Detection and Quantification of the Gaseous and Particulate Fukushima Fission Products at Orangeburg, South Carolina

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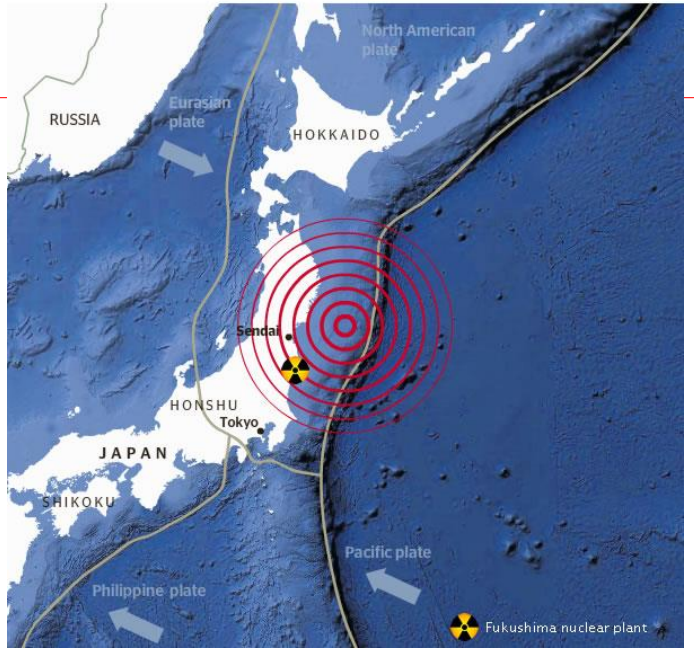
2011 Tohoku Earthquake and Tsunami

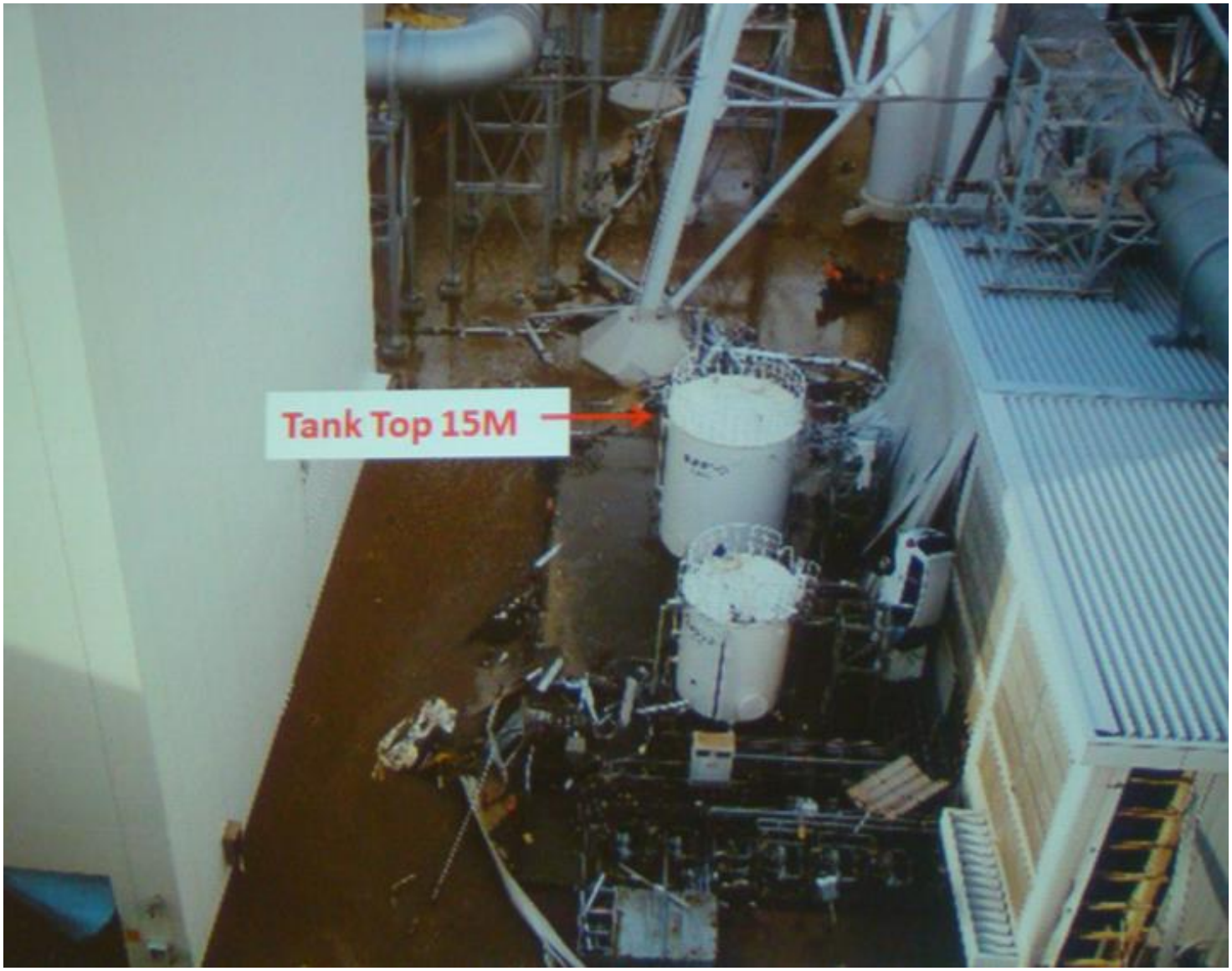
- At 14:46, March 11, 2011, a 9.0 Richter-scale earthquake struck at 32 km underwater depth, 180 km from the coast of Honshu island.
- Forty-one minutes later (15:27), tsunami waves with maximum height of 14 m arrived and hit the northeast coast of Japan.
- The impacts up and down the northeast coast resulted in 15,885 deaths, 2,623 missing, and 6,148 injured. Vast damage and destruction to buildings, highways, and constructions.
- It was the most powerful known earthquake ever happened in Japan, and one of the five most powerful earthquakes in the world since modern record-keeping began in 1900.



Incoming Wave behind Reactor Building (3/11 15:42)

Incoming Wave behind Reactor Building (3/11 15:43:54)

Incoming Wave behind Reactor Building (3/11 15:47)

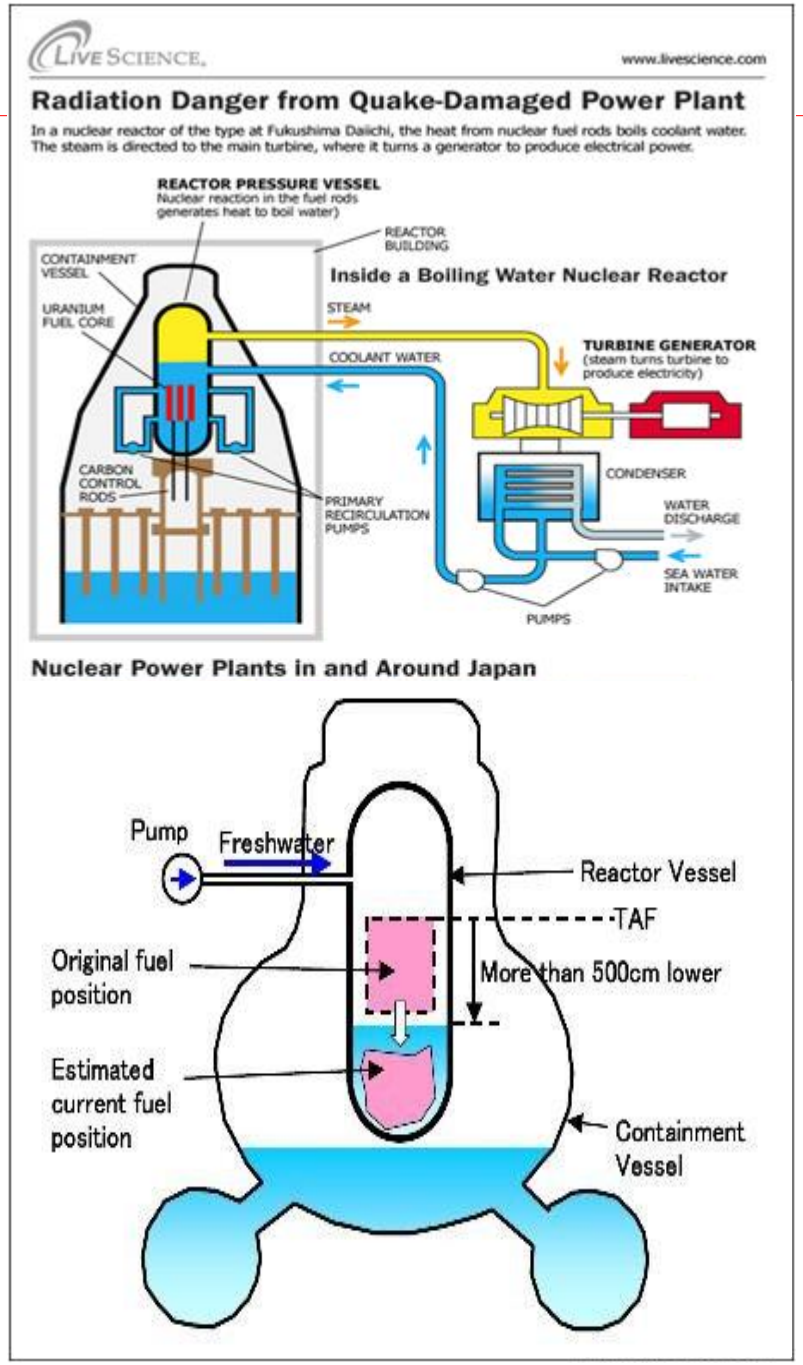


Fukushima Nuclear Power Plant on March 13

from the US Nuclear Energy Institute

2011 Fukushima Daiichi Nuclear Accident

- Fukushima Daiichi Power Plant Complex, run by TEPCO, contains 6 BWR with the total power of 5,480 GWe (GE Mark I containments). Units 1-3 were in operation, Units 4-6 not in operation on 03/11/2011.
- The scram took place successfully in Reactors 1, 2, and 3 right after the quake (14:46). The Tokyo electricity grid was damaged and the site lost the AC power. Emergency generators kicked in to continue providing power for cooling the reactors.
- The tsunami waves inundated the site 41 min after the quake. The seawall can only protect the sites from 5.7 m waves. Only one diesel generator survived from the flood, and continued to cool Units 5 and 6. Reactors 1 to 4 lost cooling water.
- LOCA occurred. The steam discharged from the RPV. The zircaloy fuel cladding of the reactor core was exposed to the air and produced large amount of H_2. Explosions happened to the buildings of Units 1, 2, 3. Explosion occurs to 4 due to H_2 mitigated from Unit 3.



Event Sequence at the Fukushima Nuclear Reactors Following the Earthquake

Events	Unit 1	Unit 2	Unit 3
Loss of AC power	+ 51 min	+ 54 min	+ 52 min
Loss of cooling	+ 1 hr	+ 70 hr	+ 36 hr
Water level down to top of fuel*	+ 3 hr	+ 74 hr	+ 40 hr
Core damage starts*	+ 4 hr	+ 77 hr	+ 42 hr
Fire pumps with fresh water	+ 15 hr	--	+ 43 hr
Hydrogen explosion (not confirmed for unit 2)	+ 25 hr (service floor)	+ 87 hr (suppression chamber)	+ 68 hr (service floor)
Fire pumps with seawater	+ 28 hr	+ 77 hr	+ 46 hr
Off-site electrical supply	+ 11-15 d		
Fresh water cooling	+ 14-15 d		

** Estimated value.*
Start time (Tsunami hit the coast): 15:27, March 11, 2011
From World Nuclear Association, Feb 22, 2012

Radioactive Release and Event Scale

IRSN (French Institute of Radioprotection and Nuclear Science)		Japanese Nuclear Emergency Response Headquarter	
Until 03/22/11		*Until 06/11*	
Rare gases:	2×10^{18} Bq	^{131}I:	1.6×10^{17} Bq
Iodine:	2×10^{18} Bq	^{137}Cs:	$0.6\text{-}1.5 \times 10^{16}$ Bq
Cesium:	3×10^{16} Bq		
Tellurium:	9×10^{16} Bq		

Nuclear Accident	Date	^{131}I-Equivalent Activity (Bq)	International Nuclear and Radiological Event Scale
Three Mile Island	03/28/1979	1.1×10^{12}	5
Chernobyl	04/26/1986	5.2×10^{18}	7
Fukushima	03/11/2011	7.7×10^{17}	7

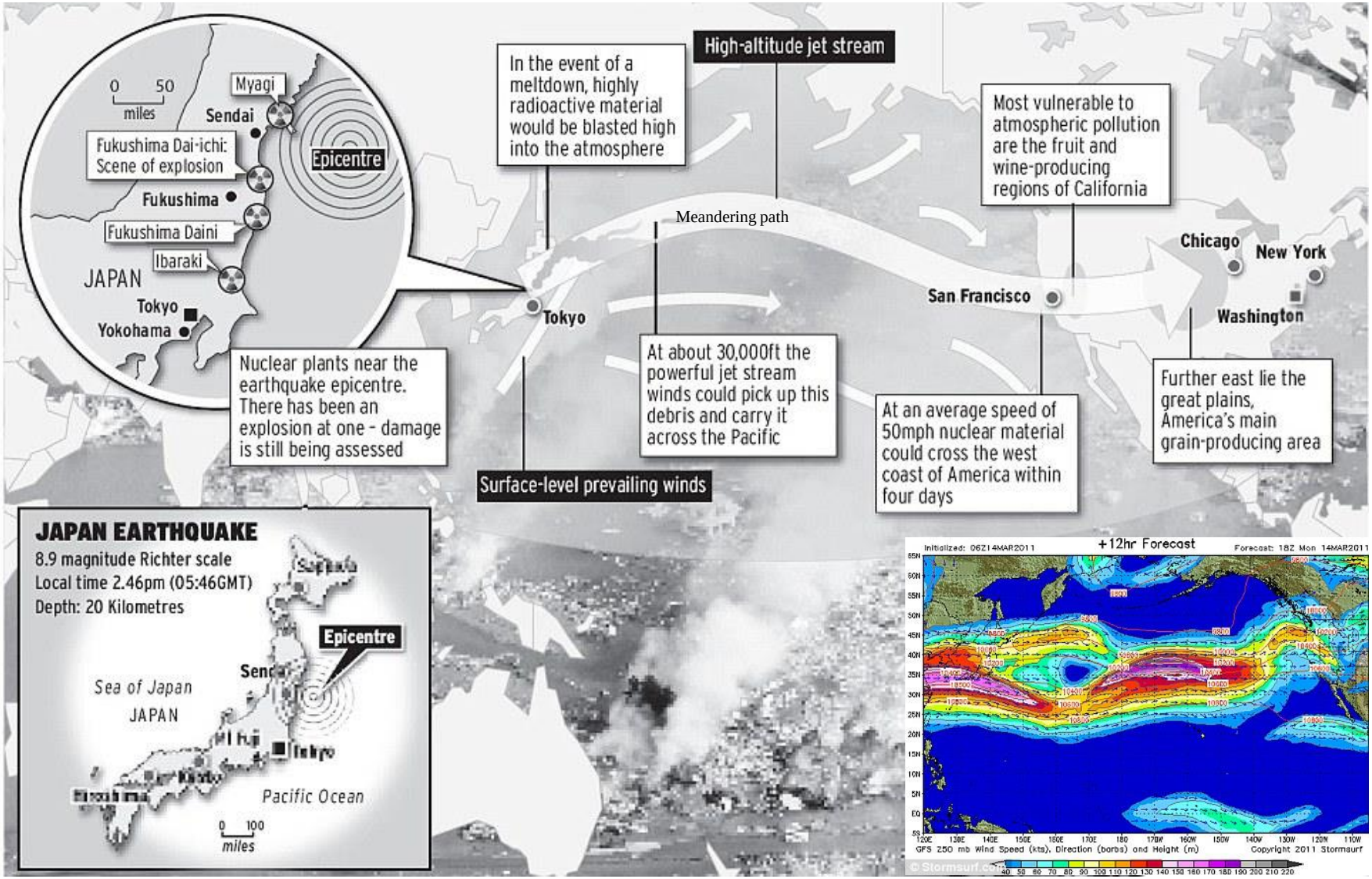
Black Smokes and Dusts Released High in the Sky

(1) (2) (3)

Second explosion rocked the crippled Fukushima Daiichi nuclear plant on 03/13/2011 (1), (2) smoke start to pour from the building housing of **Unit 3** before (3) as the building collapsed, the black plume stretched up into the sky.

From http://www.dailymail.co.uk/news/article-1366341/America-nuclear-alert-As-Japan-begs-U-S-help-fears-fallout-reach-West-Coast.html#ixzz1Gds7HDTv

The Jet Stream Takes 4 Days to Cross the Pacific Ocean



From D. Derbyshire and R. Shears, Mail Online March 15, 2011, http://www.dailymail.co.uk/news/article-1366341/Japan-tsumani-earthquake-America-nuclear-accident-radiation-alert.html#

Sampling Site

- Rooftop of Sojourner Truth Hall at SC State. The rooftop is routinely locked.
- The site is 47 m high. The highest place in an area of 160-km diameter in the SC Coastal Plan. It is 110 km from the Atlantic Ocean.
- It is a good site for monitoring the air pollutions.

Time (GMT-5, DST)	Events	Location
03/10	Earthquake & Tsunami	Fukushima, Japan
03/11-13	Fukushima Unit 1, 2, and 3 Exploded	
↓	The jet streams can carry the radioactive materials to the west coast of the USA in 4 days	Pacific Ocean
03/15-17	Glass-fiber sample GF#1, 49.0 hrs	SC State, USA
03/17-18	Glass-fiber sample GF#2, 27.9 hrs	
03/18-21	Glass-fiber sample GF#3, 67.2 hrs	
03/21-23	Glass-fiber sample GF#4, 47,7 hrs	
03/23-25	GF#5, 48.5 hrs	
03/25-29	GF#6, Carbon cartridge CC#1, 94.6 hrs	
Until May	Sampling continues …	

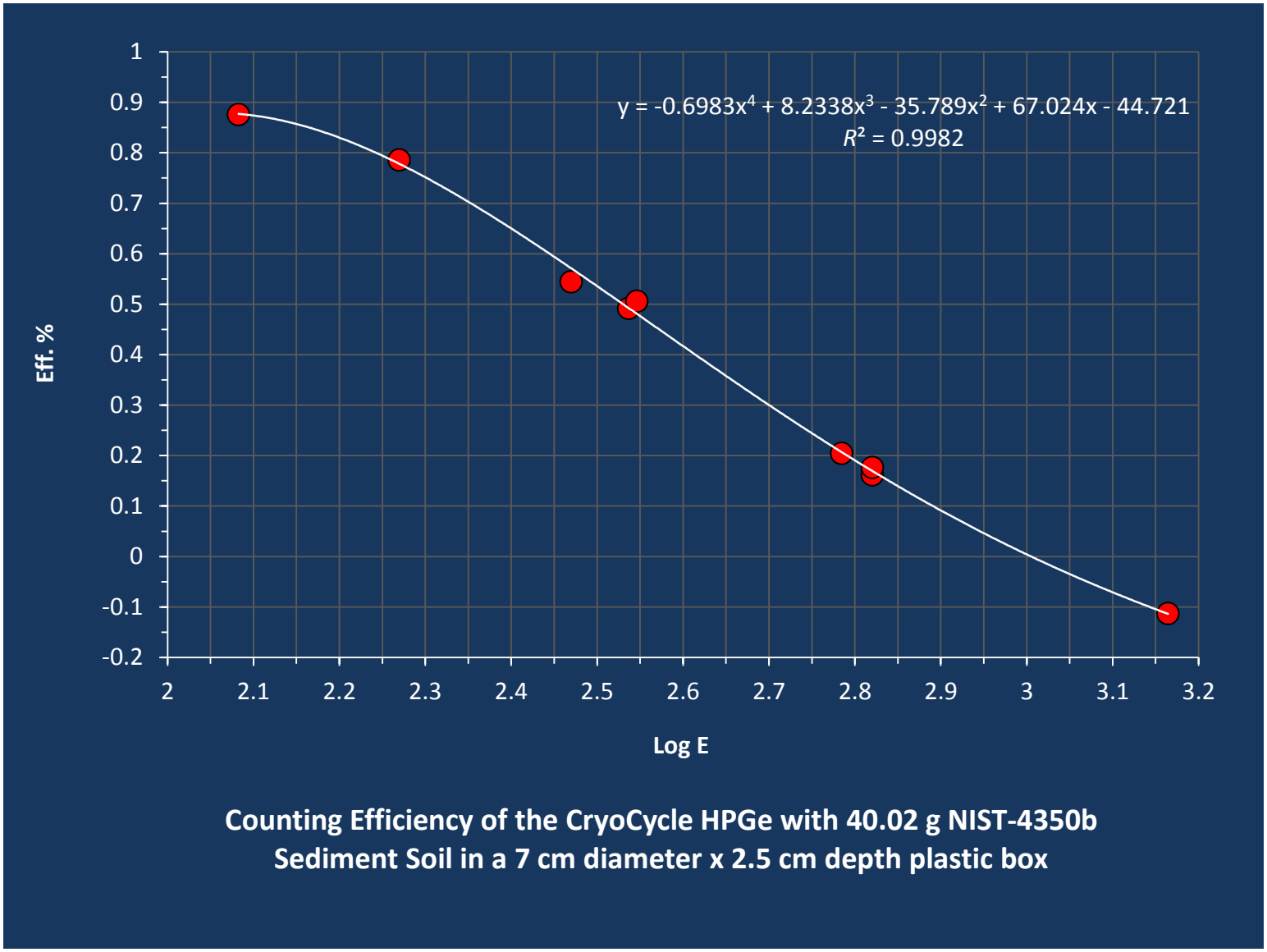
Air Sampler and Filters

- Air sampler:
  Hi-Q PSU-X-GN-R&WS. Capacity: 127 L/min.
  Gooseneck top: 55-78” high.
- Collection location:
  Rooftop of Truth Hall at SC State
  47 m high
- Collection time:
  ~48 hrs for each sample
- Flow rate:
  80 L/min
- Glass fiber filter (started from 03/15/11):
  Hi-Q FP20063-47, 2" diameter
  Porous size ~0.3 micron
- Carbon fiber filter (started from 03/25/11):
  TC-30, 2¼" diameter×1" thick, 20×30 Mesh, activated coconut-shell carbon powders impregnated TEDA (tri-ethylene di-amine). Especially designed for retention of gaseous iodine (92.5% retention of CH_3I)

HPGe Detectors for γ-Spectrometry

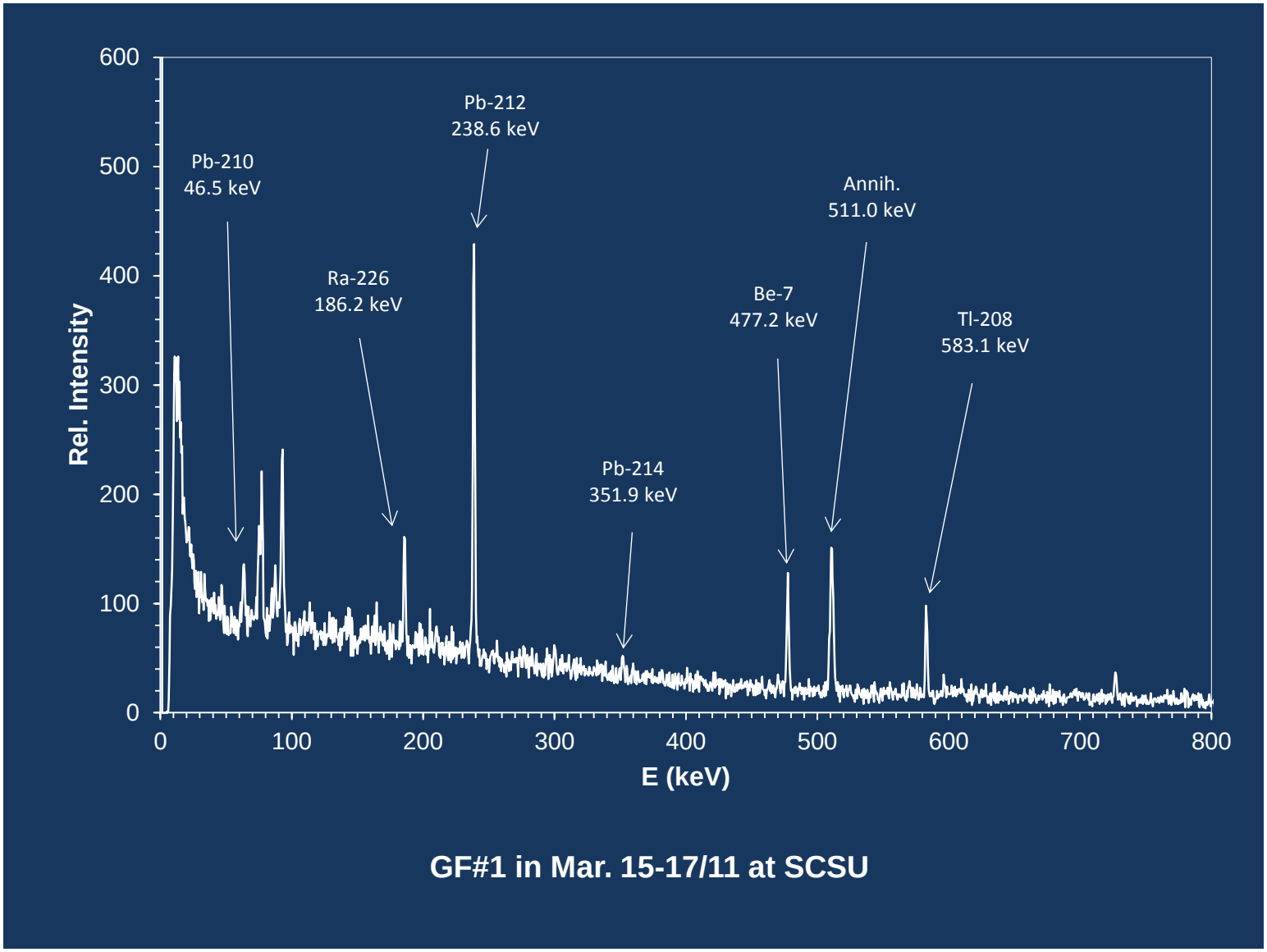
Type	Location	Fission nuclides to be analyzed	Calibration	Specifications
Coaxial Type	SCSU, Orangeburg	• Particulate ^{131}I (in glass fiber). • Gaseous ^{131}I (in carbon cartridge).	• Calibrated by the ^{152}Eu soil standard (NIST-4350b) with the same geometries as those of the glass fiber and carbon cartridge samples. • Calibrated efficiency has ~8% (2σ) uncertainty.	• Canberra GC1020. • Relative eff. ≥ 25%. • Energy resolution ≤ 1.8 keV@1.33 MeV). • Cooled by a cryocycle (Cryo-Cycle™ CC-VD).
Well Type	USC, Columbia	• Particulate ^{134}Cs (in glass fiber). • ^{137}Cs (in glass fiber).	• Calibrated by many standards over years. • Calibrated efficiency has < 5% (2σ) uncertainty.	• Princeton Gamma Tech, IGW-11022. • Relative eff. ~ 21%. • Energy resolution ~ 2.1 keV@1.33 MeV. • Cooled by a LN cryostat.

Efficiency Calibration for the Carbon Cartridge Samples

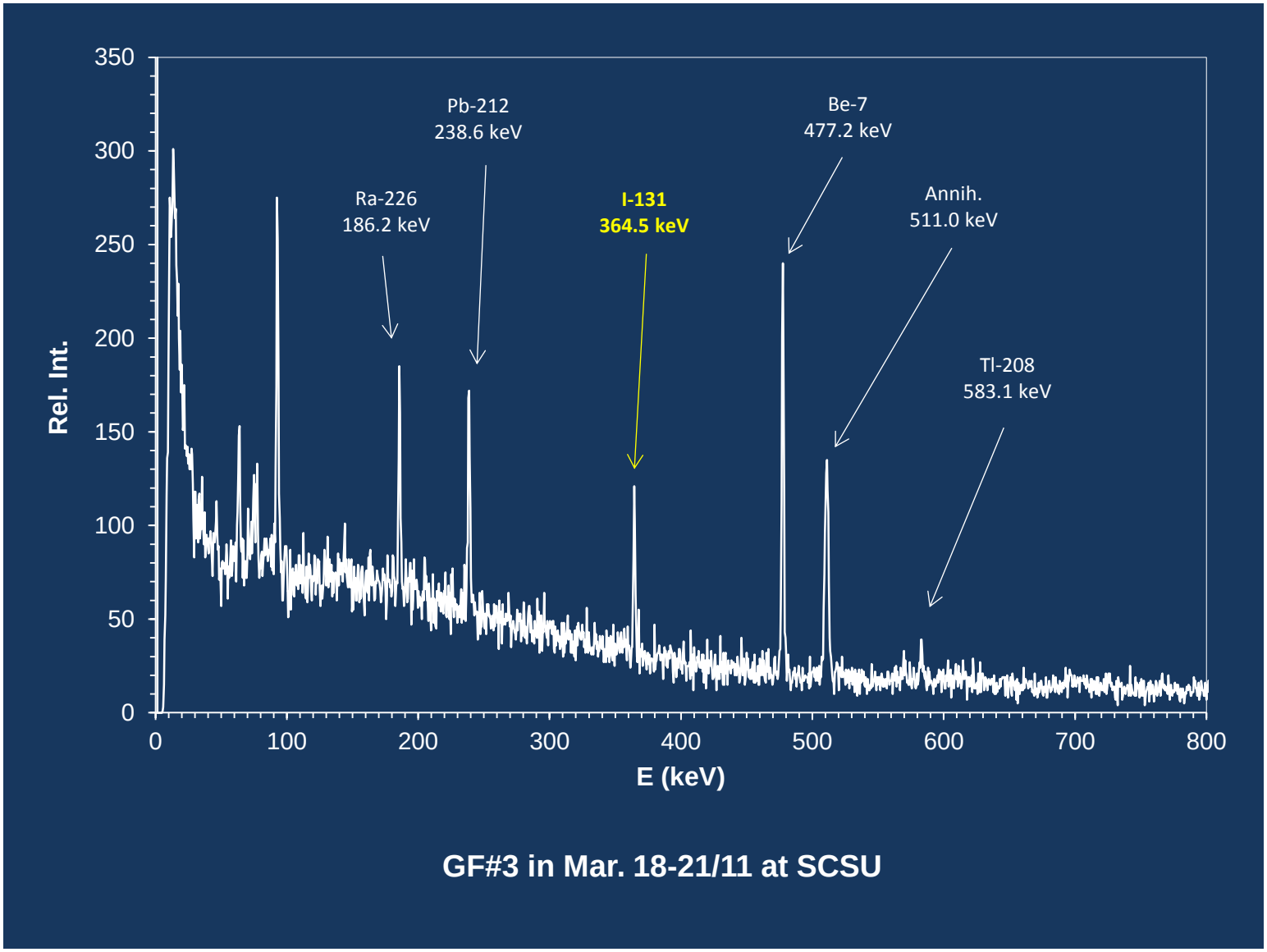


Counting Efficiency of the CryoCycle HPGe with 40.02 g NIST-4350b Sediment Soil in a 7 cm diameter x 2.5 cm depth plastic box

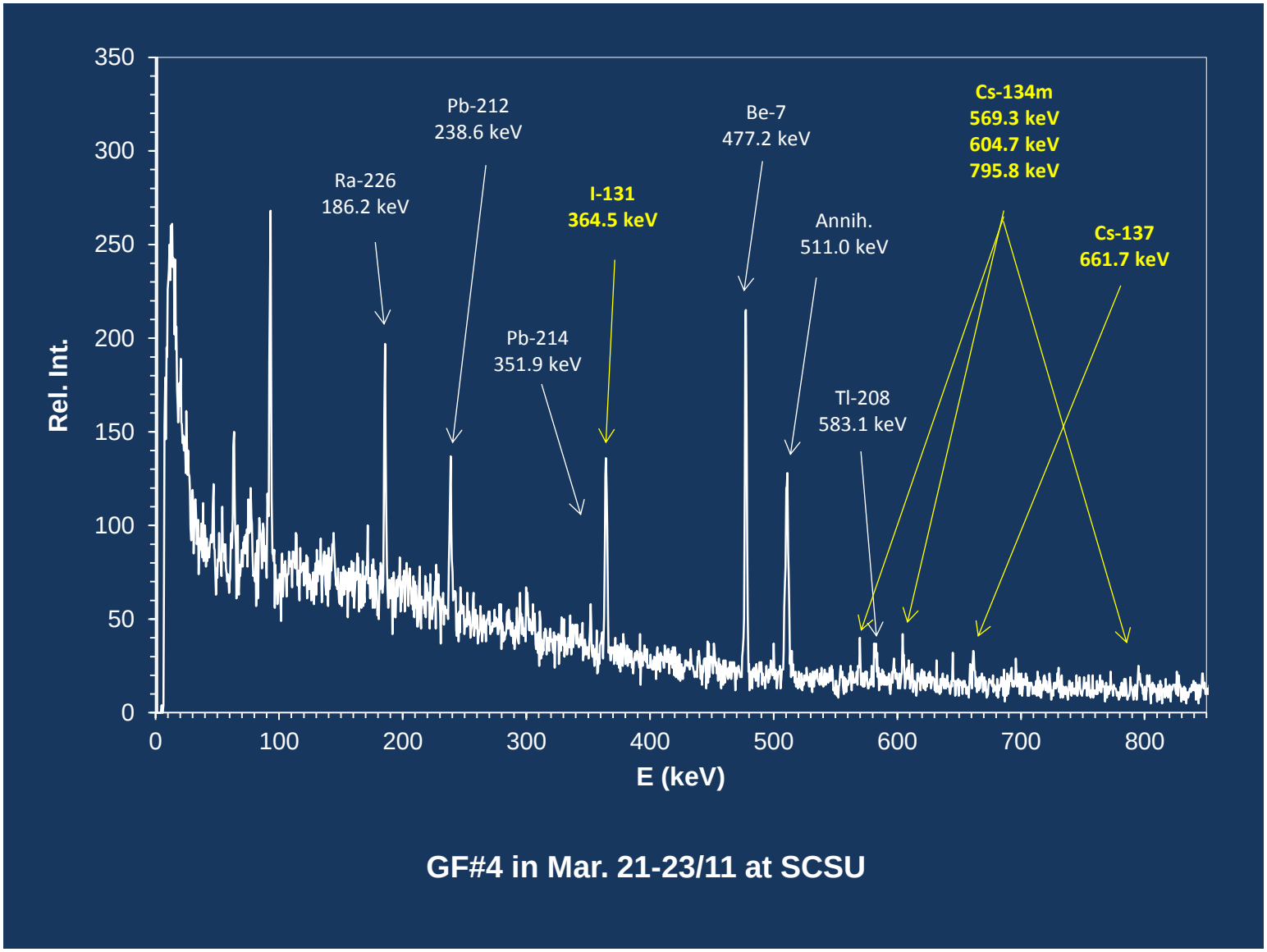
γ-Spectrum of Glass-fiber Sample GF#1 by the Coaxial Type



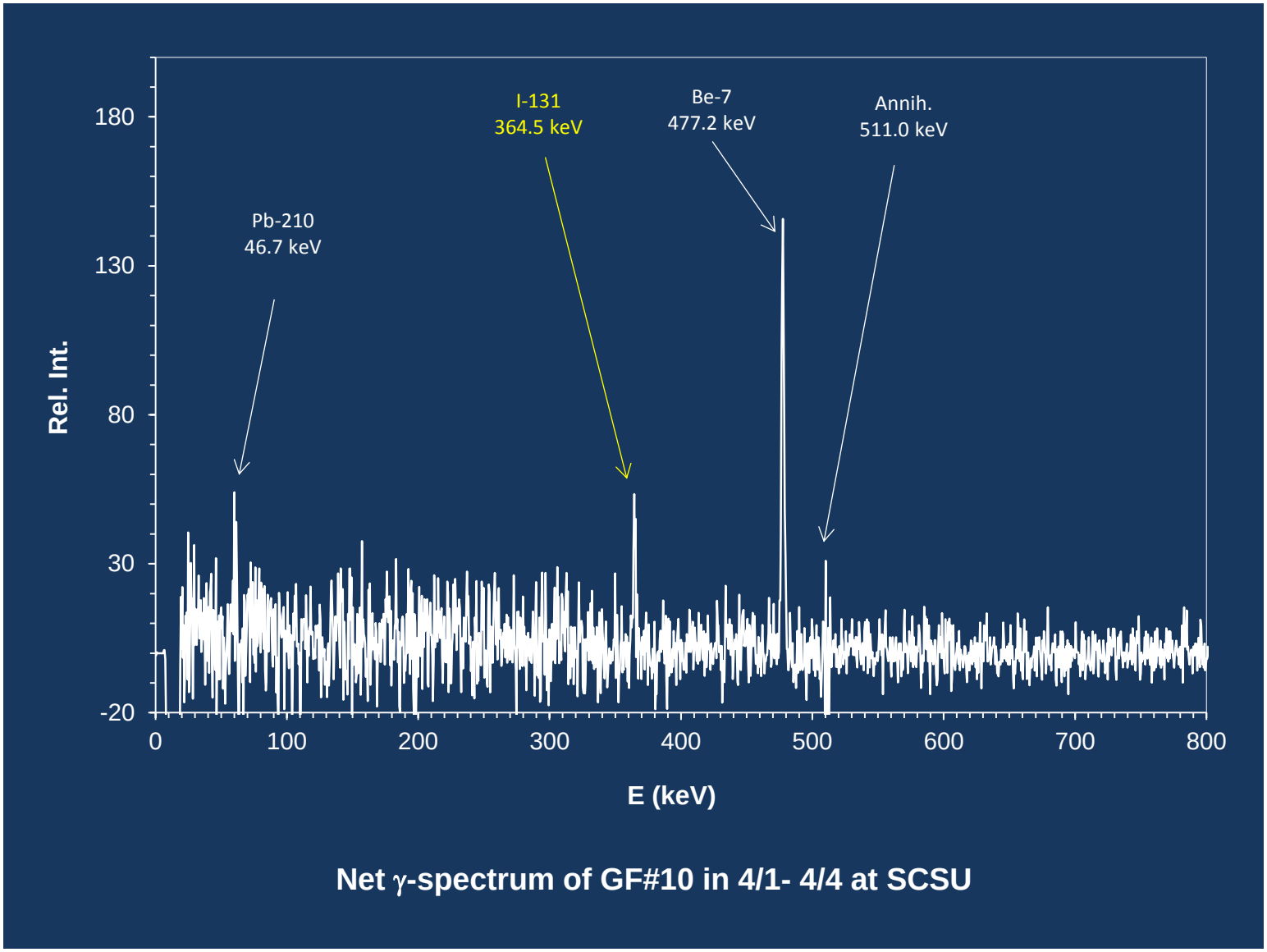
γ-Spectrum of Glass-fiber Sample GF#3 by the Coaxial Type



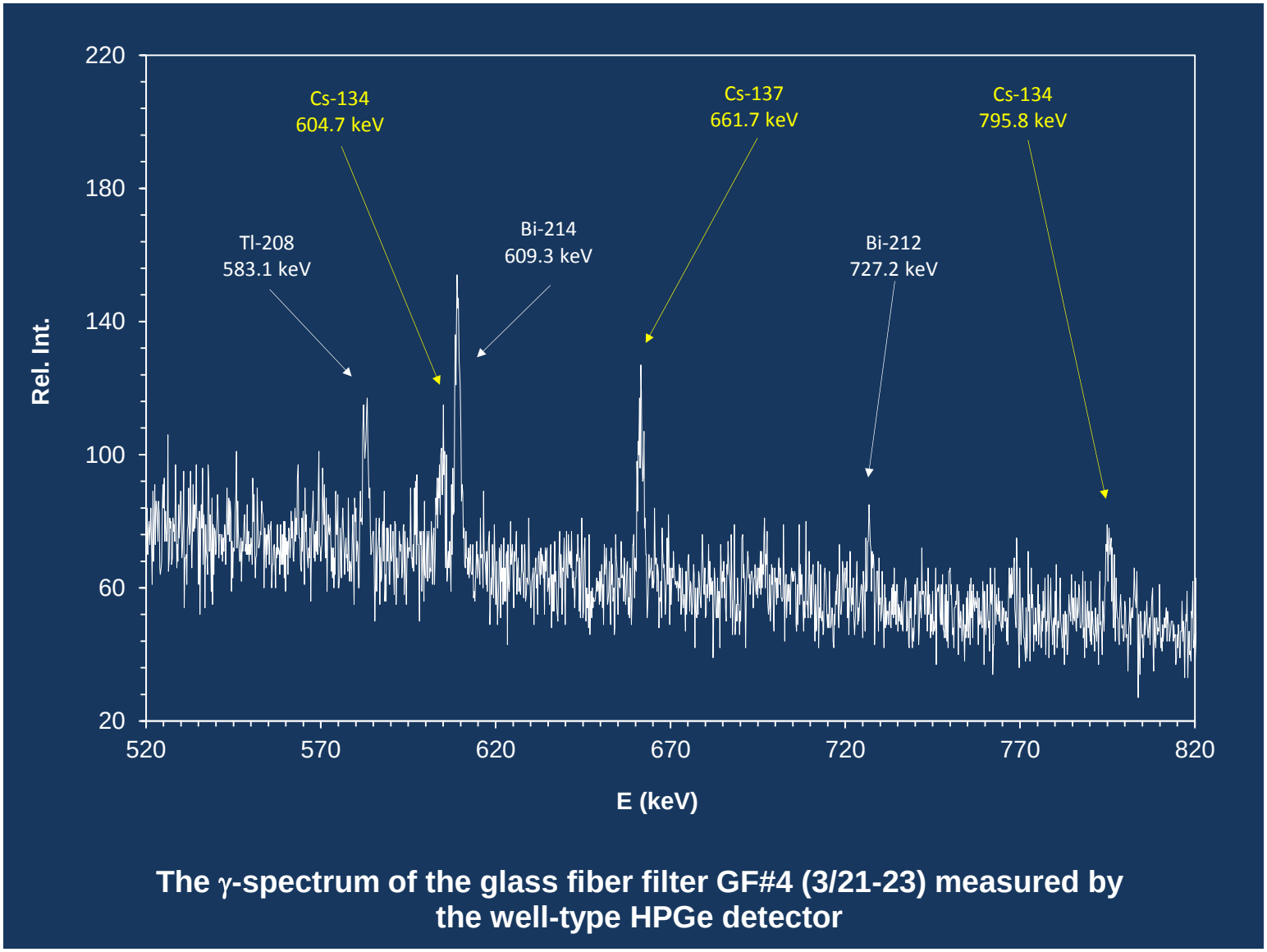
γ-Spectrum of Glass-fiber Sample GF#4 by the Coaxial Type



Net γ-Spectrum of Glass-Fiber Sample GF#10 by the Coaxial Type

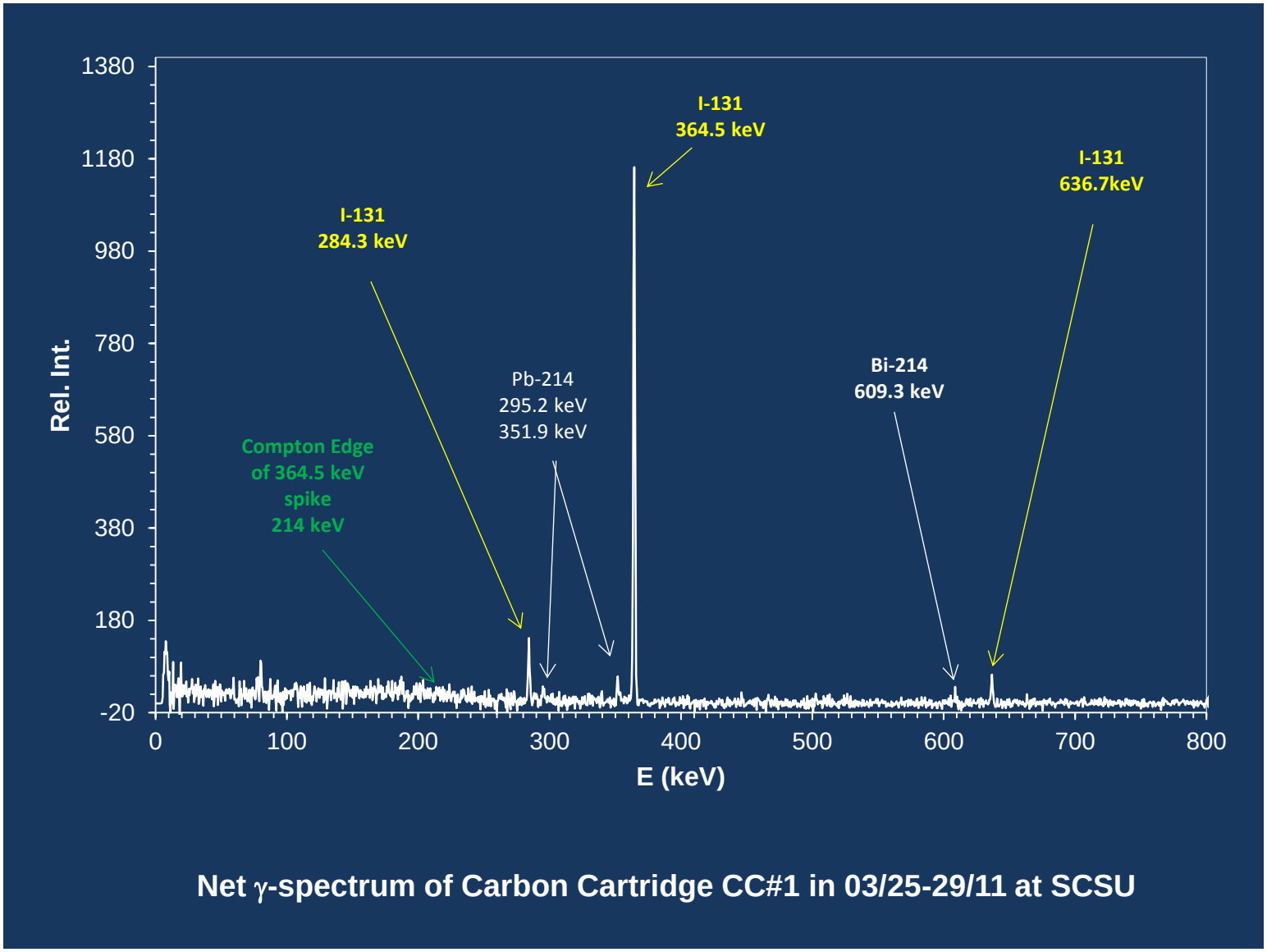


γ-Spectrum of Glass Fiber Sample GF#4 by the Well Type



The γ-spectrum of the glass fiber filter GF#4 (3/21-23) measured by the well-type HPGe detector

γ-Spectrum of the Carbon Cartridge Sample CC#1



Evaluation of the γ-lines of Interest

ROI: Coaxial Type: 4096 for 2.0 MeV. Peak position error: ±1 ch. Peak area: 10 ch. Background: 5 ch on each side.

Well Type: 8,196 ch for1.8 MeV. Peak position error: ±2 ch. Peak area: 20 ch. Background: 10 ch on each side.

FOM: Figure-Of-Merit (ε^2/B). To evaluate the sensitivity of the detector to a specific γ-line.

MDA: Minimum Detectable Activity at 95% confidence level.

$$MDA = \frac{2.71 + 3.29\sqrt{B(1+\frac{n}{2m})}}{I_\gamma \varepsilon R_f S t_c}$$

n: Channel number of the peak.

m: Channel number of the background on each side of the peak.

I_γ: γ-intensity of the γ-ray of the radionuclide.

ε: Detection efficiency of the radionuclide.

R_f: Retention efficiency of the filter of the radionuclide.

λ: Decay constant of the radionuclide.

S : Sample size. For *MDA_A*$_c$, $S = V$. For *MDA_A*$_s$, $S = W$.

t_c : Counting time.

* *Gilmore, G. (2008). Practical gamma-ray spectrometry, Chichester, West Sussex, UK: John Wiley & Sons. Chapter 5.*

Ch	E	Control	Sample	Net
719	359.91	30	25	-5
720	360.41	33	36	3
721	360.91	39	42	3
722	361.41	38	36	-2
723	361.91	39	39	0
724	362.41	26	60	34
725	362.91	31	136	105
726	363.41	38	313	275
727	363.92	28	502	474
728	364.42	30	671	641
729	364.92	36	590	554
730	365.42	23	321	298
731	365.92	29	139	110
732	366.42	27	70	43
733	366.92	29	37	8
734	367.42	32	36	4
735	367.92	36	32	-4
736	368.42	24	27	3
737	368.92	30	30	0
738	369.42	27	24	-3

Evaluation of the γ-peaks for Quantitative Analysis

HPGe Detector [b]	Radionuclide (Filter material)[c]	Half Life	Retention Efficiency R_f (%)	Peak position E (keV)	Region of interest *ROI* (ch#1-ch#2)	γ-Intensity I_γ (%)	Counting Efficiency ε (%)	Baseline count rate B (mcps)	*FOM* (cps^{-1})[d]	*MDA_A*$_c$ ($mBq\ m^{-3}$)[e]	*MDA_A*$_s$ ($Bq\ g^{-1}$)[f]
Coaxial-type HPGe	^{131}I (C Cartridge)	8.04 d	92.5	364.5	719-738	81.2	3.8	0.37	3.9	0.052	--
	^{131}I (Glass fiber)	8.04 d	100	364.5	719-738	81.2	5.2	0.39	6.8	0.036	0.62
	^{7}Be (Glass fiber)	53.3 d	100	477.6	945-964	10.3	4.6	0.24	8.9	0.253	4.4
	^{137}Cs (Glass fiber)	30.2 y	100	661.7	1332-1331	85.1	2.9	0.17	4.8	0.042	0.74
	^{134}Cs (Glass fiber)	2.6 y	100	795.8	1580-1599	85.4	2.2	0.13	3.7	0.048	0.84
Well-type HPGe	^{131}I (Glass fiber)	8.04 d	100	364.5	1910-1957	81.2	15	0.52	48	0.0075	0.13
	^{7}Be (Glass fiber)	53.3 d	100	477.6	2594-2641	10.3	12	0.35	40	0.066	1.1
	^{137}Cs (Glass fiber)	30.2 y	100	661.7	3617-3664	85.1	9.5	0.25	36	0.0085	0.15
	^{134}Cs (Glass fiber)	2.6 y	100	795.8	4363-4410	85.4	8.0	0.20	33	0.0090	0.16

[a] Samples CC#2 and GF#4 were used for the calculation of the parameters of the carbon cartridge and glass-fiber filter samples, respectively

[b] The multichannel analyzer (MCA) of the coaxial-type detector has 4096 channels over 2 MeV. The MCA of the well-type detector has 8192 channels over 1.8 MeV.

[c] The radionuclides (filter material) in italic print were not selected for the calculation of radioactivity.

[d] *FOM*: Figure of Merit. FOM = ε^2/B.

[e] *MDA_A*$_c$: the minimum detectable radioactivity concentration.

[F] *MDA_A*$_s$: the minimum detectable specific radioactivity.

Calculation of the Radioactivity

$$A = \frac{N_s - N_c}{I_\gamma \varepsilon R_f S t_c (\frac{1 - e^{-\lambda t_c}}{\lambda t_c}) e^{-\lambda t_s}}$$

N_s: Net counts of the photopeak

N_c: Net counts of the pristine sample (glass-fiber filter/carborn cartridge)

I_γ: γ-intensity of the γ-ray of the radionuclide

ε: Detection efficiency of the radionuclide

R_f: Retention Efficiency of the filter of the radionuclide

λ: Decay constant of the radionuclide

t_s : Time elapse from the mid-point of sample collection to the beginning of gamma counting

t_c : Counting time

S : Sample size (volume of air or weight of the particulate mass)

Cascade Summing

When ^{134}Cs decays into ^{134}Ba, 604.7 keV and 795.8 keV γ-rays are emitted in a cascade way within 5.2 picoseconds of one another (Shanks, et al 2012). The chance that these two γ-rays are coincidently counted as one event at 1400.5 keV increases with the geometric counting efficiency. The effect of cascade summing on the radioactivity can be corrected by:

$$A' = \frac{A}{1 - T_2 \varepsilon_2 K_2}$$

A: ^{134}Cs radioactivity before the correction of cascade summing.

T_2: Ratio of the γ-intensities (I_γ) of the 604.7 and 795.8 keV γ-rays, $T_2 = 0.974/0.865 = 1.14$

ε_2: Counting efficiency of the 604.7 keV γ-line. $\varepsilon_2 = 9.5\%$.

K_2: Ratio of the total and neat peak areas of the 604.7 keV γ-line. $K_2 \approx 1.04$

A': ^{134}Cs radioactivity after the correction of cascade summing. $A' \approx 1.11A$

Uncertainty of the Activity

1. If $N \geq 1.645\sqrt{B\left(1+\frac{n}{2m}\right)}$,

   $N\ (= Ns - N_c)$ is significant.

   Its 2σ Uncertainty $= \pm 1.96\sqrt{N}$.

2. If $0 < N < 1.645\sqrt{B\left(1+\frac{n}{2m}\right)}$,

   N is not significant.

   The upper limit for 95% confidence levele $= 1.645\sqrt{B\left(1+\frac{n}{2m}\right)}$.

* *Gilmore, G. (2008). Practical gamma-ray spectrometry, Chichester, West Sussex, UK: John Wiley & Sons. Chapter 5.*

Radioactivities by the Coaxial-type HPGe

Sample #	*Sampling Time* (GMT-5.00, DST) Midpoint of the Sampling	V (M^3) Air Volume	W (mg) particulate weight	ρ (mg m^{-3}) particulate density	$A_{c131,\,g}$ (mBq m^{-3})[a]	$A_{c131,\,p}$ (mBq m^{-3})[a]	$A_{s131,\,p}$ (Bq g^{-1})[b]	$A_{c7,\,p}$ (mBq m^{-3})[c]	$A_{s7,\,p}$ (Bq g^{-1})[c]	$\frac{A_{c131,g}}{A_{c131,p}}$
GF#0	25Sep2010 14:36	208(4)	6.2(4)	0.030(2)	-	*0.00(2)*	*0.0(6)*	5.5(7)	139(22)	-
GF#1	16Mar2011 12:20	218(4)	5.3(4)	0.024(2)	-	*0.00(2)*	*0.1(10)*	4.7(6)	192(33)	-
GF#2	18Mar2011 1:52	132(3)	14.2(4)	0.108(4)	-	*0.02(6)*	*0.2(6)*	5.1(8)	47.6(82)	-
GF#3	20Mar2011 1:35	329(6)	28.4(4)	0.086(2)	-	0.35(5)	4.1(6)	6.7(7)	77.8(104)	-
GF#4	22Mar2011 11:15	230(5)	13.2(4)	0.057(2)	-	0.92(11)	16(2)	7.4(9)	129(21)	-
GF#5	24Mar2011 12:01	237(5)	7.3(4)	0.031(2)	-	0.90(11)	29(4)	5.1(6)	165(26)	-
GF#6, CC#1	27Mar2011 11:00	457(9)	9.9(4)	0.22(1)	5.0(4)	1.0(1)	47(5)	2.7(3)	126(19)	4.8(6)
GF#7, CC#2	30Mar2011 10:45	226(5)	3.3(4)	0.015(2)	3.8(3)	0.64(10)	44(10)	4.7(6)	319(63)	6.0(11)
GF#8, CC#3	1Apr2011 1:06	139(3)	2.5(4)	0.018(3)	3.3(3)	0.80(14)	43(10)	3.3(6)	181(46)	4.1(8)
GF#9, CC#4	3Apr2011 2:02	324(6)	5.3(4)	0.016(1)	4.4(3)	0.10(3)	5.9(18)	6.2(7)	376(59)	46(14)
GF#10, CC#5	5Apr2011 13:35	212(4)	3.7(4)	0.017(2)	2.5(2)	0.12(4)	6.7(24)	7.0(8)	400(72)	21(7)
GF#11, CC#6	7Apr2011 12:03	228(5)	4.3(4)	0.019(2)	2.5(2)	*0.01(7)*	*0.5(39)*	7.9(9)	417(71)	*232(1600)*
GF#12, CC#7	10Apr2011 0:53	343(7)	10.5(4)	0.031(1)	0.44(6)	*0.02(6)*	*0.8(20)*	4.9(6)	161(24)	*18(45)*
GF#13, CC#8	12Apr2011 13:03	225(5)	14.0(4)	0.062(2)	0.18(5)	0.04(2)	0.64(40)	7.7(9)	124(18)	4.5(31)
GF#14, CC#9	14Apr2011 12:35	231(5)	6.0 (4)	0.026(2)	0.44(7)	*0.03(7)*	*1(3)*	8.3(9)	320(49)	*18(52)*
GF#15, CC#10	17Apr2011 0:28	341(7)	11.1(4)	0.033(1)	0.15(3)	0.03(2)	0.96(51)	5.8(6)	177(25)	4.6(26)
GF#16, CC#11	19Apr2011 12:15	226(5)	6.4(4)	0.028(2)	0.20(4)	*0.01(5)*	*0.5(18)*	7.8(9)	277(42)	*14(50)*
GF#17, CC#12	21Apr2011 11:57	228(5)	8.5(4)	0.037(2)	*0.01(9)*	*0.02(4)*	*0.4(12)*	7.0(8)	187(28)	*0.7(55)*
GF#18, CC#13	23Apr2011 23:37	343(7)	4.7(4)	0.014(1)	*0.01(5)*	*0.005(20)*	*0.3(16)*	5.7(6)	419(67)	*2(16)*
GF#19, CC#14	26Apr2011 11:27	226(5)	4.6(4)	0.020(2)	*0.04(10)*	*0.01(5)*	*0.6(23)*	3.9(5)	193(35)	*3(15)*
GF#20, CC#15	29Apr2011 11:53	231(5)	6.6(4)	0.029(2)	*0.005(70)*	*0.01(5)*	*0.5(17)*	6.4(8)	224(35)	*0.3(50)*
GF#21, CC#16	1May2011 1:17	354(7)	9.2(4)	0.026(1)	*0.02(5)*	*0.01(3)*	*0.2(11)*	3.8(5)	145(22)	*3(20)*
GF#22, CC#17	3May2011 11:39	211(4)	3.6(4)	0.017(2)	*0.01(11)*	*0.01(4)*	*0.5(45)*	3.8(5)	448(81)	*1(16)*
GF#30, CC#25	29May2011 10:35	237(4)	3.4(4)	0.011(2)	*0.00(11)*	*0.00(4)*	*0.0(17)*	7.6(9)	264(39)	*1(30)*

[a] $A_{c131,\,g}$ *and* $A_{c131,\,p}$: Radioactivity concentrations of ^{131}I in gaseous and particulate phases, respectively.

[b] $A_{s131,\,p}$: Specific radioactivity of ^{131}I in particulate phase.

[c] $A_{c7,\,p}$ *and* $A_{s7,\,p}$: Radioactivity concentration and specific radioactivity of particulate ^{7}Be, respectively.

Radioactivities by the Well-type HPGe

Sample #	*Sampling Time* (GMT-5.00, DST) Midpoint of Sampling	$V(M^3)$ Air Vol.	$A_{c131,\,p}$ (mBq m^{-3})[a]	$A_{s131,\,p}$ (Bq g^{-1})[a]	$A_{c134,\,p}$ (mBq m^{-3})[b]	$A_{s134,\,p}$ (Bq g^{-1})[b]	$A_{c137,\,p}$ (mBq m^{-3})[c]	$A_{s137,\,p}$ (Bq g^{-1})[c]	$A_{c7,\,p}$ (mBq m^{-3})[d]	$A_{s7,\,p}$ (Bq g^{-1})[d]	$\frac{A_{c131,\,p}}{A_{c131,\,g}}$
GF#1	16Mar2011 12:20	218(4)	*0.01(7)*	*0.1(7)*	*0.01(18)*	*0.01(27)*	*0.002(6)*	*0.1(3)*	5.5(6)	*225(28)*	*4(86)*
GF#4	22Mar2011 11:15	230(5)	0.85(10)	15(2)	0.083(11)	1.5(2)	0.11(1)	1.9(2)	6.2(5)	108(15)	0.75(13)
GF#7	30Mar2011 10:45	226(5)	0.67(8)	46(8)	0.11(1)	7.4(13)	0.12(2)	9.1(15)	4.5(2)	307(40)	0.92(14)
GF#8	1Apr2011 1:06	139(3)	0.85(11)	47(10)	0.09(2)	5.1(12)	0.10(2)	5.7(12)	3.7(2)	204(35)	0.90(20)
GF#12	10Apr2011 0:53	343(7)	*0.00(7)*	*0.1(20)*	0.020(3)	0.65(12)	0.014(3)	0.43(8)	4.3(2)	169(11)	1.4(4)
GF#15	17Apr2011 0:28	341(7)	0.03(1)	0.75(14)	0.014(3)	0.43(9)	0.015(3)	0.42(7)	5.0(3)	186(11)	0.93(3)

[a] $A_{c131,\,p}$ *and* $A_{s131,\,p}$: Radioactivity concentration and specific radioactivity of particulate ^{131}I, respectively.
[b] $A_{c134,\,p}$ *and* $A_{s134,\,p}$: Radioactivity concentration and specific radioactivity of particulate ^{134}Cs, respectively.
[c] $A_{c137,\,p}$ *and* $A_{s137,\,p}$: Radioactivity concentration and specific radioactivity of particulate ^{137}Cs, respectively.
[d] $A_{c7,\,p}$ *and* $A_{s7,\,p}$: Radioactivity concentration and specific radioactivity of particulate ^{7}Be, respectively.

Data Quality

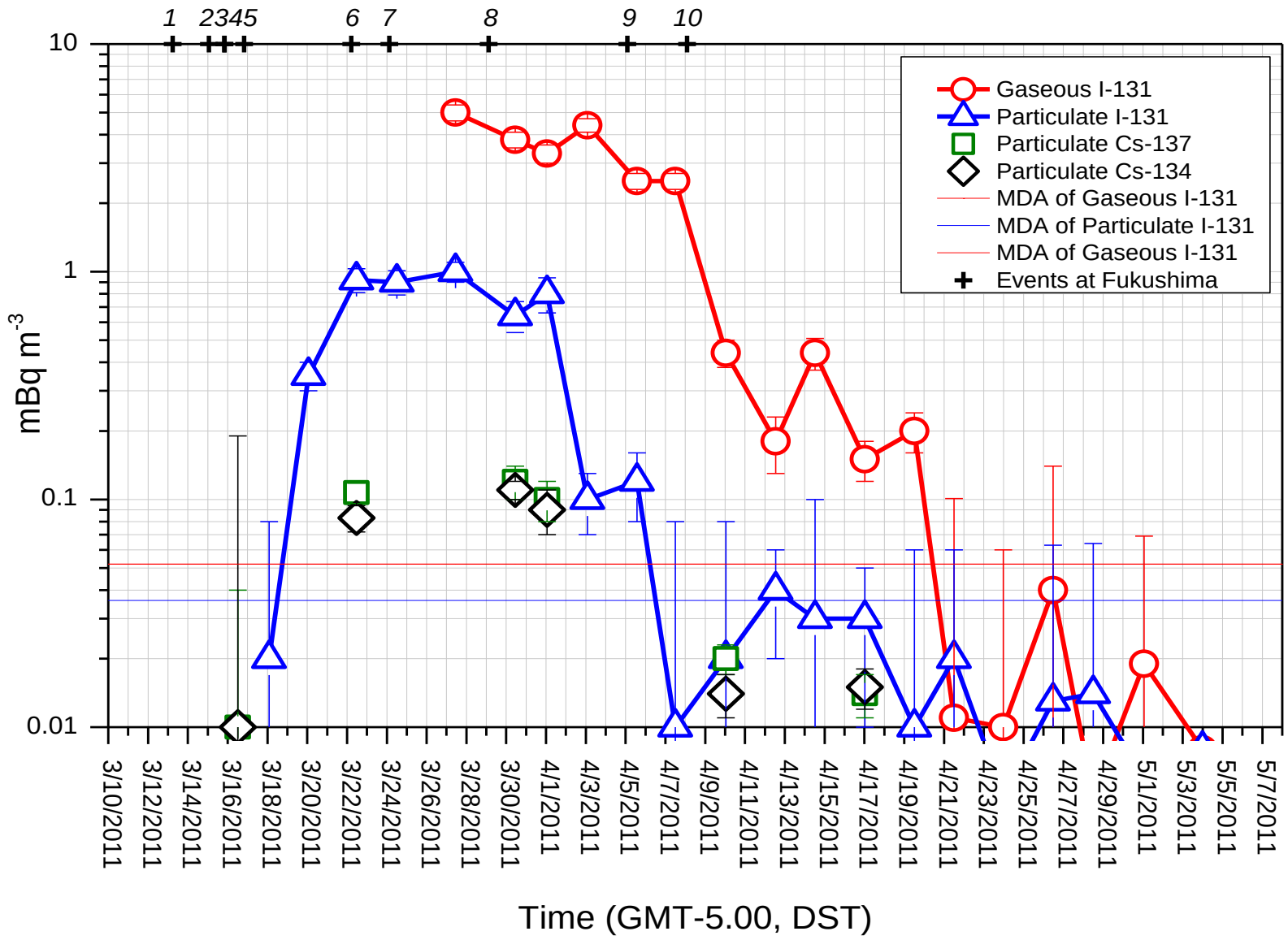
1. It was found that the measured ^{131}I activities by the two detectors were the same within the detection error.

Sample #	GF#4	GF#7	GF#8	GF#12	GF#15
Coaxial-type HPGe at SCSU	0.92 ± 0.11	0.64 ± 0.10	0.80 ± 0.14	*0.02 ± 0.06*	0.03 ± 0.02
Well-type HPGe at USC	0.85 ± 0.10	0.67 ± 0.08	0.85 ± 0.11	*0.00 ± 0.07*	0.03 ± 0.01

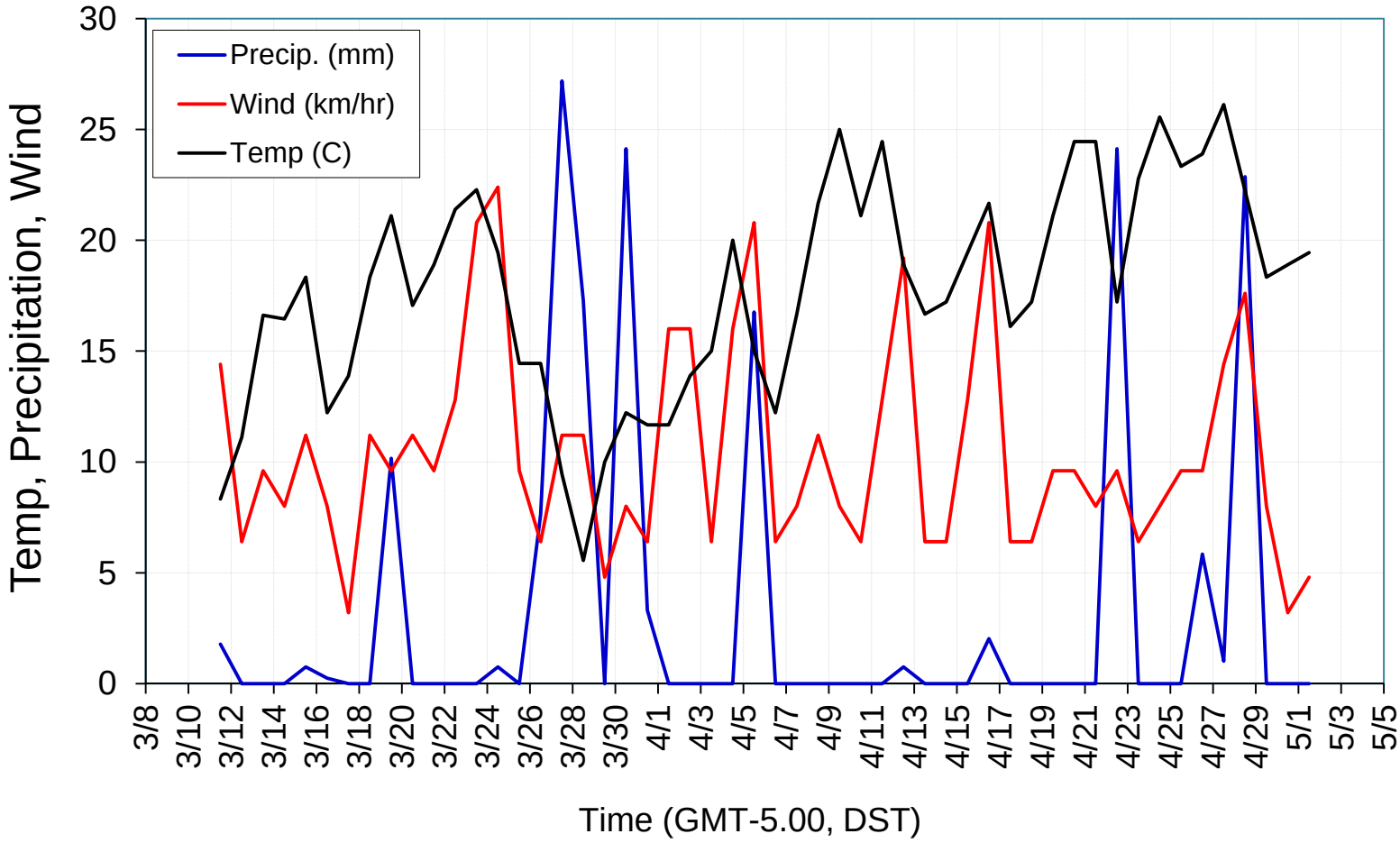
2. For ^{7}Be, the measured data agrees with the data in literature

Origin	Location	Experimental period	$A_{c7,p}$ range
Azahra et al (2004)	Granada, Spain	1993 -1997	3 - 8 mBq m^{-3}
Buraeva et al (2007)	Rostovon-Don, Australia	2001 - 2005	2 - 7 mBq m^{-3}
Yoshimori et al (2003)	Japan	2001	2 - 8 mBq m^{-3}
This study	Orangeburg, SC	03/15 - 05/31, 2011	2.7 - 8.3 mBq m^{-3}

Histogram of the Fission Products at Orangeburg, US



Weather Conditions during the Observation



Weather Conditions vs. Time

Correlation Coefficient (R) between the Nuclear Fallout and Weather Conditions

	Temp (K)	Precip. (mm)	Wind Speed (km h^{-1})	Parti. Den. (mg m^{-3})	$A_{c131,g}$ (mBq M^{-3})	$A_{c131,p}$ (mBq m^{-3})	$A_{s131,p}$ (Bq g^{-1})	$A_{c137,p}$ (mBq m^{-3})	$A_{s134,p}$ (Bq g^{-1})
Temp (K)	**1.0**	-0.2	-0.2	0.1	**-0.9**	-0.5	**-0.7**	0.4	-0.1
Precip. (mm)	-0.2	**1.0**	-0.1	-0.3	0.3	0.2	0.4	-0.2	0.1
Wind Speed (km h^{-1})	-0.2	-0.1	**1.0**	-0.2	0.2	0.1	0.1	0.1	0.2
Parti. Den. (mg m^{-3})	0.1	-0.3	-0.2	**1.0**	-0.5	0.0	-0.2	0.2	**-0.7**
$A_{c131,g}$ (mBq m^{-3})	**-0.9**	0.3	0.2	-0.5	**1.0**	**0.8**	**0.8**	-0.4	0.2
$A_{c131,p}$ (mBq m^{-3})	-0.5	0.2	0.1	0.0	**0.8**	**1.0**	**0.9**	-0.3	-0.3
$A_{s131,p}$ (Bq g^{-1})	**-0.7**	0.4	0.1	-0.2	**0.8**	**0.9**	**1.0**	-0.5	-0.1
$A_{c137,p}$ (mBq m^{-3})	0.4	-0.2	0.1	0.2	-0.4	-0.3	-0.5	**1.0**	0.3
$A_{s134,p}$ (Bq g^{-1})	-0.1	0.2	0.2	**-0.7**	0.2	-0.3	-0.1	0.3	**1.0**

*Correlation coefficient of X and Y arrays of values is obtained by:

$$R = Correl(X,Y) = \frac{\sum(x-\bar{x})(y-\bar{y})}{\sqrt{\sum(x-\bar{x})^2 \sum(y-\bar{y})^2}}$$

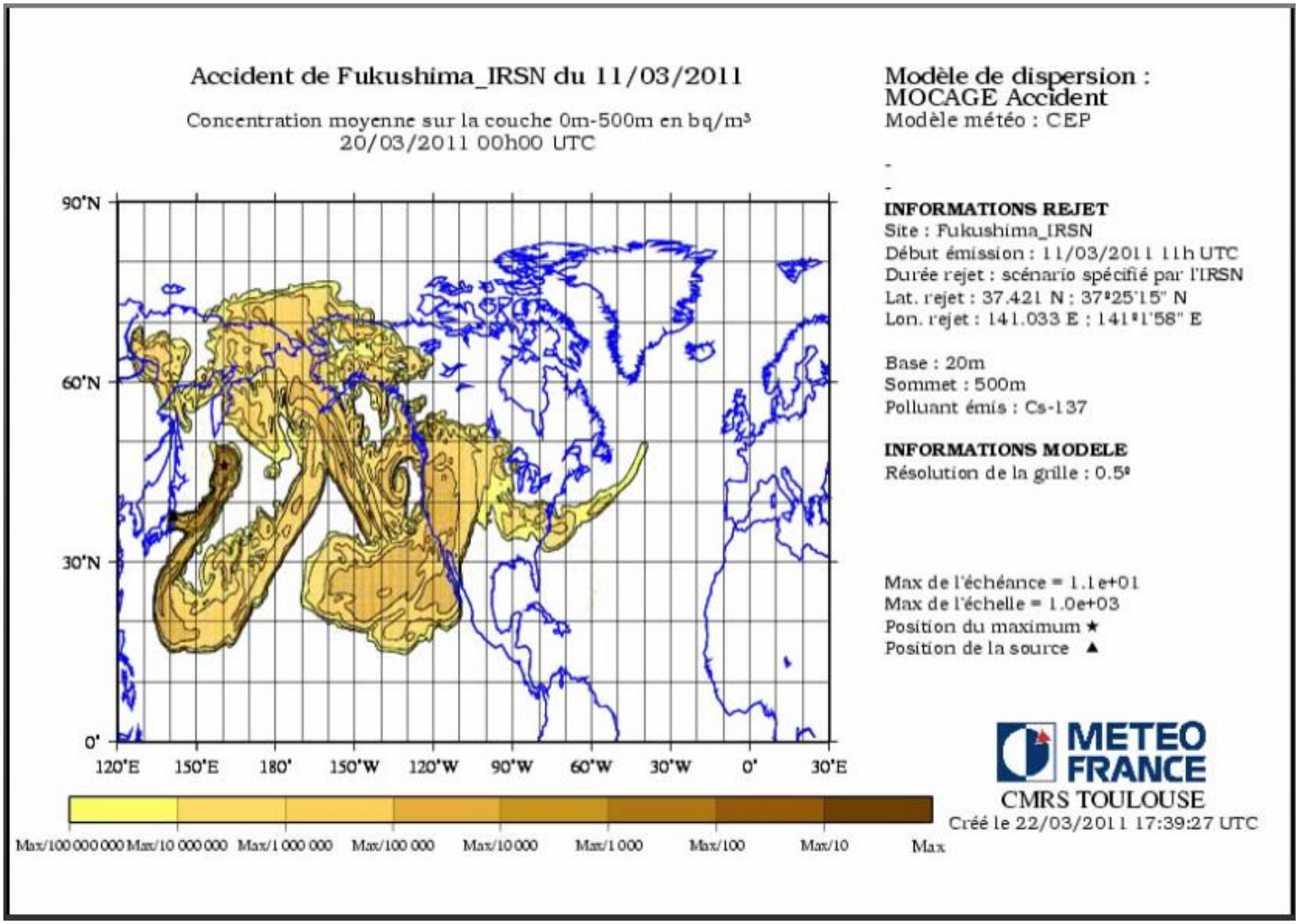
It is assumed that when $|R| \geq 0.7$ (or $R^2 \geq 0.5$), the X and Y arrays are assumed correlated. In the table, all the Rs with the absolute value being equal to or larger than 0.7 are in bold print.

Arrival Times of the Fukushima Gaseous and Particulate Fallout

No.	Events at Fukushima in Tokyo Time (GMT+9.00)	Events in US Eastern DST time (GMT-5.00, DST)	Plateau and spikes on the histogram of $A_{c131,p}$ (GMT-5.00, DST)	Time Lap from the Events (Day)	Plateau and spikes on the histogram of $A_{c131,g}$ (GMT-5.00, DST)	Time Lap from the Events (Day)
1	03/12 15:36 Unit1 explosion	03/13 4:36	22 -26 March	**7-10**	--	--
2	03/14 11:15 Unit3 explosion	03/15 0:15				
3	03/15 6:00 Unit4 explosion	03/15 9:00				
4	03/15 6:14 Unit2 blast sound	03/15 9:14				
5	03/16 5:45 Unit4 on fire	03/16 18:45				
6	03/21 5:00 Unit3 gray/black smokes	03/22 4:00	04/01 1:06	**9**	03/03 2:02	**11**
7	03/23 13:00 Unit3 gray/black smokes	03/24 2:00				
8	03/28 12:42 Cooling pumps stopped working on Reactors 1-4	03/29 1:42	04/05 13:35	**7**	04/07 12:03	**9**
9	04/04 Unit2 release 10k tons radioactive water to sea	04/5 1:00	04/12 13:03	**8**	04/14 13:35	**10**
10	04/7 Radiation released in a 7.1 magnitude aftershock	04/08 1:00	04/17 0:28	**9**	04/19 12:15	**11**

Conclusion: The particulate fission products arrived at Orangeburg in **8** days. The gaseous fallout arrived in **10** days.

IRSN Dispersion Model of the Fukushima Nuclear Fallout



IRSN. Dispersion model of the radioactive releases in the atmosphere on worldwide scale. 2011*b*. Available at http://www.irsn.fr/EN/news/Documents/IRSN_Dispersion-model-radioactive-releasesin-atmosphere-worldwide-EN.pdf. Accessed 21 July 2012.

Isotope Ratio of $^{134}Cs/^{137}Cs$ in Particulate Phase

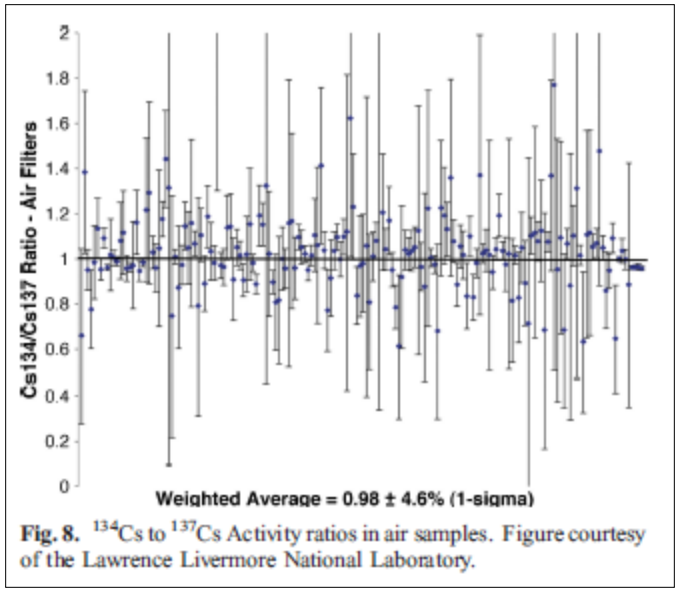


Fig. 8. ^{134}Cs to ^{137}Cs Activity ratios in air samples. Figure courtesy of the Lawrence Livermore National Laboratory.

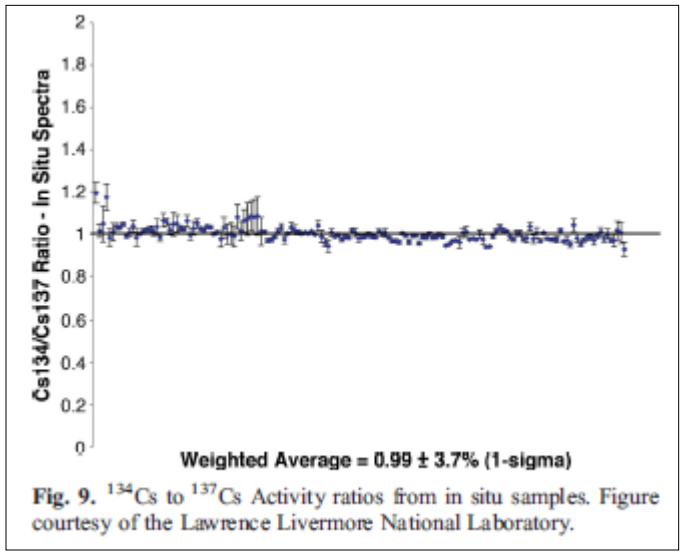


Fig. 9. ^{134}Cs to ^{137}Cs Activity ratios from in situ samples. Figure courtesy of the Lawrence Livermore National Laboratory.

Japan:

0.98 ± 4.6% (178 air samples)

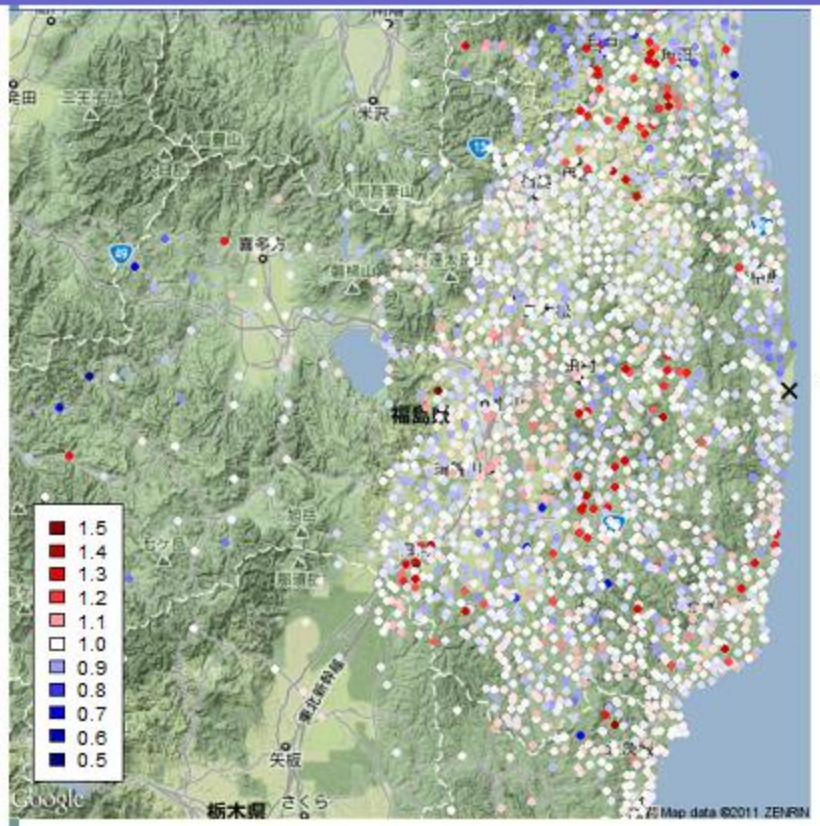
0.99 ± 3.7% (158 in-situ γ-spec samples)

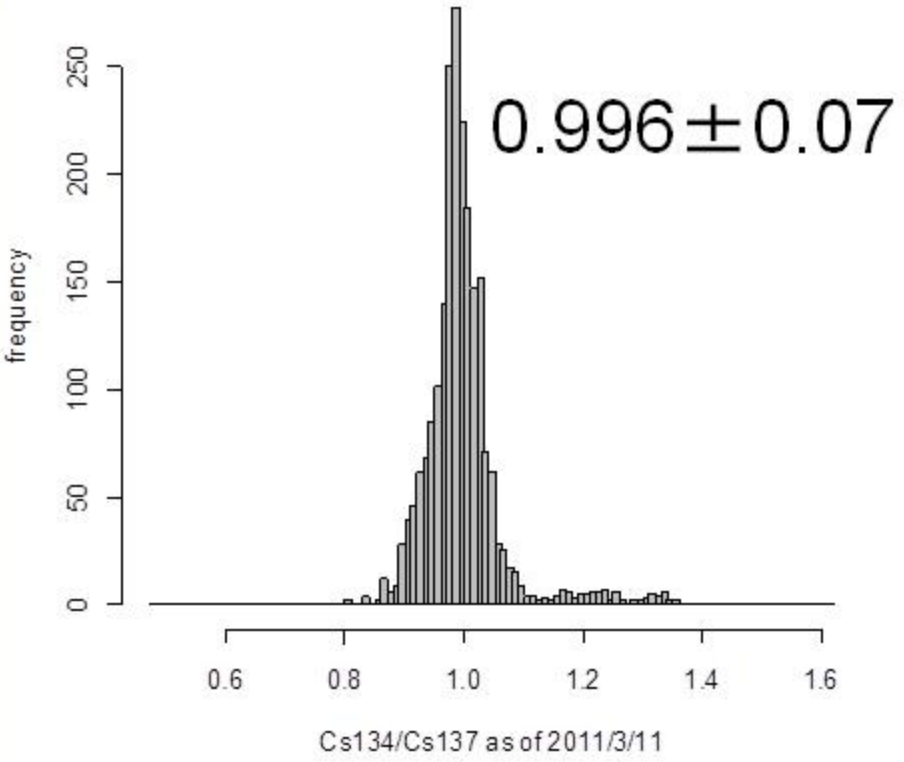
0.996 ± 0.07 (>1000 soil samples from an area of over 100 km)

Orangeburg:

0.98 ± 0.26 (five measurements of glass-fiber 22 March to 17 April based on five measurements)

Frequency distribution of $^{134}Cs/^{137}Cs$





- Estimated burnup : 17.2±1.5 [GWd/tHM]

[9] http://www.mext.go.jp/b_menu/shingi/chousa/gijyutu/017/shiryo/__icsFiles/afieldfile/2011/09/02/1310688_1.pdf

Activity Ratios of Gaseous ^{131}I to Particulate ^{131}I

Isotope Radio at Wako City and Chiba City, Japan

Date	$A_{131,g} / A_{131,p}$
March 20	0.8
March 20-21	1.0
March 22-23	2.1
March 23-25	3.5

Isotope Ratio at Orangeburg, SC

Date	$A_{131,g} / A_{131,p}$
March 27-30	5.5
April 3-5	33

Conclusion: The $A_{131,g}/A_{131,p}$ ratio increased significantly, suggesting two possibilities:

1) Particulate ^{131}I evaporated to the gaseous phase. The evaporation increased with time.
2) Particulate ^{131}I deposit faster than gaseous ^{131}I, The deposition of particulate ^{131}I increased with time compared with that of the gaseous ^{131}I.

Activity Ratios of ^{131}I to ^{137}Cs in Particulate Phase

$A_{131,p}$ / $A_{137,p}$ at Wako City and Chiba City, Japan	
Date	$A_{131,p}$ / $A_{137,p}$
March 14-15	13-23
March 20-21	7-18
March 22-23	147-231
April 9-10	15-16

Decay-Corrected $A_{131,p}$ / $A_{137,p}$ at Orangeburg, SC	
Date	$A_{131,p}$ / $A_{137,p}$
March 14	15-25
March 22	11-12
March 24	17-19
April 9	4.0-4.2

Conclusion: The $A_{131,g}/A_{131,p}$ ratio decreased slightly right after the H_2 explosions but decreased much as time past, suggesting two possibilities:

1) Particulate ^{131}I evaporated to the gaseous phase. The evaporation increased with time.
2) The deposition should not be the case because Cs should deposit more readily than iodine from the particulate phase.

Transport of the Radionuclides through the Atmosphere

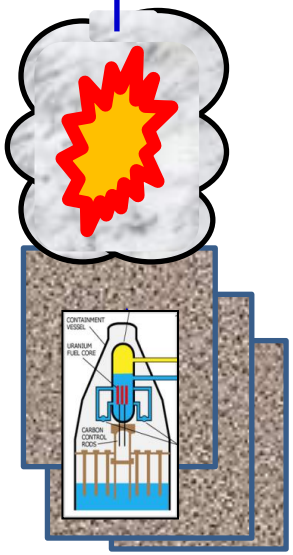
Jet Stream Transport

$I_2 + H_2O \rightarrow HI + HIO$, $I_2 + HR \rightarrow RI + HI$

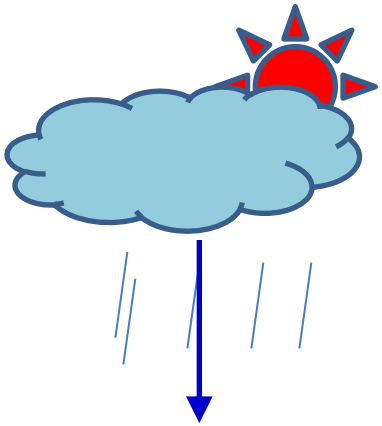
$HI + O_3 \rightarrow HIO_3$, $I_2 + O_3 \rightarrow I_2O_5$

$CsI + H_2O \rightarrow CsOH + HI$

H_2 Explosion HI, CsI, RI, I_2, HIO, $H\cdot$, $I\cdot$ HIO_3, I_2O_5



Steam Production From primary containment vessel HI, CsI



Nuclear Fallout

Gaseous phase

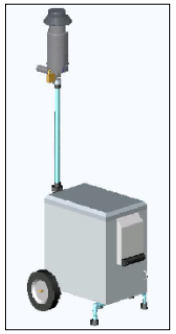
⇅

Aerosol phase

Air Sampling

Particulate-I: HI, I_2, CsI, HIO, HIO_3, I_2O_5

Gaseous-I: RI, I_2, HI



Conclusion: The particulate iodine spilled by the H_2 explosions contained more oxidized species IO^-, I_2O_5, IO_3^-, which are hard to turn into gaseous Iodine. The particulate iodine produced from the following events contained more reduced species and the element, HI, CsI, I2, which are easier to evaporate into the gaseous phase RI, I_2, HI.

Radiation Dose at Orangeburg

In its 10 CFR Part 20, US NRC, based on ICRP's data, defines the "effluents" of radionuclides for the assessment and control of dose to the public. The "effluent" concentration values are equivalent to the radionuclide concentrations which, if inhaled or ingested continuously over the course of a year, would produce a total effective dose equivalent of 50 mrem (0.5 mSv).

The effluents for airborne ^{131}I, ^{134}Cs, and ^{137}Cs are each 7.4 Bq m^{-3}.

The maximum airborne ^{131}I, ^{134}Cs, and ^{137}Cs at Orangeburg was 6.0 (gas & aerosol), 0.11, and 0.12 mBq m^{-3} for < 3 weeks.

Consider the time factor, the dose equivalent of the fission products from Fukushima accident at Orangeburg were: 23, 0.43, and 0.47 nSv, respectively.

Radionuclides	^{131}I	^{134}Cs	^{137}Cs
US NRC's Effluent (Bq m^{-3})	7.4	7.4	7.4
Effective dose equivalent for one year (mSv)	0.5	0.5	0.5
Maximum Fukushima fission products concentrations at Orangeburg for < 3 weeks (Bq m^{-3})	6.0×10^{-3}	1.1×10^{-4}	1.2×10^{-4}
Maximum dose equivalent from Fukushima fission products at Orangeburg (mSv)	23×10^{-6}	4.3×10^{-7}	4.7×10^{-7}

Conclusion

- Both gaseous and particulate fallout (^{131}I, and $^{134,137}Cs$) from the Fukushima nuclear accident were observed at Orangeburg, South Carolina from 15 March until 30 May 2011.
- It was found that the maximum radioactivity concentrations of gaseous and particulate ^{131}I were 5.0 ± 0.4 and 1.0 ± 0.1 mBq m^{-3}, respectively; the highest concentrations of ^{134}Cs and ^{137}Cs were about 10 times less than that of the particulate ^{131}I.
- Based on the plateaus and spikes in the histograms, the arrival times of the particulate and gaseous fractions of the Fukushima fallout were determined as 8 and 10 days, respectively.
- The average activity ratio of ^{134}Cs to ^{137}Cs was found to be 0.98 ± 0.26, which agrees with the literature data observed in Japan
- According to the analysis of radioactivity ratios in gaseous and particulate phases, the phase transfer and chemical states of iodine was studied.

Acknowledgements

- The research was supported by US DOE Environmental Management. DOE Award#: DE-EM0000594.
- Dr. Timothy A. DeVol, CHP, provided professional advice and review on evaluation of the radiation dose from airborne radionuclides. Dr. DeVol is Toshiba Professor of Nuclear Engineering in the Environmental Engineering & Earth Sciences, College of Engineering and Science, Clemson University.

Comparison of the Dose in Daily Life

US NRC Doses in Daily Life	Dose (mSv)	Eq. to the Fukushima dose at Orangeburg
One dental X-ray	0.015	6.3×10^2
Annual safe drinking water (EPA)	0.04	1.7×10^3
One chest X-ray	0.1	4.2×10^3
Annual dose from one's body	0.4	1.7×10^4
Annual public dose (NRC)	1	4.2×10^4
Annual US Average natural background	3.1	1.3×10^5
Annual US Average background	6.2	2.6×10^5
One whole body CT	10	4.2×10^5
Annual nuclear whole body dose limit	50	2.1×10^6

Background Radiation and Regulations

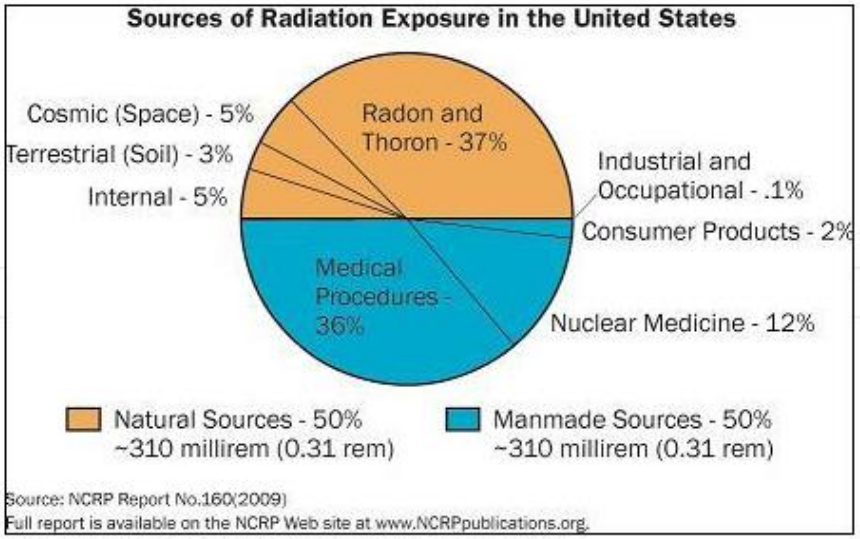
Highest ^{131}I level detected 50m above SCSU Campus: ~0.3 pCi/M^3 (3/25-29/11)

Nuclear Regulatory Commission (NRC) Limits for ^{131}I in Air:

200 pCi/M^3 (for effluent air to which the public could exposed)
20,000 pCi/M^3 (for occupational air exposure)

Nuclear Regulatory Comission (NRC) Limits for ^{131}I in Water:

1 million pCi/M^3 (for effluent water)
10 million pCi/M^3 (for monthly average release to seweas from medical facilities)

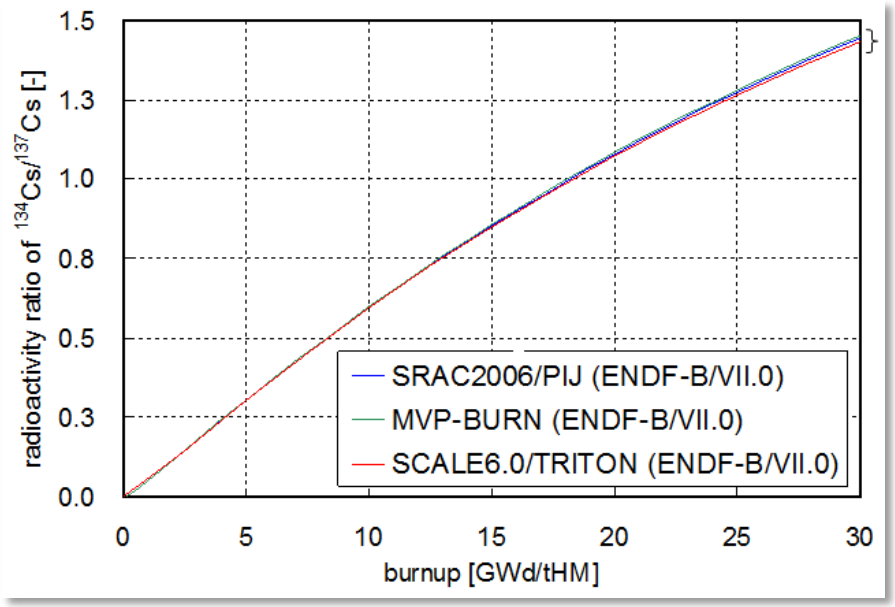


Radioactivity Ratio: A_{134}/A_{137}

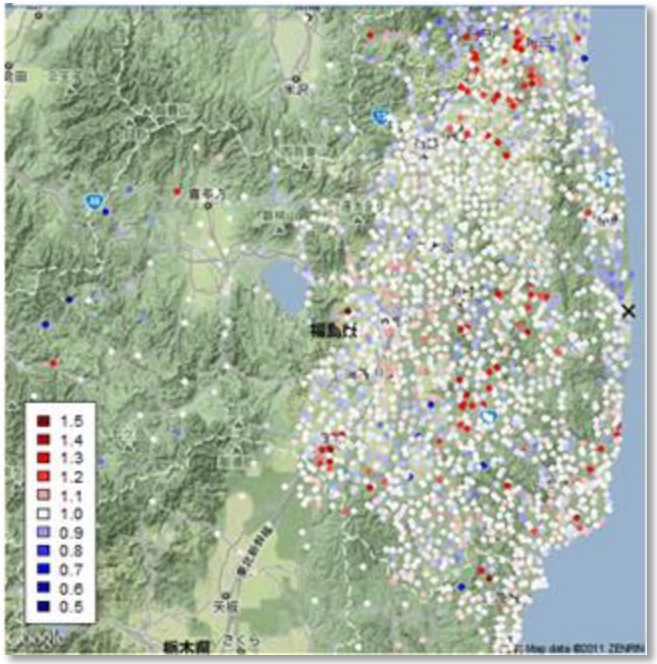
From the average of 5 aerosol samples measured by the well-type HPGe detector:

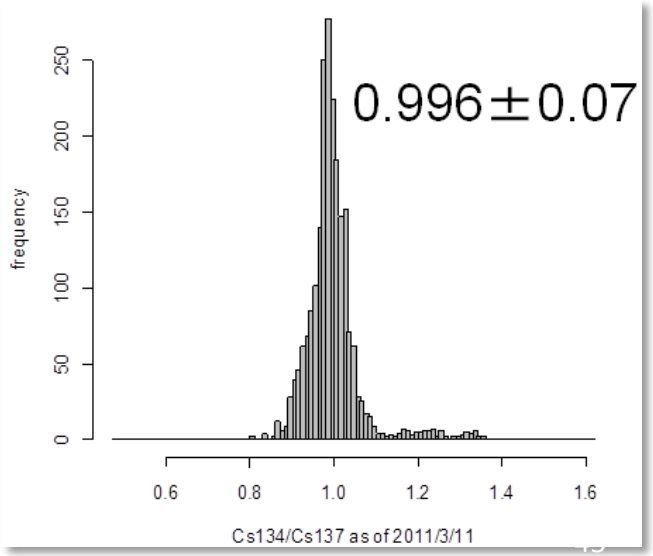
$$A_{134}/A_{137} = 0.98 \pm 0.26$$

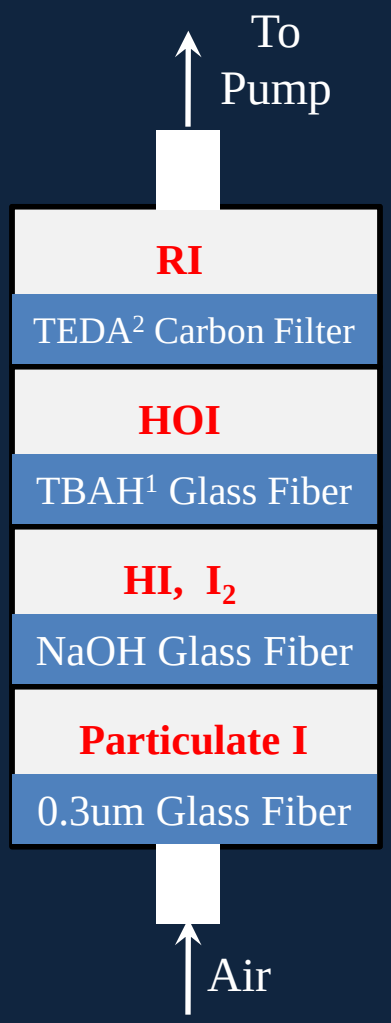
This agrees with the ratio from over thousand measurements in Japan



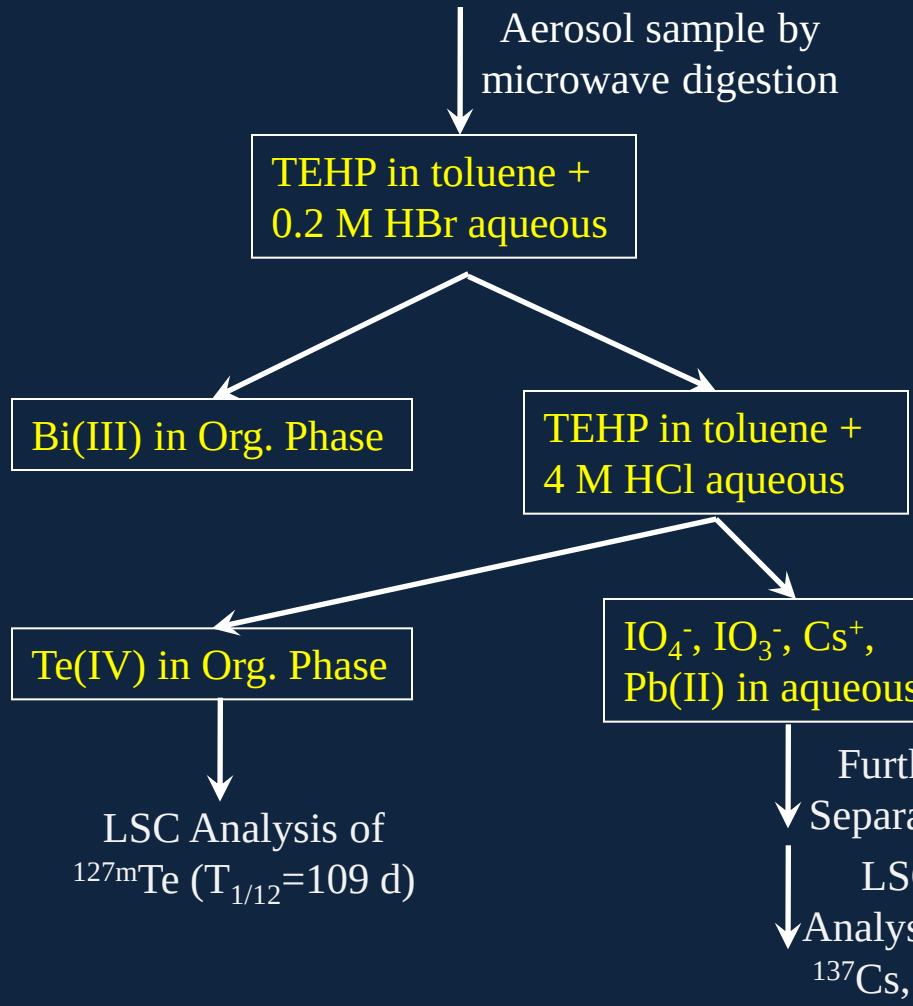
From A_{134}/A_{137} = 1, the burnup of the reactor units were estimated to be: 17.2 ± 1.5 GWd/tHM. – *Endo, T. et al Sato, S., Yamamoto, A, Nagoya University*.







1 Tributylene hydroxide, 2 TriEthyl Diamine
Hans-Eike G~ibler and Klaus G. Heumann, "Determination of atmospheric iodine species using a system of specifically prepared filters and IDMS", Fresenius J Anal Chem (1993) 345 : 53 - 59



G. S. Desai and V. M. Shinde, "Extraction and Separation Studies of Tellurium(IV) with Tris-(2 Ethyl Hexyl) Phosphate ", Talanta, Vol. 39, No. 4, pp. 405408, 1992

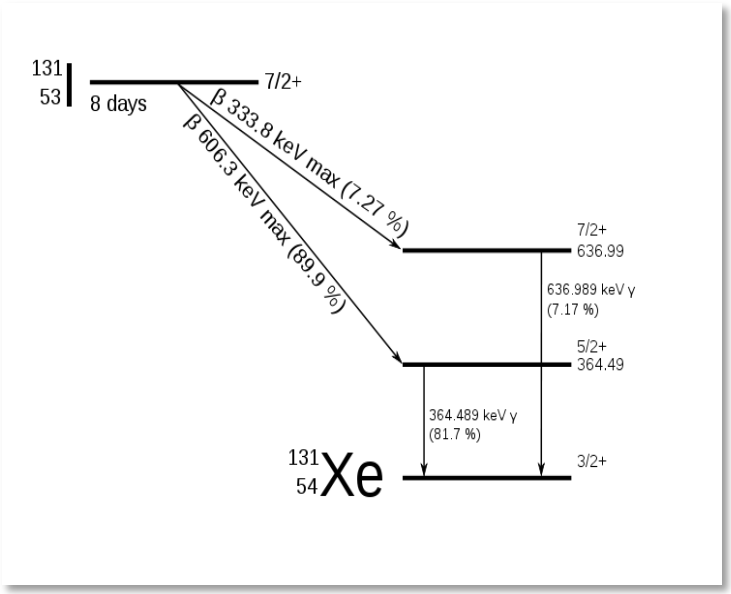
Radioactive Decay of ^{131}I

^{131}I decays with a half-life of 8.02 days by emitting b-particles and γ-rays. On decaying, ^{131}I most often (89% of the time) expends its 606.3 keV of decay energy by transforming into the stable ^{131}Xe in two steps, with a g-transition following rapidly after the β-decay:

$$^{131}_{53}\mathrm{I} \rightarrow {}^{131}_{54}\mathrm{Xe}^{*} + \beta + \bar{\nu}_e \quad \mathrm{Q} = 606\,\mathrm{keV}$$

$$^{131}_{54}\mathrm{Xe}^{*} \rightarrow {}^{131}_{54}\mathrm{Xe} + \gamma \quad \mathrm{Q} = 364.5\,\mathrm{keV}$$

the primary emissions of ^{131}I decay are thus β-particles with a maximal energy of 606.3 keV (89% abundance, others 248 - 807 keV) and 364 keV γ-rays (81% abundance, others 723 keV). β-decay, as always in this process, also produces an antineutrino, which carries off variable amounts of the β-decay energy.



Estimation of the Detection Limit

$$LLD = (2.71 / T_S) + 3.29 (R_b / T_b + R_b / T_s)^{1/2}$$

R_b : count rate of the natural background

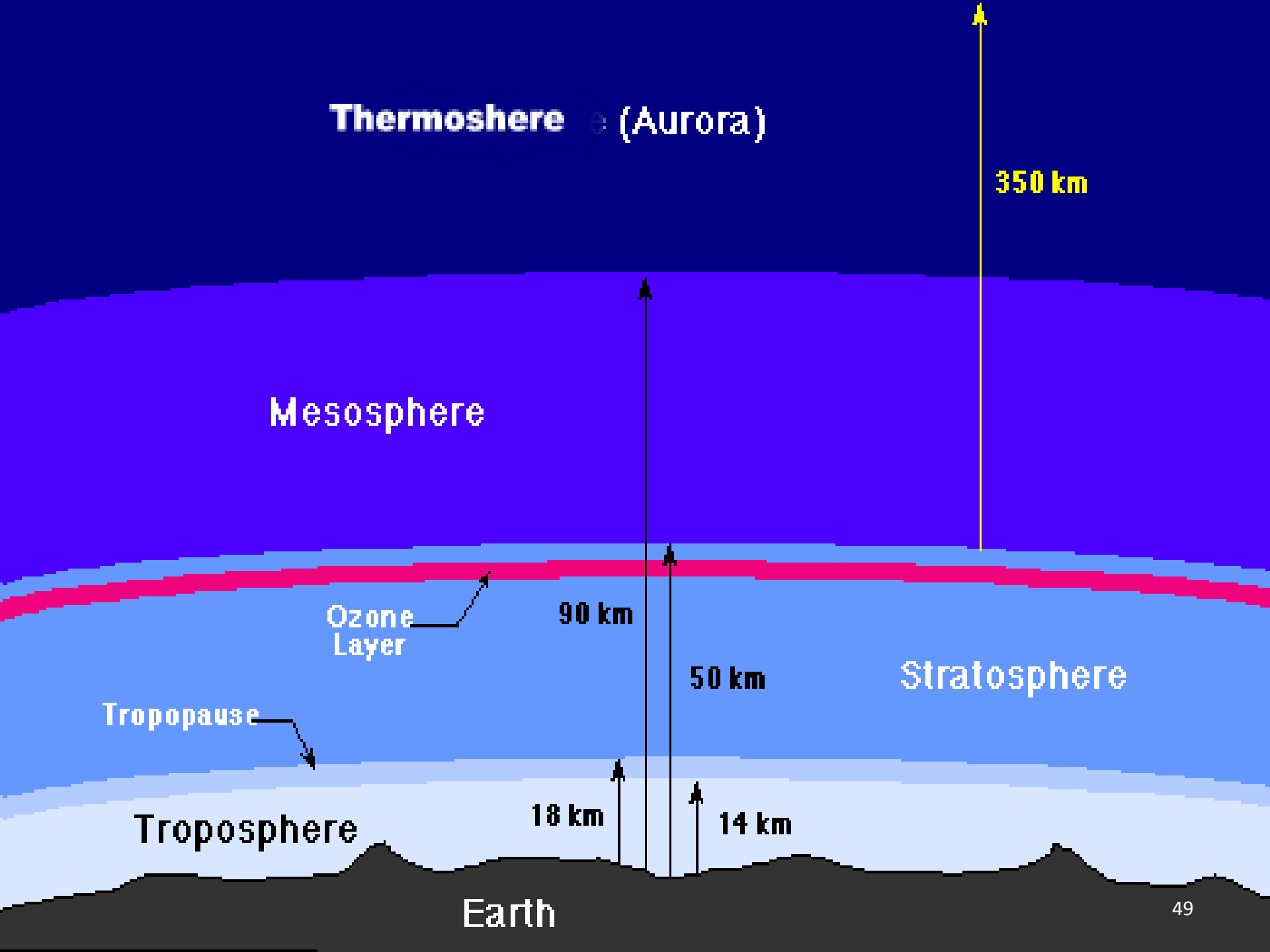
T_b : time of the background count

T_s : time of the sample count

ε : detection efficiency of the counter

Co-axle Detector : ^{131}I at 364.5 keV: 0.01 cpm, ^{137}Cs at 661.7 keV: 0.005 cpm

Well-type Detector : ^{131}I at 364.5 keV: 0.004 cpm, ^{137}Cs at 661.7 keV: 0.002 cpm


Thermoshere
(Aurora)
350 km
Mesosphere
Ozone
Layer
90 km
50 km
Stratosphere
Tropopause
18 km
14 km
Troposphere
Earth