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An air-mass trajectory study of the transport of radioactivity from Fukushima to Thessaloniki, Greece and Milan, Italy

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HIGHLIGHTS

- Comparison studies of the Fukushima fallout in air over Milano, Italy and Thessaloniki, Greece.
- Detailed time series of ^{131}I and ^{134},^{137}Cs monitored in the air over the two regions of investigation.
- HYSPLIT back trajectories analysis for interpretation of the variations of measured radionuclides.
- Confirmation of the westerly Japanese origin of the considered air masses.

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ABSTRACT

Analyses of ^{131}I, ^{137}Cs and ^{134}Cs in airborne aerosols were carried out in daily samples at two different sites of investigation: Thessaloniki, Greece (40° N) and Milan, Italy (45° N) after the Fukushima accident during the period of March–April, 2011. The radionuclide concentrations were determined and studied as a function of time. The ^{131}I concentration in air over Milan and Thessaloniki peaked on April 3–4, 2011, with observed activities 467 μBq m^{-3} and 497 μBq m^{-3}, respectively. The ^{134}Cs/^{137}Cs activity ratio values in air were around 1 in both regions, related to the burn-up history of the damaged nuclear fuel of the destroyed nuclear reactor. The high ^{131}I/^{137}Cs ratio, observed during the first days after the accident, followed by lower values during the following days, reflects not only the initial release ratio but also the different volatility, attachment and removal of the two isotopes during transportation due to their different physico-chemical properties. No artificial radionuclides could be detected in air after April 28, 2011 in both regions of investigation. The different maxima of airborne ^{131}I and ^{134},^{137}Cs in these two regions were related to long-range air mass transport from Japan, across the Pacific and to Central Europe. Analysis of backward trajectories was used to confirm the arrival of artificial radionuclides following atmospheric transport and processing. HYSPLIT backward trajectories were applied for the interpretation of activity variations of measured radionuclides.

1. Introduction

A large amount of radioactive materials were discharged into the atmosphere after the Fukushima Dai-ichi Nuclear Power Plant accident in Japan caused by a magnitude 9.0 earthquake and a subsequent tsunami on March 11, 2011. The Fukushima nuclear accident in Japan is the worst nuclear disaster following the 1986 Chernobyl accident. The Japanese Nuclear and Industrial Safety Agency (NISA) estimated that the total activity released into the atmosphere from reactors of the Fukushima Dai-ichi NPP were approximately 1.6×10^{17} Bq for ^{131}I and 1.5×10^{16} Bq for ^{137}Cs (NERH, 2011), which correspond to about 10% of the total amount released by Chernobyl accident, 1.76×10^{18} Bq for ^{131}I and 8.5×10^{16} Bq for ^{137}Cs, respectively (NEA, 2002 as cited by Kim et al., 2012). Stohl et al. (2011) reported higher releases for ^{137}Cs, 6.25×10^{16} Bq. The radioactive air was estimated to have been diluted by a factor of 10^5–10^6

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after transporting for five days across the Pacific Ocean to North America (Diaz et al., 2011).

Fukushima accident showed once again how fast airborne radionuclides can be transported over long distances, under certain meteorological conditions. Fission products released from the Fukushima plant site since March 12, 2011 have been detected around the northern hemisphere in few days and also in the southern hemisphere about one month later (CTBTO, 2011). Radioactive materials were first detected in Eastern Russia on March 14 (CTBTO, 2011) and three days later on the west coast of the U.S. and Canada (Bowyer et al., 2011; Diaz et al., 2011). The contaminated air masses were mainly transported across the Pacific Ocean towards the North American continent and Europe (Lozano et al., 2011; Lujanienė et al., 2012a,b; Manolopoulou et al., 2011; Masson et al., 2011; Paatero et al., 2012; Pittauerová et al., 2011; Povinec et al., 2012a; Tositti et al., 2012) and towards Central Asia (Bolsunovsky and Dementyev, 2011). The most comprehensive radionuclide data over Europe has been collectively described by Masson et al. (2011).

Right after the Fukushima reactor accident monitoring activities were promptly organized worldwide. The data discussed in the present work includes the observations of both Fukushima related radionuclides and ^{7}Be of natural origin; these observations were performed in Milan (45.49° N, 9.20° E), Italy by LASA (Laboratorio Acceleratori e Supercoduttivita Applicata) Laboratory of the INFN (Instituto Nationale di Fisica Nucleare) Sez. of Milan and UNIMI (Università deli Studi di Milano) and in Thessaloniki (40.50° N, 22.90° E), Greece by the Nuclear Physics Laboratory of Aristotle University of Thessaloniki.

In the present work detailed time series of radioiodine and radiocaesium isotopes monitored in the air over Milan and Thessaloniki at ground-level sites, are reported. These time series give the extent of contamination in Northern Greece and Northern Italy due to Fukushima fallout and allow interpretation of the measured activities at the site of investigation resulting from a complicated air mass transport.

2. Material and methods

The air sampling was carried out with High Volume air samplers and glass fiber filters as collection substrate. Aerosol samples were collected for 23 h. The 1 h interval between the measurements was necessary to avoid overheating and possible failure of the pump motor. The flow rate was about 100 m^3 h^{-1} resulting in a total daily air volume throughput on average 2200 m^3. The collection substrate was glass fibre filters type TFAGF 810 with dimensions 20.3 cm × 25.4 cm and collection efficiency of 99.98% for particles as small as 0.3 μm in diameter. The air filters were folded and compressed by means of a hydraulic press to obtain a cylinder of 4.7 cm diameter and 3 mm height. All samples were measured using HPGe detectors with 15% relative efficiency for samples from Milan and with 42% efficiency in the case of the samples taken in Thessaloniki.

Both laboratories are involved in intercomparison tests organized by IAEA to assure the compatibility of the air volume sampling and low-background gamma spectroscopic measurements. The gamma ray energies used during the analysis were 364 keV, 662 keV and both 604 and 795 keV for the determination of ^{131}I, ^{137}Cs and ^{134}Cs, respectively. In the case of ^{134}Cs, corrections were applied due to coincidence effect.

All activities given in the text and figures are decay-corrected to the mean time of the sampling interval. The overall uncertainties are mainly attributed to statistical counting errors 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 concentrations in surface air

In Milan region, an increasing atmospheric radioactivity was observed on an air filter taken on March 30, 2011. The first evidence of Fukushima fallout was confirmed with ^{131}I and ^{137}Cs measured in precipitation at two sampling sites in Milan on March 28, 2011, with concentrations of ^{131}I and ^{137}Cs in the rainwater equal to 0.89 Bq L^{-1} and 0.12 Bq L^{-1}, respectively. The first ^{131}I detection in Thessaloniki was on March 24, 2011. The arrival time of the first measurable Fukushima air pollutants was in good agreement with the IRSN and Meteo-France predictions, presented several days before (IRSN, 2011). Increasing levels were generally noticed during March 28th to March 30th in central Europe, while a second peak was also detected between April 3rd and April 5th (Masson et al., 2011).

The maximum ^{131}I activity concentration, 467 μBq m^{-3} observed in Milan, on April 3–4, 2011 was almost similar to the highest value, 497 μBq m^{-3}, observed in Thessaloniki, on April 4, 2011 (Manolopoulou et al., 2011) and to the highest observed value (490 μBq m^{-3}) in Athens, 37° N (Kritidis et al., 2012). However these values are lower than those observed at northern latitudes; for example 810 μBq m^{-3} in Svalbard, 78° N (Paatero et al., 2012) and 3700 μBq m^{-3} observed in Lithuania, 54° N (Lujanienė et al., 2012a,b).

The time evolution of ^{131}I concentrations in the airborne particulate form from the regions of Milan and Thessaloniki is presented in Fig. 1. The maximum ^{131}I activity concentration of 497 μBq m^{-3} was detected on April 4, 2011 in Thessaloniki, with a second maximum of 425 μBq m^{-3} on April 10, 2012. In the region of Milan, three peaks were detected; on April 3, April 7 And April 14, 2011, with a decreasing time trend. It must be specified that at both sampling sites we measured only the fraction of ^{131}I bound to aerosol particles. The ratio value of the ^{131}I concentrations in gaseous phase to the ^{131}I concentrations in aerosol particles has been defined equal to 3.74, according to measurements in Athens (Potiriadis et al., 2011). This value corresponds to a ratio of gaseous to total ^{131}I equal to 0.79. 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.

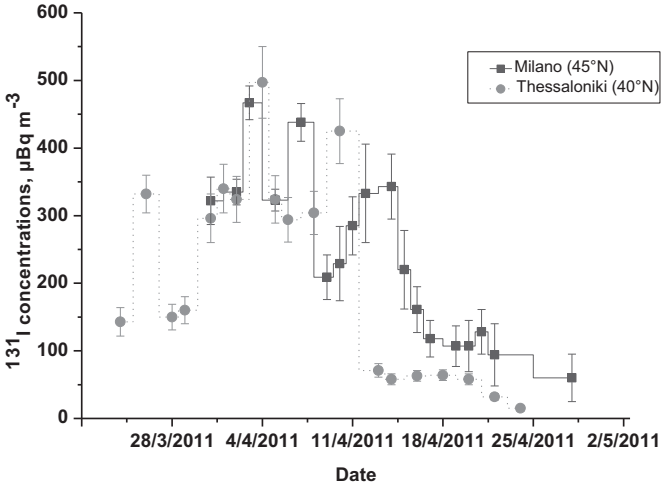

Fig. 1. Time dependence of the ^{131}I concentration measured in the airborne particulate form by the two laboratories located in Milan (45°) and Thessaloniki (40°).