An attempt to identify stratosphere-troposphere transport of ozone on the basis of beryllium (^{7}Be) activity concentration

E. Krajny[1], L. Osrodka[1], M. Wojtylak[1], M. Pajek[2] & B. Michalik[3]
[1]*Monitoring and Modelling of Air Pollution Department, Institute of Meteorology and Water Management NRI, Poland*
[2]*Satellite Remote Sensing Centre, Institute of Meteorology and Water Management NRI, Poland*
[3]*Central Mining Institute, Silesian Centre for Environmental Radioactivity, Poland*

Abstract

The aim of this study was to identify a situation of stratospheric to tropospheric air mass transport (STT), and then attempt to answer the question whether beryllium (^{7}Be) can be a good marker of stratospheric ozone (O_3) origin in the surface boundary layer of the atmosphere (SL) and its role in the tropospheric ozone budget. The sets of data covering the yearly period are examined such as: the daily activity concentration of ^{7}Be and lead (^{210}Pb), the hourly concentration of ground-level ozone O_3 and the meteorological parameters measured in SL in a meteorological station located in the town of Raciborz in southern Poland. The data obtained had undergone statistical and Fourier analysis. The data obtained had undergone Fourier analysis in order to eliminate the periodicity associated with, inter alia, the annual variability of solar radiation incoming to the Earth's surface. Then, based on statistical analysis potential days in which very likely was the occurrence of the so-called phenomenon "stratospheric intrusion" (SI) were identified. The selected periods were then analysed in terms of synoptic assessment of the impact of meteorological conditions on levels of ^{7}Be and O_3. For the selected case a detailed analysis was carried out based on the surface and upper air observations, satellite remote sensing, profile data and ground-level ozone. The use of trace methods based on the identification of concentrations of naturally occurring radionuclides as ^{7}Be and ^{210}Pb in SL of the atmosphere is

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complementary to the meteorological analysis for the identification of vertical inflow of air masses rich in ozone from stratosphere to troposphere.
Keywords: beryllium (7Be), ground-level ozone (O3), meteorology, surface layer, satellite remote sensing, stratospheric-tropospheric transport (STT), stratospheric intrusion (SI).

1 Introduction

Among the many radioactive substances monitored in ambient air beryllium (^{7}Be) is an element which may be used a tracer for air mass flow and vertical interchange dynamics between the stratosphere and troposphere [1–3]. Beryllium ^{7}Be (Z=4) is a naturally occurring cosmogenic element emitting gamma radiation (477.6 keV) and with a half-life of approximately 53.2 days. Essentially, it forms mainly from spallation processes, in the lower stratosphere (~70%) and upper troposphere (~30%), as a result of a reaction of high-energy protons or neutrons, present in cosmic radiation, with light nuclei, mostly carbon ^{12}C, nitrogen ^{14}N and oxygen ^{16}O [4–6]. The transfer of ^{7}Be to the troposphere and the variability of its concentration in the surface boundary layer (SL) depends on the seasonal changes of such processes as: (1) intensity of stratosphere-troposphere air mass exchange, (2) vertical transport in the troposphere, (3) horizontal transport from temperate and subtropical latitudes to polar latitudes and the equatorial zone, (4) gravitational subsidence (dry deposition) and washing out by precipitation (wet deposition) [7]. The downflow of beryllium from the stratosphere to the troposphere occurs mostly via interruptions in tropopause, appearing near the of stratospheric jet streams during the phenomenon of the tropopause folding. The penetration of cosmogenic elements to the troposphere is most intense in spring. This is connected with a decrease of tropopause thickness or even its decay in intermediate seasons, particularly in polar latitudes. These issues, widely studied globally, have yielded several publications concerning Poland to a various extent [8–10] so far. Lead ^{210}Pb (half-life 22.4 years) is present in SL mainly due to decay of radon (^{222}Rn) exhalated from top ground layer.

2 Measurement site and method

The measurements of daily beryllium (^{7}Be) and lead (^{210}Pb) activity concentration in surface layer air (SL) about 1.5 m height above ground level were performed on Raciborz meteorological station (geographical coordinates: latitude 50°03’42” N, longitude 18°11’30” E, 206 m height above sea level), that belongs to The National Weather Service network (IMWM NRI, Poland). The sampling programme started on 1st of June 2011 and ended 31st of May 2012. Samples of atmospheric aerosols were collected on a filter Petrianova (size 25 x 25 cm) using a pump Dwarf 100x9 Finnish company Senya (http://www.senya.fi Air Samples for Radiation Detection). During 24-hours were typically pumped about 3000 m^{3} (air flow of 140 m^{3}/h) of air. Filters with collected aerosols after waiting a few days for the decay of short radon (^{222}Rn) progeny, were formed by giving them the shape of a cylinder with a diameter of about 0.8 cm and 5 cm height (i.e. 2.5 cm^3) and then measured. The activity concentrations of ^7Be as well ^{210}Pb in filters were determined using an high-efficiency gamma rays spectrometry. HPGe Well type detector having a 50% relative efficiency was used. Activity concentration of ^7Be and ^{210}Pb were calculated based on the 477.56 keV peak (efficiency 10%) and 46.5 keV (efficiency 4.25%) respectively. Simultaneously monitoring of hourly concentration of ground-level ozone O_3 was carried out with AirPointer equipment and meteorological conditions monitoring in accordance with the guidelines WMO. The AirPointer is type approved according to the European norms for the O_3 (http://www.recordum.com).

3 Method of data analysis

The selection of episodes of stratospheric intrusion (SI) and their development was based on two criteria: mathematical and meteorological. Mathematical analysis based on statistics and Fourier analysis were used to identify intrusions of ozone from the stratosphere to the troposphere using ground-based data on concentrations of radionuclides, ozone ground-level and meteorological conditions. Meteorological criterion includes an examination of the possible occurrence of SI episodes based on synoptic analysis and interpretation of satellite data.

3.1 Mathematical analysis

The concentrations of beryllium ^7Be, lead ^{210}Pb and ground-level ozone O_3 were Fourier and statistically analyzed. The Fourier analysis was applied to eliminate the periodicity associated with the annual variability of solar radiation incoming to the Earth's surface. No periodic variability of concentrations of ^7Be and O_3 were analysed after subtracting the component of the annual one. It was assumed that the whole analyzed period is considered as interval [0, 2π]. In the set of the data identified points parallel i.e. daily data. These points are approximated using the following trigonometric polynomial

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

that is determined coefficients a_k and b_k , and the fifth component corresponds to the annual variability

$$a_k = \frac{1}{L}\sum_{i=0}^{2L-1}\cos(kx_i)f(x_i) = \frac{1}{L}\sum_{i=0}^{2L-1}\cos(k\frac{\pi ik}{L})f(x_i)$$
$$b_k = \frac{1}{L}\sum_{i=0}^{2L-1}\sin(kx_i)f(x_i) = \frac{1}{L}\sum_{i=0}^{2L-1}\sin(k\frac{\pi ik}{L})f(x_i) \qquad where\ k = 0,1,2,...,n \tag{2}$$

As a result, the amplitude spectrum is calculated for a particular frequency

$$c_k = \sqrt{a_k^2 + b_k^2} \tag{3}$$

The visual analysis shows that ^7Be and O_3 concentrations and some meteorological parameters in surface layer of air, shown clear annual variation. Average time series plus annual component determine the typical annual course of parameter. The annual component should be eliminated from the time series in order to receive a more pronounced course of the parameter by equation:

$$Y_i^{(j)} = X_i^{(j)} - \left(\frac{a_0}{2} + a_r \cdot \cos\left(\frac{r \cdot \pi \cdot i}{L}\right) + b_r \cdot \sin\left(\frac{r \cdot \pi \cdot i}{L}\right)\right) \quad where \quad i = 0,\ldots,2L-1 \tag{4}$$

Large positive values indicate the period of increase the level a given parameter and negative values of the decrease this parameter. Then, it is assessed whether the parameter value is greater or lesser than the average. To identify potential episodes SI uses four parameters ^{7}Be, ^{210}Pb, $^{7}Be/^{210}Pb$, O_3 in the surface layer of air. Four-dimensional centroid point has been taken (2.278 mBg/m^3, -0.706 mBg/m^3, 7.2, 7.06 $\mu g/m^3$). It has been designated based on quantile 0.9 for ^{7}Be, O_3, $^{7}Be/^{210}Pb$ and 0.1 for ^{210}Pb. Then the method of the k-nearest neighbours used to identify SI episodes [11].

3.2 Meteorological analysis

Very interesting and important for forecasters are situations connected with tropopause folding and stratospheric intrusions. The use of ozone, water vapour and thermal channels of METEOSAT SEVIRI instrument in form of RGB colour composites allows for detection and monitoring of such cases. Examples of satellite products for detection of such phenomena are described. The main mechanisms of stratosphere – troposphere exchange in middle altitudes are tropopause folds. This intrusion of stratospheric air deep into the troposphere is connected usually with upper tropospheric jet streams. The valuable information about stratospheric/tropospheric interaction is provided by the WV 6.2 μm channel of the SEVIRI radiometer located on the Meteosat geostationary satellite. The feature on the image – relatively narrow dark stripes [12] lying typically on the cyclonic side of the jet axis could be the indicator of sinking, dry air which could be of stratospheric origin. These dark stripes are often accompanied by potential vorticity (PV) anomalies, produced by the intrusion of stratospheric air into the upper troposphere. Potential vorticity is a concept corresponding to potential temperature; a measure of potential for vorticity in an air parcel. Stratospheric air is characterized by high PV and low relative humidity. Because potential vorticity is the product of vorticity and stability, it can be used to determine the height of the tropopause. When defined in this manner, the tropopause can be called by the more specific name of dynamic tropopause. Potential vorticity unit ($PVU = 10^{-6} \cdot K \cdot s^{-1} \cdot kg^{-1} \cdot m^{-2}$) PVU is > 3 in the stratosphere and < 3 in the troposphere. In mid-latitudes the tropopause is mostly defined as 2 PVU, but also values 1 and 1,5 are used [13]. According to the WMO definition of dynamical tropopause PV > 1.6 PVU at the tropopause indicate the presence of air mass having stratospheric origin [14]. In favourable synoptic conditions the transport mechanism of the ozone rich stratospheric air could lead to increase of the tropospheric ozone in the low part of the atmosphere [15]. The dark stripes are the most frequent features but not the only ones. The other features which could represent sinking, stratospheric air are vortices at different scales on the WV images, like small scale dark circles called "eyes". Meteosat9 SEVIRI radiometer with its 12 spectral channels allows us to generate advanced products – RGB colour composites. The visual interpretation of these images is much more user friendly comparing the analysis of the single satellite channels. One of the EUMETSAT RGB Standard Composition "Airmass" [16] is the combination of 4 channels: WV6.2, WV7.3, IR9.7 and IR10.8 [12]. Significant influence of WV 6,2 μm channel helps to detect position of jet streams and areas of dry, descending, possible stratospheric, ozone rich air, with high PV values (in reddish colour). It is also useful to discriminate different airmasses and to detect typical WV features.

4 Results and discussions

4.1 Temporal variation of ^{7}Be

^{7}Be and O_3 both show a very similar annual and seasonal cycle. Both data sets show the maximum concentration in spring and summer, and the minimum in late autumn and winter. The average concentration of ^{7}Be was 3.366±1.801 mBq/m^3 in Raciborz. The average monthly concentrations of ^{7}Be were in the range of 5.075–1.802 mBq/m respectively for May 2012 and December 2012 The results agree with results obtained at other location, particularly in temperate latitude (tab. 1).

Table 1: ^{7}Be activity concentrations in air in Raciborz and other sites worldwide.

City (Country)	^{7}Be [mBq/m^3] min – max	Reference
Malaga (Spain)	2.5–14.9	[17]
Thessaloniki (Greece)	0.47–12.7	[18]
Uppsala (Sweden)	0.5–9	[19]
Belgrade (Serbia)	0.6–18.3	[20]
Edinburgh (Unit Kingdom)	0.63–6.54	[21]
Monaco (Principality of Monaco)	0.93–13.1	[22]
Osaka (Japan)	3–9	[23]
Detroit (USA)	1.5–9.8	[24]
Brisbane (Australia)	1.2–8.7	[25]
Daejeon (Korea)	1.3–7.7	[26]
Kuwait (Kuwait)	0.2–14.9	[27]
Raciborz (Poland)	0.37–9.18	This work

The analysis of data shows the large amplitude fluctuations in the activity concentrations of ^{7}Be during the year (fig. 1). This is probably due to annual variations related to the annual cycle of solar radiation intensity variations. Statistical analysis of the concentrations of ^{7}Be and meteorological parameters in Raciborz showed that Pearson linear correlation coefficients (r) were positive for sunshine (0.576), maximum temperature air (0.511), average temperature air (0.445), minimum temperature air (0.317), and negative for relative humidity (-0.663), total cloudiness (-0.414), wind speed (-0.241). All obtained correlation coefficients are statistically important for the significance level $\alpha = 0.05$.

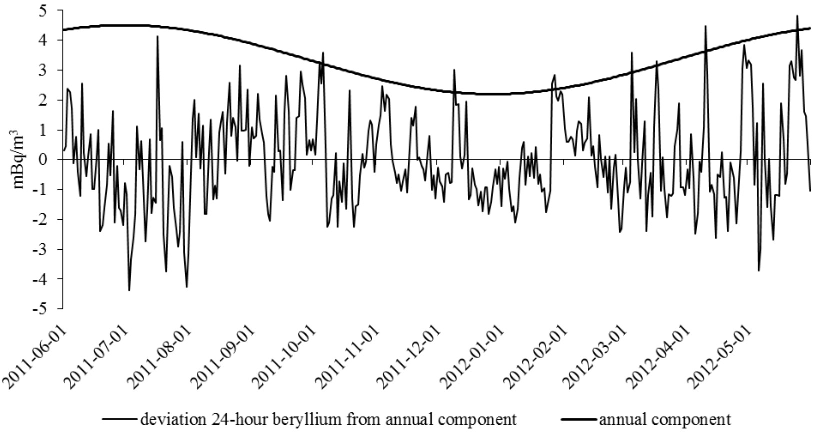

Figure 1: ^{7}Be activity concentrations in air in Raciborz.

4.2 ^{7}Be as trace stratospheric intrusion – selection of episodes

Using the mathematical analysis described above the period from 01.06.2011 to 31.05.2012 was selected , including few days with the potential for transport of ozone from troposphere to the stratosphere (SI) in Raciborz (tab. 2).

Table 2: Dates of potential episodes of transport of ozone from the stratosphere to the troposphere and deviation concentrations of ^{7}Be, ^{210}Pb, O_3 from their typical course in Raciborz.

Date	Mathematical analysis			
	Deviation from the typical course of concentration			
	^{7}Be [mBq/m^3]	^{210}Pb [mBq/m^3]	$^{7}Be/^{210}Pb$	O_3 [μg/m^3]
10.06.2011	2.563	-0.079	10.3	7.7
25.06.2011	**1.639**	**-0.105**	**7.5**	**5.7**
19.10.2011	2.331	-0.499	4.4	5.8
9.12.2011	**3.014**	**-0.752**	**10.1**	**4.4**
5.03.2012	3.584	-0.272	8.1	1.9
25.05.2012	4.838	-0.062	16.4	7.7

Among the thus selected day SI only two 25.06.2011 and 9.12.2012 were also identified independently by meteorological analysis. In the next part of the paper presents a detailed description of the episode SI 25.06.2011.

4.3 Stratospheric intrusion: case study 25th June 2011

On the 25th June 2011 Polish territory was under the influence of weak high-pressure wedge for a cool front which moving in the east (fig. 2). Over the southern part of the country was a zone of strong jet stream with north-west direction. The height of the tropopause over the southern Polish was about 7.8 km. Multiple changes of wind direction from south-east to north-west and back to south-east occurring at heights of 5 to 20 km facilitates stratospheric air transport from subtropical to higher latitudes and its mixing with tropospheric air.

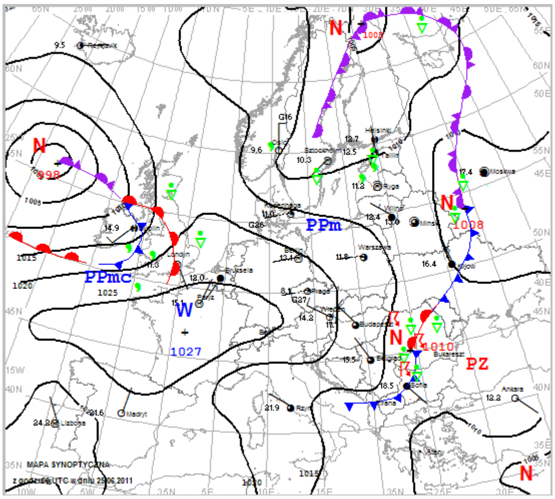

Figure 2: Surface weather chart 25.06.2011 0 UTC (source: IMWM-NRI).

Satellite images taken in the water vapour channel reveal the existence of a narrow zone of settling dry air currents over Poland, moving downstream of the behind edge of frontal zone, before the left, anticyclonic edge of the jet stream. Meridional isentropic cross-section indicative of cold air masses drainage confirms this claim. For that day the height of PV=1.5 (i.e. height in hPa, where the potential vorticity equals 1.5 PVU) was not available, but at least a few available products indicates stratospheric intrusion such as: isotachs 300 hPa – jet stream, wind for 300 hPa – a vectors wind very strong, geopotential height of the pressure level 300 hPa (500 and 700 hPa) – settling, inflow of air masses with NNW direction. Vertical cross section show examples of parameters (fig. 3). On the other hand temperature advection 700 hPa indicated cold air influx. The 25th June of 2011 at 0600 UTC two dark stripes situated on west and east fringe of Poland are clearly seen (fig. 4). The positive vorticity advection and the high upper wind speed (> 50 m/s) in jet streak indicate possibility of the tropopause folding and the stratospheric intrusion. Later on, at the 1800 UTC synoptic situation is still favourable to the dry, cold air intrusion. Reddish colour at the Airmass RGB composition shows that it could be ozone rich air mass from stratospheric origins (fig. 5).

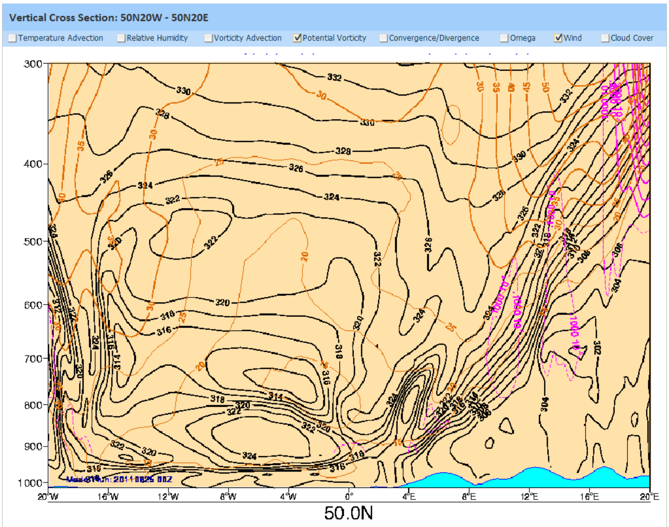

Figure 3: Vertical cross section of potential vorticity – pink line and wind speed – brown line for 1200 UTC 25.06.2011 (source: www.eumetrain.org Copyright EUMETSAT).

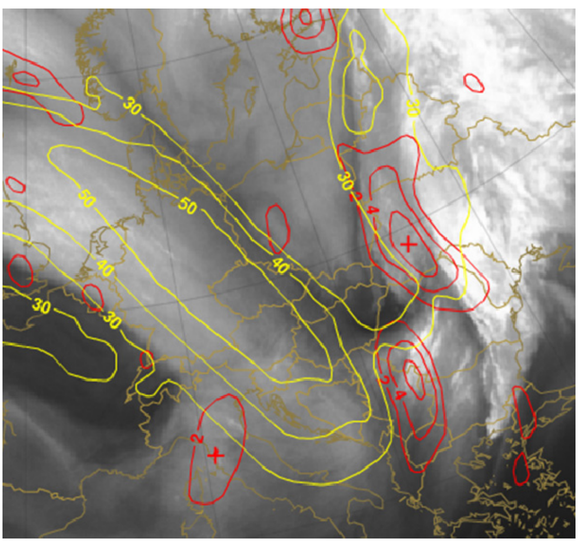

Figure 4: Meteosat WV 6.2 µm channel superimposed by cyclonic vorticity advection PVA – red lines and isotachs at 300 hPa (m/s) – yellow lines for 0600UTC 25.06.2011 (source: www.eumetrain.org Copyright EUMETSAT).

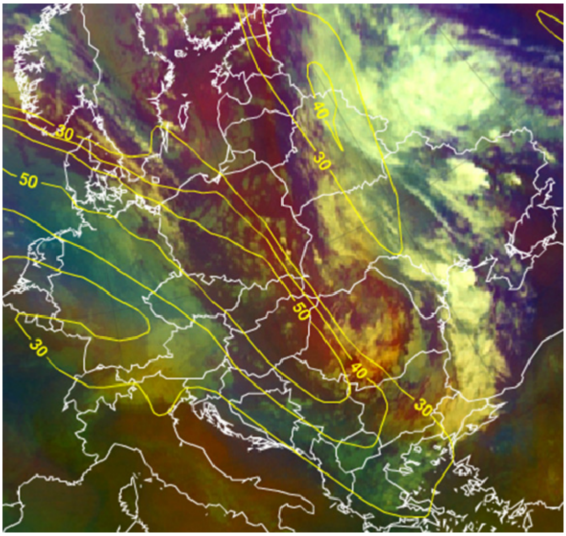

Figure 5: Meteosat RGB "AirMass" superimposed by isotachs at 300 hPa (m/s) – yellow lines for 1800 UTC 25.06.2011 (source: www.eumetrain.org Copyright EUMETSAT).

The above described situation favorable the transfer of air masses from the stratosphere to the troposphere by the mechanism described by Danielsen [1], which shows a cross section through the atmosphere and difference level of the tropopause on both sides of the axis of the jet stream. An additional factor in this case is the change of wind direction in the upper troposphere and lower stratosphere conducive additionally mixing of stratospheric air masses with tropospheric.

4.4 Discussion

The occurrence of natural radioactivity in the surface layer of the troposphere is of high cognitive importance for understanding of the physical and chemical processes occurring in the atmosphere. Due to different origin of ^{7}Be and ^{210}Pb, the value of the ^{7}Be/^{210}Pb ratio may serve as indication of the height which the air mass is transported from and whether the air is of continental or marine origin, it also allows determination of the average residence time of the aerosol in the atmosphere. The activity of ^{7}Be may also be used as an indicator of the share of ozone of stratospheric origin in its total concentration in surface layer air [28, 29]. The study been shown significant annual variation of activity concentration beryllium ^{7}Be, demonstrating advantage radiative processes in the formation of these concentrations though not necessarily associated with solar activity.

Application of the method of filtering the input data due to the dominant annual component has enabled the development of a methodology to identify situations in which the concentration of radionuclides ^{7}Be could be in connection with vertical transport of air masses from the stratosphere to troposphere. If this phenomenon was accompanied by additionally elevated ozone concentrations and specific meteorological conditions such situations could be classified as a flow of stratospheric ozone into the troposphere. The studies carried out with a year of data samples allowed us to identify six cases corresponding to episodes of vertical transport of ozone from the stratosphere to the troposphere, i.e. stratospheric intrusion (SI). Among these potential episodes SI selected mathematical method, only two were clearly positively verified by the meteorological analysis.

5 Conclusions

The paper investigates the possible contribution SI evens to the observed higher than normal ozone ground-level. The analysis led to the following conclusions:

1. The use of trace methods based on the identification of concentrations of ^{7}Be and ^{210}Pb in SL of the atmosphere is complementary to the meteorological analysis for the identification of vertical inflow of air masses rich in ozone from stratosphere to troposphere.
2. The study showed that the course of concentration of radionuclides, like the course of tropospheric ozone concentrations and meteorological elements has a clear annual periodicity associated with the radiative climate factors. The use of filtration methods of data by removing the annual component makes it much easier to identify situations related to the vertical transport of air masses and stratospheric ozone. This method can be used to identify the initial condition of the stratospheric intrusion.
3. The qualitative analysis for selected cases was done. It showed that most valuable information about stratospheric-tropospheric interaction by transport STT was provided with use of the WV 6,2 μm channel. Usage of additional information about wind speed and vorticity in the upper troposphere helps to distinguish between stratospheric intrusion SI and dry arctic air flow.

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