MONITORING OF ^{7}Be ATMOSPHERIC ACTIVITY CONCENTRATION USING SHORT TERM MEASUREMENTS

by

Sotiria M. A. PAPANDREOU, Marilia I. SAVVA, Konstantinos L. KARFOPOULOS, Dimitrios J. KARANGELOS, Marios J. ANAGNOSTAKIS*, Simos E. SIMOPOULOS

Nuclear Engineering Department, School of Mechanical Engineering,
National Technical University of Athens, Athens, Greece

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The activity concentration of ^{7}Be in atmospheric aerosol can exhibit seasonal variations due to various physical processes taking place in the troposphere and stratosphere, as well as due to solar activity. An investigation of these variations has been carried out at the Nuclear Engineering Department of the National Technical University of Athens over a two year period (3/2008-4/2010). In the framework of this study, sampling and analysis methods were appropriately selected to allow for the observation of short-term ^{7}Be air activity concentration variations, using a 4-hour sampling interval, while taking in consideration type A and type B uncertainties introduced in the measurements. In order to study the role of precipitation in surface air ^{7}Be activity concentration variations, a procedure for collecting and analyzing rainwater was developed. The techniques used in the present study allowed for the observation of seasonal and diurnal ^{7}Be concentration variations, as well as correlations between ^{7}Be activity concentration and the meteorological parameters of air temperature and relative humidity.

Key words: beryllium-7, diurnal variations, atmospheric aerosol

INTRODUCTION

Beryllium-7 is a short-lived ($T_{1/2}$ = 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) [1, 2]. Its production rate depends on the variations of the geomagnetic field and solar activity (11-year Sun cycle). About 1% of the ^{7}Be produced in the atmosphere is deposited on the surface of the Earth [3-5], as its mean residence time is ~28.2 days in the troposphere [6], reaching up to 2 years in the stratosphere [7, 8].

This radionuclide, due to its convenient half-life, serves as a tracer for the study of vertical and horizontal transfer of air masses, determination of global circulation models [1] , as well as for the study of aerosol particle transport [9]. It can also be very useful for the investigation of atmospheric pollution in case of gaseous pollutant aerosol particles [10, 11].

Seasonal variations of ^{7}Be activity concentration have been observed in several works [12-17] and have been attributed to:

- exchange of air between the stratosphere and troposphere,
- seasonal variations in the rate of vertical mixing within the troposphere,
- seasonal variations in the rate of poleward transport of air masses from middle latitudes, and
- scavenging by precipitation.

In most of these studies, the weekly or monthly average of ^{7}Be activity concentration has been determined.

The Nuclear Engineering Department of the National Technical University of Athens (NED-NTUA) has established an environmental monitoring program since 1986 [18]. Its main purpose is the continuous monitoring of outdoor radioactivity with emphasis on radon decay product concentrations. During the last two years, the monitoring scheme has been extended and modified to include ^{7}Be activity concentration in air and rainwater samples. The use of a short sampling interval, of an order of a few hours, has been of particular interest, as this will enable the monitoring of short-term variations.

The present work gives an overview of the results of this 2-year monitoring program and an investigation in correlations corresponding with meteorolog-

* Corresponding author; e-mail: managno@nuclear.ntua.gr

ical and radiological parameters. A study of the diurnal variations of ^{7}Be concentration is also presented.

INSTRUMENTS AND METHODS

Air sampling

The NED-NTUA is situated at the NTUA University campus in the vicinity of Athens (37º58'41.61" N, 23º47'5.58" E, 195 m above sea level). The NED--NTUA environmental monitoring program includes systematic air sampling with a DH-50810E type high volume (~80 m^3/h) air sampler, manufactured by F&J Specialty Products, Inc., permanently installed on the laboratory 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 borosilicate glass fiber filters 8" 10" (FP10M, F&J Specialty Products, Inc.) are used. The duration of air sampling for the determination of ^{7}Be activity concentration is 4 hours. Upon the termination of the sampling, the collected filter is analyzed by gamma spectrometry, according to the procedure described in the section on gamma spectroscopic analysis.

Within the framework of this study, 93 air filters were collected and analyzed for monitoring seasonal ^{7}Be variations and for investigating the correlation between ^{7}Be activity concentration and meteorological parameters. The sampling covered the period from March 2008 to April 2010, with approximately one 4-hour sample collected per week. Sampling start time was between 8:00-10:00 a. m., therefore all samples refer to the same time period during the day. In addition, 3 sets of 7 sequential filters were collected during two typical summer days (03/07/2008 and 17/07/2008) and during a single rainy spring day (13/4/2009), in order to study the ^{7}Be diurnal variations.

Rainwater sampling

Rainwater sampling is another component of the NED-NTUA ^{7}Be monitoring program. The following procedure, developed by the NED-NTUA, based on standard rainwater sampling techniques [19], is used for rainwater collection and analysis:

– during rainfall, rainwater is collected in plastic basins installed on the NED-NTUA building roof covering an area of about 1 m^2, while rainfall height is simultaneously being monitored,
– after the rainfall event, the sampling basins are rinsed with 1N HNO_3 and the rainwater is transferred to holding containers,
– the rainwater is then filtered through 90 mm diameter Whatman no. 42 paper filters in a Buchner funnel, rinsing the containers with 1N HNO_3. One filter per 2 L of water is used. The collected filters are stacked together and analyzed by gamma spectrometry to determine the ^{7}Be activity concentration in suspended particulates, and
– the filtrate is evaporated at a low temperature to a volume of less than 282 cm^3 and transferred to the standard 282 cm^3 container, used at NED-NTUA for gamma-spectroscopic analysis. Distilled water is used to rinse the filtrate container.

Two different samples are produced from each rainfall collection by applying this procedure: a "filter sample" and a "volume sample". In both samples, ^{7}Be activity concentration is determined by gamma spectrometry. The activity concentration of the collected rainwater is then calculated by adding the concentrations determined from both the filter and volume samples.

This dual sample approach is followed in order to minimize the analytical error due to sample inhomogeneity during gamma spectroscopic analysis, as any particles present in an unfiltered water sample will tend to settle down to the bottom of the container. The geometric efficiency for this thin layer of particles will be considerably greater than that of a homogeneous source used for volume geometry calibrations, possibly leading to an overestimation of the activity for radionuclides that are preferentially attached to particulate matter.

The processing of samples with volumes up to 20 L can be performed with minimal effort within a few days following this procedure. Sample volumes can, in principle, be increased by increasing the size of the rainwater collector. However, the large number of filters required makes gamma spectroscopic analysis impractical. A total of 6 rainfall events were studied during this study by using the said procedure, with sample volumes ranging from less than 1 L to up to 17 L.

Gamma spectroscopic analysis

For the gamma-spectroscopic analysis of the samples collected during this study three types of samples were used: (1) an air filter sample of 8" 10", (2) a 90 mm filter for rain water filtration, and (3) a cylindrical volume sample (282 cm^3) for the analysis of the rainwater filtrate. All samples were analyzed for ^{7}Be activity concentration determination, using the 477.6 keV photopeak, while air filters were also used for radon progeny activity concentration when possible, using the 295.2 keV and 351.9 keV (^{214}Pb), and 609.3 keV, 1120.3 keV, and 1764.5 keV (^{214}Bi) photons.

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 standard signal processing electronics. 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 [20].

The analysis of the 8" 10" air filters was carried out in two steps:

– *determination of radon progeny activity concentration*. Immediately upon the termination of the sampling procedure, filters are folded to produce a rectangular, 4" 5" geometry. The folded filters are then measured for 2 hours and the activity concentration of the short-lived radon progeny is determined. The results of the radon progeny count are beyond the scope of this work and are not presented in the study, and
– *determination of the ^{7}Be activity concentration*. After the 2 hours analysis, a second spectrum is collected over an 8-18 hours interval. During the initial 2 hours of analysis, radon progeny has almost completely decayed, allowing for better ^{7}Be detection conditions in terms of the continuous background, lower limit of detection and measurement uncertainty (type A). For this spectroscopic analysis, the filter is folded to a rectangular geometry of 6.77 cm 6.35 cm 0.65 cm and packed in a container made of plexiglas, as shown in fig. 1, so as to improve filter positioning repeatability and detection efficiency for the ^{7}Be photons.

Using this geometry, a full energy peak efficiency of 0.075 0.003 (1) is attained for the 477 keV ^{7}Be photons. This value is 40% higher compared to the rectangular 4" 5" geometry, allowing for a lower measurement uncertainty for the same analysis time. With normal air ^{7}Be activity concentration levels (5-6 mBq/m^{3}), an 8 hours spectroscopic analysis is sufficient for the ^{7}Be measurement uncertainty better than 10% (1).

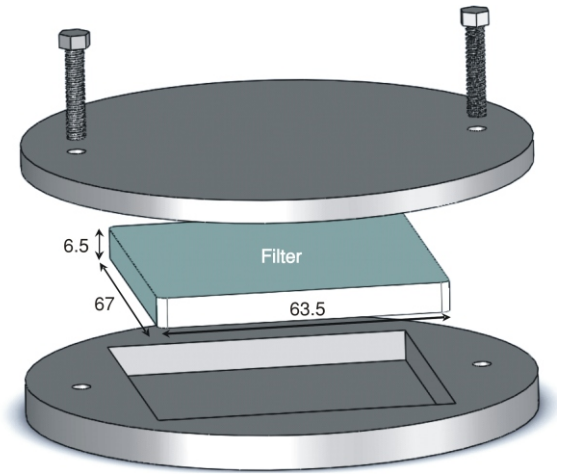

Figure 1. The geometry used for air filter analysis (all dimensions are given in mm)

As for the 90 mm filter sample and the volume sample produced from the rainwater sampling procedure, the spectroscopic analysis is quite straightforward.

Calibration of the gamma ray spectrometry system

Different calibration methods were applied for the different sample geometries used, depending on the availability of certified calibration sources.

As for the volume geometry used for the rainwater spectroscopic analysis, the detector system was experimentally calibrated by means of a certified Amersham QCY.48 mixed nuclide solution. For the 4" 5" filter geometry, laboratory calibration sources were applied. For the geometry presented in fig. 1, a numerical method based on Monte Carlo simulations was used, as certified calibration sources for this geometry were not available. For this purpose, the Monte Carlo code PENELOPE was used [21]. For the detector model, the geometrical characteristics provided by the detector manufacturer were used together with Ge dead layer thickness data that were experimentally determined. For the determination of dead layer thickness, an iterative algorithm based on the comparison of experimental and numerical full energy peak efficiency was applied. These efficiencies were determined for two reference geometries that approximate the filter geometries used in the present study, namely: (1) a cylindrical volume source (height: 10 mm, diameter: 72 mm) filled with a QCY.48 mixed nuclide solution and (2) an IAEA certified filter source (diameter: 47 mm), spiked with a mixed radionuclide solution. For the detector model that was finally adopted, experimental and numerical efficiencies of the reference geometries do not differ by more than 2.5% for photons in the energy region 279 keV to 850keV. The validated detector model was then used for the determination of 477.6 keV photons (^{7}Be) peak efficiency for filter geometries used in this work.

Uncertainty analysis

A realistic assessment of uncertainties is crucial for the identification of variations in ^{7}Be activity concentration. The measurements performed in the present work are accompanied by a total combined uncertainty that can be separated in type A and type B uncertainty components [22]. As far as 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.

For the analysis of air filter samples, several potential sources of type B uncertainty have been identified [22]. In the present work, the uncertainty of the efficiency calibration procedure and the airflow

measurement uncertainty were taken into account. Calibration uncertainty, including the calibration source activity uncertainty, was estimated equal to 4% (1). The airflow measurement uncertainty was estimated to be less than 1% (1) 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 volume measurement and efficiency calibration were considered as sources of type B uncertainty. Efficiency calibration uncertainty was estimated equal to 4% (1) for the filter sources, as described previously, and equal to 3.2% (1) for the volume sources. Rainwater sample volumes were determined by weighing, with an uncertainty less than 1% (1).

It should be noted that, when comparing individual measurements that have been produced by the same measuring apparatus, type B uncertainty components will be correlated and should not be taken in account. Therefore, only type A should be taken in consideration. A simple method for the identification of variations in ^{7}Be activity concentration is by comparing individual measurements using the *u*-score, calculated as [23]

$$u\text{-score} \quad \frac{|\text{Value}_2 \quad \text{Value}_1|}{\sqrt{\text{unc}_1^2 \quad \text{unc}_2^2}} \qquad (1)$$

In this formula, Value_1 and Value_2 are the individual measurements under comparison and unc_1 and unc_2, the corresponding type A uncertainties at the 1 level. The measurements are considered as statistically different when *u*-score is <1.96 at a 95% confidence level.

RESULTS AND DISCUSSION

Seasonal variations

The ^{7}Be activity concentrations measured in the 93 air filters collected are presented in fig. 2, together with measurement (type A) uncertainty (1).

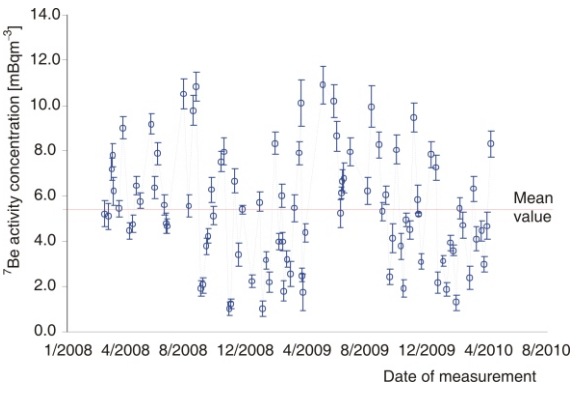

Figure 2. Beryllium-7 air activity concentration for a two year period

The mean value of ^{7}Be activity concentration measurements is 5.4 mBq/m^3, standard deviation 2.5 mBq/m^3, and range is from 1.0 0.4 mBq/m^3 to 10.9 0.8 mBq/m^3. These values are consistent with ^{7}Be activity concentrations in ground-level air reported by long-term studies using large sampling intervals performed in similar latitudes [13, 16, 24, 25, 26].

A seasonal trend is apparent in the data presented in fig. 2, with concentrations reaching a maximum during the summer months and a minimum in the winter months. This trend is made clearer in fig. 3, where data from the present study are compared to data obtained by other researchers at the same latitude (~38 °N) on a monthly average basis [16, 24].

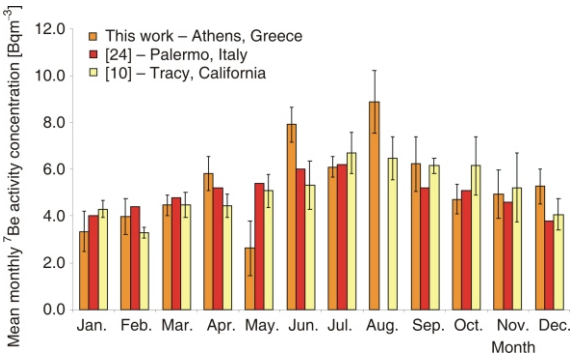

Figure 3. Mean monthly activity concentration of ^{7}Be

Mean concentrations of all samples collected for each month of the year throughout the duration of the project are compared with values from literature in fig. 3. It can be seen that seasonal variations similar to those reported in this work have also been observed in Tracy, California, and Palermo, Italy, and have been attributed to the higher vertical air-mixing rate within the troposphere [16, 17, 24] and the elevation of tropopause during the warm summer months [27].

Increased ^{7}Be scavenging by precipitation has also been proposed as a mechanism to explain the seasonal minima observed during the winter months. The short time scale technique applied in the present work is well suited for demonstrating this effect, as shown in fig. 4, presenting the ^{7}Be activity concentration for a two-month period (15/08/2008 to 24/10/2008), together with the local daily rainfall height (as obtained from the NTUA meteorological station*).

Measurements taken after a rainfall event indicate lower ^{7}Be concentrations, which is to be expected

* The automatic meteorological station of NTUA is maintained by the ITIA Research Team from the Department of Water Resources and Environmental Engineering of the School of Civil Engineering of NTUA

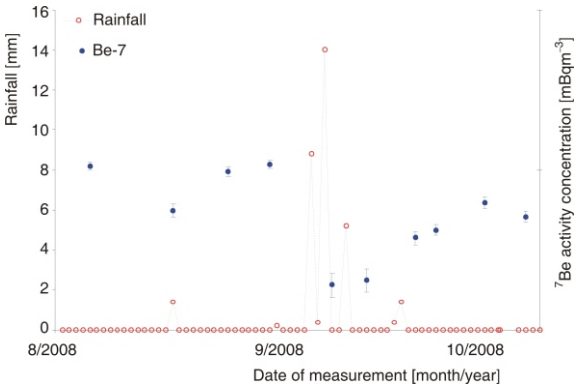

Figure 4. The effect of wet deposition on ^{7}Be activity concentration

due to atmospheric rinsing, while periods without rainfall lead to a gradual increase of measured ^{7}Be concentrations.

In a similar way, it can be seen in fig. 3 that the highest concentrations and the lowest standard deviation values of ^{7}Be are obtained during summer. Furthermore, small standard deviations indicate stability in the measured values as a result of negligible precipitation during these months.

Correlation between ^{7}Be activity concentration and meteorological parameters

Possible correlations between the activity concentration of ^{7}Be and meteorological parameters: air temperature, atmospheric pressure, and relative humidity during measurement are examined in figures 5-7. The main advantage of the sampling procedure used at NED-NTUA compared to other studies is that the sampling time scale is compatible with the time scale of meteorological parameters variations.

From figs. 5-7 it can be concluded that a correlation does exist between ^{7}Be concentration and temperature and between ^{7}Be concentration and relative humidity. The linear correlation coefficients (r) are 0.57 (t_0 = 6.6 for 91 degrees of freedom) and –0.45 (t_0 = = –4.8 for 91 degrees of freedom), respectively. A double-sided student test was applied in order to test the hypotheses

$$H_0: \rho \quad 0$$
$$H_1: \rho \quad 0$$

The critical values at the 95%, and 99% levels are equal to $t_{0.95} = 1.99$, and $t_{0.99} = 2.63$, respectively, therefore the null hypothesis is rejected in both cases. It should be noted that the correlation between ^{7}Be concentration and relative humidity is negative, *i. e.* the concentration decreases with increasing humidity. This can probably be attributed to the low concentrations observed after a rainfall event, when relative humidity is high. No correlation between ^{7}Be activity concentration and atmospheric pressure was observed, as the linear correlation coefficient r is not different from 0 to a statistically significant degree ($r = 0.11$, $t_0 = 1.06$ for 91 degrees of freedom). These conclusions are consistent with other studies [9, 27].

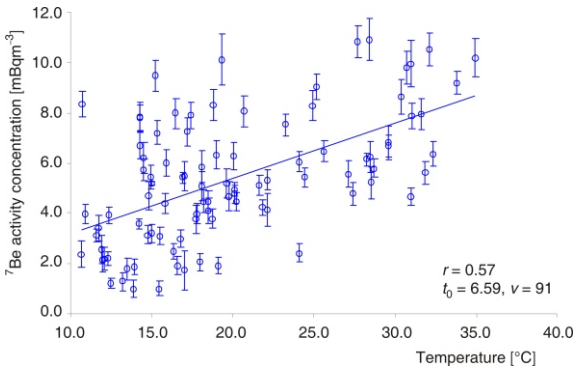

Figure 5. Beryllium-7 activity concentration *vs.* air temperature

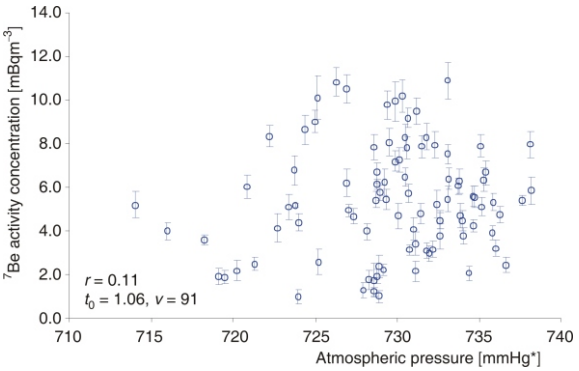

Figure 6. Beryllium-7 activity concentration *vs.* atmospheric pressure
*1 mm Hg 133.322 Pa

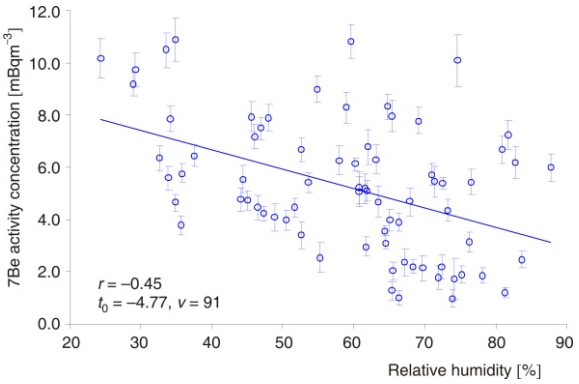

Figure 7. Beryllium-7 activity concentration *vs.* relative humidity

Correlation between ^{7}Be activity concentration and solar activity

In order to study the effect of solar activity on the measurements conducted in this work, the correlation between the ^{7}Be average monthly activity concentration and the monthly number of sunspots* is examined in fig. 8. The calculated linear correlation coefficient r is statistically equal to 0 and thus no correlation can be concluded, though correlation between the ^{7}Be activity concentration and the number of sunspots has been well established by studies extending to longer time periods [24]. This lack of correlation should be attributed to the short time period (2 years) covered by the present study, which represent only a small fraction of the 11-year solar cycle. Furthermore, the time period covered by this study corresponds to the beginning of the solar cycle following the 2008 minimum in the number of sunspots, when ^{7}Be concentration in the atmosphere is at a maximum.

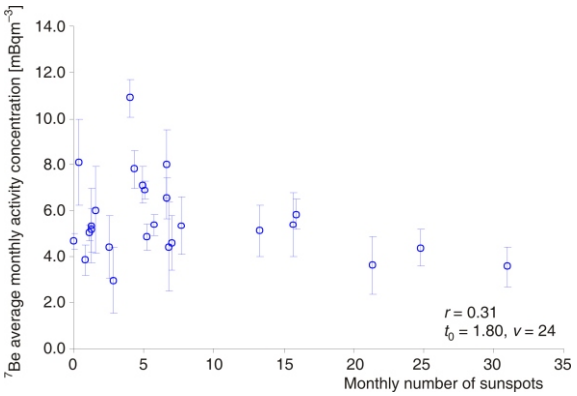

Figure 8. Correlation between ^{7}Be mean monthly concentration and monthly number of sunspots

Diurnal variations

In order to study the diurnal variations of ^{7}Be concentration, three sets of measurements were conducted: two on typical summer days and one on a rainy spring day. Each set of measurements consisted of 7 consecutive air filters.

The results of these three sets of measurements are presented in fig. 9, together with their type A uncertainty (1).

From the measurements presented in fig. 9 it can be easily concluded that a diurnal variation is observable during a typical summer day. This variation is confirmed by performing u-tests using formula (1). The small number of time profiles does not allow for

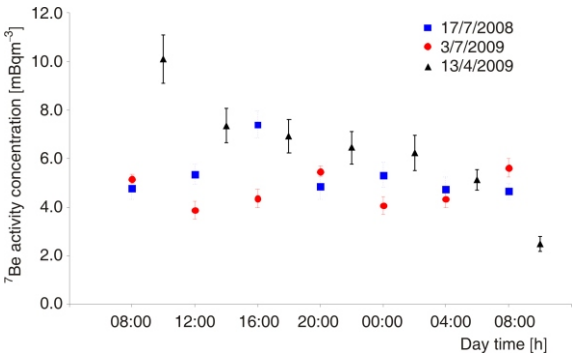

Figure 9. Diurnal variations of ^{7}Be activity concentration

any definite conclusions or trends of ^{7}Be concentration for the time period examined. For the rainy day time profile (13/04/09), it should be noted that the rain started during the last filter sampling. The variation is more intense during this set of measurements – as expected – due to rainout and washout effects.

Rainwater samples

Six rainfall events were studied in this work. The results of these measurements are presented in tab. 1. The table includes details on the sampling date, rainfall height, sampling time, mass of the collected water and ^{7}Be activity concentration measured in both the filter and volume sources, according to the procedure described in the section on rainwater sampling. Activity concentration is accompanied by its type A uncertainty (1).

It is obvious that ^{7}Be is detected in both samples (filter sample and filtrate volume sample), meaning that ^{7}Be activity concentration in suspended particulates is a crucial factor of the measurement. In cases when rain-

Table 1. Beryllium-7 activity concentration measured in rainwater samples

Sampling date	Rainfall height [mm]	Sampling time [h]	Total sample volume [L]	^{7}Be activity concentration	
				Filter sample [BgL^{-1}]	Volume sample [BqL^{-1}]
7-13/Feb/2009	19.1	144	17.5	3.24 0.06	Not detected*
14/Apr/2009	2.2	1	2.0	0.41 0.05	0.2
15/Apr/2009	3.1	1	2.8	0.48 0.05	Not analyzed
4/Jul/2009	11	1	0.6	0.14 0.09	Not analyzed
1/Dec/2009	3.6	2	3.3	0.09 0.02	0.48
2/Dec/2009	12.1	5	11.1	0.17 0.02	1.26

* MDA was calculated for this spectrum according to [28] and was found to be equal to 0.082 Bq/L (confidence level 95%)

* Data retrieved from the site of the Space Weather Prediction Center, Boulder, CO, National Oceanic and Atmospheric Administration, US Deptt. of Commerce: http://www/swpc.noaa.gov/ftpmenu/indices.html

water is measured without filtration and volume geometry is used, suspended particulates may settle down, leading to an overestimation of ^{7}Be activity concentration in rainwater, as explained in the section on rainwater sampling. The results presented in tab. 1 are consistent with measurements reported by other researchers [29-31]. Although significant variations in ^{7}Be concentrations can be observed, no further interpretation was attempted, due to the limited volume of data available, particularly when the complex and dynamic nature of rainfall effects is taken in account. It is, however, interesting to note that in the two rain samples collected on December 2009, ^{7}Be concentration in the filters was very low. Though this applies to only two rainwater samples from the same rain event and thus no definite conclusion can be reached, this very low ^{7}Be concentration may be attributed to a reduced dust load on the filters, due to significant rainfalls occurring previously in the vicinity of Athens which might have reduced the air suspended particulates content.

CONCLUSIONS

Within the frame of the ^{7}Be monitoring program at the NED-NTUA, a protocol for the monitoring of ^{7}Be short-term activity concentration variations was established and applied. The whole procedure allows for the observation of both seasonal and diurnal concentration variations of ^{7}Be by keeping type A and B uncertainties low.

The measurements conducted over a two year period allow for the determination of mean monthly concentrations of ^{7}Be. Correlations between the ^{7}Be activity concentration and meteorological parameters of air temperature and relative humidity were determined, while no correlation can be claimed between the ^{7}Be activity concentration and the atmospheric pressure. The correlation between the ^{7}Be activity concentration and the number of sunspots could not be statistically confirmed, probably due to the short-term measurements and the fact that these measurements were conducted during the lowest of the 11-year cycle of solar activity. The two years' air and rainwater ^{7}Be concentration measurements presented in this work are in good agreement with other studies.

It has been demonstrated that diurnal variations in ^{7}Be activity concentration can be monitored using the techniques presented. However, more measurements are required in order to develop a model describing ^{7}Be diurnal behavior.

The results of the technique that was applied for the analysis of rainwater samples indicate that there is a basis for measuring ^{7}Be activity at both the filters and the volume samples. However, more measurements and a more thorough investigation that would take into account various meteorological conditions is needed in order to reach a definite conclusion.

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**Сотирија М. А. ПАПАНДРЕУ, Марилија И. САВА,
Константинос Л. КАРФОПУЛОС, Димитриос Ј. КАРАНГЕЛОС,
Мариос Ј. АНАГНОСТАКИС, Симос Е. СИМОПУЛОС**

МОНИТОРИНГ ^{7}Be У ПРИЗЕМНОМ СЛОЈУ АТМОСФЕРЕ КОРИШЋЕЊЕМ КРАТКОРОЧНИХ МЕРЕЊА

Концентрација ^{7}Be у приземном слоју атмосфере показује сезонске варијације услед физичких процеса који се догађају у тропосфери и стратоффрери, као и соларне активности. Истраживања ових варијација обављена су на Одсеку за нуклеарну инжењерију, Државног техничког универзитета у Атини, у периоду од две године (март 2008 – април 2010). У оквиру ове студије, методе за узорковање и анализу правилно су одабране како би се омогућило посматрање варирања концентрације коришћењем четворочасовног интервала узорковања, узимајући у обзир мерну несигурност типа А и Б. У циљу одређивања утицаја падавина на варијацију концентрације ^{7}Be у приземном слоју атмосфере, развијена је метода за сакупљање и анализу падавина. Такође, развијен је поступак који омогућава посматрање варирања сезонске и дневне концентрације ^{7}Be и корелацију између вредности концентрације ^{7}Be и метеоролошких параметара, као што су температура ваздуха и релативна влажност ваздуха.

Кључне речи: берилијум-7, дневна варијација, атмосферски аеросоли