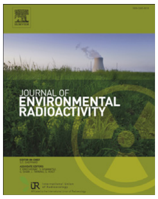
Journal of Environmental Radioactivity 155-156 (2016) 55–62

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad



Identification of airborne radioactive spatial patterns in Europe – Feasibility study using Beryllium-7

M.A. Hernández-Ceballos*, G. Cinelli, T. Tollefsen, M. Marín-Ferrer

European Commission, Joint Research Centre (JRC), Institute for Transuranium Elements (ITU), Via Enrico Fermi 2749, Ispra, VA 21027, Italy

ARTICLE INFO

Article history:
Received 25 August 2015
Received in revised form
27 January 2016
Accepted 7 February 2016
Available online 23 February 2016

Keywords:
Cluster methodology
Monthly index
^{7}Be
Tropopause height
Europe

ABSTRACT

The present study proposes a methodology to identify spatial patterns in airborne radioactive particles in Europe. The methodology is based on transforming the activity concentrations in the set of stations for each month (monthly index), due to the tightly spaced sampling intervals (daily to monthly), in combination with hierarchical and non-hierarchical clustering approaches, due to the lack of a priori knowledge of the number of clusters to be created. Three different hierarchical cluster methodologies are explored to set the optimal number of clusters necessary to initialize the non-hierarchical one (k-means).

To evaluate this methodology, cosmogenic beryllium-7 (^{7}Be) data, collected between 2007 and 2010 at 19 sampling stations in European Union (EU) countries and stored in the Radioactivity Environmental Monitoring (REM) database, are used. This methodology yields a solution with three distinguishable clusters (south, central and north), each with a different evolution of the ^{7}Be monthly index. Clear differences between monthly indices are shown in both intensity and time trends, following a latitudinal distribution of the sampling stations.

This cluster result is evaluated performing ANOVA analysis, considering the original ^{7}Be activity concentrations grouped in each cluster. The statistical results (among clusters and sampling stations within clusters) confirm the spatial distribution of ^{7}Be in Europe, and, hence, reinforce the use of this methodology. Finally, the impact of tropopause height on this grouping is successfully tested, suggesting its influence on the spatial distribution of ^{7}Be in Europe.

For airborne radioactive particles the analysis gave valuable results that improve knowledge of these atmospheric compounds in Europe. Hence, this work addresses a methodology to a grouping of airborne sampling stations, 1) allowing a better understanding of the distribution of ^{7}Be activity concentrations in the EU, and 2) serving as a basis for further investigation of the heterogeneity of airborne radioactivity concentrations in Europe.

1. Introduction

The airborne radioactive particles have an important radiological significance because of their health impact effects (e.g. Radon (Bowie and Bowie, 1991)) or because they may act as tracers for atmospheric processes (e.g. ^{7}Be and ^{210}Pb (Lozano et al., 2012). Due to their importance, there are worldwide networks for radionuclides measurements (e.g. EUropean Radiological Data Exchange Platform (EURDEP) and Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO)) with the purpose of monitoring its temporal and spatial variability.

These networks are composed of stations placed in a wide range of geographic locations. Many authors have demonstrated the impact of topographic (mountains, valleys, plateaus) (Gerasopoulos et al., 2003) and meteorological conditions (Krajny et al., 2014) on the variability of radioactive particles measurements. In Europe, many studies have described radioactive concentrations in surface air at single sampling stations (Carvalho et al., 2013; Tositti et al., 2014) pointing out the large set of heterogeneous sampling stations in terms of airborne radioactivity concentrations. This fact is also well observed considering recent analysis carried out (Kulan, 2006; Hernández-Ceballos et al., 2015).

A comprehensive analysis of radionuclides distribution at European level identifying spatial distribution patterns has not yet been undertaken. This analysis would allow a better understanding of the heterogeneity in activity concentrations measured in the EU,

---

* Corresponding author.
E-mail address: miguelhceballos@gmail.com (M.A. Hernández-Ceballos).

http://dx.doi.org/10.1016/j.jenvrad.2016.02.006

and it would allow the identification of areas in which radioactive concentrations present similar patterns. This information would be useful, for instance, to identify and discriminate anomalies (false positive and negative) in measurements carried out in sampling stations.

Considering these benefits, with the purpose of exploiting the similarities/dissimilarities among sampling stations in the EU, we propose a methodology based on 1) transforming the activity concentrations in each station to monthly index values, and 2) combining hierarchical and non-hierarchical clustering approaches, to identify spatial patterns in airborne radioactive particles.

In the case of airborne radioactive particles, the use of different tightly spaced sampling intervals (daily, weekly, monthly, quarterly) and the non-homogeneity in the definition of the sampling period (start-end) limit the direct use of the local sampling databases in cluster analysis. In this sense, a previous homogenization process of the databases is required. To overcome this restriction, the calculation of the monthly index is suggested and tested in the present work to be taken as reference to apply cluster analysis on radioactivity concentrations. This dimensionless magnitude has been previously used to study airborne radioactive concentrations (Todorovic et al., 1999; Kulan, 2006), because it is appropriate for the direct comparison of seasonal fluctuations with highly differing activities and/or substantial long-term changes.

On the other hand, cluster analysis is widely applied in research studies in order to define groups that help to simplify and understand a large quantity of information on the basis of a specific aggregation variable (e.g. Adame et al., 2012; Wegner et al., 2012). Two different approaches based on these algorithms can be used to simplify information, viz. hierarchical and non-hierarchical (Everitt, 1980). In the present work, following the suggestions of Maitra et al. (2010), and recently applied in Austin et al. (2013), these two algorithms are combined, based on the use of hierarchical cluster methodologies to initialize the non-hierarchical one (k-means).

To evaluate this methodology, we have used the ^{7}Be activity concentrations measured in surface air from 2007 to 2010 by EU Members States in 19 sampling stations and stored in the Radioactivity Environmental Monitoring (REM) database at the Institute for Transuranium Elements of the European Commission, Joint Research Centre (JRC) in Ispra (Italy) (http://rem.jrc.ec.europa.eu/). The validity of the results is evaluated statistically by applying the t-test and ANOVA analyses.

Taking into account the positive correlation between the tropopause height and the concentration of ^{7}Be in the surface air reported in Ioannidou et al. (2014), the present work also performs a first approach to understand the impact of this parameter on the ^{7}Be spatial distribution in EU. The positive correlation between both parameters would reflect both downward transport of upper tropospheric air within anticyclonic conditions and lower scavenging rates.

Therefore, this study intends to address the following issues:

- To evaluate a methodology for further clustering research in case of tightly spaced sampling intervals;
- To identify regions with similar behaviour of ^{7}Be activity concentrations in the EU;
- To suggest the influence of tropopause height on the space distribution of ^{7}Be activity concentrations.

2. Materials and methods

2.1. Database and sampling methods

Within the European Commission, the Radioactivity Environmental Monitoring database (REMdb) is a unique resource of consistently structured information for anyone wishing to handle, analyse and compare environmental radioactivity data. The data have been obtained from data transmitted directly to the Commission by the national competent authorities. More information on the REM database can be found on the web site (http://rem.jrc.ec.europa.eu/RemWeb/activities/Remdb.aspx). The ^{7}Be database stored in the REMdb is public until 2006, and the access to the data corresponding to the 2007–2011 period can be granted only after explicit request.

Taking as reference the set of stations belonging to the sparse monitoring network of REMdb (34 sampling stations in total), which integrates a number of representative locations in each EU country with high-sensitivity measurements of ^{7}Be, the selection of the stations in this work is a compromise between 1) working with the maximum number of stations, and 2) having a representative set of ^{7}Be data to depict its yearly, seasonal and monthly variability in each station, in order to obtain an illustrative set of results.

With the aim to achieve these requirements, the ^{7}Be measurements between 2007 and 2010 at the 19 sampling locations indicated in Table 1 and shown in Fig. 1 were used in the present work. These measurements were mainly collected in weekly sampling intervals, although in Helsinki were mainly obtained with daily temporal resolution. This reference period has been selected to use a large number of available stations with weekly data availability greater than 60% (more than 136 considering the total number of weeks during 2007–2010 (228)). Table 1 shows that most of the stations have a greater availability of data than the expected one (even above 90%), ensuring an adequate representativeness of the results.

Airborne particulate sampling was carried out by pumping air through filters at a flow rate of several hundred cubic metres per day. Further information on the procedure to collect aerosol samples from ground level air at each sampling station can be consulted in the Supplementary Material (Table S1).

2.2. Calculation of monthly index

The heterogeneity of the temporal sampling measurements within each station limits the direct comparison of data among them. For this reason, to be able to apply a cluster analysis it is necessary to transform the measurements. In this work, we have used the monthly index defined in Ferris (1998) and Todorovic et al. (1999).

Briefly, Eq. (1)–(3) show the way to calculate the monthly index. It is calculated as the ratio between mean monthly activity and the overall average activity in the selected period. For each time period, the activity is expressed in percent (%) of the monthly average. The monthly index values for the selected period are estimated using the mean value of the weekly activity (W_n), the average of the weekly means over one year (OAPA) and the weekly index (W_i) values as follows,

$$OAPA = \frac{\sum_{n=1}^{52} W_n}{N} \tag{1}$$

$$Weekly\ Index\ (W_i) = \frac{W_n}{OAPA} \tag{2}$$