Atmospheric Dispersion of Radionuclides from Fukushima Dai-Ichi Nuclear Accident and Dose Assessment

FINNISH METEOROLOGICAL INSTITUTE

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FACOLTÀ DI SCIENZE MATEMATICHE,
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Fukushima Nuclear Accident

11 March 2011 earthquake 9.0 and tsunami
- >500 square kilometers flooded
- 20,000 lives lost

Worst civil nuclear accident since Chernobyl in 1986
- Reactor cores severely damaged
- Large radioactive releases
  (about 10-20% of Chernobyl)

Dispersion of Radionuclides

12 March 2011
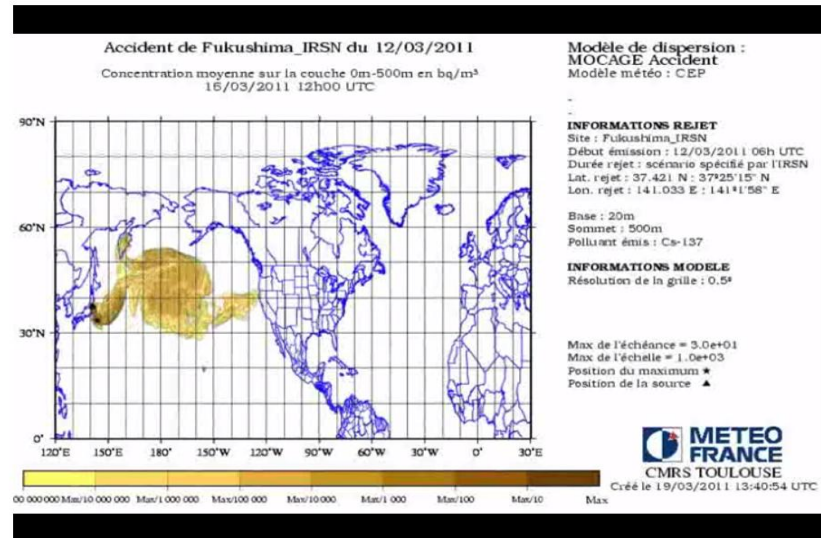

after 12 March 2011

Foreward trajectories modelling,
IRSN, FLEXPART, NOAA Hysplit model
Projection for the plume

Speed at which
risk and crisis information
flows through the Media

20 years ago:	24 hours
10 years ago:	4 hours
Today (2015):	4 minutes

Dispersion of Radionuclides - Projections
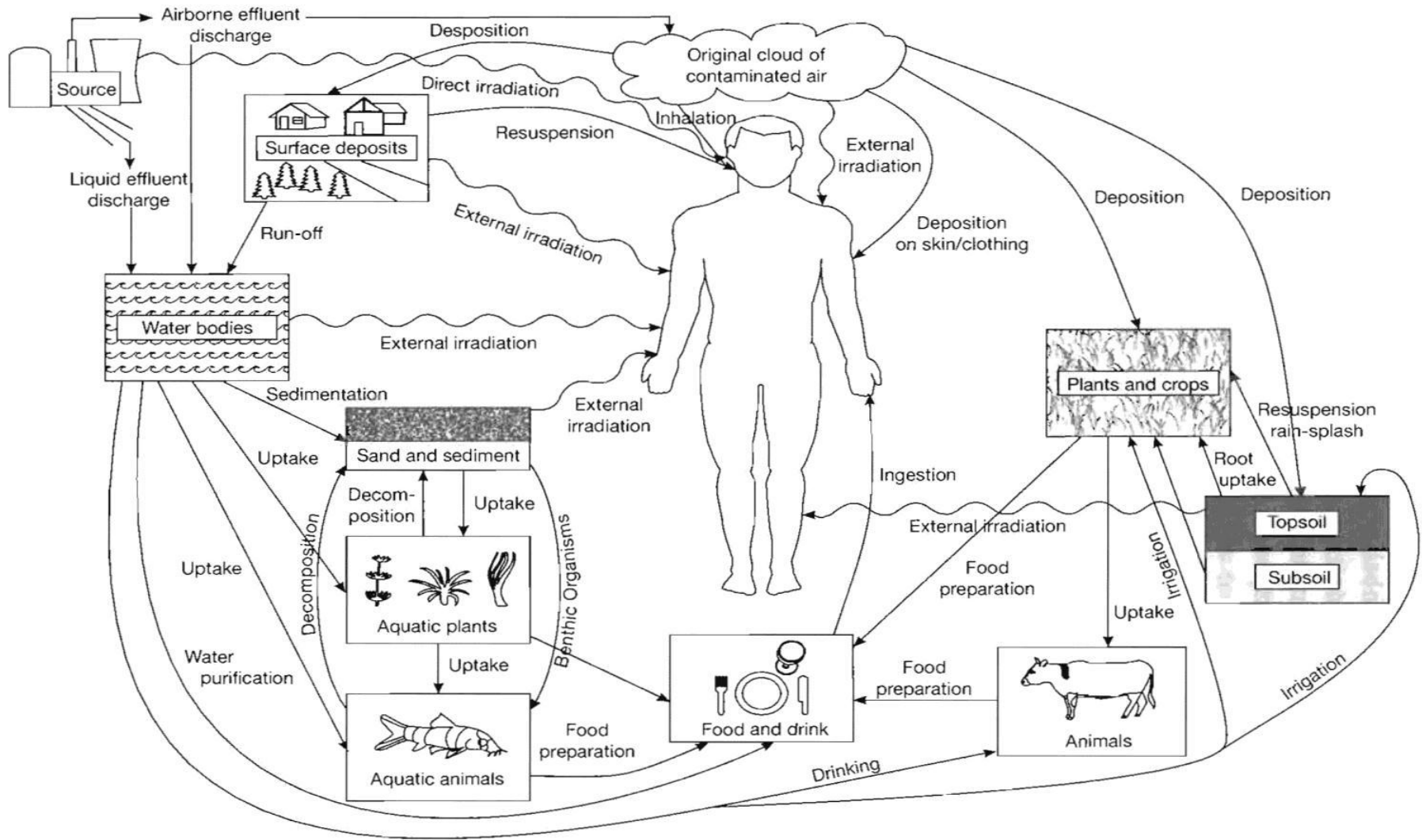

The plume reached the east coast of **North America after 3 days**

For information regarding the monitoring of radioactive material reaching the United States, please visit the EPA:http://epa.gov/japan2011
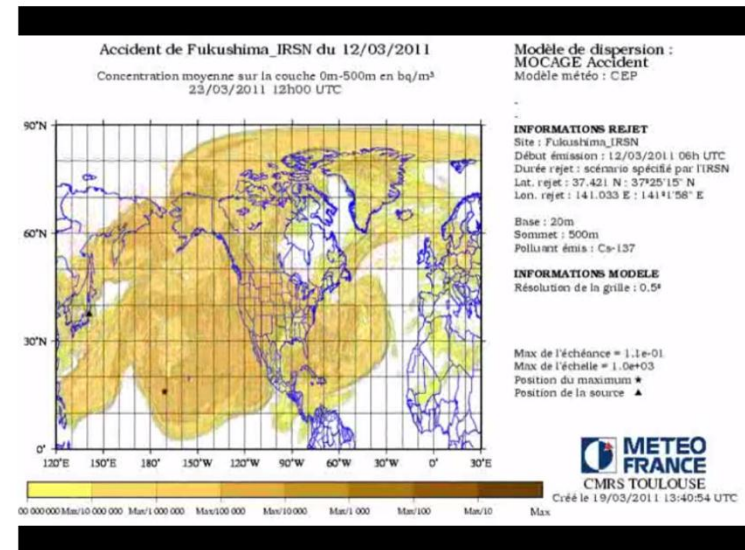
Expecting the plume in Central Europe **at around March 24** and this is exactly what happened

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

HV air Sampler (Staplex TFIA-2)

Flow rate: 1.6-1.7 m³/min (60cfm)
Sampling duration: 23 h
Total Volume: 2400-2700 m³
Air Volume Uncertainty (2σ): 30-50 m³
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.
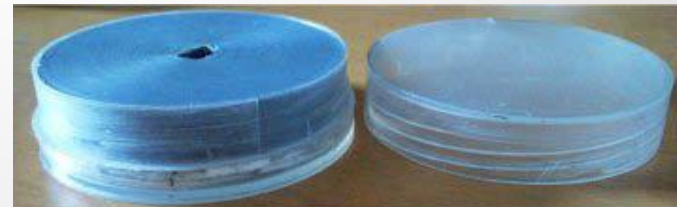

"8x10"

Air-filters analysis by gamma ray spectra

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

^{131}I- (364 keV)
^{137}Cs-137(662 keV)
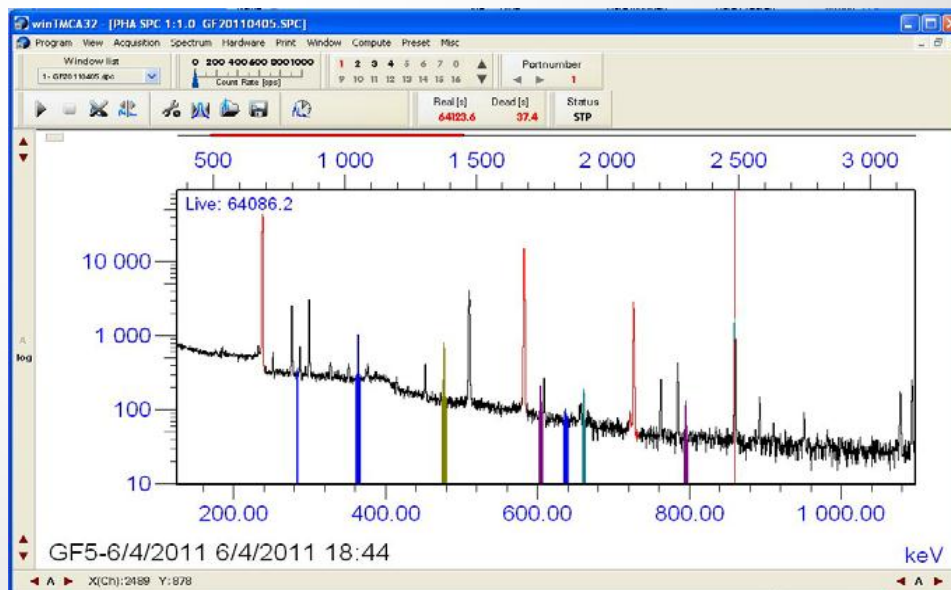^{134}Cs (605 keV & 796 keV) correction due to summation effect

Air-filters analysis by gamma ray spectra

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

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

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

Date of sampling	Fallout isotopes in surface air				
	^{131}I $\mu Bq\ m^{-3}$	^{137}Cs $\mu Bq\ m^{-3}$	^{134}Cs $\mu Bq\ m^{-3}$	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

^{131}I <467 μBq m^{-3}

^{137}Cs <63 μBq m^{-3}

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

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

$^{131}I < 500\ \mu Bq\ m^{-3}$

The maximum ^{131}I activity concentration observed in Milan, Italy was almost similar with the highest value (497 μBq m^{-3}) observed in Thessaloniki, Greece, and the highest observed value (490 μBq m^{-3}) in Athens Greece
but was lower than the one observed (810 $\mu Bq\ m^{-3}$) in Svalbard [Paatero et al., 2012] and in Lithuania (3700 $\mu Bq\ m^{-3}$, [Lujaniene et al., 2012]).

$^{137}Cs < 63\ \mu Bq\ m^{-3}$

The max ^{137}Cs activity concentration at Milan, Italy was 63 $\mu Bq\ m^{-3}$, while in Thessaloniki, Greece was 145 $\mu Bq\ m^{-3}$ and in Svalbard as high as 675 $\mu Bq\ m^{-3}$.

In most sampling sites is measured only the fraction of ^{131}I bound to aerosol particles.

The ratio value of of gaseous to total ^{131}I equal to 0.79, according to measurements in Athens (Potiriadis et al., 2011).

Masson et al. (2011) reported that measurements taken near Fukushima showed that the average ratio of gaseous to total ^{131}I was around 0.71, while European measurements showed an average gaseous to total ^{131}I ratio of 0.77.

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

Fukushima fallout isotopes at Thessaloniki
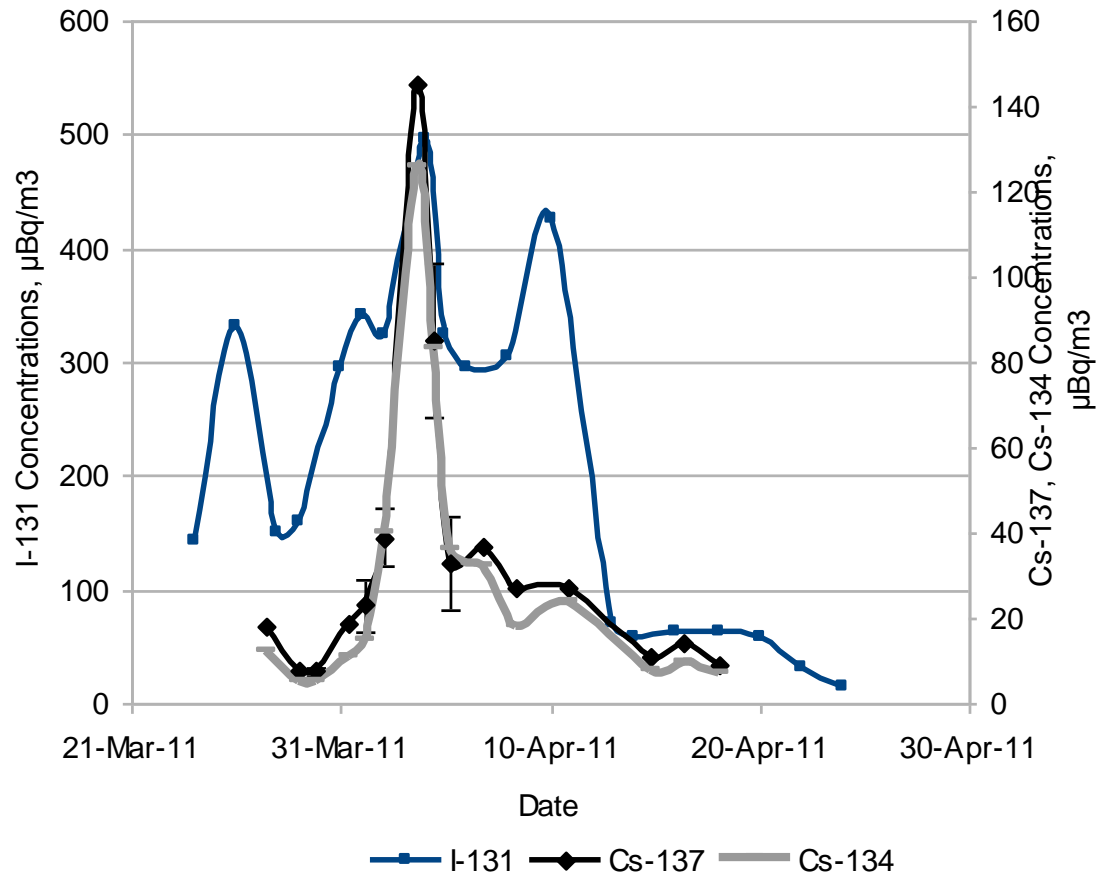

$^{134}Cs/^{137}Cs =1$

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

According to Kirchner et al. (2012) the isotopic compositions observed in Europe correspond to emissions from units 1 and 3 of the destroyed Fukushima reactor.

For comparison after the Chernobyl accident the ratio was about 0.5

$^{131}I/^{137}Cs$ activity ratio

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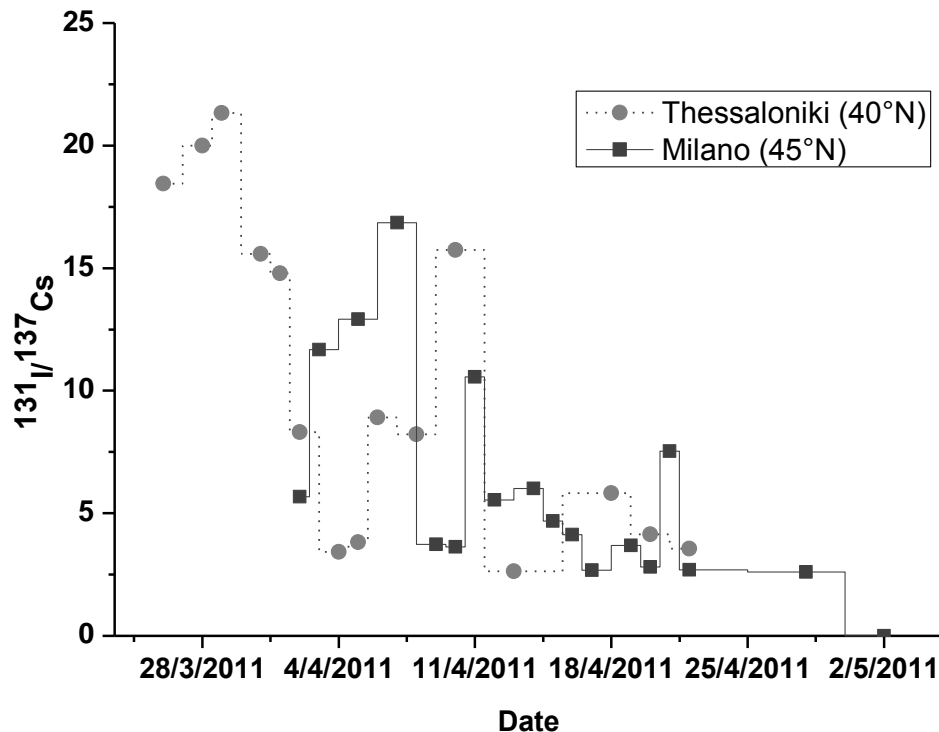
### $^{131}I/^{137}Cs$ decreases with time

The high $^{131}I/^{137}Cs$ ratio, observed during the first days after the accident, followed by lower values later on, reflects not only the initial release ratio but also differences in volatility, attachment and removal of two isotopes during transportation due to their different physico-chemical properties.

The presence of more than one peak of ^{131}I and ^{137}Cs could indicate "fresh" air masses that arrive from Fukushima in both regions up to the end of April/first days of May.

^{131}I atmospheric concentrations
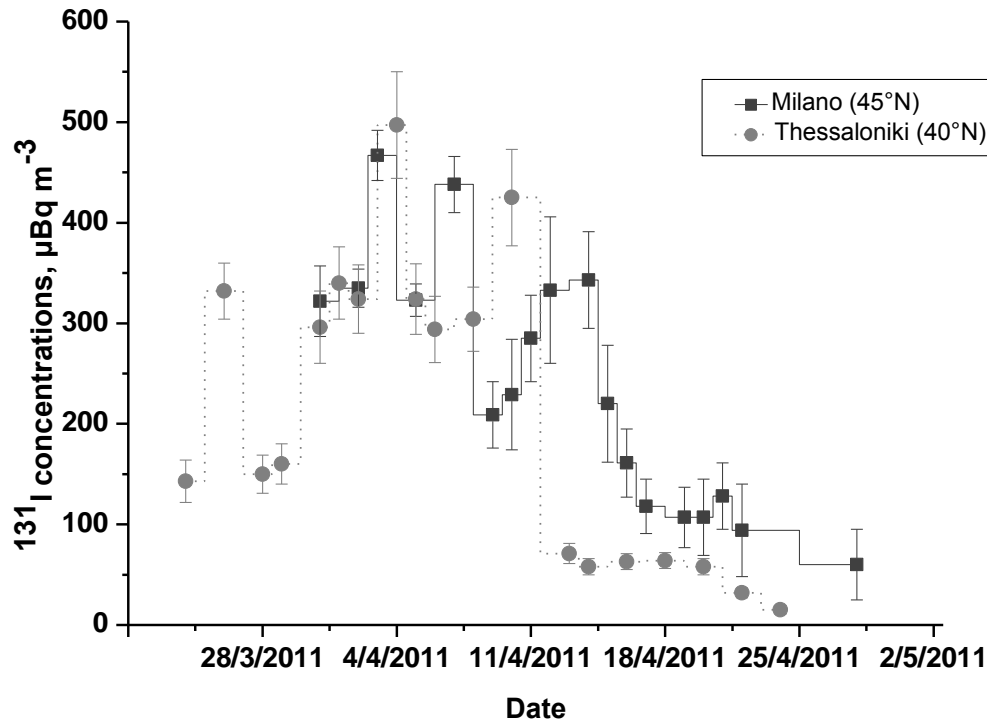
^{131}I maximum 3-4 April
~500 µBq m^{-3}

Similar maximum values have been observed during 3-4 April, 2011 in both regions, despite the considerable distance and the possible differences of climatic parameters that may influence the air concentrations of nuclides.

But in general, there is a shift trend in the peak days.
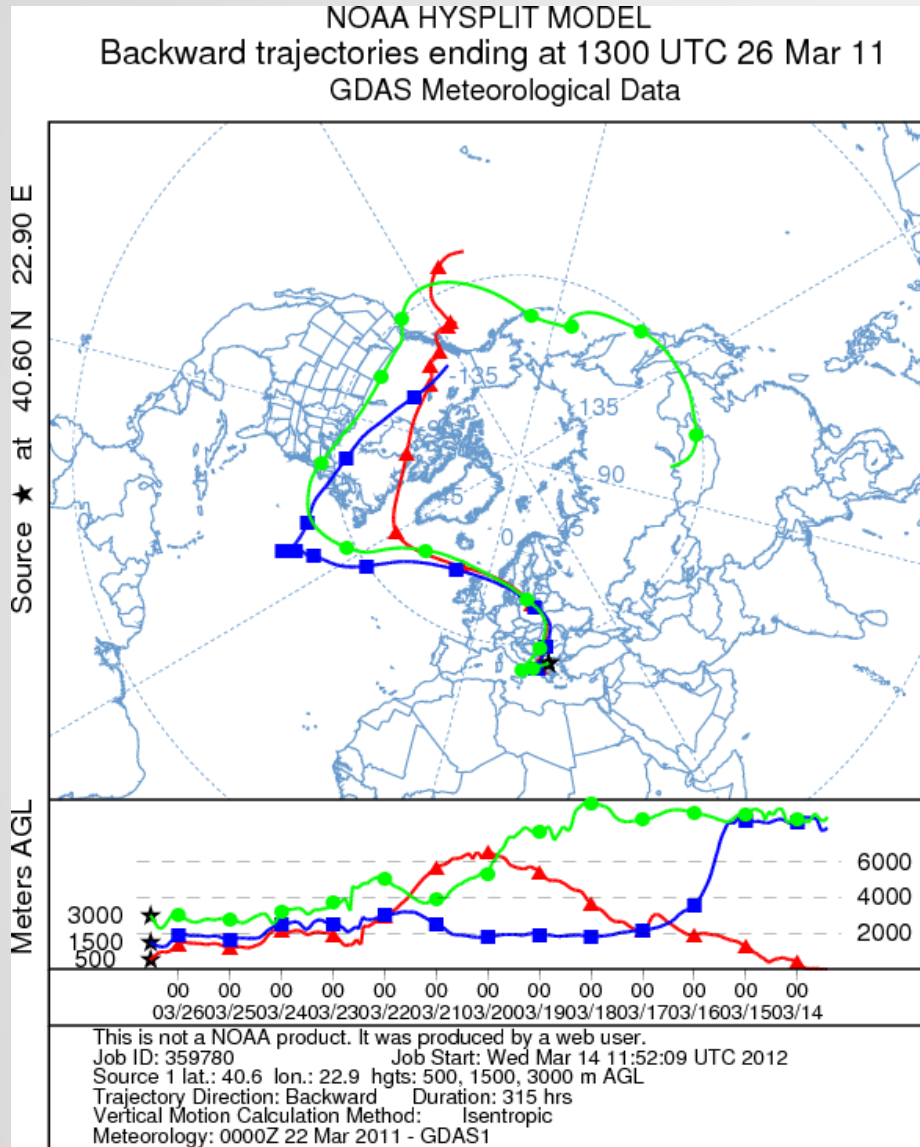
Back trajectories analysis
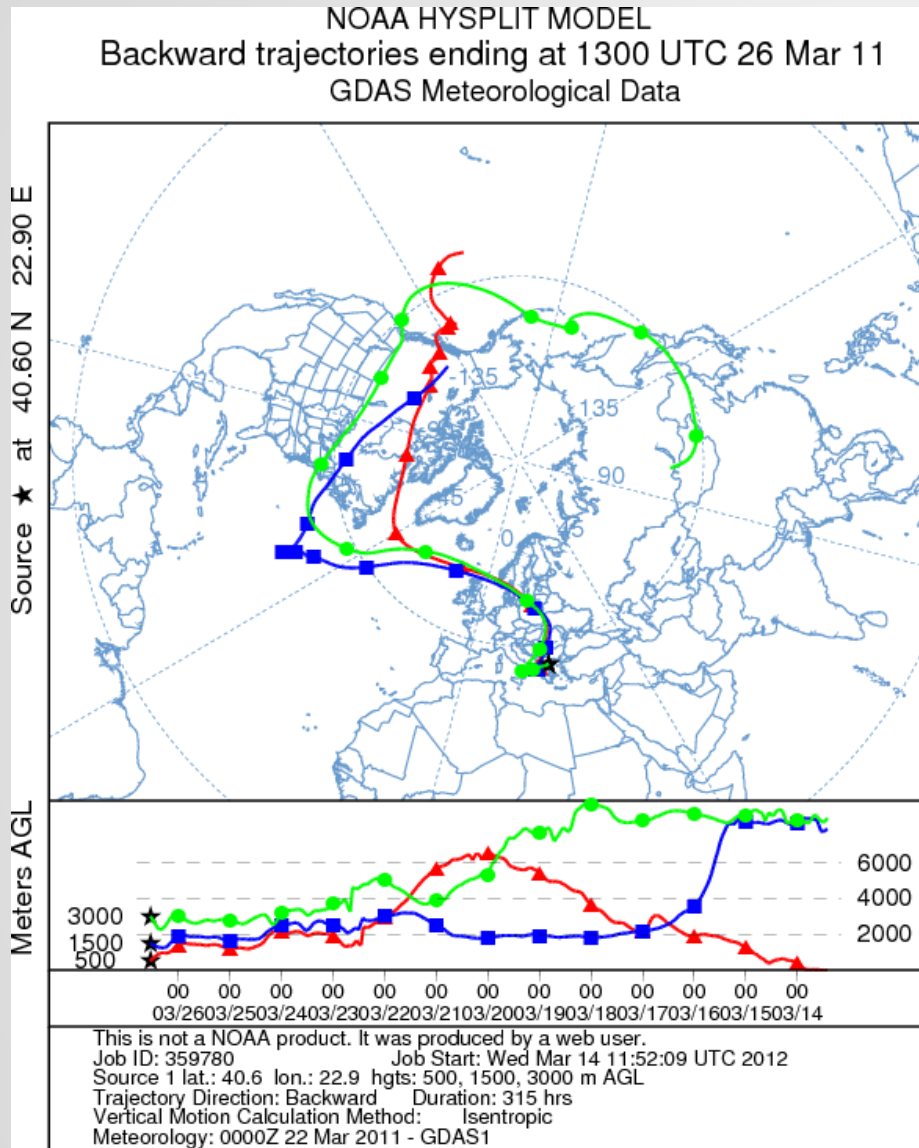
The NOAA HYSPLIT model was used to assess the transport pattern and to explain the deviation in radionuclide activity concentrations found.

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.
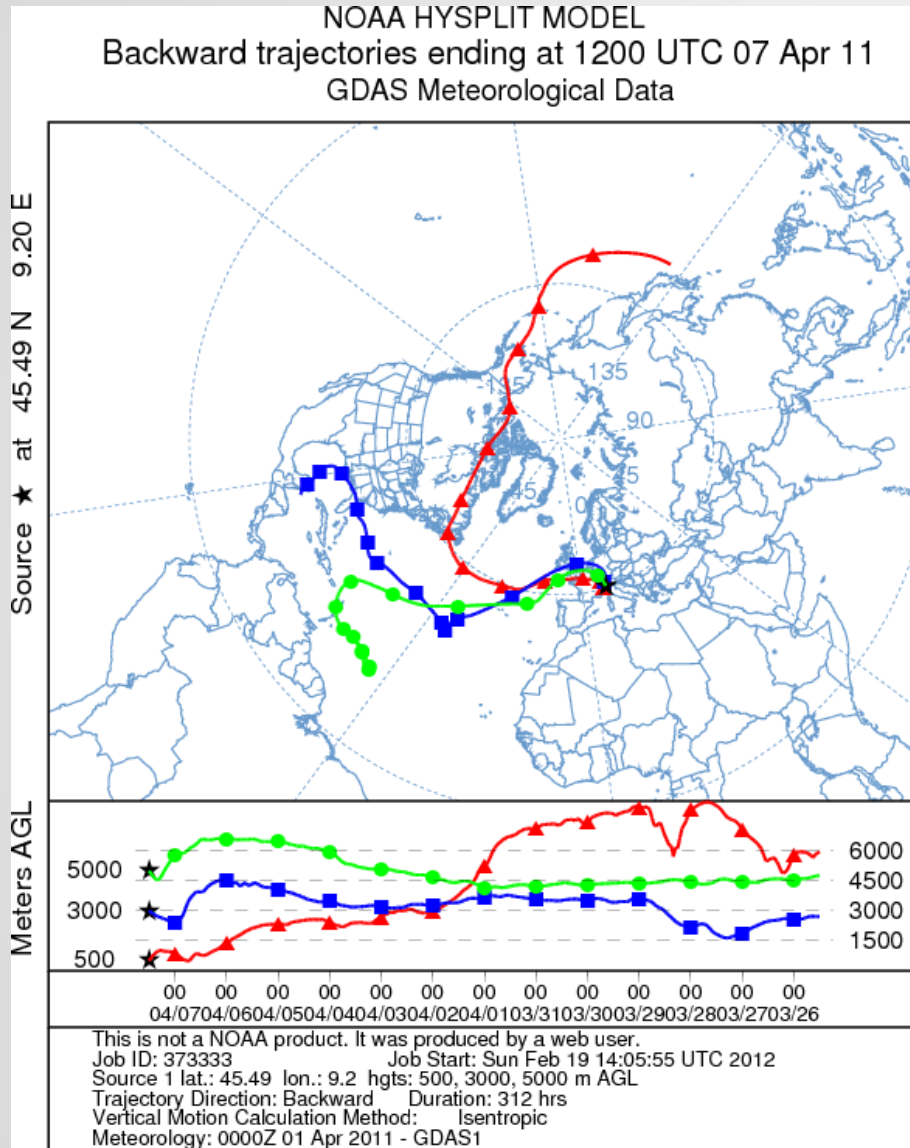
Back trajectories analysis

NOAA HYSPLIT MODEL
Backward trajectories ending at 1300 UTC 26 Mar 11
GDAS Meteorological Data

This is not a NOAA product. It was produced by a web user.
Job ID: 359780   Job Start: Wed Mar 14 11:52:09 UTC 2012
Source 1 lat.: 40.6 lon.: 22.9 hgts: 500, 1500, 3000 m AGL
Trajectory Direction: Backward   Duration: 315 hrs
Vertical Motion Calculation Method: Isentropic
Meteorology: 0000Z 22 Mar 2011 - GDAS1

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.
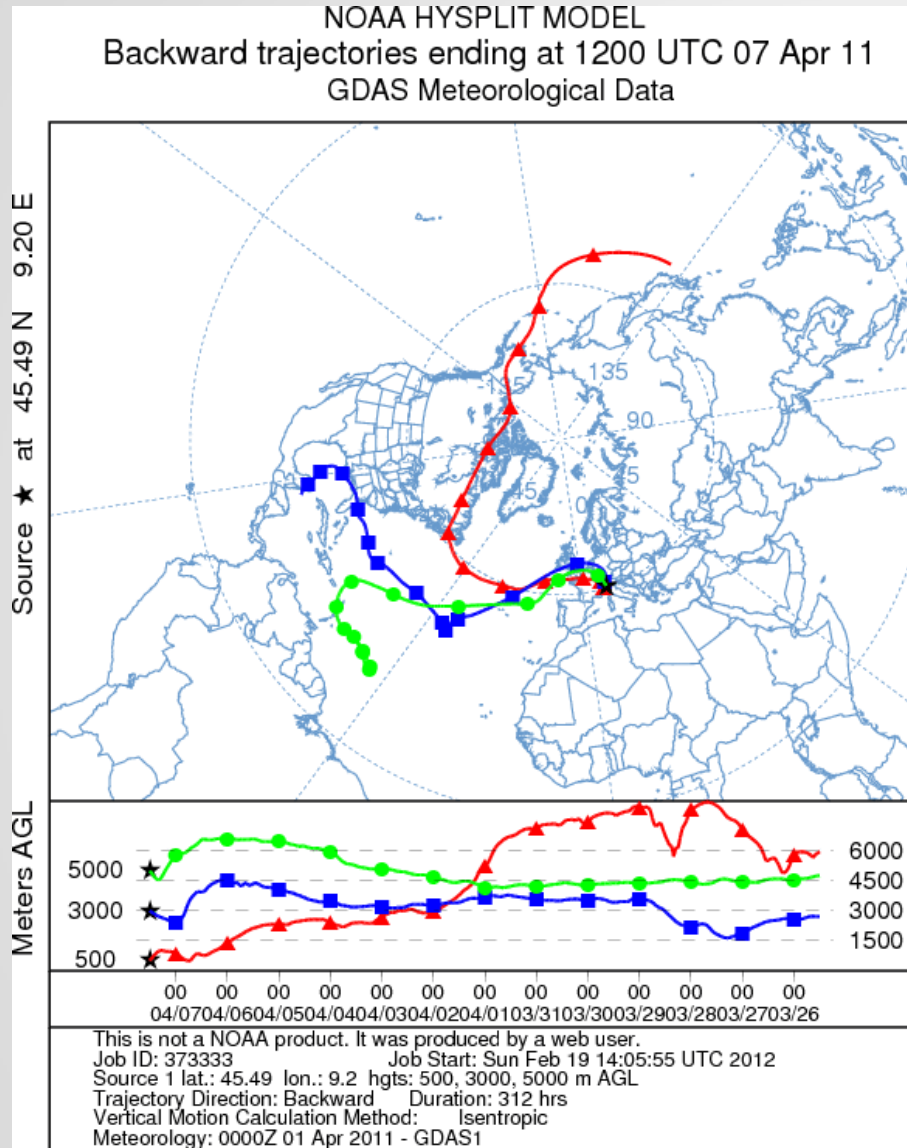
Back trajectories analysis

NOAA HYSPLIT MODEL
Backward trajectories ending at 1200 UTC 07 Apr 11
GDAS Meteorological Data

This is not a NOAA product. It was produced by a web user.
Job ID: 373333   Job Start: Sun Feb 19 14:05:55 UTC 2012
Source 1 lat.: 45.49 lon.: 9.2 hgts: 500, 3000, 5000 m AGL
Trajectory Direction: Backward   Duration: 312 hrs
Vertical Motion Calculation Method: Isentropic
Meteorology: 0000Z 01 Apr 2011 - GDAS1

An example of transported air mass at **07 of April 2011** at Milan, Italy 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

NOAA HYSPLIT MODEL
Backward trajectories ending at 1200 UTC 07 Apr 11
GDAS Meteorological Data

Although the second maximum of concentration of ^{131}I that observed at Milan on 7th 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
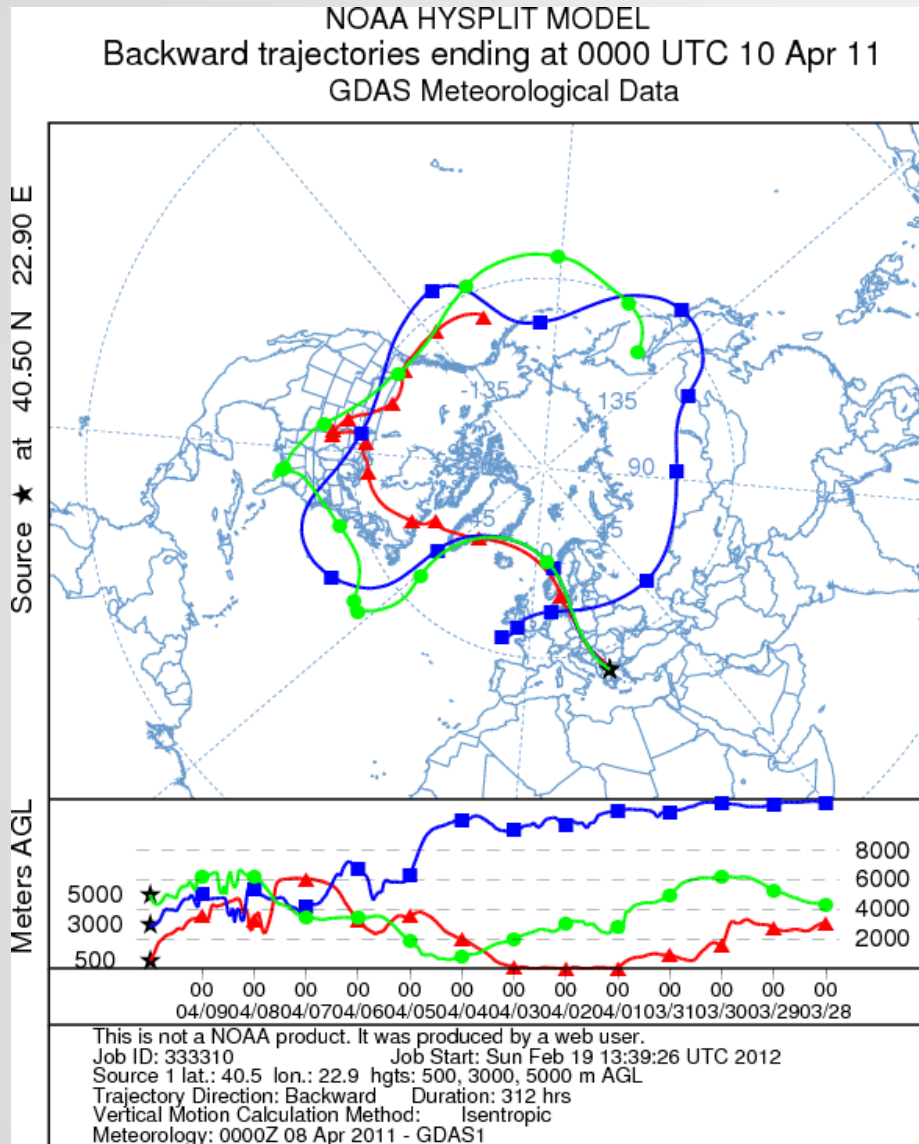
On **10th of April** back-trajectory analysis showed a direct transfer from Fukushima across the Pacific Ocean, a transport through the North Pole and a pathway through the Greenland and Iceland to Thessaloniki.

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.

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

<div align="center">Fallout isotopes in rainwater samples</div>

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 < 1Bq 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).

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

Samples collected at 3000 m s.l.m.

Monte Moro - Macugnaga, Italy

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

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]	-

Total surface: 1 m^2

Total mass: 0,35-0.45 kg

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

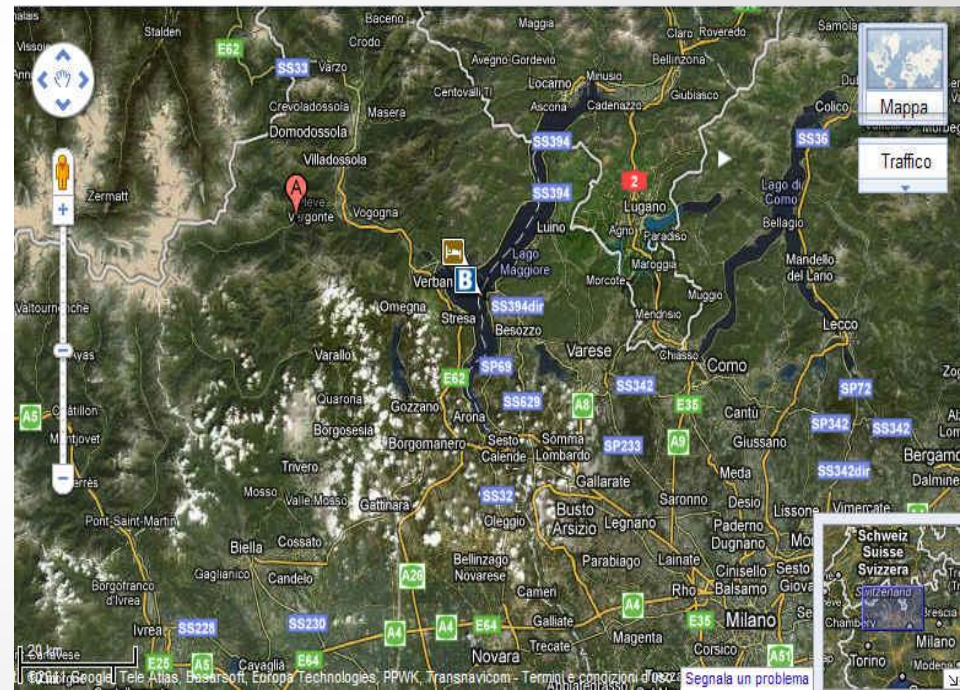| Site	Date of Sampling	Fallout isotopes in soil samples			
		^{131}I Bq kg^{-1}	^{137}Cs Bq kg^{-1}	^{134}Cs Bq kg^{-1}	ratio ^{137}Cs/^{134}Cs
Segrate	30/03/11	0.63±0.29	12.26±0.70	0.83±0.30	0.07
Senago uncovered	04/04/11	0.57±0.25	85.17±4.40	0.29±0.13	0.0034
Senago covered	04/04/11	<0.15[a]	84.65±4.40	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

^{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

	Goat Milk				Cow Milk			
Date of Sampling	^{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	

Fallout isotopes in milk samples

a. MDA
b. Critical level

Greece:

^{131}I   1200±350 mBq L^{-1}

^{137}Cs   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:

$$E = E_{est} + \sum_j h(g)_{j,ing} J_{j,ing} + \sum_j h(g)_{j,inh} J_{j,inh} < 1 \, mSv \, a^{-1}$$

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}

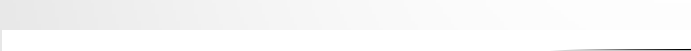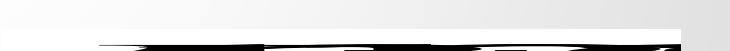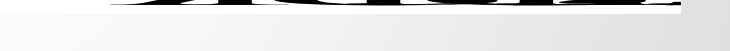| Nuclide	age < 1 a $h(g)_{ing}$	age > 17 a $h(g)_{ing}$	age < 1 a$^{(*)}$ $h(g)_{inh}$	age > 17 a$^{(*)}$ $h(g)_{inh}$
I-131	$1{,}8\ 10^{-7}$	$2{,}2\ 10^{-8}$	$7{,}2\ 10^{-8}$	$7{,}4\ 10^{-9}$
Cs-137	$2{,}1\ 10^{-8}$	$1{,}3\ 10^{-8}$	$8{,}8\ 10^{-9}$	$4{,}6\ 10^{-9}$
Cs-134	$2{,}6\ 10^{-8}$	$1{,}9\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

$^{(*)}$ 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.
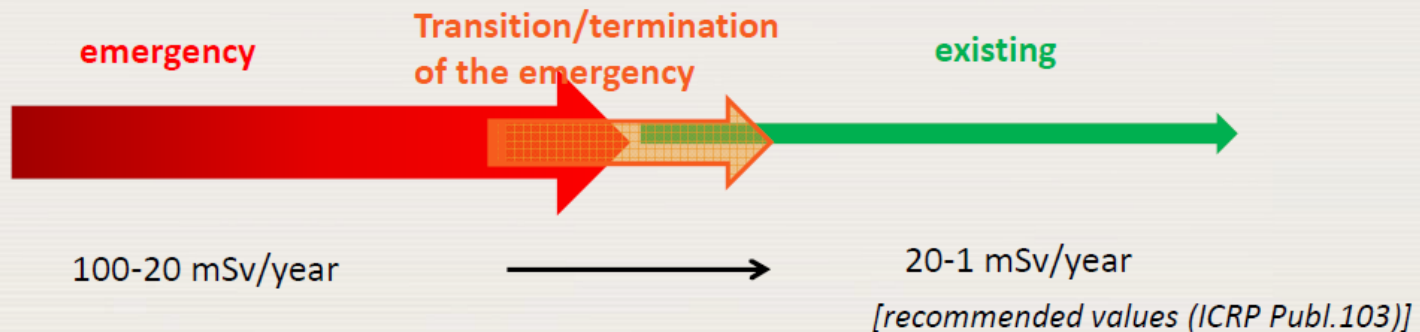
Effective doses due to different pathways

Pathways	age < 1 a	age > 17 a
Air		
Water		
Goat Milk		
Cow Milk		

< 1mSv y^{-1}

Fukushima Nuclear Accident

Point 5: Transition from Emergency to Existing Exposure Situation

By whom and **at which values** are reference levels set?
(an example for situation after accident)

emergency

Transition/termination of the emergency

existing

100-20 mSv/year → 20-1 mSv/year
[recommended values (ICRP Publ.103)]

Government, regulatory body or another relevant authority

↓

selects an **appropriate values** to <u>secure necessary activities be undertaken</u>
or
(**adopts** necessary **amendments** to existing laws (regulations) to <u>secure it</u>)

IAEA

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Fukushima Nuclear Accident

	Nuclear Weapons Tests (atm+ocean)	Reprocessing (ocean)	Chernobyl (atm+ocean)	Fukushima (atm+ocean)
3H	186 000	1000	10	?
14C	213	10	?	?
^{131}I			1760	(130-160)+5
^{137}Cs	950+600	50	85+16	(13-15)+(3-6) direct
				5-10 deposition
^{90}Sr	620+380	7	~20	0.14+(1-2)
^{239,249}Pu	10	1	~1	?

Povinec, Hirose, Aoyama: Fukushima Accident: Environmental Impact, Elsevier, 2013, 400p

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CONCLUSIONS

➢ The Fukushima plume was detected in Milan, Italy and Thessaloniki, Greece

➢ 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