Atmospheric Environment 87 (2014) 108–122

Contents lists available at ScienceDirect

Atmospheric Environment

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

12 years of ^{7}Be and ^{210}Pb in Mt. Cimone, and their correlation with meteorological parameters

L. Tositti [a], E. Brattich [a,b,*], G. Cinelli [b,c], D. Baldacci [a]

[a] Environ. Chemistry and Radioactivity Lab., Dept. of Chemistry "G. Ciamician", Alma Mater Studiorum University of Bologna, Via Selmi 2, 40126 Bologna, BO, Italy
[b] Dept. of Biological, Geological and Environmental Sciences – Section of Geology, Alma Mater Studiorum University of Bologna, Piazza di Porta San Donato 1, 40126 Bologna, BO, Italy
[c] European Commission, DG JRC, Institute for Transuranium Elements, Via E Fermi 2749, I-21027 Ispra, VA, Italy

HIGHLIGHTS

- ^{7}Be and ^{210}Pb activities over the period 1998–2011.
- Seasonal/interannual variations of ^{7}Be and ^{210}Pb and of their activity ratio.
- Frequency distribution of radiotracers and of other atmospheric variables.
- Correlations of radiotracers with meteorological and compositional atmospheric variables.
- Possible use of the studied radiotracers as potential proxies is examined.

ARTICLE INFO

Article history:
Received 12 August 2013
Received in revised form
4 January 2014
Accepted 6 January 2014
Available online 15 January 2014

Keywords:
Atmospheric radiotracers
^{7}Be
^{210}Pb
Stratosphere-to-troposphere exchange
Ambient aerosol
Saharan Dust
PM10

ABSTRACT

Simultaneous measurements of airborne radionuclides ^{7}Be and ^{210}Pb, together with aerosol mass load PM_{10}, have been routinely carried out at the Global WMO-GAW station of Mt. Cimone (Italy, 2165 m a.s.l., 44° 12′ N, 10° 42′ E) from 1998 to 2011. The experimental activity was started with the purpose of gaining a better understanding of the vertical and horizontal transports taking place at this site affecting the atmospheric chemical composition. The time series of the collected data is presented and discussed in this paper. The ^{7}Be concentrations in this period are in the range 0.05–15.8 mBq m^{-3} with the presence of two distinct relative maxima during winter/spring and summer, suggesting an origin from different physical processes. The ^{210}Pb concentrations collected during the period are in the range 0.05–2.30 mBq m^{-3} and are characterized by a single maximum during the warm period. The $^{7}Be/^{210}Pb$ ratio was in the range 0.5–127.8 and is characterized by a maximum during the cold period. The frequency distributions of the three parameters and the seasonal/interannual variabilities are investigated and presented.

1. Introduction

In the course of the last decades airborne radionuclides have long been investigated within the framework of atmospheric science. Initially the focus concerned the emission of artificial radioactivity during weapon testing (see for example chapter 9 of Eisenbud and Gesell, 1997; Pállson et al., 2013) which pointed out to the scientists both the safety issues connected with radioactivity hazard as well as the remarkable efficiency of atmospheric transport processes at the global scale. It was soon recognized that atmospheric radioactivity had also a not negligible background component capable to trace both the gaseous and the particulate phases enabling the quantitative description of fundamental processes of atmospheric dynamics. Airborne radioactivity has long been playing a relevant role in the study of atmospheric transport processes as detectable from the frequency of scientific publications (Burton and Stewart, 1960; Junge, 1963; Reiter et al., 1971; Gaggeler, 1995; Arimoto et al., 1999; Turekian and Graustein, 2003; WMO-GAW Report n. 155, 2004; Dibb, 2007; Papastefanou,

* Corresponding author. Environ. Chemistry and Radioactivity Lab., Dept. of Chemistry "G. Ciamician", Alma Mater Studiorum University of Bologna, Via Selmi 2, 40126 Bologna, BO, Italy.
E-mail addresses: laura.tositti@unibo.it (L. Tositti), erika.brattich@unibo.it (E. Brattich), giorgia.cinelli@jrc.ec.europa.eu (G. Cinelli), daniela.baldacci@unibo.it (D. Baldacci).

1352-2310/$ – see front matter © 2014 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.atmosenv.2014.01.014

2008; Rastogi and Sarin, 2008; Froehlich and Masarik, 2010; Sykora and Froehlich, 2010; Lozano et al., 2011).

At present nuclear safety is still a basic issue at the global scale as demonstrated by the follow up of Chernobyl and Fukushima accidents (see for example Papastefanou et al., 1988; Hötzl et al., 1992; Davison et al., 1993; Vakulovsky et al., 1994; chapter 12 of Eisenbud and Gesell, 1997; Diaz Leon et al., 2011; Lozano et al., 2011; Manolopoulou et al., 2011; Masson et al., 2011; Pittauerová et al., 2011; Tositti et al., 2012; Ioannidou et al., 2013) or as a result of episodic cases such as the fall of nuke-fed satellites (Cosmos 954, Kosmos 1402, see for example chapter 12 of Eisenbud and Gesell, 1997) or the accidental melting of orphan sources/metals scraps in high temperature processes (e.g., Algeciras accident, Krysta and Bocquet, 2007; on this occasion ^{137}Cs, a radionuclide which is usually below detection limit at Mt. Cimone, was detected at this site in two samples at very low levels). Moreover the need for monitoring potential violations of the Nuclear Ban Treaty has recently promoted the constitution of a global network for artificial radioactivity whose efficiency has been successfully tested following the recent Fukushima emergency (Masson et al., 2011; Hernández-Ceballos et al., 2012; Thakur et al., 2013).

Cosmogenic and naturally occurring radionuclides have long been investigated either per se or as normalizing and reference factors in the study of artificial radioactivity or again both as efficient tracers in environmental science and as geochronometers. Altogether this field of research has been crucial to the comprehension of several basic processes such as interhemispheric transport, stratosphere-to-troposphere exchange (STE) and time scales of atmospheric dynamics, while posing the basis for basic concepts in environmental science such as biogeochemistry, environmental monitoring management and exposure/dosimetry concepts presently extended to stable “classic” pollutants such as for example ozone and/or airborne particulate matter.

At present though the application of radiotracers constitutes a niche approach, the simultaneous use of artificial and natural radiotracers still provides a solid background for the characterization of atmospheric transport (Arimoto et al., 1999; Paatero and Hatakka, 2000; Dueñas et al., 2011) the testing of atmospheric models (Koch et al., 1996; Liu et al., 2001, 2004; Heinrich and Jamelot, 2011; Christoudias and Lelieveld, 2013) as well as in supporting source apportionment of pollutants (Li et al., 2002; Cuevas et al., 2013).

Among the most used naturally occurring radionuclides there are ^{7}Be, ^{210}Pb, ^{222}Rn and others included in the group of the key atmospheric components that should be routinely monitored within the WMO-GAW network (WMO-GAW Report n. 155, 2004). In particular the importance of ^{210}Pb and ^{7}Be relies upon their distinct natural sources. ^{210}Pb (half-life, $T_{1/2} = 22.1$ years) is supplied to the atmosphere at ground level by the radioactive decay of its precursor, ^{222}Rn ($T_{1/2} = 3.83$ days). As the ^{222}Rn flux from the ocean is negligible, ^{210}Pb is considered a continental tracer of air masses (Balkanski et al., 1983; Turekian et al., 1983; Baskaran, 2011). In contrast, ^{7}Be is a relatively short lived ($T_{1/2} = 53.3$ days) radionuclide of cosmogenic origin, produced by cosmic ray spallation reactions with light atmospheric nuclei of nitrogen and oxygen (Usoskin et al., 2009): most of the ^{7}Be production ($\sim$75%) occurs in the stratosphere while the remaining part ($\sim$25%) is produced in the troposphere, and particularly in the upper troposphere (Johnson and Viezee, 1981; Usoskin and Kovaltsov, 2008). The production rate of ^{7}Be has a latitudinal dependence (Ioannidou et al., 2005), while it has a negligible dependence from season and longitude, but a remarkable variation due to the 11-year solar cycle (Hötzl et al., 1991; Megumi et al., 2000; Cannizzaro et al., 2004; Ioannidou et al., 2005; Leppanen et al., 2012). Once formed, ^{7}Be and ^{210}Pb undergo rapid association onto submicron-sized aerosol particles both peaking in the accumulation mode (Papastefanou and Ioannidou, 1995; Winkler et al., 1998; Gaffney et al., 2004; Ioannidou et al., 2005). Thereafter, ^{7}Be and ^{210}Pb are removed from the atmosphere by wet and dry scavenging of the carrier aerosol (Feely et al., 1989; Kulan et al., 2006). Most of the ^{7}Be produced in the stratosphere does not readily reach the troposphere because of its short half-life compared to the longer residence times of aerosols in the stratosphere (which, depending on the size of the particles, is equal to one or more years as estimated from Hamill et al., 1997; Rasch et al., 2008). In fact, the relatively high production rates of ^{7}Be in the upper troposphere (UT), combined with transport from the lower stratosphere (LS) to the upper troposphere usually maintain a steep vertical concentration gradient between the upper and the lower troposphere (Feely et al., 1989). Nevertheless, the UT–LS may cause high ^{7}Be concentrations in the surface air, easily detectable at a high altitude stations such as for example Mt. Cimone station (Bonasoni et al., 1999, 2000a, 2000b; Cristofanelli et al., 2003, 2006; Cristofanelli et al., 2009a).

Due to the similar tropospheric physico-chemical behaviour, variations in the ^{7}Be/^{210}Pb ratios reflect both vertical and horizontal transport in the atmosphere. Because of the different origins of the two radionuclides, the use of the combination of ^{7}Be and ^{210}Pb as activity ratio has been shown to provide clearer information about the origin of the air masses (Graustein and Turekian, 1996; Bonasoni et al., 2000a, 2000b, 2004; Zheng et al., 2005), and its seasonal variability over continents has been studied for examining vertical exchange transport processes (Koch et al., 1996). The simultaneous measurements of ^{7}Be and ^{210}Pb, together with their ratio can provide useful information about the vertical motion of air masses as well as on convective activity in the troposphere (Brost et al., 1991; Koch et al., 1996; Lee et al., 2004; Tositti et al., 2004; Lee et al., 2007). Recently, Lozano et al. (2012) studied the different synoptic patterns and air masses types associated to ranges of ^{7}Be and ^{210}Pb activity concentrations in the southwestern Iberian Peninsula, indicating the differences between the arrival of maritime and continental air masses and confirming that both radionuclides can be used as two independent atmospheric transport markers.

In this work we present a basic overview of the time series of ^{7}Be and ^{210}Pb collected at the WMO-GAW station of Mt. Cimone from 1998 until 2011. This activity has been already the object of several papers devoted to specific topics, in particular the use of ^{7}Be in STE (Stratosphere-to-Troposphere Exchange), a rather classic application of this radionuclide, though not thoroughly understood yet (Bonasoni et al., 1999, 2000a, 2000b; Cristofanelli et al., 2003; Cristofanelli et al., 2006; Cristofanelli et al., 2009a). The follow up of Fukushima accident was also investigated at this station and results have been recently published either at the European scale in a collective paper by Masson et al. (2011) or at the regional scale comparing the radionuclidic pool at two nearby stations including Mt. Cimone (Tositti et al., 2012). Finally another recent paper concerned the PM_{10} matrix in which pioneristically all the radionuclides herein treated are measured since the beginning of this experiment, providing a long-term overview of PM_{10} behaviour in the core of the Mediterranean region (Tositti et al., 2013). This paper presents and discusses a statistical analysis of frequency distributions, seasonality, interannual variation, correlations of the ^{7}Be, ^{210}Pb (and their ratio) data of acquired from 1998 to 2011 at the WMO-GAW station of Mt. Cimone, with the purpose of gaining better insights into the different physical mechanisms at the basis of their variabilities.

2. Material and methods

2.1. Measurement site

Mt. Cimone station (44°12′ N, 10° 42′ E) is located on top of the highest peak of the Italian Northern Apennines (2165 m a.s.l.). The