Fission product activity measurements in air particulate filters collected after Fukushima accident at Palermo, Italy

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Abstract. In the framework of routine operation of AGN-201 COSTANZA Nuclear research reactor, measurements of radionuclide air concentration are periodically performed by filtering an high volume of air through paper filters and by using HPGe gamma-ray spectrometry. After Fukushima accident, a series of samplings was carried out with a daily frequency so to detect the possible arrival of air radioactive contamination and to follow its evolution.
Particulate collection was performed by suction of atmospheric air through 45cm × 45cm Sofiltra-Poelman HYN-75 (Bleu type) cellulose filter paper using a high-volume air sampler located on the roof of our department 20 m above ground-level. The sampling time was generally set to 14 h from 6 p.m. to 8 a.m. the next day; the filtered air volume is typically 10,000-13,000 m^3. After particulate sampling, the filters were sprayed with a suitable fixer, cut into strips, folded and pressed into 6 cm side and 0.7 cm thickness packets by a 15-t press. These samples are then measured by HPGe gamma spectrometric systems. The analysis of the spectrometric measurements highlights the presence of ^{131}I, ^{134}Cs, ^{136}Cs , ^{137}Cs and, only for a sample, traces of ^{132}Te-^{132}I. The trend of air concentration values shows a rapid initial increase, related to the most significant release from Fukushima plant, followed by a steady decrease in the values caused by air masses dilution and, only for ^{131}I and ^{136}Cs, radioactive decay. From a dose to population point of view, no significant values are obtained. Compared with the values determined immediately after the Chernobyl accident, the concentration values are from 1,000 to 100,000 times lower while, for only ^{137}Cs, are comparable with the ones highlighted after the Algeciras (Spain) accident.
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KEYWORDS: Air Particulate, Fukushima accident, radionuclide concentration.

1. Introduction

A research activity on environmental radioactivity monitoring was held for many years at Energy Department of Palermo University. The analysis started with the detection of radioactive contamination produced by atmospheric nuclear tests in the early sixties of the last century and continued with measurements oriented to determine natural or artificial radioactive compounds in air particulate as a way of transport of various pollutants [1-14]. Particularly interesting are measurements and monitoring activities conducted in the aftermath of the Chernobyl accident [2-5]. For these activities, since 1979 until a few years ago, a sampling station equipped with an high volume sampler (about 15000 m^3/day) was extensively used.
Following the notices on the accident in Fukushima and on a possible contamination in the air, the frequency of sampling was intensified (up to become at least daily) and the samples were analyzed with at least two spectrometric systems.
In the following it will show the results of measurements of fission products concentration in the air of Palermo related to Fukushima plant accidental release. The results of the monitoring activity confirm that air concentration values have no significance from a dose point of view, being very far from the limits of concentration in the air provided by rules with reference to population dose.

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2. Materials and Methods

The sampling of air particulates was carried out from March 21, 2001 until the first ten days of May 2011. The time period was bounded between the report on a possible release of radioactive substances from the plant in Fukushima (Japan), after about 10 days of the earthquake and the "tsunami" that hit the power station, and the second ten days of May 2011 when airborne concentrations were below the detection limits of the used measurement instruments.
The suctions were made with a high volume sampler (9,000 ÷ 14,000 m^3 of filtered air per sample), located about 20 m above ground level on the roof of the Department, and using a cellulose filter Sofiltra Poelman HYN-75 (Bleu type) of size 45cm x 45cm. To avoid excessive clogging of the filter, the sampling time was set at 14 h, from 6 pm to 8 am the next day (with timer), with the exception of longer time sampling during holiday periods. At the end of sampling, the filter was cut, packed and pressed so to obtain a size 6 cm × 6 cm × 0.7 cm, a measurement geometry called "packet-sample" and used for gamma-ray spectrometry analysis with HPGe detectors.
During the first week of monitoring were also carried out some suctions during the day so to recognize the possible arrival of contaminated air and to evaluate the change in concentration throughout the day. Once performed the initial evaluations, it was found that the amount of radionuclide activities deposited on a filter were rather small and it was decided to perform a single daily sampling with a more long time in order to have more activity on a filter. In this way, the probability to have activity values greater than the detection limits was improved.
The filters were measured with two spectrometric systems equipped with ORTEC HPGe detectors, relative efficiency 60% and 35%, respectively. For each spectrometric system an efficiency calibration was available, performed a few years ago with the use of different methods. So, any systematic errors in the determination of radionuclide activities on the filter can be detected. However, similar results are obtained from the analyses of spectra detected on the same filter with both the detectors, which confirms the goodness of the calibration and measurement procedures. Minimum Detectable Activity values (MDA), for some energies taken into account, are given in Table 1. As regards the procedure for determining the efficiencies refer to what is already specified in [10].

Table 1: MDA evaluation for main artificial radionuclides identified in the samples taken after Fukushima accident.

Radionuclide	Energy (keV)	Emission probability (%)	MDA (Bq)	Air concentration (microBq/m^3) (*)
^{131}I	364	81,2	0,021	1,5 -2,3
^{134}Cs	604	97,6	0,018	1,3-2
^{137}Cs	662	85,2	0,015	1,1-1,7

(*) with the hypothesis of 9,000÷14,000 m^3 of filtered air volume.

3. Results and discussion

Figs. 1-4 show the time dependence of airborne concentrations of ^{131}I (only particulate matter), ^{134}Cs, ^{136}Cs and ^{137}Cs measured from March 24, 2011 (daylight sampling), in which there was the first detection of ^{131}I, until the first days of May 2011, when airborne concentrations were comparable with detection limits of the spectrometer systems used. The first detection of ^{131}I was recorded less than 13 days after the accident, and probably a few days after the release from damaged plant. The airborne concentration values were very low but significantly higher than MDA reported in Table 1. Lower airborne concentration values were computed for ^{134}Cs and ^{137}Cs in the first sample in which they were identified, taken a few days later. We want here to point out that a reliable determination of airborne concentrations of this order of magnitude is a difficult task and can only be done through the filtration of large volumes of air, while it seems less important to have an high sensitivity of the spectrometer systems. The airborne concentration trends are characterized by a sudden increase and a gradual decrease in concentrations, the last attributed to a progressive dilution of concentrations in air masses, and also for ^{131}I and ^{136}Cs to radioactive decay.

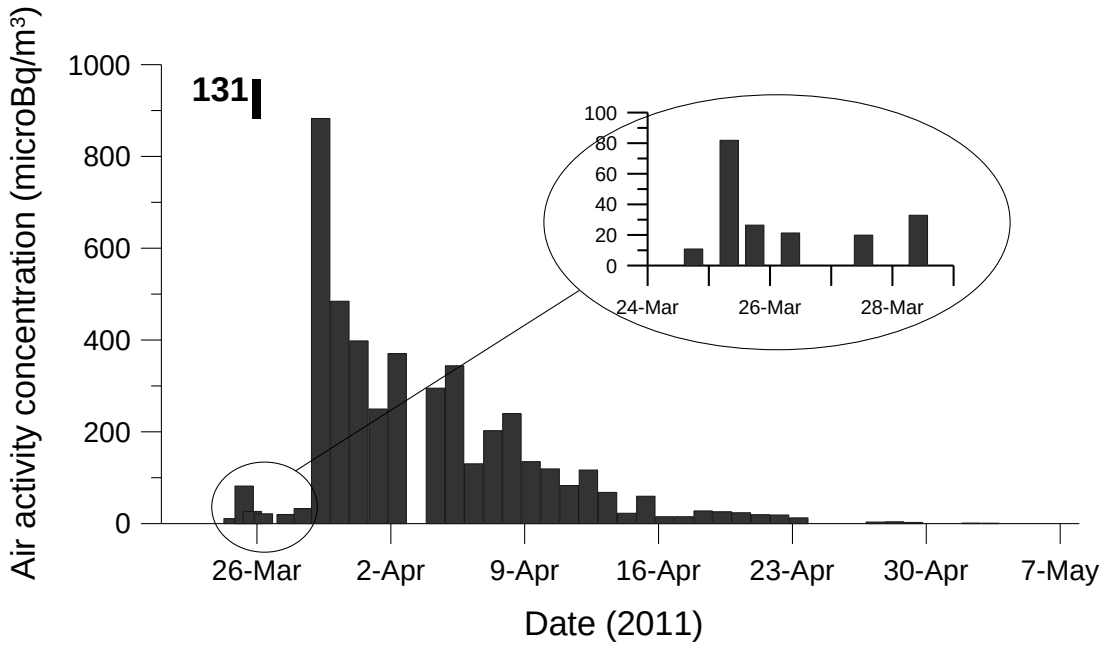

Figure 1: Time dependence of ^{131}I air particulate concentration values.

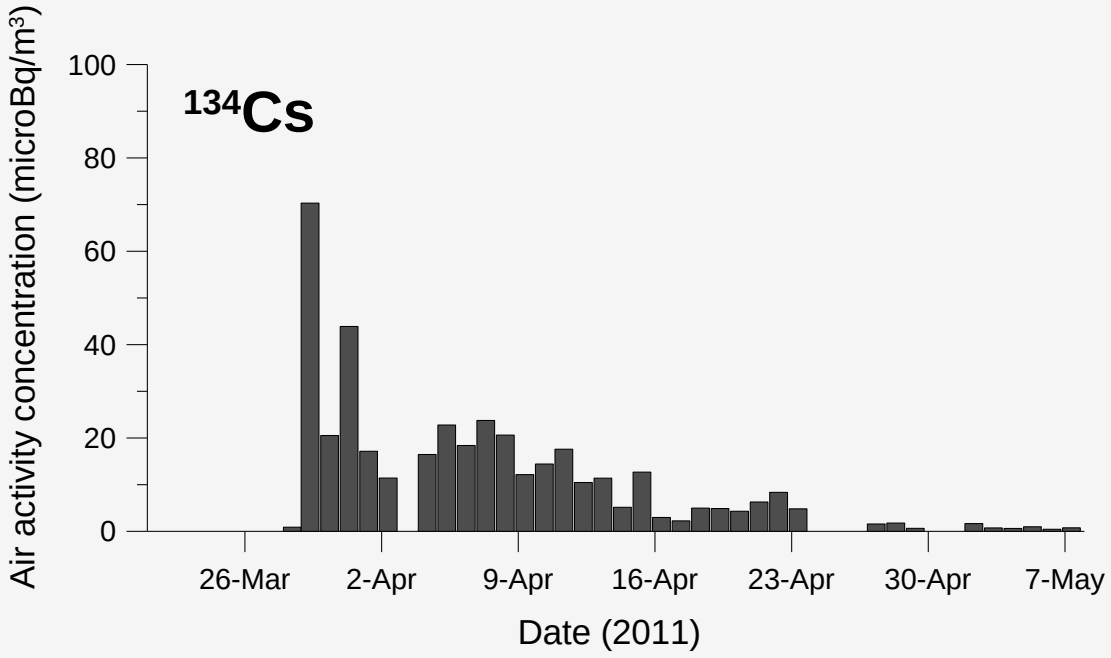

Figure 2: Time dependence of ^{134}Cs air particulate concentration values.

In Figs. 5-8 are given gamma-ray spectra recorded with the HPGe detector ORTEC GEM50195 (relative efficiency 60%) before the first detection (sampling of March 23-24, 2011), the one related to the first detection of artificial radionuclides (sampling of March 24, 2011), the one immediately before the unexpected increase on air concentration values (sampling of March 27-28, 2011) and the last regarding the sample in which were found higher activities and maximum values of airborne concentrations were evaluated (sampling of March 28-29, 2011). In the latter we note the presence of other fission products, as ^{132}I-^{132}Te, no longer detected in other measurements.

The activity ratios for the identified radionuclides can give information about source of radioactive contamination and possibly on the origin of air masses. In fact, as occurred during the measurements following the Chernobyl accident, $^{134}Cs/^{137}Cs$ activity ratio was almost constant if air contamination is due to the release which originates from the nuclear plant. Instead, $^{134}Cs/^{137}Cs$ activity ratio may be affected by a contribution of ^{137}Cs with different origin, such as the residuals of nuclear tests in the atmosphere. In our site, it is known [10] that high levels of ^{137}Cs associated with Saharan dust contaminated by nuclear tests by France in the early sixties of the last century are sampled when is prevalent the sirocco wind (coming from North Africa) [14].

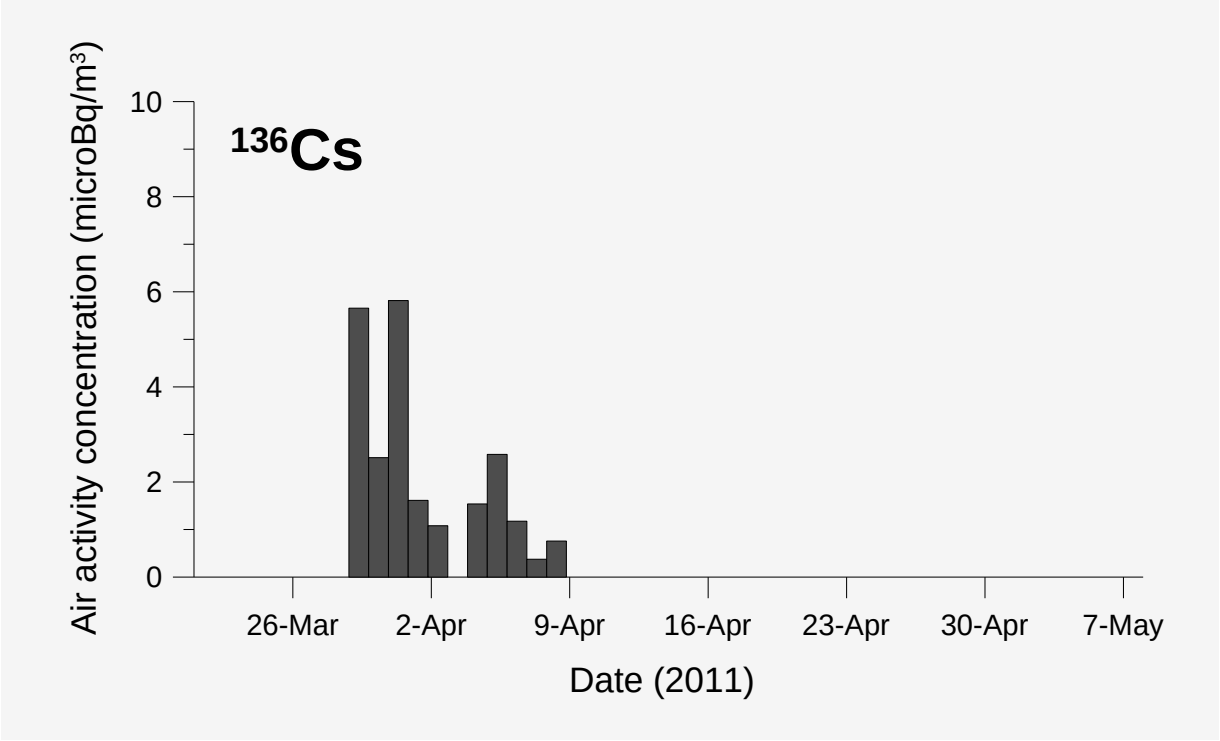

Figure 3: Time dependence of ^{136}Cs air particulate concentration values.

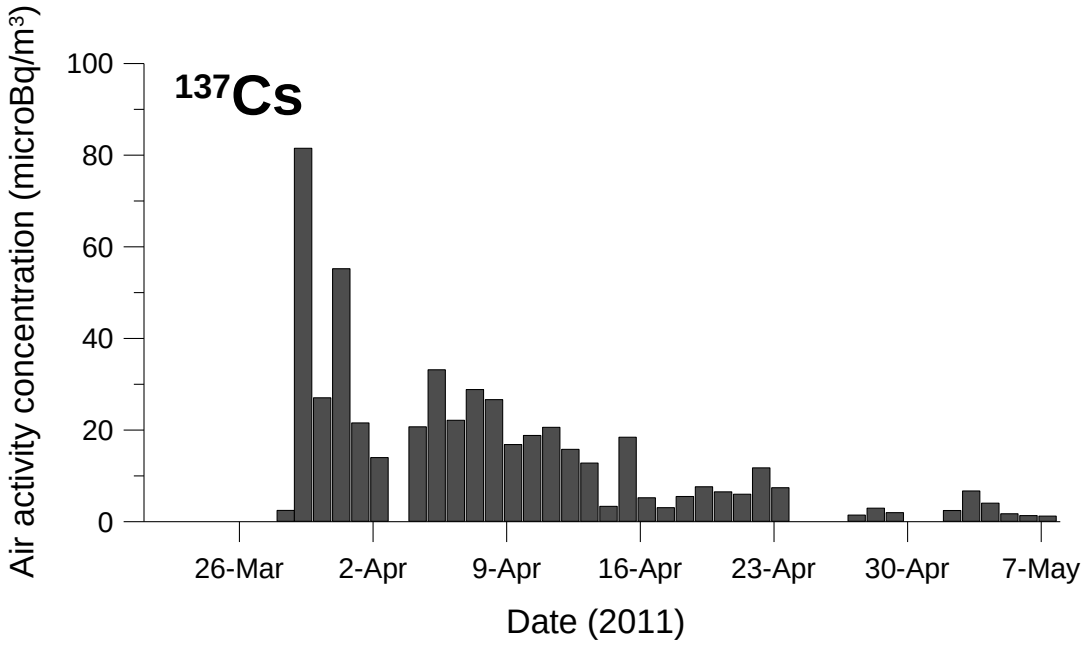

Figure 4: Time dependence of ^{137} Cs air particulate concentration values.

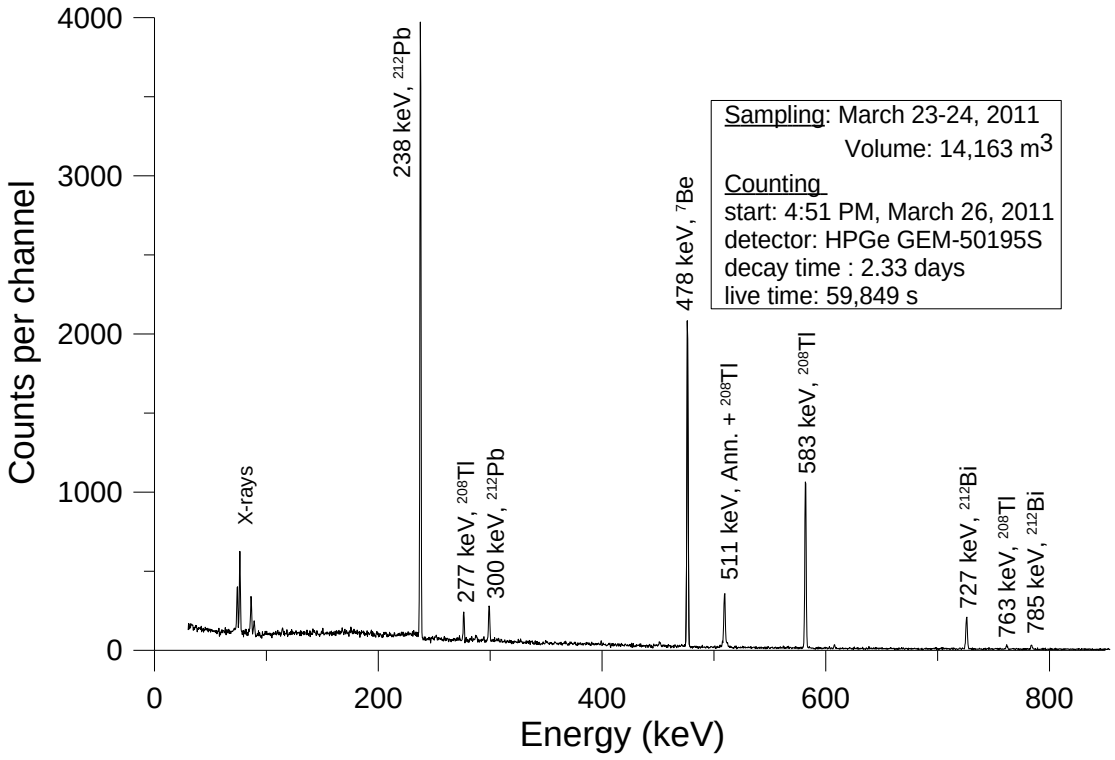

Figure 5: Gamma-ray spectrum detected on the air particulate filter taken during the night between March 23 and March 24, 2011. Photopeaks highlighted are all related to environmental radioactivity.

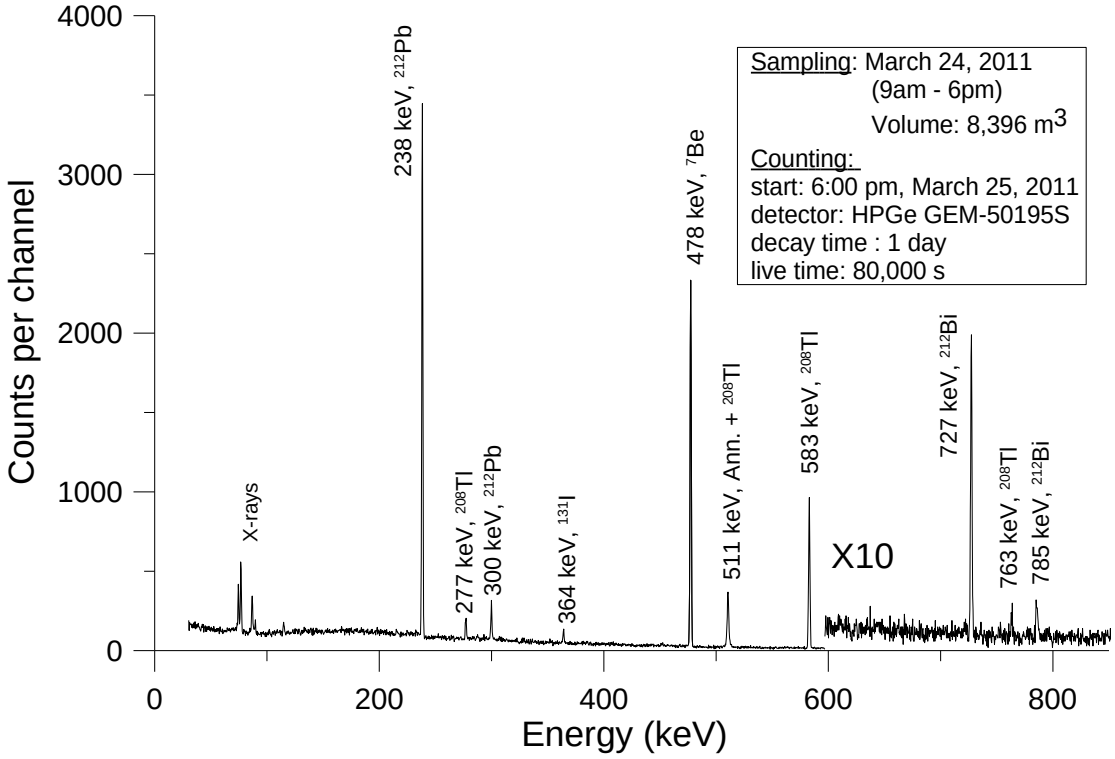

Figure 6: Gamma-ray spectrum detected on the air particulate filter taken during the day on March 24, 2011.

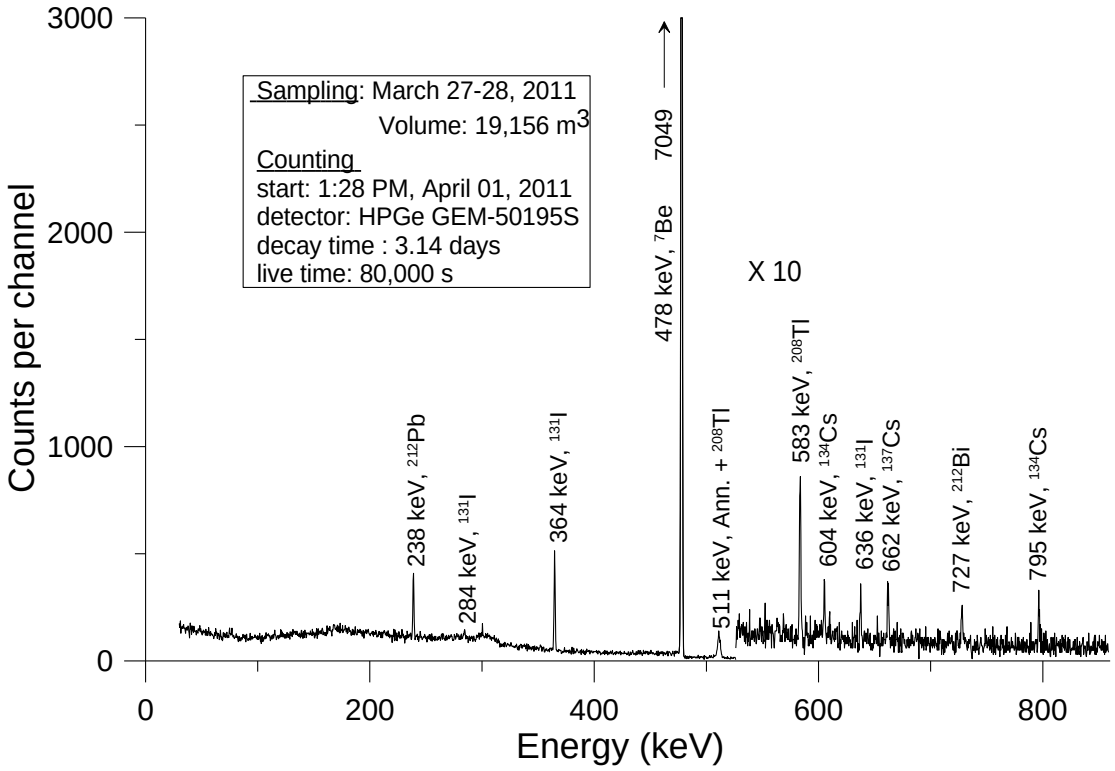

Figure 7: Gamma-ray spectrum detected on the air particulate filter taken during the night between March 27 and March 28, 2011.

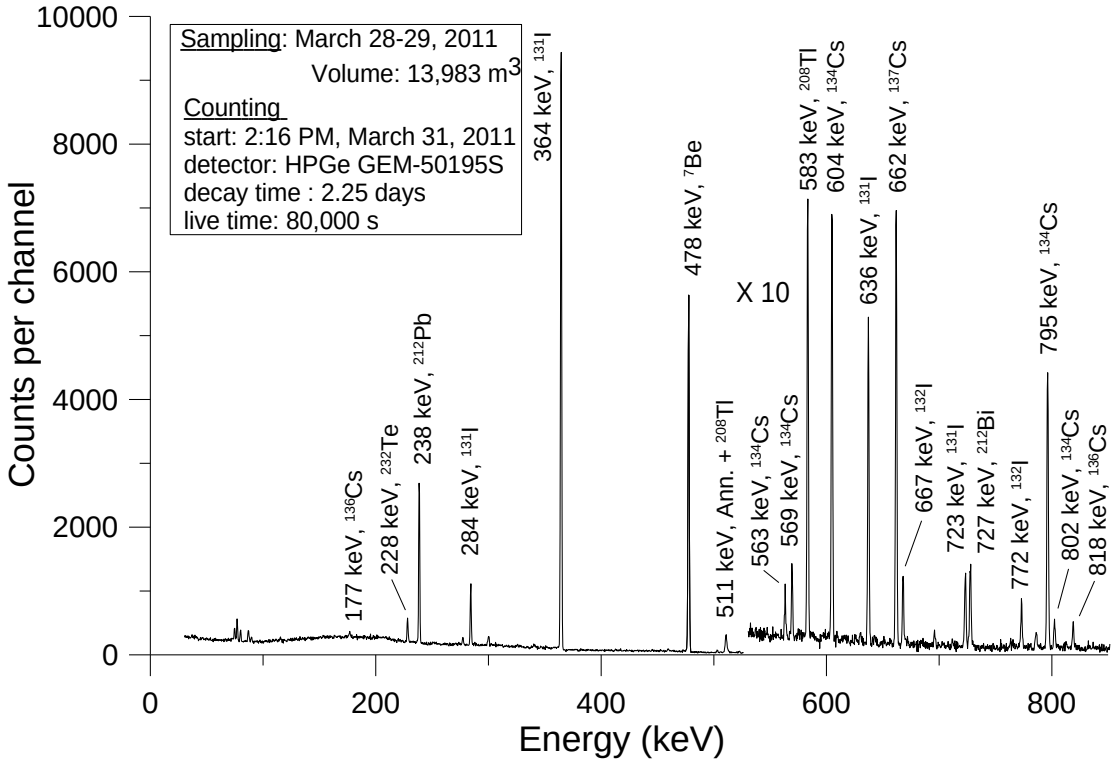

Figure 8: Gamma-ray spectrum detected on the air particulate filter taken during the night between March 28 and March 29, 2011.

Fig. 9 shows the correlation between ^{134}Cs and ^{137}Cs activity on the samples taken after March 28, 2011. The best value of 0.807 was obtained with a linear fit with a correlation coefficient R^2= 0.99. This value is quite different from the one detected after the Chernobyl accident (0.59 [2,3,5]), which can be attributed to different values of burn-up of the two plants at time of the accident. However, if the time dependence of the activity ratio is considered (Fig. 10), we can note a value almost constant and equal, on average, to 0.76 in the period March 25- April 28, 2011. Fluctuating values are registered on March 24 and during April 28 – May 7, 2011 time period. The first low value is probably due to a time difference of arrival and sampling of the cloud, while measurements following April 29 are characterized by a significant amount of residual ^{137}Cs nuclear tests in the Sahara. For example, Fig. 11 shows a gamma-ray spectrum measured on a sample of atmospheric particulate collected between 2 and 3 May 2011 which highlighted a significant difference on peak counting related to main emissions of ^{137}Cs and ^{134}Cs, the last almost comparable with the detection limit. This particulate sample, as those immediately preceding and following, are taken with weather conditions characterized by temperatures higher than the seasonal average and a strong sirocco wind that carries large amounts of dust in the air.

As regards $^{131}I/^{137}Cs$ and $^{136}Cs/^{137}Cs$ activity ratios, Figs. 12 and 13 show the results of a linear best fit to the experimental data, with a value of 9.56 and 0.079 , respectively.

In order to assess population dose, committed effective dose related only to inhalation of particulates was computed with the assumption that the concentration in the air for the whole day is equal to measured value at end of sampling. Dose values were very low, 8×10^{-4} μSv for the ^{131}I (considering only the particulates), 2×10^{-4} μSv for ^{134}Cs and 4×10^{-4} μSv for ^{137}Cs. These values are of no radiological significance. Even if we consider infants (age <1 a), values are still no significant from a radiological point of view, i.e. 8×10^{-3} μSv, 7×10^{-4} μSv and 1.4×10^{-3} μSv respectively.

Finally, ratios of about 1/ 2700 for the ^{131}I (only particulate matter), about 1 / 14000 for ^{134}Cs and 1/22800 for ^{137}Cs were inferred by comparing the maximum concentration values with those found in Palermo after the Chernobyl accident (1986). The maximum value of the ^{137}Cs concentration in air is comparable with the maximum value recorded after the accident at Acerinox plant in Algeciras (Spain) in 1998 [10].

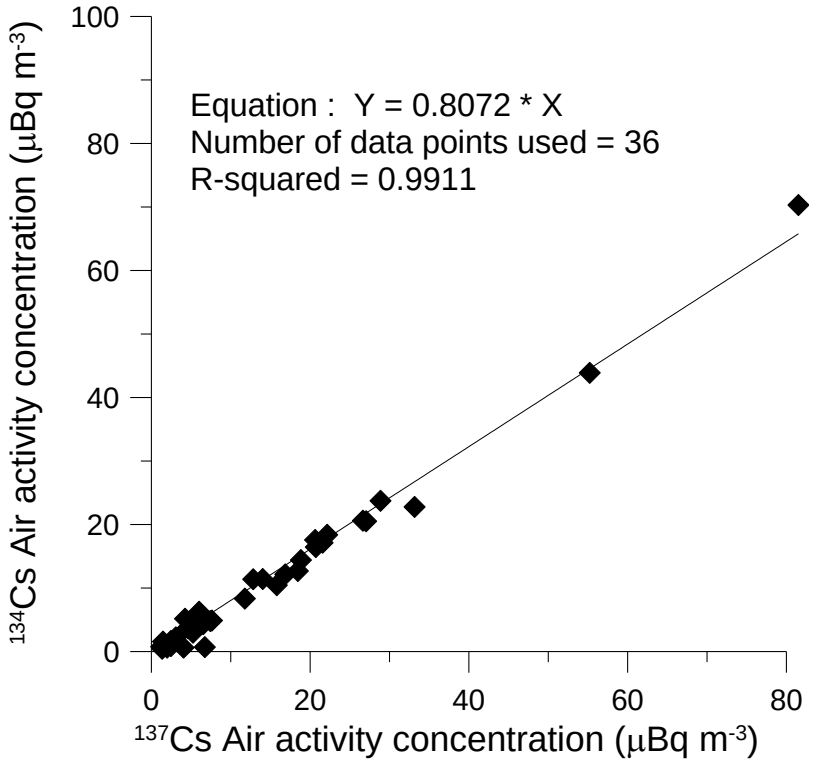

Figure 9: Correlation between ^{134}Cs and ^{137}Cs air activity concentrations .

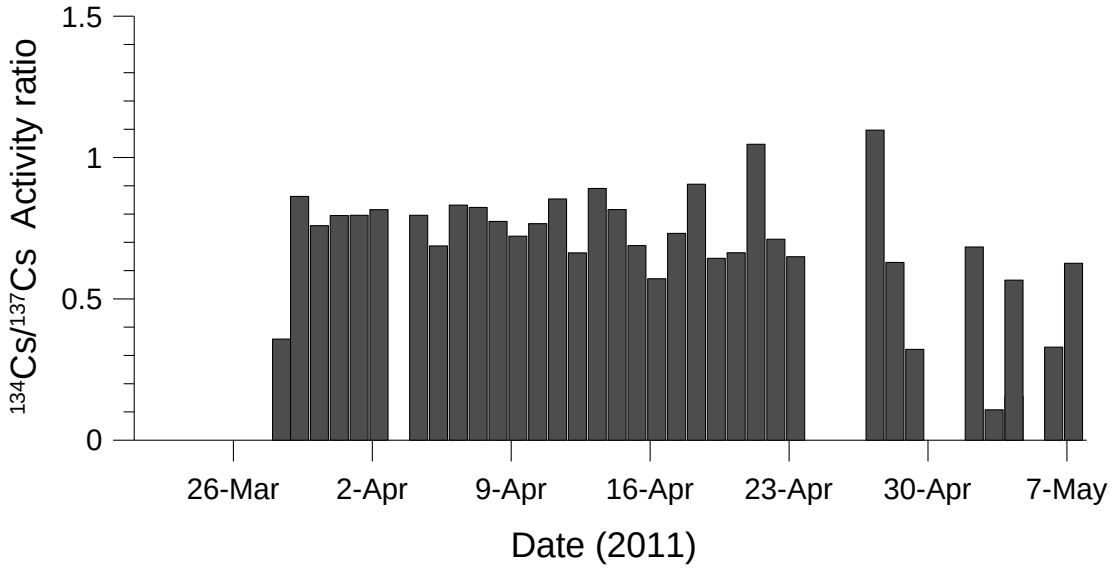

Figure 10: Time dependence of $^{134}Cs/^{137}Cs$ activity ratio.

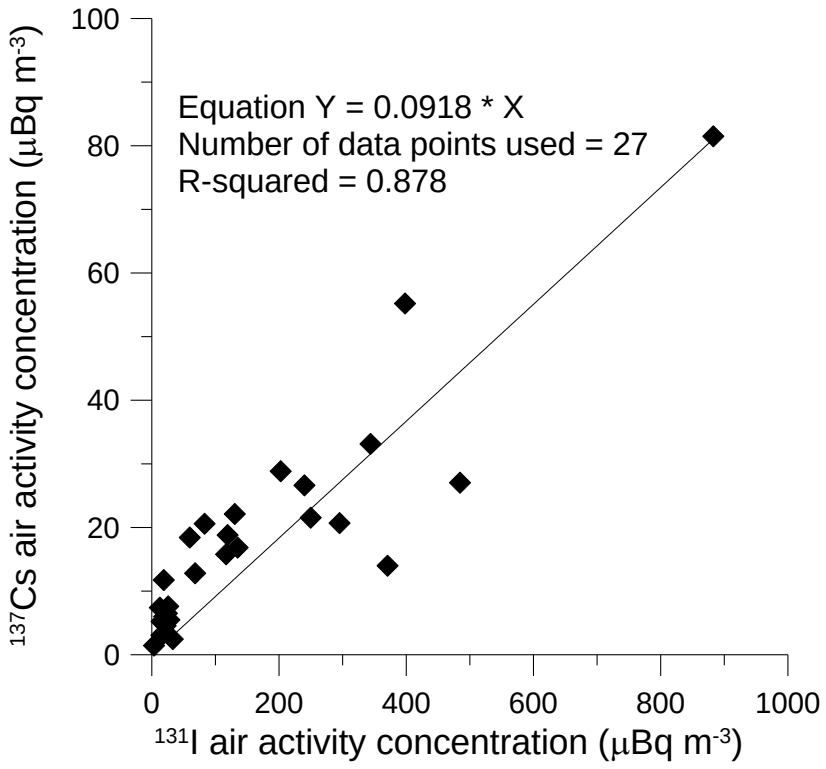

Figure 11: Correlation between ^{131}I (only particulate matter) and ^{137}Cs air activity concentrations.

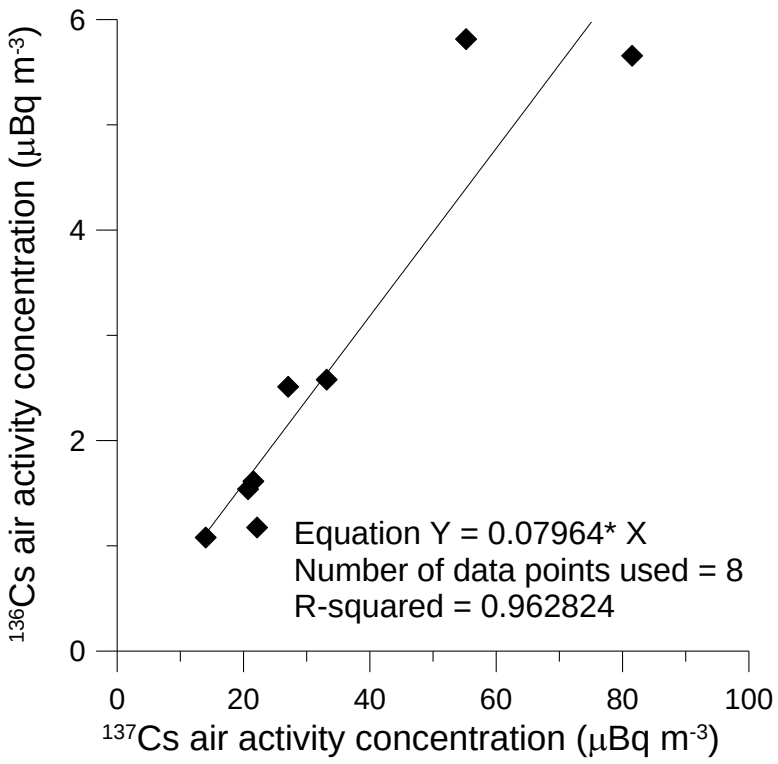

Figure 12: Correlation between ^{136}Cs and ^{137}Cs air activity concentrations .

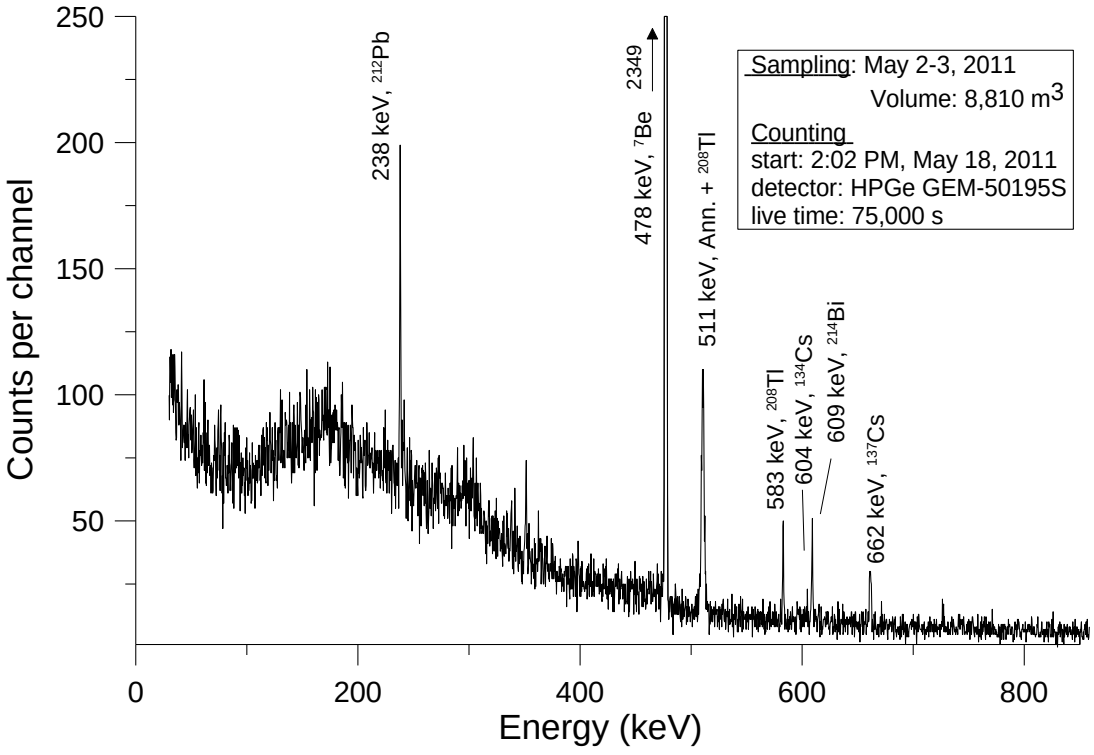

Figure 13: Gamma-ray spectrum detected on the air particulate filter taken during the night between May 2 and May 3, 2011.

4. Conclusion

Following the Fukushima accident resulting the earthquake and tsunami of March 11, 2011 that hit Japan, weak airborne concentrations of ^{131}I, ^{134}Cs, ^{136}Cs and ^{137}Cs were detected in the air of Palermo, Italy. The measured maximum concentrations are of the order of 1/1000 to 1/100000 of those recorded immediately after the Chernobyl accident and are comparable with those highlighted after the accident at Acerinox (Spain) plant. These low concentration values can be measured in air without complex sample preparation only by means of a high volume sampler able to collect radionuclide activity amounts above MDAs of the measurement equipment used. The radioactive contamination of the air of Palermo in the period March - May 2011 was no significant from a radiological point of view and may be expected which has also affected equally the entire Sicily region.

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