

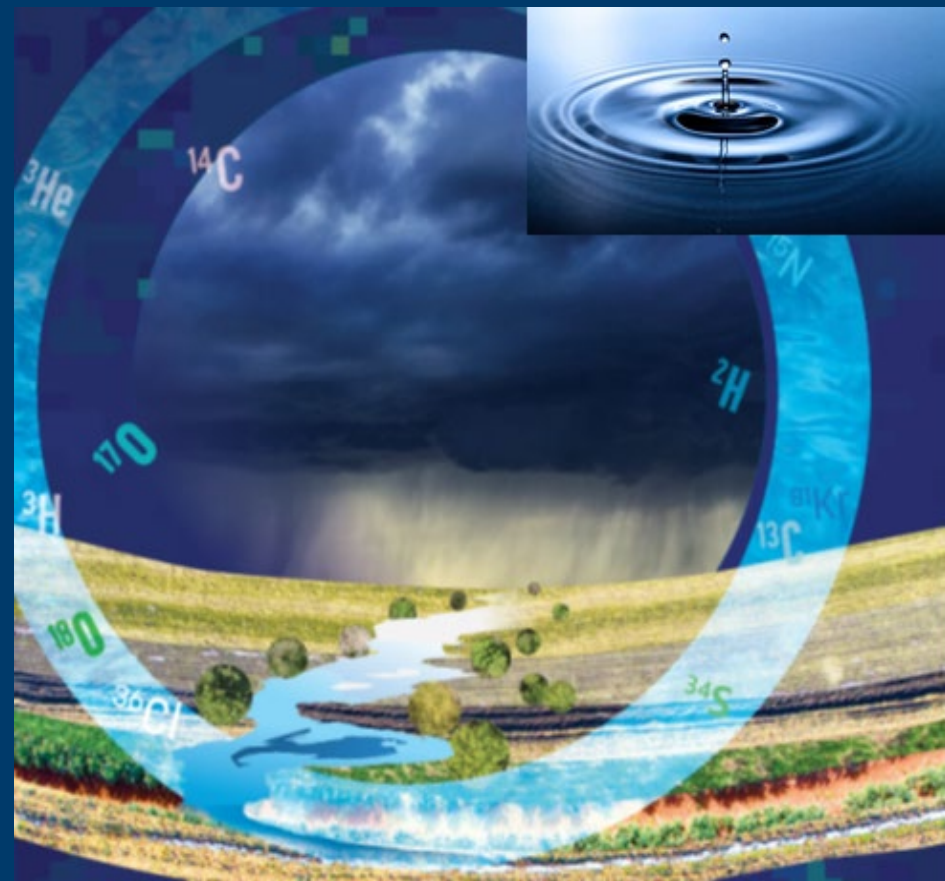
Characterization and analysis of groundwater recharge through tritium measurements

Ferrara Fiere, September, 24 2020

Chiara Telloli, Stefano Salvi, Antonietta Rizzo, Alberto Ubaldini

Methods and Techniques for Nuclear Safety, Monitoring and Traceability Laboratory (TNMT)
Fusion and Technology for Nuclear Safety and Security Department (SICNUC)
Nuclear Safety Security and Sustainability Division (FSN)



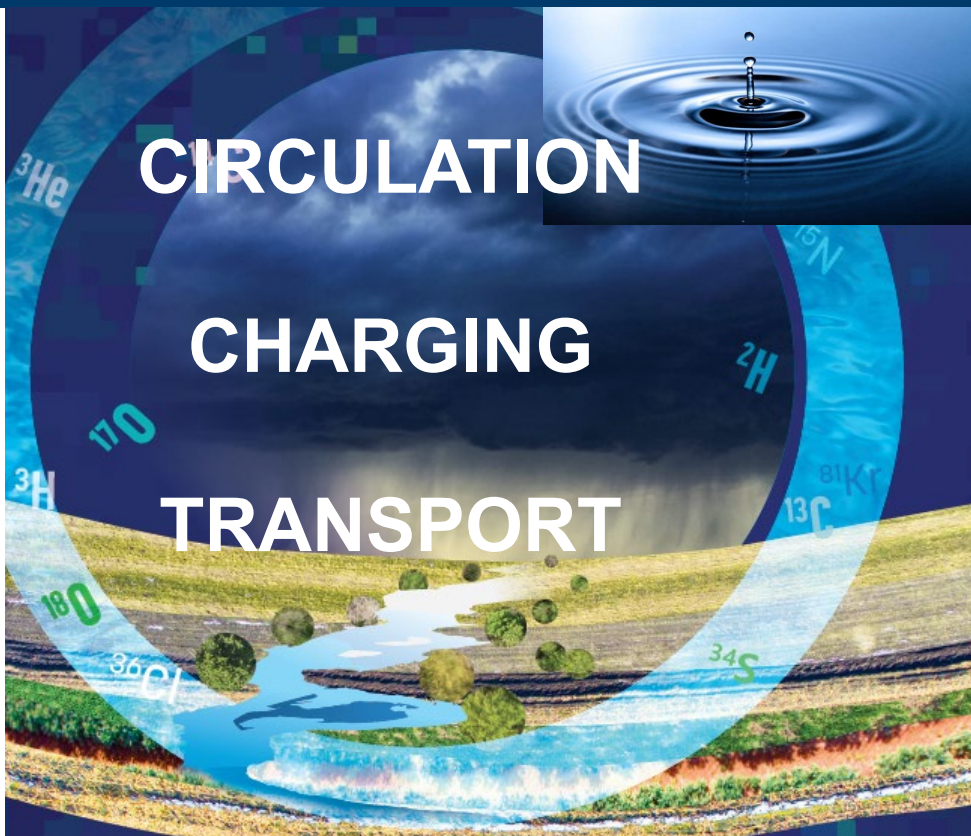


The key to good water management is knowledge: how much water we have, where it comes from, where it flows, what age

"young" water is easier to exploit it, "old" water is more precious and needs to be protected but young water is even more vulnerable

Isotopes around us

Hydrology studies the distribution and movements of groundwater and the influence that the geological environment has on their accumulation and circulation



Isotope hydrology uses stable and radioactive isotopes in water to trace its processes

ISOTOPES

STABLE

^2H ^{13}C ^{15}N ^{18}O

FRACTIONATION

TRACERS

origin/genesis of aquifers

RADIOACTIVE

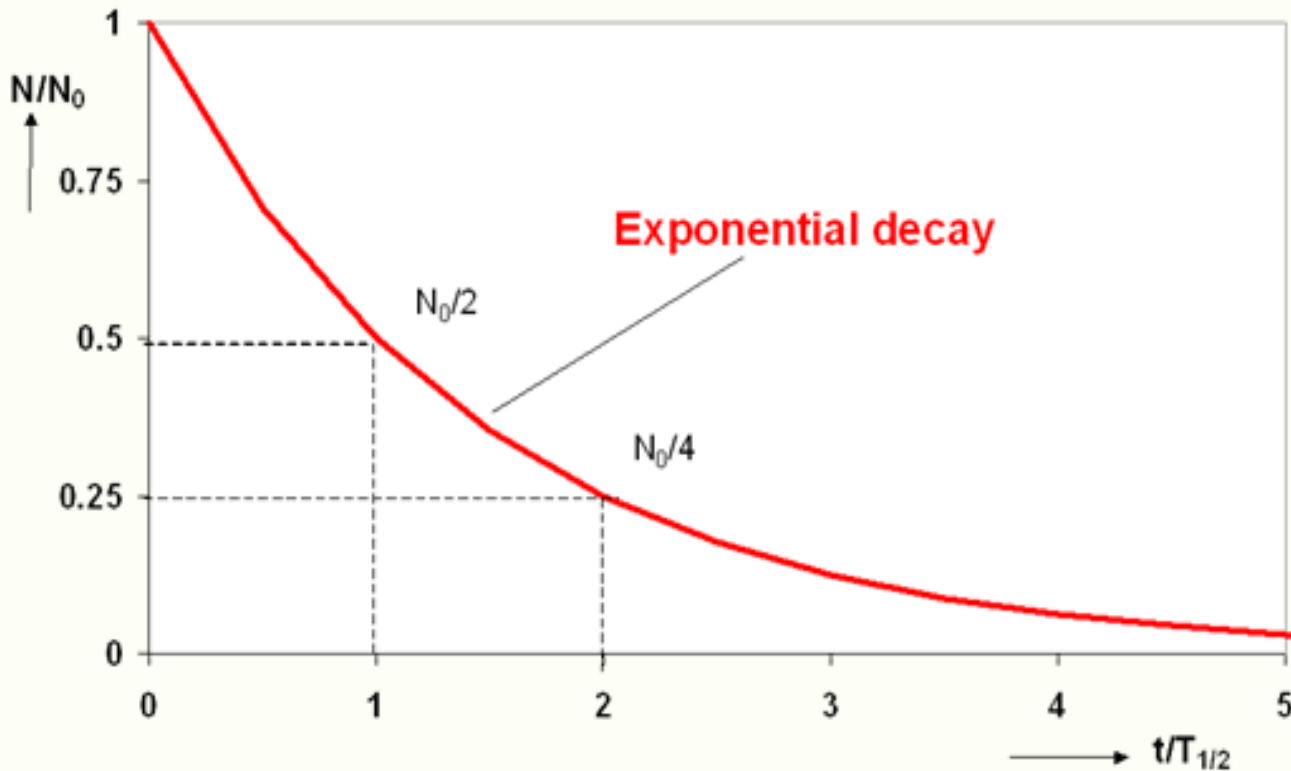
^3H ^{14}C ^{36}Cl ^{81}Kr

RADIOACTIVE DECAY

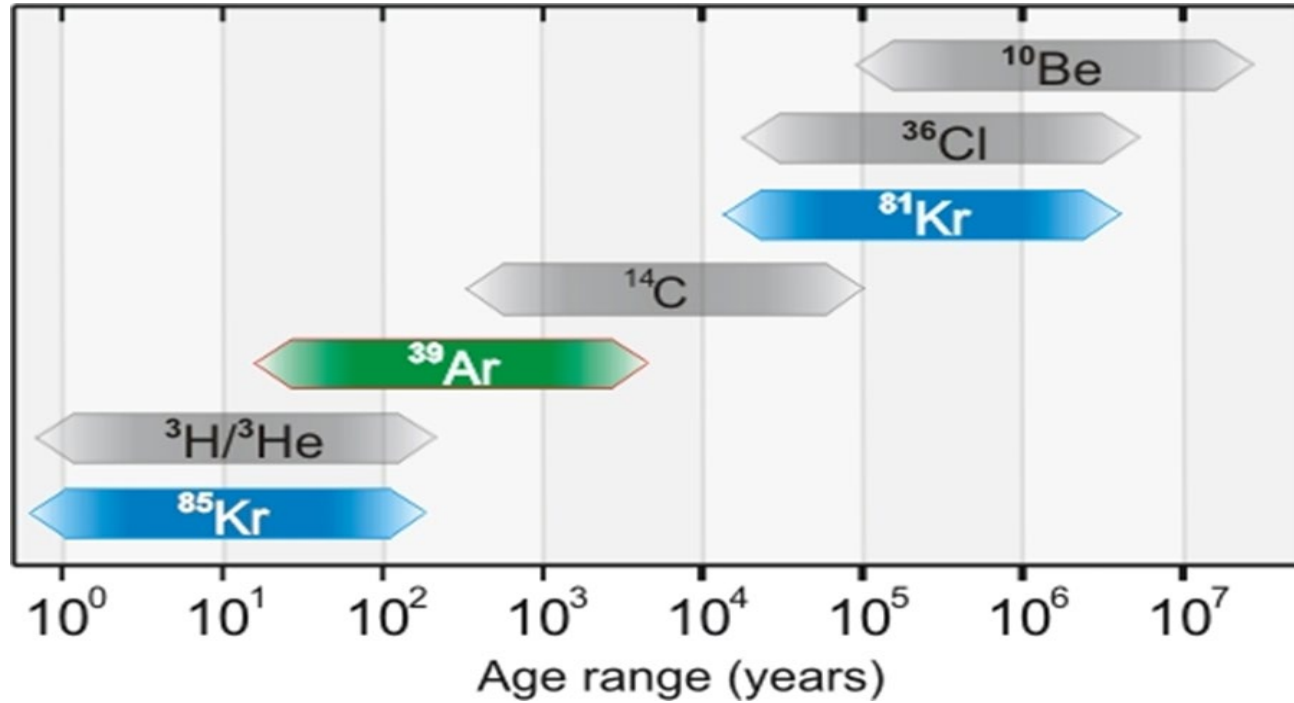
CLOCK

hydrogeological processes

Decay of radioactive isotopes



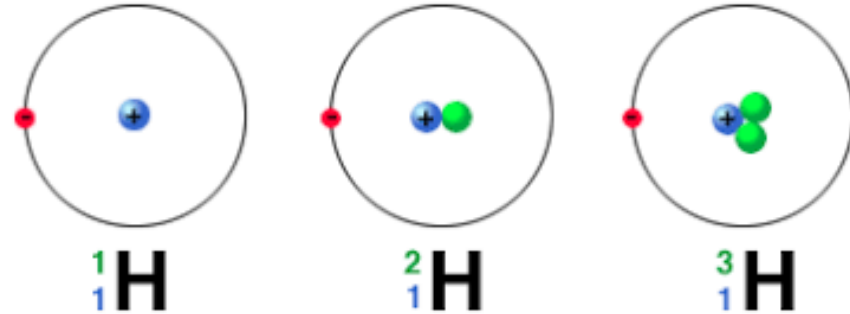
Radioactive isotopes - clock



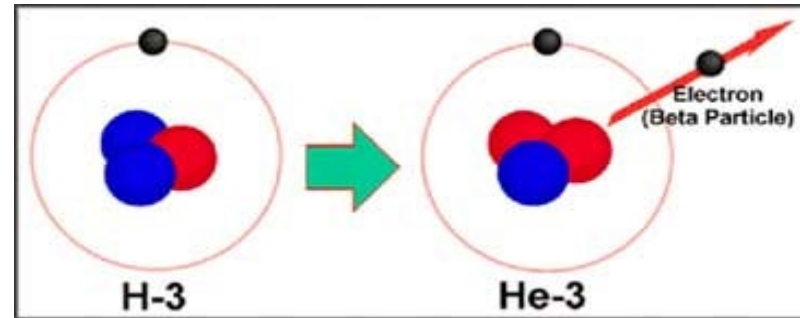
Tritium (^3H)



is an unstable isotope of the H element



which by emission of β -particles decays into a He atom (^3He)



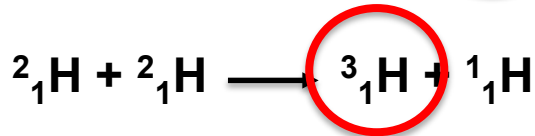
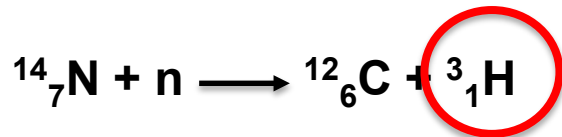
The half-life is approximately 4500 days (12.3 years)

Tritium (^3H)



It is an element mainly of cosmogenic origin:

it is generated naturally in the atmosphere by the interaction between cosmic ray and N or ^2H present in the stratosphere and upper troposphere

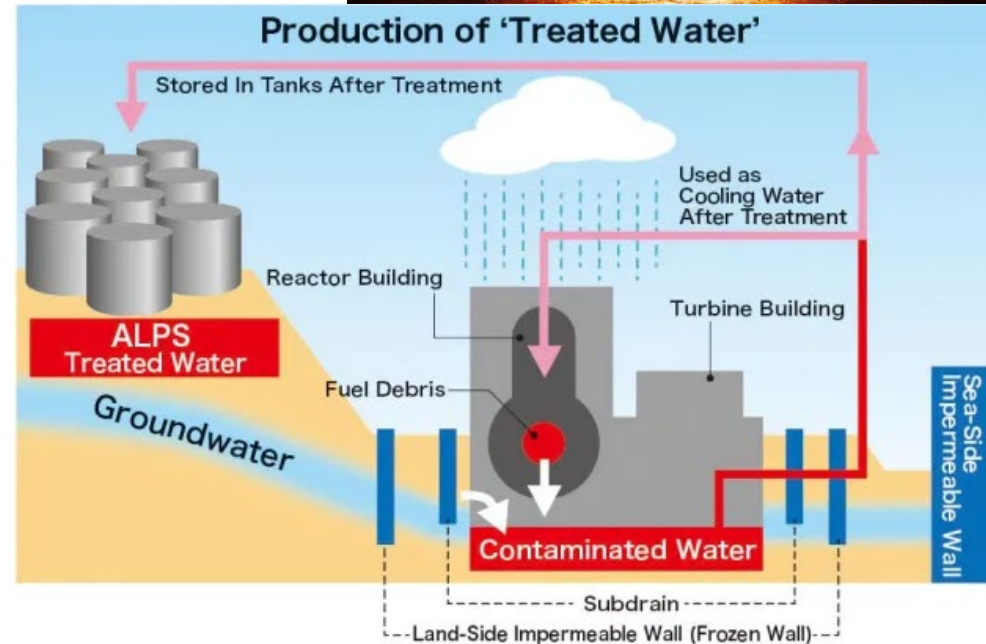


Tritium (^3H) anthropogenic

Nuclear tests from 1952 to 1963
and **nuclear power plants**.

These events changed the geochemical footprint of rainfall with significant increases stopped with the international agreement (CTBT) that banned all nuclear tests.

<https://www.ctbto.org/>



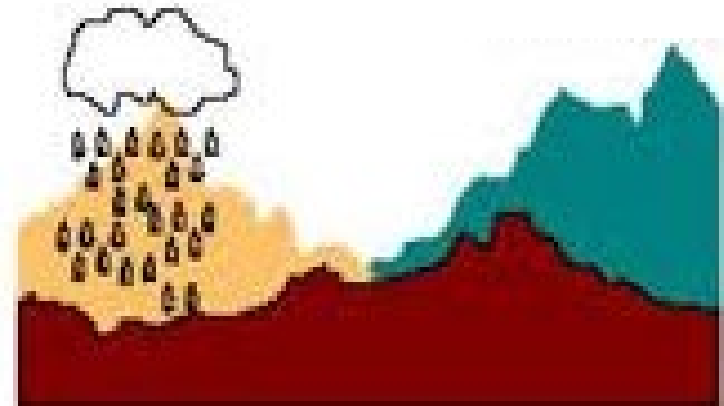
Tritium (^3H)



^3H combined with O present in the air produced tritiated water (HTO) in the atmosphere

- greater molecular mass than H_2O molecule (^3H is heavier than ^1H or ^2H)
- ↓
- shorter residence time in atmosphere because of precipitation.

*4 million curies (~ 0.41 kg)
produced per year*



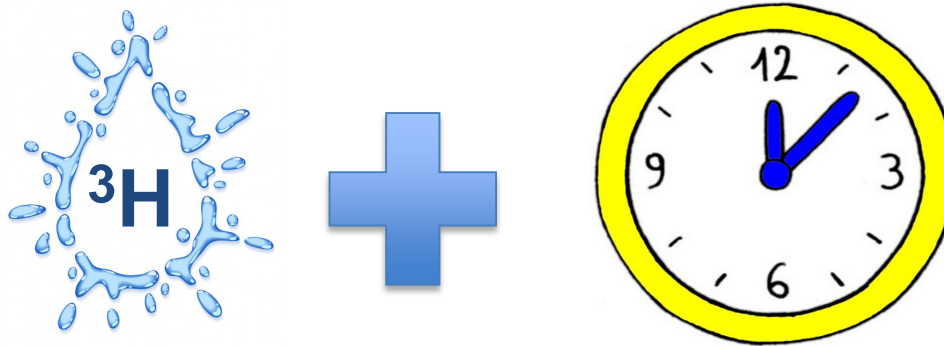
World inventory of natural tritium from cosmic ray interactions is approximately 70 million curies

Tritium (^3H)



The amount of ^3H in surface and groundwater depends:

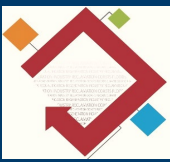
- on the amount of ^3H in the atmosphere when precipitation is formed
- on the time it takes for the water to go from the surface to the groundwater



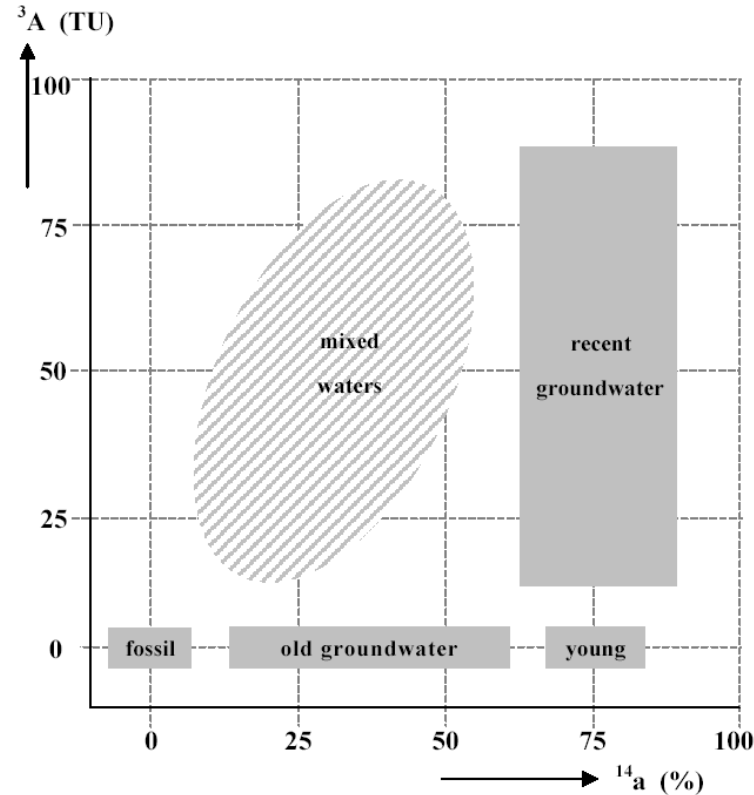
*short periods of residence in atmosphere + short period of decay
= ^3H concentrations in rains are low and almost constant ($\sim 70 \times 10^6$ Ci)*

(US Department of Energy 2002, Annual Report)

Tritium (^3H)



The decay of ^3H , e.g. in groundwater, produces its continuous loss which, in the absence of recharge (e.g. rainwater), causes a decrease.



^3H

^{14}C

^{36}Cl

RADIOACTIVE ISOTOPES

BOMB PEAK

TRITIUM IN PRECIPITATION

^3H concentrations of natural origin in the precipitations are $0.1 \div 0.6 \text{ Bq/l}$

^3H in precipitation increased 1000 times in N hemisphere

1950

1960

1970

1980

1990

2000

2010

2500

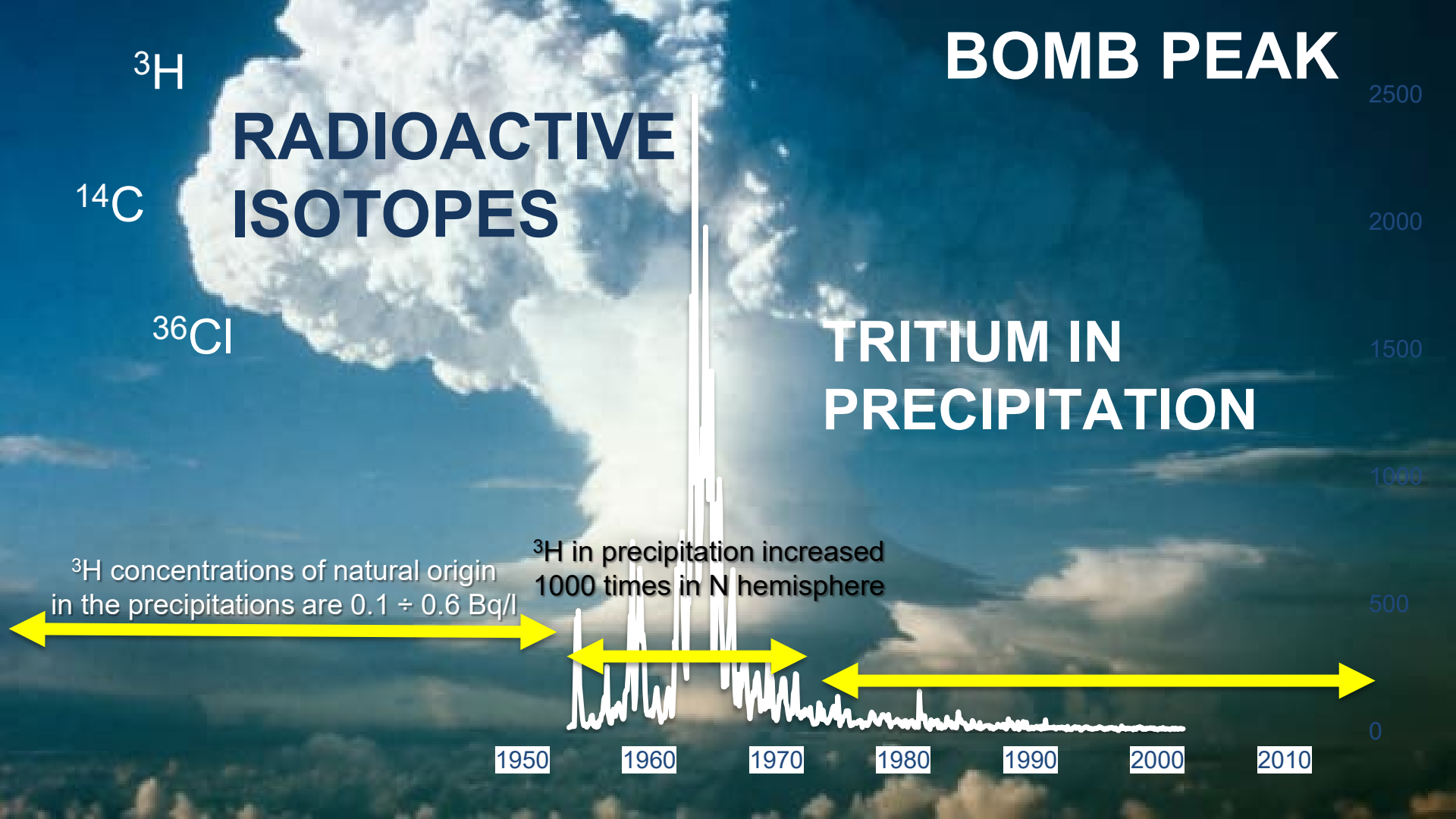
2000

1500

1000

500

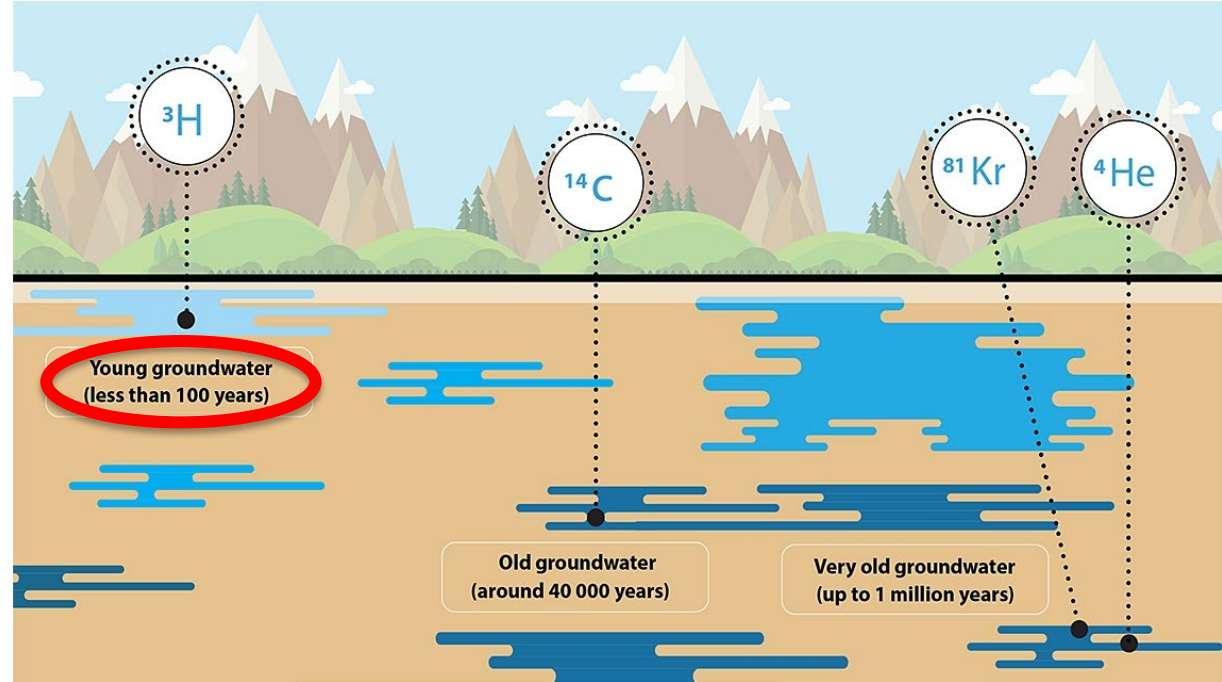
0



Tritium (^3H)



Due to the relatively short T1/2, groundwater infiltrated before 1950 does not contain ^3H .



Analyzing the concentrations of ^3H in groundwater it is possible to trace the age of the aquifer and / or define if the aquifer is polluted by anthropogenic activities.



The analysis of these phenomena requires good capacity of HTO measurements in concentrations $< \text{d.l.}$ (August, 2 2017 Decree)

This is possible only through a ^3H enrichment procedure and analysis with low-level liquid scintillation spectrometer



ENEA's Environmental Traceability and Radiometry Laboratory (FSN-SICNUC-TNMT)



Fusion and Technology for Nuclear Safety and Security Department



Nuclear Safety and Sustainability Division



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ENEA's Environmental Traceability and Radiometry Laboratory (FSN-SICNUC-TNMT)



TRACEABILITY LABORATORY
(www.tracciabilita.enea.it)
Scientific Manager: Antonietta Rizzo

ENEA's Environmental Traceability and Radiometry Laboratory (FSN-SICNUC-TNMT)



is at the forefront of the analysis of radioisotopes in the environment, including low-concentration of ^3H analyses (ISO 9698:1989)



**Brasimone
Research Center**



**Bologna
Research Center**

ENEA's Environmental Traceability and Radiometry Laboratory (FSN-SICNUC-TNMT)



ENEA's Environmental Traceability and Radiometry Laboratory (FSN-SICNUC-TNMT)



- The laboratory is equipped with
- electrolysis system consisting of 20 steel cells
 - cooling system
 - DC power supply
 - temperature control unit
 - three multiple distillation batteries for tritium samples
 - dryer for drying electrolytic cells
 - Glassware



ENEA's Environmental Traceability and Radiometry Laboratory (FSN-SICNUC-TNMT)



HTO sample mixed with scintillating liquid, so that the molecules excited by the radioactive emissions, returning to the stable level, emit by fluorescence a light radiation in the visible field.



- **3 trolleys** each containing 20 vials and a **sample elevator** that lifts the vial and places it in the measuring chamber
- several studies indicate that **Teflon vials** give the best results in terms of sample storage, interference with measurement, quality/price ratio.





QUANTULUS

low background instrument

that allows to carry out analyzes even in non-underground laboratories (not shielded from naturally radiation) thanks to **an active and a passive shielding**



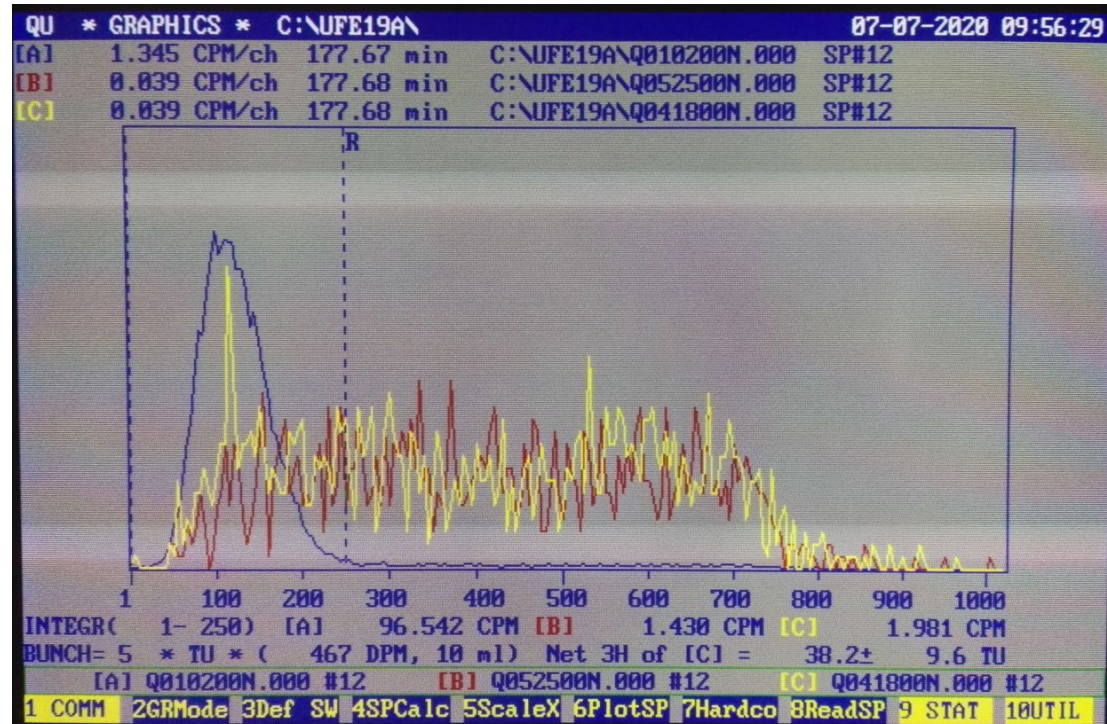
ENEA's Environmental Traceability and Radiometry Laboratory (FSN-SICNUC-TNMT)



The amplitude of the electrical pulse is converted into a digital value for each of the channels

(multi-channel analyzer covering an energy range from 0 to 2000 KeV)

CPM vs channel (energy)

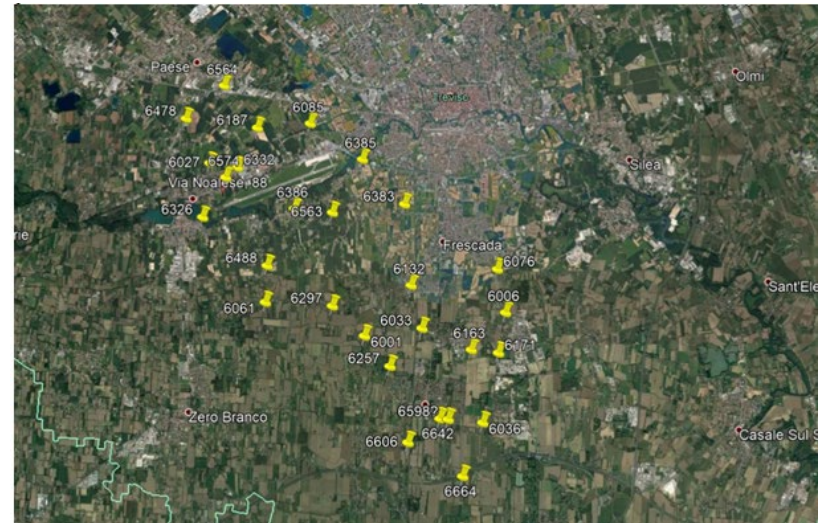
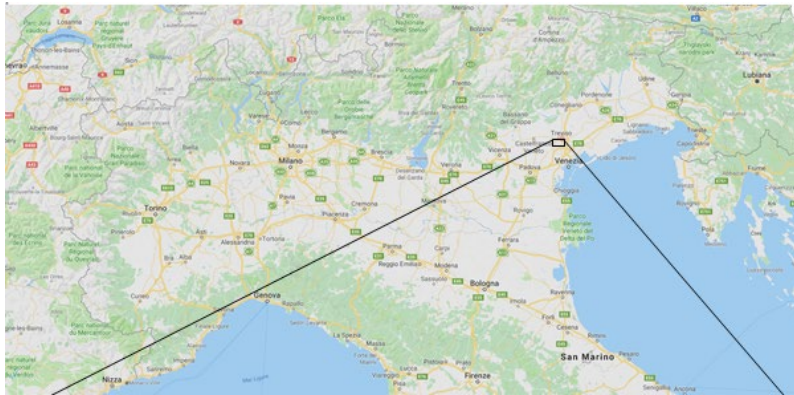


Treviso case study



Dep. Physics and Earth Sciences, Ferrara University + ARPAV Treviso

determination and evaluation of the concentrations
of ^3H in groundwater samples in the south-area of Treviso



Treviso case study

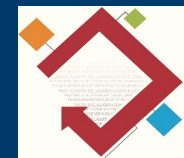


^3H activity	Age estimated
No ^3H	Long time of residence, recharge time > 70 years
$^3\text{H} < 0.8 \text{ TU}$	50 – 70 years
$0.8 < ^3\text{H} < 5 \text{ TU}$	10 – 50 years
$5 < ^3\text{H} < 7 \text{ TU}$	1 – 10 years (general value of rainwater)
$7 < ^3\text{H} < 15$	possible presence of external contributions
$^3\text{H} > 15 \text{ TU}$	evidence presence of external contributions

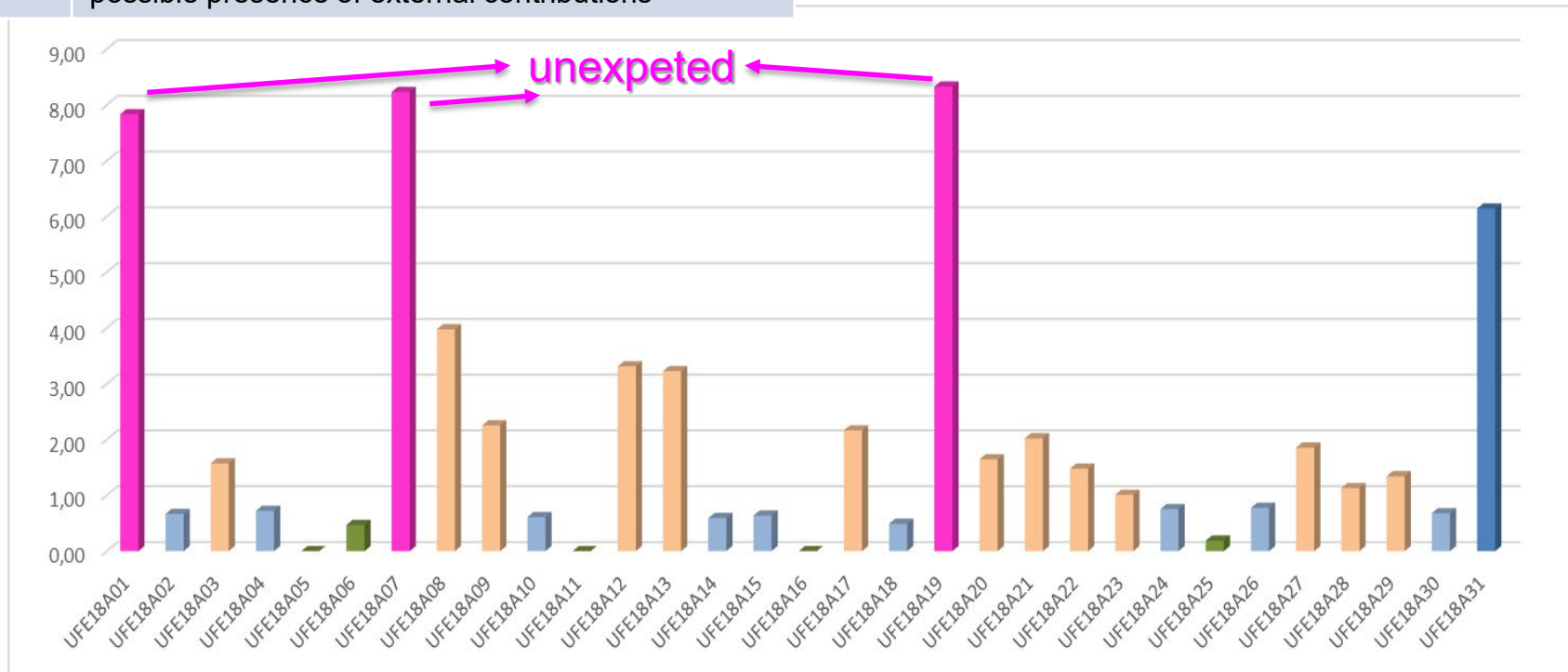
Clark e Fritz (1997)

1 TU = 1 ^3H atom
for 10^{18} H atoms
= 0.118 Bq/L

e.g. leachate from landfills hosting photoluminescent objects
or from hospital wastewater or biological laboratories



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*The samples show isotopic value
in accordance with the young age of water recharge*



- possible contributions of surface waters (climate change)
- differences in recharge, reflecting the change in permeability of gravelly sediments of High and Middle Plains compared to less permeable sediments of the transition area.

**Treviso
aquifer is
vulnerable**

ISOTOPES

AGE

GENESIS

RECHARGE SPEED

VULNERABILITY

The study of ^3H concentration in groundwater is a very useful tool for the determination of recent exchanges with surface water or the presence of recent anthropogenic contributions.



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Remtech EXPO – September, 24 2020 – Chiara Telloli (ENEA Bologna)



August, 2 2017 Decree

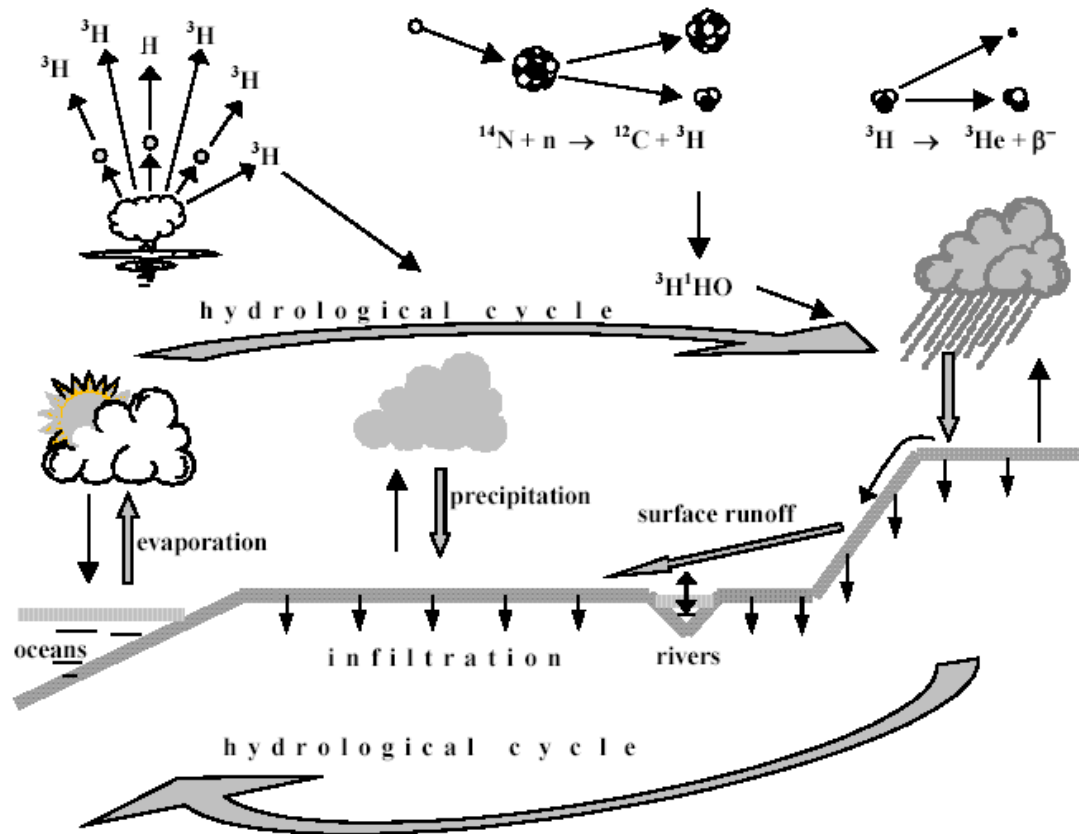
standardized the health protection requirements from the presence of radioactive substances in the waters intended for human consumption defined by Legislative Decree 28/2016

Legislative Decree 31/2001

provides for the obligation to verify the value of two parameters relating to the radioactivity content in drinking water:

- the concentration of tritium (< 850 UT)
- the total indicative dose, related to ingestion (< 0.1 mSv / year)

trizio



	^1H	^2H	^3H
Stabilità	stabile	stabile	radioattivo
Abbondanza naturale	0.99985	0.00015	$< 10^{-17}$
Attività specifica nat.			$< 1.2 \text{ Bq/L water}$
Decadimento/figlio			$\beta^+ / ^3\text{He}$
Tempo dimezz. (T/2)			$12.320 \pm 0.022 \text{ a}$
Cost. decadimento (λ)			$5.576 \times 10^{-2} / \text{a} = 1/18.33 \text{ a}^{-1}$
Max energia β			18 keV
Abb. ciclo idrog.		250‰	Da 0 to 10^{-16}
Report		$^2\delta$, $\delta^2\text{H}$ o δD	^3A
In		‰	TU, Bq/L H_2O $^3\text{H}/^1\text{H} = 10^{-18}$ (1 TU = 0.118 Bq/L)
Standard int.		VSMOW	NBS-SRM 4361
valore assoluto		$^2\text{H}/^1\text{H} = 0.00015575$	$^3\text{H}/^1\text{H} = 6600 \text{ TU}$