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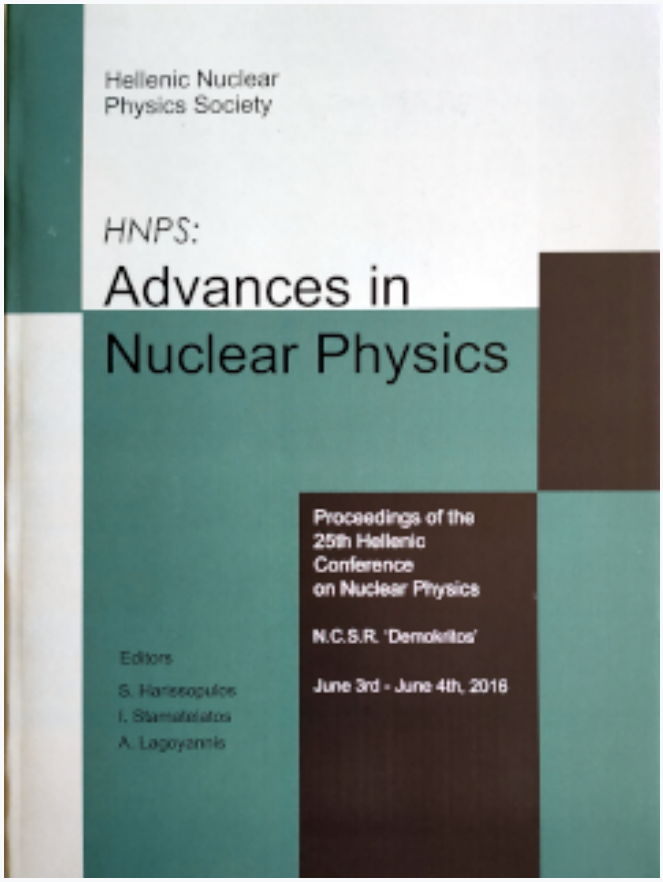

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Activity size distribution of radioactive nuclide ^{7}Be at different locations and under different meteorological conditions

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Abstract The activity size distributions of the natural radionuclide tracer ^{7}Be in different size fractions (<0.4 μm, 0.4-0.7 μm, 0.7-1.1 μm, 1.1-2.1 μm, 2.1-3.1 μm, 3.1-4.2 μm, 4.2-5.8 μm, 5.8-9.0 μm >9.0 μm) were determined at different site places in Northern Italy. Samplings were carried out during the four different seasons of the year 2011. The aim of this work was to define any differences due to the different environments and different meteorological conditions and clarify the main parameters influencing the activity size distribution of radioactive aerosols.

Keywords radioactive aerosols, AMAD, cosmogenic radionuclides, urban pollution.

INTRODUCTION

As well known, the radioactive nuclide ^{7}Be is a cosmic ray produced radionuclide ($t_{1/2}$ = 53.3 d) which is formed in the upper troposphere and lower stratosphere by spallation reactions of light atmospheric nitrogen and oxygen nuclei with cosmic rays (Lal et al. 1958). Once it is formed, it is rapidly associated with submicron aerosol particles, condensing on the non-radioactive species, and participates in the formation and growth of the accumulation mode (from 0.07 to 2 mm) aerosol, which is the major reservoir of pollutants in atmosphere. The aerosol becomes radioactive so the fate of ^{7}Be will become the fate of the carrier aerosols and it can be used as an index of pollution conditions (Papastefanou and Ioannidou 1996). The object of this work is to analyse the pollution in correlation with different environments and meteorological conditions and the influence of the main parameters on the activity size distribution of radioactive aerosol.

STUDY AREAS

Four different locations in the North part of Italy were chosen for sampling: (a) a suburban – industrialised area (Segrate, Milan) – the reference station, (b) an urban area in downtown Milan, (c) a rural-residential area (Ispra), near Maggiore Lake and distant 60 km from Milano, and (d) a rural area at Monte Rosa mountain at 1300 m asl and distant 150 km from Milano. The first station at Segrate has been chosen as a reference station: so each sampling at the other three sampling stations was carried out simultaneously with the sampling at the reference station.

SAMPLING PROCEDURE AND INSTRUMENTATION

Aerosol samplings, for measuring the activity concentration of the short lived cosmogenic radionuclide ^{7}Be ($t_{1/2}$ = 53.3 d) were carried aut in the open air.

The total experimental collection period lasted almost a full year (from February 2011 to December 2011), covering all seasons of the year 2011. The length of each collection period was 7-10 days with regulated air flow rate of about 28.3 L min^{-1} (1cfm) and collected air volume of about 300-400 m^3. The duration of sampling have been chosen in order that the concentrations of ^{7}Be in each size range is above minimum detectable activity. All samplings have been carried out by two compatible 1ACFM 9-stages cascade impactors and with Efficient Cutoff Diameters (ECD) of 0.4, 0.7, 1.1, 2.1, 3.3, 4.7, 5.8 and 9 μm.

In Fig. 1 are reported two cascade impactors during the first stage of the experiment, devoted to verify the compatibility of the two instruments.

Fig. 1. The two impactors used in the experiment during the measurements made to test the reciprocal compatibility.

Acetate cellulose filters were used as collection substrates (0.8 mm pore size Sartorius – Germany of 8.2 cm diameter). No additional coating (oil or grease) was used. As an example, in Fig. 2 is shown a set of filters at the end of collection period.

Meteorological data concerning the temperature T(°C), Relative Humidity (RH%) and amount of precipitation (rain/snow) during each sampling period were obtained from the Meteorological Stations of the Regional Agency for the Environmental Protection, which head the four locations of sampling.

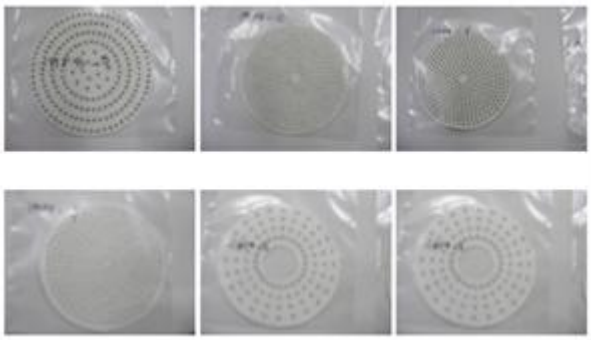

Fig. 2. A set of acetate cellulose filters used in the experiment at the end of collection period.

At the end of the collection procedure, the filters, used as plane sources, were measured for ^{7}Be activity (E_γ = 477 keV) using two high resolution, high relative efficiency (42%), low – background HPGe detectors. The statistical uncertainty ranges from about 12% - 18% for the activities on the impactor stages 5 -9 (finer particles) to about 20 – 30% for the activities on the impactor stages 1 – 4 (coarser particles).

RESULTS AND DISCUSSION

The activity median aerodynamic diameter (AMAD) ranged from 0.40 μm το 1.05 μm during winter period, between 0.50 to 0.73 μm during spring period, between 0.47 to 0.69 μm during summer period and between 0.50 to 0.96 μm during autumn period.

Lower AMAD values are recorded during summer period which in general is characterized by low relative humidity conditions and high temperatures. It seems that during winter period the highest AMAD values are recorded. These results support the results of previous investigations, reported in the literature for entirely different environments and under different meteorological conditions (Ioannidou, 2011, Papastefanou and Ioannidou 1996, Yu and Lee 2002).

In all stations, except the one in Ispra region near the Maggiore Lake, the AMAD values were anticorrelated with ^{7}Be activities as shown in Fig. 3, while they are correlated with RH%, as it is demonstrated in Fig. 4. The observed correlation between the AMAD values and RH% can be due to the intense condensation process during high RH% conditions, resulting in increased particle sizes and higher scavenging rates of aerosols.

In Segrate region and in downtown stations which are located in most polluted environment the AMAD values of ^{7}Be aerosol particles are greater than the less polluted environment and far from industrial activities regions like ISPRA and Monte Rosa mountain. These results combined with the reported PM2.5 and PM10 in the regions of investigation support our assumption that under polluted environment the radioactive aerosol particles are connected with greater aerosol particles. This result confirms that the radioactive nuclides can serve as an index of air polluted conditions.

It can be noticed also that AMAD values in the winter campaigns are greater than those obtained during the summer campaign. During winter time, in fact, the greatest atmospheric pollution phenomena take place (thermal inversion, condensation, fog, etc.), because of both worse climatic conditions and domestic heating.

The data demonstrate also that the pollution rate between the two areas (metropolitan and suburban-industrialised) is similar and ^{7}Be activity concentrations and AMAD values are almost equal.

All measurements show a similar trend in all locations, except rural-residential (Ispra) areas, with higher ^{7}Be concentrations associated to the finest particulate. However, the relative concentrations differ sensibly between the urban-industrialised (Milan and Segrate) and the rural-residential (Ispra) areas, being, the values found in these last, much lower.

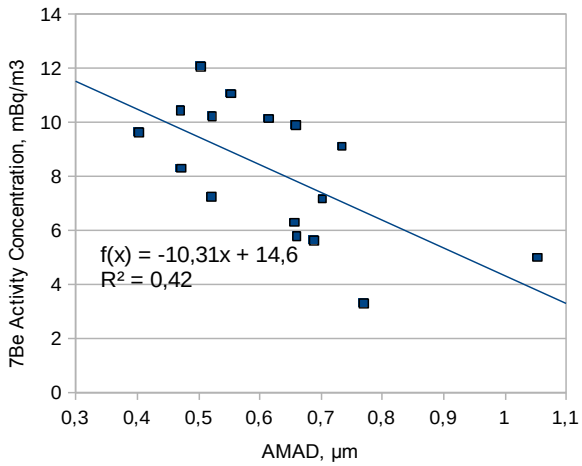

Fig. 3. Anticorrelation relationship between ^{7}Be concentration and AMAD parameter.

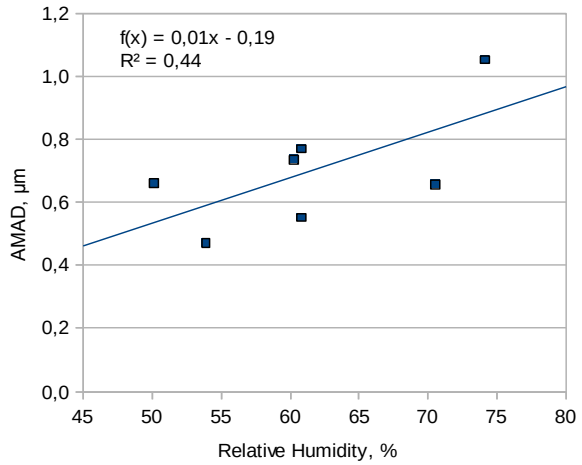

Fig. 4. Correlation relationship between RH% and AMAD parameter.

CONCLUSIONS

To the best of our knowledge this is the first study of activity size distribution of a radioactive nuclide simultaneously at different environments and during four seasons of a year.

In particular the sampled four different locations in north part of Italy were: (a) a suburban – industrialised area (Segrate, Milan) – the reference station, (b) an urban area in

downtown Milan, (c) a rural-residential area (Ispra), near Maggiore lake and (d) a rural area at Monte Rosa mountain (1300 m asl). The results obtained from the measurements of the activity size distribution of ^{7}Be in ambient aerosol at these four different locations demonstrate that the greatest part of ^{7}Be aerosol is associated with fine particulate in all stations except one in Ispra region, probably due to its proximity to Maggiore Lake. As reported in many other publications, also in this study the AMAD values were anticorrelated with ^{7}Be activities while they were correlated with RH%.

Moreover it was found that the two more polluted places – University of Milano and LASA in Segrate – present the same behaviour in respect to the other two cleaner environmental places from the point of view of the AMAD values. In particular in the most polluted environment the AMAD values were greater.

Studying the AMAD values as a function of the seasons, LASA and Milano presented similar values during winter and spring and the greater values during winter, while in LASA was observed the lowest AMAD values during summer. During winter period the AMAD value in ISPRA was the lowest

In this work an attempt to find any association with the possibility to use the activity size distribution of the radioactive nuclide ^{7}Be as an index of air pollutant conditions was made. The obtained results confirm conclusions present in literature, even if this is the first time that the comparison of the behavior for meteorological and pollutant aspect of different places benefits of the ability of a simultaneously sampling.

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