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Relationship between the ^{7}Be and O_3 in the surface layer of air in Gydnia and Warsaw (Poland)

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Abstract
Beryllium ^{7}Be appears in the surface layers of the atmospheric boundary layer (ABL) as a result of stratosphere to troposphere air mass exchange (STE). Stratosphere to troposphere air mass transport (STT) can sometimes be accompanied by the origin of stratospheric ozone (O_3). The aim of this study was to attempt to answer the question as to whether ^{7}Be can be a good marker of stratospheric ozone origin in the concentration of ground-level ozone in the surface ABL. Eight sets of data covering the period 2002–2009 were examined: weekly concentrations of ^{7}Be and hourly O_3 measured in the ground level of the atmosphere. The measuring sites were at two different locations in Poland: Gdynia and Warsaw. Both locations show the maximum ^{7}Be concentration in spring and summer, and the minimum in late autumn and winter. In 2002–2009, the average concentration standard deviations of ^{7}Be were 3.336±1.312 mBq/m^3 and 2.907±0.030 mBq/m^3, respectively, for Gdynia and Warsaw. The data obtained were subjected to statistical and frequency analysis (FA) in order to eliminate the periodicity the seasonal and annual variability of solar activity and to estimate the the relationship between ^{7}Be and O^3.
Keywords: *beryllium (^{7}Be), ozone (O_3), $^{7}Be/O^3$ ratio, stratospheric intrusion (SI).*

1. Introduction

Galactic cosmic rays (GCRs) and solar energetic particles (SEPs) interact with the Earth's atmospheric nuclei and produce many isotopes through nuclear interactions, within a range of half-lives from several minutes to millions of years. One of them is beryllium (^{7}Be), a short-lived (half-life ~53.3 days) cosmogenic gamma emitting (~477.6 keV) radionuclide. It is produced in the stratosphere (~70%) and upper troposphere (~30%) by spallation reactions of energetic protons with atmospheric oxygen O (Z=8), nitrogen N (Z=7) and carbon O (Z=6). The level of activity in the stratosphere ^{7}Be is variable in time and space and depends on the intensity of cosmic rays (CRs) and solar activity. Solar activity changes with an average cycle length of 11 years (with variations from 9 to 14 years). The greater is the intensity of the CRs, the greater the solar activity and the intensity of the solar wind. However, the solar wind weakens the intensity of the GCR component. This phenomenon should have an impact on the concentration of ^{7}Be in the surface layer of the atmospheric boundary layer (ABL). The ^{7}Be produced in the stratosphere comes into the troposphere by processes of air mass exchange between the troposphere and the stratosphere (STE) [1]. The main predictor controlling the seasonality of ^{7}Be concentrations is the height of the tropopause, reflecting more vertical transport from upper tropospheric levels into the lower troposphere during the warm seasons than during the cold seasons. The estimated removal time in the stratosphere is much longer than the half-life of ^{7}Be, suggesting that most stratospheric ^{7}Be decays within the stratosphere and a large part of the surface ^{7}Be is of tropospheric origin. Since most ^{7}Be resides in the stratosphere, it has been used in many studies as a tracer of stratosphere to troposphere air mass transport (STT) [see e.g. 2]. Thus, air coming from the upper troposphere, and especially the stratosphere, can be identified by its enhanced ^{7}Be levels. The mean residence time (MRT) of ^{7}Be in the stratosphere has been reported e.g. by Thomas et al. [3] as 1 year, while the MRT in the troposphere is estimated to be approximately 25–40 days (range 3–60 days) [4, 5, 6]. The average ^{7}Be concentration in the lower stratosphere is in the range from approximately 150 to 200 mBq/m^3, while in the upper troposphere air is 40–83 mBq/m^3 [7, 8, 9], and surface air concentrations average a few mili-becquerels per cubic metre, as widely reported in the literature [10]. The ^{7}Be atoms transported from the stratosphere or formed in the troposphere, soon became attached to ambient aerosol particles in accumulation mode (from 0.07 to 2 μm, average 0.3 μm) [6]. The tropospheric residence time of these aerosols can vary from a few days to a few weeks,

depending on meteorological conditions [11]. Thus, air coming from the upper troposphere and especially the stratosphere can be identified by its enhanced ^{7}Be level. Feely et al. [1] reported that there are four factors controlling the ^{7}Be seasonal variation: (a) the wet scavenging process (dry deposition plays a minor role), (b) STE, (c) the vertical atmospheric motion in the troposphere, and (d) the air mass horizontal transport from middle latitudes into high latitudes. The variations in the surface layer ^{7}Be concentration generally depend on static factors, such as geographical location, and dynamic factors, such as CR variations, solar activity, cosmogenic production and in the lower troposphere, radioactive decay and meteorological conditions, including the dynamics of air mass exchange between the stratosphere and troposphere. Some studies have used the correlation between ^{7}Be and ozone O_3 to quantify the contribution of stratospheric O_3 to ground level O_3 [12]. According to numerous studies which have reported the O_3 annual cycle, the two types of situation are classified into episodes of high level ozone with a maximum in summer and the ones that have a spring maximum [13]. The aim of the present paper was to study the origin and annual course ^{7}Be in the near surface air at Gdynia and Warsaw during the period 2002–2009 and answer the question as to whether ^{7}Be can be a good an indicator of stratospheric ozone origin in the surface layer of the ABL. The analysis was conducted for the years 2002–2009 because for this period were available full database both for ozone as well as for beryllium.

2. Material and methods

The measurement results of the radioactive contamination of the environment were collected from a network of high-volume aerosol sampling stations, type ASS-500 [14]. The Central Laboratory for Radiological Protection (CLOR) operates the network of automatic State Environmental Monitoring (PMS) in Poland and that is supervised by the National Atomic Energy Agency (PAA). The weekly activity concentrations of radionuclide beryllium ^{7}Be were measured in a network of laboratories equipped with high-efficiency gamma rays spectrometry using an HPGe detector. The technical parameters of the stations ASS-500 are as follows: filter Petrianov FPP-15-1.5 with area 0.2 m^2, system drying filter – halogen radiators infrared, flow-meter – vortex, air flow volume 50 000 – 90 000 m^3 per week, 1.5 m a.g.l. height of air sampling. The sampling stations were located at Gdynia – a coastal city, situated at latitude 54°31'N and longitude 18°33'E, about 2 m above sea level (a.s.l.), at an average direct distance from the seashore 1 km – and at Warsaw – a continental city, situated at latitude 52°18'N and longitude 20°59.5'E, about 82 m a.s.l., at an average direct distance from the seashore of 262 km. Ozone ground-level O_3 is measured using a UV absorption spectrophotometer. The reference method for the measurement of ozone is that described in EN 1425:2005 "Ambient air quality – standard method for the measurements of the concentration of ozone by ultraviolet photometry" which agrees with Directive 2008/50/EC. The monitor is located in the station-type urban background localized in Gdynia (54°33'39''N, 18°29'36''E, 70 m a.s.l.) and Warsaw (52°13'28''N, 21°1'8''E, 112 m a.s.l.). Ozone concentrations have been obtained within the framework of the PMS tasks and have been made available by the Main Inspectorate of Environmental Protection (GIOŚ). The ozone measurement stations were located about 8 km to the north-east and 10 km to the north from the measurement of beryllium for Gdynia and Warsaw, respectively. The database contains 1-hour ozone ground-level concentrations. Solar activity was assessed on the basis of the number of sunspots (SSN). Daily SSN data were obtained from the website Solar Influences Data Analysis Center (SIDC). The main measurement period covered the eight years from January 2002 to December 2009. All the data obtained were statistical analysis (comprehensive regression analysis) and frequency analysis (FA). The data were analysed for the weekly or monthly average values.

The concentrations of beryllium ^{7}Be, ground-level ozone O_3 and the ratio $^{7}Be/_{3}O$ concentration, and the SSN were analysed Fast Fourier Analysis (FFA) in the period 2002–2009, together with the amplitude of the frequency of the annual and multiyear component ^{7}Be. It was assumed that the whole analysed period was considered as an interval $[0, 2\pi]$. These points were approximated using the following trigonometric polynomial:

$$Q_n(x) = \frac{a_0}{2} + \sum_{k=1}^{n} (a_k \cos(kx) + b_k \sin(kx)) \quad x \in [0,2\pi]$$

where a_0, …, a_k, b_1, …, b_n are real coefficient, n∈N, n < L.

The coefficients of the trigonometric polynomial that minimize the discrete least squares error are given by:

$$a_k = \frac{1}{L}\sum_{i=0}^{2L-1} \cos(kx_i) f(x_i)$$

$$= \frac{1}{L}\sum_{i=0}^{2L-1} \cos\left(k\frac{\pi ik}{L}\right) f(x_i) \qquad (1)$$

$$b_k = \frac{1}{L}\sum_{i=0}^{2L-1} \sin f(x_i)$$

$$= \frac{1}{L}\sum_{i=0}^{2L-1} \sin\left(k\frac{\pi ik}{L}\right) f(x_i)$$

$$k = 0,1,2,\dots,n$$

$f(x_i)$ where i = 0, …, 2L-1 are weekly concentrations. As a result, the amplitude spectrum is calculated for a particular frequency:

$$c_k = \sqrt{a_k^2 + b_k^2},$$

where k=0, …, 2L-1, and we can extract the component correspond to the annual variability.

3. Results

3.1 Frequency analysis

Figures 1 and 2 show the graph of the results of the frequency analysis FA for Gdynia and Warsaw.

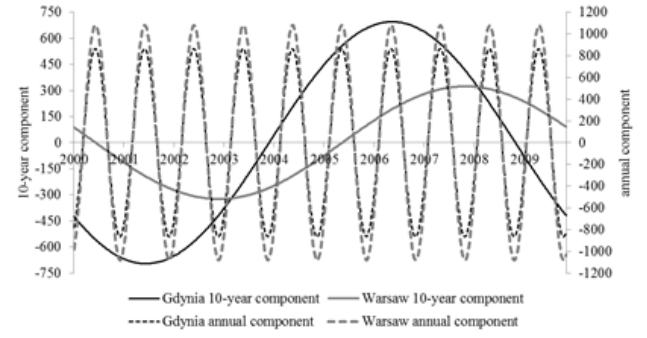
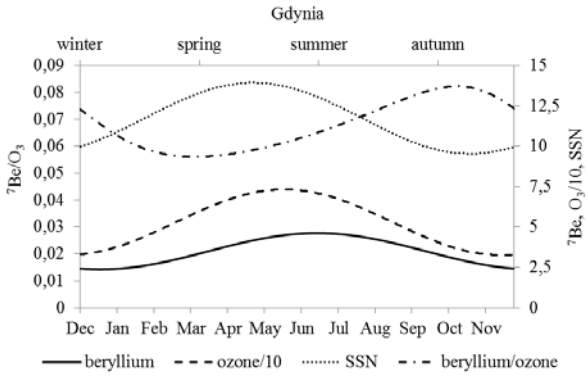


Fig. 1: Amplitude of the frequency for the annual and 10-year component of ^{7}Be for Gdynia and Warsaw calculated for the period 2000–2009.



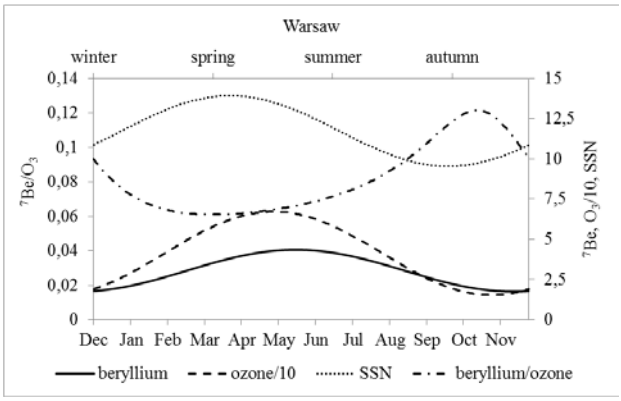


Fig. 2: Annual course amplitude of the frequency for ^{7}Be, O_3, ^{7}Be/O_3 and sunspot number (SSN) for Gdynia (upper figure) and Warsaw (bottom figure) calculated for the period 2000–2009 (winter: Dec, Jan. Feb, spring: Mar, Apr, May, summer: Jun, Jul, Aug, autumn: Sep, Oct, Nov).

FA confirmed the relations known from the literature:

- in general there are clearly marked variations in the annual concentrations of beryllium at both stations, the value of this component is slightly higher for Warsaw than Gdynia;
- the 10-year course of beryllium is quite significantly marked but is higher for Gdynia than Warsaw; the phase for the maximum concentration of beryllium in Warsaw is delayed by approximately 2.5 years from that of Gdynia;
- the annual course for beryllium and ozone at both stations is similar, although more pronounced for ozone;

- the maximum ozone concentration occurs in spring (April/May) and the minimum in autumn (October/November);
- the maximum beryllium concentration occurs in summer (June) and the minimum in autumn (October/November);
- the ratio of the concentrations of beryllium to ozone is highest in autumn (September/October) and lowest in spring (April);
- the annual course od SSN shows a maximum in April and a minimum in October;
- the regularity of the course concentrations of beryllium and ozone in the year indicate a significant impact of the radiation factor on the production and vertical transport of these substances.

3.2 Temporal variations of ^{7}Be and O_3

The monthly average values of ^{7}Be and O_3 measured at the two sites, Gdynia and Warsaw in Poland, over the period 2002–2009 are given in Figure 3. The ^{7}Be concentration is inversely correlated to the variations in SSN. ^{7}Be and O_3 both show a very similar annual and seasonal cycle, wherein higher concentrations were found in Gdynia (Figures 4–7). Both data sets show the maximum in ^{7}Be concentration in spring and summer, and the minimum in late autumn and winter. The average concentration standard deviations in 2002–2009 of ^{7}Be were 3.336±1.312 mBq/m^3 and 2.907±0.030 mBq/m^3, respectively for Gdynia and Warsaw. The average monthly concentrations of ^{7}Be were in the range of 2.241–4.188 mBq/m^3 (Gdynia) and 1.850–3.993 mBq/m^3 (Warsaw) and exhibited maximum specific activity in spring and summer and a minimum in winter. The results agree with results obtained at other locations in the world (see Table 1 for examples).

Table 1: Average ^{7}Be activity concentration in air in Poland and other countries.

City (Country)	^{7}Be [mBq/m^3]	Reference
Maryland (USA)	3.3–5	[15]
Osaka (Japan)	3–9	[16]
Kuwait (Kuwait)	0.2–14.9	[17]
Uppsala (Sweden)	0.5–9	[18]
Detroit (USA)	1.5–9.8	[19]
Thessaloniki (Greece)	0.47–12.7	[20]
Belgrade (Serbia)	0.6–18.3	[21]
Edinburgh (United Kingdom)	0.63–6.54	[22]
Brisbane (Australia)	1.2–8.7	[23]
Malaga (Spain)	2.5–14.9	[24]
Barcelona (Spain)	2.4–4.7	[25]
Islamabad (Pakistan)	1.14–3.17	[26]
Daejeon (Korea)	1.3–7.7	[27]
Monaco (Principality of Monaco)	0.93–13.1	[28]
Gdynia (Poland)	0.96–-9.37	This work
Warsaw (Poland)	0.52–7.04	

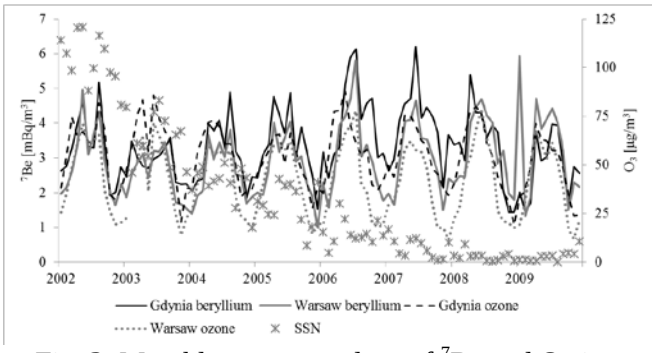


Fig. 3: Monthly average values of ^{7}Be and O_3 in Gdynia and Warsaw and sunspot number (SSN) over the period 2002–2009 (data point O_3 for February 2003 in Warsaw is missing).

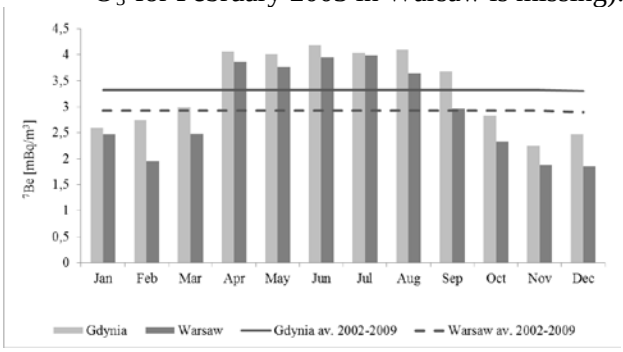


Fig. 4: Monthly average values of ^{7}B in Gdynia and Warsaw in the period 2002–2009 (lines represent the average value over the examined period).

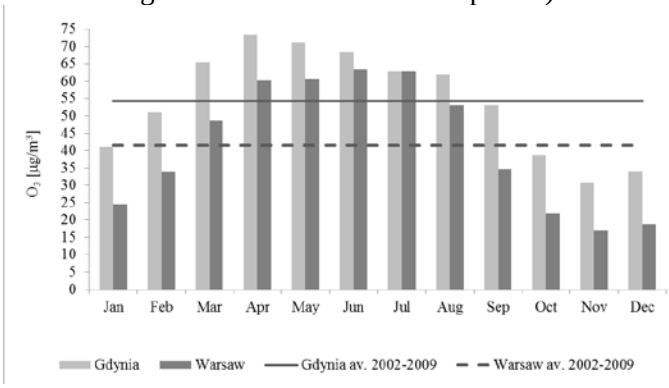


Fig. 5: Monthly average values of O_3 in Gdynia and Warsaw in the period 2002–2009 (lines represent the average value over the examined period).

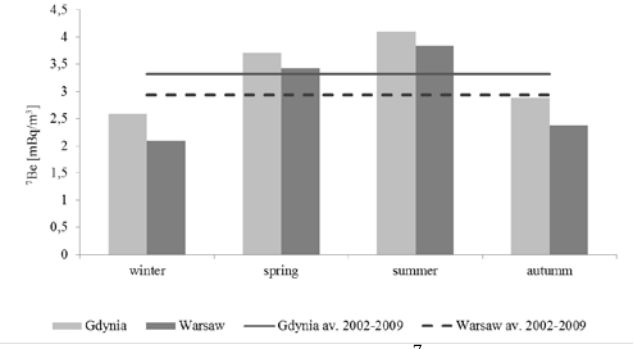


Fig. 6: Seasonal average values of ^{7}Be in Gdynia and Warsaw in the period 2002–2009 (lines represent the average value over the examined period).

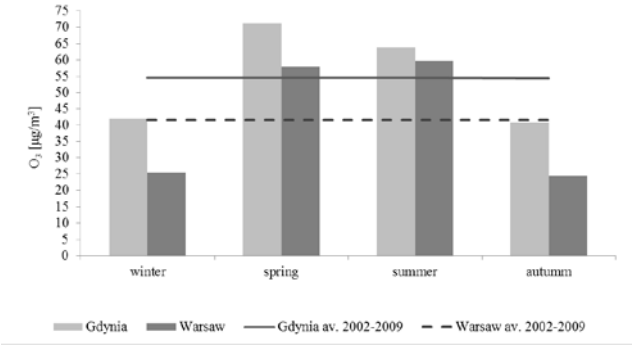


Fig. 7: Seasonal average values of O_3 in Gdynia and Warsaw in the period 2002–2009 (lines represent the average value over the examined period).

The monthly average O_3 values in the examined period were 54 µg/m^3 (Gdynia) and 42 µg/m^3 (Warsaw). The ranges of values were 19–87.7 µg/m^3 (Gdynia), and 11–79 µg/m^3 (Warsaw). The O_3 annual cycle obviously shows minima in winter and maxima in summer. To further analyse the relationship between ^{7}Be and O_3, seasonal indices were calculated. Table 2 shows a monthly and seasonal index representing the ratio of the monthly average value and the overall average value for the examined period. The value index for both study sites shows a characteristic annual pattern with maximum in the summer season and minimum in the winter season.

Table 2: Monthly and seasonal indices for ^{7}Be and O_3 in Gdynia and Warsaw in the period 2002–2009.

Station		Gdynia		Warsaw	
		^{7}Be	O_3	^{7}Be	O_3
Month					
winter	Dec	0.76	0.61	0.65	0.44
	Jan	0.78	0.75	0.79	0.58
	Feb	0.83	0.93	0.70	0.81
spring	Mar	1.02	1.21	0.96	1.14
	Apr	1.25	1.33	1.33	1.39
	May	1.21	1.37	1.33	1.55
summer	Jun	1.24	1.24	1.35	1.45
	Jul	1.17	1.14	1.30	1.46
	Aug	1.14	1.14	1.18	1.32
autumn	Sep	1.06	0.98	0.97	0.84
	Oct	0.85	0.72	0.81	0.57
	Nov	0.69	0.60	0.62	0.43
Quarter					
winter		0.79	0.76	0.72	0.61
spring		1.16	1.30	1.21	1.36
summer		1.18	1.17	1.28	1.41
autumn		0.87	0.77	0.80	0.61

Table 3 contains the results of the correlation between ^{7}Be and O_3. The linear Pearson correlation coefficients were calculated for the whole data set and for the seasons. The calculated overall correlation coefficient between ^{7}Be and O_3 is moderate for Gdynia and strong for Warsaw. The correlation is the highest for the monthly average maximum O_3. There is no very significant correlation in winter. The large correlation coefficient in autumn could indicate that ^{7}Be and the maximum O_3 are regulated by the same processes. One possible explanation is the existence of the downward transport from higher altitudes. However, monthly or weekly ^{7}Be allow only for limited analysis STE. Daily and hourly values of ^{7}Be are more appropriate.

Table 3: Linear Pearson correlation coefficient of ^{7}Be and O_3 (different font indicating *medium* and **large** value).

^{7}Be		Gdynia	Warsaw
Average O_3	all	**0.54**	**0.72**
	winter	***0.33***	-0.09
	spring	0.14	**0.53**
	summer	0.03	***0.38***
	autumn	***0.40***	**0.59**
Average maximum O_3	all	**0.58**	**0.73**
	winter	***0.33***	-0.07
	spring	0.16	**0.52**
	summer	0.15	0.29
	autumn	***0.48***	**0.67**
Maximum 1 hour O_3	all	**0.53**	**0.71**
	winter	0.16	-0.06
	spring	0.15	***0.43***
	summer	0.17	0.29
	autumn	***0.40***	**0.67**

3.3 $^{7}Be/O_3$ an atmospheric tracer

To identify the presence of O_3-rich stratospheric air at ground level, several studies have demonstrated the feasibility of using ^{7}Be as a natural radioactivity tracer of the upper atmospheric air. In analogy with the work by Ajtic et al. [29], a same approach is here proposed for the assessment of stratospheric intrusion SI phenomenon using in situ measurements of ^{7}Be and O_3. Taking into account that the descent of stratospheric air masses takes about a month, and assuming that the wet scavenging of ^{7}Be and mixing of air masses can be ignored over that period, the $^{7}Be/O_3$ ratios were calculated. Dutkiewicz and Husain [12] reported that the ratio of 0.2 mBq/µg gave 0.134 mBq/µg as a tropospheric $^{7}Be/O_3$ ratio, and the limits from Stohl et al. (2000) of 0.1 mBq/µg and 0.155 mBq/µg gave 0.067 mBq/µg and 0.104 mBq/µg, respectively. Figure 6 shows the ratios that were calculated for monthly average, monthly average maximum O_3 and monthly 1 hour maximum O_3. Comparing the monthly average ratio ($^{7}Be/O_3$) for Gdynia and Warsaw to the ratio based on Dutkiewicz and Husain [12], or Stohl [2] indicated many occurrences of stratospheric air sampling (Figure 8). The same results were obtained for the ratio ^{7}Be and weekly average O_3, weekly average maximum O_3, and weekly 1 hour maximum O_3. Examination of the monthly or weekly ^{7}Be and O_3 ratios confirmed only the possibility that the origin of stratospheric ozone is in the surface layer ABL.

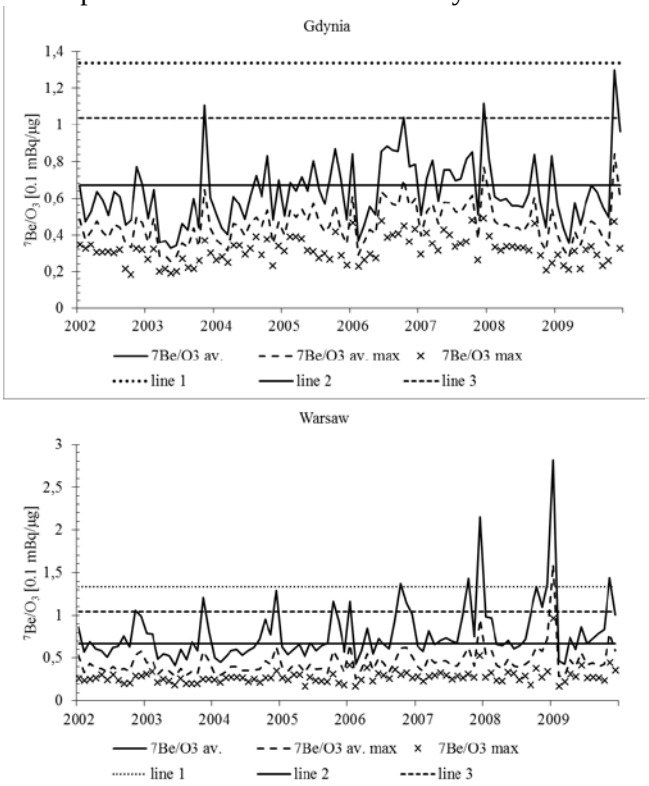


Fig. 8: Ratio of monthly mean ^{7}Be measured in Gdynia (upper figure), Warsaw (bottom figure) and monthly average ($^{7}Be/O_3$ av. grey line) and monthly average maximum O_3 ($^{7}Be/O_3$ av. max grey dotted line) and monthly 1 hour maximum O_3 ($^{7}Be/O_3$ max asterisks). Line 1 represents 0.134 mBq/µg, calculated according to Dutkiewicz and Husain

(1979), lines 2 and 3 represent the ratio limits of 0.067 mBq/μg and 0.104 mBq/μg calculated according to Stohl et al. (2000).

To find a situation conducive to the transport of ozone from the stratosphere to the troposphere a method based on Fourier analysis was also used. The analysis of the measurement data of ozone and beryllium ^{7}Be was performed by using FFA. Then, from the initial values, the component responsible for the annual cycle of ^{7}Be and O_3 was subtracted. In this way, the concentrations of ^{7}Be and O_3 independent of seasonal changes were obtained. In the next step, the periods in which a significantly positive deviation was noted for both O_3 and ^{7}Be concentrations and in relation to their normalization courses were found. In this period, situations in which the observed ratios of the weekly concentration of ^{7}Be to the weekly average maximum ozone concentration were high, were sought. Based on the analysis of synoptic and weather conditions, in particular the vertical distribution of potential temperature, relative humidity were identified, which potentially could be a phenomenon of STT. In 80% of selected cases, low relative humidity (< 20%) was identified at altitudes of about 3000 m above ground level. These situations were classified as cases of so-called "stratospheric intrusion" (SI). In the years 2002–2009 23 such cases were identified at the measuring station in Gdynia and nine cases for the station in Warsaw. The likely STE could be in June 2008 (Gdansk and Warsaw) and July 2009 (Warsaw). Most probably the situation occurred in the second ten days of July 2009. The next stage of the research will be a detailed analysis of the stratospheric–tropospheric phenomenon.

4. Conclusions

We can draw the following conclusions:

- ^{7}Be beryl shows annual course with maximum during spring/summer season, where the concentration is higher at both sites in Gdynia and Warsaw, and agrees with the results obtained at other locations in the world, particularly locations on the same latitude.
- The correlation coefficient between ^{7}Be and average O_3 generally is significant in autumn, which implies that their concentrations are driven by the same processes, and not very significant in winter.
- Analysis of the relationship between monthly and weekly concentrations of ^{7}Be even with 1-hour ground-level ozone does not allow unambiguous identification of the contribution of stratospheric ozone in its total concentration in the surface layer of the ABL. Daily and hourly values of ^{7}Be are more appropriate for this research and a more detailed study should be performed.
- Studying the monthly or weekly ^{7}Be/O_3 ratio only allows the identification of potential STE situations and the appearance of the origin of stratospheric ozone in the surface layer of the ABL.

In summary, ^{7}Be can be one of the indicators of stratospheric ozone in the surface layer of the ABL, but on its own is not a very good indicator. As a result, on the basis of the STE, not all situations, such as satellite remote sensing, upper atmospheric sounding, back trajectories, and meteorological conditions, are then confirmed in the course of the synoptic situation. STT is a temporary phenomenon and sometimes occurs on a local level in a vertical column of atmosphere. In the study of STT phenomena, data plays an important role, the best being short-term data (hourly, daily). On the other hand, it is difficult to determine the existence of an STE situation without upper airborne observations and measurements.

Appendix

Appendixes, if needed, appear before the acknowledgment.

Acknowledgments

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