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BERYLLIUM-7 SURFACE CONCENTRATION EXTREMES IN EUROPE †

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Abstract. *Seasonal and spatial patterns of extremely high beryllium-7 surface concentration recorded over the 2001–2010 period across Europe are investigated. The beryllium-7 measurements for 14 sites are taken from the Radioactivity Environmental Monitoring Database. The maxima and minima in the annual cycle of the beryllium-7 surface concentration occur later in the year as the latitude of the measurement site decreases. Extremely high beryllium-7 surface concentrations are defined here as values greater than the 95th percentile in each measurement site. Most of the extremes occur over the March–August period. At least 10 % of the total number of extremes appear during autumn and winter, with an exception of Vienna, where all the extremes took place during spring and summer. The regional spread of extremes common to pairs of measurement sites points to an existence of three distinct regions in Europe: north of 55 °N, between 45 °N and 55 °N, and south of 45 °N. Although the beryllium-7 concentration records are significantly correlated across all the investigated sites, the strongest correlations are found within the identified regions.*

Key words: *beryllium-7, annual cycle, maximum concentrations, seasonal distribution, correlations, Europe*

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1. Introduction

Beryllium-7 (Be-7) is a cosmogenic natural radionuclide produced in spallation processes in the upper troposphere and lower stratosphere (Lal and Peters, 1967). Once produced, Be-7 rapidly attaches to fine particles (e.g., Papastefanou and Ioannidou, 1995; Winkler et al., 1998), and thereon it is subject to the same removal mechanisms as its carrier aerosol (primarily wet and secondarily dry deposition) (Ioannidou and Papastefanou, 2006; Papastefanou and Ioannidou, 1991; Pham et al., 2011). Descent of air masses brings Be-7 from its production altitude down to the Earth's surface. Hence, the abundance of Be-7 at the surface is a result of its transport history – the duration of descent is one key factor while passage through precipitation regions is the other major determinant (Feely et al., 1989). Due to its relatively long half-life of 53.22 days, the Be-7 concentration in the air can be taken as a tracer of subsidence from the upper troposphere–lower stratosphere region (e.g. Husain et al., 1977; Zanis et al., 1999, 2003). However, since the Be-7 concentration in the air depends strongly on removal mechanisms of its carrier aerosol, and since about a third of its production takes place in the upper troposphere (e.g. Usoskin and Kovaltsov, 2008), it cannot be considered as an unambiguous stratospheric tracer (e.g. Cristofanelli et al., 2006).

Beryllium-7 surface concentration shows an annual cycle with the maxima occurring in spring and summer (e.g. Ajtić et al., 2008, 2013; Hernández-Ceballos et al., 2015; Sarvan et al, 2016; Steinmann et al., 2013; Todorovic et al. 1999, 2005). In spring, relatively frequent stratospheric intrusions enrich the troposphere with air masses of high Be-7 concentration (Cristofanelli et al., 2006; Elbern et al., 1997; Feely et al., 1989). In summer, on the other hand, it is the efficient vertical mixing within the troposphere that brings air rich in Be-7 from the upper troposphere down to the surface (Feely et al. 1989; Gerasopoulos et al., 2001, 2003). Isolated cases of high Be-7 surface concentration have been investigated, mostly in alpine stations in Europe (Cristofanelli et al., 2006; Elbern et al., 1997; Gerasopoulos et al., 2001; Stohl et al., 2000; Zanis et al., 1999). Ajtić et al. (2016) demonstrated that in Helsinki, Finland, around 10 % of extremely high Be-7 surface concentrations occurred outside the warm season, i.e. in autumn and winter. Further, Hernández-Ceballos et al. (2017) investigated events of high Be-7 concentrations in Spain, and showed that some cases happened during the colder half of the year.

This paper discusses the overall pattern of high Be-7 surface concentration occurrences in a region of Europe between 37–69 °N and 6 °W – 28 °E. The temporal and spatial distribution of these occurrences are investigated. Mutual correlations between the Be-7 measurements performed in different locations are also analyzed.

2. Material and Methods

2.1. Beryllim-7 database

The radioactivity measurements stored in the Radioactivity Environmental Monitoring Database (REMdb) were used in this analysis. This database is supported by the Radioactivity Environmental Monitoring and Emergency Preparedness and Response (REM&EPR) group of the Knowledge for Nuclear Safety, Security & Safeguards Unit of the Joint Research Centre (JRC) in Ispra, Italy, and the database is stored online.

The REM database contains environmental radioactivity measurements since 1984. The Be-7 activity concentrations are available for 34 European stations (sparse Network) in which high-sensitivity measurements are performed. The Be-7 measurements stored in the REMdb are public for the data until 2006, while the access to the data for the 2007–2015 period can be granted only after explicit request. Further information on the measurement procedure and the list of the contributing National Competent Authorities is available in Hernández-Ceballos et al. (2015) and in the REMdb monitoring reports (https://rem.jrc.ec.europa.eu/RemWeb/Reports.aspx).

2.2. Study area

In this analysis, 14 measurement sites from the sparse REM database were selected. The sites cover a region of Europe in the 37–69 °N latitudinal belt, with the longitude between 6 °W and 28 °E (Fig. 1). The altitude of these sites is between 4 m and 411 m above sea level, with Madrid as the only exception at 715 m.

This choice of the sites was based on the similarities in the sampling patterns: approximately one measurement per week was performed in each location over the 2001–2010 period, and around 500 data points in total were collected per each site.

2.3. Methodology

First, extremes were defined as the measurements with the Be-7 specific activity above the 95^{th} percentile of the total set of measurements in each sampling location. The 95^{th} percentile marks the value in a set of data whereby 95 % of the values are less than this value (NIST/SEMATECH e-Handbook of Statistical Methods http://www.itl.nist.gov/div898/handbook//prc/section2/prc252.htm, 9 April 2017). In other words, only 5 % of the values are greater than the 95^{th} percentile. Thus, in our analysis, the highest 5 % of the measurements per site were taken as the extremes. For each site, the number of available measurements was around 500, and the highest 5 % gave around 25 extremes.

Fig. 1 Measurement locations with their latitude and longitude, and the total number of measurements over 2001–2010 (in parenthesis).

Using the 95th percentile in each sampling location provides a threshold for extremes which is specific for each set of measurements, and has an advantage over a constant limit (such as 8 mBq/m^3 in Cristofanelli et al. (2006), Hernández-Ceballos et al. (2017) and Stohl et al. (2000)), since it includes an inherent spatial dependence in the Be-7 distribution in Europe – e.g. the measurement sites in the south show ~2.5 higher concentrations than in the north of the continent (Hernández-Ceballos et al., 2015).

In addition to looking into the extremes at each location, dates of simultaneous Be-7 extremes at multiple measurement sites were also investigated. These results were limited in a sense that they looked only into the extremes coinciding on the very same day. As a prerequisite for this, the measurements also had to be performed on the very same day. To overcome this limitation, a period of +/- 3 days around each extreme was also marked as an extreme event. Intersections of periods thus generated gave common Be-7 extremes as events occurring within a week at multiple measurement locations.

To investigate a relation between the mean monthly Be-7 specific activities measured at different locations, a normality test was first performed. Since most of the data arrays were not normally distributed, Spearman's correlation coefficients and their statistical significance (significance level 0.05) were calculated. Only statistically significant coefficients are given here.

3. Results and Discussion

Differences in the annual cycles at the 14 stations were investigated first. Overall, the Be-7 concentrations were higher in the south, and the maximum and minimum of the annual cycle shifted to later in the year as the latitude decreased. Thus, the locations in the north (latitude greater than 50 °N) reached the maximum in May, in midlatitudes (40–50 °N) the maximum occurred in June and July, while in the south (latitude less than 40 °N) in August. The minimum of the annual cycle shifted from November in the northern stations to January for the southern stations.

To illustrate the meridional differences between the locations, seasonal patterns for a northern (Ivalo, 68.64 °N; 27.57 °E), midlatitude (Vienna, 48.22 °N; 16.35 °E) and southern site (Madrid, 40.45 °N; -3.69 °E) are given in Figure 2. The Be-7 surface concentration is less in Ivalo than further south, in Vienna and Madrid. At Ivalo, Vienna and Madrid, the maxima in the annual cycle occur in May, July and August, respectively, while the minima occur in November, December and January, respectively.

The latitudinal dependence of the Be-7 specific activity in surface air has been observed before (e.g. Doering and Saey, 2014; Feely et al., 1989; Hernández-Ceballos et al., 2015, 2016a; Kulan et al., 2006; Persson and Holm, 2014). Doering and Saey (2014) also reported a disparity in the timing of the maximum annual concentrations between four stations covering a latitudinal belt of 25° in Australia.

3.1. Seasonal characteristics of the Be-7 extremes

The Be-7 extremes were grouped into seasons: autumn (September, October and November), winter (December, January and February), spring (March, April and May), and summer (June, July and August). Figure 3 shows the seasonal distribution of the extremes for the 14 selected stations.

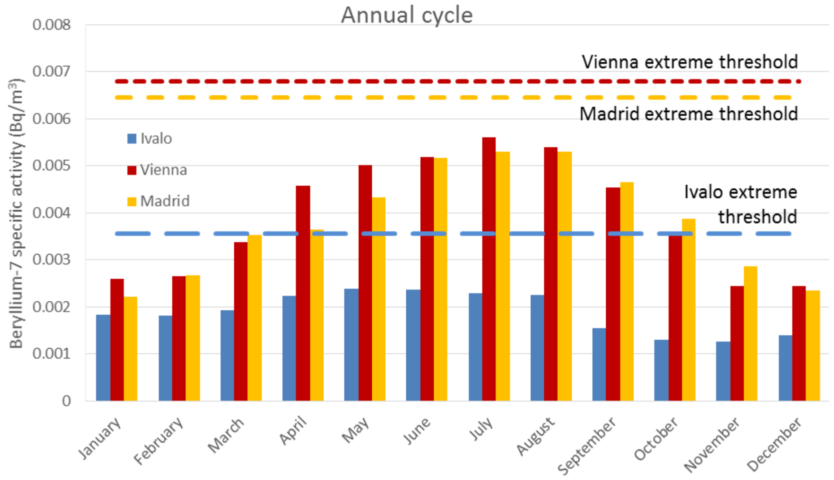

Fig. 2 Monthly mean Be-7 specific activities over the 2001–2010 period (bars) and extreme thresholds (95th percentile) (dashed lines) for Ivalo (blue), Vienna (red) and Madrid (yellow).

As expected, most of the extremes occurred during the warmer seasons. With an exception of Risoe, Bilthoven and Offenbach, the highest number of extremes was noted in summer. In Risoe, Bilthoven and Offenbach, however, most of the extremes occurred in spring, and specifically during the month of May: in Risoe, 6 of the total 13 spring extremes; in Bilthoven, 8 out of the total 16 spring extremes; and in Offenbach, 7 out of the total 11 spring extremes. In all the three stations, the annual maximum of the Be-7 specific activity occurred in the month of May.

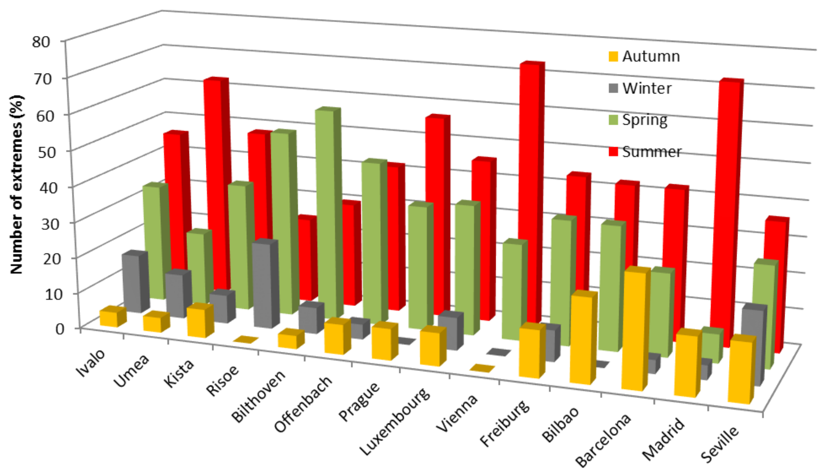

Fig. 3 Number of extremes in the Be-7 specific activity (as percent of the total number of extremes) per season.

Vienna was the only measurement site with no extremes during the colder half of the year. South of Vienna, however, there was a noticeable increase (higher than 10 %) in the percentage of the autumn maxima (Fig. 3). These autumn maxima constituted 23 % and 31% of the total number of extremes in Bilbao and Barcelona, respectively. Most of autumn extremes were observed in the month of September. For example, all four autumn extremes in Seville occurred in that month. In both Seville and Madrid, the maximum of the annual cycle happened in August which was later than in the other locations.

The timing of the annual maximum reflects on a different seasonal contribution to the total number of extremes. For example, as mentioned above, the southern stations exhibited the annual maximum in August, and at the same time, extreme Be-7 concentrations often occurred in September, giving a high contribution of the autumn extremes to the total.

To adjust for the time shift in the annual cycle, we propose a definition of “colder half of the year” that is specific for a given measurement location: a six-month period over which the mean monthly Be-7 concentration in surface air is lower than the median of the monthly means (this same criterion was previously used in Ajtić et al., 2016).

Figure 4 gives an example of the proposed definition for a northern and southern station. In Ivalo, the northern station, September to February are the months with the mean Be-7 concentration lower than the monthly median. In Madrid, the southern site, the cold half of the year starts in November and lasts until the following April. According to this definition, the months that traditionally fall into autumn, September and October, would be encompassed in the warmer half of the year for a number of measurement sites.

3.2. Regional distribution of common Be-7 extremes

Common (within +/- 3 days, as explained in Section 2.3) extremes were used to identify the spatial pattern of the highest Be-7 activity concentration occurrences.

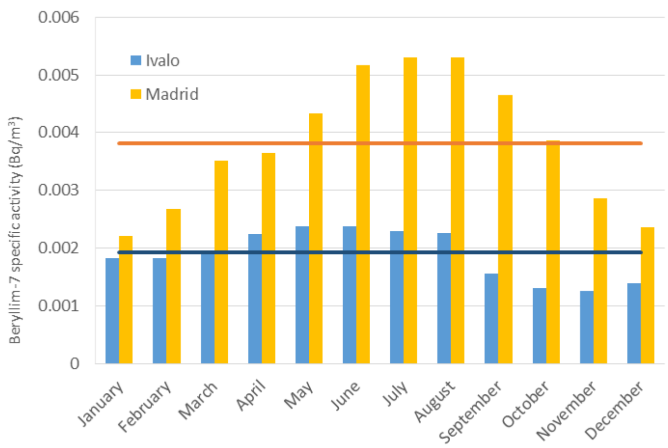

Fig. 4 Monthly mean Be-7 specific activities (bars) for Ivalo (blue) and Madrid (yellow) over the 2001–2010 period. The dark blue and red lines denote the median of the monthly means for Ivalo and Madrid, respectively.

First, for each measurement site, the number of mutual extremes with every other site was calculated. For example, for Umea, 37.5 % and 41.7 % out of the total 24 extreme events coincided with the extremes in Ivalo and Kista, respectively, while for all other sites, the percentage was less than 20 %. Similarly, for Risoe, 52 %, 36 % and 32 % of the total 25 extremes coincided with the extremes in Kista, Bilthoven and Prague, respectively, while the percentages for the other sites were less than 30 %.

It is worth noting that since one extreme event in any given location can coincide with the extremes in any number of other locations, the percentages need not sum to 100 %. Overall, there were 91 pairs of sites, and the highest percentage of mutual extremes was 52 % for the Kista–Risoe pair. Further, there were 16 pairs of the sites (18 % of the total 91 pairs) with the mutual extremes of 30 % or higher; and 6 pairs of the sites (8 % of the total 91 pairs) with the mutual extremes of 40 % or higher. These two values of the mutual extremes, 30 % and 40 %, were therefore taken as an indication of a strong and very strong association between the sites.

The spatial pattern of common extremes more frequent than 30 % divided Europe into three regions: north of 60 °N, between 45 °N and 60 °N, and south of 45 °N (Fig. 5). The northernmost region, encompassing Ivalo, Umea and Kista, seemed rather isolated from the rest of the locations. Ivalo and Umea showed a relatively strong isolation, while Kista shared some common extremes with more southern sites, Risoe and Prague. Similarly, common extremes more frequent than 30 % were almost uniform within the latitudinal belt 45–60 °N, in a sense that almost each site had common extremes with every other site in the belt. In other words, there was a strong association between all the sites within this region. Furthermore, this latitudinal belt was completely isolated from the southernmost locations. There were no preferred concurring sites south of 45 °N.

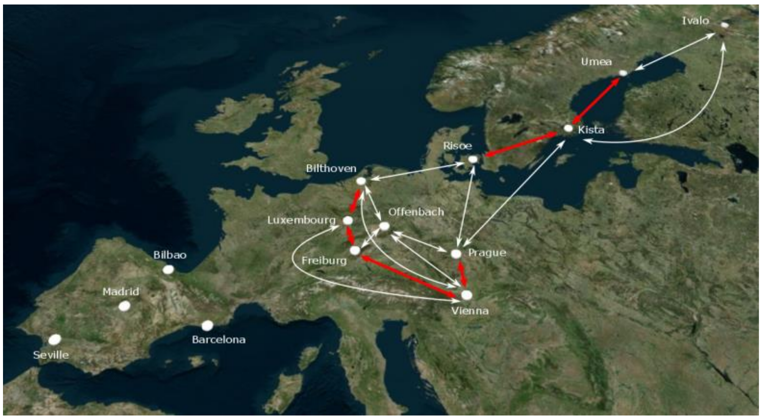

Fig. 5 Graphical representation of common extreme occurrences. The arrows show occurrences more frequent than 30 %; less frequent occurrences are not plotted. The occurrences of around 40 % or larger are given in red.

The northern separation line could alternatively be drawn south of Risoe, rather than between Risoe and Kista, since Risoe seems to be more strongly linked with Kista than with the other measurement sites (Fig. 5). In this case, the separation line would be closer to 50 °N which arose as a dividing latitude in the comparison of the annual cycles. These results agree very well with the three regions of distinct Be-7 concentration behavior identified by Ajtić et al. (2015) and Hernández-Ceballos et al. (2016b).

Ajtić et al. (2015) investigated an association between the Be-7 surface concentration and tropopause height at 21 measurement sites across Europe, while Hernández-Ceballos et al. (2016b) used cluster analysis technique on the monthly index to identify spatial patterns of similar Be-7 surface concentrations. These two studies and our present study used different methods to investigate the abundance of Be-7 in surface air, but got the same conclusion on its spatial variability, pointing out the existence of three different regions: northern, midlatitude and southern Europe.

3.3. Correlations between the measurement sites

To investigate whether the regional grouping of the measurement sites reflected on the inter-site correlations, we calculated the Spearman's correlation coefficients for the arrays consisting of the Be-7 monthly means (Tab. 1). The coefficients showed:

1) in general, the Be-7 specific activities across the investigated region of Europe were significantly correlated;

2) the magnitude of the correlation decreased with distance between the sites; and

3) all of the sites in midlatitudes registered strong correlations (with the coefficients larger than 0.7), but some strong correlations also existed within the northern and southern regions.

Table 1 Correlation matrix for the measurement sites (denoted by the first three letters of their names, except for Bilthoven – Bilt). The correlation coefficients larger than 0.7 and less than 0.5 are given in bold and light grey, respectively. The correlation coefficients for the sites with common extremes more frequent than 40 % (represented by the red arrows in Fig. 5) are given in red.

	Ume	Kis	Ris	Bilt	Off	Pra	Lux	Vie	Fre	Bil	Bar	Mad	Sev
Iva	**0.77**	**0.71**	0.52	0.42	0.46	0.50	0.47	0.46	0.37	0.40	0.33	0.37	0.38
Ume		**0.87**	0.61	0.50	0.52	0.61	0.52	0.58	0.45	0.40	0.44	0.49	0.42
Kis			**0.74**	0.60	0.62	**0.71**	0.61	0.67	0.57	0.49	0.48	0.56	0.51
Ris				0.61	0.48	0.57	0.48	0.55	0.47	0.40	0.34	0.35	0.41
Bilt					**0.83**	**0.77**	**0.82**	**0.72**	**0.73**	0.64	0.62	0.65	0.53
Off						**0.85**	**0.84**	**0.83**	**0.79**	0.60	0.65	**0.74**	0.58
Pra							**0.78**	**0.90**	**0.76**	0.56	0.68	**0.78**	0.57
Lux								**0.75**	**0.74**	0.61	0.64	0.67	0.55
Vie									**0.76**	0.53	**0.70**	**0.72**	0.59
Fre										0.68	**0.75**	**0.71**	0.60
Bil											0.67	0.68	0.66
Bar												**0.80**	0.61
Mad													0.67

Further, the measurement sites with frequent common extremes (Fig. 5) were strongly mutually correlated (Tab. 1). However, the correlation was not conspicuously higher than for the other pairs of locations. This could imply that the extreme Be-7 events might be caused by specific meteorological conditions, different from the average European patterns. This result is in agreement with the study of Hernández-Ceballos et al. (2017) where some high Be-7 surface activities over the Iberian Peninsula were associated with unusually fast stratosphere-to-troposphere exchange.

4. CONCLUSION

The annual cycles of the Be-7 specific activities in surface air observed at 14 European stations showed that, apart from a greater abundance of Be-7, the southern stations also had a shift of the annual maximum and minimum towards later in the year: the maximum moved from May for the northern stations, to August for the southern stations, while the minimum moved from November to January. Extremely high Be-7 surface concentrations also showed differences in their seasonal pattern. For example, no extreme concentrations occurred during autumn and winter in Vienna, while in all other locations, at least around 10 % of the total extremes occurred during these two seasons.

Nevertheless, the similarities in the investigated Be-7 arrays, as quantified by Spearman's correlation coefficients, were significant for all pairs of the measurement sites. The degree of correlation generally decreased with the distance between the locations.

Our analysis showed that the investigated European sites could be grouped in three regions with similar Be-7 characteristics in terms of the occurrence of the extremes and annual patterns: north of 55 °N, between 45 °N and 55 °N, and south of 45 °N. Within the northern and midlatitude regions, the extremes in the Be-7 surface concentrations frequently coincided, and the correlations between the locations were generally very strong. The southern region, on the other hand, showed a lack of common extremes, and generally lower correlations. However, this result could possibly depend on the exclusion of the measurement sites south of Madrid. This exclusion arose from our choice of the measurement sites which was based on the similarity in the sampling patterns.

A future analysis, based on the methodology presented in this paper, could expand the investigated region of Europe by including more sampling locations. The spatial coverage in our study was limited not only by different sampling frequencies of the available datasets, but also by the number of contributing sites to the REMdb. An inclusion of the Be-7 surface concentration measurements conducted in other parts of Europe would not only broaden the spatial coverage but also add more confidence in the obtained results.

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EKSTREMNO VISOKE VREDNOSTI SPECIFIČNE AKTIVNOSTI BERILIJUMA-7 U PRIZEMNOM SLOJU ATMOSFERE U EVROPI

Sezonska i prostorna raspodela ekstremno visokih specifičnih aktivnosti berilijuma-7 u prizemnom sloju atmosfere analizirane su u ovom radu. Merenja tokom 2001–2010. godine na 14 evropskih stanica preuzeta su iz REMdb–baze podataka Monitoring radioaktivnosti u životnoj sredini. Prvo su razmotreni godišnji ciklusi specifične aktivnosti berillijuma-7 koji pokazuju da se godišnje maksimalne i minimalne srednje mesečne vrednosti pomeraju ka kasnijim mesecima kako se smanjuje geografska širina merne stanice. Za ekstremno visoke vrednosti specifične aktivnosti berilijuma-7 uzete su vrednosti veće od 95-og percentila, koji je izračunat za svaku stanicu posebno. Najveći broj ovih ekstrema događa se od marta do avgusta, a najmanje 10 % od ukupnog broja ekstrema tokom jeseni i zime. Izuzetak je merna stanica Beč na kojoj su se svi ekstremi dogodili tokom proleća i leta. Rasprostranjenost ekstrema koji su zajednički za parove mernih mesta, ukazuje da postoje tri različita regiona u Evropi: severno od 55 °N, između 45 °N i 55 °N, i južno od 45 °N. Iako su merenja specifične aktivnosti berilijuma-7 značajno korelisana za sve analizirane stanice, najveći koeficijenti korelacije dobijeni su u okviru ovih regiona.

Ključne reči: *berilijum-7, godišnji ciklus, maksimalne koncentracije, sezonska raspodela, korelacije, Evropa*