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Variations of ^{210}Pb concentrations in surface air at Thessaloniki, Greece (40°N)

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Abstract

Atmospheric concentrations of ^{210}Pb were measured over the year 2009 in ground level air at Thessaloniki, Northern Greece (40°62′ N, 22°95′E). The mean activity concentrations of ^{210}Pb in surface air have been found to be $671 \pm 213\,\mu\mathrm{Bq\,m^{-3}}$. The highest values of monthly atmospheric concentrations of ^{210}Pb were observed in the autumn and the lowest in the spring period. The higher values of ^{210}Pb during autumn were attributed to frequent inversion conditions of the surface layers, resulting in an enrichment of radon and its decay products in surface air. The lower values during the winter months might be due to the low emanation of radon from the frozen or snow-covered soil. The minima of ^{210}Pb concentrations during spring might reflect on higher washout during this period, which results in less emanation of radon from saturated with water soil, resulting in less production of ^{210}Pb near ground-level air. The relative high values during summer are probably due to the higher ^{222}Rn exhalation from the ground and due to the higher air mixing within the troposphere, which has as a result to carry down to the surface layer ^{210}Pb whose origin is older air masses which entered into the free troposphere.

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1. Introduction

Lead-210 is the first long-lived ($t_{1/2} = 22.6\,\mathrm{y}$) decay product of ^{222}Rn, the first gas in the ^{238}U decay chain. Because ^{222}Rn is mainly exhaled from land surfaces and has a half-time $t_{1/2} = 3.82\,\mathrm{d}$, ^{210}Pb is primarily produced in the lower troposphere over continents.

Since atmospheric ^{222}Rn is a chemically inert and unscavenged and as a result is not removed from the atmosphere by physical or chemical mean, and its half-life is much less than the mixing time of the atmosphere, its concentrations are greatest near the land surface and decrease with both altitude and distance from land. As a result the main source of ^{210}Pb in the free troposphere is a quiescent ascent of its gaseous ^{222}Rn parent from the ground and upward transport of subspecies mobilized by resuspension and motion with the atmospheric masses. Advective transport of radon-rich air from the boundary layer can reach the upper troposphere or even the stratosphere creating a significant stratospheric ^{210}Pb reservoir above the tropopause. Most of ^{210}Pb in the stratosphere is due to the decay of ^{222}Rn injector convectively mainly in the tropics [1] while, other sources are the volcanic eruptions, which may introduce the ^{210}Pb to the stratospheric level [2].

The unique features of this radionuclide with altitudinal distinct sources, makes it ideal tool to depict transport processes in the whole atmosphere [3–5], suggesting that improved understanding of their atmospheric distributions obtained from detailed measurements will facilitate refinement and validation of global circulation models.

Lead-210 provides a useful tool for the study of the behavior of aerosols because, soon after the formation of ^{210}Pb atoms, the atoms become irreversibly attached to submicron aerosol particles. [6] estimated the (AMAD) of ^{210}Pb of $0.53\,\mu$m and [7] $0.56\,\mu$m. ^{210}Pb has been extensively used to determine the mean residence time of atmospheric aerosols [8], to trace chemical compound that may have broadly similar source distributions, for example sulphur [9], and to study the transport of continental aerosols across the sea or into Polar Regions [10, 11].

The concentrations of ^{210}Pb in the air decrease with elevation from the ground, due to its higher production rate in the lower troposphere. The vertical distribution of ^{210}Pb in the atmosphere has been studied by [12]. The distribution of ^{210}Pb in the atmosphere shows both spatial and temporal variations, depending on the geographical location, atmospheric circulation and scavenging processes.

The main objective of this work was to determine the factors influencing ^{210}Pb concentrations in surface air in the region of Thessaloniki, Greece

(40°62′ N, 22°95′E). The results of a one year period are presented and analyzed in combination of meteorological parameters.

2. Instrumentation

Lead-210 concentrations were measured by air sampling; using Staplex high-volume air samplers with Staplex type TFAGF 810 glass-fiber filters $8'' \times 10''$ and having 99.28% collection efficiency for particles as small as $0.3\,\mu$m. This design involves a regulated air-flow rate of 1.7–1.92 $\mathrm{m^3\,min^{-1}}$ (60–68 $\mathrm{ft^3\,min^{-1}}$). The length of each collection period was 24 h. Air samplings were carried out once a week on the roof (20 m above the ground, and 52 m a.s.l.), at the Faculty of Science building in the center of the city of Thessaloniki, Greece.

After the collection procedure, the filters are folded and compressed by means of hydraulic press at up to 3 tons to give a cylinder 5.8 cm diameter and 2 mm height. All the samples were measured for ^{210}Pb activity ($E_\gamma = 46.50$ keV) using a Ge planar detector, active area 2000 $\mathrm{mm^2}$, thickness 20 mm, energy resolution (FWHM) 400 eV at 5.9 keV or 700 eV at 122 keV, Be window 0.5 mm thin. Blank filters were regularly checked. The average total uncertainty for ^{210}Pb is almost 10%.

3. Results and discussion

3.1. ^{210}Pb concentration levels in surface air

Weekly measurements of ^{210}Pb activity concentration on aerosol particles were performed at ground level in outdoor air in Thessaloniki, Greece during the year 2009. Aerosol sampling started on January 2009. Mean activity concentrations of ^{210}Pb in surface air have been found to be $671 \pm 213\,\mu\mathrm{Bq\,m^{-3}}$, while the values range between 55–2063 $\mu\mathrm{Bq\,m^{-3}}$.

The average ^{210}Pb annual concentration measured in this work is in agreement with data reported in literature for continental locations. For the 10-year period 1982 to 1992, [13] reported mean values for ^{210}Pb to be $640 \pm 250\,\mu\mathrm{Bq\,m^{-3}}$ (range 290–1290 $\mu\mathrm{Bq\,m^{-3}}$) in ground level air at Munich–Neuherberg (48°8′N, 11°35′E), [14] reported a mean value of $540 \pm 210\,\mu\mathrm{Bq\,m^{-3}}$ (range 240–1400 $\mu\mathrm{Bq\,m^{-3}}$) for the region of Malaga, Spain (36.7°N, 4.5°W), comparable with our data. Lower values are reported from both lower and higher latitudes. [15] found a mean value of surface concentrations of $370 \pm 60\,\mu\mathrm{Bq\,m^{-3}}$ at El-Minia, Egypt (28°04′N,

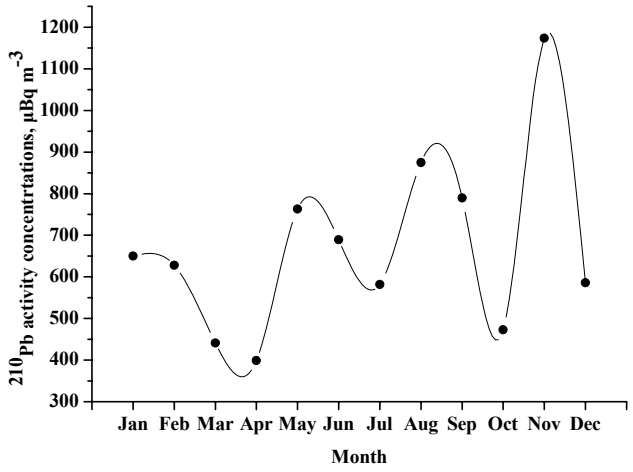

Figure 1: Mean monthly atmospheric concentrations of ^{210}Pb over year 2009.

30°45′E), [16] reported a mean value of 284 ± 150 at Islamabad, Pakistan (33.38°N, 73.10°E). While a 30-year long series of airborne ^{210}Pb in Finland gave a mean value of $282 \pm 257\,\mu\mathrm{Bq\,m^{-3}}$ and $237 \pm 226\,\mu\mathrm{Bq\,m^{-3}}$ for southern and northern Finland respectively [11].

Time variations of ^{210}Pb surface air concentrations were studied. Atmospheric processes and meteorological parameters that may contribute to these variations are discussed below.

3.2. Variability of ^{210}Pb concentrations in surface air

The concentration of ^{210}Pb in surface air fluctuate considerably. ^{210}Pb weekly concentration values showed a great variability ranging from 55 to $2063\,\mu\mathrm{Bq\,m^{-3}}$. The variability is mainly due to local prevailing meteorological conditions of the atmosphere during the sampling period. Since the day-by-day concentrations of ^{210}Pb in surface air show strong variations due to interactions on daily or even shorter time scales, considering the mean monthly concentrations these variations are minimized. Averaging the data of ^{210}Pb atmospheric concentrations on a monthly basis the mean monthly concentrations of ^{210}Pb over a year are calculated.

Figure 1 presents the mean monthly atmospheric concentrations of ^{210}Pb over year 2009. From Fig. 1 it is evident that the monthly atmospheric concentrations of ^{210}Pb in surface air over the year 2009 varied by a factor of 3 during the year, showing a seasonal trend with the highest values being

observed in the autumn months (up to 1173.5 μBq m^{-3} during November) and the lowest in the spring period (399 μBq m^{-3} during April). Also high values were observed during summer.

The higher values of ^{210}Pb during autumn might be attributed to frequent inversion conditions of the surface layers, resulting in an enrichment of radon and its decay products in ground-level air. The lower values during the winter months might be due to the low emanation of radon from the frozen or snow-covered soil [17]. The minima of ^{210}Pb concentrations during March and April might reflect on higher washout, since the higher precipitation amount took place during these months, which has as a result to remove ^{210}Pb aerosols efficiently. Moreover, the emanation of radon is strongly diminished when the soil is saturated with water, resulting in less production of ^{210}Pb near ground level air. The relatively high value during May of the year 2009 might attributed both to the lapse of any rain during sampling period and to the relative high temperatures during this month of the year 2009. The relatively high observed values of ^{210}Pb during the summer period is probably due to higher air mixing within the troposphere which has as a result to carry down to the surface layer ^{210}Pb whose origin is older air masses which entered into the free troposphere in combination with higher emanation of ^{222}Rn from local soil. These high observed values of ^{210}Pb concentrations in surface air during summer are in agreement with results reported [18], which observed that if the height of the tropopause is higher than 12 km (summer period), the concentrations of ^{210}Pb near the ground and in the middle troposphere are increased. [14] reported higher values of ^{210}Pb during summer in the region of Malaga, Spain.

Local meteorological parameters such as wind speed, air temperature, relative humidity, and rainfall were also monitored at the same site. No correlation between weekly ^{210}Pb concentration values and wind speed in the same period was found. Relative Humidity, RH (%), though it may slightly affect radionuclide concentrations in surface air, particularly through removal of atmospheric dust with associated radionuclides, does not seem to control the concentration of ^{210}Pb in surface air. No correlation was observed between the mean monthly ^{210}Pb activity concentrations and Relative Humidity, RH (%). Also, mean monthly ^{210}Pb activity concentrations were not correlated with temperature, T (°C).

A strong rainfall event during the sampling period results in decreased activity concentrations in surface air, while low precipitation rates during drizzling result in higher decreased surface air activity concentrations [19]. The low observed concentrations of ^{210}Pb during spring period coincidence with periods of rain, except May where no rainfall occurs during sampling

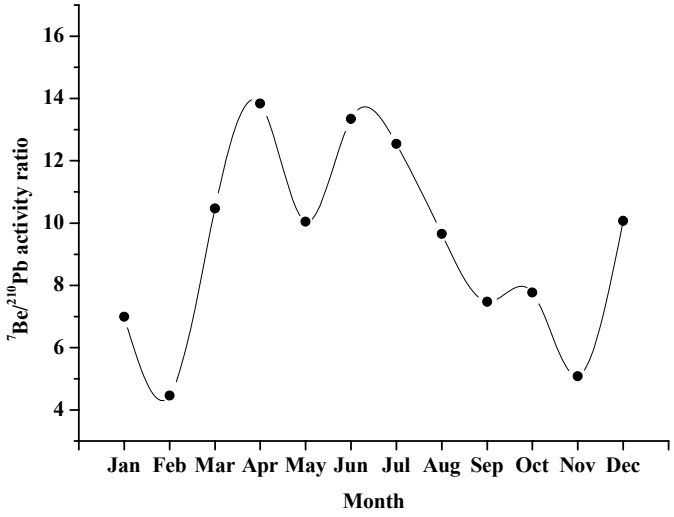

Figure 2: Mean monthly ^{7}Be/^{210}Pb activity ratio over year 2009.

periods. The very low value of 55 μBq m^{-3} that was observed during November 2009 was coincident with rainfall during the whole sampling period. However, the removal of ^{210}Pb from the atmosphere by rainfall events seems to be invalidated immediately after the event and the ^{210}Pb concentration in air to be reestablished rapidly. No correlation was observed between the mean monthly ^{210}Pb activity concentrations and rainfall.

3.3. ^{7}Be/^{210}Pb activity ratio

^{210}Pb data have been combined with data of ^{7}Be during year 2009. Simultaneous measurements of these two nuclides provide a powerful tool in atmospheric studies. The ^{7}Be/^{210}Pb ratio can provide information on the atmospheric removal behavior of these nuclides as well as information on whether these two radionuclides can be used as independent racers. Changes in ^{7}Be/^{210}Pb ratio would serve as an indicator of vertical transport in the atmosphere. These changes are presented and clarified below.

The different mechanisms that govern the levels of concentrations of ^{7}Be and ^{210}Pb in surface air for geomagnetic latitude over 40°N result in different ^{7}Be/^{210}Pb activity ratios. The observed monthly average ^{7}Be/^{210}Pb activity ratio varies by a factor of 3 during the year 2009 (Fig. 2) in very good agreement with the findings reported by [5] for the northern hemisphere continental sites. The monthly ^{7}Be/^{210}Pb activity ratio values show a broad spring – summer maximum peaking in April and June with an abnormal

low value during May, probably due to the abnormal high value of ^{210}Pb during May. The lowest activity ratio values were observed from September to February, with a high value during December, explained by the relative high ^{7}Be observed values during this month.

The lowest during the autumn – winter periods were attributed to the almost steady state conditions during those seasons. Frequent inversion condition during these months reflects on both radionuclides, since the winter stability tends to isolate surface air from the ^{7}Be sources in the upper troposphere and the opposite for the ^{210}Pb, to isolate the upper troposphere from its near ground level sources, resulting in lower concentrations for ^{7}Be and higher concentrations for ^{210}Pb. On the other hand, the high activity ratio values during the summer months were attributed to the increased dry convective mixing. Finally, since the stratospheric intrusions in situations of tropopause folding events during spring period result in a significant increase of ^{7}Be in the troposphere during this period, while have negligible reflection on ^{210}Pb concentration, we may conclude that the ratio ^{7}Be/^{210}Pb during spring period it could be an index of stratospheric contribution.

4. Conclusions

The highest values of monthly atmospheric concentrations of ^{210}Pb were observed in the autumn and the lowest in the spring period. The higher values of ^{210}Pb during autumn were attributed to frequent inversion conditions of the surface layers, resulting in an enrichment of radon and its decay products in ground-level air. The lower values during the winter months might be due to the low emanation of radon from the frozen or snow-covered soil. The minima of ^{210}Pb concentrations during spring might reflect on higher washout. The relative high values during the summer period were attributed to the maximum observed ^{222}Rn exhalation from the ground surface and to the higher air mixing within the troposphere

The monthly ^{7}Be/^{210}Pb activity ratio values show a broad spring – summer maximum. High values of ^{7}Be/^{210}Pb activity ratio seem to associate with upper atmosphere sources and low values with continental boundary layer sources. Frequent inversion condition during autumn period, and the strong vertical removal of air masses within the troposphere during summer months are both phenomena that result in both radionuclides, while stratosphere-troposphere air exchange reflects only the ^{7}Be concentrations in the spring period, as a consequence the ^{7}Be/^{210}Pb ratio during that season could be an index of stratospheric contribution.

The high variability of ^{210}Pb concentration on a weekly scale and its seasonal variation points at the importance of long-term measurements for a reliable ^{210}Pb activity concentration assessment in outdoor air.

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