Radionuclides from Fukushima accident in
Thessaloniki, Greece (40°N) and Milano, Italy (45°)

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ABSTRACT

^{131}I, ^{137}Cs and ^{134}Cs were observed in environmental samples in Milano (40°N), Italy and Thessaloniki (45°N), Greece, soon after the nuclear accident in Fukushima, Japan. The radionuclide concentrations were determined and studied as a function of time. In Thessaloniki the ^{131}I in air was observed for the first time on March 24, 2011. In Milano, the first evidence of Fukushima fallout has been confirmed with ^{131}I and ^{137}Cs measured in wet precipitation collected two days later. The maximum ^{131}I activity concentration in air of 467±25 µBq m^{-3}, observed in Milano on April 3-4, 2011, was almost similar to the highest value of 497±53 µBq m^{-3} observed in Thessaloniki. The ^{134}Cs/^{137}Cs activity ratio values in air were around 1 in both regions. Soil, grass and milk samples were contaminated with ^{131}I and ^{137}Cs at a low level. Finally, a dose assessment for these two areas showed clearly that the detected activities in all environmental samples were far below levels of concern.

Keywords: nuclear accident; Fukushima accident; radioactive nuclides; radioactive fallout

1. Introduction

As a consequence of the Fukushima accident, resulting from the big earthquake and Tsunami of March 2011, significant amounts of radionuclides were released to the terrestrial and marine environments [1]. Fukushima accident showed once again how fast airborne radionuclides can be transported over great distances: emissions of radionuclides were high enough to be detected all over the northern hemisphere, indicating not only local and regional but also global impact of the accident [2-13].

The most important radionuclides detected in Europe due to Fukushima accident were ^{137}Cs ($T_{1/2}$=30.6 a), ^{134}Cs ($T_{1/2}$= 2.06 a) and ^{131}I ($T_{1/2}$= 8.02 d). Under normal conditions, only ^{137}Cs remains in the atmosphere for long time, due to its relative long half life. In Europe, ^{137}Cs from the Chernobyl accident was still measurable before the Fukushima accident at trace levels viz. below 1 µBq m^{-3} [14, 15], while ^{134}Cs due to Chernobyl accident had not been measured in the atmosphere since the middle of the 1990s.

Immediately after the Fukushima accident, monitoring activities were promptly organized worldwide. The data discussed in the present work includes the observations of Fukushima related radionuclides in Milano (45.49° N, 9.20° E), Italy by LASA (Laboratorio Acceleratori Supercoduttivita Applicata) Laboratory of the INFN (Instituto Nationale di Fisica Nucleare) of Milano and of UNIMI (Università degli Studi di Milano) and in Thessaloniki (40.50° N, 22.90° E), Greece by Nuclear Physics Laboratory of Aristotle University of Thessaloniki, giving the extent of contamination in Northern Italy and in Northern Greece due to Fukushima fallout.

The activities of ^{131}I, ^{134}Cs and ^{137}Cs carried out in air, precipitation, soil, grass and milk at the two sites are presented here. Moreover, an evaluation of the effective dose for population was done based on the values obtained.

2. Material and Methods

Analysis for ^{131}I, ^{137}Cs and ^{134}Cs in airborne aerosols were carried out in daily samples for both sites during March-April, 2011. Air sampling was carried out with high volume air samplers and glass fiber filters. The aerosol samples were collected for 23 hours, with one hour interval between the

collections, time interval necessary for avoiding overheating and possible destruction of the motor pump. The flow rate was about 60 cfm resulting in a total daily air volume throughput of average 2200 m^3. The collection substrate was glass fiber filters type TFAGF 810 with dimension "8 x 10" with collection efficiency of 99.98% for particles as small as 0.3 μm in diameter [16].

The wet deposition was determined by exposing collection funnels to the atmosphere during the rainfall events. At the end of the rainfall, the contents of the funnels were collected and acidified with 1 mL orthophosphoric acid 85% per litre of rainwater, in order to prevent any loss by absorption to the vessel [17].

Surface soil samples were collected from a depth of 0-7 cm (upper 7 mm) and from an area of 1 m^2. Grass samples were collected from a surface area of 1 m^2 and at the same time as soil sampling.

All samples were counted using HPGe detectors at about 50% relative efficiency in case of Milano samples and with 42% efficiency in case of Thessaloniki [18, 19]. All detectors were calibrated with reference sources and standard geometries with overall uncertainties no more than 3%. In order to avoid corrections for the different geometry configuration, all samples were measured in the same geometrical assembly as the calibrated sources. In particular 1 L Marinelli geometry was used for rainwater, milk, grass and soil samples; the air filters were folded and compressed by means a hydraulic press up to 3t to obtain a cylinder of 4.7 cm in diameter and 3mm in height.

The two Laboratories performed intercomparison tests in order to assure: a) the compatibility of the sampling procedures by checking and comparing the calibration and the air samplers; b) the quality of low–background gamma spectrometry measurements, which is guaranteed by intercomparison measurements organized by international organizations like IAEA and EU. Gamma ray energies used for the analysis were 364.489 keV for the determination of ^{131}I, 661.660 keV for ^{137}Cs and both 604.721 and 795.864 keV for ^{134}Cs [20]. In the case of ^{134}Cs, corrections due to coincidence effect were applied [21].

All activities given in the text and figures are decay-corrected at the mean time of the sampling interval. The overall uncertainties in the determined activities are attributed mainly to the statistical counting and sampling uncertainties, while the contribution of calibration uncertainty is less than 3%.

3. Results and Discussion

3.1. ^{131}I, ^{137}Cs and ^{134}Cs activity concentration in surface air

In Milano region increased atmospheric radioactivity concentration for ^{131}I (322±36 μBq m^{-3}) was observed on an air filter taken on 30 March 2011, while the maximum activity concentration of 467±25 μBq m^{-3} was recorded on April 3-4. The first evidence of Fukushima fallout was confirmed with ^{131}I and ^{137}Cs measured in precipitation at two sampling sites at Milano on 28 March with the concentrations of ^{131}I and ^{137}Cs in the rainwater equal to 0.89±0.11 Bq L^{-1} and 0.12±0.09 Bq L^{-1}, respectively.

The ^{131}I was detected initially (143±21 μBq m^{-3}) in Thessaloniki on 24 March 2011. The maximum ^{131}I activity concentration of 497±53 μBq m^{-3} was detected on 4 April while a second maximum of 425±48 μBq m^{-3} on 10 April.

The time evolution of ^{131}I activity concentration in the airborne particulate from the two regions is presented in Fig.1. It is clear that in both regions similar maximum values have been observed during 3-4 April despite the considerable distance and the possible differences in climatic parameters that may influence the concentrations of radionuclides in air, but there is a tendency of a shift in the peak days.

In Figs 2 and 3 ^{137}Cs and ^{134}Cs time evolution activity concentrations respectively in the airborne particulate for the two regions are reported. The highest values of ^{137}Cs and ^{134}Cs in air over Milano, 63±29 μBq m^{-3} and 61±22 μBq m^{-3} respectively, were observed on April 10 and over Thessaloniki, 145±18 μBq m^{-3} and 126±11 μBq m^{-3} respectively, on April 4. In most of the cases the observed maxima values for ^{137}Cs and ^{134}Cs do not follow the maxima of ^{131}I, with the only exception is in the region of Thessaloniki on 4 April where the highest values of ^{131}I, ^{137}Cs and ^{134}Cs were

observed. In both regions of investigation the ^{134}Cs/^{137}Cs ratio values remained almost stable at about 1 throughout the period of measurements. This value is in agreement with the measurements of High Energy Accelerator Research Organization at Tsukuba, Japan [22] and, in according with Kirchner et al. [23], it corresponds to the isotopic composition observed in Europe related to emissions of units 1 and 3 of the destroyed Fukushima reactor. As well known this ratio value is related to the burn-up history of the damaged nuclear fuel of the destroyed nuclear reactor: for comparison after the Chernobyl accident the ratio was about 0.5 [24-26].

The presence of more than one peak of ^{131}I and ^{137}Cs (Figures 1,2) demonstrates that the radioactive plume was continuously transferred from Fukushima, Japan to Europe up to end of April and/or first days of May. These results are in agreement with values for ^{131}I and ^{137}Cs reported from other European countries [4, 9 – 11, 13, 27 – 31].

The ratio of ^{131}I/^{137}Cs in air in Milano was 11.7 on April 3-4, reached the higher value of 16.8 on April 7-8 and then declined to 2.6 on April 28. The ratio of ^{131}I/^{137}Cs in air in Thessaloniki was first 18.4 on March 26 and then decreased to 3.4 on April 4. These data confirm that the radioactive cloud was first richer in ^{131}I, as iodine is a more volatile element than cesium.

After first days of May in both regions there was no more detectable presence of these artificial radionuclides. These data are consistent with the ones observed over the European continent [9] and show a rather large degree of homogeneity of the plume, following redistribution process in the troposphere.

3.2. ^{131}I, ^{137}Cs and ^{134}Cs activity concentration in rainwater samples

Wet deposition by rain is known to be the most effective transfer path for airborne radioisotopes to ground and water bodies. Wet precipitation affects the levels of deposition in the land, including grass consumed by animals that produce milk, and could affect the levels of the radioactivity in the tap water.

Convective storms on 29 March and 14 April, 2011 in Thessaloniki and 28 March and 12-15April, 2011 in Milano were responsible for carrying the bulk of the radioactive material from the atmosphere to the ground.

The ^{131}I activity concentration in samples collected in Thessaloniki is ranging between 0.69 ± 0.18 Bq L^{-1} to 0.096 ± 0.026 Bq L^{-1} while in Milano was 0.89 ± 0.11 Bq L^{-1} and 0.72 ± 0.13 Bq L^{-1} in the samples collected in two different **regions near the city of Milano** on 28 March and 0.29 ± 0.09 Bq L^{-1} after the precipitation of 15 April. The observed ^{131}I values in rainwater in all the sites were below 1 Bq L^{-1}, two orders of magnitude less than the limit of 170 Bq L^{-1} [32], fixed by the Food and Drug Administration (FDA) as intervention level for ^{131}I in drinking water and infant milk. So, all tap water in Northern Greece and Italy were completely safe in terms of its ^{131}I content, taking also into account that rain water undergoes to many dilutions before becoming tap **water.**

3.3. ^{131}I, ^{137}Cs and ^{134}Cs activity concentration in soil, grass and milk samples

^{131}I was not detected in soil samples in Thessaloniki, while in Milano the maximum ^{131}I activity concentration observed was as high as 0.20±0.13 Bq kg^{-1}. No ^{134}Cs was observed from the region of Thessaloniki, while in Milano ^{134}Cs activity concentration was below 1 Bq kg^{-1}. ^{137}Cs activity concentration as high as 46 Bq kg^{-1} was observed in Thessaloniki and as high as 25 Bq kg^{-1} in Milano.

Grass was contaminated with ^{131}I and ^{137}Cs at a low level. The observed values of ^{131}I and ^{137}Cs in grass samples were in Milano between 0.037± 0.013 - 0.135 ± 0.049 Bq kg^{-1} and 0.047± 0.030 -0.089 ± 0.032 Bq kg^{-1} respectively, while in Thessaloniki between 0.84 ± 0.13 - 1.35 ± 0.17 Bq kg^{-1} and 0.33 ± 0.11 - 0.38 ± 0.05 Bq kg^{-1} respectively. The wide range of the observed values in the two sites was probably due to the different masses collected per area as a consequence of the different growth rate at the different sampling days, the surface of the grass is not homogeneous, the leaf blades

do not present a wide surface area, while the deposited particles on the grass cannot remain attached to the leaves due to the wind, that was present in that days.

Iodine-131 was detected and measured in fresh goat and cow milk from rural areas in both sites. The highest value of ^{131}I activity concentration, 1.2 Bq L^{-1}, was observed in Thessaloniki on 30 March. The reported ^{131}I values in milk samples in Milano were below 1 Bq L^{-1}. After the end of May **there was no more detectable presence of** ^{131}I in milk samples.

Finally, the relatively high activity concentration of ^{137}Cs and the very small ratio ^{134}Cs/^{137}Cs values in all soil, grass and milk samples, far below the value of 1 (that was observed in atmospheric data), in combination with the absence of ^{134}Cs in most of these samples, is an indication that ^{137}Cs is the one remaining from Chernobyl and past global fallout rather than due to Fukushima fallout.

4. Dose Assessment

After the Fukushima accident a number of dose assessments have been carried out for the populations living in the north-west fallout zone of the Fukushima nuclear accident, by MEXT in Japan, DOE/NNSA in USA, IRSN in France [33, 34], with quite similar projected dose values. In the more affected regions the estimated projected doses reach particularly significant values, some of them even above 200 mSv, which are no longer in the range of "low doses" according to UNSCEAR 2000 definition [35]. The level of external projected doses in upcoming years is up to 4 Sv lifetime in the high-contaminated areas of 30 MBq m^{-2} of ^{137,134}Cs.

On the contrary, the radioactive plume that reached European countries has only small amounts of radioactive isotopes. However, these isotopes, that were observed at low-level in the air boundary layer, were deposited by wet and dry deposition and have contaminated the land, and as a consequence the whole food chain. So the radioisotopes of cesium and iodine were found above their detection limits in all environmental samples but very far below levels of concern. Based on the observed values in our regions the total doses have been calculated.

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

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

where,

E_{est} is the effective dose for exposure;

$J_{j,ing}$ and $J_{j,inh}$ 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.

The evaluation of the committed dose is done using the maximum detected concentration values measured in the environment for ^{131}I, ^{134}Cs and ^{137}Cs in a hypothetical case when the radionuclide intakes are assumed to be constant for one year and taking into account the h(g) coefficients for population of age younger than 1 year old and older than 17, as reported in the Italian Radioprotection Law [36]. In the calculation we assumed the annual individual usage factors for external exposure, inhalation and consumption of foods reported in NCRP-123 [37]. Even in this worst scenario, the values obtained with an extremely high conservative model confirm that the dose contributions of Fukushima fallout due to different pathways to Italian and Greek population were at least one order of magnitude less than the limit of 1 mSv a^{-1}. Furthermore and as a comparison, the derived dose equivalent from inhalation of airborne materials is very small compared to 1.25 mSv a^{-1} derived from inhalation of natural radiation sources, stemming mainly from radon 1.15 mSv a^{-1} and thoron 0.10 mSv a^{-1} [35].

5. Conclusions

Determination of ^{131}I, ^{137}Cs and ^{134}Cs in environmental samples were carried out in Thessaloniki (40°N) and in Milano (45°N) after the Fukushima accident during the period of March-May, 2011. All samples had very low levels of radioactive material, consistent with releases from the damaged Japanese nuclear reactors.

Activity concentrations of radionuclides in air measured at two different sites were almost similar taking into account the experimental errors. In these samples the ^{134}Cs/^{137}Cs ratio, throughout the measurement period, remained almost stable at about 1, value that is the fingerprint that these radionuclides are related to Fukushima disaster. On the contrary the relative high activity concentration of ^{137}Cs, the very small ratio ^{134}Cs/^{137}Cs far below the value of 1 in all soil, grass and milk samples, in combination with the absence of ^{134}Cs in most of these samples is an indication that the ^{137}Cs detected in these samples is related to the one remaining from Chernobyl and past global fallout in soil and as a consequence in grass and milk rather than due to Fukushima fallout.

The estimated committed doses for population related to the contributions of Fukushima fallout due to different pathways were at least one order of magnitude less of the limit of 1 mSv a^{-1}, even if the calculations are made using high conservative assumptions. In conclusion, radioisotopes of caesium and iodine were found above their detection limits in all environmental samples, but well below levels of concern.

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Figure Captions

Fig. 1 Time dependence of the ^{131}I activity concentration measured in airborne particulates at the two laboratories in Milano and Thessaloniki

Fig. 2 Time dependence of the ^{137}Cs activity concentration measured in airborne particulates by the two laboratories in Milano and Thessaloniki

Fig. 3 Time dependence of the ^{134}Cs activity concentration measured in airborne particulates by the two laboratories in Milano and Thessaloniki
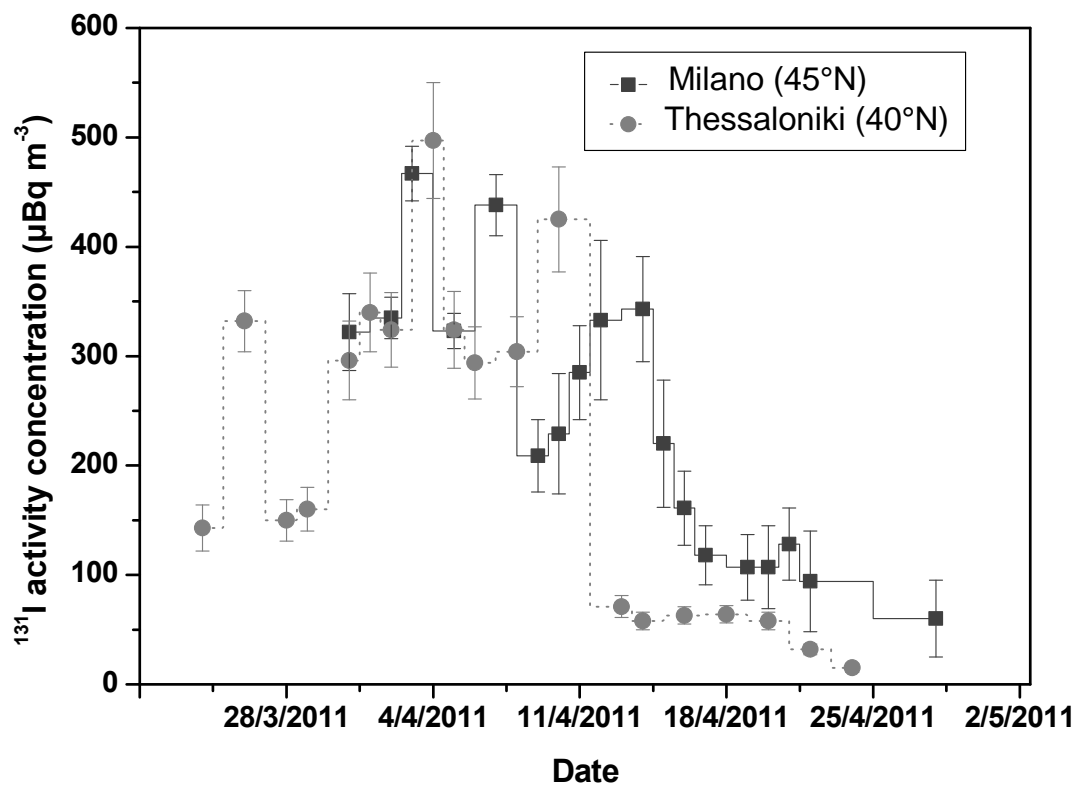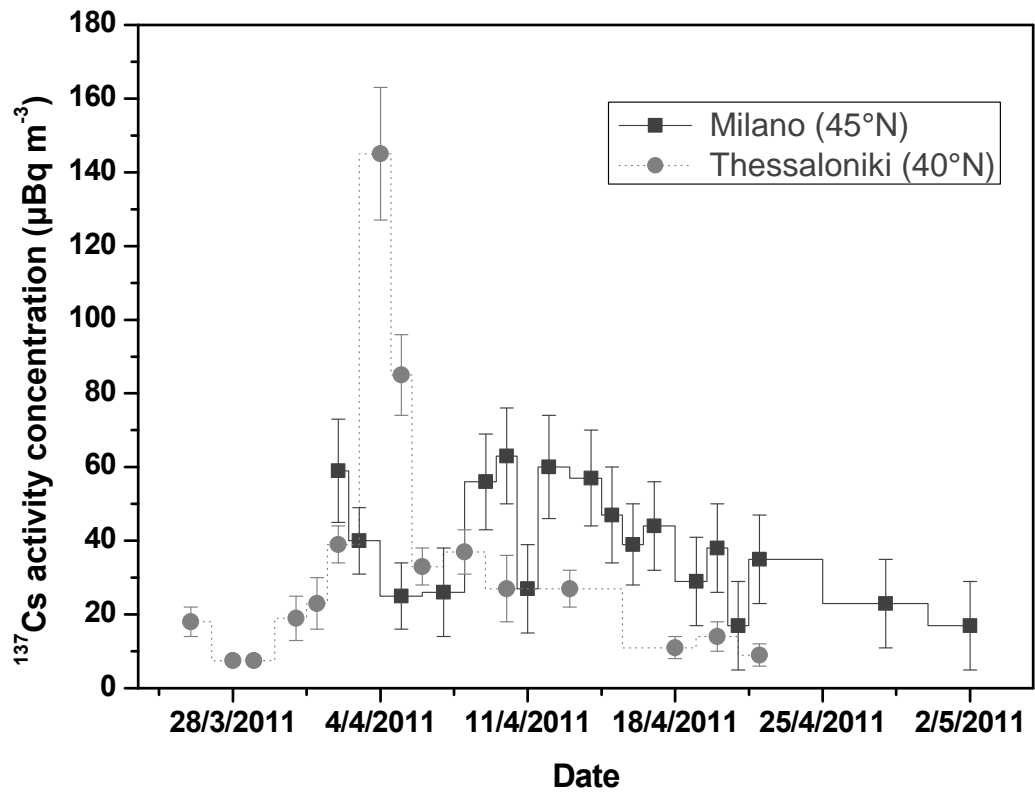
Fig. 1

Fig. 2

Fig. 3