LONG RANGE TRANSPORT OF FUKUSHIMA RADIOACTIVE PLUME TO EUROPF

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Nuclear Physics Lab

Laboratory Equipment on Radiation Measurements

2 HPGe (42%)

1 HPGe (20%)

1 Ge-planar

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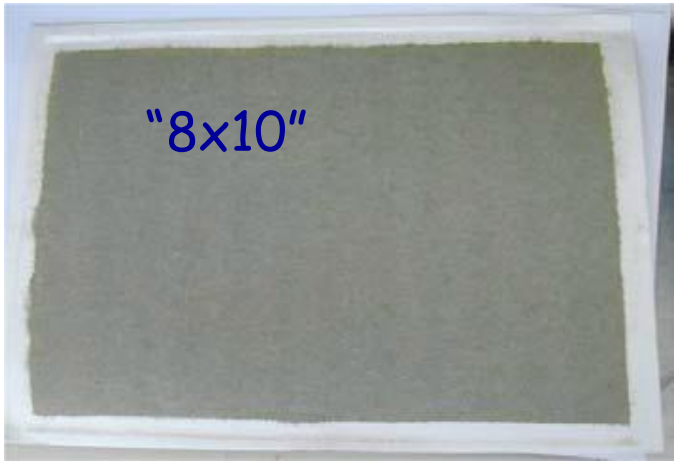
Activity concentrations of ^{131}I, ^{137}Cs and ^{134}Cs in air

HV air Sampler (Staplex TFIA-2)

Flow rate: 1.6-1.7 m^3/min (60cfm)
Sampling duration: 23 h
Total Volume: 2400-2700 m^3
Air Volume Uncertainty (2σ): 30-50 m^3
Position: Thessaloniki 40°N, Milano 45°N

Glass Fiber Filters TFAGF810

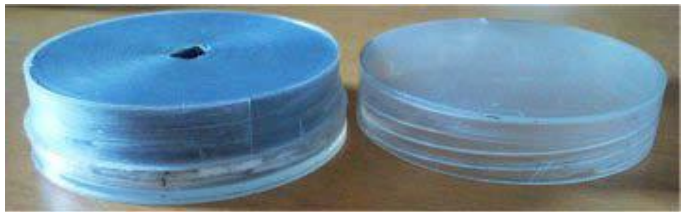
Very high retention of fine particles. 99.98% retention efficiency of 0.3 micron particles.



Air-filters analysis by gamma ray spectra

Air filters are measured at least 10h after the end of sampling

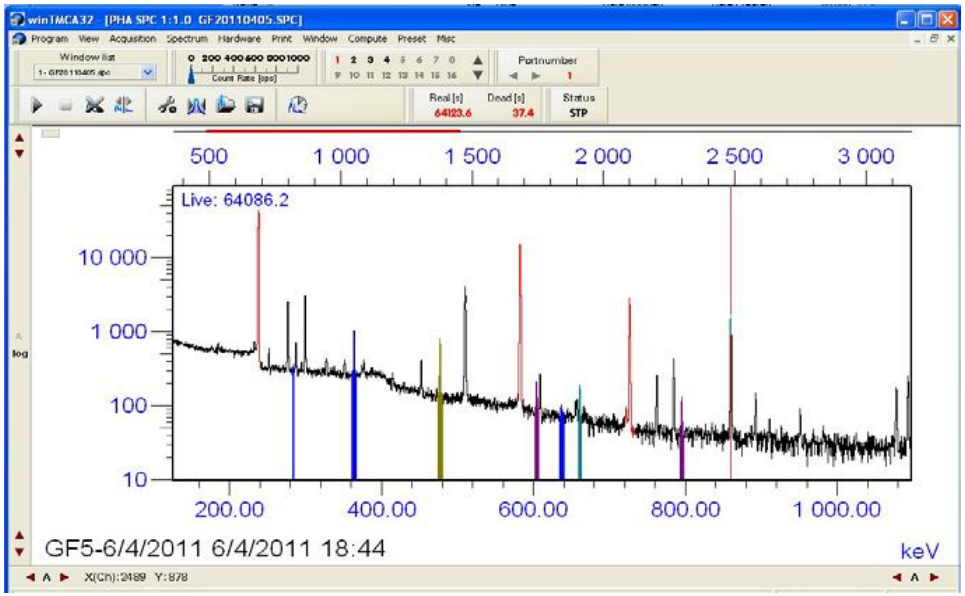
I-131 (364 keV)
Cs-137(662 keV)
Cs134 (605 keV & 796 keV) correction due to summation effect



gamma-spectroscopy system

Ge Detector: Relative efficiency 42.3%
FWHM 1.8 keV(1332 keV Co-60)

g-spectroscopy,
picoSPEC2 (Target)
based in DSP technology



Activity concentrations of ^{131}I, ^{137}Cs and ^{134}Cs in air

Date of sampling	Fallout isotopes in surface air				
	^{131}I μBq m^{-3}	^{137}Cs μBq m^{-3}	^{134}Cs μBq m^{-3}	ratio $^{134}Cs/^{137}Cs$	ratio $^{131}I/^{137}Cs$
31/03/11	322±35	< 29[a]	< 26[a]	-	-
02/04/11	335±89	59±42	56±37	0.95	5.7
03/04/11	467±25	40±9	37±8	0.92	11.7
05/04/11	323±16	25±9	27±9	1.09	12.9
07/04/11	438±28	26±17	25±15	0.98	16.8
09/09/11	209±33	56±30	54±24	0.97	3.7
10/04/11	229±55	63±30	61±22	0.97	3.6
11/04/11	285±43	[b]27±18	[b]23±14	0.90	10.6
12/04/11	333±73	60±38	56±30	0.94	5.6
14/04/11	343±48	57±26	54±23	0.95	6.0
15/04/11	220±58	47±27	42±20	0.89	4.7
16/04/11	161±34	39±10	[b]13±7	0.33	4.1
17/04/11	118±27	44±17	31±13	0.69	2.7
19/04/11	107±30	29±16	40±14	1.38	3.7
20/04/11	107±38	38±16	23±12	0.62	2.8
21/04/11	128±33	[b]17±16	27±14	1.59	7.5
22/04/11	94±46	35±16	< 11[a]	-	2.7
28/04/11	[b]60±35	[b]23±16	< 12[a]	-	2.6
29/04/11	< 41[a]	< 11	33±14	-	-
30/04/11	< 19[a]	< 16[a]	< 12[a]	-	-
3/05/11	< 9[a]	17±16	22±14	1.34	-

[a] MDA
[b] Critical Level

$^{134}Cs/^{137}Cs$ =1

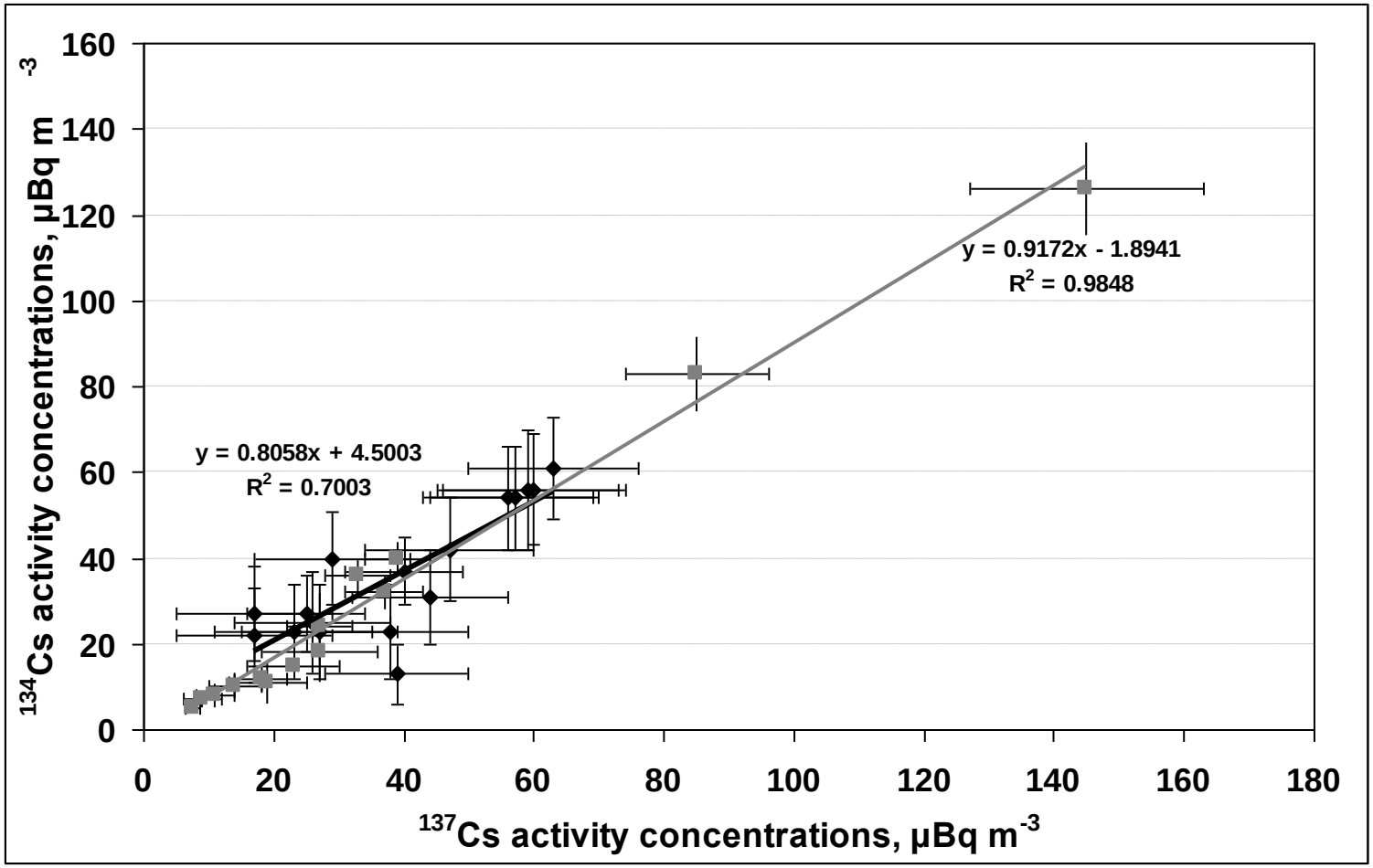
Related to the burn-up history of the nuclear fuel of the destroyed nuclear reactor

^{131}I <500 μBq m^{-3}

$^{131}I/^{137}Cs$ decrease with time

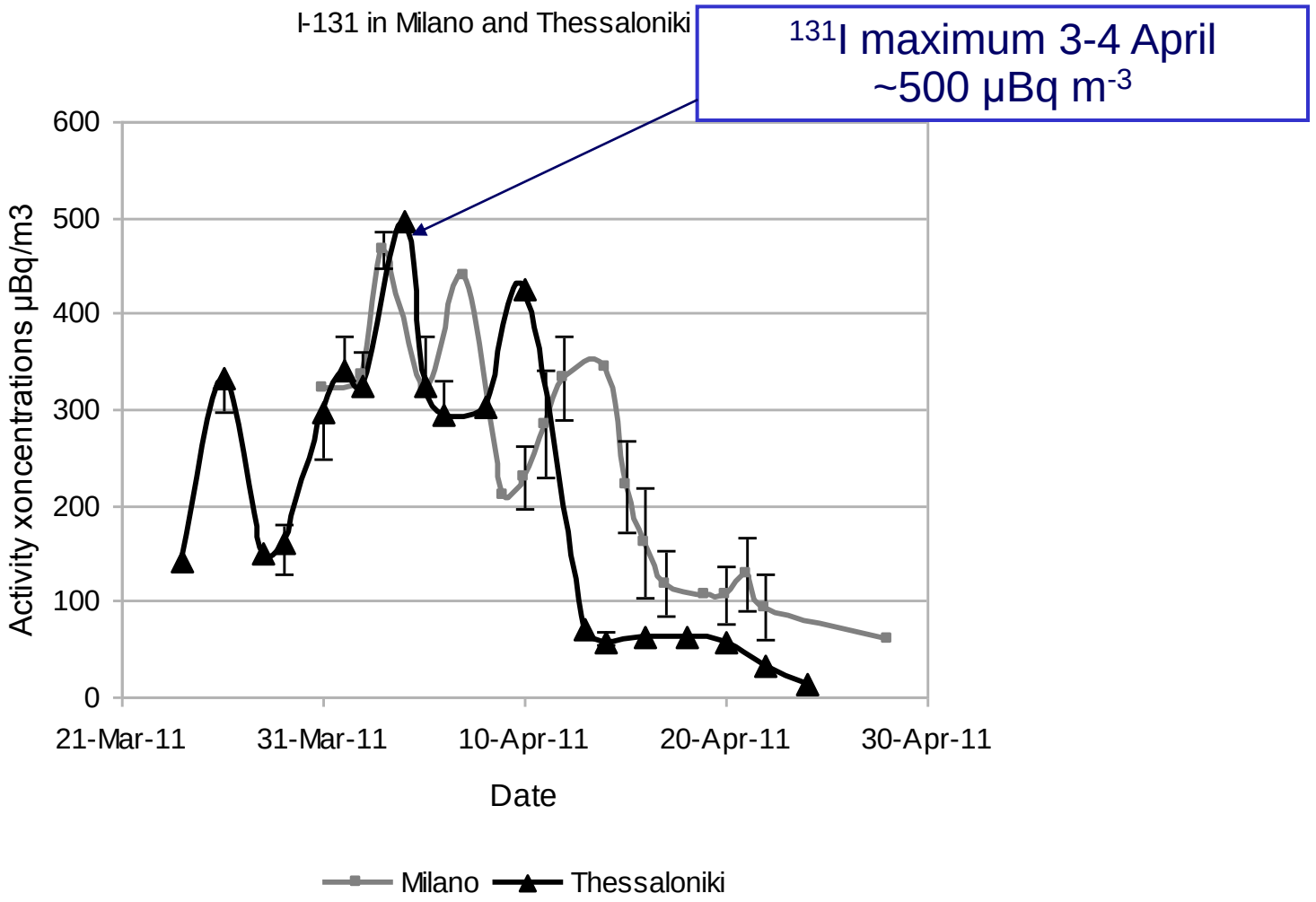
Reflects the different volatility, attachment and removal of the two isotopes during transportation due to their different physico-chemical properties.

$^{134}Cs/^{137}Cs$ activity ratio = 1

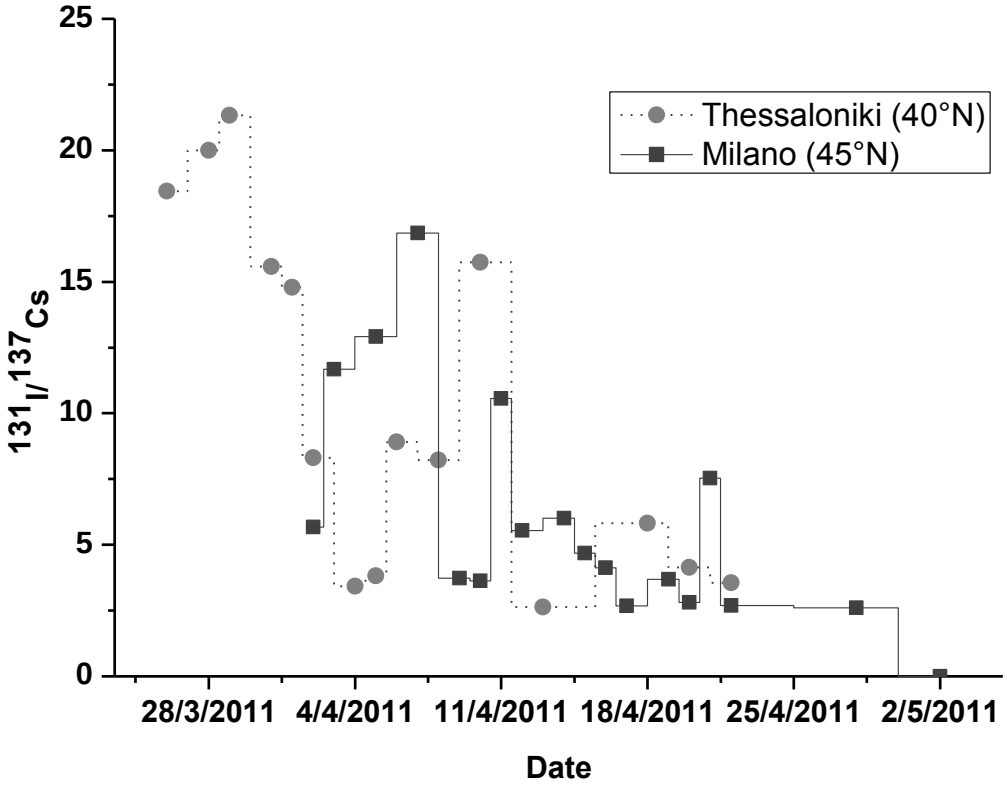


^{134}Cs concentrations versus ^{137}Cs concentrations in Milan (45°), Italy (black dots) and Thessaloniki (40°), Greece (gray dots)

^{131}I atmospheric concentrations

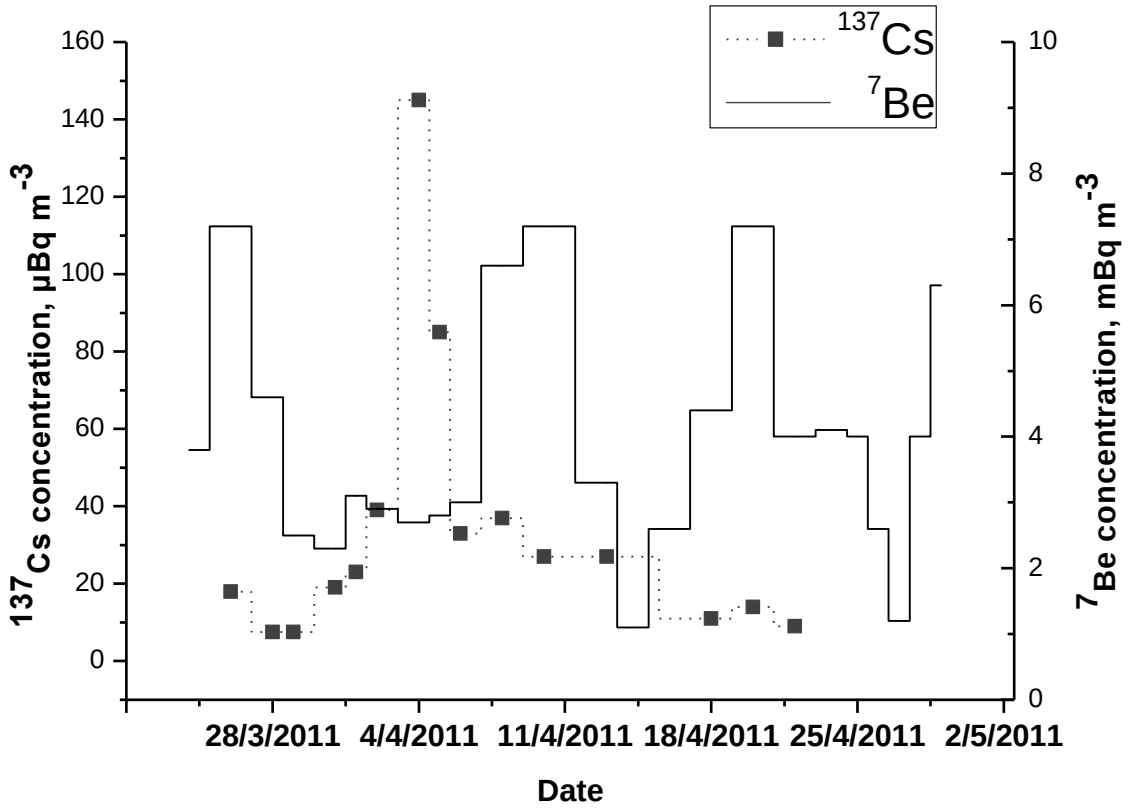


$^{131}I/^{137}Cs$ atmospheric concentrations

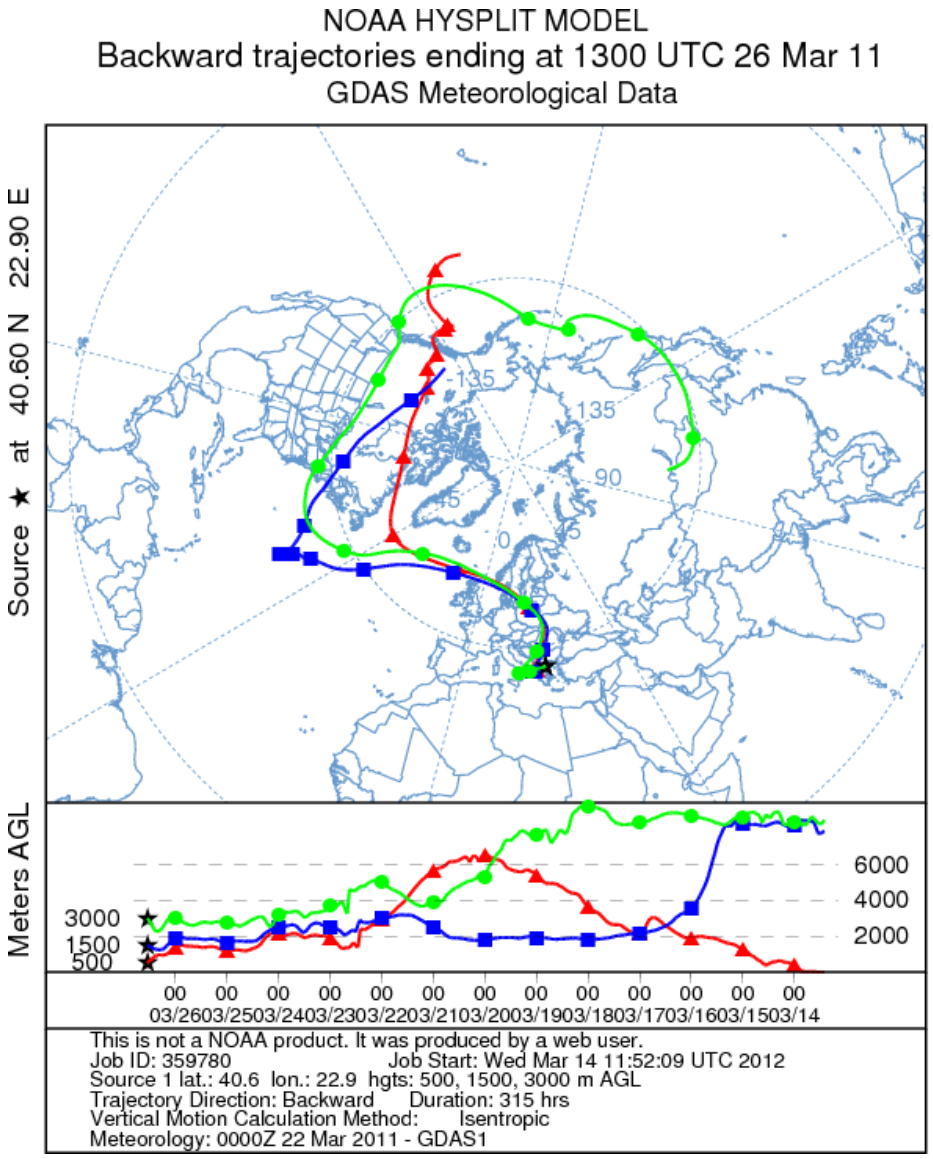


$^{131}I/^{137}Cs$ activity ratio in Milan (45°) and Thessaloniki (40°)

^{137}Cs and ^{7}Be activity concentrations in Thessaloniki (40°N)



Back trajectories analysis

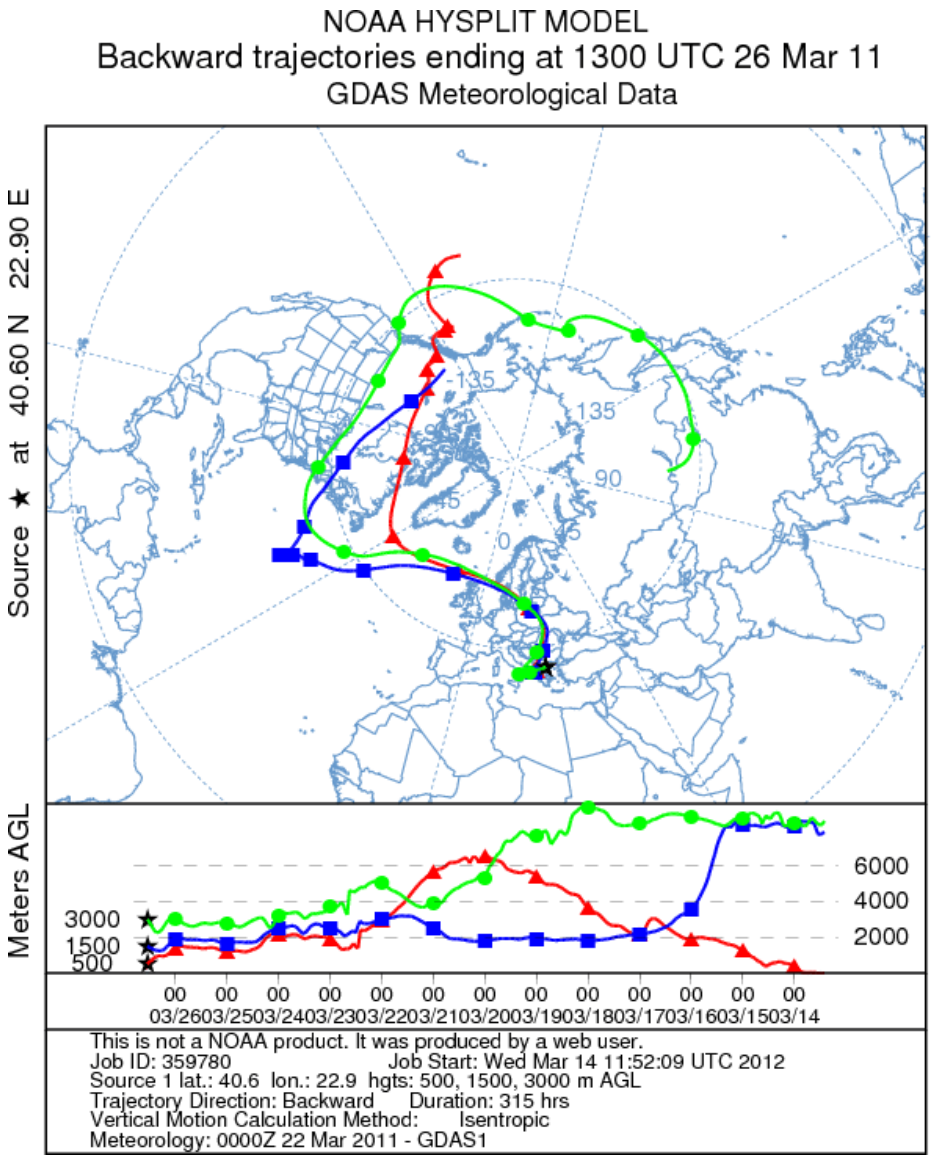


The NOAA HYSPLIT model was used to assess the transport pattern and to explain the deviation in radionuclide activity concentrations found.

Thirteen days (312) back trajectories were calculated for different arrival height and for 12 UTC time.

The trajectories are labeled every 24h by a filled symbol.

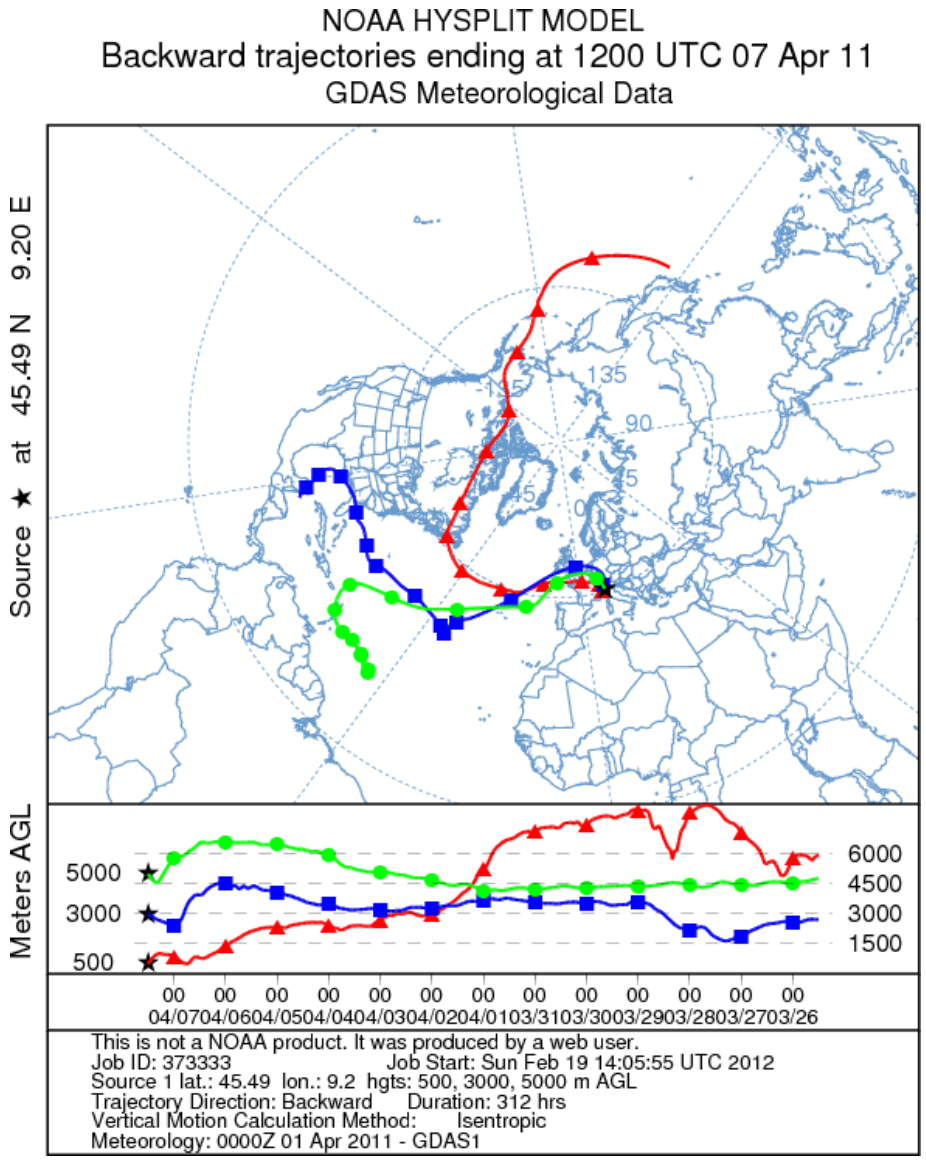
Back trajectories analysis



The first maximum in concentration of ^{131}I (332 μBq m^{-3}) was observed at Thessaloniki on **26th of March** 2011. Air masses were lifted rapidly and transported over the North America to Europe at height of 500 m.

Air masses were also traveled at higher atmosphere levels from Japan. It is also possible that radioactive particles were transported at higher altitudes and may have been removed in the lower layer of the atmosphere due to various reasons, e.g. rainfall characteristics, fog formation or growth of aerosol particles and their deposition.

Back trajectories analysis

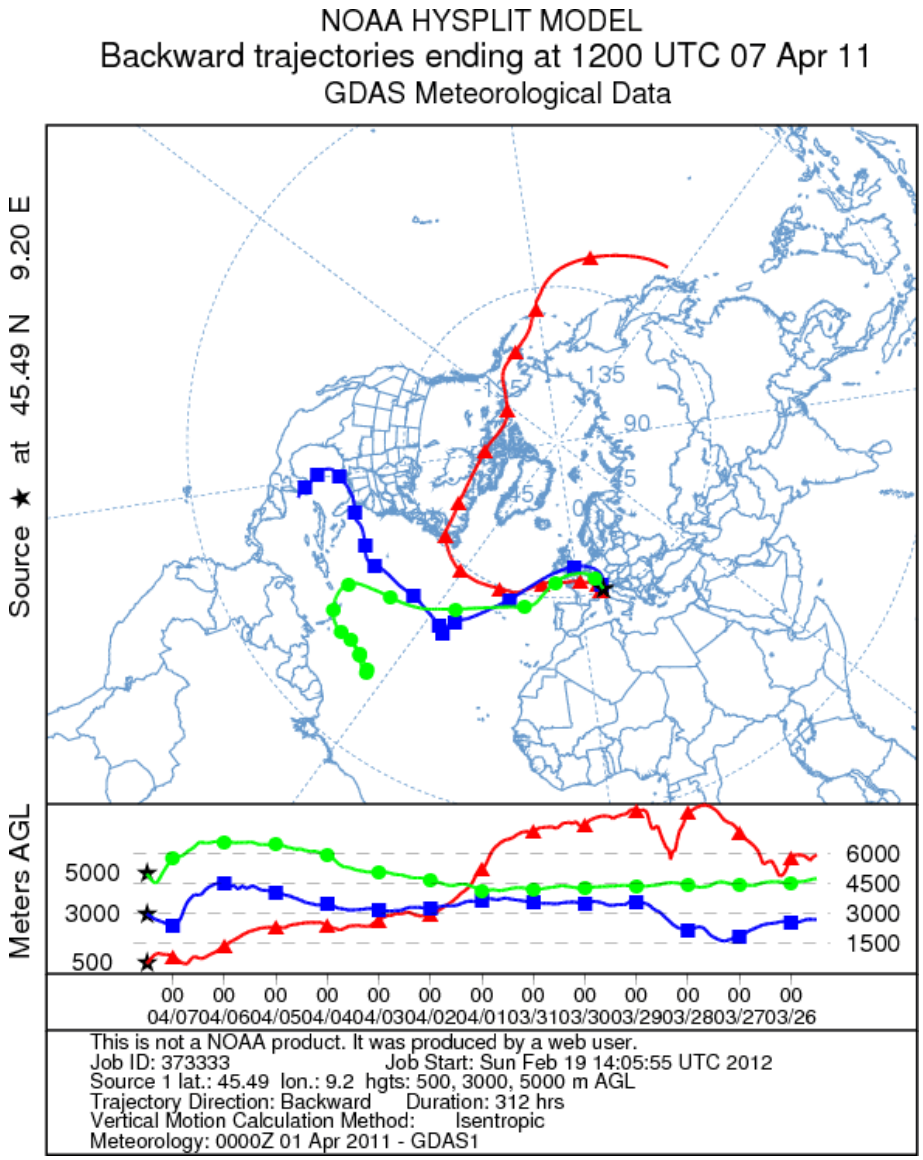


An example of transported air mass at **07 of April 2011** at Milano is presented.

The results showed a direct transfer from Fukushima across the Pacific Ocean, a transport through the North Pole and a pathway through the Greenland and Iceland at height of 500 m to Milano.

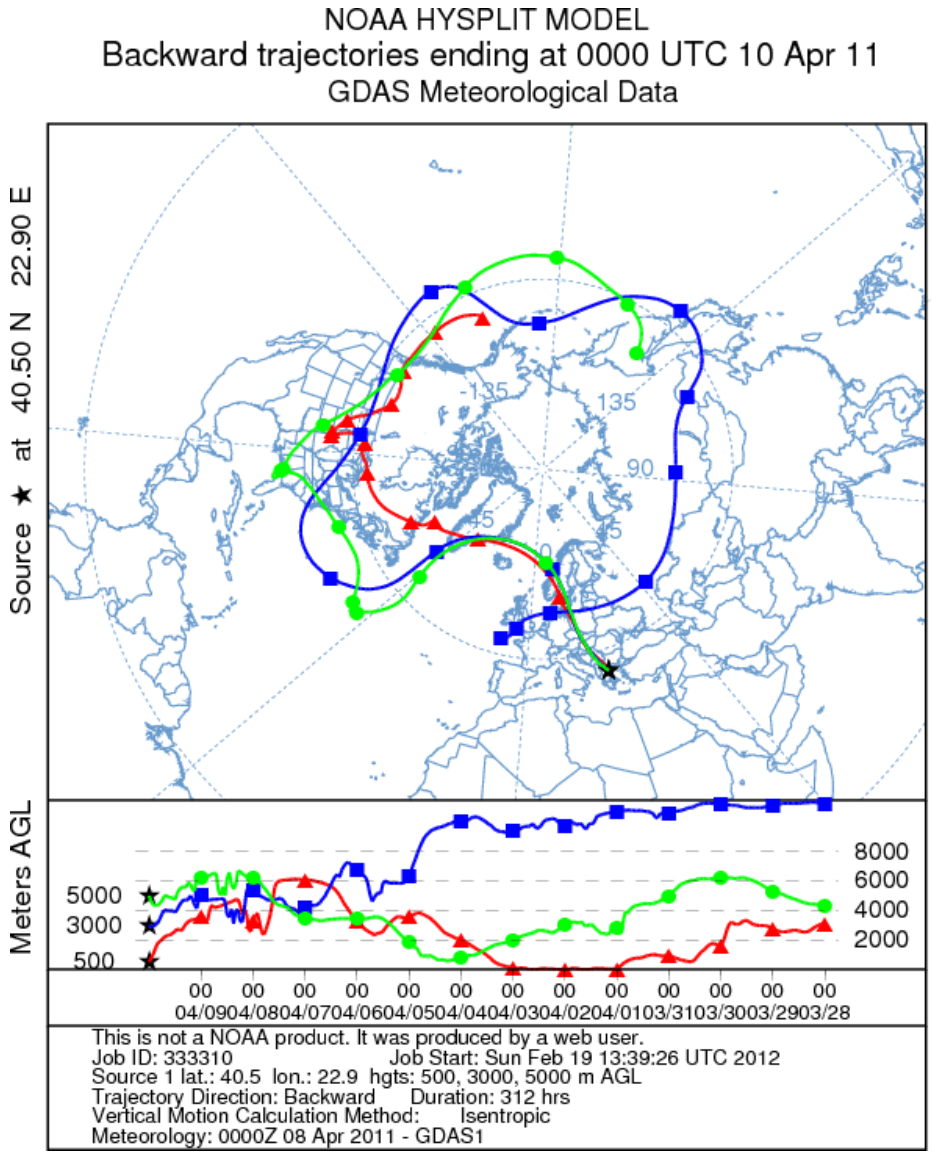
The air masses at higher altitudes were rapidly transported, while the air masses at 500 m exhibited rather slow transport.

Back trajectories analysis



Although the second maximum of concentration of ^{131}I that observed at Milan on 07th of April can be attributed to the advection of air masses from Japan at altitude of 500 m, however, the back-trajectory analysis **for the same day at Thessaloniki indicates no transport of air masses from Japan**, at least for height of 500 and 1500 m.

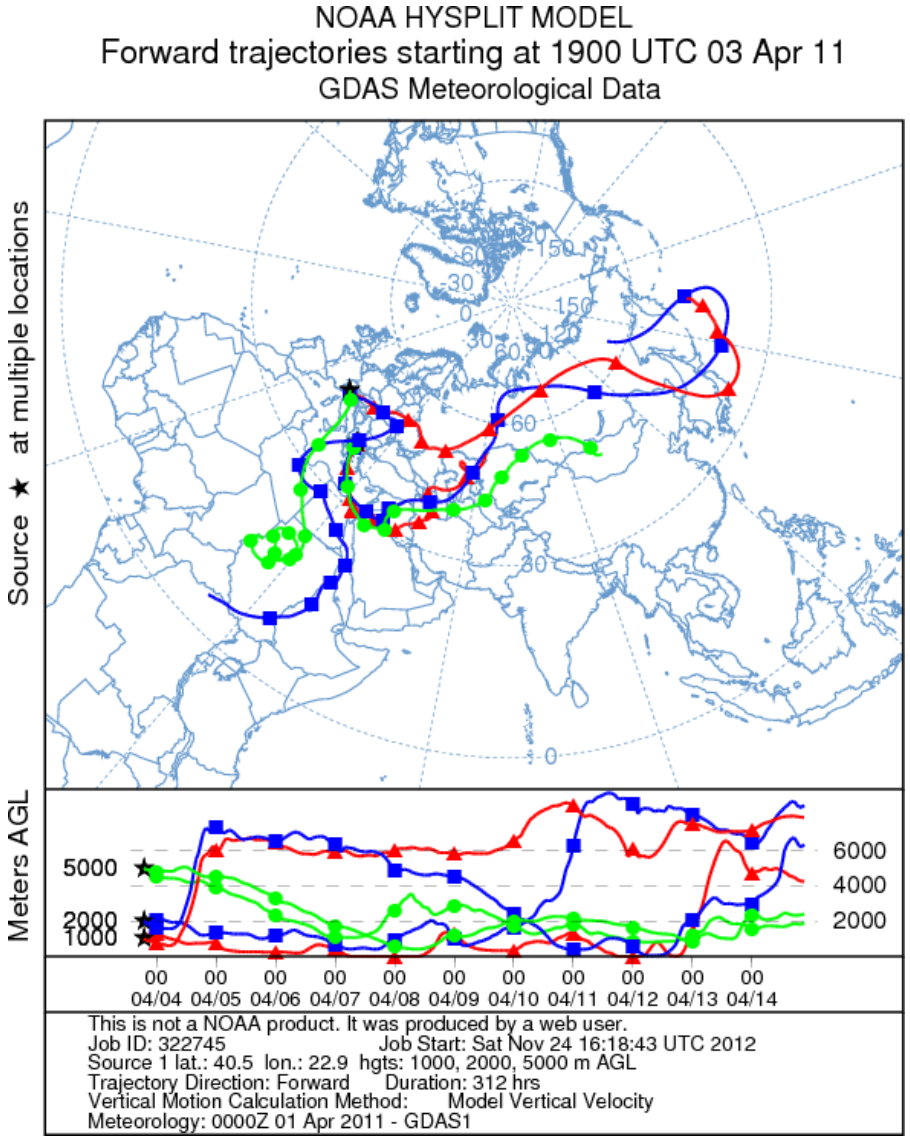
Back trajectories analysis



On **10th of April** back-trajectory analysis showed a direct transfer from Fukushima across the Pacific Ocean, a transport through the North Pole and a pathway through the Greenland and Iceland to Thessaloniki.

The air masses on that day reach Thessaloniki from Northwest direction and this is possible the reason why no maximum concentration was observed at Milan.

Back trajectories analysis



On **April 3, 2011** at 19:00 UTC the back-trajectory analysis indicates the transport of air masses from Japan both in Italy and in Greece but at different arrival heights. As Fig. 8 shows the air masses started above Japan at around 2 km. In Italy the air mass moved down, travelled near the ground and arrived above Italy at height of 1 km. On the other hand, the air mass after being near the ground for almost 2 days rose to arrive at height of 2 km above Thessaloniki. Both transport pathways can explain the maximum concentrations that were observed at the regions of study.

Wet and Dry deposition

The most effective transfer path for airborne radioisotopes to the ground

Thessaloniki

29/3/2011	0.18cm
I-131:	1.21±0.31 Bq m^{-2}
14/4/2011	0.48 cm
I-131:	0.46±0.13 Bq m^{-2}
18/4/2011	0.91 cm
I-131: < 30 /m	
Cs-137, Cs-134: < 40 /l	

Wet deposition of ^{131}I, ^{137}Cs and ^{134}Cs

Fallout isotopes in rainwater samples

Site	Date of Sampling	Volume	Surface area m^2	^{131}I mBq L^{-1} (*Bq m^{-2}*)	^{137}Cs mBq L^{-1} (*Bq m^{-2}*)	^{134}Cs mBq L^{-1} (*Bq m^{-2}*)
Segrate	28/03/11	0.685	0.1739	891±115 (*3.51±0.45*)	[b]122±89 (*0.48±0.35*)	< 58[a] (*<0.23*)
Senago	28/03/11	0.500	0.1739	725±133 (*2.08±0.38*)	< 11[a] (*<0.03*)	< 86[a] (*<0.25*)
Segrate	12/04/11	0.016	0.5217	<36[a] (*<0.011*)	859±435 (*0.271±0.137*)	<308[a] (*<0.097*)
Segrate	15/04/11	0.925	0.5217	291±87 (*0.52±0.15*)	45±32 (*0.08±.0.06*)	57±26 (*0.10±.0.05*)

[a] MDA
[b] Critical Level

$^{131}I < 1 Bq\ L^{-1}$

The Food and Drug Administration (FDA) fixed intervention level for ^{131}I in drinking water and infant milk, to 170 Bq L^{-1} while in Japan, the ^{131}I limit for consumption of tap water is 100 Bq L^{-1} for infants, and 300 Bq L^{-1} for adults (RIKEN, 2011).

Dry deposition of ^{131}I, ^{137}Cs and ^{134}Cs

Date of Sampling	Fallout isotopes in dry deposition samples			
	^{131}I Bq m^{-2}	^{137}Cs Bq m^{-2}	^{134}Cs Bq m^{-2}	ratio $^{134}Cs/^{137}Cs$
06/04/11	0.40±016	0.24±0.11	< 0.05[a]	-

Dry Deposition:

^{131}I: 11% of wet

^{137}Cs: 50% of wet

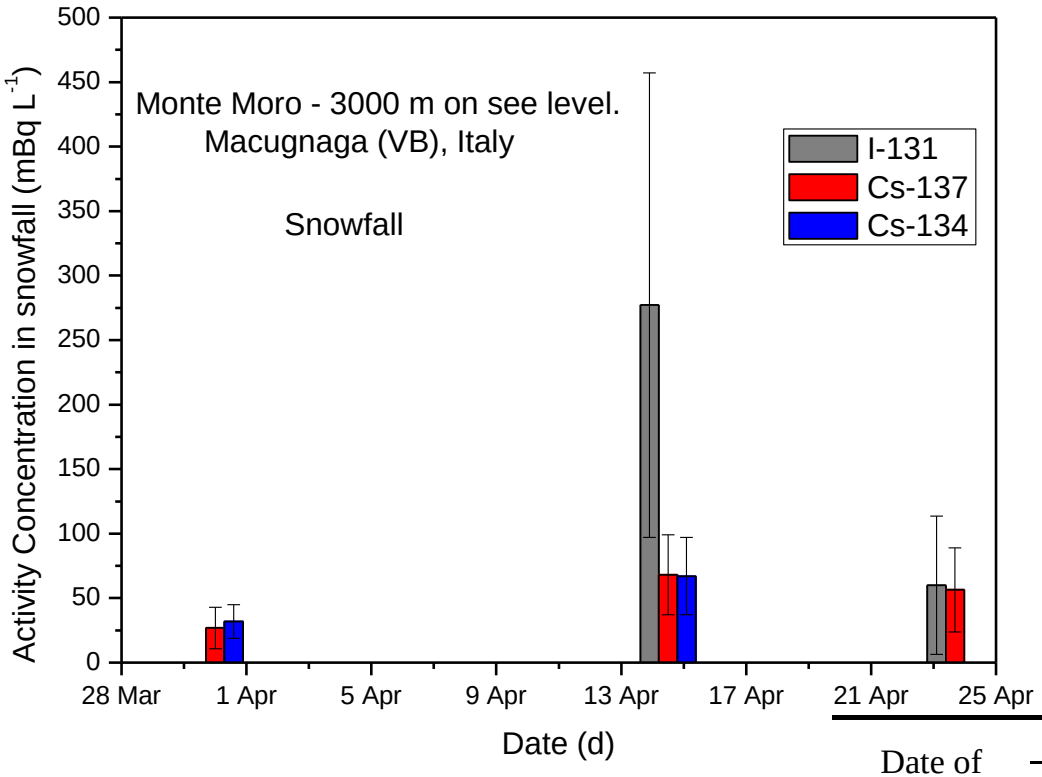
- If these data are compared with the ones obtained by rainwater, it is clear that the dry deposition of ^{137}Cs and ^{134}Cs is greater than that of ^{131}I.
- This can be explained because, contrary to the iodine mainly found in gaseous form, caesium is rapidly bound to aerosols and thus highly subject to dry deposition.

Activity concentrations of ^{131}I, ^{137}Cs and ^{134}Cs in snow

Samples collected at 3000 m s.l.m.

Monte Moro - Macugnaga (VB), Italy

Activity concentrations of ^{131}I, ^{137}Cs and ^{134}Cs in snow



Date of Sampling	Fallout isotopes in snowfall samples			
	^{131}I mBq L^{-1}	^{137}Cs mBq L^{-1}	^{134}Cs mBq L^{-1}	ratio $^{134}Cs/^{137}Cs$
28/03/11	< 12.04[a]	< 8.98[a]	< 6.92[a]	-
31/03/11	< 20.88[a]	27±16	[b]32 ±13	1.19
14/04/11	277±180	68±31	67±30	0.98
23/04/11	[b]60±53	56±33	< 29[a]	

^{131}I, ^{137}Cs and ^{134}Cs in grass

Total surface: 1 m^2

Total mass: 0,35-0.45 kg

Date of Sampling	Fallout isotopes in grass samples			
	^{131}I mBq kg^{-1}	^{137}Cs mBq kg^{-1}	^{134}Cs mBq kg^{-1}	ratio $^{134}Cs/^{137}Cs$
30/03/11	66±24	47±19	<18[a]	-
06/04/11	[b]37±33	60±20	[b]21±17	0.3
13/04/11	<51[a]	[b]41±35	<30[a]	-
20/04/11	[b]135±119	89±32	<21[a]	-

Surface deposition

^{131}I: 0.016-0.029 Bq m^{-2}

^{137}Cs: 0.014-0.026 Bq m^{-2}

Dry Deposition:

^{131}I: 0.40 Bq m^{-2}

^{137}Cs: 0.24 Bq m^{-2}

^{131}I, ^{137}Cs and ^{134}Cs in soil

Site	Date of Sampling	Fallout isotopes in soil samples			
		^{131}I Bq kg^{-1}	^{137}Cs Bq kg^{-1}	^{134}Cs Bq kg^{-1}	ratio $^{137}Cs/^{134}Cs$
Segrate	30/03/11	0.63±0.29	12.26±0.70	0.83±0.30	0.07
Senago uncovered	04/04/11	0.57±0.25	85.17±4.40	0.29±0.13	0.0034
Senago covered	04/04/11	<0.15[a]	84.65±4.40	0.47±0.28	0.01
Segrate[b]	06/04/11	0.85±0.34	18.73±1.02	[c]0.48±0.27	0.03
Segrate	13/04/11	0.95±0.60	18.65±1.04	< 0.21[a]	-
Segrate	20/04/11	1.99±1.32	19.08±1.05	< 0.19[a]	
Segrate	04/05/11	< 0.24[a]	9.62±0.56	0.45±0.19	0.05
Segrate	11/05/11	< 0.21[a]	11.99±0.63	< 0.06	
Segrate	18/05/11	< 0.48[a]	24.95±1.30	< 0.07	

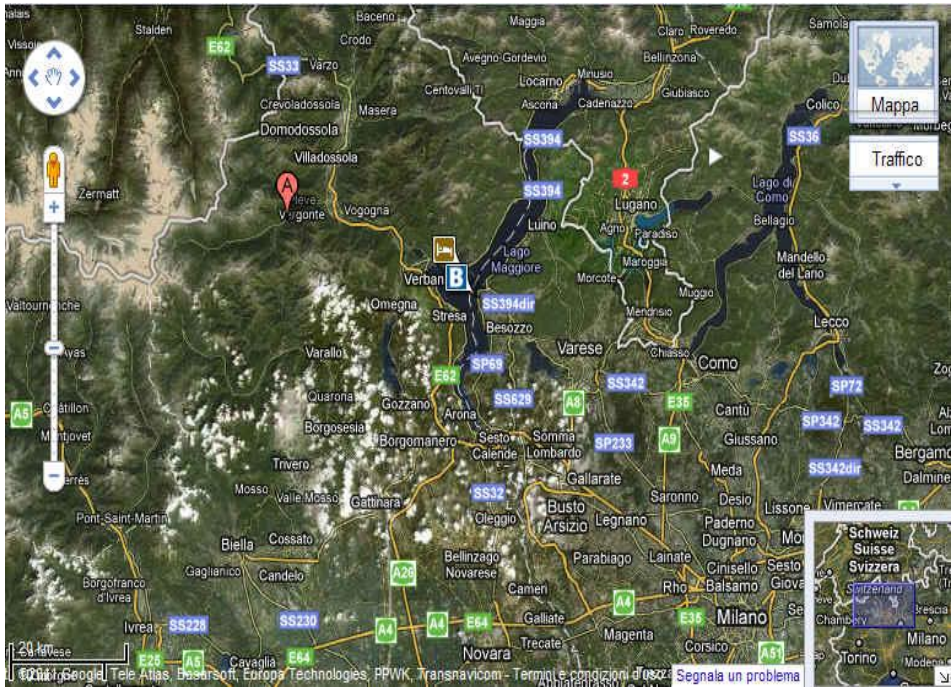
[a] MDA
[b] Sample taken in an unplowed area
[c] Critical Level

Dry Deposition:

^{131}I: 0.70 Bq m^{-2}

^{131}I, ^{137}Cs and ^{134}Cs in milk

Samples of sheeps and cows milk
collected in Val Anzasca (VB), Italy at 400 m s.l.m.



^{131}I, ^{137}Cs and ^{134}Cs in milk

Fallout isotopes in milk samples								
Date of Sampling	Goat Milk				Cow Milk			
	^{131}I mBq L^{-1}	^{137}Cs mBq L^{-1}	^{134}Cs mBq L^{-1}	Ratio $^{134}Cs/^{137}Cs$	^{131}I mBq L^{-1}	^{137}Cs mBq L^{-1}	^{134}Cs mBq L^{-1}	Ratio $^{134}Cs/^{137}Cs$
9/04/11	246±107	481±52	< 33[a]	-	208± 97	333±44	< 31[a]	
1/05/11	101±68	506±48	< 26[a]		[b]68±67	421±44	< 31[a]	
8/05/11	87±72	448±47	< 26[a]		< 40[a]	263±39	< 26[a]	
16/05/11	< 24[a]	526 ±50	< 30[a]		< 38[a]	302±47	67±35	0.22
21/05/11	[b]77±73	527±63	[b]59±44	0.11	< 53[a]	684±54	< 28[a]	
29/05/11	60 ±46	474±47	69 ±26	0.15	110±58	473±44	< 27[a]	
05/06/11	< 25[a]	398±44	< 33[a]		< 34	354±41	< 27[a]	
11/06/11	< 68[a]	378±55	< 34[a]		77±68	279±37	41±24	0.15
20/06/11	< 32[a]	298±37	< 22[a]		< 28[a]	197 ±35	< 22[a]	
26/06/11	< 29[a]	460±45	< 25[a]		81±60	283±64	< 34[a]	
03/07/11	< 28	796±67	[b]48±30		< 32 [A)]	296±36	< 0.23	

a. MDA
b. Critical level

I-131 1200±350 mBq L^{-1}

Cs-137 150±30 mBq L^{-1}

Dose assessment

The limit of the effective dose for the population is fixed for the Italian Low of Radioprotection in 1 mSv y^{-1}
(Italian Government Legislative Decree, 1995).

The evaluation of the effective dose is done by the relation:

where, E_{est} is the effective dose for exposure;
$Jj,_{ing}$ and $Jj,_{inh}$ are the intake activity (Bq) by ingestion and by inhalation of radionuclide *j*, respectively;
$h(g)_{j,\ ing,}$ $h(g)_{j,\ inh}$ (Sv Bq^{-1}) are the coefficients of committed dose for unit of intake by ingestion and/or by inhalation for the population of age group *g*, due to radionuclide *j*.

Dose assessment

Coefficients of committed dose for unit of intake by ingestion and/or by inhalation for the population of age group *g*, for the radionuclides of interest, per unit of intake – Sv Bq^{-1}

Nuclide	age < 1 a $h(g)_{ing}$	age > 17 a $h(g)_{ing}$	*age < 1 a*$^{(*)}$ $h(g)_{inh}$	age > 17 a$^{(*)}$ $h(g)_{inh}$
I-131	$1{,}8\ 10^{-7}$	$2{,}2\ 10^{-8}$	$7{,}2\ 10^{-8}$	$7{,}4\ 10^{-9}$
Cs-137	$2{,}1\ 10^{-8}$	$1{,}3\ 10^{-8}$	$8{,}8\ 10^{-9}$	$4{,}6\ 10^{-9}$
Cs-134	$2{,}6\ 10^{-8}$	$1{,}9\ 10^{-8}$	$1{,}1\ 10^{-8}$	$6{,}6\ 10^{-9}$

(*) Fast Type of Absorption

Annual individual usage factors for external exposure, inhalation and consumption of foods. reported in NCRP-123 publication

Pathways - External *and* Inhalation	**Unit**	**Exposure**
Inhalation	$m^3\ a^{-1}$	8 000
Pathways - Ingestion	**Unit**	**Intake**
Water and beverages	$L\ a^{-1}$	800
Milk	$L\ a^{-1}$	300

Dose assessment

The evaluation of the Effective Dose was done using the highest concentration value for ^{131}I, ^{134}Cs and ^{137}Cs measured (Tables 1, 2, 7), and are taken into account only the h(g) coefficients for population of age less than 1 year old and greater than 17 a.

Effective doses due to different pathways

Pathways	***age < 1 a***	**age > 17 a**
Air	[illegible]	[illegible]
Water	[illegible]	[illegible]
Goat Milk	[illegible]	[illegible]
Cow Milk	[illegible]	[illegible]

< 1mSv y^{-1}

CONCLUSIONS

- The Fukushima plume was detected all over Europe
- The presence of more than one peaks of ^{131}I and $^{137,134}Cs$ is an index that air masses continuously transferred from Fukushima, Japan till the end of April, 2011.
- HYSPLIT backward trajectories interpreted the measured atmospheric concentrations
- The relative high concentrations of ^{137}Cs in grass, soil and fresh milk samples, correspond to Chernobyl fallout
- ^{131}I and $^{137,134}Cs$ isotopes were found above their detection limits in all environmental samples but very far below levels of concern

Publications on Fukushima accident

The 1st publication in *Journal of Environmental Radioactivity* about Fukushima accident was from our Nuclear Physics laboratory.

A publication as collaboration of all Greek laboratories with GAEC published at *Radiation Measurements* Journal.

A combined publication between Milano and Thessaloniki lab there is in *JRNC*.

A combined publication between Milano and Thessaloniki lab with air mass trajectories analysis published at *Atmospheric Environm.* journal.

ENVIRONMENTAL Science & Technology

ARTICLE

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Tracking of Airborne Radionuclides from the Damaged Fukushima Dai-Ichi Nuclear Reactors by European Networks

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Received: May 19, 2011
Accepted: August 2, 2011
Revised: July 21, 2011