TRITIUM and the ENVIRONMENT

SOURCES

MEASUREMENT

and TRANSFER

Ph GUETAT, CEA

Thanks for their help to C Douche, JC Hubinois, N. Baglan ,

D Galeriu, Ph. Davis, W Raskob

SOURCES 1 - 1g = 0.358 PBq

- *Natural :*
  - 1300 PBq 3.5 kg at equilibrium
  - 72 PBq/an – 200 g/a -
- *Atmospheric nuclear tests*
  - 190 000 PBq north 420kg
  - 50 000 Pbq south 140kg remain 40kg 2007
- *Reprocessing Plants*

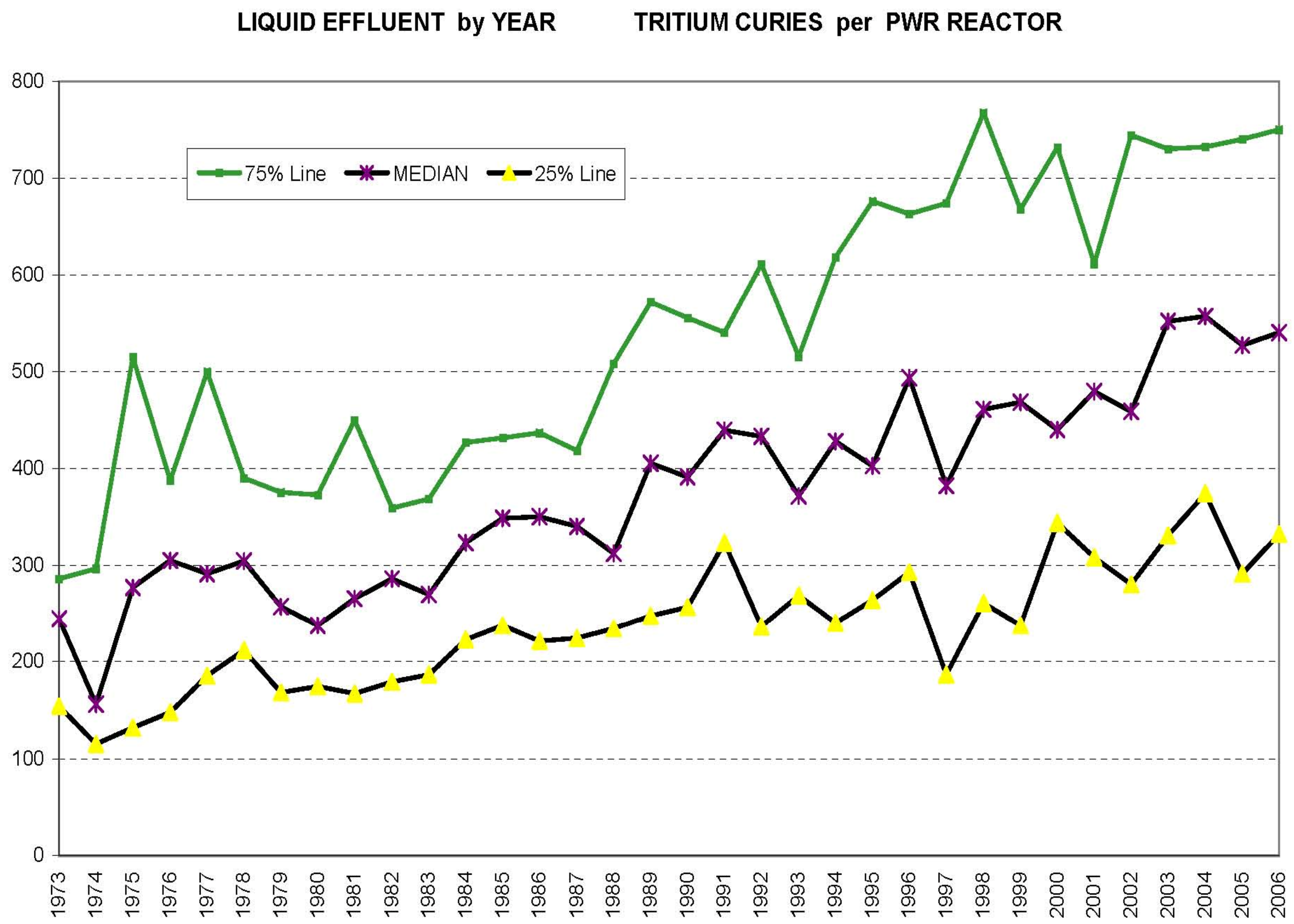
0,4 PBq.(GW.a)$^{-1}$ = 1 g.(GW.a)$^{-1}$ as liquid release

- La Hague - *sea* 10 PBq.an^{-1} 30g - *air* 0,07
- Sellafield - *sea* 2 to 3 PBq.an^{-1} 8g - *air* 0,6 to 0,2

SOURCES 2 - 1g = 0.358 PBq

- *Fission reactors releases* (TBq.a^{-1}. GW^{-1})

	Gas	liquid
– GCR	4	200-300
– HWR	100-1000 = 1g	100-500 = 1g


LIQUID EFFLUENT by YEAR
TRITIUM CURIES per PWR REACTOR
75% Line
MEDIAN
25% Line
0
100
200
300
400
500
600
700
800
1973
1974
1975
1976
1977
1978
1979
1980
1981
1982
1983
1984
1985
1986
1987
1988
1989
1990
1991
1992
1993
1994
1995
1996
1997
1998
1999
2000
2001
2002
2003
2004
2005
2006

SOURCES 3 - 1g = 0.358 PBq

- ***Industrial and Small users***
  - Amersham 0.5 PBq.a^{-1} before 2000 0.1 PBq.a^{-1}
  - R&D biology – labelled compounds : french stock 0.5 PBq 1g
    - Small amounts,
    - Incinerators; surface disposal - with^{14}C
  - Tritium lighting devices

EXIT – other paintings 0,1-0,5 TBq/device (1mg/device)

- ***Future users***
  - Laser (ie : LMJ) few tritium gas (mg.a^{-1})
  - Fusion reactors, use of : JET 20g - ITER 1,5 kg.an^{-1}

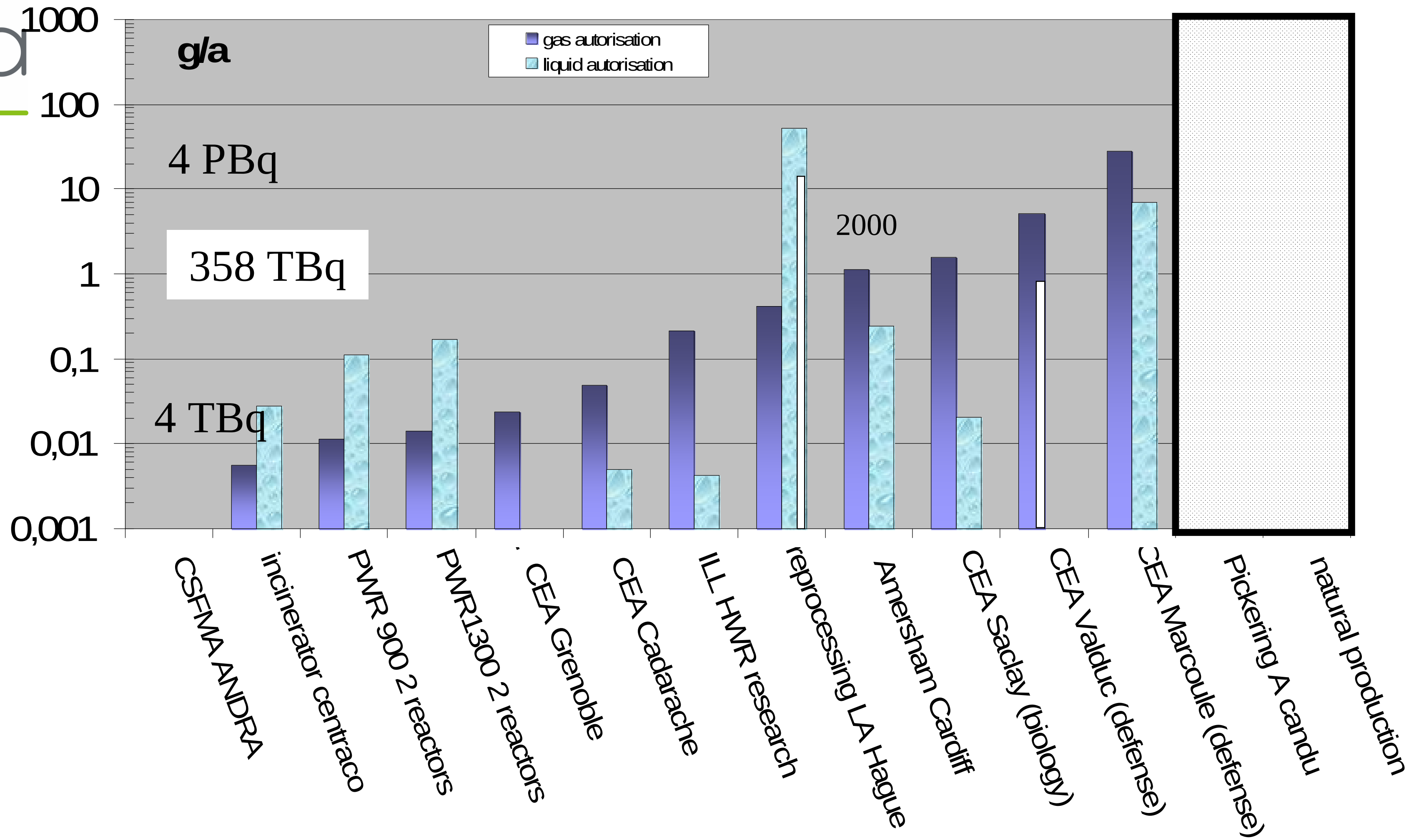
SOURCES 4 - 1g = 0.358 PBq

- *Waste and waste disposals*
  - – Sea dumping 1967-1982 : 20 PBq 60g
  - – In France very small amounts in surface disposal
    - CSM 9 PBq 30 g
    - CSFMA < 4 PBq <10 g
    - CSTFA VLLW (ANDRA) low acceptance criteria.
  - – Graphite : small outgassing (• 10^{-7} a^{-1}?)
    subsurface

    20 000 t - • 5 PBq (2007)
  - – Hulls & nozzles : 20 TBq / t

    small outgassing (< 10^{-6} a^{-1}) type B
  - – CEA tritium waste : 5PBq after treatment + storage
  - – Rods B_4C, sodium cold trap… storage treatment

AUTHORIZED RELEASE TRITIUM [gramme]



Production of tritiated releases and waste in the future

– Depending on :
    - Ø Conception of process
    - Ø Treatments of air and waste

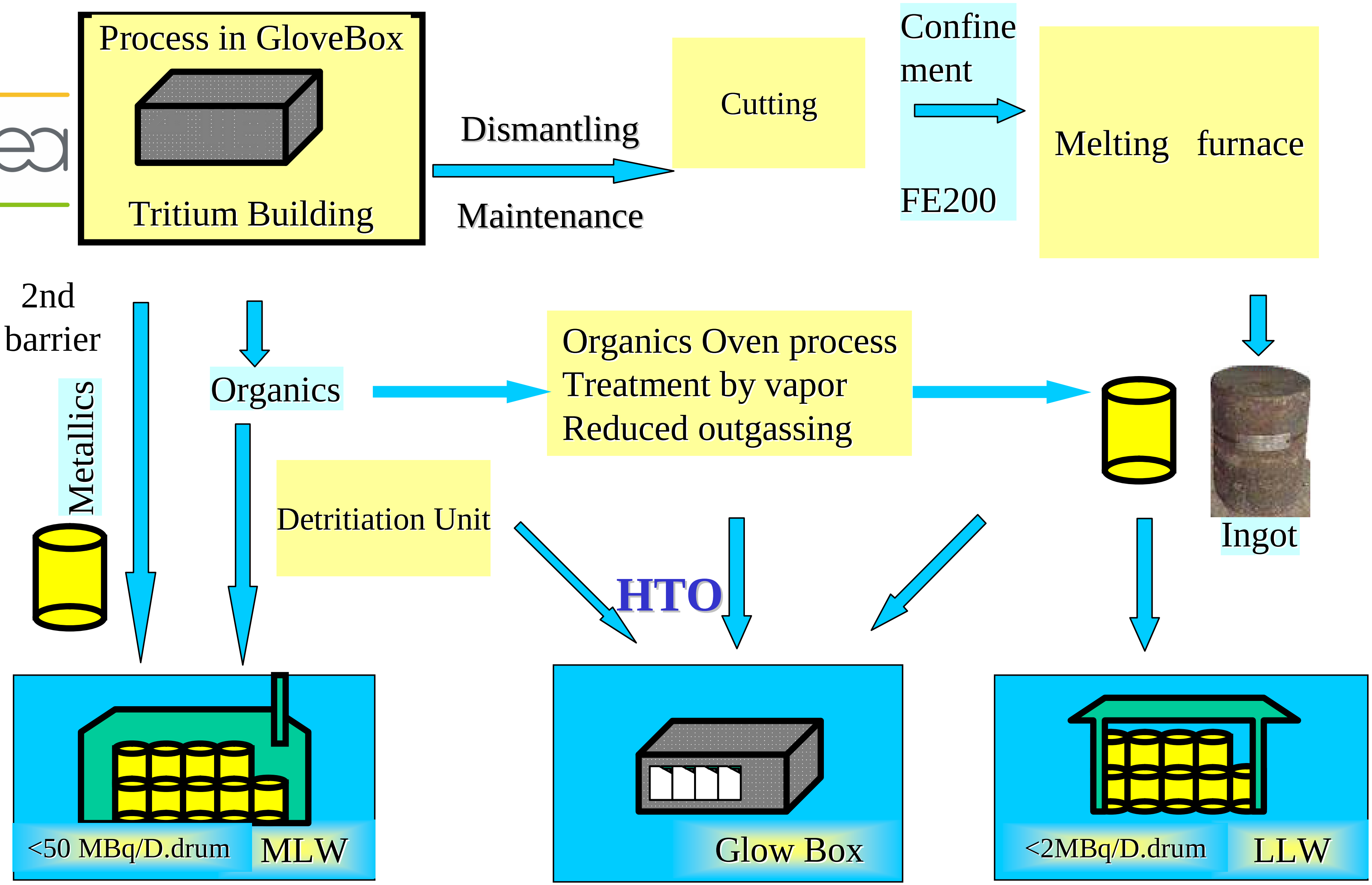
In Tritium buildings

- Avoid dilution
  - Limit volumes of air
  - Limit water vapor in air => dry air
- Limitation of leaks
- Detritiation of air - recycling

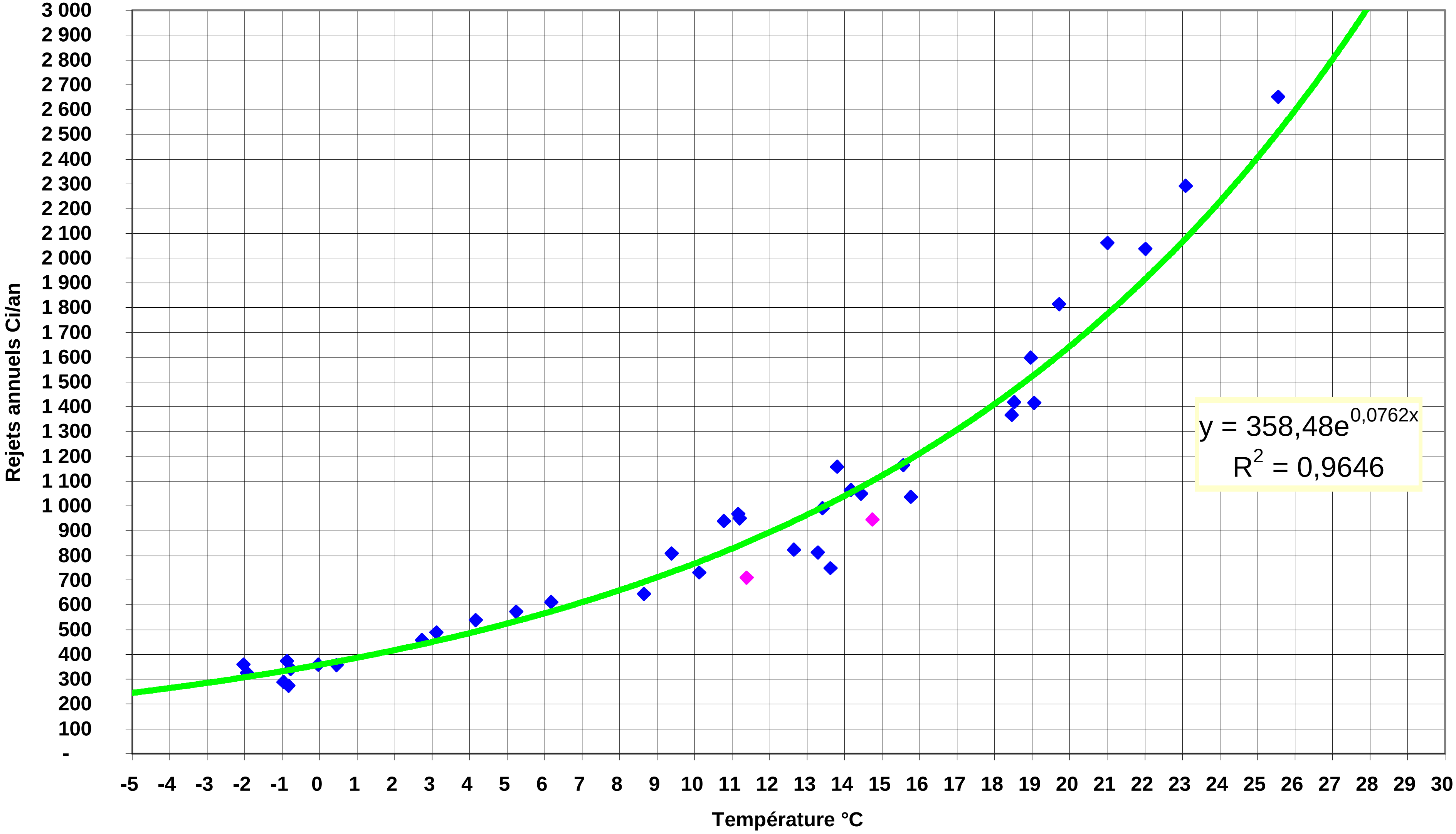
- Waste management a major source of Tritium release

- Detritiation of very low activity water not reasonable

Tritiated Solid waste treatment on VALDUC



TRITIUM RELEASE Ci/Year Fct (Temp. °C) : MA Waste Storage



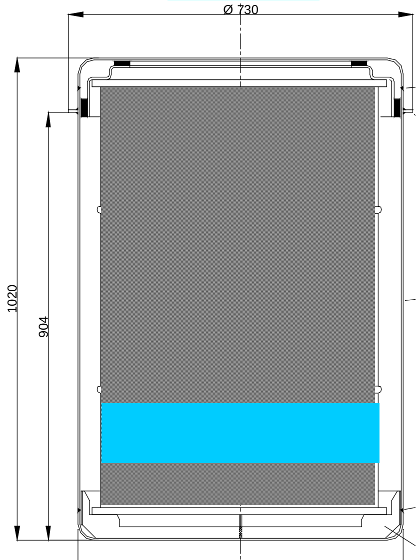
Container for tritiated waste

- Reversibility
- Confining
- Internal atmosphere Analysis

Drum 200 L

overdrums

RCDT2



EPICEA

FE200

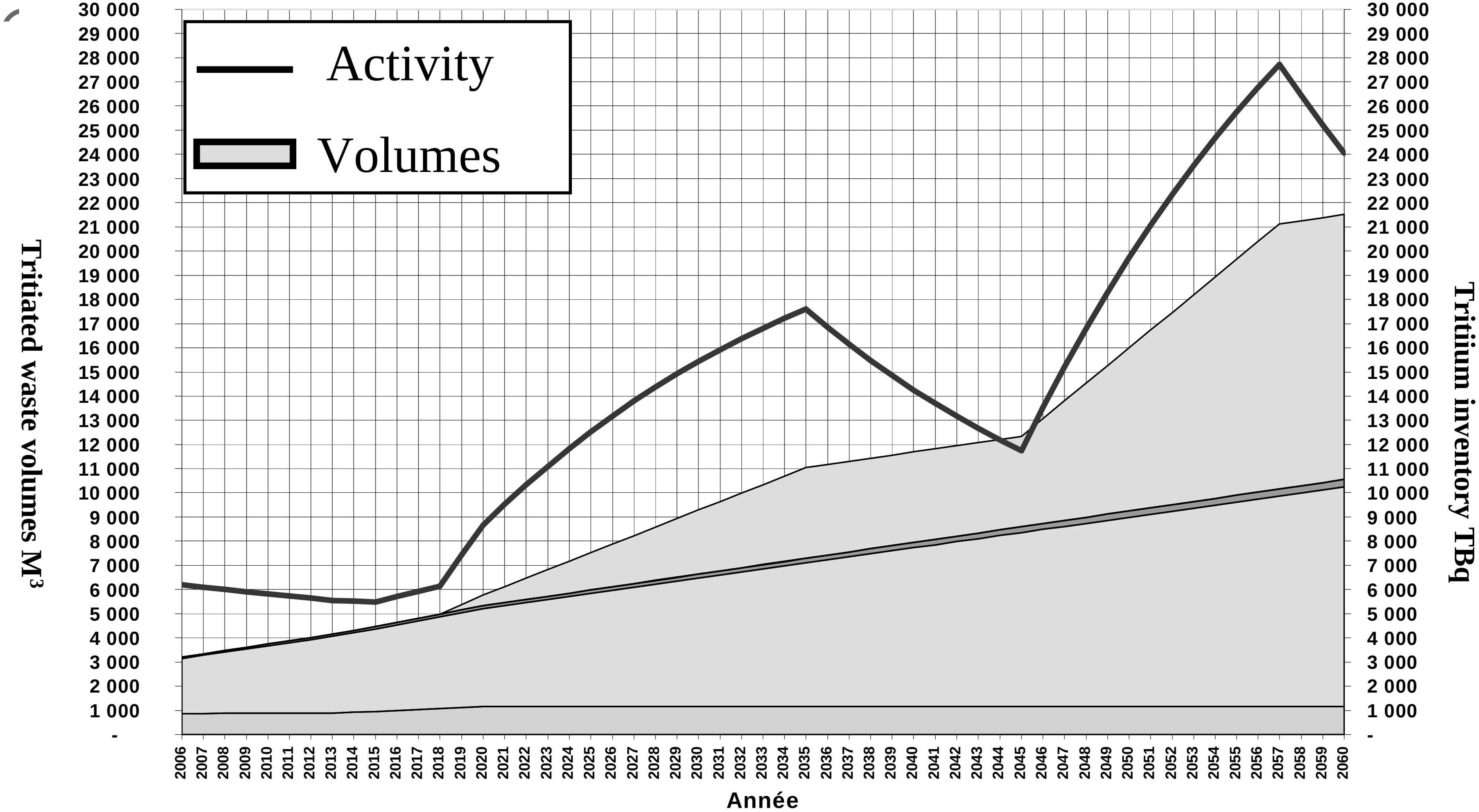
Drum 200 L	overdrums	RCDT2	EPICEA	FE200
elastomer joint	**Elastomer joint**	**welded**	**Metal joint**	
10^{-3} Pa.m³/s	**10^{-5} Pa.m³/s**	**10^{-9} Pa.m³/s**	**10^{-7} Pa.m³/s**	
60 €	**1 400 €**	**• 1 500 €**	**5 000 €**	**12 200 €**

NB. initial costs of the drum

Prospective cumulated volumes & Tritium activity of waste in France

1 - Small amounts of tritiated waste

2 – Program law N° 2006-739 28th juin 2006 - storage solutions…



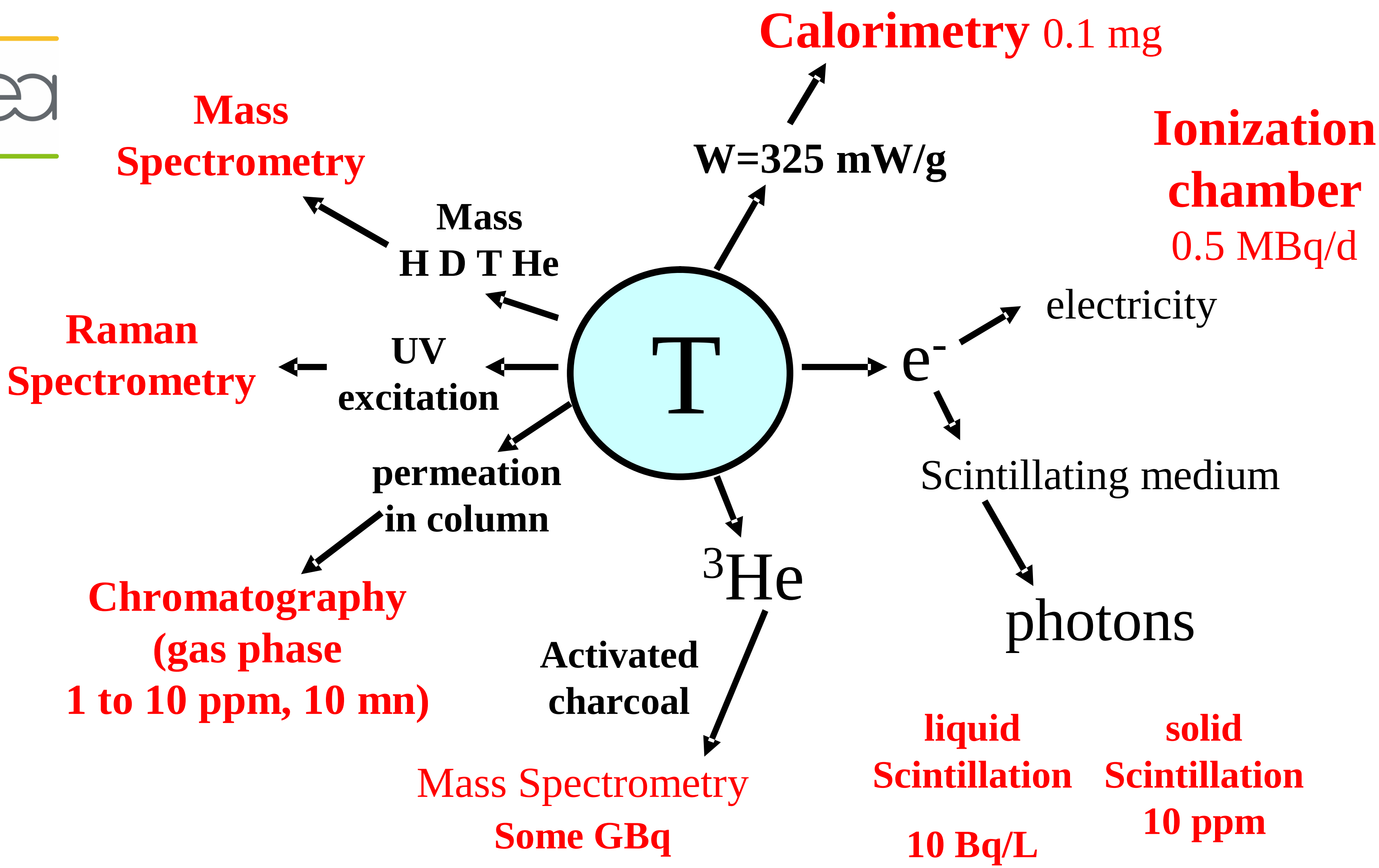
CONCLUSIONS for SOURCES

- Natural Tritium production is higher than man-made release
- Atmospheric nuclear test multiplied by 100 at world scale and more.
- Reprocessing plants = main sources (sea)
- Heavy water reactor produces much more Tritium (D-T) than PWR, and PWR (boron) more than BWR (ternary fission). T production increases
- Industrial use :
  - AMERSHAM : specific transfer for dissolved Organic Compounds in estuary environment
  - Lighting devices : in US landfills, many places >20000 pCi/L
- Waste :
  - Tritium in many types of waste : activity and outgassing
  - Very few in surface disposal in France.
  - Air tight containers are expensive.
- Fusion use : The use increases but no fundamental change for release
  what outgassing for waste ?

Measurement

- Preparation of sample depending on the physicochemical form
  - Gas : HT HTO other
  - Liquide (Water) : pure or not (distillation)
  - Solid : T_2 HT HTO OBT (E OBT and NE OBT)
    - Bio
    - Metal
    - Powder - hydrides

2 - Principles - What is measured ?



3 - Objectives of measurements

- **Inventories :**
    - Calorimetry
    - solid Scintillation
- **Releases :** ***alarm or precision***
    - Ionization chamber
    - liquid Scintillation
- **Isotopy, impurities**
    - Mass spectrometry
    - Gas chromatography
    - Raman spectrometry
- **Waste** : ***outgassing or inventory***
    - ionization chamber,
    - He3
    - Surface contamination, wipe test + scintillation
- **Environment :**
    - Liquid scintillation, electolytic enrichment

4 - Liquid scintillation

- Principle : A scintillating cocktail transforms • (e^-) in photons, use of gauging curves to determine efficiency of counting before tritium activity calculation

- Detection levels :
- Survey : 10 $Bq.L^{-1}$ 2 hours for measurement
- Very low level : 1 $Bq.L^{-1}$ NE OBT (1,5day/sample).

- Reference water : $A<0,2\ Bq.L^{-1}$

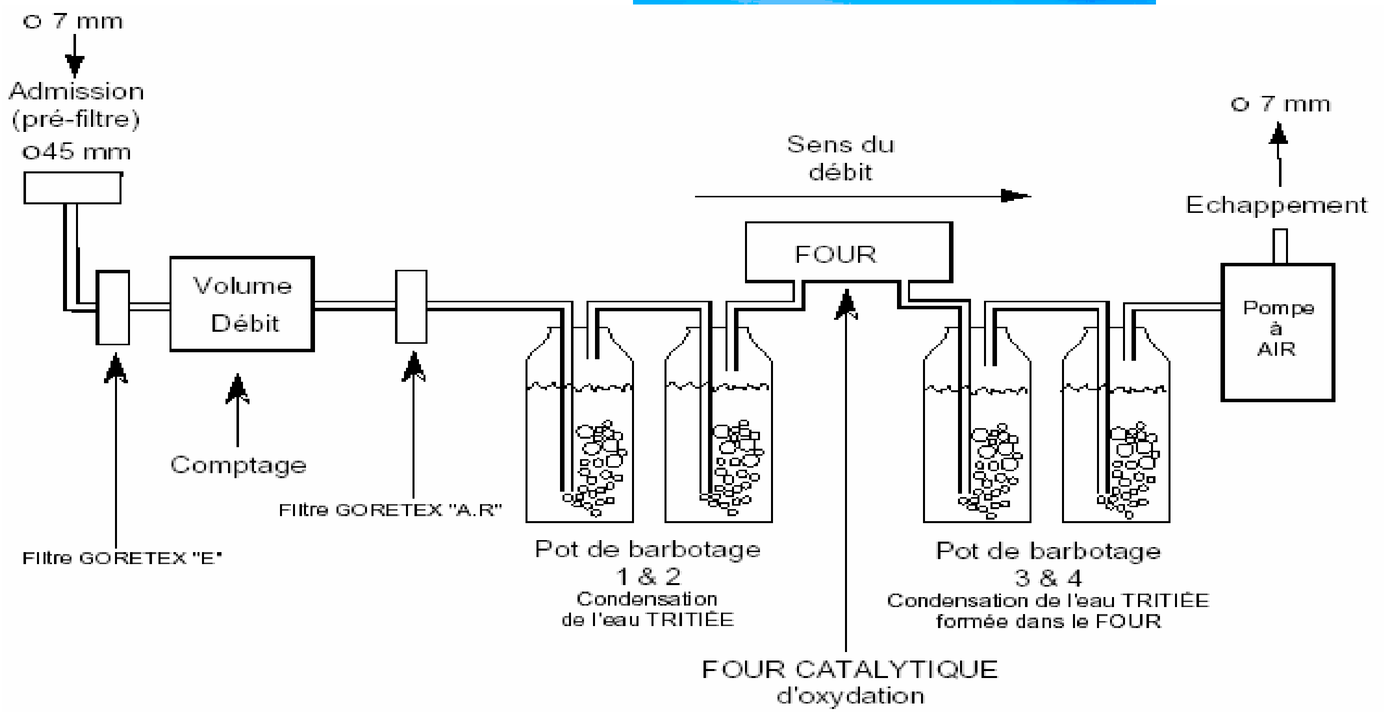
- Pure Water : no salt, no color, no other nuclides - or distillation

- Contamination problems
  - Controls for measure, distillation, lyophilisation, burning
  - Radon, C14…
  - Chemiluminescence
  - Static electricity

4.1 - Chemical Forms and sampling for tritium measurement in air

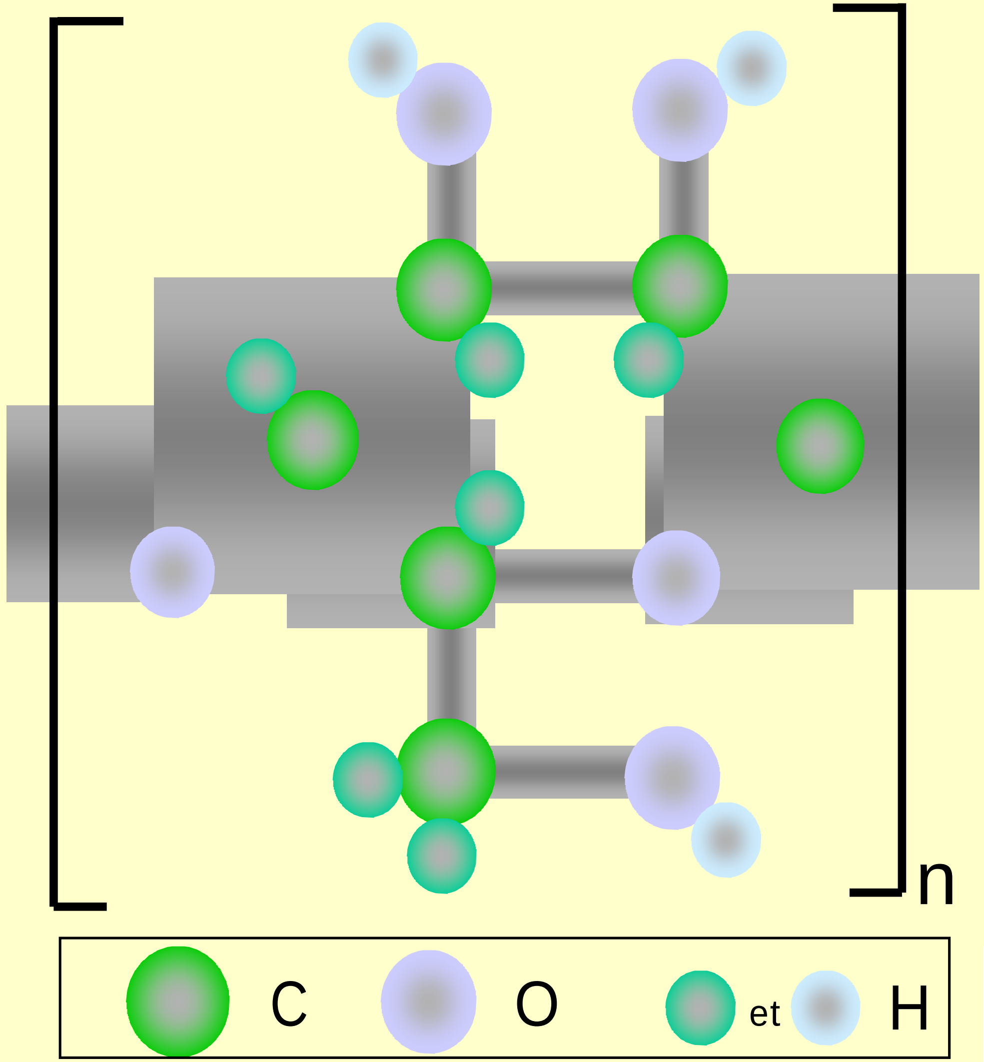
- Gas : tritiated water vapor HTO; hydrogen gas (HT) and tritiated methane.
  - sampling :
    - Active pump + bubbling

Le Marc 7000 ®



4-2 - Organically bound tritium : definition

Example of the cellulose molecule

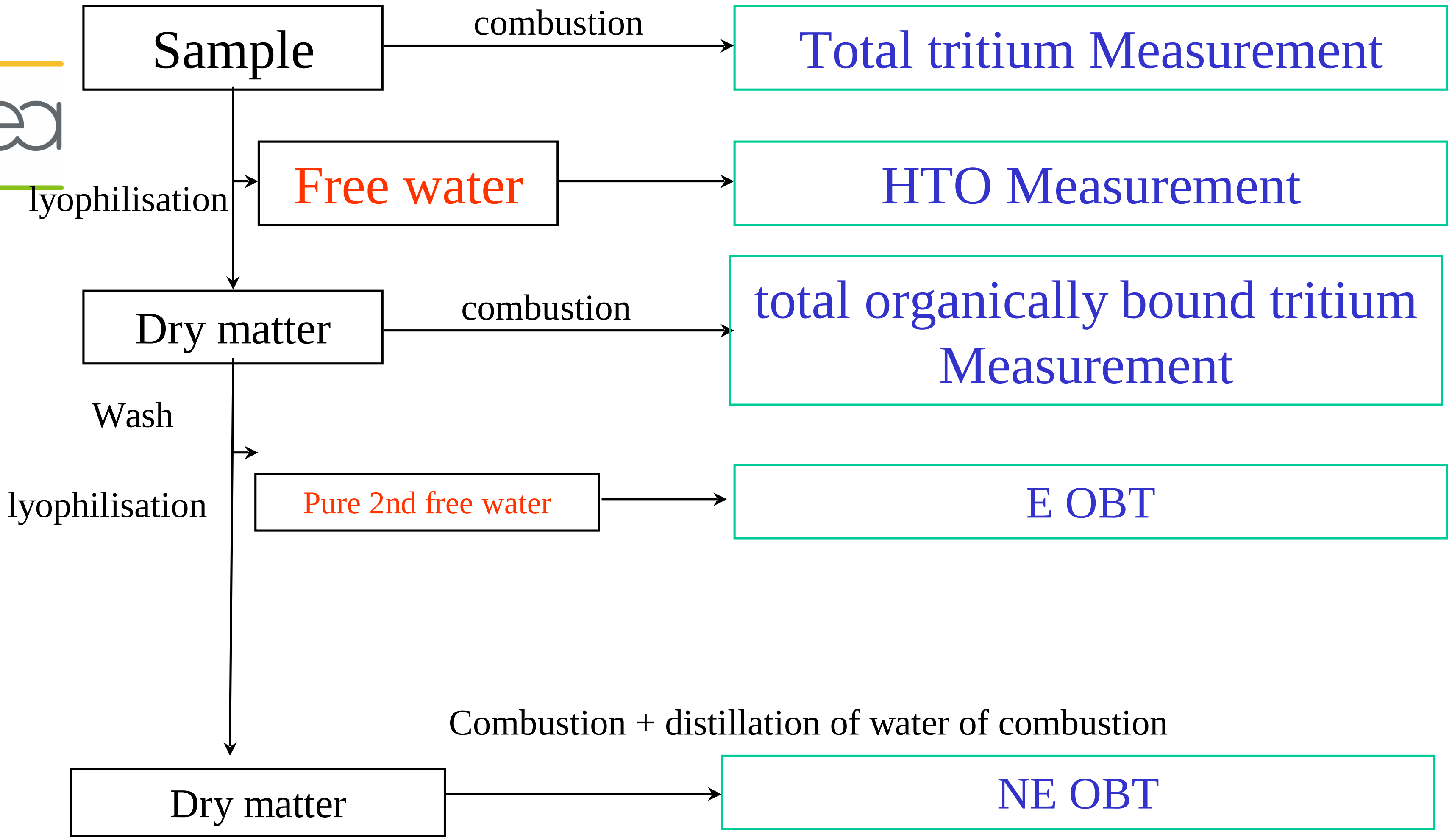


2 categories of hydrogen

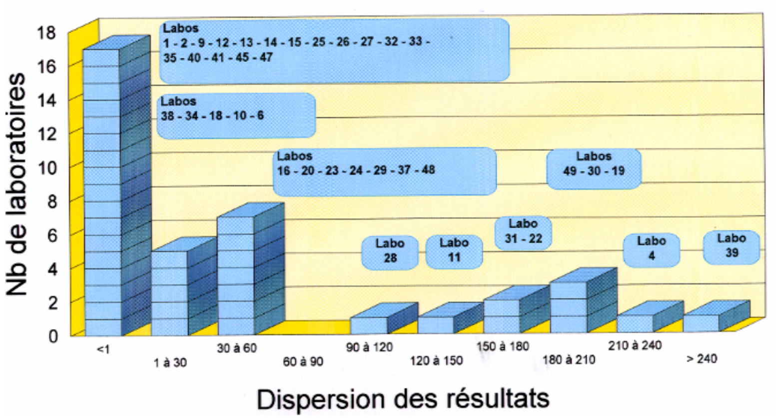
- Bound to O, N or S : weekly bound high capacity of exchange with H or T of environment = <u>exchangeable organic tritium</u>

- Bound to C : strong covalent bound, small capacity of exchange with H or T of environment
 Non Exchangeable organically bound <u>tritium</u>.

4-3 solid sample Preparation for measurement



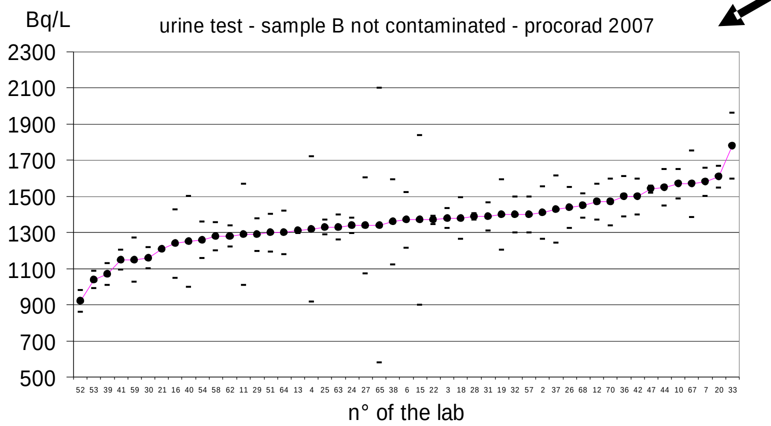
5 - Procorad intercomparisons

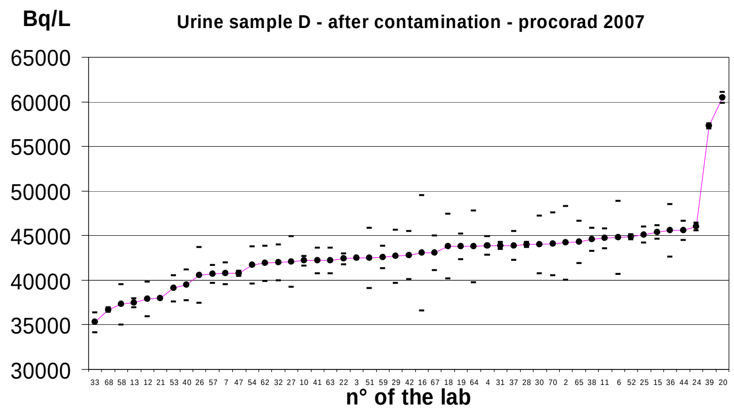


Procorad 1996

pollution

Procorad 2007





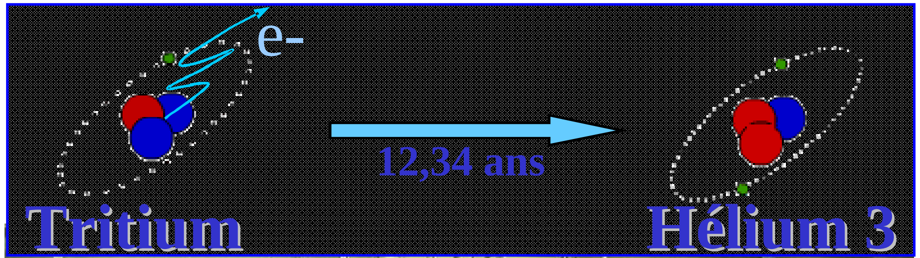
Ionisation Chamber

confining during 30 minutes

tritium out-gasing GCC
LD = 0,5 MBq/Day/Drum

Method He3

confining during some hours



Total activity LD = some GBq/drum

Conclusions for measurement

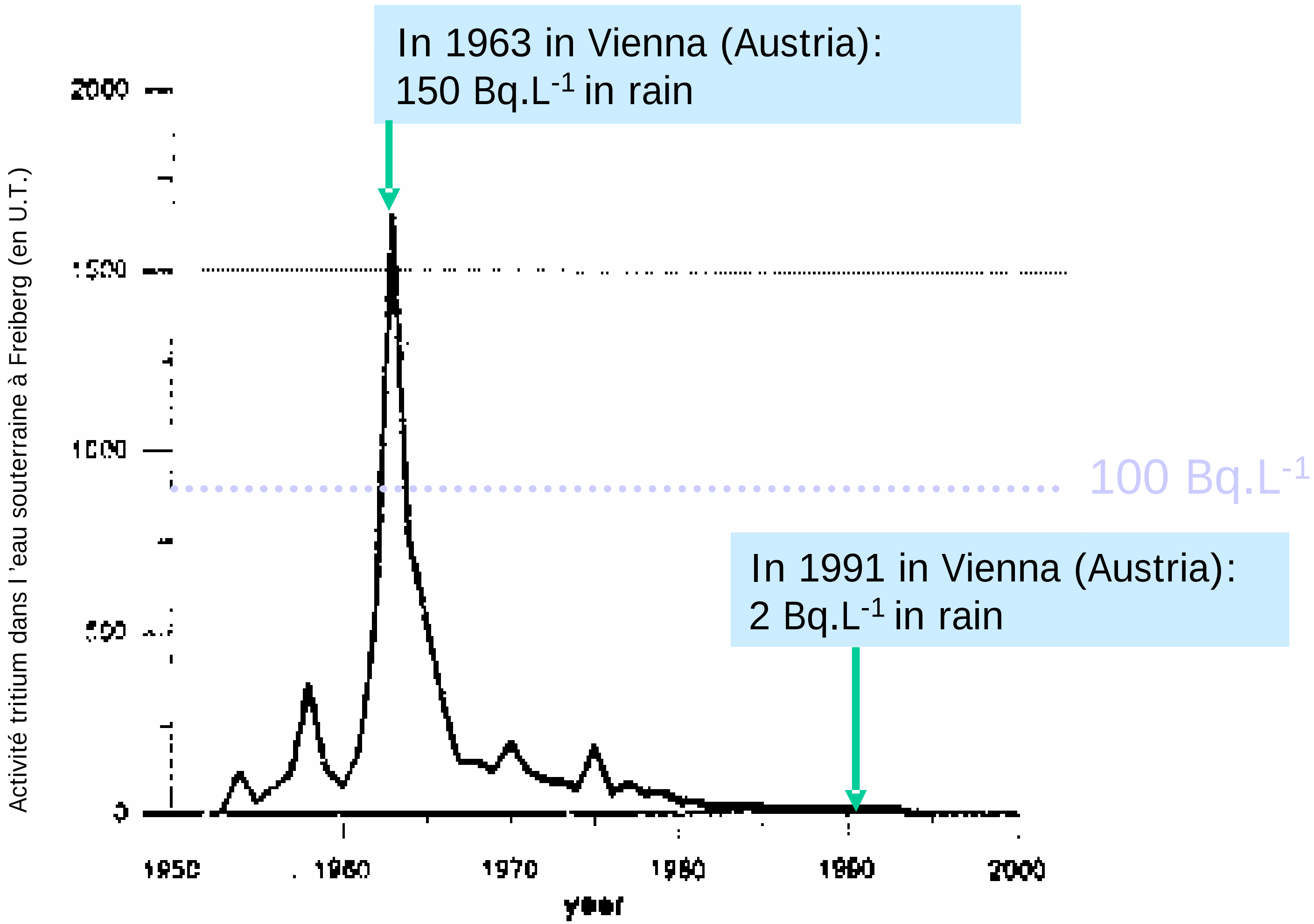
- Measurement apparatus for large inventories and large volumes to improve
- Easy to measure at a low level but need to have a good preparation
- Preparation takes time for solid material (case of incident ?)
- Not Easy to reach ANDRA's requirements for disposal

- LEVELS in ENVIRONMENT

10 –activities of rain and surface waters in the past

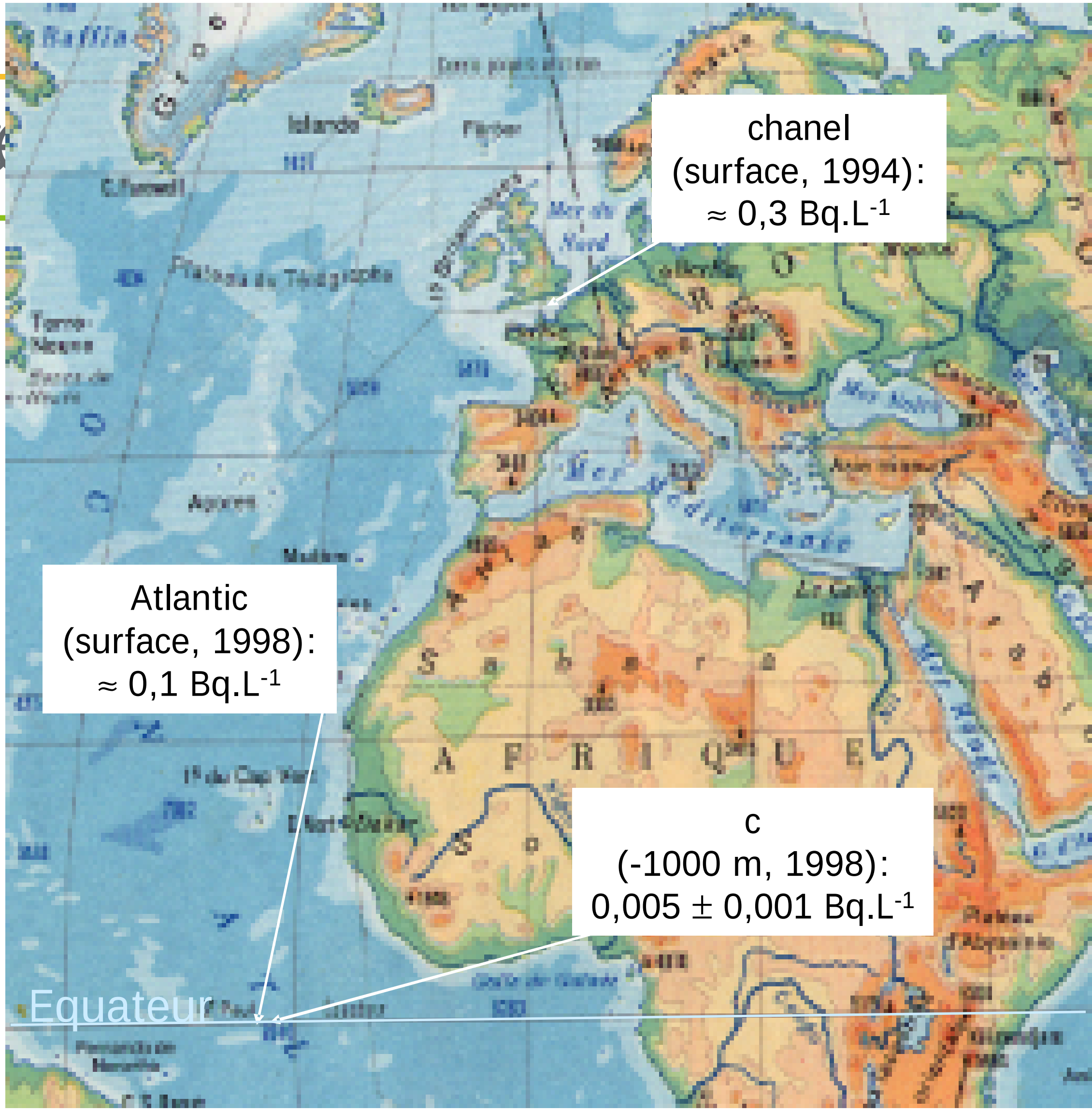
Effet des essais nucléaires atmosphériques

1 U.T.
(Tritium Unit)
= 0,118 Bq.L^{-1}



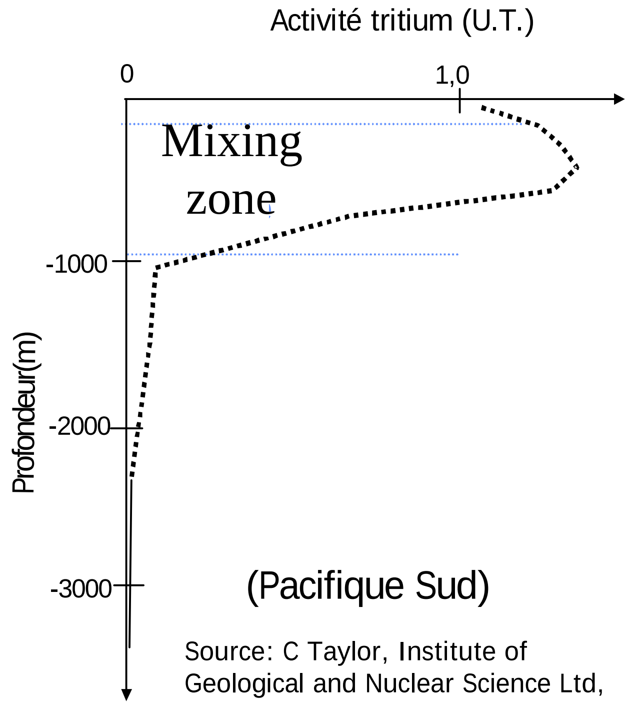
Référence : O. Nitzsche, D. Hebert, proc. Lowrad, p.106, 1996.

11. Distribution : sea and oceans



Repartition of T :
- 87% in oceans,
- 12% in continental waters,
- 1% in l atmosphere (HT et HTO)

Evolution of tritium activity versus depth:



Source: C Taylor, Institute of Geological and Nuclear Science Ltd, Lower Hutt, New Zealand (1997).

- Tritium transfer in environment
  - Continuous release **TRS 364**
  - Accidental release

Inhalation + transcutaneous : few mn- h

Air-vegetable contamination HTO : few h - days

Air-vegetable OBT : few weeks

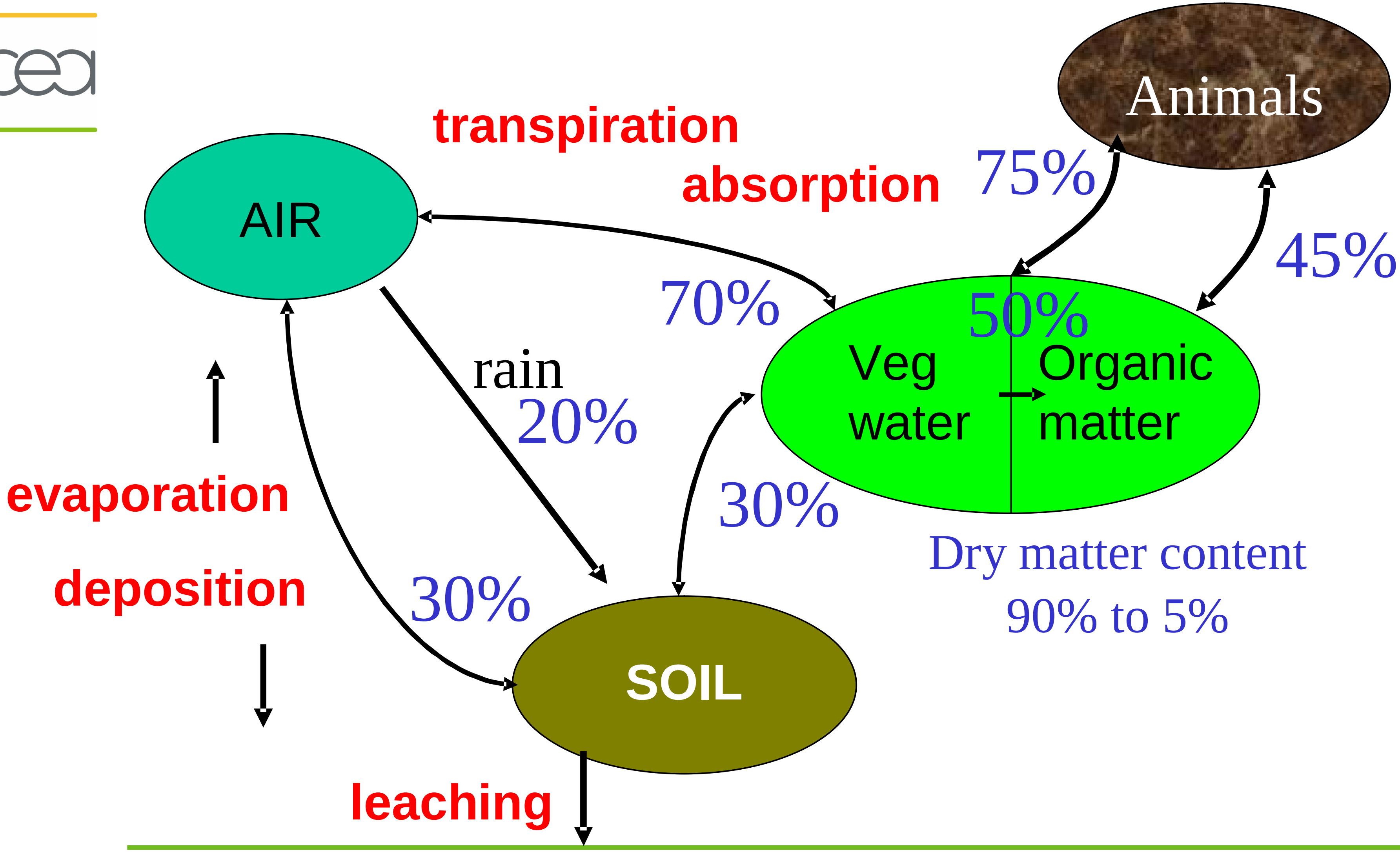
Soil (to reach equilibrium with air) : few months

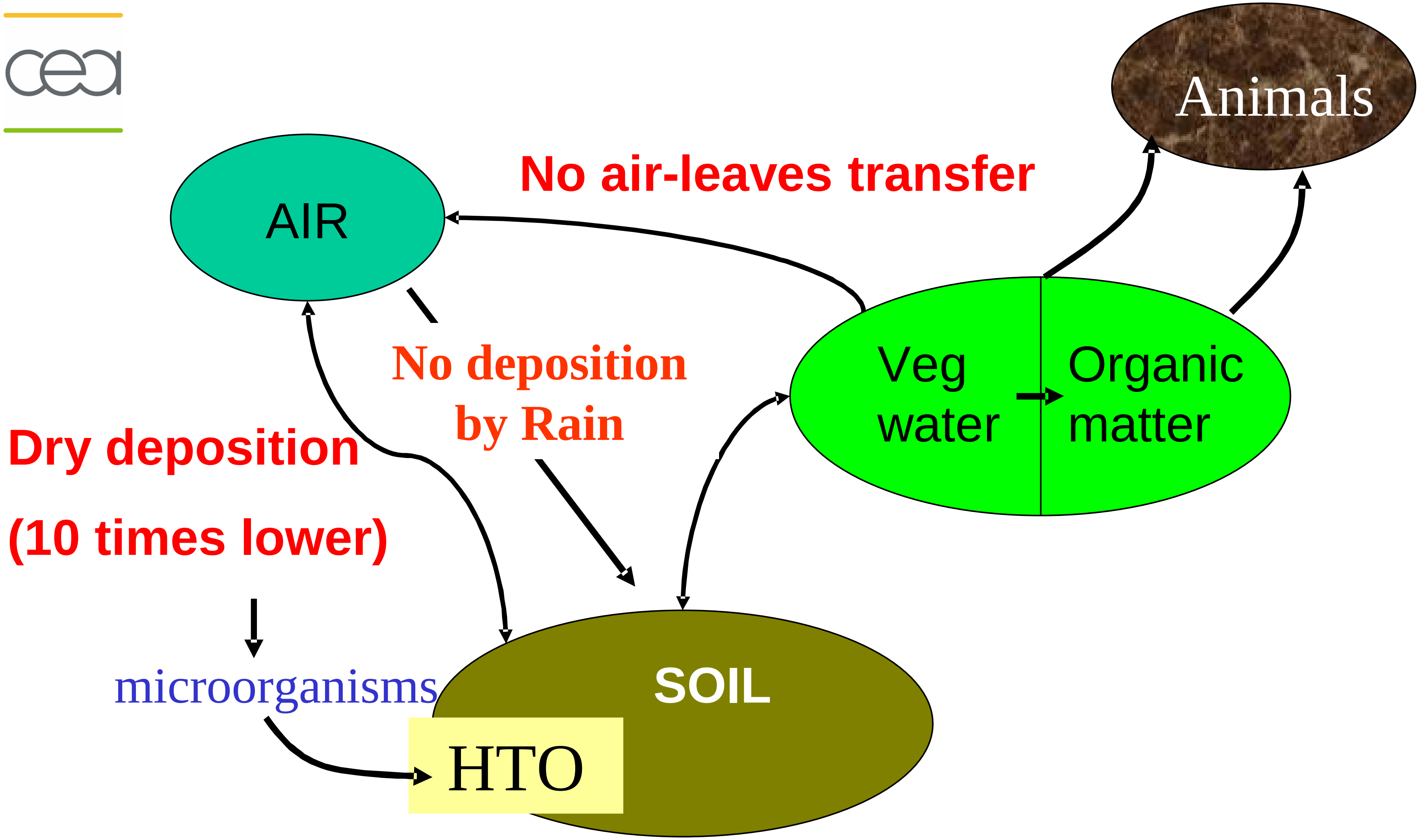
- HTO and OBT in vegetable from soil water : in equilibrium
- Vegetable HTO in equilibrium with soil water within 2 days

Animal products : more or less in equilibrium with fodder – Grass : few weeks of delay

A global IDEA of the tritium (HTO) pseudo equilibrium

« X% = order of magnitude of water concentration ratio »

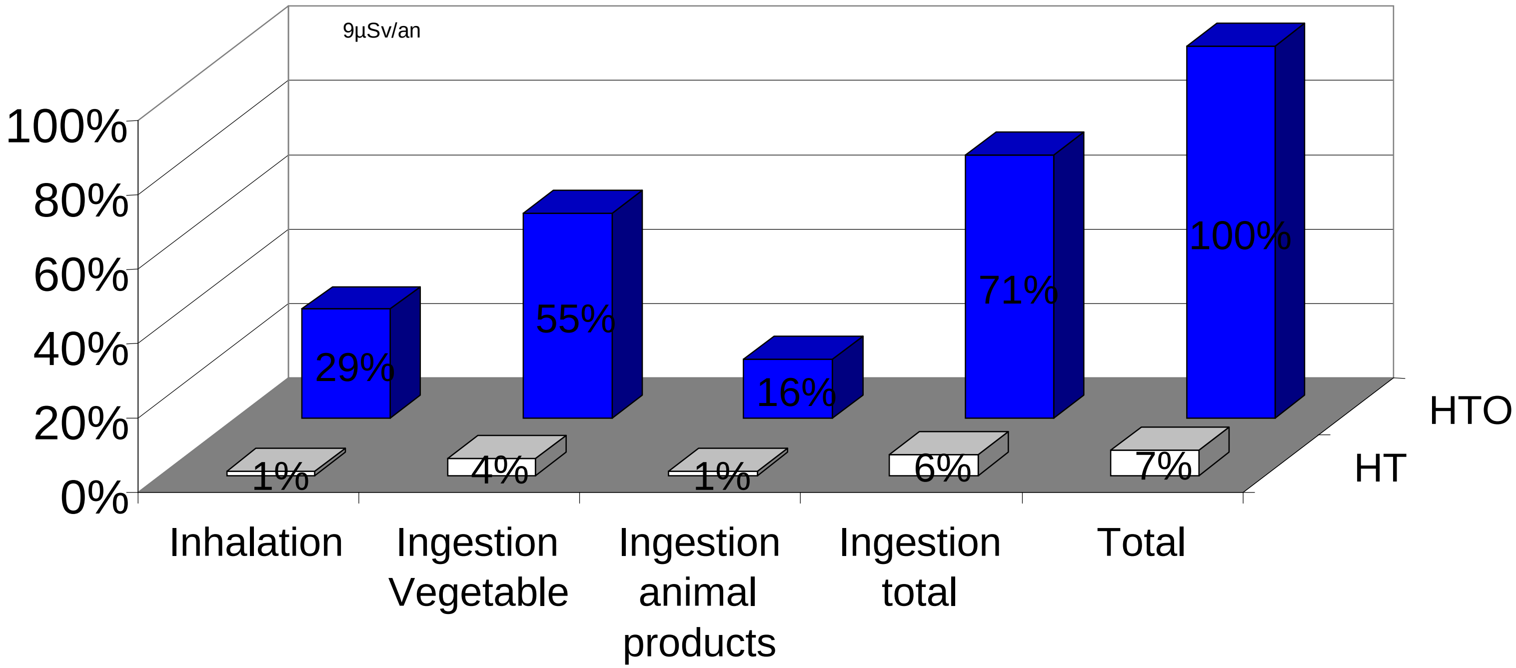



Animals
No air-leaves transfer
AIR
No deposition
by Rain
Veg
water
Organic
matter
Dry deposition
(10 times lower)
microorganisms
SOIL
HTO

HTO vs. HT Predicted Dose

10g =370 TBq/y; **chronic**; DCART

$\chi/Q=1\times10^{-6}$ s.m^{-3} total = 9 µSv



For similar source terms, an HT release has a 1-10 % dose impact compared with HTO

Water equivalent factor

1 kg **100g** **0.9L**

Fresh weight =dry weight + water

		2 to 18	
Protein	7 % H	=>	63% H_2O
Fat	12 % H	=>	108%
Carbohydrate	6 % H	=>	54%

Weq • 50% - 60%

Final assessment for dose assessment

$$C^{total}_{plant.fw} = [K_{air} + K_{soil})].[K_{OBT} + K_{HTO}].C_{air.w}$$

$$C^{total}_{plant.fw} = \boxed{[H_{r\,air}} + \boxed{0.3(1 - H_{r\,air})]}.\boxed{[(1 - H_{plant}).W_{eq}.D_p} + \boxed{H_{plant}}].C_{air.w}$$

$$C^{total}_{plant.fw} = [H_{air} + 0.3(1 - H_{air})]C_{air.w}.[(1 - H_{pl}).0.3 + H_{pl}]$$

$$C_{green\ veg} = (0.7_{air} + 0.09_{soil}).(0.06_{obt} + 0.8_{HTO})\ C_{air.w}$$

$$C_{grain\ cereal} = (0.7_{air} + 0.09_{soil}).(0.24_{obt} + 0.2_{HTO})\ C_{air.w}$$

Conclusions for normal releases

- IAEA document TRS 364 – 2008 and corresponding tecdoc– an up-to-date synthesis for normal tritium release assessment.

- For HTO :
  - At each step of the transfer chain, dilution occures
  - Vegetable Ingestion is clearly dominant
  - Direct Air-plant pathway dominant

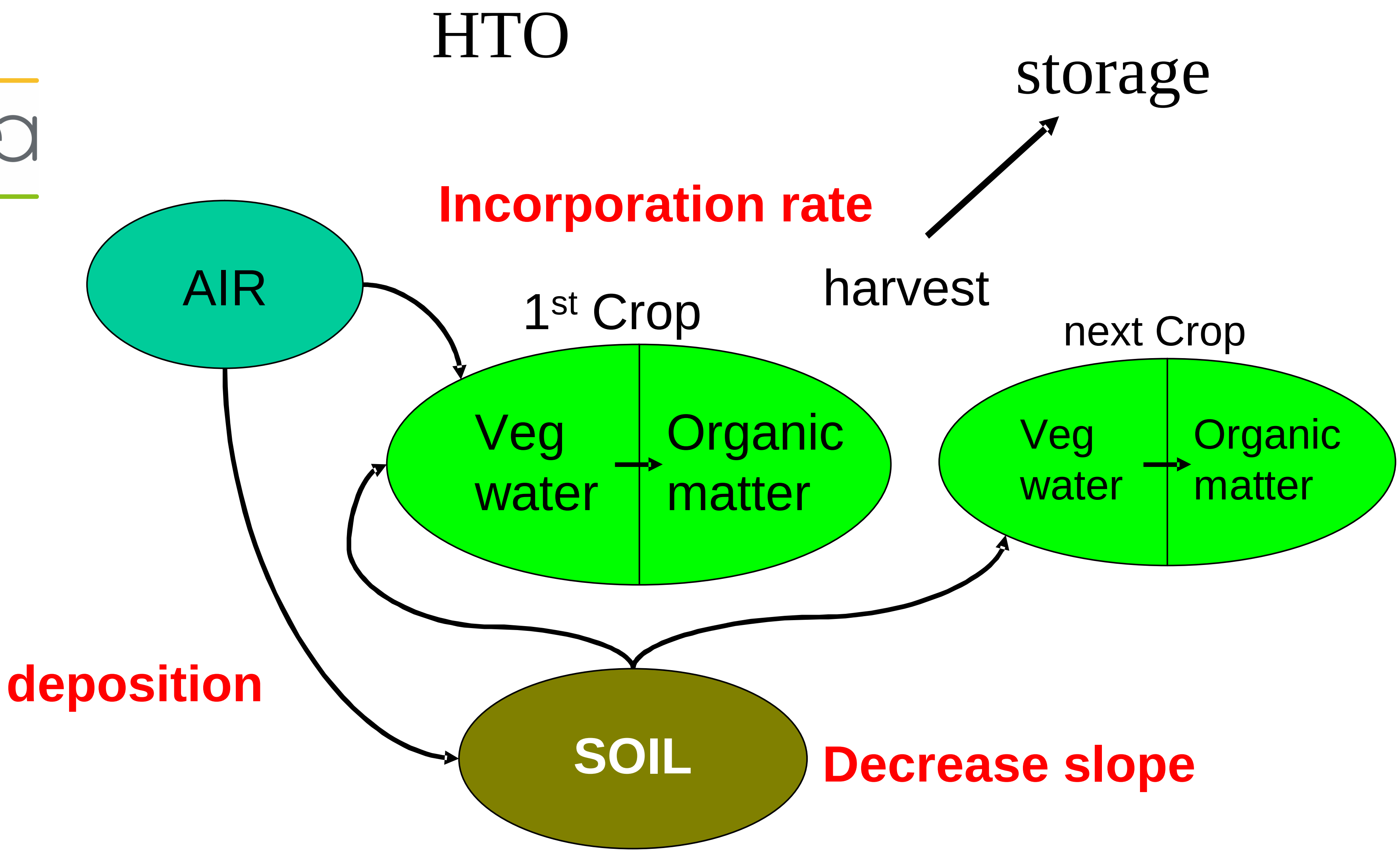
- a HT release has a 1-10 % dose impact compared with HTO
  - Exposure comes then from HTO converted in soil

- Tritiated water of plant follows air concentration and so is not stable
- OBT integrates air concentration

- ACCIDENTAL RELEASE
  ACCUTE RELEASE

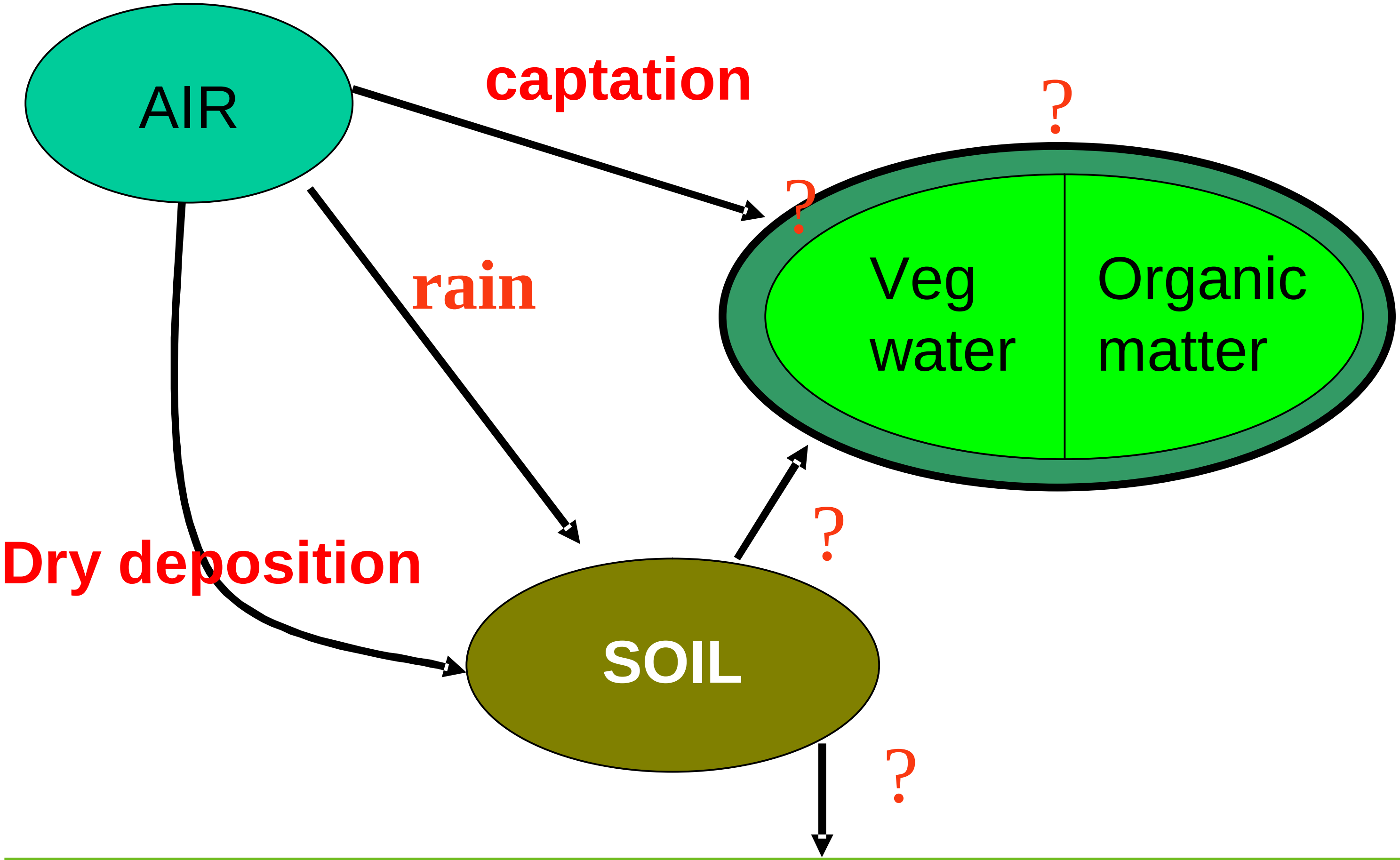
- Dynamic models

The different mechanisms involved in plants contamination : accute release



Tritiated particulates – dust & metallic hydrides

- Captation and deposition depending on particles' size



Do we know enough about Tritium impact ?

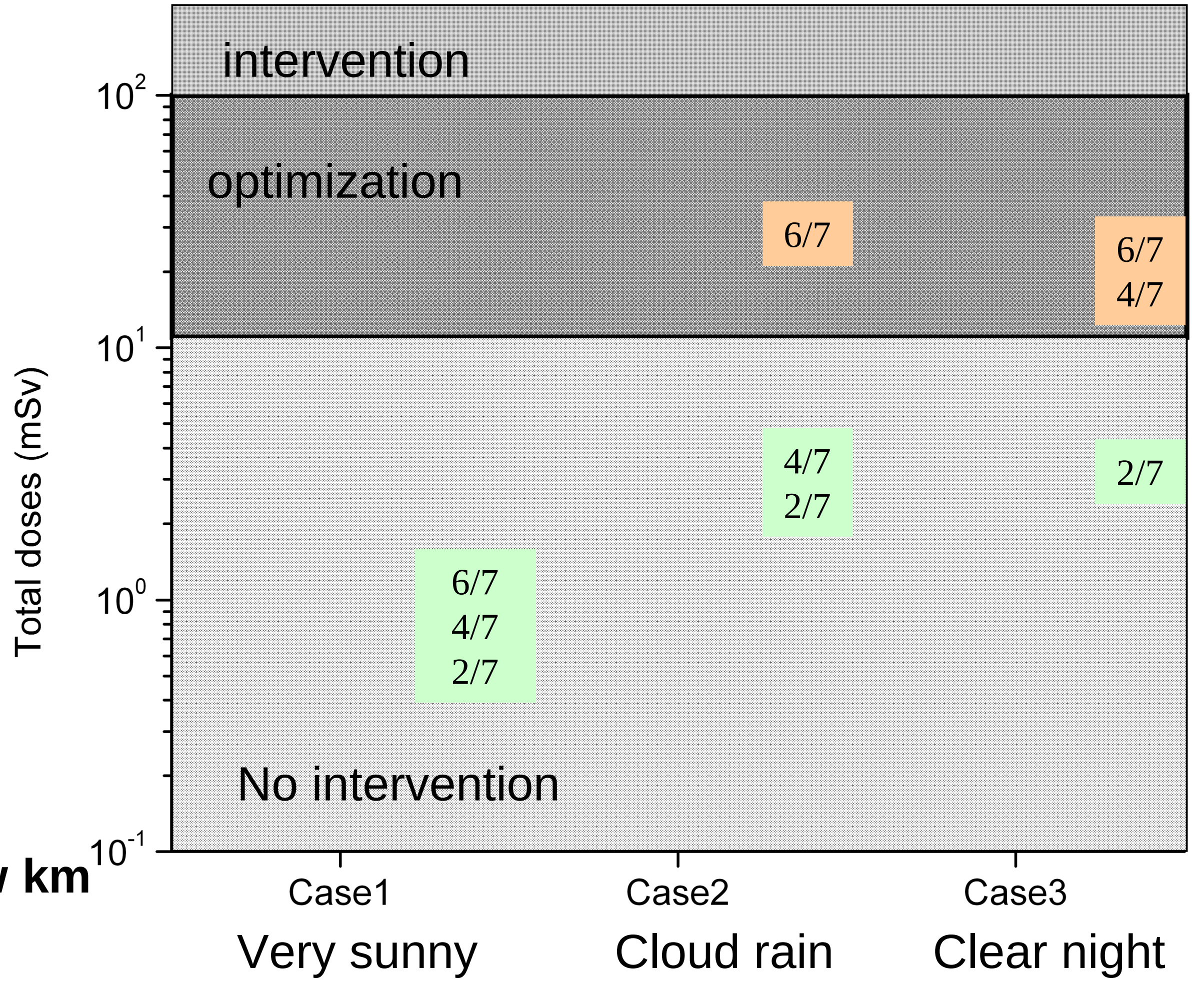
Strong influence of atmospheric diffusion conditions

Some conditions where optimization is required

- OK for fine sunny day
- Factor 10 for rain
- > Factor 10 for night

10 g of HTO = the frontier.

optimization limited to few km



Dose details by pathway –

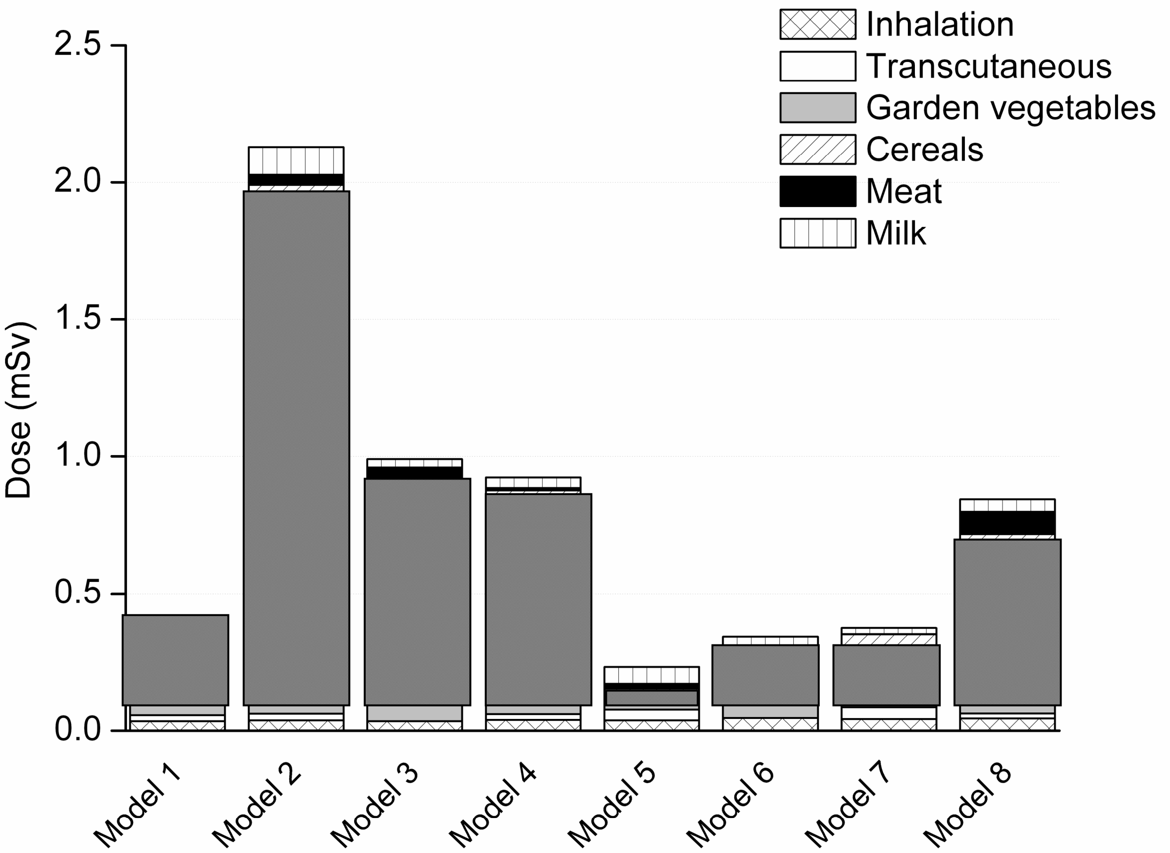
(At normalized conc.)

Vegetable Ingestion main pathway

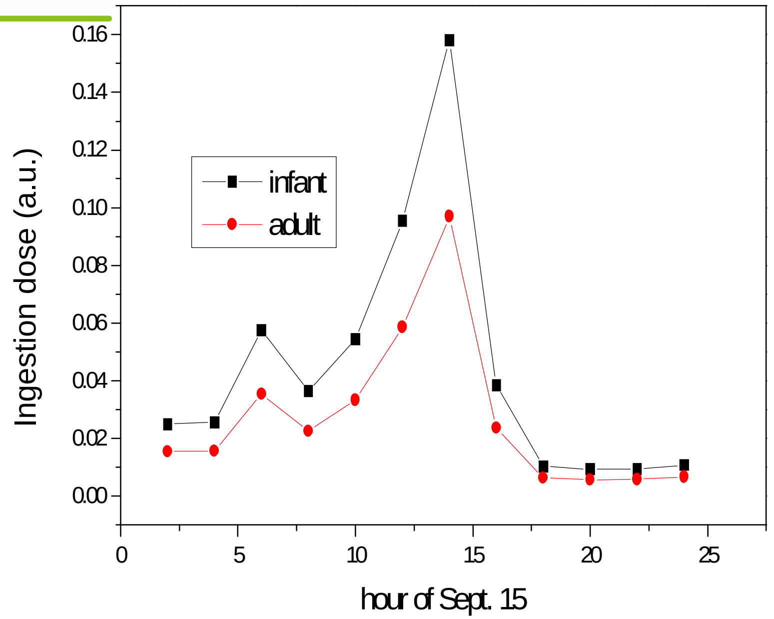
Garden veg or cereal

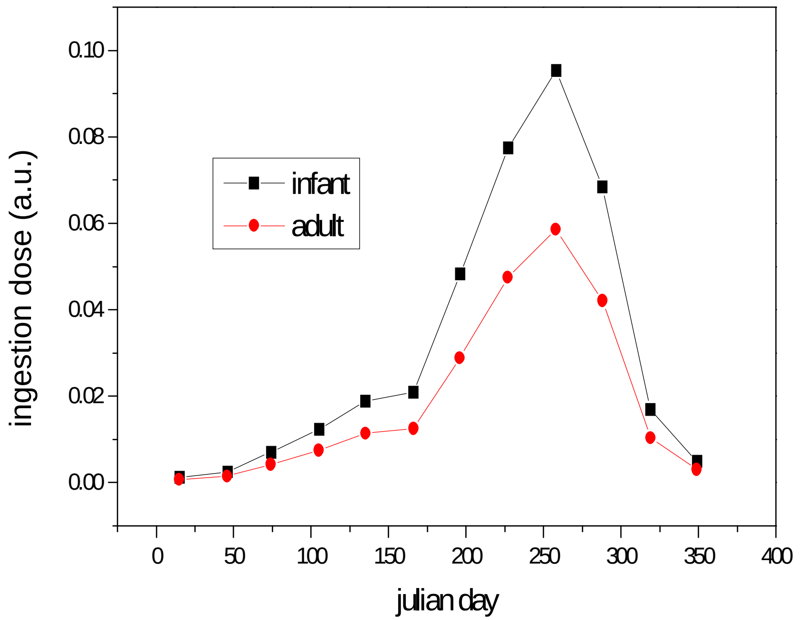
With rain no change.

At night, still food.



Diurnal and seasonal effects on ingestion dose after an accidental tritium release calculated with RODTRIT)





Dose is given in arbitrary units (a.u). The plants and animals were exposed to a constant HTO air concentration for one hour.

Relative importance of mechanisms

- Presently not enough certitude about acute release.

- Strong Rain may increase quite a lot the soil pathway, (but not the total exposure) ?

- Even with a strong rain (15mm), air pathway remains dominant in IAEA-EMRAS exercise.

- OBT generally dominant because of cereals – is that realistic ?

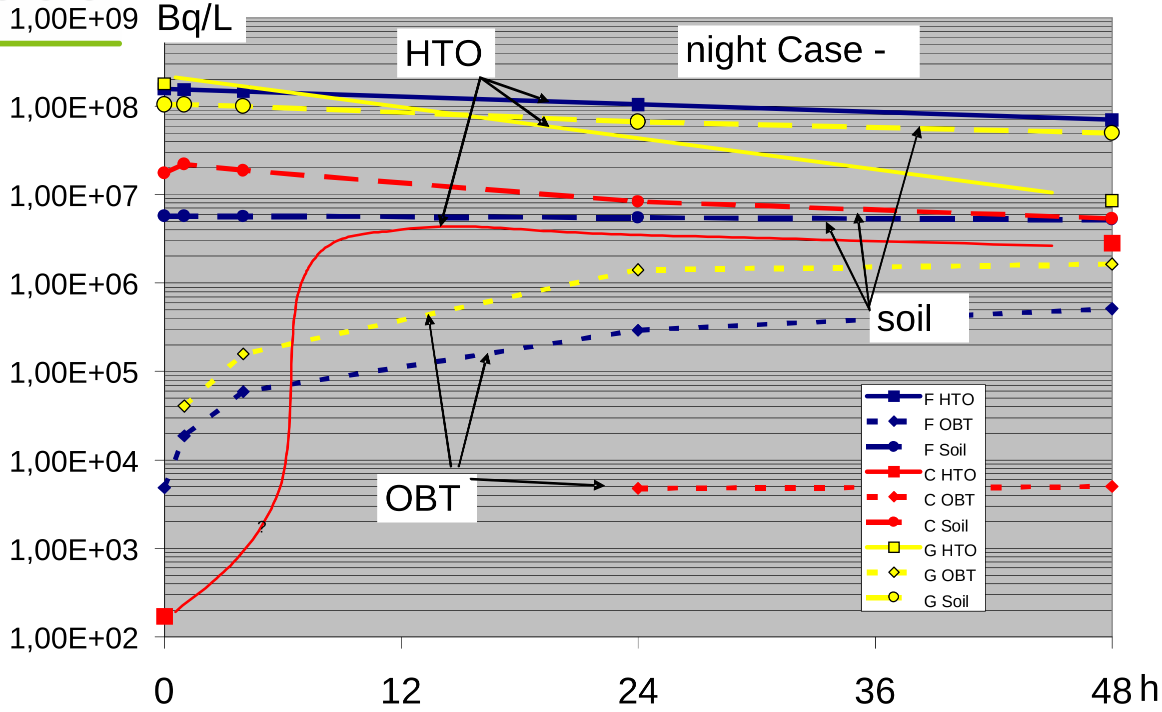
- Dose has to be integrated over one agricultural season because of soil pathway.

Question about scenario :case of wheat

- Ingestion of wheat is a specific problem for tritium :
  - The effect of cereals is reinforced by the storage and one year of consumption.

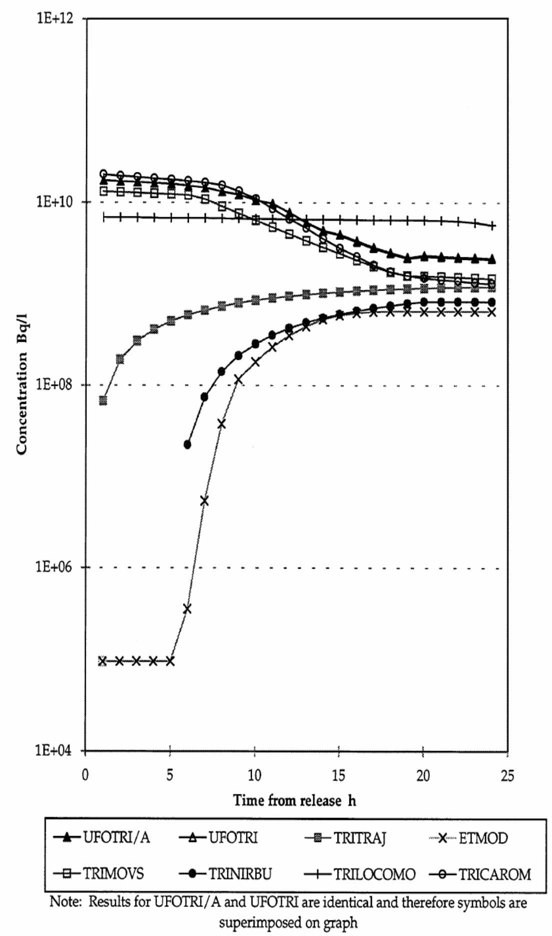
- What is covered by the name Cereals ?
  - 260g of cereals : flour, Bread, rice, corn- flakes…
  - Question of dry or wet weight.
    - 260g/d of flour ó 430 g/d of bread
  - The use of a single point in the wind axe is possible for a garden but has to be changed at least to a surface of a field for industrial food productions. That has a strong effect at short distance.
  - Sensible pathway only for few weeks per year.

Example of uncertainties for night assessment



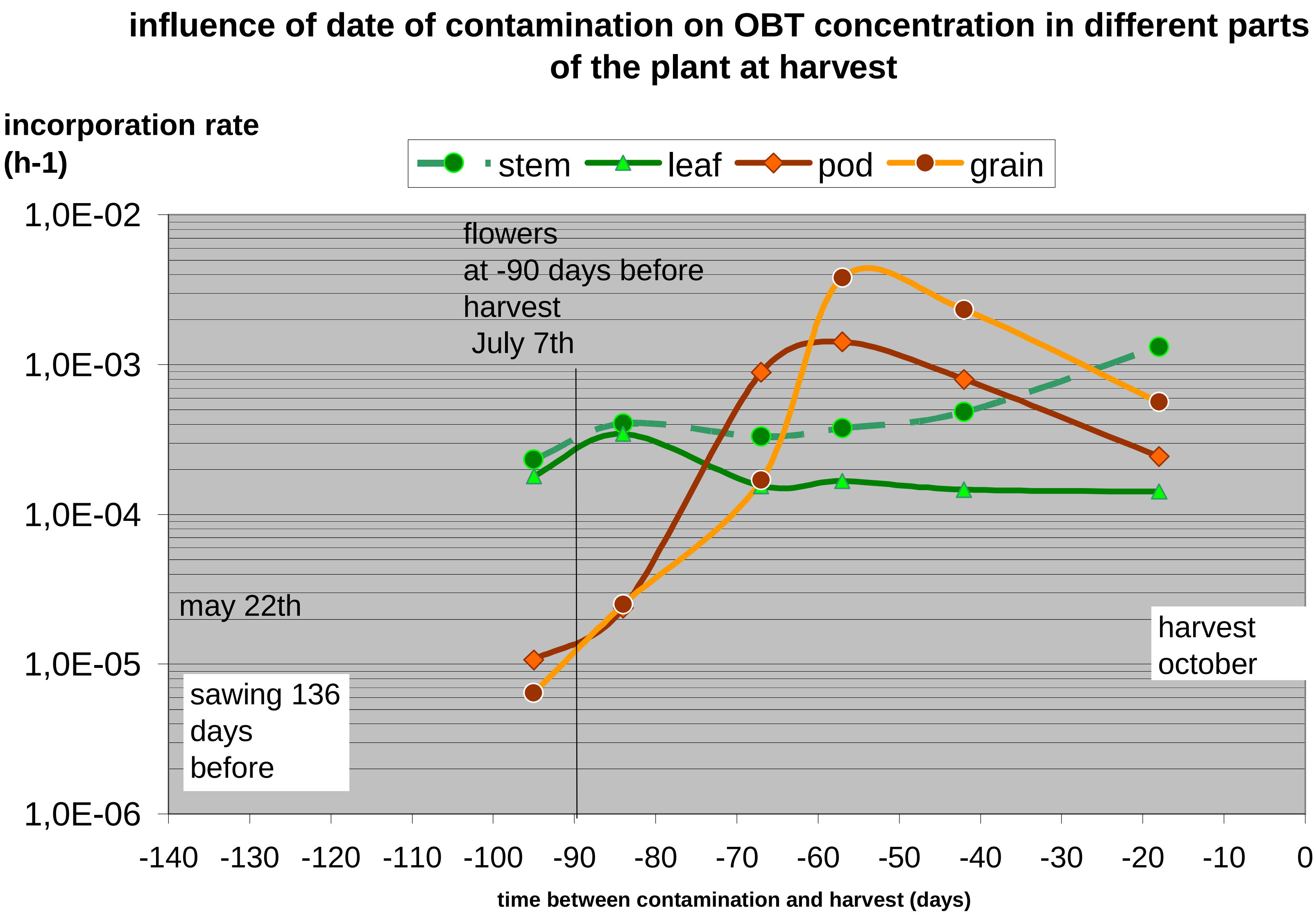
- Level reached

- slopes



. Concentration of HTO in tissue water of leafy vegetables following the night-time release, 1–24 h.

Agricultural data to develop (sojabean example)



Regulation : Reference level for food limitation after accident

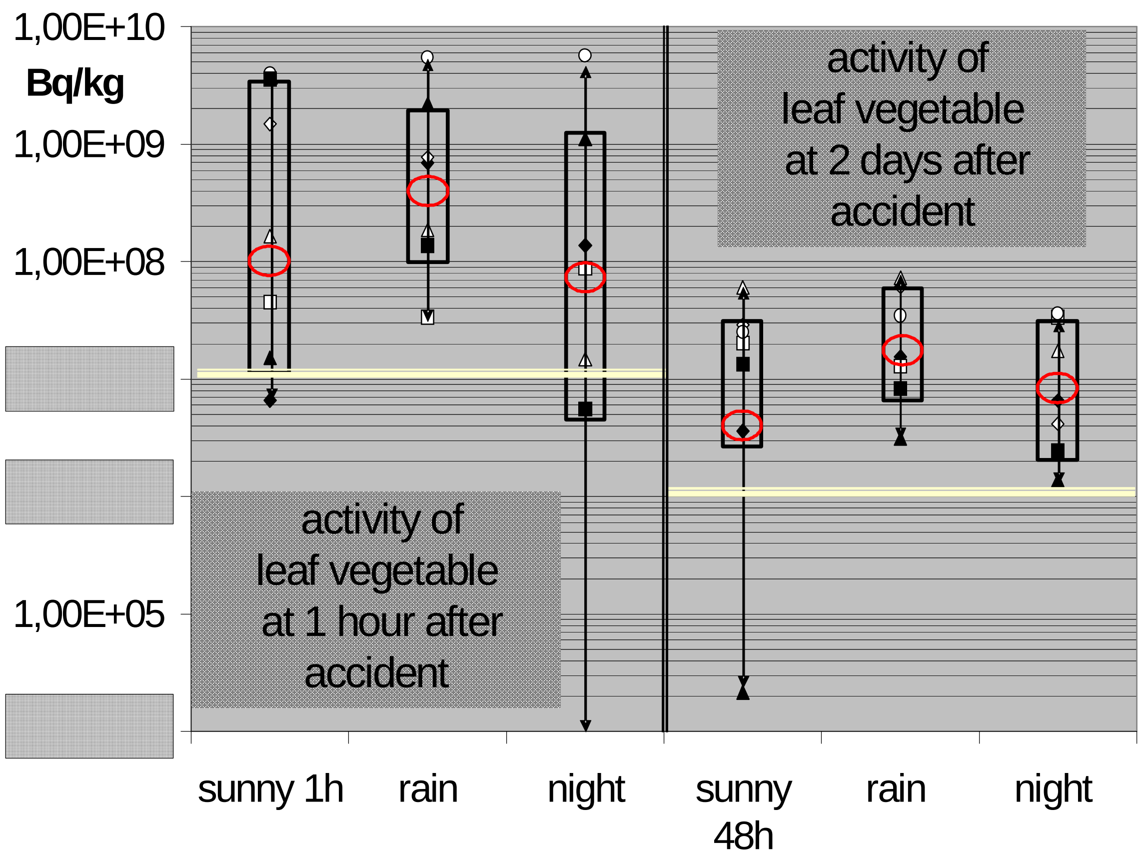
(calculated for 1mSv in 1 year after accident 10% local and 650 kg/y)

10^7 to define area concerned the 1st day

10^6 to precise area in the following days

(After 2 days equilibrium reached with soil water)

10^4 of Codex alimentarius far too low



Conclusions for environment : R&D

- Tritium does not concentrate in food chain

About models

- Variability remains very large in case of accident especially in rain and night cases.
- Modification needed for wheat modelization - realistic approach

About experimental Data

- Translocation of organic matter from leaves to edible part of the vegetable.
- Case of the night for experimental data.
- What about Tritiated particulates ?

About modelers

- The present Tritium scientific community is very small,
- has to synthesize what is absolutely needed in models for acute release.
- This community could disappear from EU in the few next years.

Conclusions for regulation ?

- what level for food trade ?

- **10^4 Bq/kg** is not the right level and is really too low

- Not Consistent with the 10 mSv ICRP level for optimization of interventions (>2 orders of magnitude)
- Not Consistent with codex alimentarius principles 1 mSv + 10% of a huge consumption + level decreasing after an accident + assessment for 1 year.
- Not Consistent with 10^4 Bq/L for WHO drinkable water in normal conditions.(0,1mSv-100%)

- Transport

4 Bq/cm^2 is not reasonable for Tritium

- Drinkable water :

WHO should be kept as reference 10^4 Bq/L. keep 100 Bq/L as an investigation value but not a definition of drinkable limit. (Cf USA)

Environmental Regulation or reference data

- 10^9 Bq.kg^{-1} - European directive radioprotection 1996
- 10^5 Bq.kg^{-1} - IAEA clearance level for waste
- 10^5 Bq.kg^{-1} - Canada OBT food trade value after accident
- 10^4 Bq.kg^{-1} - Codex alimentarius proposal after accident
- 10^4 Bq.L^{-1} - WHO for drinkable water
- 740 Bq.L^{-1} USA reference value for watertable 20000 pCi.l^{-1}
- 100 Bq.L^{-1} - EU for distribution water control
- 10 to 10000 Bq.L^{-1} normal T activity in the vicinity of a plant
- 10 Bq.L^{-1} - reference value for ANDRA storm rain basin
- 1 to 10 Bq.L^{-1} - normal tritium activity of water
- 4 Bq.cm^{-2} - beta value for transport

Thank you for your attention