

Radioactive pollution in Athens, Greece due to the Fukushima nuclear accident

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ABSTRACT

As a result of the nuclear accident in Fukushima Dai-ichi power plant, which started on March 11, 2011, radioactive pollutants were transferred by air masses to various regions of the Northern hemisphere, including Europe. Very low concentrations of ^{131}I , ^{137}Cs and ^{134}Cs in airborne particulate matter were measured in Athens, Greece during the period of March 24 to April 28, 2011. The maximum air concentration of ^{131}I was measured on April 6, 2011 and equaled $490 \pm 35 \mu\text{Bq m}^{-3}$. The maximum values of the two cesium isotopes were measured on the same day and equaled $180 \pm 40 \mu\text{Bq m}^{-3}$ for ^{137}Cs and $160 \pm 30 \mu\text{Bq m}^{-3}$ for ^{134}Cs . The average activity ratio of $^{131}\text{I}/^{137}\text{Cs}$ in air was 3.0 ± 0.5 , while the corresponding ratio of $^{137}\text{Cs}/^{134}\text{Cs}$ equaled 1.1 ± 0.3 . No artificial radionuclides could be detected in air after April 28, 2011. Traces of ^{131}I as a result of radioactive deposition were measured in grass, soil, sheep milk and meat. The total deposition of ^{131}I (dry + wet) was $34 \pm 4 \text{ Bq m}^{-2}$, and of ^{137}Cs was less than 10 Bq m^{-2} . The maximum concentration of ^{131}I in grass was $2.1 \pm 0.4 \text{ Bq kg}^{-1}$, while ^{134}Cs was not detected. The maximum concentrations of ^{131}I and ^{137}Cs in sheep milk were $1.7 \pm 0.16 \text{ Bq kg}^{-1}$ and $0.6 \pm 0.12 \text{ Bq kg}^{-1}$ respectively. Concentrations of ^{131}I up to $1.3 \pm 0.2 \text{ Bq kg}^{-1}$ were measured in sheep meat. Traces of ^{131}I were found in a number of soil samples. The radiological impact of the Fukushima nuclear accident in Athens region was practically negligible, especially as compared to that of the Chernobyl accident and also to that of natural radioactivity.

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1. Introduction

Since 1960, the Environmental Radioactivity Laboratory (ERL) is engaged with a permanent routine monitoring of the environmental radioactivity in Greece. This includes measurements in air, radioactive deposition, surface/drinking water and selected food samples. The results are regularly reported to the Greek Atomic Energy Commission. The initial damage in Fukushima Dai-ichi power plant, caused mainly by the enormous tsunami, related to the magnitude 9.0 earthquake of March 11, 2011 in Japan, was followed by a series of failures and explosions in a number of power units. As a result, very high quantities of radioactive material escaped to the atmosphere and spread initially across the entire Northern hemisphere. The first traces of Fukushima pollutants were detected in Europe on March 20 (IRSA, 2011). During April 2011, traces of Fukushima pollutants were detected in some regions of the Southern Pacific as well. As soon as the serious magnitude of the nuclear accident became obvious, despite the distance of about 8500 km from the accident place, ERL initiated a series of higher sensitivity measurements in air, aimed to detect radioactive

concentrations well below the 1 m Bq m^{-3} level. The first measurable concentrations of ^{131}I in air occurred in March 24, 2011, in good accordance with the predictions of the French IRSN, based on the application of a model for large scale atmospheric dispersion (IRSN, 2011). The results of our measurements during the period March 24 – April 28, 2011, including activity concentrations in air, deposition, grass, sheep milk and meat, are presented in this study.

2. Materials and methods

The coordinates of our air sampling station are: $37^{\circ}59'43.21'' \text{ N}$, $23^{\circ}48'57.16'' \text{ E}$. The air samples were collected with the high volume ESM Andersen sampler, combined with the 230 Series HV Cascade Impactors. The aerosol size classification by aerodynamic diameter was $>7 \mu\text{m}$, $2.5\text{--}7 \mu\text{m}$ and $<2.5 \mu\text{m}$. The back-up filter was Whatman 41 Quantitative, with an active sampling area of $20 \times 25 \text{ cm}$. The nominal air flow rate was $66 \text{ m}^3 \text{ h}^{-1}$. The sampling duration varied between 20 and 24 h, while the total air volume sampled was typically within $1350\text{--}1500 \text{ m}^3$. The air filter was consequently cut to form a cylinder of $D = 70 \text{ mm}$ and $H = 2 \text{ mm}$, which coincided with one of the measuring geometries, for which the high resolution gamma-spectrometry system has been calibrated.

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The filters were measured by use of a gamma-spectrometry system, based on the HpGe Eurisy EGPC 30-185-R detector of relative efficiency of 30% and energy resolution of 1.9 keV at 1.33 MeV, located inside a 10 cm cylindrical lead shield (System 1). The spectrum analysis software was Interwinner 4.1, adjusted roughly to 0.25 keV per channel. The measurement started typically 3 h after the end of sampling, in order to achieve a significant reduction of the activity of the short-lived decay products of ^{222}Rn , collected in the filter, which improved the peak-to-Compton ratio in the energy regions of interest. The gamma peaks used were 364.5 keV for ^{131}I , 661.6 keV for ^{137}Cs , 604.7 and 795.8 keV for ^{134}Cs . The duration of each gamma-spectrometry measurement varied between 20 and 48 h, depending on the observed counting statistics. The values for ^{131}I were decay-corrected and the data presented in this paper correspond to the midpoint of the sampling period. Depending on the sampling and measuring time, the lower limits of detection varied within 20–30 $\mu\text{Bq m}^{-3}$ for ^{131}I and 26–40 $\mu\text{Bq m}^{-3}$ for ^{137}Cs .

The radioactive deposition was collected by use of a steel sampler with area of 1 m^2 , in which the presence of a thin layer of distilled water was maintained during the whole duration of sampling. Depending on the intensity of the rainfall, different parts of the water sample were used for gamma-spectrometry analysis with a Marinelli beaker of 1000 mL volume. A second high resolution measuring system was used in this case, with a Canberra HpGe detector of 20% relative efficiency and energy resolution of 1.9 keV at 1.33 MeV, located inside a 10 cm thick cylindrical lead shield. The spectrum analysis was made by use of the Ortec Maestro II software, adjusted at 0.5 keV per channel (System 2). The system was calibrated for this geometry by use of a multi-nuclide standard solution.

The same System 2 and the same Marinelli sample geometry described above were used for the measurements of grass, sheep milk and sheep meat samples. Four samples of sheep milk from the Athens region were analyzed. The selection of these specific products reflects our experience from the impact of the Chernobyl accident in Greece, when grass appeared to be the earliest and very significant indicator of the radionuclide transfer to the surface biota, while sheep milk and meat were the products with highest concentrations of ^{131}I , measured in food during the early phase.

Sixteen grass samples from Athens region and from Lesbos island were collected and analyzed. In Athens, grass was collected from the region of NCSR "Demokritos", where air concentrations and radioactive deposition were measured, and from a pasture region near Grammatiko, where sheep milk and meat were collected as well. The Lesbos samples were collected in the frame of a different study, which coincided in time with the period of interest. The grass samples were collected from an area of 1 m^2 and only the part above the soil was kept for measurement. The soil was collected to a depth of 5 cm. The same sample depth was used by us after the Chernobyl accident. The sampling areas were selected to coincide with regions of our detailed post-Chernobyl sampling. The soil radioactivity was measured in the same system, by use of a cylindrical geometry of $D = 70$ mm and $H = 15$ mm. The same system and geometry were used for measurement of sea sediment samples.

Surface, near shore seawater samples were collected from a marine area near Athens – the Southeastern prefecture of Sunion. This sampling point was selected because the renewal of the water of Saronikos Gulf, which is adjacent to the metropolitan Athens region, occurs from this area (Hopkins and Coachman, 1975). In this case, a chemical separation of cesium was applied, followed by gamma-spectrometry analysis.

Finally, a significant number of imported products, originating from Japan and the wider Pacific region were analyzed, in the frame

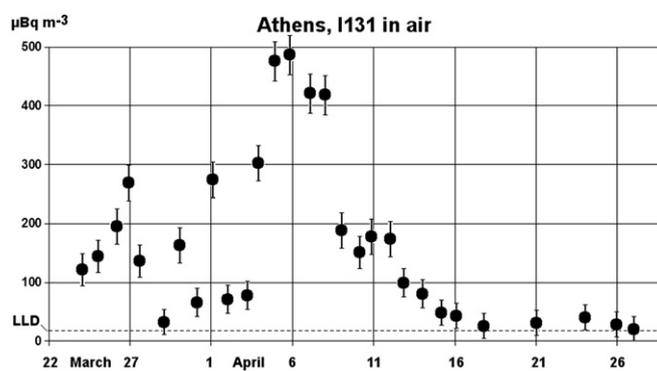


Fig. 1. Concentrations of particulate ^{131}I in air during the period March 24 - April 28, 2011.

of specific post-Fukushima EU directives (starting with the EU Commission Regulation No 351/2011 of April 11, 2011). Most of the samples were measured in System 2, by use of the 1000 mL Marinelli geometry. In this case, the duration of measurements was typically 5 h.

3. Results and discussion

The arrival time of the first measurable Fukushima air pollutants was in good agreement with the predictions of IRSN/MeteoFrance, presented several days before (IRSN, 2011). Concentrations of particulate ^{131}I (the form bound to aerosol) above the lower limit of detection (LLD) were measured during the period March 24 – April 28, 2011 (Fig. 1). The values of the combined standard uncertainties are shown as well. The highest value of $490 \mu\text{Bq m}^{-3}$ ($\pm 35 \mu\text{Bq m}^{-3}$ based on counting statistics) was measured on April 6, 2011. The integrated concentration of particulate ^{131}I during this period was $4.7 \text{ m Bq d m}^{-3}$. Our sampling method did not allow the collection of iodine in gaseous form, which, according to (Potiriadis et al., 2011), contributed significantly to the total concentration of ^{131}I in air.

Concentrations of ^{137}Cs and ^{134}Cs above the LLD level were measured during the period March 24 – April 14, 2011 (Fig. 2). The highest values of ^{137}Cs and ^{134}Cs were $180 \pm 40 \mu\text{Bq m}^{-3}$ and $160 \pm 30 \mu\text{Bq m}^{-3}$ respectively and were measured in April 6, 2011, the same day when the maximum value of ^{131}I was observed. The integrated concentration of ^{137}Cs during the mentioned period was lower than 1.3 mBq d m^{-3} , where "lower" reflects the inclusion of certain values lower than the LLD. The corresponding quantity for ^{134}Cs is difficult to estimate, due to the restricted number of values above the LLD.

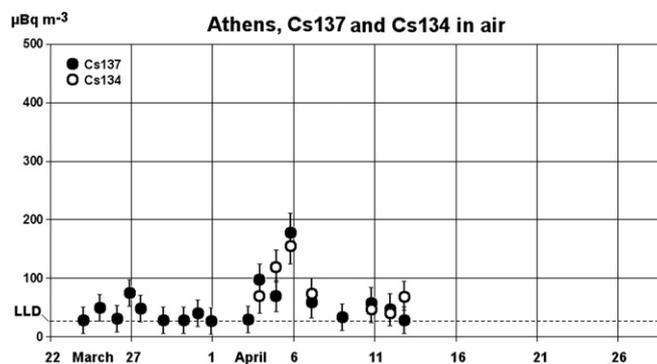


Fig. 2. Concentrations of ^{137}Cs and ^{134}Cs in air during the period March 24 - April 13, 2011.

The mean ratio of the measured air concentrations of $^{131}\text{I}/^{137}\text{Cs}$ was 3.0 ± 0.5 . The corresponding ratio of $^{137}\text{Cs}/^{134}\text{Cs}$ was 1.1 ± 0.3 . Both values are based on subsets of data with higher counting statistics.

Sampling and measurement of air continues until now (November 2011), on a weekly basis. No artificial radionuclides were detected in air after April 28, 2011.

Our results were compared with those, reported by two other laboratories for the Athens region. The maximum concentration of ^{131}I , measured by the Greek Atomic Energy Commission (GAEC) in a sampling location only 600 m distant from the ERL one, was $585 \pm 70 \mu\text{Bq m}^{-3}$, which is relatively close to our $490 \pm 35 \mu\text{Bq m}^{-3}$ value, taking into account the counting uncertainties (GAEC, 2011; Potiriadis et al., 2011). The maximum values of ^{137}Cs and ^{134}Cs measured by the GAEC, were 181 ± 42 and $158 \pm 20 \mu\text{Bq m}^{-3}$ respectively. These values are practically identical with the maximum observed by us. The integrated concentration of ^{131}I , as derived from the GAEC data, was 5.6 mBq d m^{-3} , a value about 20% higher than ours (4.7 mBq d m^{-3}).

The data of the Nuclear Engineering Department, National Technical University of Athens (NED/NTUA), with a sampling station located about 4 km South-West from this of ERL, are also in reasonable agreement with our data (NTUA, 2011). The integrated concentration of ^{131}I , as derived from the NED/NTUA data, was 5.5 mBq d m^{-3} , a value 17% higher than ours.

The results reported by two Thessaloniki laboratories, located about 300 km north of Athens, were not expected to be quite comparable with ours. Our Chernobyl experience shows that the considerable distance and the possible differences of climatic parameters may influence the air concentrations of radionuclides (GAEC, 1986a, 1986b). Nevertheless, the maximum concentration of $^{131}\text{I} = 497 \pm 53 \mu\text{Bq m}^{-3}$, reported by the Atomic and Nuclear Physics Laboratory (ANPL), Aristotle University of Thessaloniki (Manolopoulou et al., 2011a, 2011b), was very close to our value of $490 \pm 35 \mu\text{Bq m}^{-3}$, while the integrated concentration of ^{131}I , as derived from the ANPL data, was 3.8 mBq d m^{-3} , a value about 20% lower than ours.

The maximum air concentrations of ^{131}I and ^{137}Cs , measured by the Nuclear Technology Laboratory (NTL), Aristotle University of Thessaloniki (Potiriadis et al., 2011) were $408 \pm 61 \mu\text{Bq m}^{-3}$ and $132 \pm 61 \mu\text{Bq m}^{-3}$ respectively. These values are respectively 17% and 26% lower than ours. Taking into account also the data of ANPL, we can conclude that the radioactive pollution of air in Thessaloniki region was slightly lower than in Athens region.

The radioactive deposition of ^{131}I , measured by us during the period of detectable air concentrations was $34 \pm 4 \text{ Bq m}^{-2}$. Due to the very low levels of air concentrations and the related deposition rates, it was not possible to determine the deposition of ^{131}I on a daily basis (with the exception of two days of strong rainfall). Instead, 4 samplings of 1 week duration each were made and decay corrections to the middle point of the sampling period were applied

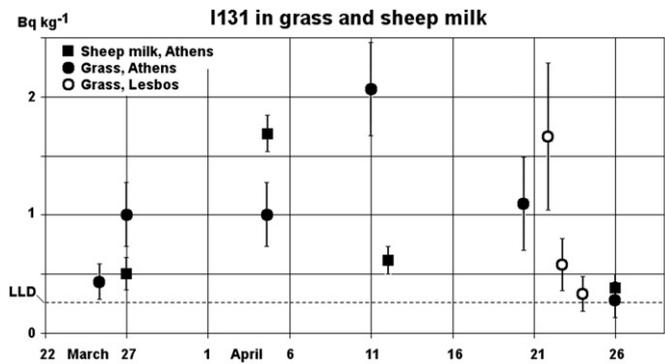


Fig. 3. Concentrations of ^{131}I in grass and sheep milk during the period March 24 - April 26, 2011.

in each case. This adds a potential systematic error to each weekly result, as long as the variability of the deposition rate, determined by the rainfall intensity during the sampling period, is not taken into account. This error was estimated, on the basis of rainfall data, and added to the one related to counting statistics. The deposition of ^{137}Cs and ^{134}Cs were not measurable, but based on the LLD levels for these radionuclides, we estimate their values to be less than 10 Bq m^{-2} for each radionuclide.

Our ^{131}I deposition value is in good agreement with the data for wet deposition of ^{131}I , provided by NED/NTUA (Savva et al., 2011).

Based on our experience from the Chernobyl accident, special attention was paid to grass, which is one of the earliest and most sensitive indicators of the transfer of radioactivity from the air to the surface flora. The concentrations of ^{131}I in grass exceeding the LLD level are shown in Fig. 3. The maximum concentration of ^{131}I in grass was $2.1 \pm 0.4 \text{ Bq kg}^{-1}$, while in 8 other samples values within $0.3\text{--}1.7 \text{ Bq kg}^{-1}$ were measured. Concentrations of ^{137}Cs above the detection limit of 0.3 Bq kg^{-1} were found in 9 samples, covering the range of $0.35\text{--}1.5 \text{ Bq kg}^{-1}$. As long as ^{134}Cs was not detected in any of these samples, we have to conclude that the major part of these concentrations is related to the ^{137}Cs pollution of soil due to the Chernobyl accident, propagating through root uptake.

Our ^{131}I concentrations in grass are close to those found by NTL/AUTH, where maximum value of $1.9 \pm 0.4 \text{ Bq kg}^{-1}$ was reported (Potiriadis et al., 2011).

Another lesson from the Chernobyl accident is the importance of sheep milk, which during May 1986 appeared to be the most sensitive indicator of the early ^{131}I pollution, given that the animals were in a regime of fresh grass pasture. The results for milk are also presented in Fig. 3. The maximum concentration of ^{131}I was $1.7 \pm 0.16 \text{ Bq kg}^{-1}$. The concentration of ^{137}Cs was above LLD in one case only ($0.6 \pm 0.12 \text{ Bq kg}^{-1}$), while ^{134}Cs was not detected. As in the case of grass, we have to attribute the observed ^{137}Cs concentration to the residual Chernobyl contamination, propagating through the grass-sheep food chain.

Table 1

Measurements in media other than air related to the impact of the Fukushima accident in Greece.

Region and number of samples	Period	Sample type, units	^{131}I min - max	^{137}Cs min - max	^{134}Cs min - max
Athens, 4	11.04–09.05.11	Soil, Bq kg^{-1}	<0.05–0.54	1.7–7.0	0.4–1.7
Lesbos, 2	23.04–25.04.11	Soil, Bq kg^{-1}	<0.05–0.40	10.6–18.0	<0.05–1.5
Athon, 3	25.05–26.05.11	Soil, Bq kg^{-1}	<0.05–1.2	<0.07–30.0	<0.05–3.0
Xanthi, 5	2010	Mushrooms, Bq kg^{-1}	<0.05	<0.07–2.1	<0.05
Xanthi, 4	May 2011	Mushrooms, Bq kg^{-1}	<0.05	<0.07–1.2	<0.05
Sunion, 3	29.03–15.06.11	Seawater, Bq m^{-3}	<0.6	3.0–3.3	<0.6
Sunion, 3	29.03–15.06.11	Sediment, Bq kg^{-1}	<0.05	0.7–1.4	<0.05
Sunion, 3	15.06.11	Fish, Bq kg^{-1} dry weight	<0.04	<0.06–0.24	<0.04

The values lower than the LLD (1σ) are presented in the form “<LLD”.

Table 2

Comparison of the radioactive contamination levels, observed after the Fukushima and the Chernobyl accidents in Athens region (GAEC, 1986a, 1986b).

Sample type, units	Isotope, form, quantity	Fukushima value	Chernobyl value	Ratio Chernobyl/Fukushima
Air, mBq m ⁻³	¹³¹ I, particulate form, maximum concentration	0.5	16000	32000
Air, mBq d m ⁻³	¹³¹ I, particulate form, integrated concentration	4.7	40000	8500
Air, mBq m ⁻³	¹³⁷ Cs, maximum concentration	0.18	4000	22000
Air, mBq d m ⁻³	¹³⁷ Cs, integrated concentration	< 1.3	10000	> 7700
Earth surface, Bq m ⁻²	¹³¹ I, radioactive deposition	34	17000	500
Grass, Bq kg ⁻¹	¹³¹ I, maximum concentration	2.1	2800	1300
Sheep milk, Bq kg ⁻¹	¹³¹ I, maximum concentration	1.7	4500	2600

Our values can be compared with the maximum of ¹³¹I in sheep milk of 4.4 Bq kg⁻¹ reported by NED/NTUA (Savva et al., 2011). NTL/AUTH reported a maximum of 2.4 Bq kg⁻¹ (Potiriadis et al., 2011). A maximum value of 1.2 Bq L⁻¹ was found by ANPL/AUTH, but the sample originated from Trikala, Central Greece (Manolopoulou et al. 2011b).

After the initial detection of ¹³¹I in sheep milk, we analyzed 6 samples of sheep tissue – 3 of viscera and 3 of muscle. In two cases, milk samples from the same herd were collected and analyzed. The maximum concentration of ¹³¹I was found in sheep viscera and was 1.3 ± 0.2 Bq kg⁻¹, while lower maximum values of ¹³¹I were measured in the muscle parts – 0.25 ± 0.12 Bq kg⁻¹. Concentrations of ¹³⁷Cs up to 1.3 ± 0.2 Bq kg⁻¹ were measured in sheep viscera, but the absence of measurable ¹³⁴Cs values indicates again the post-Chernobyl origin of ¹³⁷Cs.

The results of other measurements, related to the impact of the Fukushima accident, are summarized in Table 1. The measurements of soil samples indicated the presence of ¹³¹I and ¹³⁴Cs. In all other cases these radionuclides were not detected. The mushroom samples included species of *Basidiomycota* named *Craterellus cornicopoides*, *Calocybe gambosa*, *Boletus edulis*, *Cantharellus cibarius* and *Agaricus bisporus* collected in Northern Greece during 2010 and on May 2011. The 2011 values were lower than those observed before the Fukushima accident. (Matzourani et al., 2001).

The activity concentrations of ¹³⁷Cs in seawater ranged between 2.9 and 3.6 Bq m⁻³, while those of ¹³⁴Cs and ¹³¹I were below the detection limits. Sediment samples were collected in Sounio and Corinthiakos Gulf, where ¹³⁷Cs concentrations varied between 0.3 and 3.1 Bq kg⁻¹, while ¹³⁴Cs and ¹³¹I concentrations were below the LLD of 0.1 Bq kg⁻¹. According to our measurements carried out before the accident, the background activity of ¹³⁷Cs in seawater Greek marine environment ranged from 2.7 to 3.7 Bq m⁻³ (Florou et al., 2010), whereas at coastal surface sediments was up to 5 Bq kg⁻¹, with an exception at Thermaikos Gulf, North Aegean Sea, where it was relatively higher (Florou et al., 2011). It can be concluded that the impact of the Fukushima accident in the Greek marine environment was not detectable.

In the frame of the ERL program of provisional services, imported products from Japan and the Pacific area were analyzed and respective certificates were issued, according to the EU Commission Regulation No 351/2011 of 11 April 2011. This included so far 82 samples of seafood (FAO 61, 51, 81), 5 samples of candies, 2 samples of rice products, 51 samples of the alga *Chlorella sp.* (used in the aquaculture), 16 samples of miscellaneous feeding stuffs and fish oil (e.g. egg-cysts of *Artemia salina* for fish juveniles). Traces of ¹³¹I and ¹³⁷Cs (up to 2 Bq kg⁻¹) were detected in a few samples of *Chlorella sp.*. In the rest of the samples, the concentrations of these radionuclides were close or below the LLD of 0.2 Bq kg⁻¹.

4. Comparison with the impact of the Chernobyl accident

After May 3, 1986, when the first polluted air masses entered Greece, ERL was the major source of official information, related to

the radiological impact of the Chernobyl accident in the country. The most important data from this period were summarized by Kritidis and Florou (2001), while a lot of details are given in a number of GAEC reports (e.g. GAEC, 1986a, 1986b).

Comparison of the activity concentrations measured after the Fukushima and Chernobyl accidents in Athens region are given in Table 2. The ratio of integrated concentrations of ¹³¹I and ¹³⁷Cs in air, observed during the two pollution events, can be used for estimation of the corresponding ratio of inhalation doses. Using the same presumptions and dose conversion factors, applied in (GAEC, 1986a), we derive an inhalation effective dose due to the Fukushima accident in Athens region equal to 1.1 nSv for adults and 1.9 nSv for 10-year children. These values are of negligible radiological importance, as long as they 6 orders of magnitude lower than those, resulting from natural radioactivity in air (mainly radon and thoron decay products). The difference between the air and deposition Chernobyl-to-Fukushima ratios for ¹³¹I are probably a result of the strong precipitation during the Fukushima period, as opposed to the absence of rainfalls in Athens during the period of strong Chernobyl air pollution. Not also, that in both cases the particulate form of ¹³¹I in air has been collected.

We do not have enough data for a reliable evaluation of the ingestion doses related to the Fukushima accident. Given the worst scenario, defined by the 1:1300 ratio of Fukushima-to-Chernobyl grass contamination, we assume that the Fukushima ingestion dose is lower than 400 nSv, which is again a negligible value, if compared to the average ingestion dose due to the natural radioactivity, which is nearly 3 orders of magnitude higher.

5. Conclusions

1. Measurable concentrations of ¹³¹I in air were detected during a period of 34 days, while ¹³⁷Cs in air was detected during a shorter period of 20 days. The maximum concentration levels of these radionuclides were about 4 orders of magnitude lower than those of the natural radioactivity in air. No traces of artificial radionuclides in air were detected after April 28, 2011.
2. The average ratio of ¹³¹I to ¹³⁷Cs activity concentrations in air was 3.0 ± 0.5. The average ratio of ¹³⁷Cs to ¹³⁴Cs activity concentrations in air was 1.1 ± 0.3.
3. The maximum concentrations of ¹³¹I measured in grass and sheep milk were more than 3 orders of magnitude lower than those measured after the Chernobyl accident. This indicates a negligible ingestion dose due to the Fukushima radionuclides. No radioactive iodine was detected in grass and sheep milk after the end of April 2011.
4. It is still possible to measure the presence of ¹³⁷Cs, originating from the Chernobyl fallout, not only in soil samples, but also in grass, as a result of the root uptake.

Based on the measured activity concentrations, we conclude that the radiological impact of the Fukushima nuclear accident in the region of Athens was extremely low. It did not require the

application of any radiation protection measures. The experience gained from this pollution event is mostly related to problems of detecting concentrations of radioiodine and radiocesium close to the detection limits. These levels are usually 1–2 orders of magnitude lower than the lowest, which can be still considered as “radiologically significant”. Therefore, a question of optimization arises, at least for scientific units, for which the radiation protection of the population is of highest priority. The optimization concerns the selection of sampling and measuring parameters, which assure precision of results compatible with the very low radiological significance of the event.

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