IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

50 years of radioactivity monitoring of the environment

IRSN/DEI/STEME/LMRE

1. IRSN
2. Radioactivity levels in the environment
3. Methodologies
4. 50 years of aerosol sampling

IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

IRSN, Radioprotection and Nuclear Safety Institute

Position of IRSN

Public expert

in charge of the scientific evaluation

of radiological and nuclear risks

In France

The expertise is based on scientific research

Main missions

- public information in the field of radiological and nuclear risks,
- support public authorities in nuclear safety and radiation protection for civil and defense activities,
- expertise available to numerous French and foreign partners and customers.


FONTENAY-AUX-ROSES
(Head office)
Nuclear defense surveys
Environment and response
Radiation protection and human health
Reactor safety
Plant, laboratory, transportation, and waste safety
OCTEVILLE - CHERBOURG
Environment
LE VÉSINET
Environment and response
Radiation protection of man
SACLAY
Plant, laboratory, transportation, and waste safety
ORSAY
Environment
PIERRELATTE
Response
Radiation protection and human health
TOURNEMIRE
AGEN
Environment and response
LES ANGLES - AVIGNON
Response
Plant, laboratory, transportation, and waste safety
CADARACHE
Environment
Prevention of major accidents
Radiation protection and human health
Nuclear defense surveys
LA SEYNE-SUR-MER
Environment


N
MAHINA - TAHITI
Environment

IRSN
INSTITUT
DE RADIOPROTECTION
ET DE SÛRETÉ NUCLÉAIRE

- Human Radiological Protection
- Fuel cycle transportation and facilities
- Reactor Safety
- Prevention of Major Accidents
- Nuclear Defense Expertise
- **Environment and emergency operations**

~ 1 700 specialists: engineers, researchers, medical doctors, agronomists, vets and technicians, competent in all the fields of nuclear safety, radiation protection and inspection of sensitive nuclear materials.

~ 70 PhD students

Missions of the Division

- Study (on-site monitoring, experiments, modelling) on behaviour and effects of radionuclides in biosphere or geosphere due to controlled or accidental releases, waste storage (disposal) or natural sources
- **Quantifying, monitoring and interpreting the radiological state of the environment**
- Radiological protection operations and assistance (radiological control in facilities, radioactive sources recovery, emergency assistance, etc.)
- Crisis management (Emergency Technical response Center, emergency tools development, etc.)
- Study of seismic risks

Study of radionuclides behaviour in Ecosystem

Main objectives :

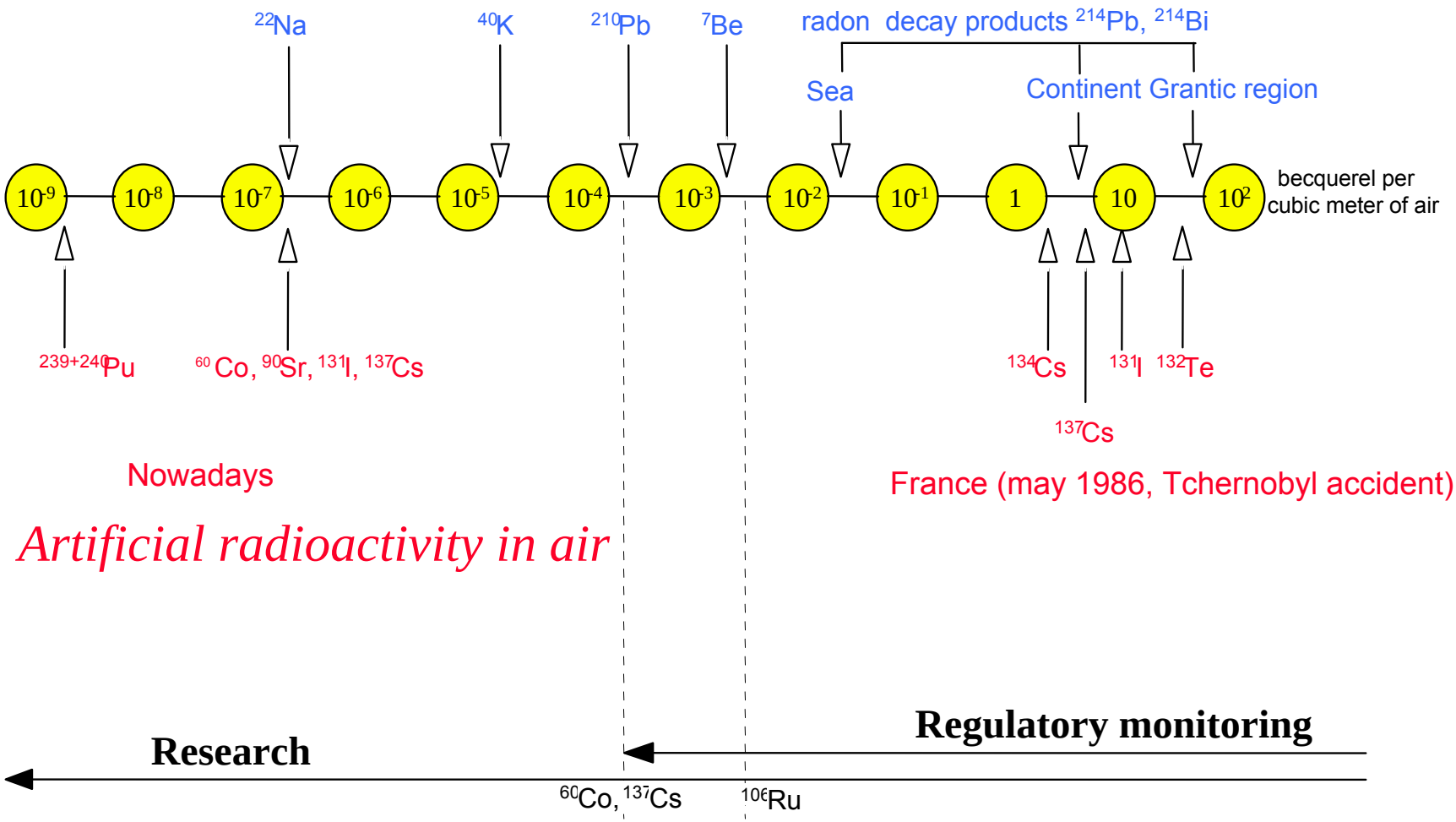
- to understand and quantify the transfer mechanisms of the natural and artificial radionuclides (eventually associated with chemical pollutants) in the ecosystem as well as their effects
- Development of numerical models to explain and quantify the behavior of radionuclides in biosphere, soil or sea
- Synthesis of knowledge and methods to develop IRSN expertise ability concerning the radionuclides-related risk on environment

Radioactivity levels in the environment

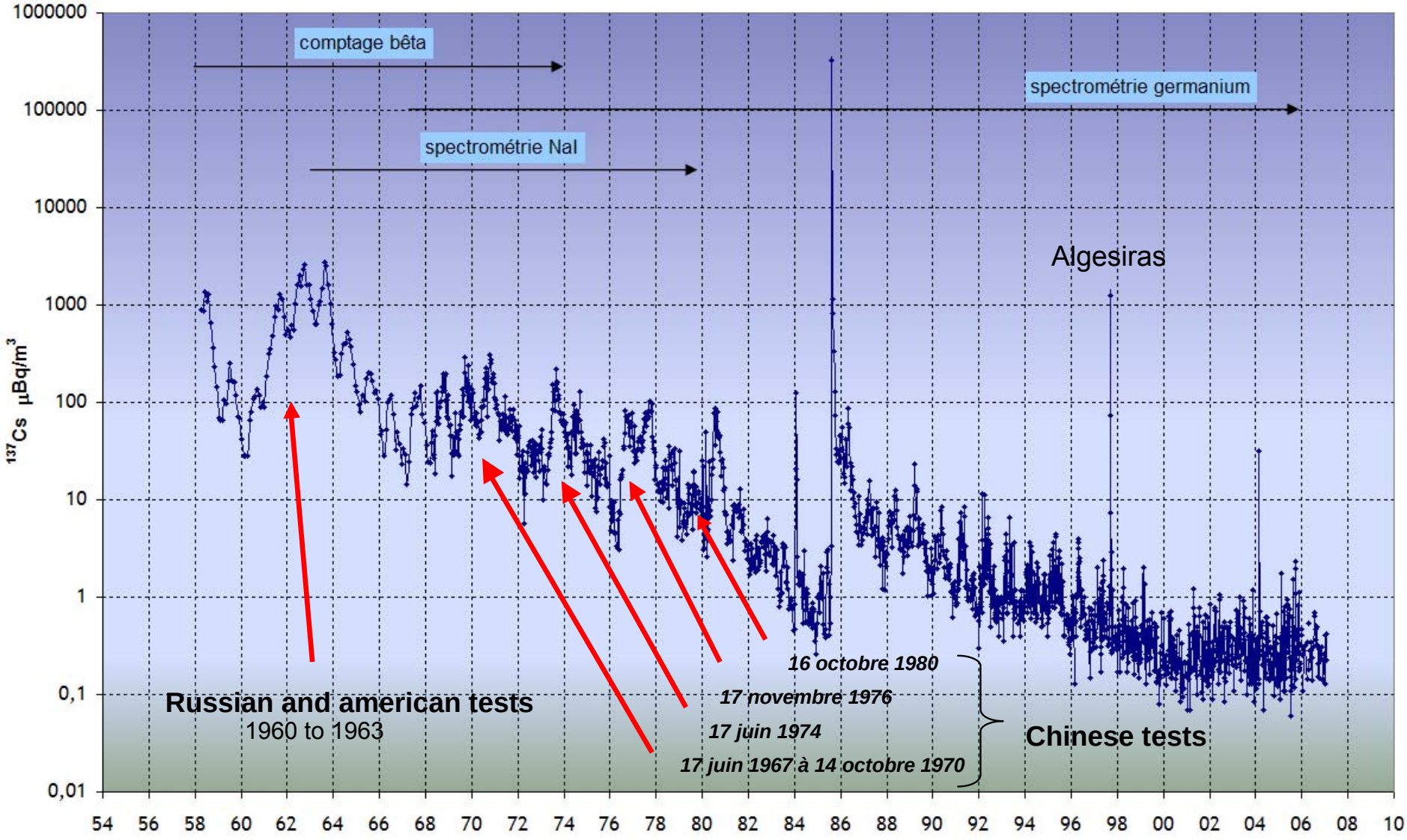
...or the need of low level measurements

Logarithmic scale of radioactivity concentrations in air in France

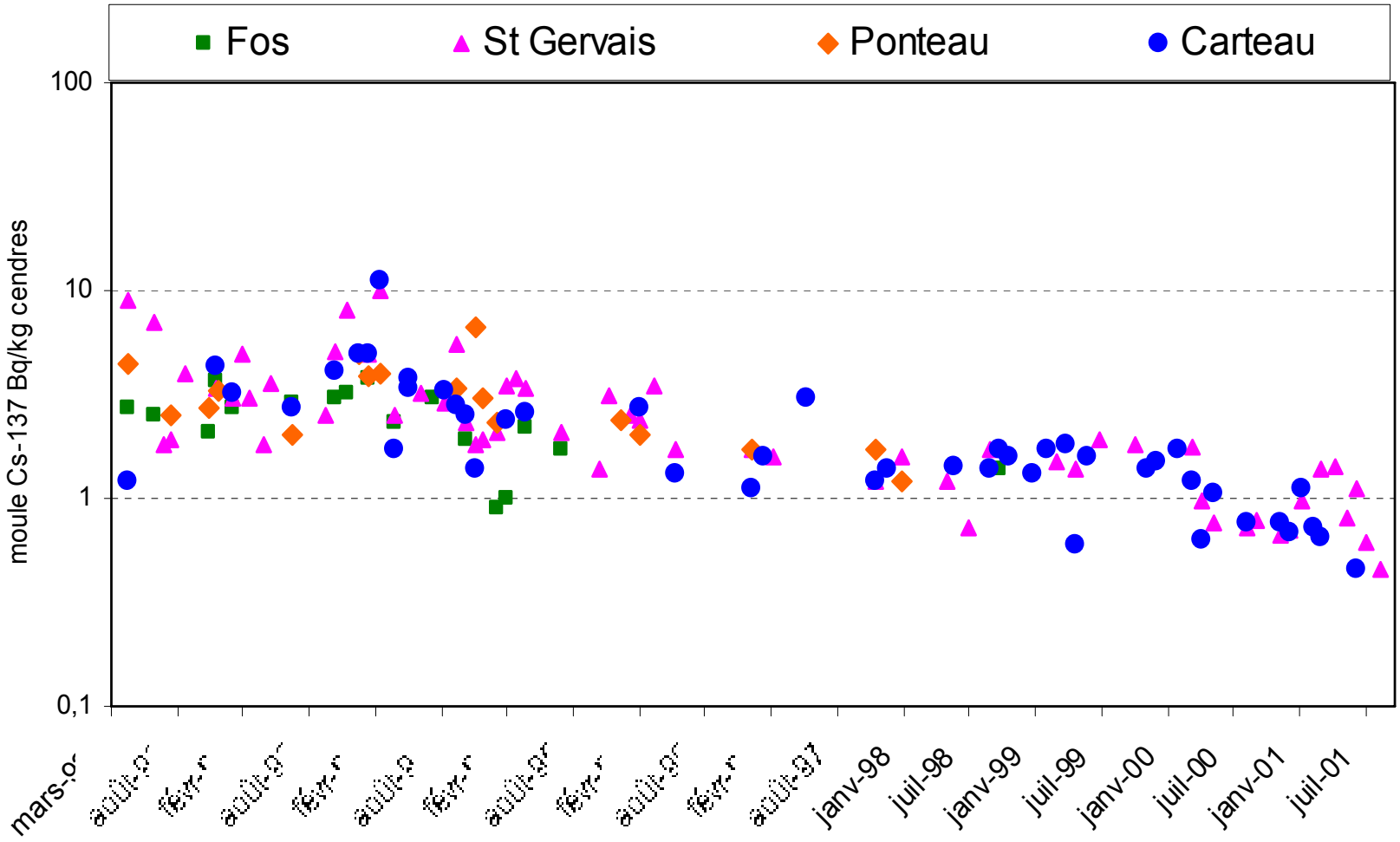
Natural radioactivity in air



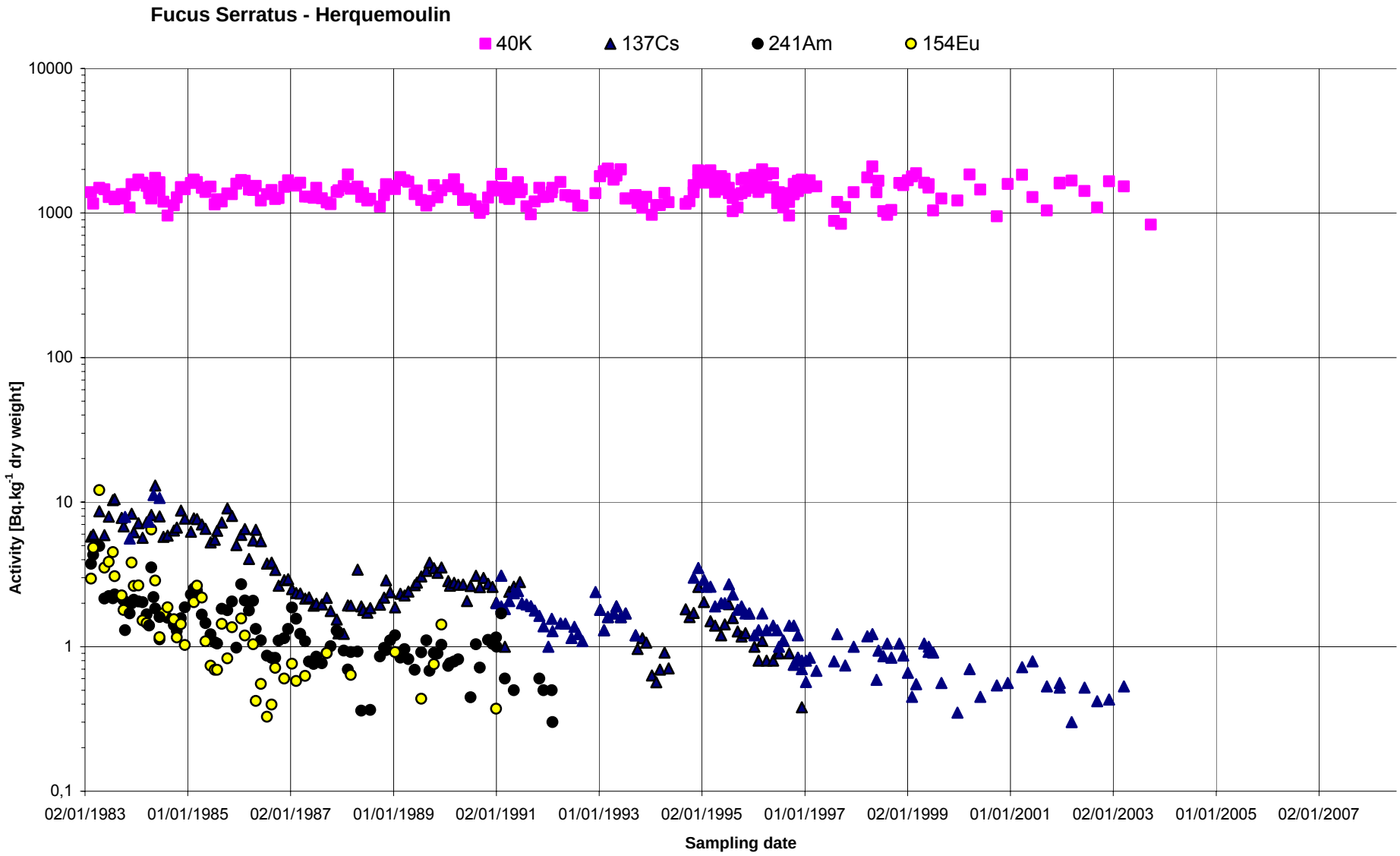
^{137}Cs in air at Orsay



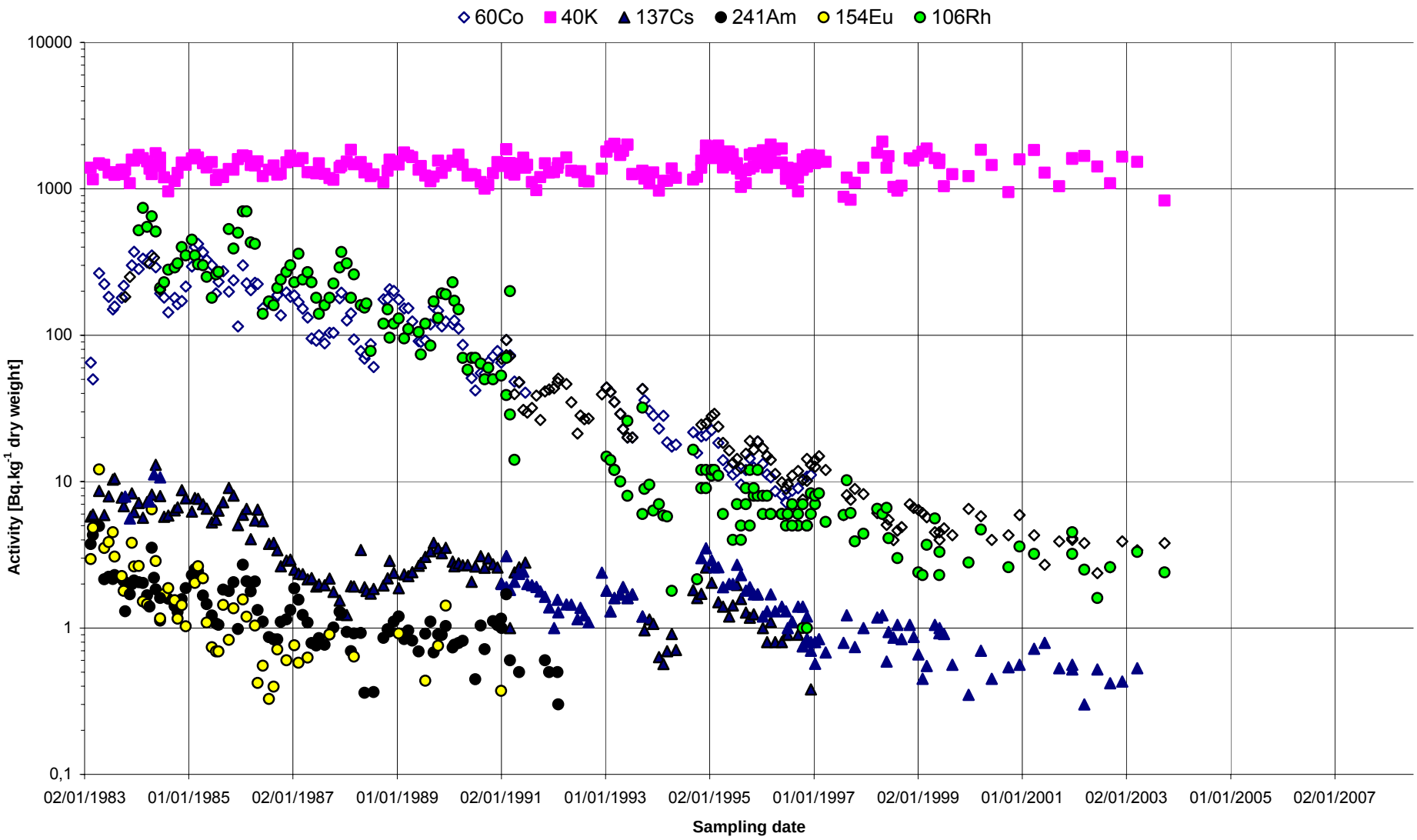
^{137}Cs: mediterranean Mussels from 1992 à 2002



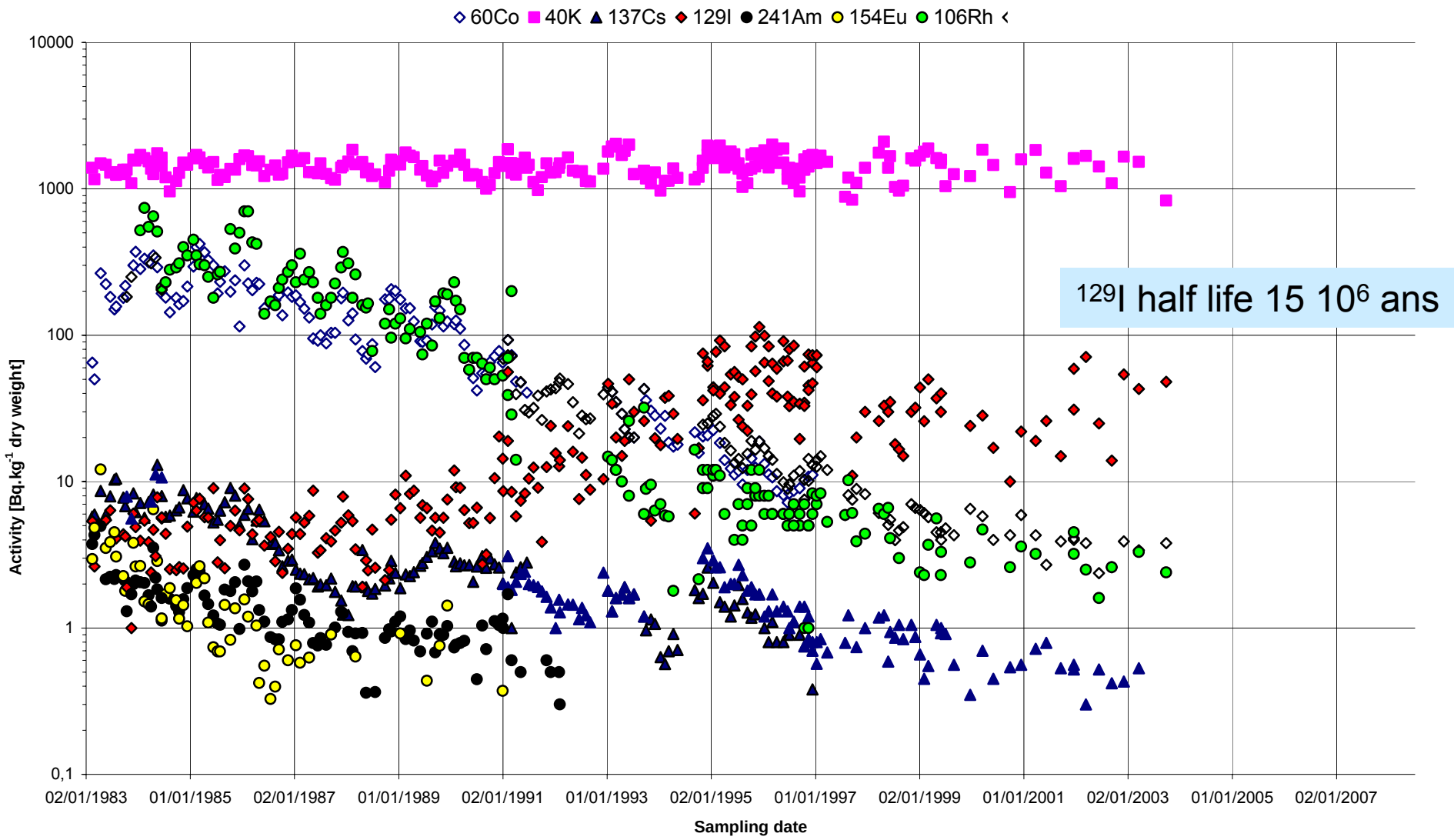
Seaweed in the English Channel (*Fucus Serratus* - Herquemoulin)



Seaweed in the Channel



Seaweed in the Channel



^{129}I half life 15 10^6 ans

What is the aim to study radionuclides in the environment at the trace level ?

Transfer studies in natural (not contaminated) environments

- **From one compartment to another (atmosphere/oceanic/terrestrial)**
- **From an organism to the other (grass->meat or milk, …)**
- **From a compartment to an organism (water >Mussels, …)**
- **-> Development / validation of models (that can explain, and predict)**

Background levels existing in the environment – away from nuclear facilities

- **in all compartments of the environment**

The overall objective : improving the prevision of the impact of the effluents in a post accidental situation

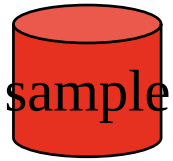
=> best available methods in order to quantify, a detection limit is not enough

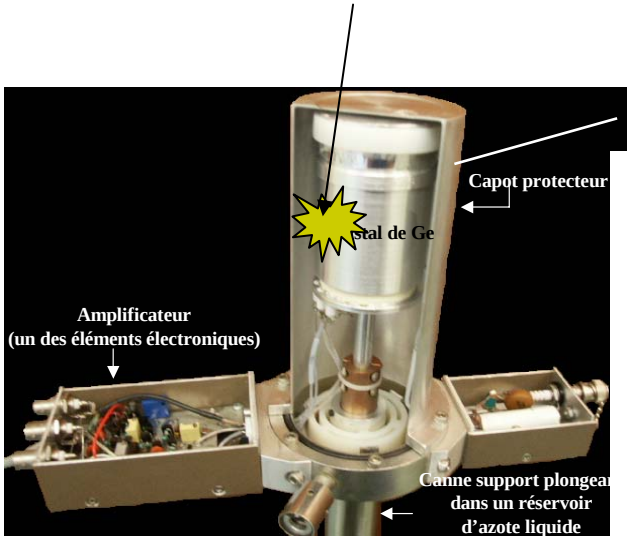
Methodologies

Collecting the sample

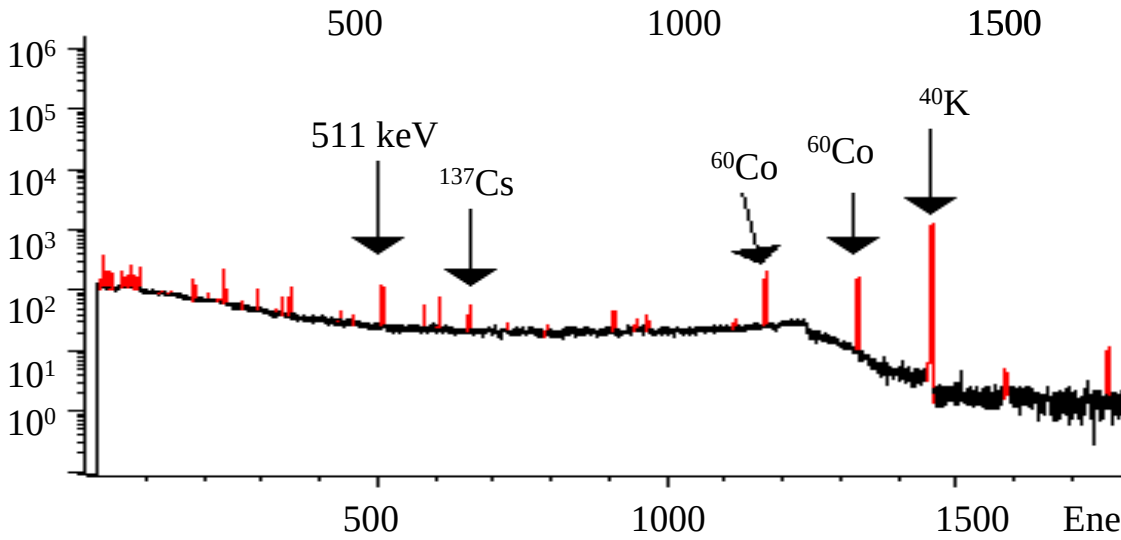
Huge “vacuum cleaner”
+ filter =
sample

Gamma spectrometry air samples





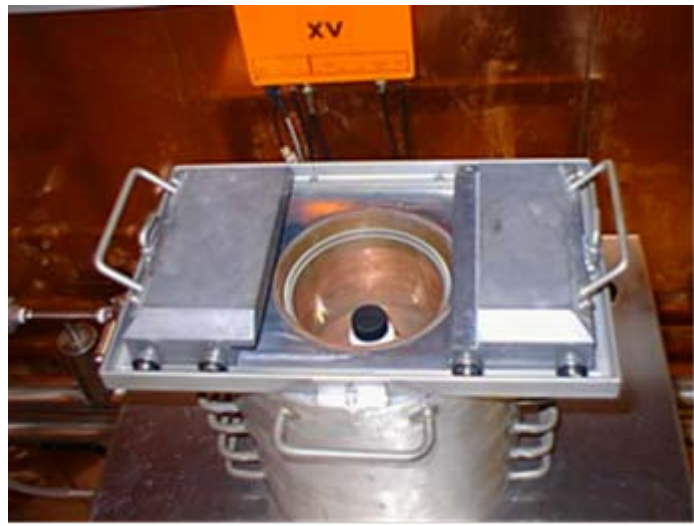
Identification
Quantification



For this monitoring we have a

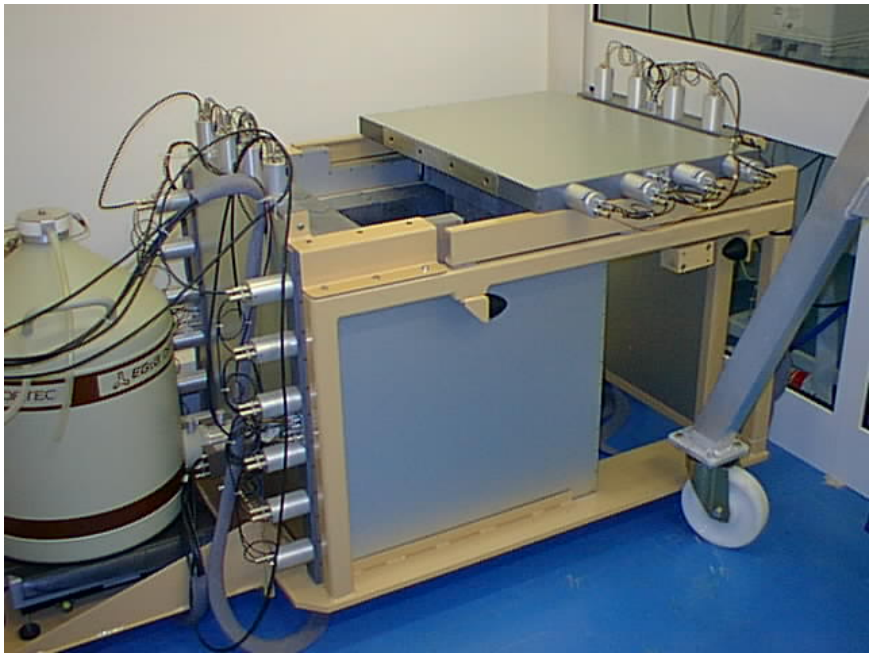
Signal / Noise problem !!!

Standards detectors at Orsay



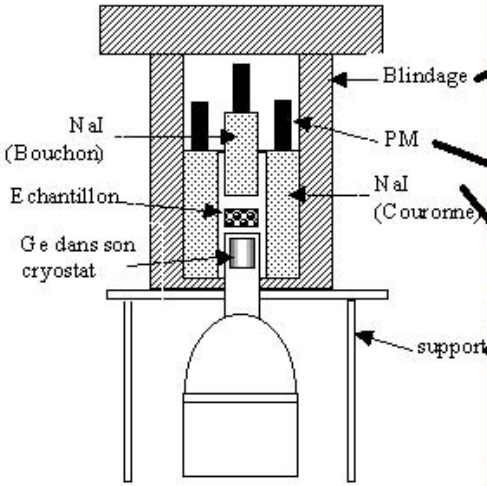
•Shielded room, large spectrometers (50% relative efficiency)
• Sous 3 m de béton (~ 10 mee)

Special detectors

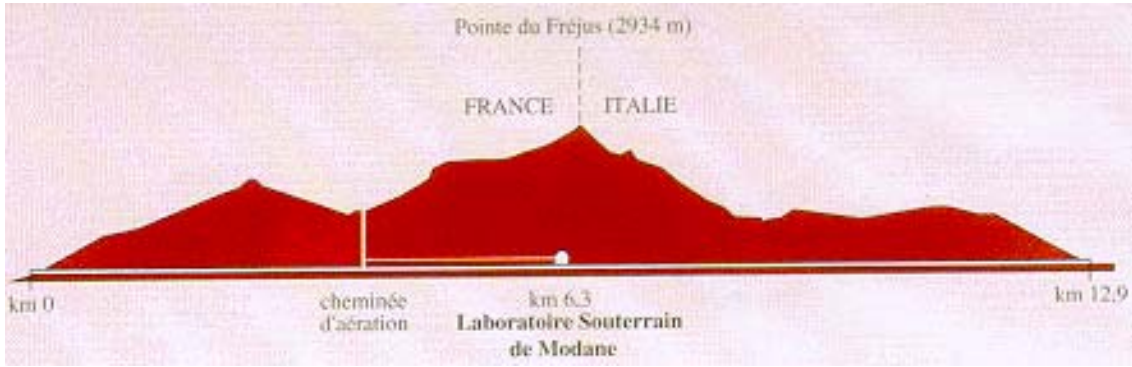


Anti Cosmic

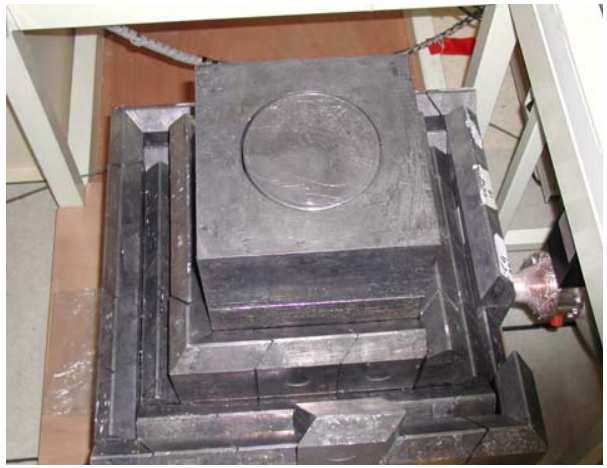
Anti Compton



Ultra low background detectors installed in theunderground laboratory of Modane



Le laboratoire est situé à 1700 m sous la pointe du Fréjus au milieu du tunnel routier

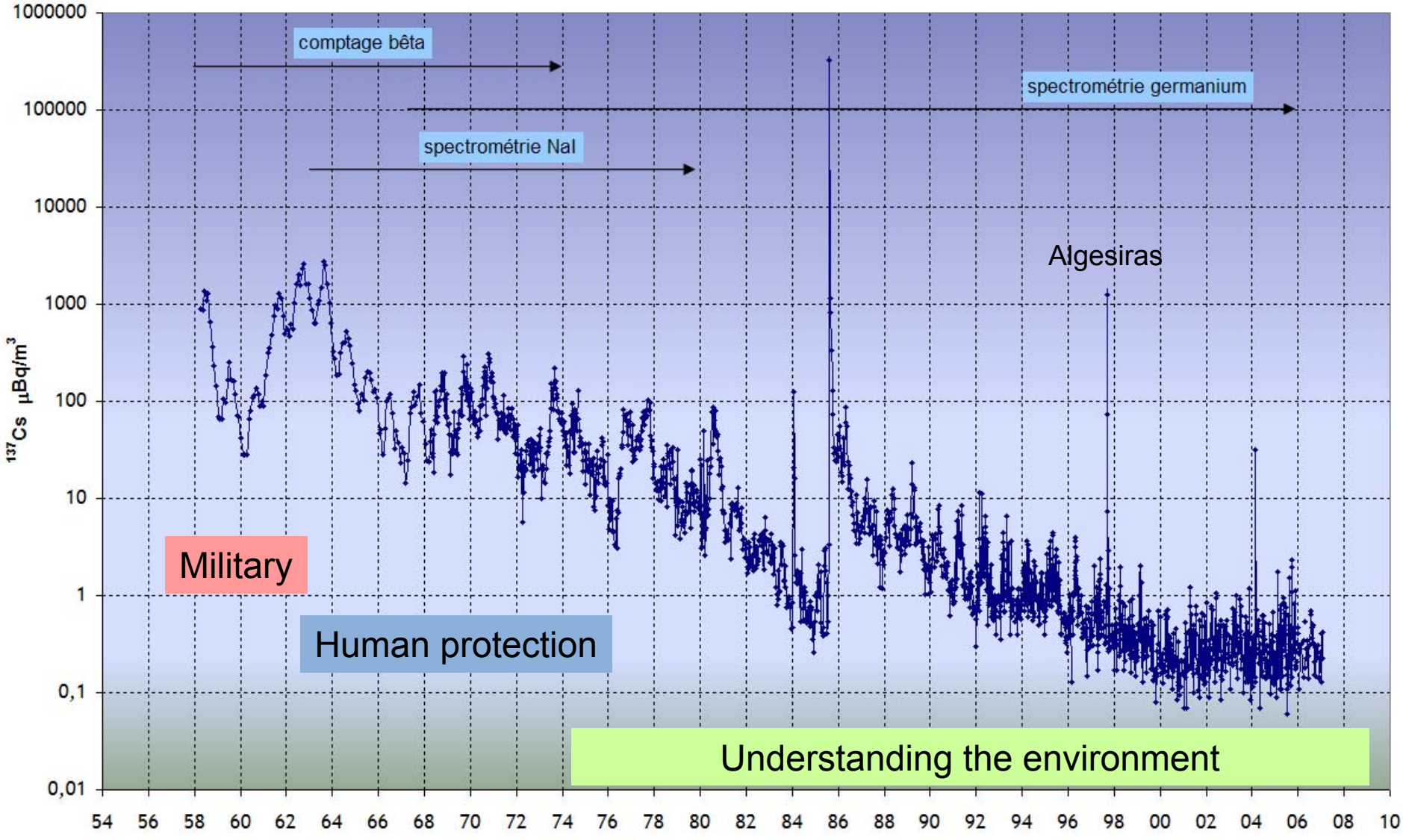


- Ge coaxial type-N 50%

- Détecteur puits 860 cm^3

50 years of aerosol sampling

Historical aims of the aerosol study



Compréhension des phénomènes de transport

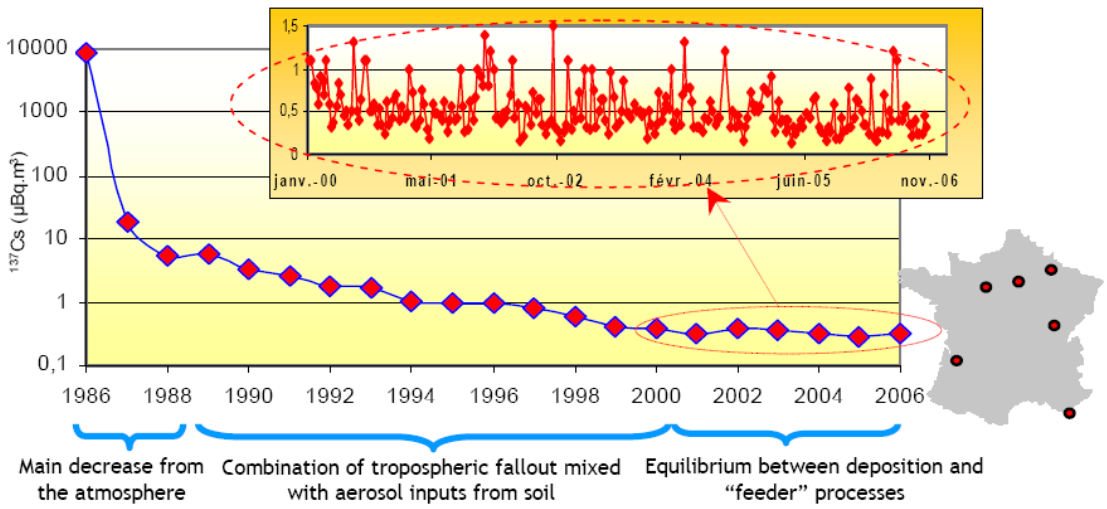


Figure 1 : Mean annual ^{137}Cs activity level in air and details on a 10-day sampling basis

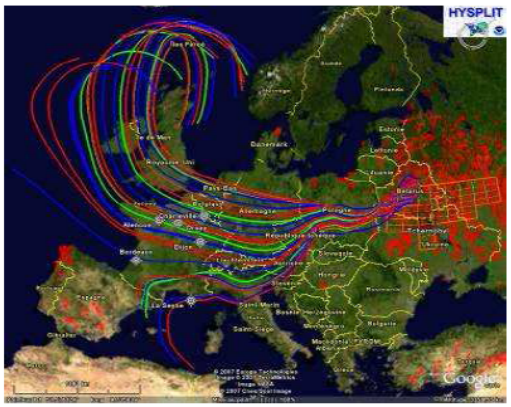


Fig. 6: Trajectories of smoke plume and fire spots

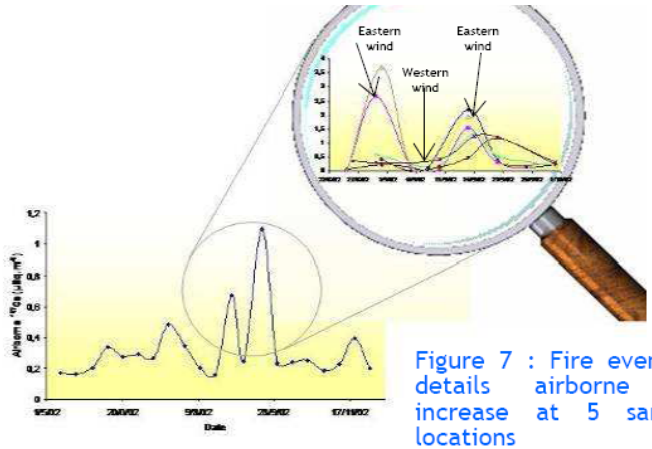
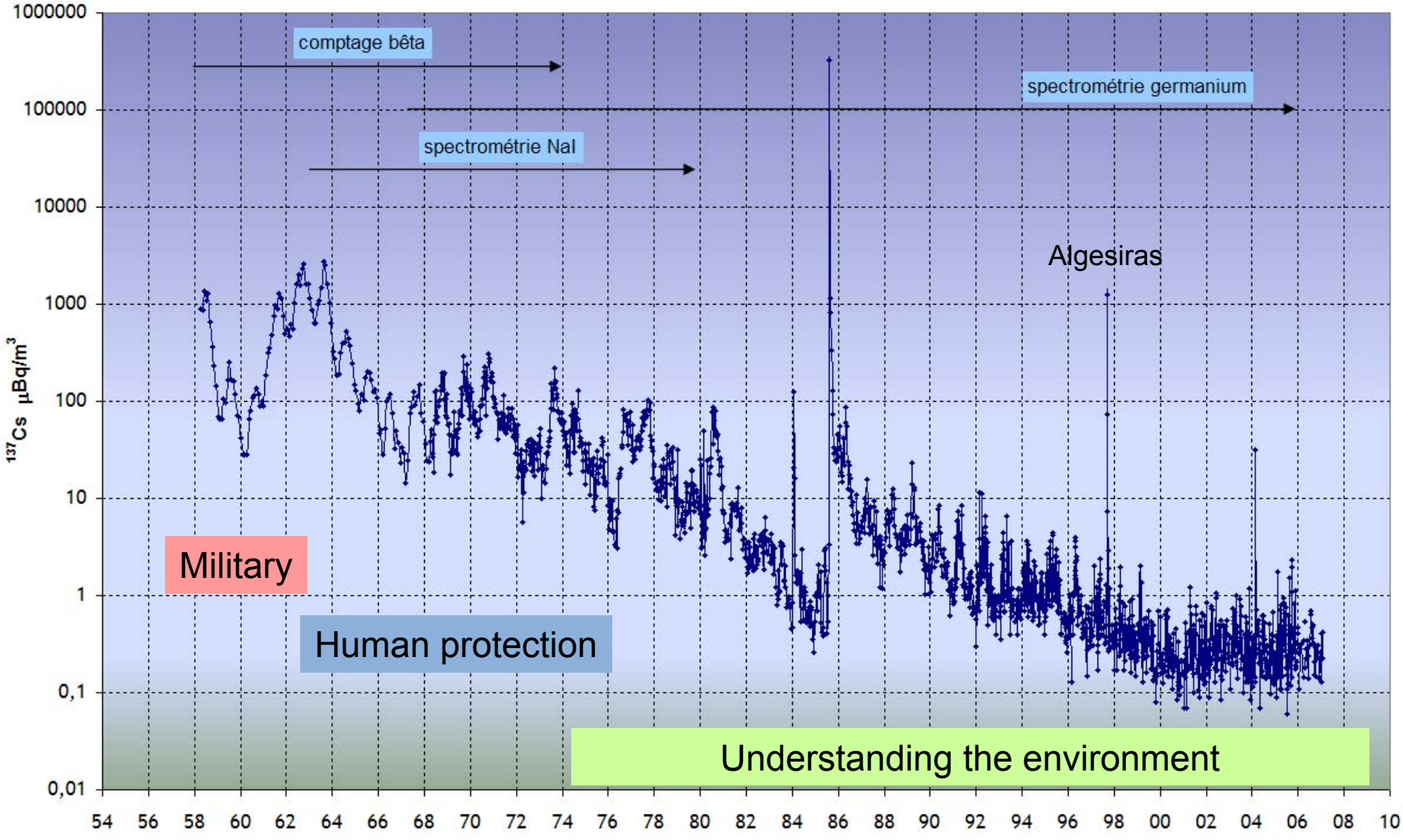
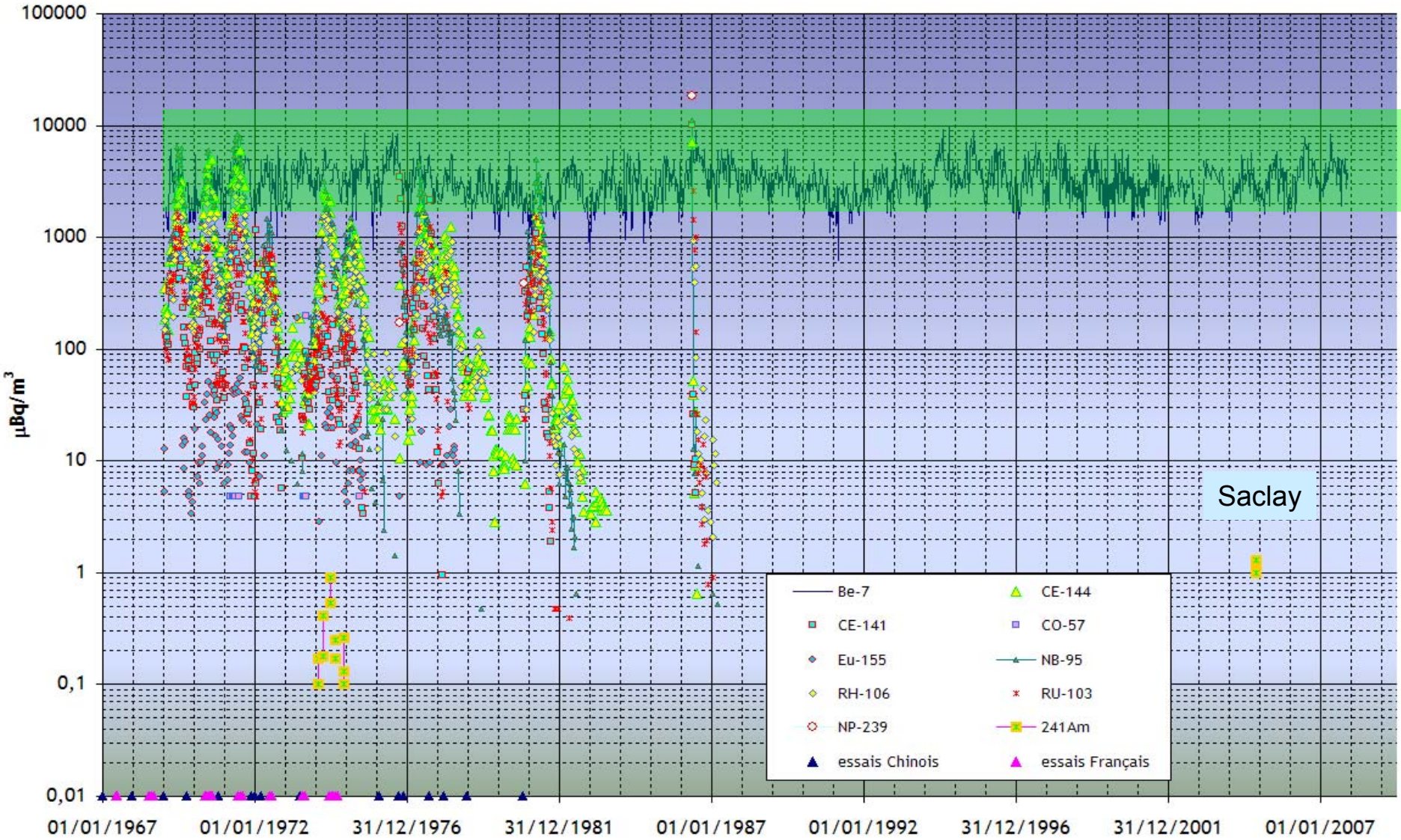


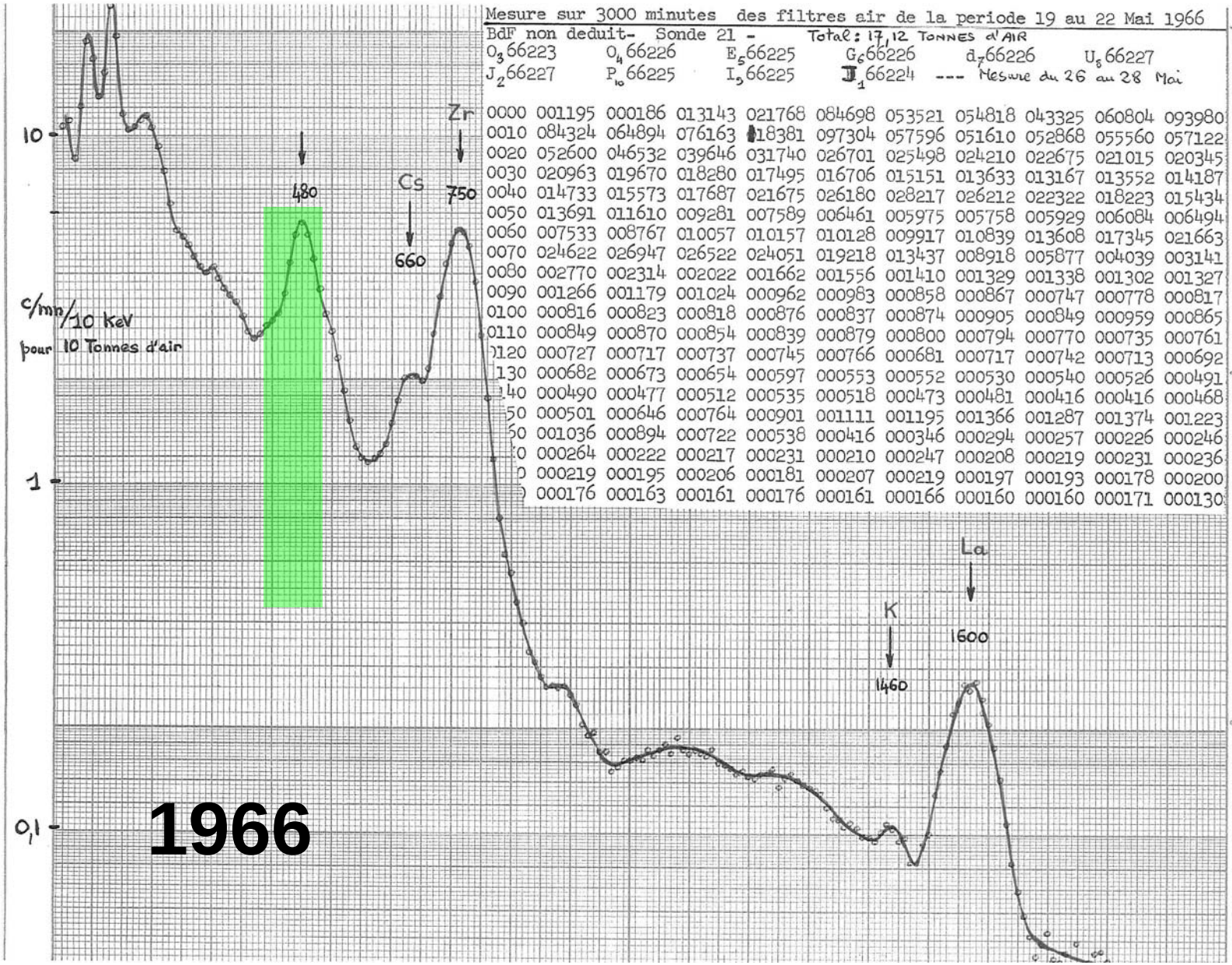
Figure 7 : Fire event and details airborne ^{137}Cs increase at 5 sampling locations

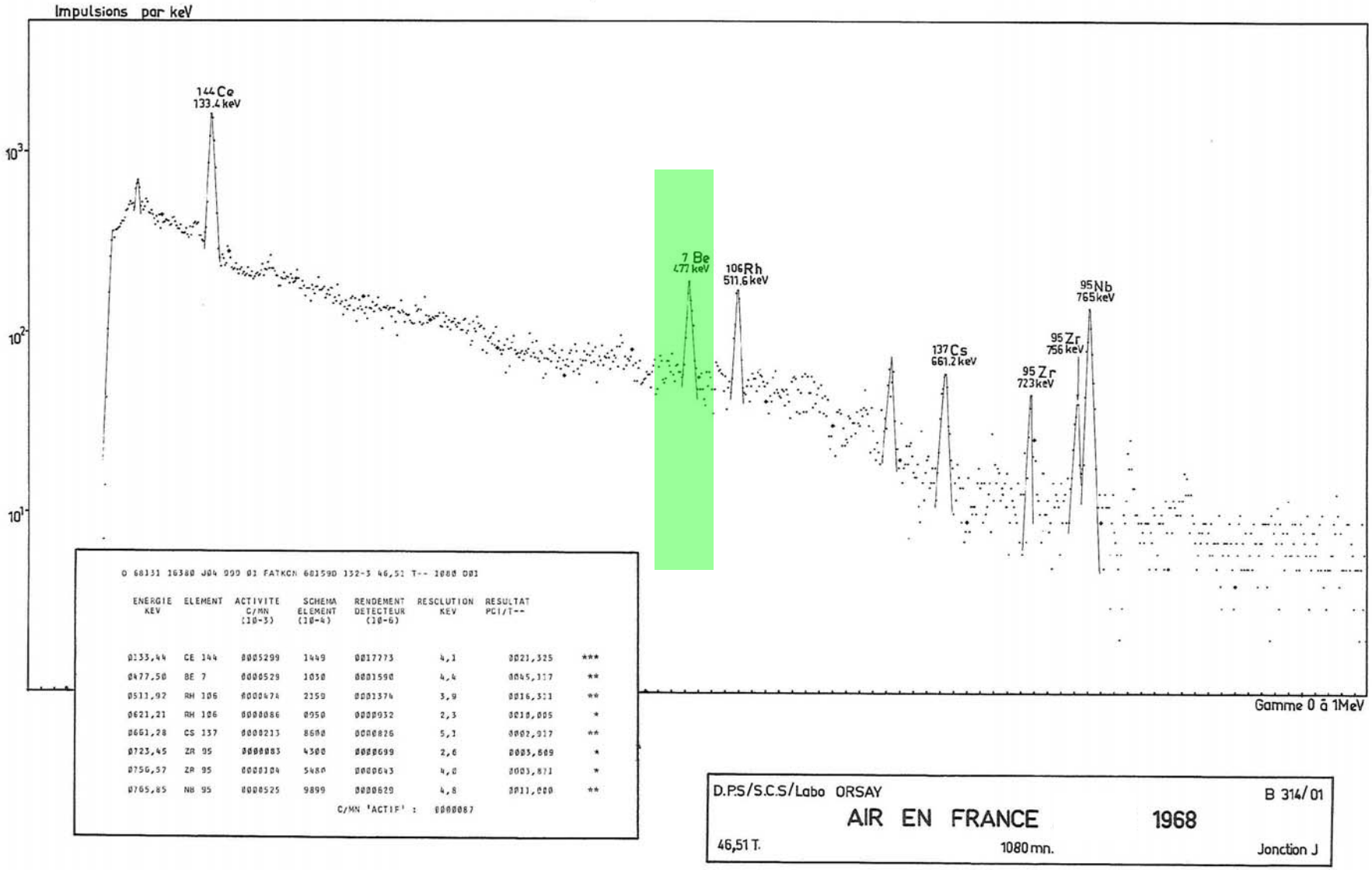
Historical aims of the aerosol study



Air at Orsay (20 km south of Paris)



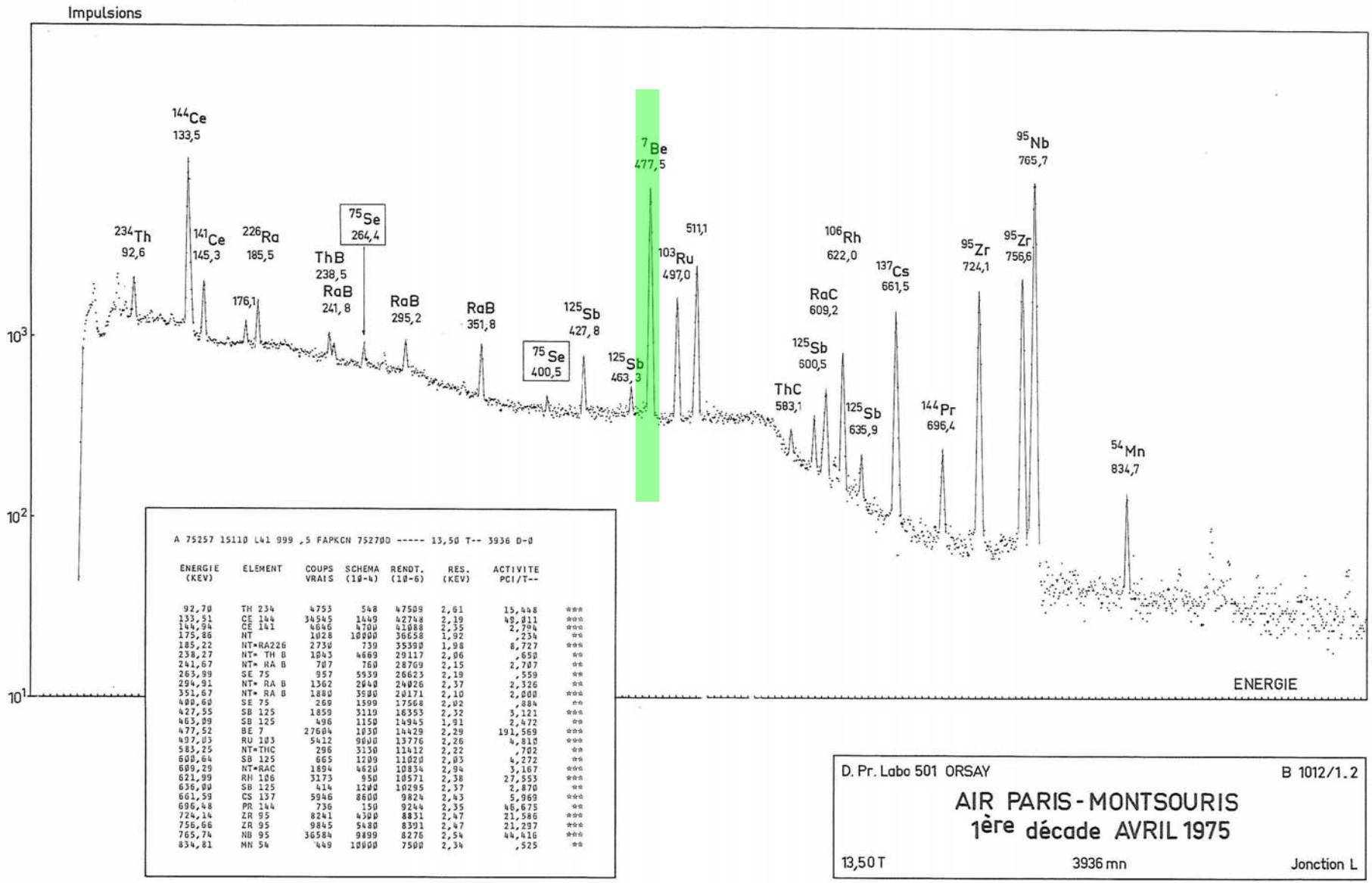

Mesure sur 3000 minutes des filtres air de la periode 19 au 22 Mai 1966
BdF non deduit- Sonde 21 - Total: 17,12 TONNES d'AIR
O_3 66223 O_4 66226 E_5 66225 G_6 66226 d_7 66226 U_8 66227
J_2 66227 P_{10} 66225 I_9 66225 $\mathbb{I}_1$ 66224 --- Mesure du 26 au 28 Mai
0000 001195 000186 013143 021768 084698 053521 054818 043325 060804 093980
0010 084324 064894 076163 018381 097304 057596 051610 052868 055560 057122
0020 052600 046532 039646 031740 026701 025498 024210 022675 021015 020345
0030 020963 019670 018280 017495 016706 015151 013633 013167 013552 014187
0040 014733 015573 017687 021675 026180 028217 026212 022322 018223 015434
0050 013691 011610 009281 007589 006461 005975 005758 005929 006084 006494
0060 007533 008767 010057 010157 010128 009917 010839 013608 017345 021663
0070 024622 026947 026522 024051 019218 013437 008918 005877 004039 003141
0080 002770 002314 002022 001662 001556 001410 001329 001338 001302 001327
0090 001266 001179 001024 000962 000983 000858 000867 000747 000778 000817
0100 000816 000823 000818 000876 000837 000874 000905 000849 000959 000865
0110 000849 000870 000854 000839 000879 000800 000794 000770 000735 000761
0120 000727 000717 000737 000745 000766 000681 000717 000742 000713 000692
0130 000682 000673 000654 000597 000553 000552 000530 000540 000526 000491
0140 000490 000477 000512 000535 000518 000473 000481 000416 000416 000468
0150 000501 000646 000764 000901 001111 001195 001366 001287 001374 001223
0160 001036 000894 000722 000538 000416 000346 000294 000257 000226 000246
0170 000264 000222 000217 000231 000210 000247 000208 000219 000231 000236
0180 000219 000195 000206 000181 000207 000219 000197 000193 000178 000200
0190 000176 000163 000161 000176 000161 000166 000160 000160 000171 000130
10
1
0,1
c/mn/10 keV
pour 10 Tonnes d'air
480
Cs
660
Zr
750
K
1460
La
1600
1966



0 68131 16380 J04 999 01 FATKCN 681590 132-3 46,51 T-- 1080 001

ENERGIE KEV	ELEMENT	ACTIVITE C/MN (10-3)	SCHEMA ELEMENT (10-4)	RENDEMENT DETECTEUR (10-6)	RESOLUTION KEV	RESULTAT PCI/T--	
0133,44	CE 144	0005299	1449	0017773	4,1	0021,325	***
0477,50	BE 7	0000529	1050	0001590	4,4	0045,317	**
0511,92	RH 106	0000474	2150	0001374	3,9	0016,321	**
0621,21	RH 106	0000086	0950	0000932	2,3	0018,005	*
0661,28	CS 137	0000213	8600	0000826	5,1	0002,917	**
0723,45	ZR 95	0000083	4300	0000699	2,6	0003,609	*
0756,57	ZR 95	0000184	5480	0000643	4,0	0003,871	*
0765,85	NB 95	0000525	9899	0000629	4,8	0011,000	**

C/MN 'ACTIF' : 0000087



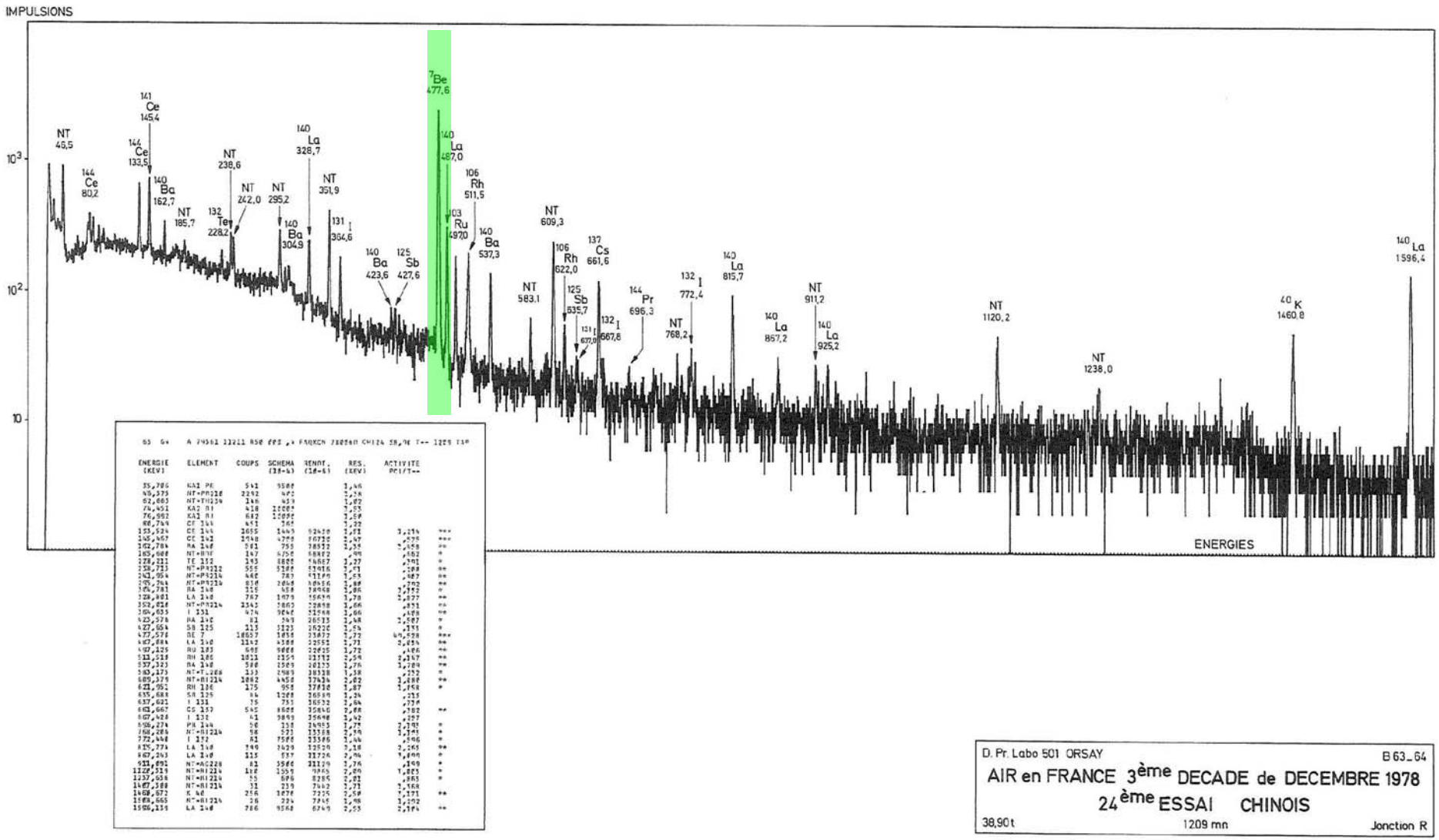
A 75257 15110 L41 999 ,5 FAPKCN 752700 ----- 13,50 T-- 3936 D-0

ENERGIE (KEV)	ELEMENT	COUPS VRAIS	SCHEMA (10-4)	RENDT. (10-6)	RES. (KEV)	ACTIVITE PCI/T--	
92,70	TH 234	4753	548	47509	2,61	15,448	***
133,51	CE 144	34545	1449	42748	2,19	49,011	***
144,94	CE 141	4646	4700	41088	2,35	2,794	***
175,86	NT	1028	10000	36658	1,92	,234	**
185,22	NT=RA226	2730	739	35390	1,98	8,727	***
238,27	NT= TH B	1043	4669	29117	2,06	,650	**
241,67	NT= RA B	707	760	28709	2,15	2,707	**
263,99	SE 75	957	5939	26623	2,19	,559	**
294,91	NT= RA B	1362	2048	24026	2,37	2,326	**
351,67	NT= RA B	1880	3930	20171	2,10	2,000	***
400,60	SE 75	269	1509	17568	2,02	,884	**
427,55	SB 125	1859	3119	16353	2,32	3,121	***
463,09	SB 125	496	1150	14945	1,91	2,472	**
477,52	BE 7	27604	1030	14429	2,29	191,569	***
497,03	RU 103	5412	9000	13776	2,26	4,810	***
583,25	NT=THC	296	3130	11412	2,22	,702	**
600,64	SB 125	665	1209	11020	2,03	4,272	**
609,29	NT=RAC	1894	4620	10834	2,94	3,167	***
621,99	RH 106	3173	950	10571	2,38	27,553	***
636,00	SB 125	414	1240	10295	2,37	2,870	**
661,59	CS 137	5946	8600	9824	2,43	5,960	***
696,48	PR 144	736	150	9244	2,35	46,675	**
724,14	ZR 95	8241	4300	8831	2,47	21,586	***
756,66	ZR 95	9845	5480	8391	2,47	21,297	***
765,74	NB 95	36584	9899	8276	2,54	44,416	***
834,81	MN 54	449	10000	7500	2,34	,525	**

D. Pr. Labo 501 ORSAY — B 1012/1.2

AIR PARIS - MONTSOURIS
1ère décade AVRIL 1975

13,50 T — 3936 mn — Jonction L



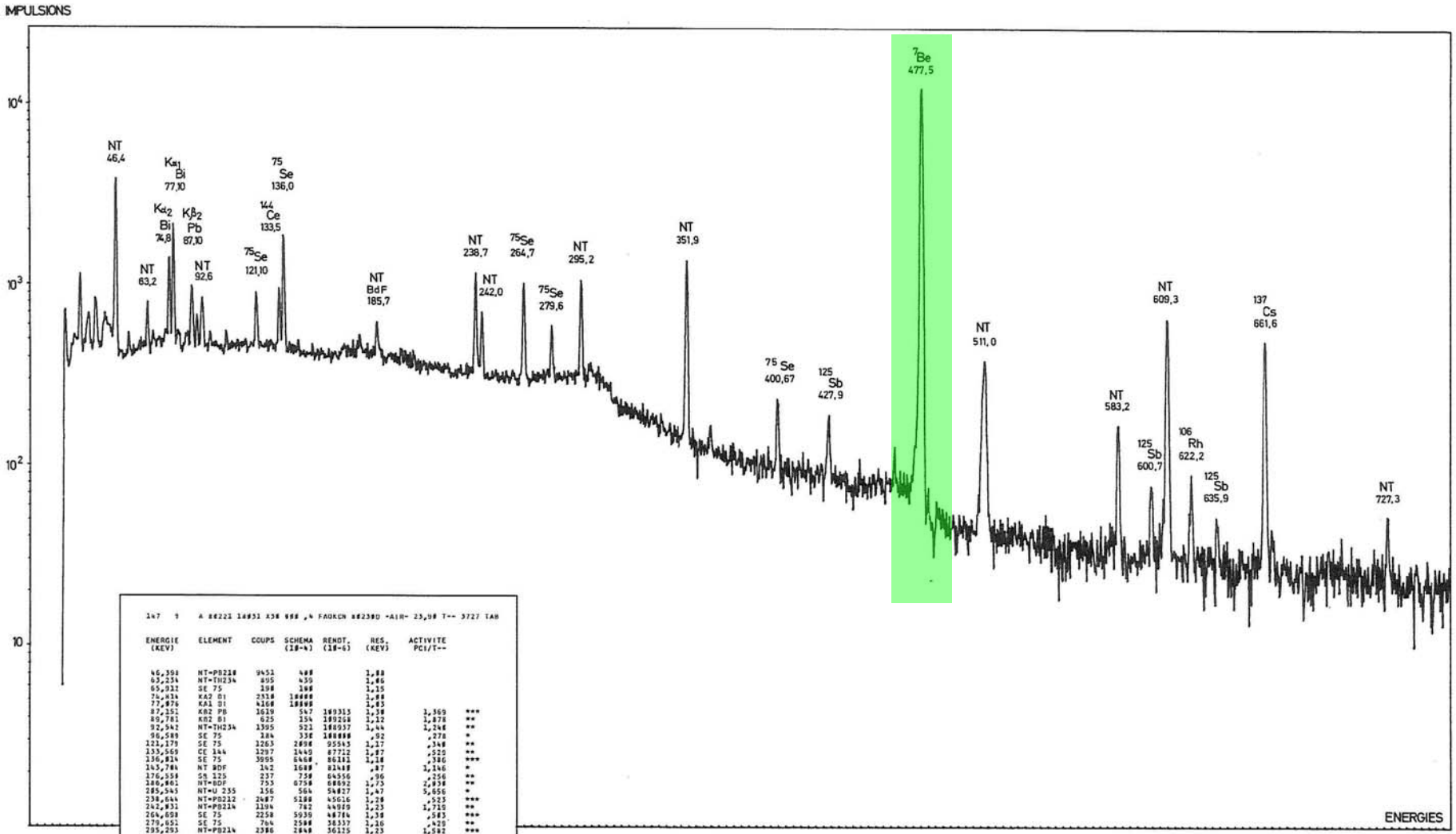
ENERGIE (KEV)	ELEMENT	COUPS	SCHEMA (38-43)	RENAT. (18-63)	RES. (KEV)	ACTIVITE PC/ST--	
35,786	KA1 PE	541	9589		1,446		
49,379	NT+PB12E	2292	442		2,24		
52,865	NT+TH234	148	459		1,02		
74,451	KA2 BI	418	25004		1,83		
76,962	KA1 BI	642	12956		1,69		
80,734	CE 144	451	765		1,22		
133,524	CE 144	1655	1443	92428	1,01	3,234	***
145,467	CE 142	2748	4769	76736	1,47	5,635	***
162,784	NA 140	541	755	28952	1,35	1,458	**
165,908	NT+B1E	147	4258	68882	.99	.552	*
178,211	TE 132	193	3885	46867	1,27	.391	*
238,713	NT+PB212	555	5386	57916	1,71	.588	**
241,954	NT+PB214	446	783	91189	1,53	.487	**
295,744	NT+PB214	836	2656	80456	1,88	.702	**
326,781	BA 140	155	458	18968	1,86	1,772	*
328,801	LA 140	767	1873	15639	1,78	1,827	**
352,016	NT+PB214	2342	5863	72858	1,66	.831	**
360,603	I 131	474	9647	52568	1,66	.458	**
425,578	BA 140	81	343	26513	1,48	1,587	*
427,654	SB 125	213	3123	26226	1,54	.331	*
477,574	BE 7	18607	3858	33877	1,72	49,538	***
487,884	LA 140	1147	4388	23552	1,71	2,454	**
497,125	RU 103	648	9868	22625	1,72	.486	**
511,518	RH 106	1821	1354	21113	1,54	2,167	**
537,323	BA 140	588	1589	20123	1,76	1,284	**
583,173	NT+T.2E8	133	2983	38338	1,58	.252	*
609,379	NT+B1214	3862	4458	37434	2,02	3,888	**
621,952	RH 106	175	955	37418	1,87	1,158	*
635,688	SB 125	44	1268	36849	1,24	.215	
637,621	I 131	35	731	36532	1,64	.234	
661,667	CS 137	545	8685	35846	1,68	2,182	**
667,424	I 132	41	9893	35698	1,42	.207	
696,274	PR 144	90	338	34985	1,72	1,191	*
768,284	NT+B1214	98	571	33368	1,58	1,341	*
772,448	I 132	81	7588	33306	1,44	.596	*
815,774	LA 140	749	3629	32520	2,18	2,265	**
867,243	LA 140	115	537	31726	2,04	1,899	*
911,091	NT+AC228	82	5866	31129	1,74	.349	*
1228,539	NT+B1214	148	1559	9865	2,06	1,825	*
1237,656	NT+B1214	55	896	8285	2,01	.865	*
1447,588	NT+B1214	31	239	7442	1,71	1,168	
1458,672	K 40	256	1676	7225	2,58	1,771	**
1588,665	NT+B1214	26	224	7245	1,98	1,252	
1596,134	LA 140	786	9568	6749	2,53	2,194	**

D. Pr. Labo 501 ORSAY B 63_64

AIR en FRANCE 3ème DECADE de DECEMBRE 1978

24ème ESSAI CHINOIS

38,90 t 1209 mn Jonction R



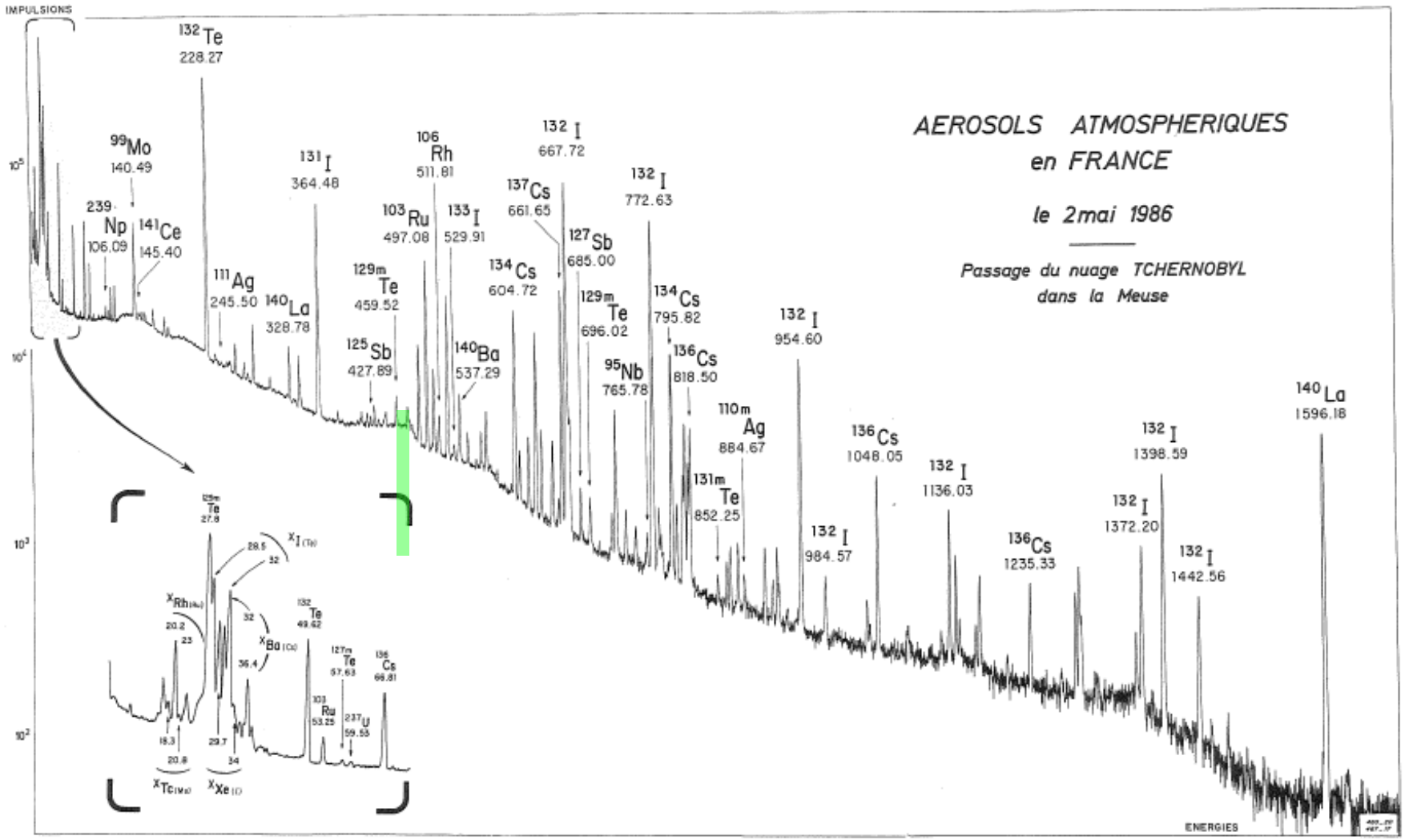
147 1 A 88223 14851 A3# 888 ,4 FAGNON 882380 -AIR- 23,9# T-- 3727 TAB

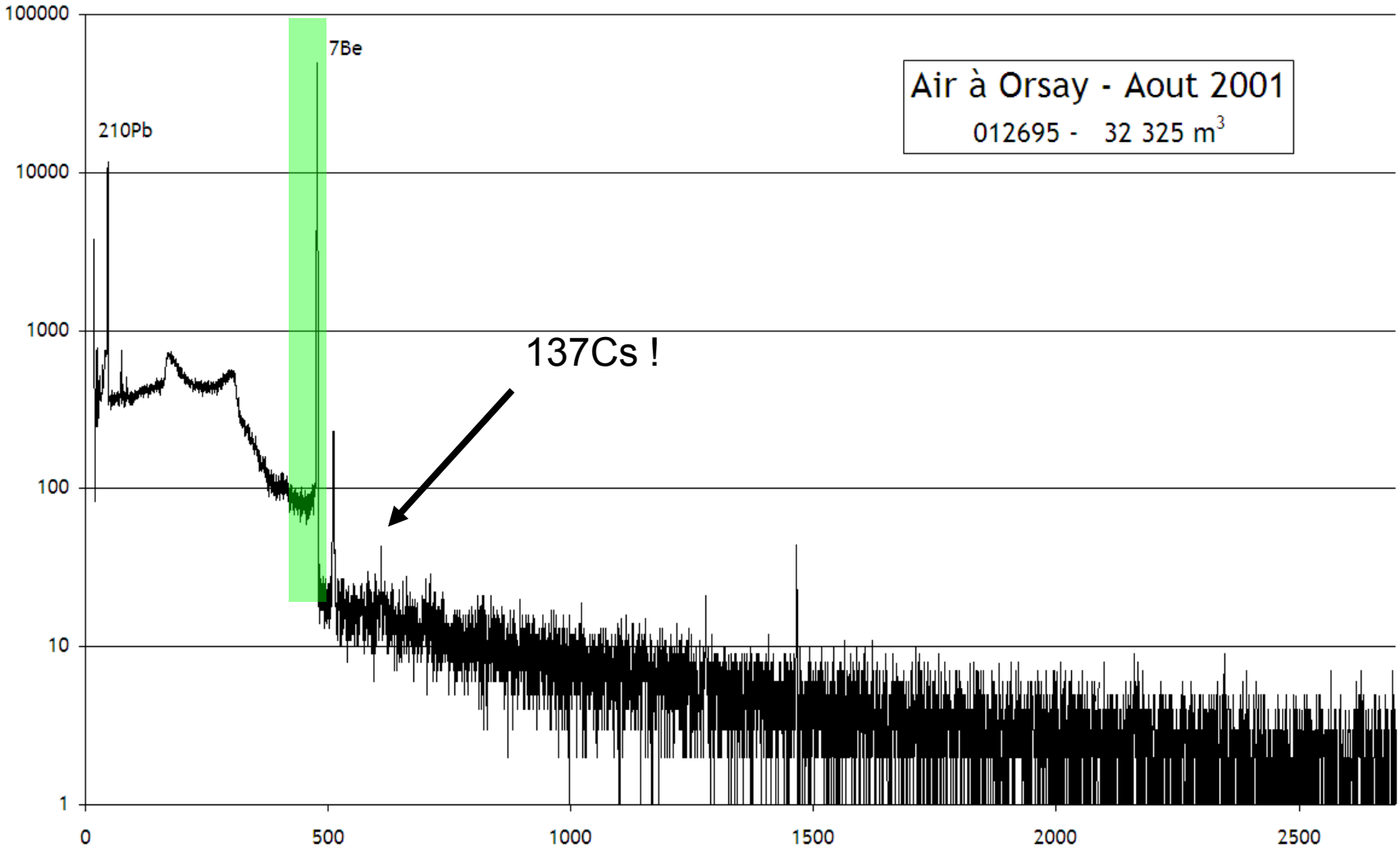
ENERGIE (KEV)	ELEMENT	COUPS	SCHEMA (10-4)	RENDT. (10-6)	RES. (KEV)	ACTIVITE PCI/T--	
46,398	NT-PB21#	9451	488		1,88		
63,234	NT-TH234	895	459		1,86		
65,932	SE 75	198	108		1,15		
74,834	KA2 BI	2334	10000		1,88		
77,876	KA1 BI	4164	18888		1,83		
87,151	KB2 PB	1619	547	189315	1,38	1,369	***
89,781	KB2 BI	625	154	189268	1,12	1,878	**
92,542	NT-TH234	1395	523	188937	1,44	1,296	**
96,589	SE 75	184	338	188888	,92	,278	*
121,179	SE 75	1263	2898	95545	1,17	,348	**
133,569	CE 144	1297	1448	87712	1,87	,525	**
136,814	SE 75	3995	6468	86182	1,18	,386	***
143,784	NT BDF	142	1688	81488	,87	1,146	*
176,358	SB 125	237	758	64556	,96	,256	**
186,861	NT-BDF	753	8758	60892	1,73	2,938	**
285,545	NT-U 235	156	564	54827	1,47	5,656	*
238,644	NT-PB212	2487	5188	45616	1,28	,523	***
242,831	NT-PB214	1194	782	44969	1,23	1,718	**
264,698	SE 75	2258	5939	48786	1,38	,543	***
279,651	SE 75	764	2588	38337	1,26	,428	**
295,253	NT-PB214	2386	2648	36135	1,23	1,582	***
299,833	NT-PB212	225	521	35525	1,83	,994	*
351,952	NT-PB214	3682	3869	29694	1,31	1,788	***
427,756	SB 125	571	5123	23549	1,48	,257	**
477,595	BE 7	43637	1458	185527	1,38	121,198	****
511,242	NT	2691		18421	2,55		***
583,192	NT-TL288	512	2989	15875	1,43	,545	**
688,688	SB 125	173	1919	15264	1,55	,386	*
680,521	NT-BI214	2357	4458	15487	1,43	1,784	***
622,128	RH 106	288	958	14614	1,45	,763	*
635,849	SB 125	73	1248	14215	1,44	,218	*
661,634	CS 137	1638	8688	13528	1,58	,886	***
727,361	NT-BI212	88	718	12835	1,17	,528	*
768,344	NT-BI214	174	523	11277	1,49	1,491	*

DPr Labo 501 ORSAY B147_09

AIR en FRANCE en MAI 1980

23,90 t 3727 mn Jonction X


IMPULSIONS
AEROSOLS ATMOSPHERIQUES
en FRANCE
le 2mai 1986
Passage du nuage TCHERNOBYL
dans la Meuse
132 Te 228.27
99 Mo 140.49
239 Np 106.09
141 Ce 145.40
111 Ag 245.50
140 La 328.78
131 I 364.48
129m Te 459.52
125 Sb 427.89
106 Rh 511.81
103 Ru 497.08
133 I 529.91
140 Ba 537.29
132 I 667.72
137 Cs 661.65
134 Cs 604.72
127 Sb 685.00
129m Te 696.02
132 I 772.63
134 Cs 795.82
136 Cs 818.50
95 Nb 765.78
132 I 954.60
110m Ag 884.67
131m Te 852.25
136 Cs 1048.05
132 I 984.57
132 I 1136.03
136 Cs 1235.33
132 I 1372.20
132 I 1398.59
132 I 1442.56
140 La 1596.18
ENERGIES


100000
10000
1000
100
10
1
0
500
1000
1500
2000
2500
7Be
210Pb
Air à Orsay - Aout 2001
012695 - 32 325 m3
137Cs !

From 1960 to 1980, the volume of sampled air x 50

- **Nowadays : 70 000m^3/5 days.**

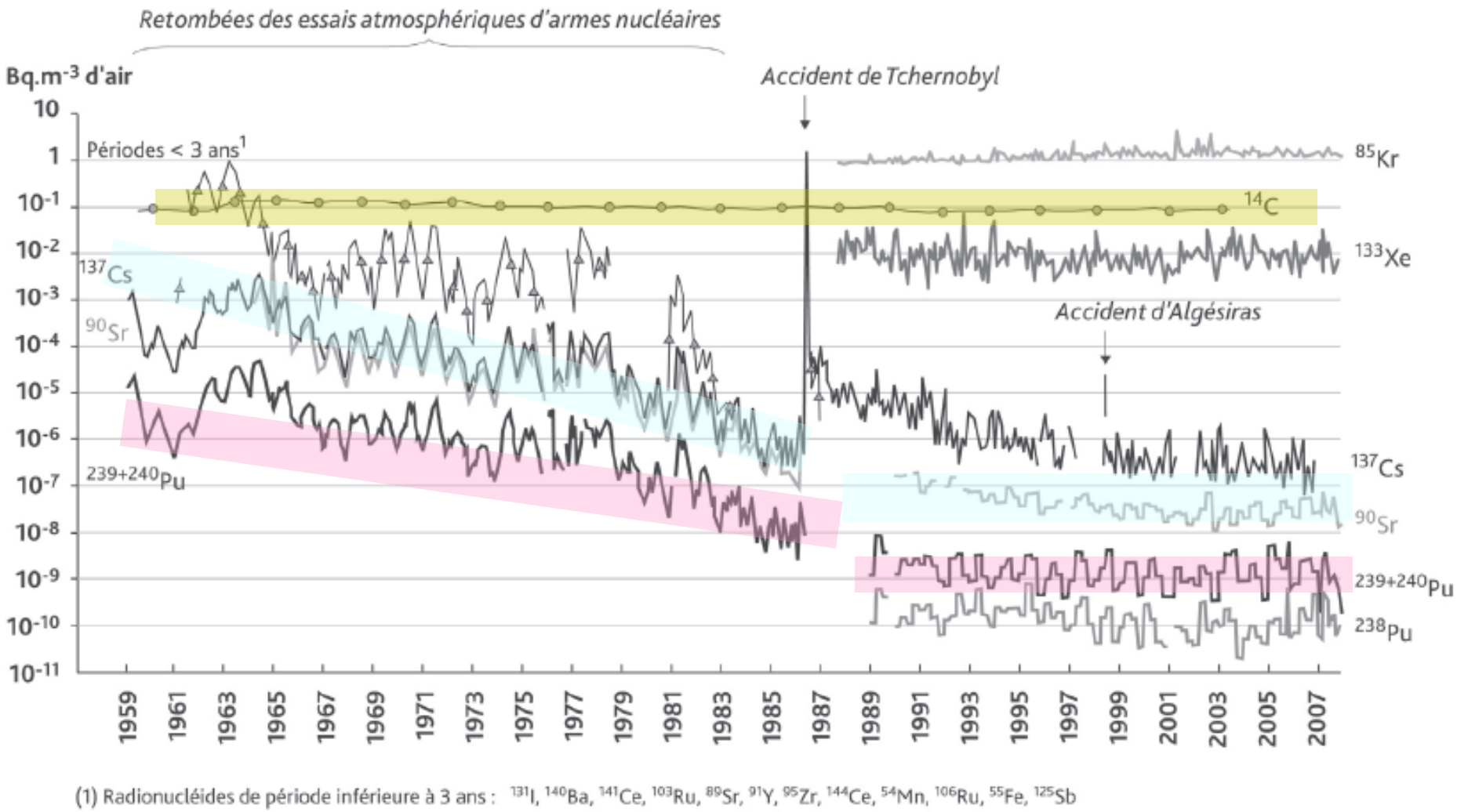
From 1970 to 1986 measuring times x 5

From 1980 to 1986 measuring efficiency x 4

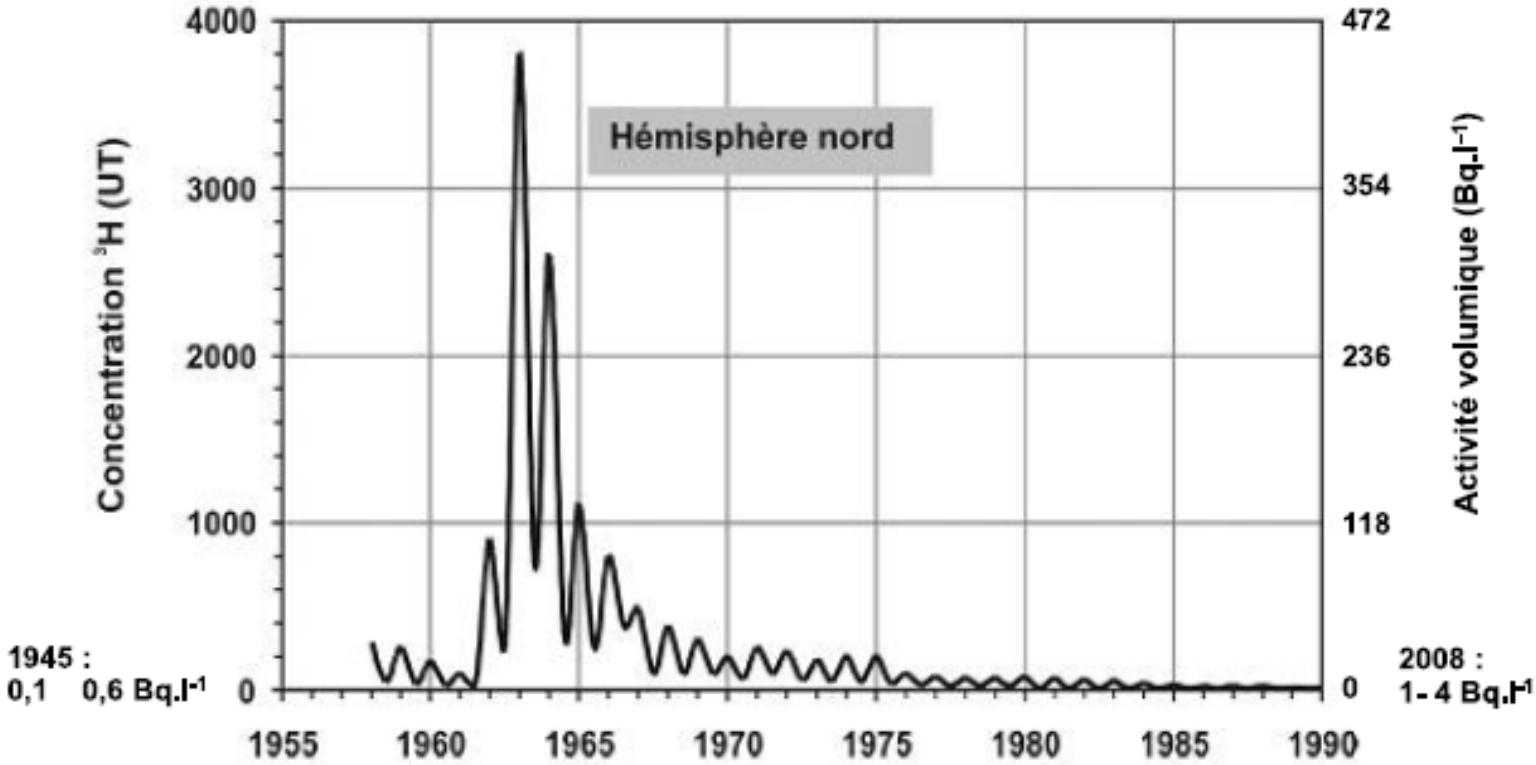
- **Then again x2 between 1986 and 2002**

From 1993 to 2004 detector backgrounds / 10

All this improvements were required : from 1960 to 2008 artificial radioactivity is / **10 000**



(1) Radionucléides de période inférieure à 3 ans : ^{131}I, ^{140}Ba, ^{141}Ce, ^{103}Ru, ^{89}Sr, ^{91}Y, ^{95}Zr, ^{144}Ce, ^{54}Mn, ^{106}Ru, ^{55}Fe, ^{125}Sb



Courbe lissée représentant la teneur moyenne en ^{3}H des précipitations au-dessus de la surface continentale de l'hémisphère Nord. ***Source = AIEA Isotope hydrology, 2006***

The low level monitoring of the radioactivity in the aerosols allows ton understand and anticipate the contamination of the environment, as the atmosphere is generally the first vehicle of this contamination.

These observations are particularly interesting over long periods as the reference to a previous state is possible.

The knowledge acquired in the routine situation allows to better forecast the behaviour of radionuclides in incidental situations

The possibility to measure the “radioactivity background” is part of the environmental monitoring program of IRSN

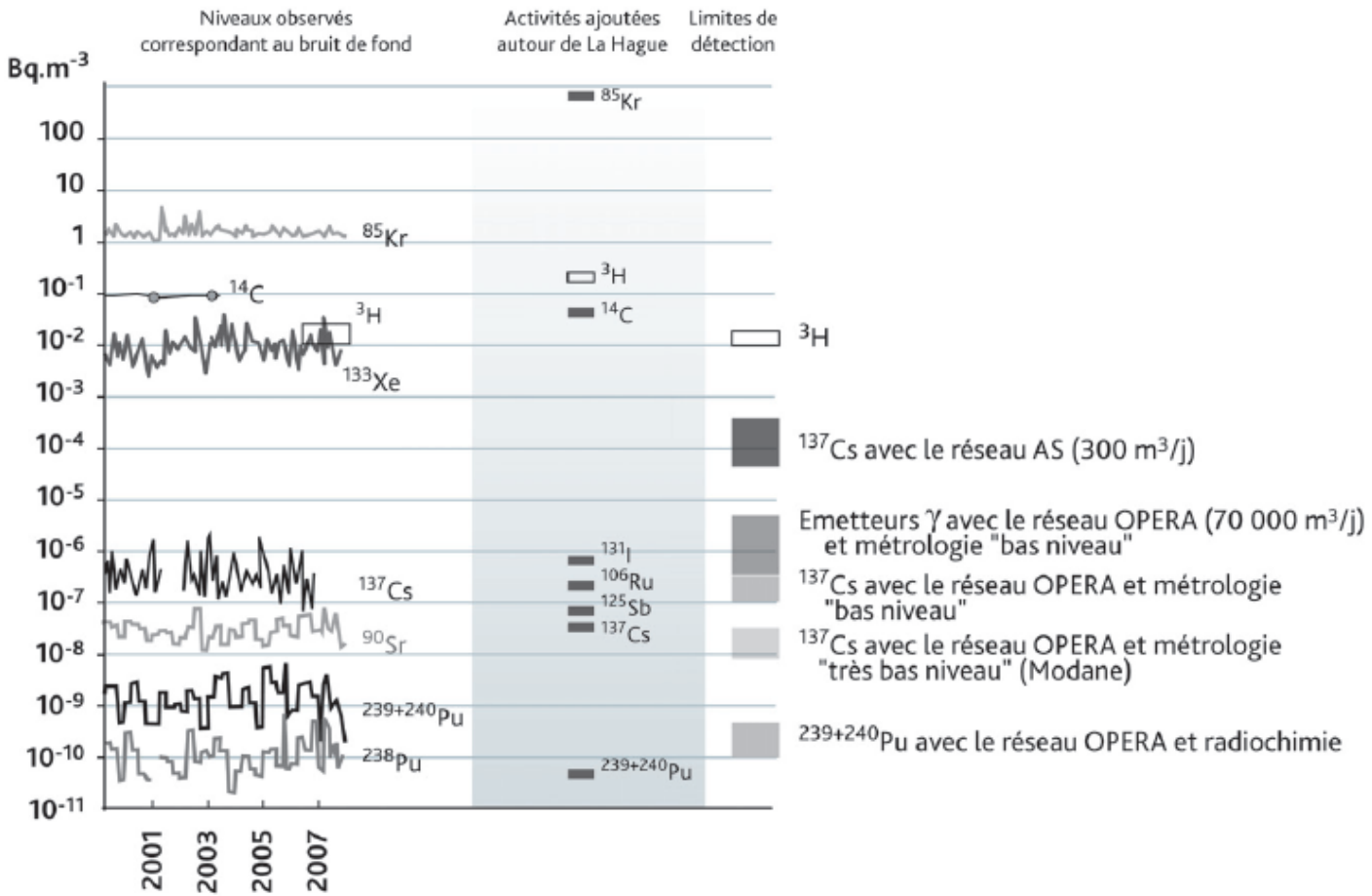


Figure 3 – ***Activités volumiques actuelles des radionucléides artificiels ou semi artificiels dans l'air, activités moyennes potentiellement ajoutées par les rejets de La Hague au niveau du groupe de référence, limites de détection de différents systèmes d'acquisition.***

Present volumic activities of anthropogenic or semi anthropogenic radionuclides, mean potentially added activities by La Hague releases for the reference group, detection levels of various acquiring systems.

Dominant impact of atmospheric Nuclear testing

Nowadays it is still the dominant component – even if it has largely decreased

Exception for a few samples taken close of nuclear facilities

The "research monitoring" job is getting tough !

even with the best available tools it is getting difficult to measure the radioactivity in the environment

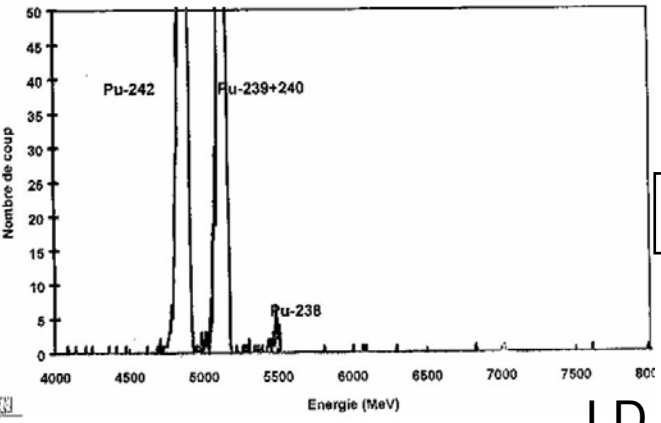
good thing, isn't it ?

Spectrométrie alpha ^{241}Am, ^{238}Pu, $^{239+240}Pu$

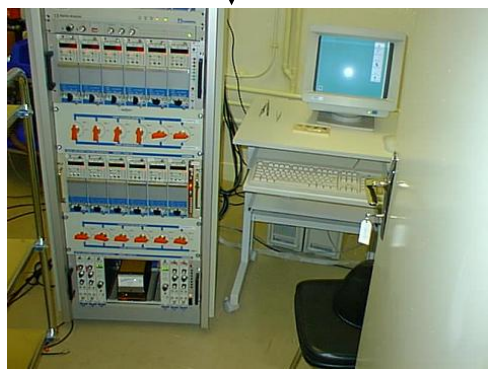
50-200 g d'échantillon sec

Extraction sélective (minéralisation, co-précipitations, purification sur colonnes)

3 à 4 semaines de préparation
puis électrodéposition



Comptage 2 semaines



LD ~ 1 mBq/kg cendres

Transformation du carbone de l'échantillon en benzène pour la mesure ^{14}C par scintil. liq.

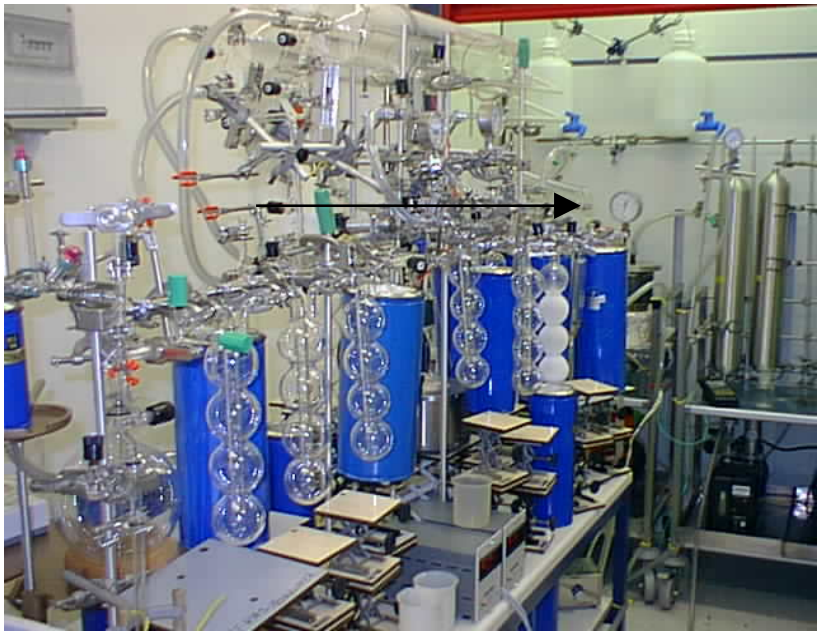
$$C + O_2 \longrightarrow CO_2$$

$$2CO_2 + 10Li \longrightarrow Li_2C_2 + 4\,Li_2O$$ *carburation*

$$Li_2C_2 + 2H_2O \longrightarrow C_2H_2 + 2LiOH$$ *hydrolyse du carbure de Li*

$$3C_2H_2 \xrightarrow[\text{contenant } Cr^{3+}]{\text{catalyseur}} C_6H_6$$ *trimérisation*

(polymérisation de l'acétylène en benzène par catalyse)



+ scintillant

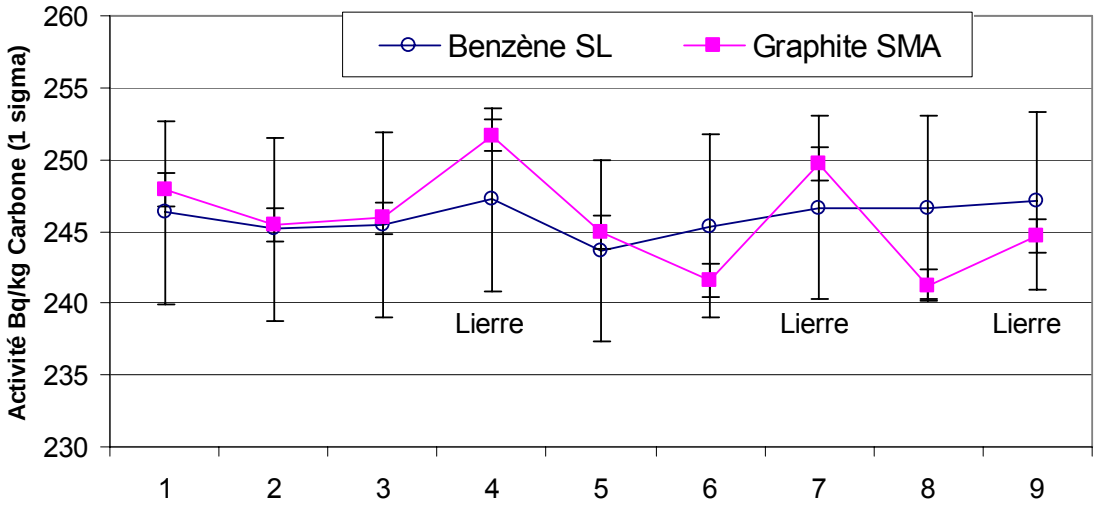
Mesure par scintillation liquide

Analyse de ^{14}C au LMRE

Prise d'essai de 5 à 20g sec
Synthèse de benzène et mesure par scintillation liquide

Difficulté rencontrée pour les solides à faible teneur en MO

=> mise en œuvre de la mesure par accélérateur au **LMC14 (prise d'essai de 10 à 100 mg)**



-> hétérogénéité des échantillons pour les faibles prises d'essais (<<1g)

Etudes récentes

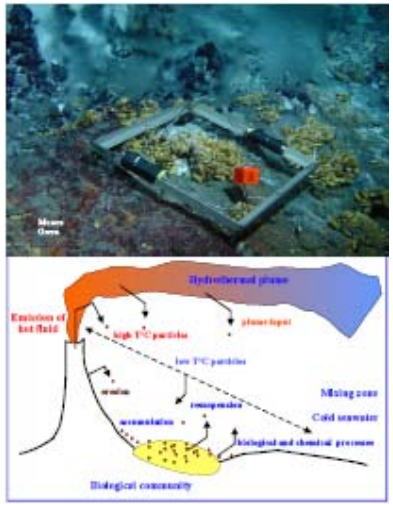

Compréhension de l’environnement

Recherche « fondamentale » : moules Hydrothermales

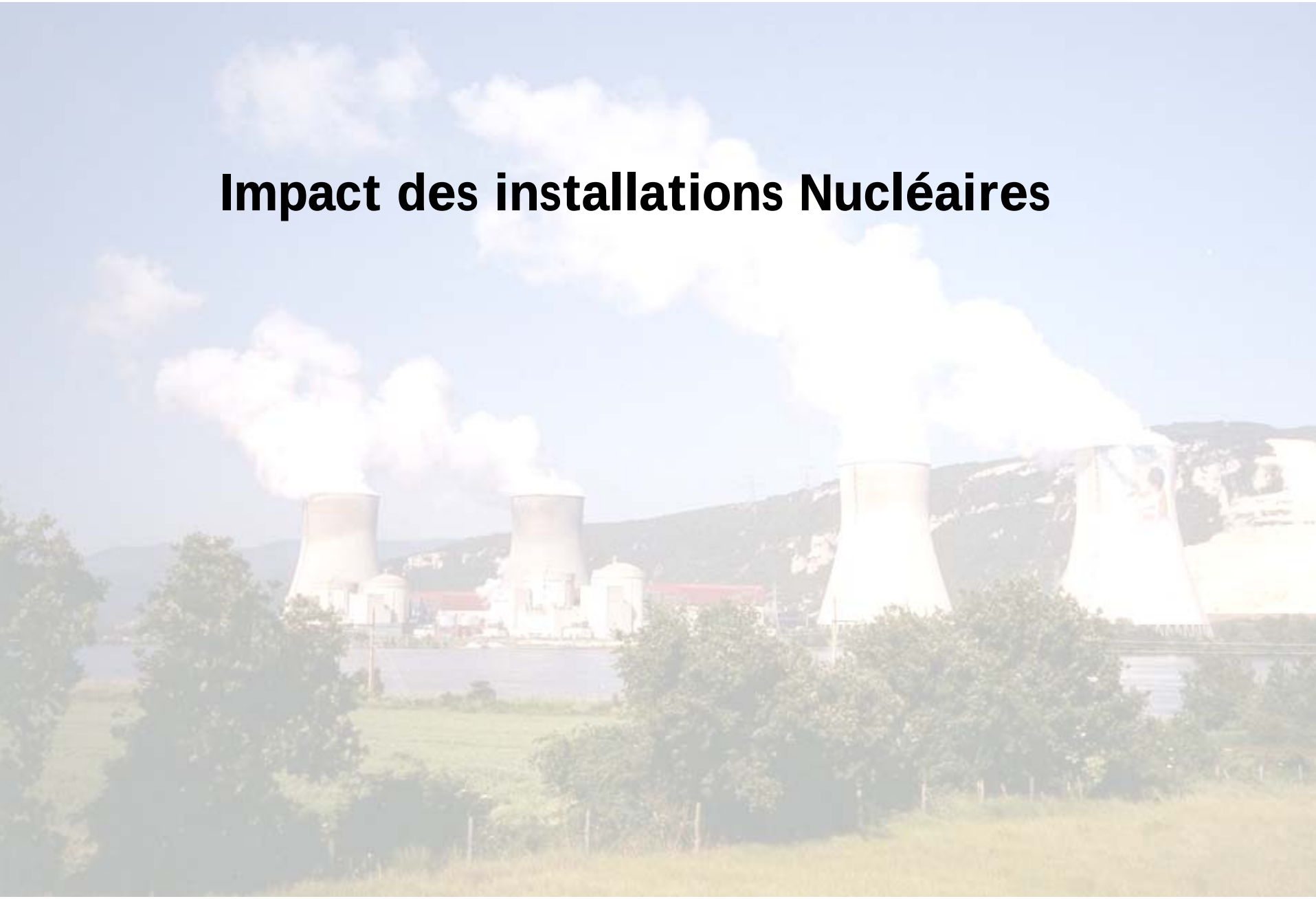
Organismes dans les fumerolles des fosses vivent dans des milieux radioactifs

Objectifs :

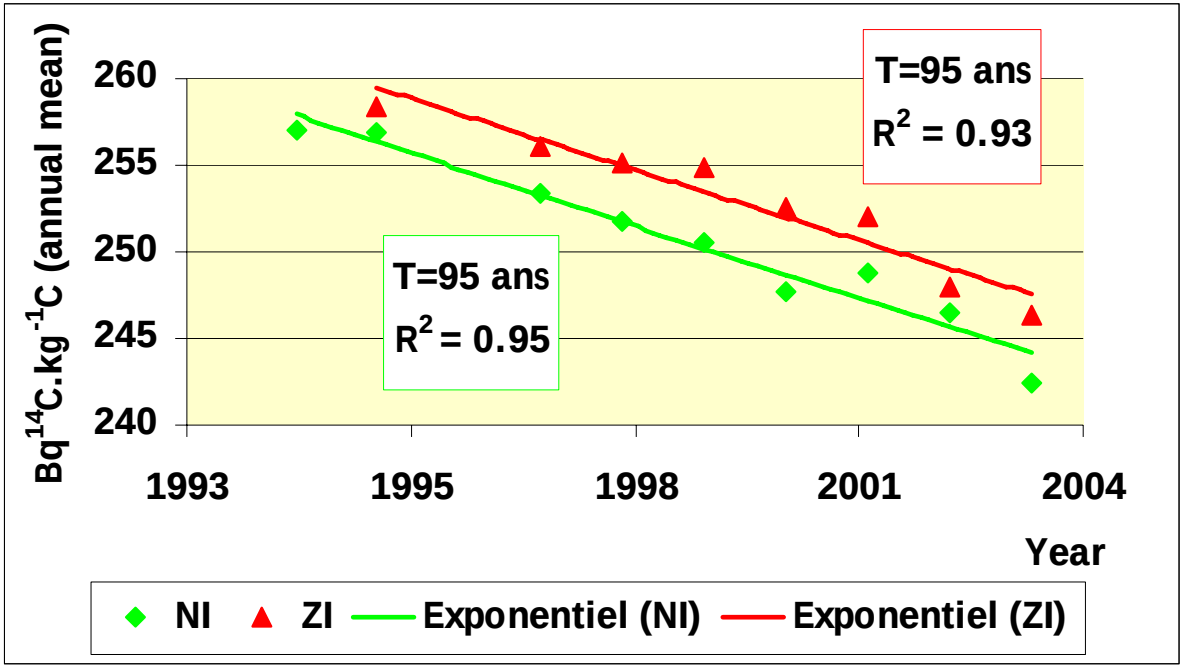
- Compréhension de leurs modes de vie
- Impact de la contamination chronique, mécanismes de protection



Impact des installations Nucléaires



Diminution du ^{14}C en milieu terrestre



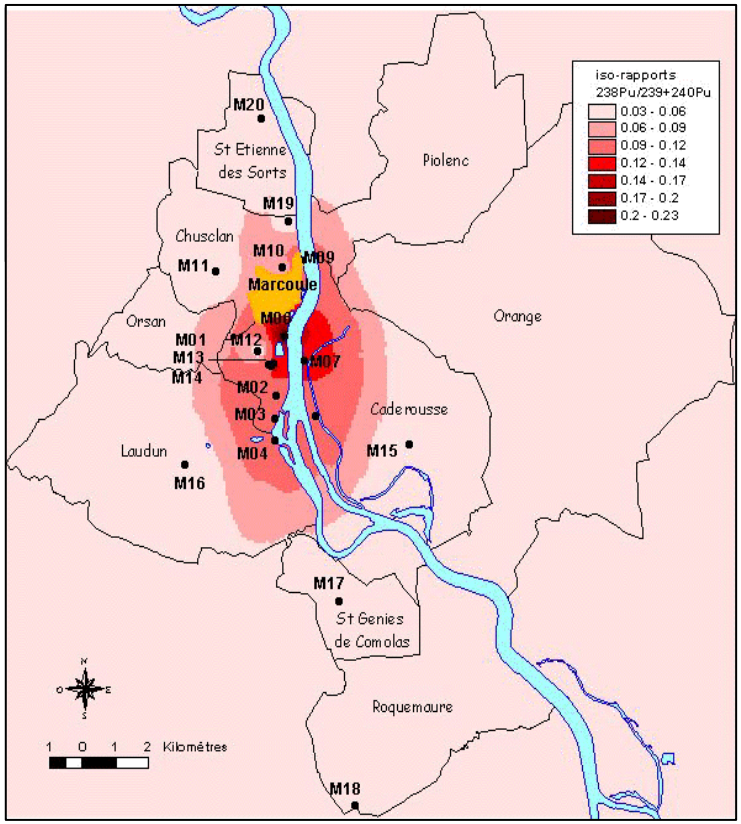
- L'activité spécifique diminue avec une constante de temps équivalente à une période apparente de 95 ans

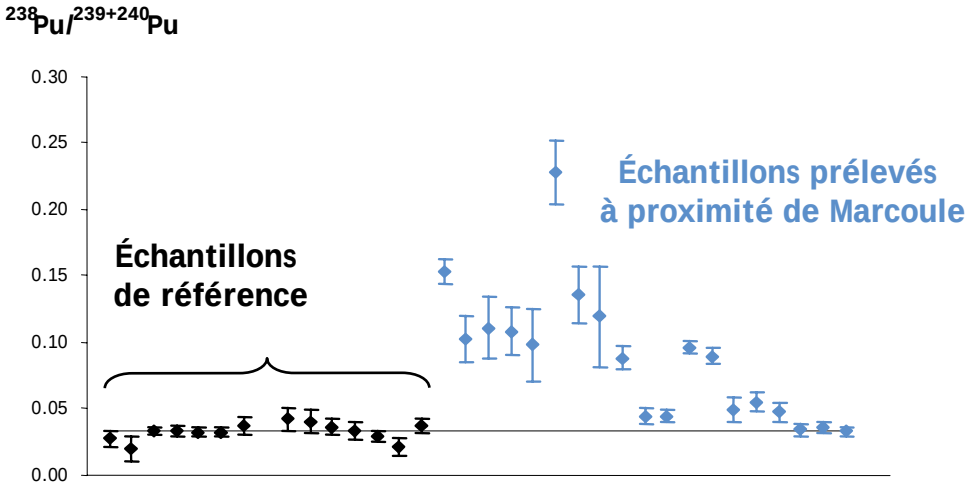
- **Le niveau moyen (NI) est en 2003 de 242 +/- 6 Bq $^{14}C.kg^{-1}C$**

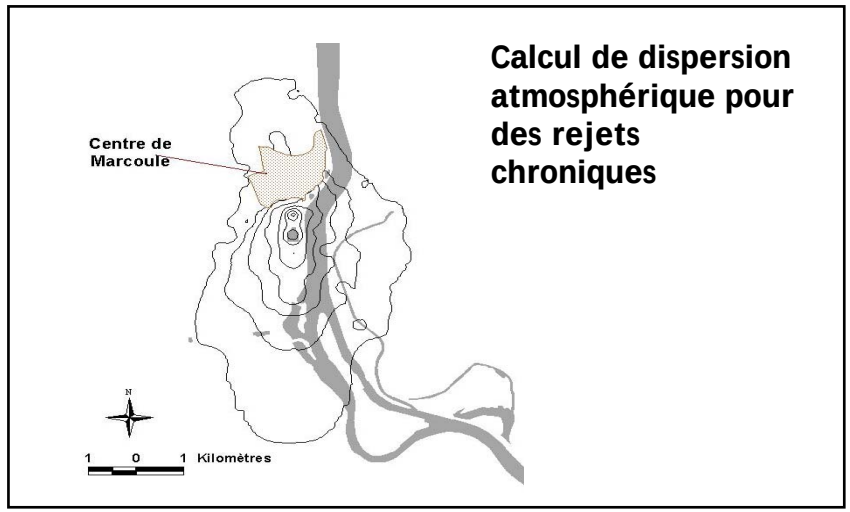
- L'hypothèse d'équilibre des rapports isotopiques $^{14}C/C$ est confirmée

Rapports d 'activités $^{238}Pu/^{239+240}Pu$ mesurés dans les mousses terrestres prélevées autour de Marcoule

Mousses terrestres : bioindicateurs







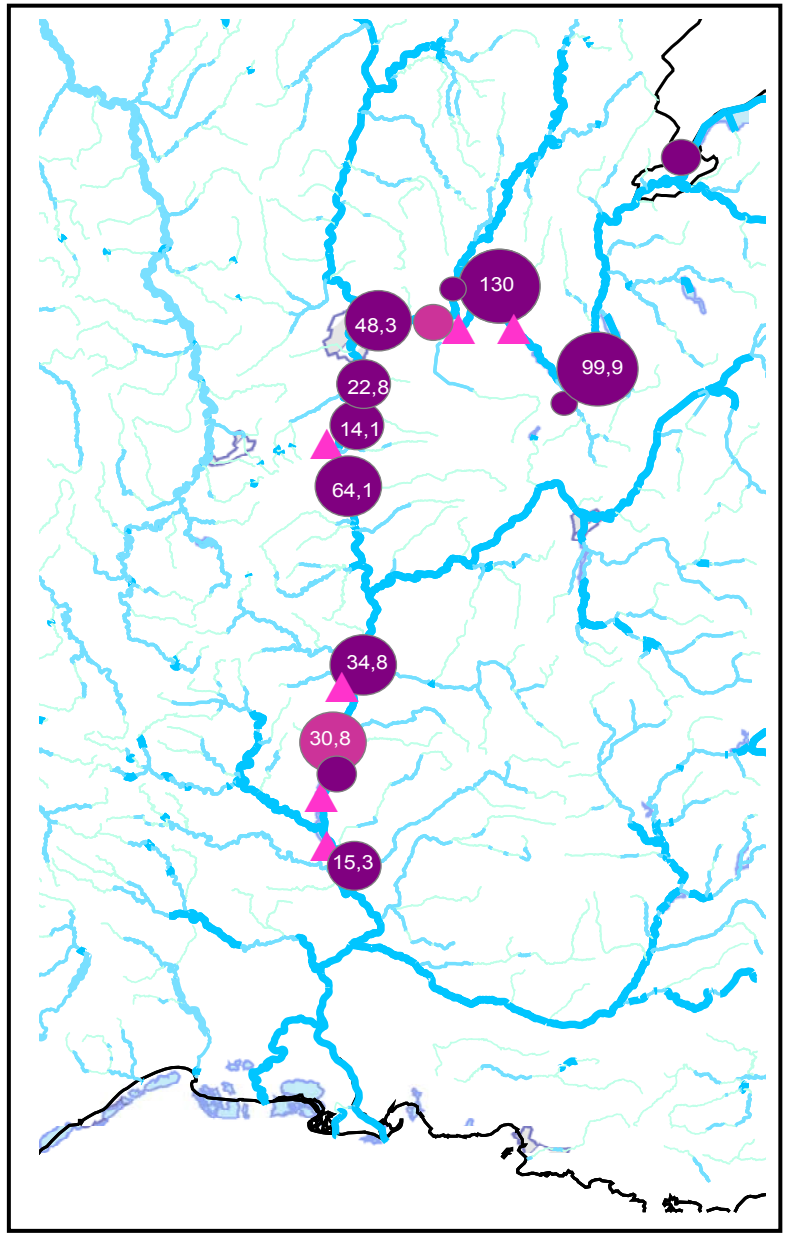
Tritium organique dans le Rhône

Figure 2

Activité en Tritium organique dans l'Eau de Combustion de phanérogames immergés en Bq.L^{-1}

- inf à 2 Bq.L^{-1}
- de 2;1 à 10 Bq.L^{-1}
- de 10,1 à 30 Bq.L^{-1}
- de 30,1 à 80 Bq.L^{-1}
- sup à 80 B.L-1
- Sites nucléaires

0 15 30 60 Kilomètres



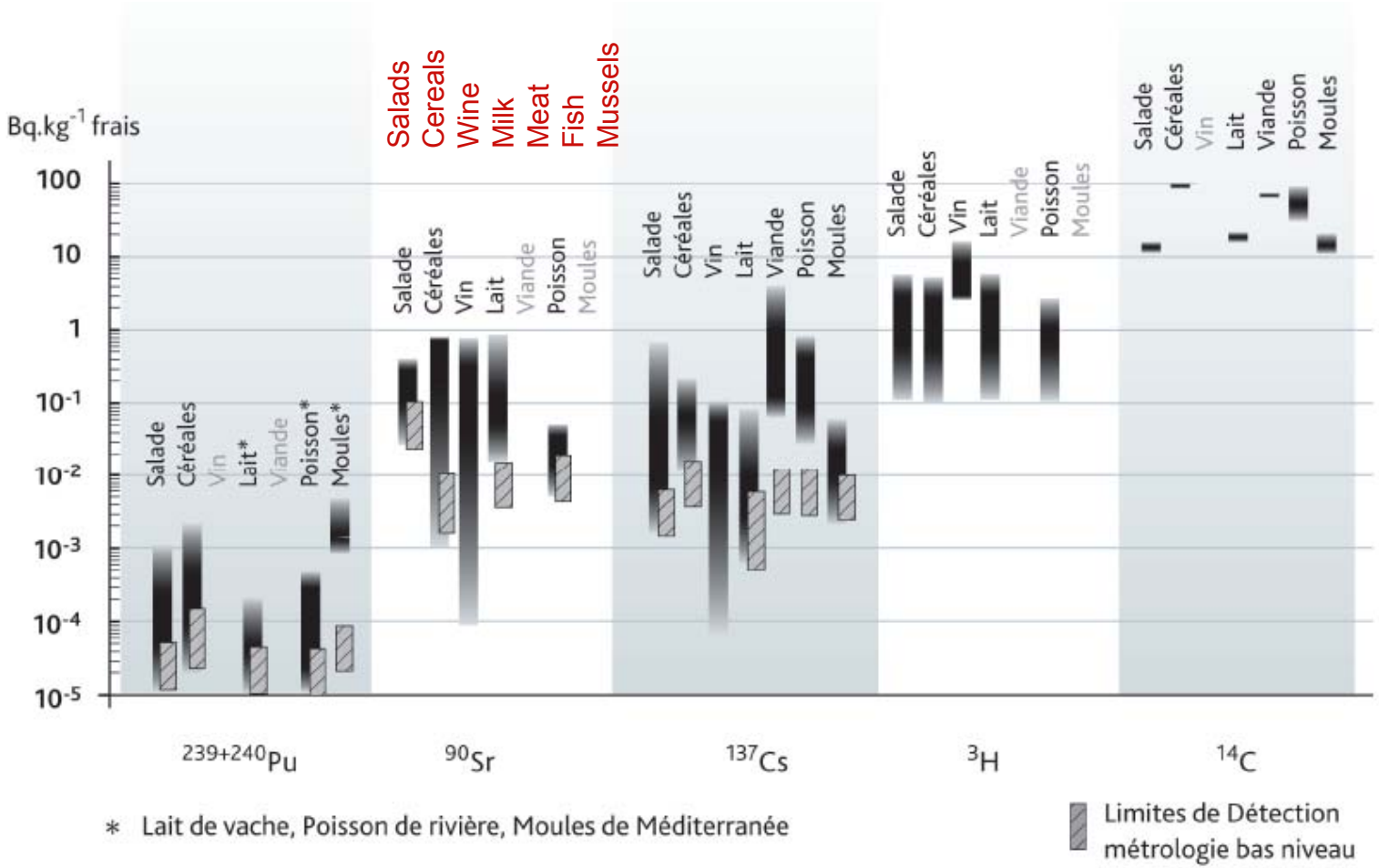


Figure 4 – *Gammes d'activités des principaux radionucléides artificiels dans les denrées et gammes des limites de détection obtenues au moyen des meilleures techniques disponibles à l'IRSN.*

Activity ranges of anthropogenic radionuclides in foodstuff and detection levels using best tools available at IRSN.

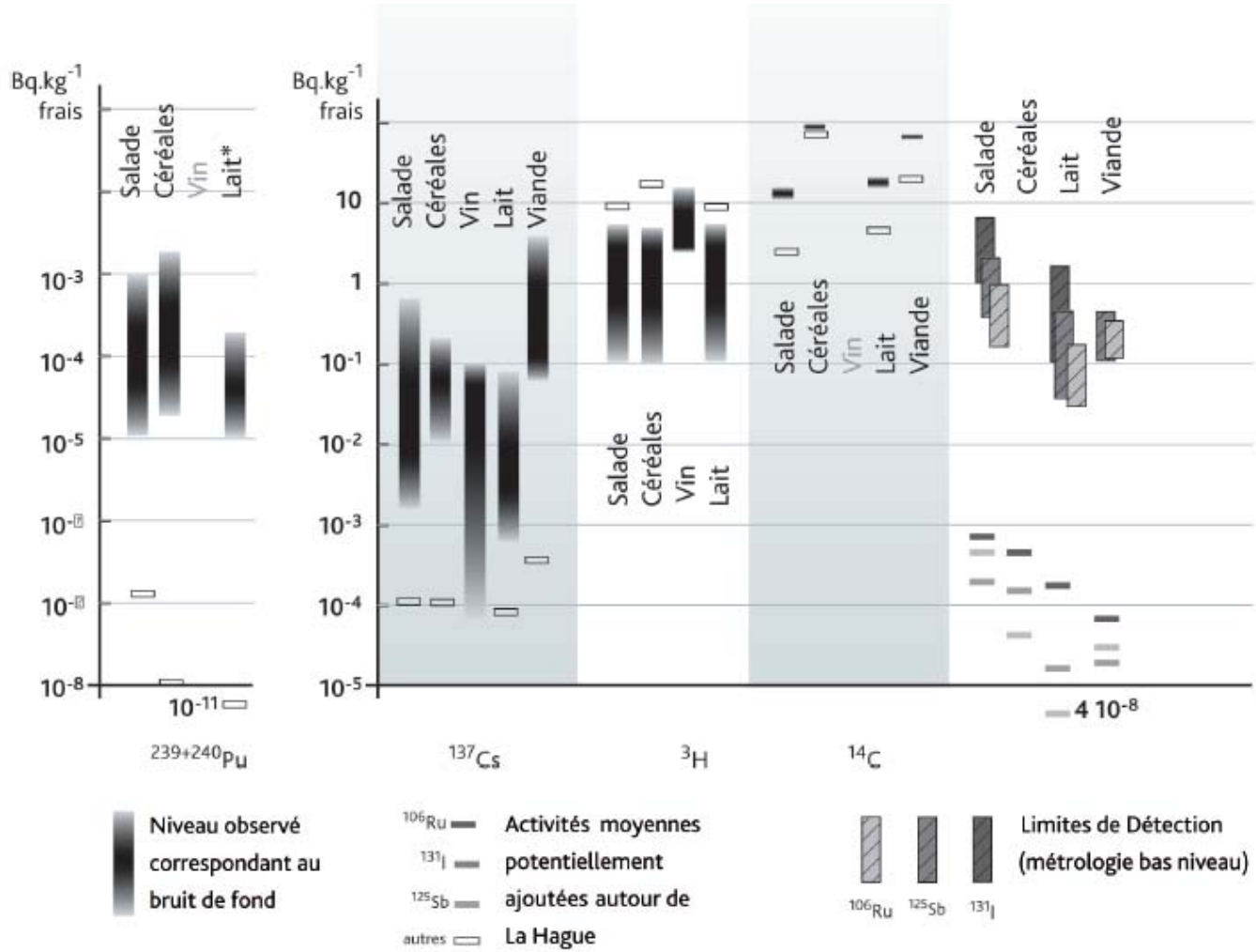


Figure 5 – ***Comparaison entre les activités relevant du bruit de fond ou rémanentes des retombées anciennes et les activités potentiellement ajoutées au niveau du groupe de référence de La Hague.***

Comparison between background activities and potentially added activities for the reference group of population around La Hague.

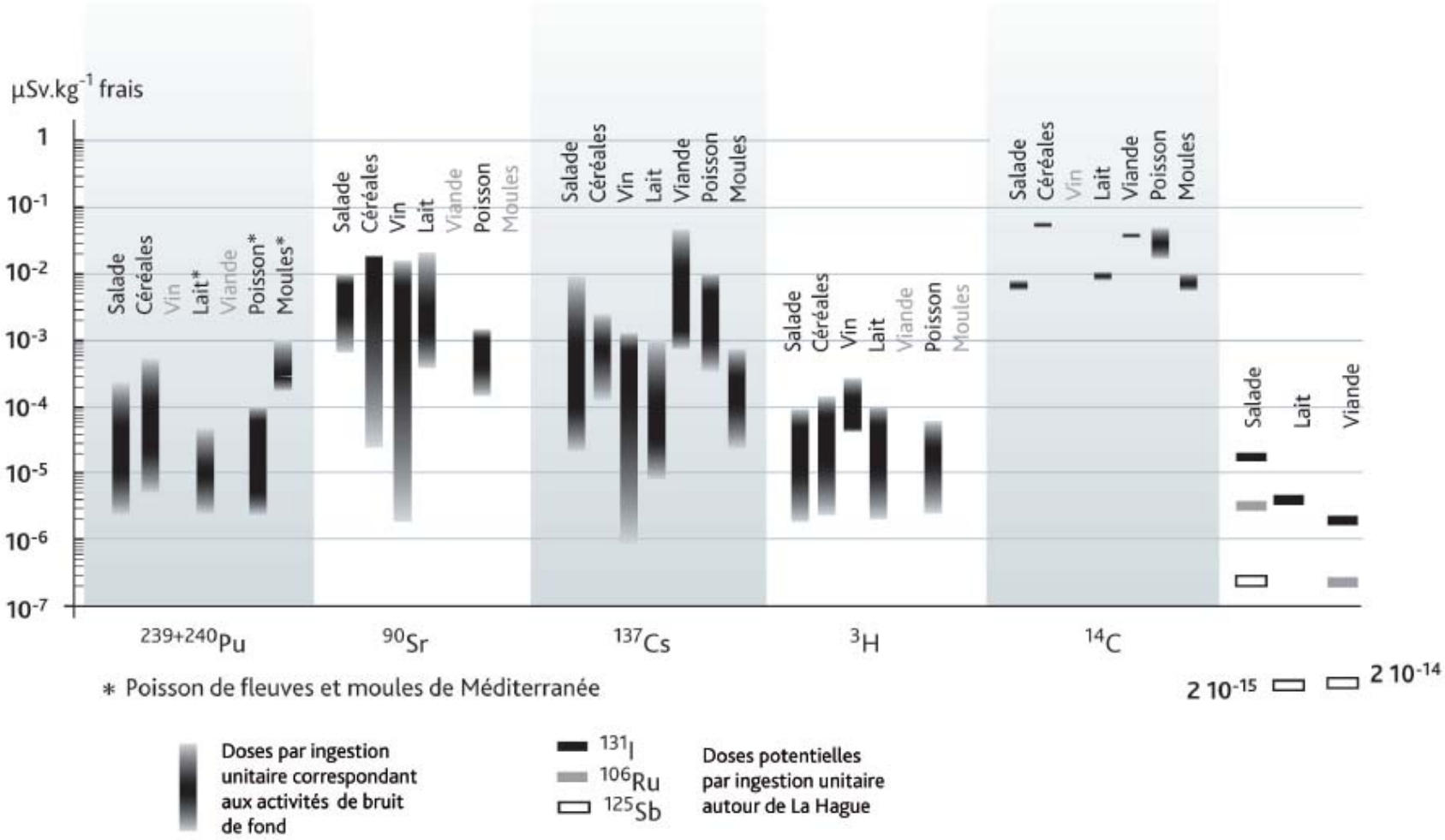


Figure 6 – *Dose efficace relative à l'ingestion par un adulte de 1 kilogramme de denrée.*
Effective doses linked to the ingestion of one kilogramme of foodstuff by an adult.