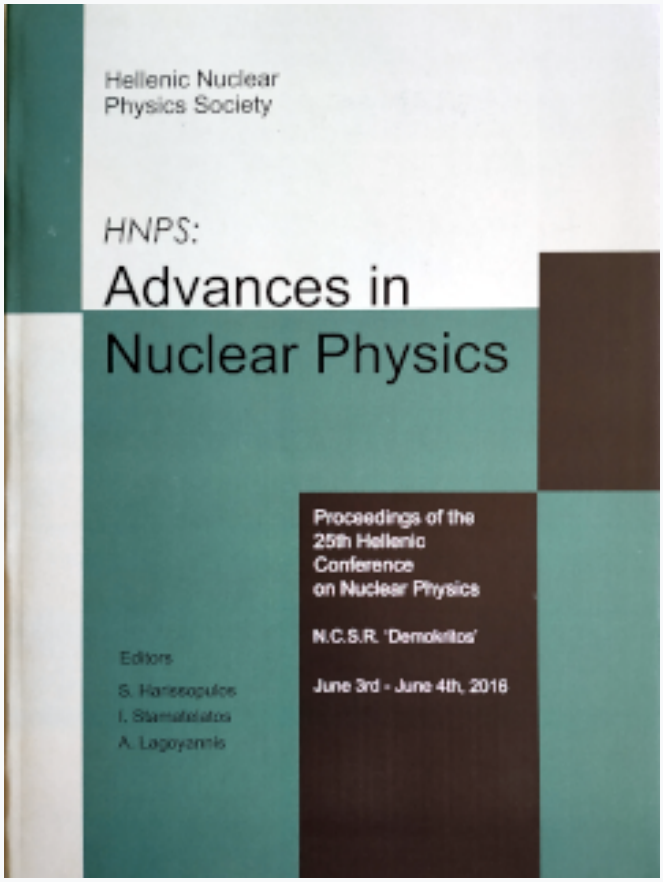
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M. Savva, M. Anagnostakis

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Determination of ^{7}Be, ^{210}Pb and ^{22}Na Activity in Air and Rainwater samples by Gamma-ray Spectroscopy

Marilia Savva*, Marios Anagnostakis

Nuclear Engineering Department, School of Mechanical Engineering
National Technical University of Athens, Iroon Polytexneiou 9, 15780,
Zografou, Athens, Greece

Abstract

The activity concentration of cosmogenic radionuclides in atmospheric aerosol has been used in many applications, such as air mass transport models and solar activity reconstructions. One of the most studied cosmogenic radionuclides is ^{7}Be which is being monitored at the Nuclear Engineering Department of the National Technical University of Athens (NED-NTUA) since many years, with high accuracy and precision. Sodium-22 is another cosmogenic radionuclide met in such studies, however less reported due to its very low activity concentration, of the order of 1 μBq/m3 [1], [2] – roughly four orders of magnitude lower than that of ^{7}Be.

Besides cosmogenic radionuclides, radionuclides of terrestrial origin like the ^{222}Rn progeny ^{210}Pb are also used in atmospheric aerosol studies. Lead-210 serves as an ideal tracer of continental air masses because ^{222}Rn emission from oceans and lakes is negligible (<1%) compared to land sources. However, its deposition exhibits geographic and seasonal variations depending on ^{226}Ra content of the top soils, soil-porosity and meteorology of a particular region [3]. Values reported for ^{210}Pb activity in the air range from 0.1 to 3.4 mBq/m^{3} [4].

During this research, an investigation regarding the detection of ^{210}Pb and ^{22}Na in atmospheric aerosol and the determination of ^{7}Be/^{22}Na ratio has been conducted at NED-NTUA. Sampling and analysis methods were appropriately selected to allow for the detection of all three nuclides in aerosol samples. In order to study the role of precipitation in surface air ^{7}Be, ^{210}Pb and ^{22}Na activity concentration, a procedure for collecting and analyzing rainwater was also developed.

From the results obtained so far it is concluded that with the sampling and analysis procedures followed, ^{210}Pb can be easily determined with reasonable accuracy. As far as ^{22}Na is concerned, it was concluded that it can be detected although with high uncertainty, since its activity concentration is within the limits of detection.

Keywords Aerosol sampling, rainwater sampling, ^{7}Be, ^{22}Na, ^{210}Pb.

INTRODUCTION

Beryllium-7 is a short-lived (half-life: 53.3 d) cosmogenic radionuclide, produced in the upper troposphere (~30%) and lower stratosphere (~70%) by spallation reactions of light atmospheric nuclei (C, N, O) with particles of the primary component of cosmic rays (protons and neutrons) [5], [6]. Sodium-22 with a half-life of 2.602 y is a cosmogenic

* Corresponding author, email: msavva@nuclear.ntua.gr

radionuclide generated in the upper atmosphere by argon spallation reactions with protons and secondary cosmic ray neutrons. The activity ratio of ^{7}Be and ^{22}Na is often used as tracer of stratosphere-troposphere vertical exchange, in global aerosol radionuclide monitoring networks. Both nuclides can be detected by means of gamma spectroscopy through their 477.6 keV (^{7}Be) and 1274.5 keV (^{22}Na) photons. Though the detection of ^{7}Be is relatively easy, ^{22}Na detection is much more difficult since its activity concentration is extremely low - about four orders of magnitude lower than that of ^{7}Be. In addition, the 1274.5 keV photopeak lies on the continuum background produced by Compton scattering of the 1460.8 keV photons emitted by ^{40}K.

Lead-210 with a half-life of 22.3 years is a long lived ^{222}Rn progeny. Since ^{222}Rn is continuously exhaled from the surface soils to the atmosphere, while its emission from oceans and lakes is negligible (<1% compared to land sources [7], [8], [9]), ^{210}Pb serves as an ideal tracer of continental air masses. However, the flux of ^{222}Rn shows significant spatial and temporal variability depending upon ^{226}Ra content of the topsoil, soil-porosity and meteorology of a particular region [10], [11].

The Nuclear Engineering Department of the National Technical University of Athens (NED-NTUA) has established an environmental monitoring program since 1986 [12]. Its main purpose is the continuous monitoring of radioactivity outdoors with emphasis on radon decay products concentrations. Since 2008, the monitoring scheme includes ^{7}Be activity concentration in air and rainwater samples [13]. In this study, the monitoring scheme was extended to include the radionuclide of terrestrial origin ^{210}Pb and the cosmogenic radionuclide ^{22}Na. It should be noted that this kind of data – especially for ^{210}Pb and ^{22}Na – are hard to find in the latitude where NED-NTUA is situated.

MATERIALS AND METHODS

Air sampling

The NED-NTUA is situated at NTUA University Campus in the vicinity of Athens (37º 58′ 41.61′′ N, 23º 47′ 5.58′′E, 195m above sea level). Its environmental monitoring program includes systematic air sampling with a DH-50810E type high volume (~80m^3/h) air sampler, manufactured by F&J Specialty Products, Inc., that is installed on the building roof ~10 m above ground. The air sampler is equipped with sensors that record airflow, atmospheric pressure and temperature during air sampling. High efficiency (>98%) rectangular glass fiber filters 8”x10” (FP10M, F&J Specialty Products, Inc.) are used.

In the framework of this study, 20 air filters were collected and analyzed, covering the period from July 2014 to April 2016, with 4 – 10 days of sampling per month (average sampled volume ~13000 m^3).

Rainwater sampling

Rainwater sampling is another component of the NED-NTUA monitoring program. For the rainwater collection and analysis the following procedure is followed:

- Rainwater is collected in plastic basins that are installed on the NED-NTUA building roof covering an area of about 1 m^2.
- At the end of the rainwater sampling, the basins are rinsed with 1N HNO_3 and rainwater is transfer to holding containers.
- Cation exchange resin is added to the rainwater sample (5 g resin per liter of rainwater.
- The sample is filtered to allow for the retention of the resin, which is then dried for ~5 days using Silica Gel.

In the framework of this study, 12 rainwater samples were collected with monthly interval. Sampling covered the period from September 2014 to March 2015 and volume sampled varied from 6 to 64 L.

Gamma spectroscopic analysis

Two types of sample were analyzed by means of gamma spectroscopy: (i) air filter samples 8"x10" and (ii) cation resin cylindrical volume samples (28.7 cm^3). All samples were analyzed for the determination of ^{7}Be, ^{210}Pb and ^{22}Na activity concentration using their 477.6 keV, 46.5 keV and 1274.5 keV photopeaks, respectively.

The gamma-ray spectrometry system used consists of a closed-end coaxial Extended Range (XtRa) Germanium detector, with a 104.5% relative efficiency, housed in an old steel shield and equipped with a Compton Suppression System [14]. For the determination of ^{7}Be and ^{210}Pb, the suppressed spectrum is used, since it leads to lower peak area uncertainties. On the contrary, ^{22}Na, which is a β^+ emitter suffering coincidence effects, is determined using the normal (unsuppressed) spectrum. Gamma spectrum analysis was performed by the in-house developed code SPUNAL. Details regarding the gamma spectroscopy analysis performed at NED-NTUA can be found elsewhere [15]. In all analyzed samples, appropriate corrections were made for the coincidence phenomenon in ^{22}Na determination.

Calibration of the gamma ray spectroscopy system

For both geometries - air filters and cation resin samples - a numerical method based on Monte Carlo simulation was applied, as certified calibration sources for these geometries were not available. For this purpose the Monte Carlo code PENELOPE was used [16]. For the detector model used in the simulation, the geometrical characteristics provided by the detector manufacturer were adopted, together with Ge dead layer thickness data that were experimentally determined. For the determination of dead layer thickness, an iterative algorithm that was based on the comparison of the experimentally and numerically obtained full energy peak efficiency for two reference geometries was applied [17]. For the detector model that was finally adopted experimental and numerical efficiencies of the reference geometries differ less than 4.0% for photons in the energy region 60 – 1500 keV [18]. The validated detector model was then used for the determination of the photopeak efficiency for the geometries used in this work.

Uncertainty analysis

A realistic assessment of uncertainties is crucial for the determination of low level activity concentrations of ^{7}Be, ^{210}Pb and ^{22}Na. The measurements presented in this work are accompanied by a combined standard uncertainty that can be separated into type A and type B uncertainty components [19]. As far as the type A uncertainties are concerned, the peak area uncertainty that was calculated by the gamma spectroscopic analysis code was considered to be the principal component. In the case of ^{210}Pb, additionally the uncertainty of the peak area in the background spectrum was taken into account.

For the analysis of air filter samples, several potential sources of type B uncertainty have been identified [19]. In this work, the uncertainty of the efficiency calibration procedure and the airflow measurement uncertainty were taken into account. Calibration uncertainty was estimated to be equal to 4.0% (1σ). The airflow measurement uncertainty was estimated less than 1% (1σ) by the manufacturer of the air sampler. All other uncertainty components were considered to have a negligible contribution.

For the analysis of rainwater samples, only sample mass measurement and efficiency calibration were considered as sources of type B uncertainty. Efficiency calibration uncertainty was estimated equal to 4.0% (1σ). Rainwater sample volumes, as well as cation resin samples mass were determined by weighing, with an uncertainty less than 1% (1σ).

For ^{22}Na analysis, the correction for the coincidence phenomenon also introduces an uncertainty of the order of 2.0% (1σ), although negligible, compared with the main uncertainty component which is the peak area uncertainty.

RESULTS AND DISCUSSION

Air sampling

The activity concentration for the radionuclides of interest measured in the 20 air filters collected are presented in Fig. 1, 2 and 3, together with the combined standard uncertainty (1σ). The calculated values for ^{7}Be are in accordance with respective activity concentrations in ground-level air reported in similar latitudes, which vary from 0.32 to 15.3 mBq/m^3 [20], [21], [22], [30]. Accordance is also observed for ^{210}Pb data with respective measurements reported by other researchers: 0.108 – 3.390 mBq/m^3 [4], [22], [30]. It should be noted however that the ^{210}Pb activity determined in the air filter is the total ^{210}Pb activity that includes ^{210}Pb that is produced in the filter from the decay of the short lived radon daughters sampled.

For the analysis of ^{22}Na, in some cases the levels of significance in the γ-spectroscopic analysis code had to be lowered for the photopeak to be analyzed (marked differently in Fig. 3). Lowering the levels of significance leads to the detection of more photopeaks with relatively higher uncertainties. However, even with lower level of significance ^{22}Na could not be detected in 15 samples. For the samples where ^{22}Na was not detected, the low limit of detection was estimated to be around 0.8 μBq/m^3. It should be noted that ^{22}Na measurements

at the same latitude are hard to find. In other studies, the reported concentrations may vary between 0.08 – 1.63 μBq/m^3 [24], [29], [31].

To further study the potential detection of ^{22}Na in atmospheric air, filter samples from three consecutive months were combined to create higher volume samples. The results from this study are given in Fig. 4.

For the samples where ^{22}Na was detected in the individual air samples, the ratio ^{7}Be/^{22}Na was determined and presented in Fig. 5. As reported in the literature, this ratio exhibits an annual variation with high values in autumn and low values in late spring/early summer [24], [25]. In this study, however, this trend is not visible due to the high uncertainties and small amount of data available.

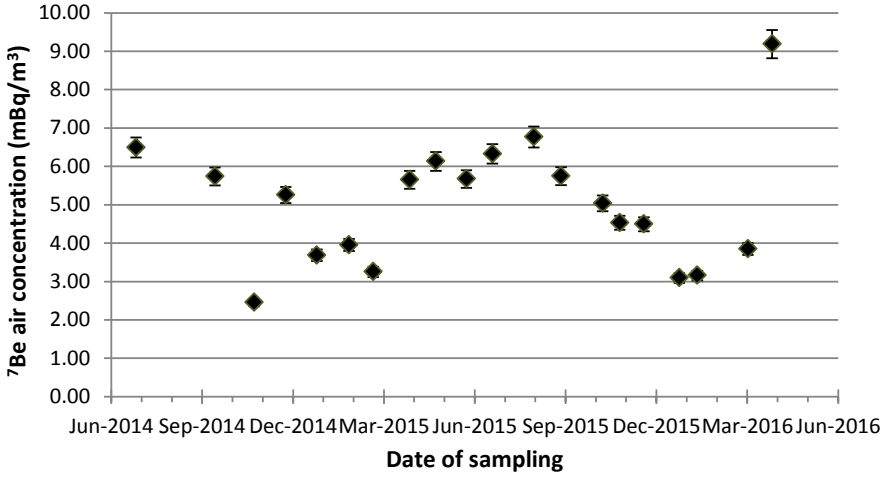


Fig. 1. Beryllium-7 activity concentration measurements in air samples

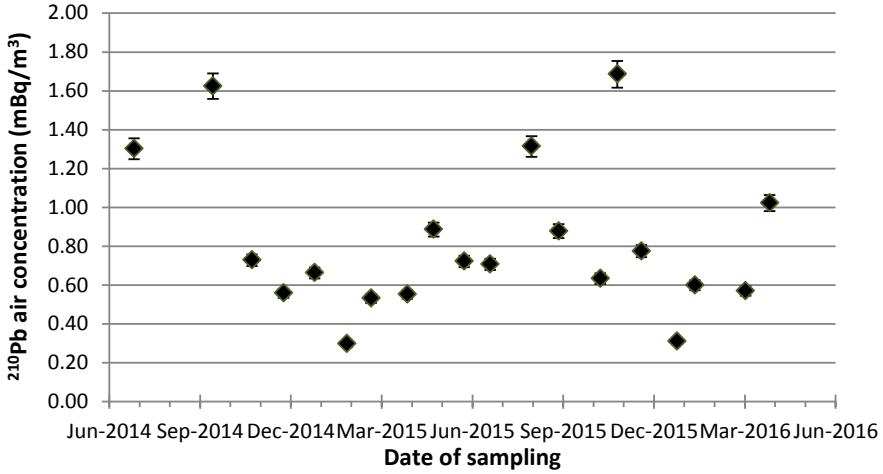


Fig. 2. Lead-210 activity concentration measurements in air samples

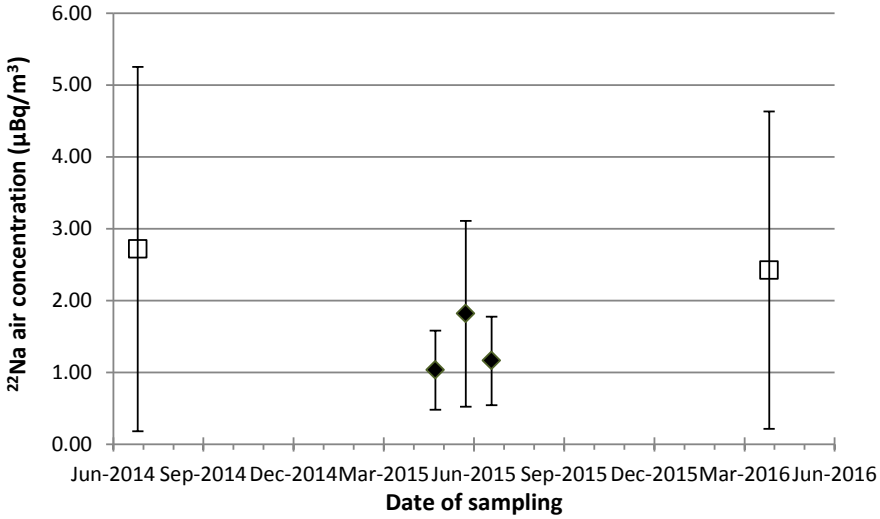


Fig. 3. Sodium-22 activity concentration measurements in air samples (measurements with lower significance levels marked with □)

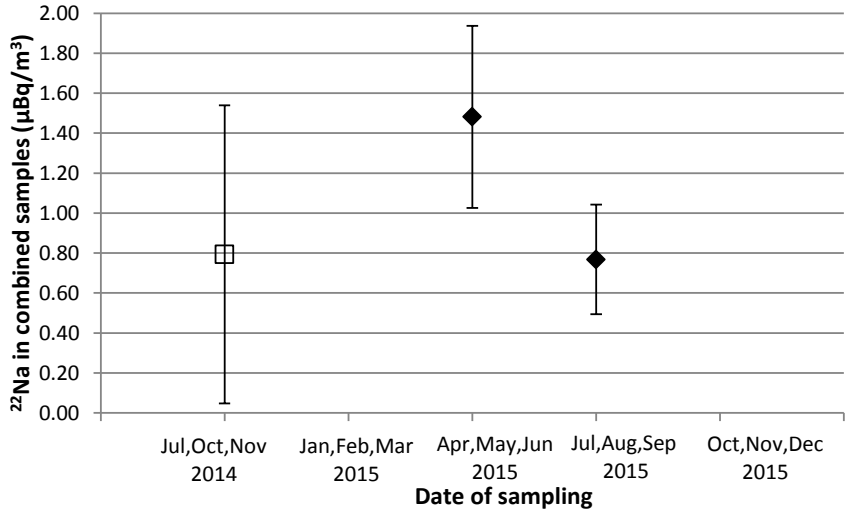


Fig. 4. Sodium-22 activity concentration in combined air samples (measurements with lower significance levels marked with □)

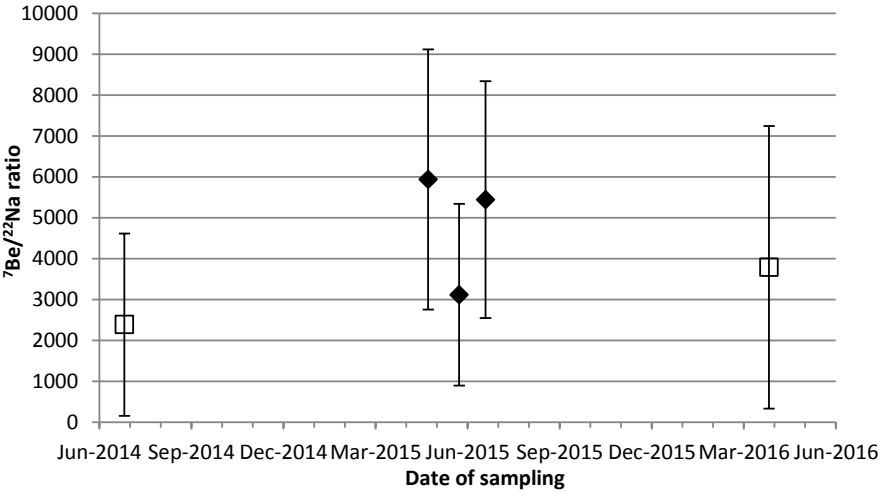


Fig. 5. Ratio of ^{7}Be/^{22}Na activity concentration in air samples (measurements of ^{22}Na with lower significance levels marked with □)

Rainwater sampling

The activity concentration for the radionuclides of interest measured in the rainwater samples that were collected are presented in Fig. 6 and 7, together with the combined standard uncertainty (1σ). The results are consistent with measurements reported by other researchers for both ^{7}Be – 0.2 to 4.1 Bq/L [27], [28] – and ^{210}Pb – 0.06 to 3.0 Bq/L [23], [32].

Sodium-22 was not detected in any rainwater samples, even after lowering the levels significance in the spectrum analysis software. The lower limit of detection for ^{22}Na was estimated to be around 0.6 mBq/L. This value is in accordance with the data reported in the literature in similar altitudes: 0.05 – 0.12 mBq/L [26].

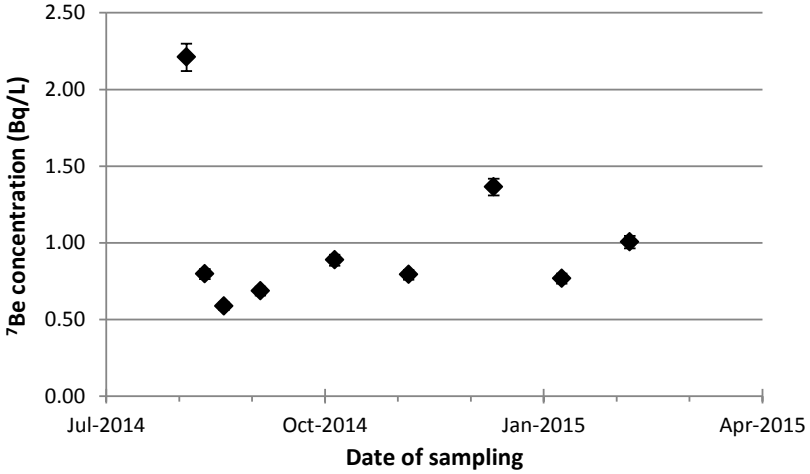


Fig. 6. Beryllium-7 activity concentration measurements in rainwater samples

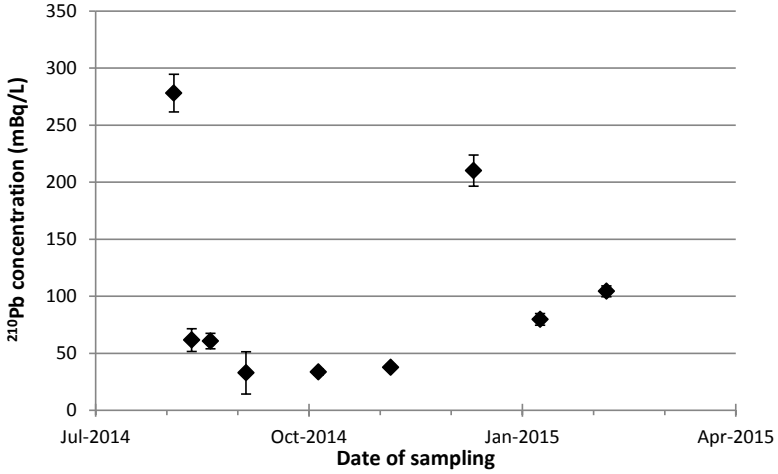


Fig. 7. Lead-210 activity concentration measurements in rainwater samples

CONCLUSIONS

The sampling and analysis procedure developed allowed for the detection of ^{7}Be with combined standard uncertainty of the order of 4% and ^{210}Pb with combined standard uncertainty of the order of 5%. Sodium-22 detection was possible only in warm periods and

higher sampled volumes with high uncertainties (>50%). The lower limit of detection for the method used was estimated to be 0.8 μBq/m^3. As far as the ratio ^{7}Be/^{22}Na is concerned, in this phase of the study conclusions regarding the seasonal variation of this ratio cannot be extracted, due to the few available data.

For rainwater samples, the sampling and analysis procedure developed allowed for the detection of ^{7}Be with combined standard uncertainty of the order of 4% (1σ) and ^{210}Pb with uncertainties from 5% to 22% (1σ). Unfortunately, the procedure followed did not allow for the detection of ^{22}Na. It is therefore concluded that the sampling protocol for water should be modified allowing for the collection of larger samples over longer periods.

From this work it is concluded that the determination of ^{22}Na activity concentration in atmospheric aerosol is possible with the equipment available at NED-NTUA and therefore the NED-NTUA aerosol monitoring program can provide reliable such data that will allow the study of its diurnal variation in the region.

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