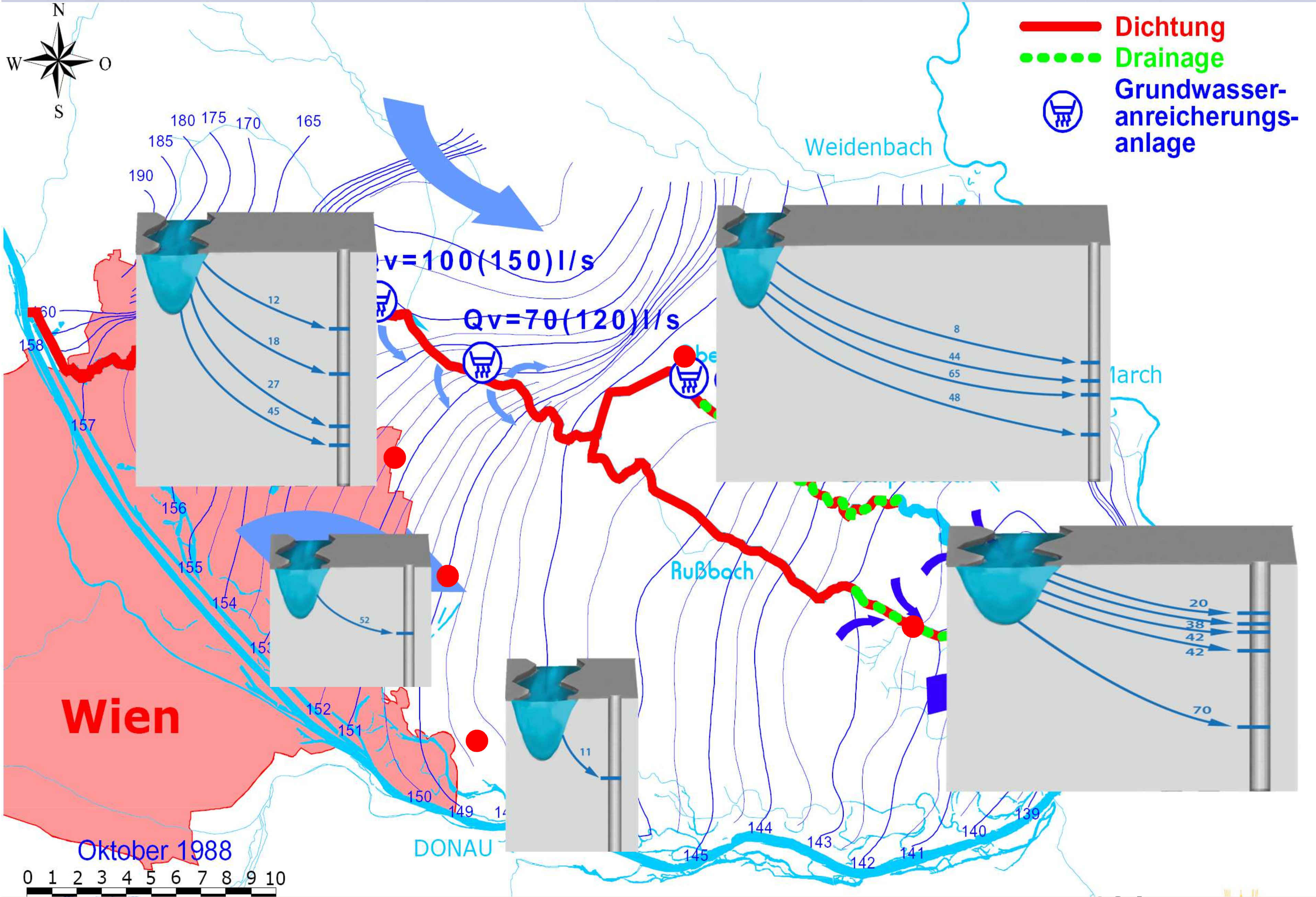
IAEA

Ref: IAEA



IAEA

Ref: M. Kralik (Austrian Env. Agency)



Dichtung
Drainage
Grundwasser-
anreicherungs-
anlage
N
W
O
S
Weidenbach
Qv=100(150)l/s
Qv=70(120)l/s
March
Rußbach
Wien
DONAU
Oktober 1988
0 1 2 3 4 5 6 7 8 9 10
190
185
180
175
170
165
160
158
157
156
155
154
153
152
151
150
149
145
144
143
142
141
140
139
12
18
27
45
52
11
8
44
65
48
20
38
42
42
70

10 km

Concluding remarks: Use of tritium in isotope hydrology

- Tritium still a useful tracer as indicator of recently recharged water (qualitative and quantitative). Simpler use than other tracers of young waters (e.g. CFCs, SF_6, noble gases)

- Combined use of tritium and ^3He offers significant advantages and time resolution to other methods

- Tritium survey at catchment level, supported with ^3He helps in understanding water flows, residence time of water, mixing patterns and assessment/forecast of pollutant behaviour in aquifers and rivers.

IAEA

Thank you !

IAEA