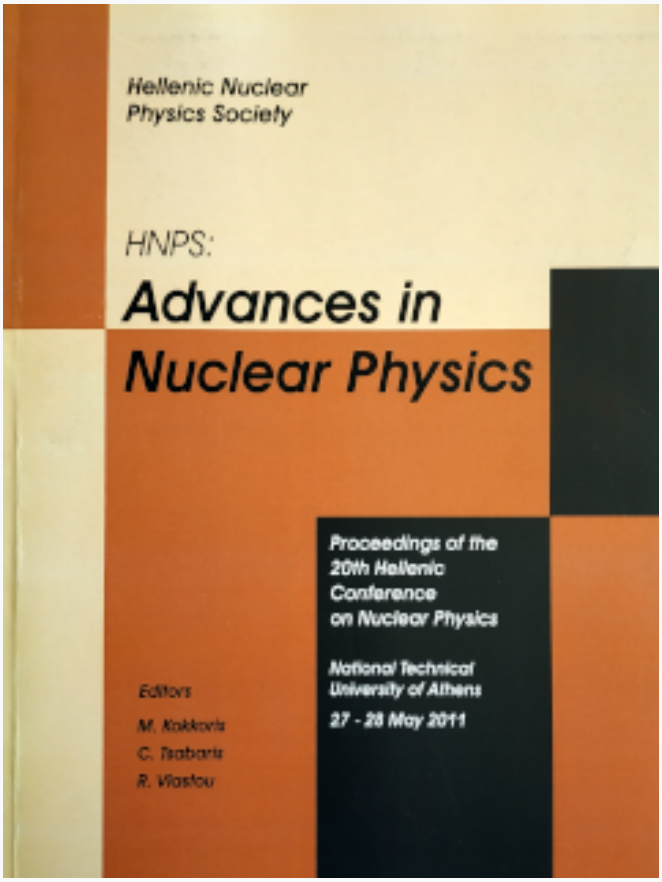
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^{131}I, ^{137}Cs, ^{134}Cs from Fukushima fallout at Milano, Italy

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Abstract

Right after the Fukushima reactor accident, a systematic air and wet sampling and analysis programme was undertaken to detect and quantitatively analyze the radionuclides in the Fukushima fallout in Milano, Italy. Radionuclides from Fukushima were first detected at Milano region in a rain water sample, collected during March 27-28, 2011 with the concentrations of ^{131}I and ^{137}Cs isotopes in the rainwater to be equal with 0.89 Bq L^{-1} and 0.12 Bq L^{-1}, respectively. The concentration of ^{131}I in goat and cow milk samples from a farm in Monte Rosa mountain, were 0.25 and 0.21 Bq L^{-1}, respectively. Increased atmospheric radioactivity was detected on air filter taken on 30 March 2011, while the maximum activity of 467 μBq m^{-3} occurred at April 3-4, 2011. A week later the activities had fallen to about 50% of peak values, with a general increasing trend over the following days. In the time period of one month after the nuclear accident, concentrations of ^{137}Cs and ^{134}Cs in air as high as 63 μBq m^{-3} and 61 μBq m^{-3}, respectively were recorded. The presence of more than one peaks of ^{131}I and $^{137,134}Cs$ till April 11, 2011, indicates that ^{131}I and $^{137,134}Cs$ were continuously transferred from Fukushima, Japan to Italy. According to the measurements, airborne activity levels remain of no concern for public health in Italy.

Keyword : nuclear accident; radioactive fallout; radiaoctive nuclides

1. Introduction

The Fukushima nuclear reactor accident which occurred on March 11, 2011 is known to have injected into the atmosphere a pulse of ^{131}I and ^{137}Cs and ^{134}Cs radionuclides. Iodine-131 and caesium isotopes were transported across the Pacific towards the North American continent and Europe despite dispersion and washout along the trip of the contaminated air mass. The activities of these radionuclides present in the subsequent fallout have been measured in different locations throughout Europe.

Starting from this day many Laboratories all over the Europe start to collect samples and obtain results in order to evaluate the contribution of the Fukushima release into different environmental matrices [1-3]. Also in Italy the National Institute for the Environmental Protection and Research [4] and the INFN and University Laboratory of Bicocca [5] and LASA Laboratory of INFN Sez. of Milano and UNIMI were involved. At LASA radionuclides from Fukushima were first detected in a rain water sample collected during the March 27-28, 2011. On March 26, 2011, the highest values of fission product radionuclides

were observed in the prefecture of Yamagata, Japan as high as 7500 Bq m^{-2} for ^{131}I and 1200 Bq m^{-2} for ^{137}Cs (IAEA, 2011)[6].

Our focus was mainly to the detection of the radioactive nuclides of ^{131}I and ^{137}Cs and ^{134}Cs. The thyroid slurps up iodine, including the radioactive kind, which immediately kills and mutates cells and can cause thyroid cancer. Its short half-life means that very little, if any, could make it across the Pacific. Radioactive caesium easily enters the food chain through milk and vegetables. If inhaled or ingested, it acts like potassium but continues to emit radiation inside the body, and can increase the risk of any form of cancer. So, our measurements were focuses on defining the concentrations of the above radionuclides in air and water, which control the main way of deposition of the radionuclides in the soil surfaces and vegetables.

2. Results and discussion

2.1 ^{131}I, ^{137}Cs and ^{134}Cs concentrations in surface air

The transferred radioactivity by the cloud formed in air over the Fukushima nuclear power plant, caused radioactive contamination to the air over Europe very early after the nuclear accident occurred.

At LASA air sampling was carried out daily, with a High Volume air sampler of Staplex. Aerosol samples were collected for 23 hours, with filters changed daily at 15:00 local time. The flow rate was about 60 cfm resulting in a total daily air volume throughput on average 2200 m^3. The collection substrate was Glass Fiber filters with a collection efficiency of 99.8% for particles as small as 0.3 μm in diameter.

All samples were counted and analyzed using HPGe-detector setups for fission products from the Fukushima fallout. The samples were measured for ^{131}I through 0.364 MeV gamma-ray, for ^{137}Cs through 0.662 MeV gamma-ray and for ^{134}Cs through 0.605 MeV gamma-ray photons. The results of air fallout radioactivity concentration from March 30 through April 12, 2011 are given in Table 1. and in Figs. 1 - 2. The results are reported at the mean time of the sampling interval.

Table 1. Activity concentrations, following the Fukushima accident, of ^{131}I, ^{137}Cs and ^{134}Cs in air at Milano, Italy

Date of Collection	^{131}I μBq m^{-3}	Error ^{131}I μBq m^{-3}	^{137}Cs μBq m^{-3}	Error ^{137}Cs μBq m^{-3}	^{134}Cs μBq m^{-3}	Error ^{134}Cs μBq m^{-3}	ratio ^{134}Cs /^{137}Cs
31/03/2011	322.49	35.39	< 29.35		< 26.37	-	-
02/04/2011	334.58	88.64	58.90	41.59	56.07	37.01	0.95
03/04/2011	467.11	24.72	39.80	8.78	36.72	7.71	0.92
05/04/2011	323.30	16.43	24.83	9.09	27.12	8.50	1.09
07/04/2011	437.89	27.88	25.73	17.42	25.23	15.25	0.98
09/09/2011	209.30	33.23	55.85	30.33	54.41	24.01	0.97
10/04/2011	229.20	54.96	63.03	29.65	61.04	21.88	0.97
11/04/2011	285.03	43.31	26.94	17.89	23.13	14.19	0.90
12/04/2011	333.16	72.53	60.06	38.32	56.16	29.74	0.94

As also evident from Fig. 1, ^{131}I reached 437 μBq m^{-3} on April 6-7, 2011. The presence of more than one peaks of ^{131}I in Fig. 1 shows that ^{131}I was continuously transferred from Fukushima, Japan.

According to the source term declared by the NISA Japanese Agency [7], around 150 PBq (1 PBq = 10^{15} Bq) of ^{131}I and 6 to 12 PBq of ^{137}Cs were release to the atmosphere. A rough estimation of the total ^{131}I inventory that have passed over Italy during this period is < 1% of

the released amount. The airborne activity levels remains of no concern for public health in Italy.

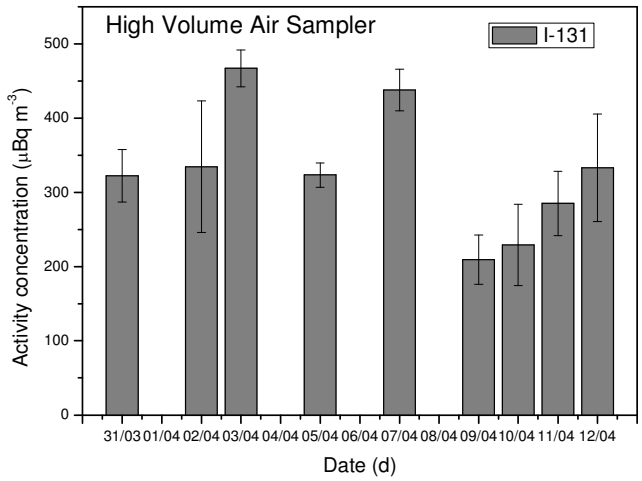


Fig. 1. Activity concentrations of ^{131}I in air at the period 30 March – 12 April, 2011

The ^{137}Cs/ ^{134} Cs ratio values were remained almost stable at about 1, throughout the time period of measurements. This value is in agree with the measurements of KEK High Energy Accelerator Research Organization at Tsukuba, Japan, for which the ratio ^{137}Cs/^{134}Cs was equal to 0.97, almost 1.0, at the period 15 March, 2011 14:39 – 15 March, 2011 17:34 hour local time [8].

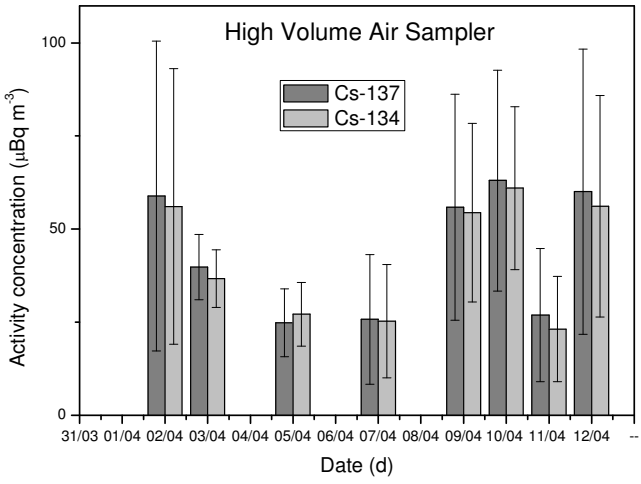


Fig. 2. Activity concentrations of ^{137}Cs and ^{134}Cs in air in the period 30 March– 12 April, 2011

2.2 ^{131}I, ^{137}Cs and ^{134}Cs concentrations in rainfall, snowfall and dry deposition samples

On March 27 and 28 a rainfall occurred, with a height of precipitation of 0.8 mm (Milano Meteorological Station report). Two samples from two different regions of the city of Milano were examined. One was collected at LASA laboratory in Segrate, at East of Milano, while the second one at Senago, a small town in the North region of the city.

The samples were measured in 1 L Marinelli geometry by HPGe detector with 40% efficiency and high resolution. The results are given in Table 2. In Fig. 3 is reported the spectrum related to the LASA sample, where the ^{131}I peak is clearly evident. The levels seen in rainwater are not expected to present any threat to public health.

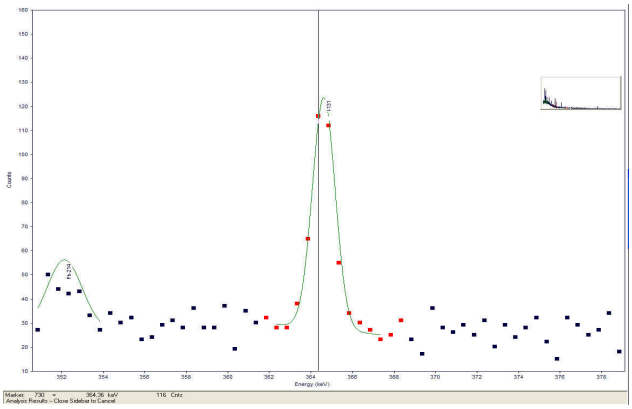


Fig. 3. gamma spectrum of rainwater sample collected at LASA Laboratory – Segrate (MI), Italy during the rainfall of 27 – 28 March 2011.

Table 2. Concentrations of ^{131}I, ^{137}Cs and ^{134}Cs in rainwater samples following the Fukushima accident at Milano, Italy

Site	Date of Sampling	^{131}I		^{137}Cs		^{134}Cs		
		activity mBq L^{-1}	error mBq L^{-1}	activity mBq L^{-1}	error mBq L^{-1}	activity mBq L^{-1}	error mBq L^{-1}	ratio $^{134}Cs/^{137}Cs$
		890.81	115.00	122.26	88.56	< 57.767	MDA	-
Senago	27-28/03/11	725.10	132.91	< 11.00	MDA	< 86.00	MDA	

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. All tap water in Northern Italy is completely safe in terms of its ^{131}I content. The rain water will be diluted many times before it becomes tap water, and the rain water itself is well below the 100 Bq L^{-1}, the limit for infants.

During the same days two snowfall samples were collected from Monte Rosa mountain at a height of 3000 m. The samples were treated exactly as the rainfall samples and the results are given in Table 3. It seems that the snow is more efficient in scavenging ^{137}Cs instead of ^{131}I.

Table 3. Concentrations of ^{131}I, ^{137}Cs and ^{134}Cs in snowfall samples following the Fukushima accident at Milano, Italy

Site	Data of Sampling	^{131}I		^{137}Cs		^{134}Cs		
		activity mBq L^{-1}	error mBq L^{-1}	activity mBq L^{-1}	error mBq L^{-1}	activity mBq L^{-1}	error mBq L^{-1}	ratio $^{134}Cs/^{137}Cs$
Monte Rosa	27-28/03/11	< 12.04	MDA	< 8.98	MDA	< 6.92	MDA	-
Monte Rosa	30-31/03/11	< 20.88	MDA	26.86	16.05	31.87*	13.09	1.19

*Critical Level

The dry deposition seems to be very important especially in case of cesium isotopes (^{137}Cs and ^{134}Cs). A sample of dry deposition that was collected 9 days after the first rainfall event of 27-28/03/2011 showed that the dry deposition of ^{137}Cs and ^{134}Cs was grater that that of ^{131}I (Table 4), if these data are compared with the ones obtained by rainwater. 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.

Table 4. Concentrations of ^{131}I, ^{137}Cs and ^{134}Cs in dry deposition following the Fukushima accident at Milano, Italy

Site	Data of Sampling	^{131}I		^{137}Cs		^{134}Cs		ratio $^{134}Cs/^{137}Cs$
		activity Bq m^{-2}	error Bq m^{-2}	activity Bq m^{-2}	error Bq m^{-2}	activity Bq m^{-2}	error Bq m^{-2}	
Segrate	06/04/2011	0.40	0.16	0.24	0.11	< 0.05	MDA	-

2.3 ^{131}I, ^{137}Cs and ^{134}Cs concentrations in milk samples

Iodine-131 was detected and measured in the goat and cow fresh milk produced and collected on April 9, 2011 at Macugnaga area (rural area), in Monte Rosa mountain. We defined almost similar ^{131}I concentration in both goat and cow milk samples (Table 5), of 0.25 and 0.21 Bq L^{-1}, respectively. The concentration of ^{137}Cs in both samples are relatively high.

Table 4. Concentrations of ^{131}I, ^{137}Cs and ^{134}Cs in goat and cow milk following the Fukushima accident at Milano, Italy

Milk type	Data of Sampling	^{131}I		^{137}Cs		^{134}Cs		ratio $^{134}Cs/^{137}Cs$
		activity mBq L^{-1}	error mBq L^{-1}	activity mBq L^{-1}	error mBq L^{-1}	activity mBq L^{-1}	error mBq L^{-1}	
Goat	09/04/2011	246	107	481	51	< 33	MDA	-
Cow	09/04/2011	208	97	333	44	< 31	MDA	-

Similar results in sheep milk, have been reported by ISPRA [4] and by the Greek Atomic Energy Commission [2]. For contrast, in Fukushima prefecture dairy farms, milk ^{131}I levels had been measured up to 1.510 Bq L^{-1} [9], while the maximum permissible level of radioactivity fixed by the European Commission to import food from Japan for ^{131}I in milk is 100 Bq L^{-1} for infant formulas

3. Conclusions

The transferred radioactivity, by the cloud formed in air over the Fukushima nuclear power plant, caused radioactive contamination to the air over Europe. In particular at LASA Laboratory of the INFN Sez. of Milano and UNIMI the maximum concentrations of ^{131}I of 467 μBq m^{-3}, were observed at April 3-4, 2011. The data measured by this Laboratory are comparable with the ones measured by the INFN Laboratory of Milano Bicocca and the ones reported by the Italian National Institute for the Environmental Protection and Research. The presence of more than one peaks of ^{131}I and $^{137,134}Cs$ indicates that ^{131}I and $^{137,134}Cs$ were continuously transferred from Fukushima, Japan to Italy till April 11, 2011. Radioisotopes of caesium and iodine were found above their detection limits, but still with no concern for public health.

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