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Air mass origin and its influence on radionuclide activities (^{7}Be and ^{210}Pb) in aerosol particles at a coastal site in the western Mediterranean

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ABSTRACT

Studies of radionuclide activities in aerosol particles provide a means for evaluating the integrated effects of transport and meteorology on the atmospheric loadings of substances with different sources. Measurements of aerosol mass concentration and specific activities of ^{7}Be and ^{210}Pb in aerosols at Málaga (36° 43′ 40″ N; 4° 28′ 8″ W) for the period 2000–2006 were used to obtain the relationships between radionuclide activities and airflow patterns by comparing the data grouped by air mass trajectory clusters. The average concentration values of ^{7}Be and ^{210}Pb over the 7 year period have been found to be 4.6 and 0.58 mBq m^{-3}, respectively, with mean aerosol mass concentration of 53.6 μg m^{-3}. The identified air flow types arriving at Málaga reflect the transitional location of the Iberian Peninsula and show significant differences in radionuclide activities. Air concentrations of both nuclides and the aerosol mass concentration are controlled predominantly by the synoptic scenarios leading to the entrance of dust-laden continental flows from northern Africa and the arrival of polar maritime air masses, as implied by the strong correlations found between the monthly frequencies of the different air masses and the specific activities of both radionuclides. Correlations between activity concentrations and precipitation are significant though lower than with air masses.

1. Introduction

Lead-210 and ^{7}Be, both naturally occurring radionuclides, have been used as tracers of air mass history owing to their unique sources. Lead-210 (half-life, $T_{1/2}=22$ years) is supplied to the atmosphere by the radioactive decay of ^{222}Rn ($T_{1/2}=3.8$ days), a noble gas produced from the decay of primordial ^{238}U. Radon-222 diffuses from soils into the continental boundary layer where it decays through several short-lived daughters to ^{210}Pb which rapidly attaches to existing aerosol particles. The ^{222}Rn flux from the ocean is negligible, and therefore ^{210}Pb is useful as a tracer of continental emissions (Turekian et al., 1983; Balkanski et al., 1993). In contrast, ^{7}Be is produced via cosmic ray spallation reactions with nitrogen and oxygen in the stratosphere and upper troposphere. Most ^{7}Be production (~67%) occurs in the stratosphere and the remaining part (~33%) is produced in the troposphere, particularly in the upper troposphere (Lal and Peters, 1967). This is largely a result of the long residence time of aerosols in the stratosphere compared with the 53.3-day radioactive half-life of ^{7}Be.

Backward-in-time trajectory analysis is traditionally used to identify synoptic-scale atmospheric transport patterns and determine potential source areas of air tracers. Backtrajectories have frequently been utilized for the interpretation of individual pollution episodes, although there are uncertainties in the computation of single trajectories arising from the input meteorological fields and the numerical models (Stohl, 1998, and references therein; Harris et al., 2005; Cabello et al.,

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2008a). The errors associated to trajectory calculation are on the order of 15–20% of the distance traveled after a few days (Stohl, 1998); however the statistical analysis of large numbers of trajectories arriving at a site over relatively long periods of time should minimize errors from single trajectories, thus increasing the accuracy of the results. Therefore, cluster analysis is now frequently used as a suitable technique to classify the air masses arriving at a study site (Moody and Galloway, 1988; Dorling et al., 1992; Brankov et al., 1998; Mattis, 2001; Jorba et al., 2004; Salvador et al., 2004; Cabello et al., 2008b). Even though backtrajectories do not account for turbulence and other atmospheric processes (Stohl et al., 2002), at the synoptic scale the representative trajectory of each cluster is in general related to different circulation types.

The concentrations of radon and radon daughters, particularly those of ^{210}Pb, are controlled namely by large scale meteorological conditions, especially through the alternate intrusion of either oceanic or continental air masses into the local surface air (Carvalho, 1995). Backtrajectory analysis shows that the duration of air mass travel over the continent before reaching a study site is strongly correlated with ^{210}Pb concentrations, as found at Geneva (Caillet et al., 2001) and Edinburgh (Likuku, 2006), and to a lesser extent at Málaga (Dueñas et al., 2009), suggesting that air masses are progressively enriched with ^{210}Pb along their pathway over land. Beryllium 7 does not present such an association, as would be expected from its cosmogenic origin.

A trajectory cluster analysis for Bermuda Island (Arimoto et al., 1999) associates the highest loadings of ^{7}Be and ^{210}Pb to transport from the NW of Bermuda, while low activities are linked to transport from the E and SE. Advected African dust and ^{210}Pb showed different seasonal and interannual trends. Results for ^{7}Be are in agreement with the work of Moody et al. (1995) that associate its highest values at Bermuda to subsiding flow behind surface cold fronts moving eastward over the North Atlantic. The influence of such downward transport at the northern Apennines and the Alps has been reported by Bonasoni et al. (2000) and Zanis et al. (2003), respectively. Episodes of high ^{7}Be concentrations in Canary Islands are also attributed to subsidence (Hernandez et al., 2008) and may be concurrent with African dust outbreaks. The association between the advection of African dust and the activities of ^{7}Be and ^{210}Pb in continental Europe, however, has not been addressed yet.

The transitional location of the Iberian Peninsula (IP) between subtropical and mid-latitudes, at the southwestern edge of the European continent and strongly influenced by its proximity to Africa, and between two large water bodies (the Atlantic Ocean and the Mediterranean Sea), induces a variety of circulations with distinct aerosol loadings (Querol et al., 2004, 2008; Salvador et al., 2004; Borge et al., 2007; Cabello et al., 2008b).

Dueñas et al. (2009) studied the correlation between the specific activities of ^{7}Be and ^{210}Pb and some meteorological variables, but the correlations found are signs of a synoptic scale weather influence rather than local effects. It is necessary therefore to determine the influence of the atmospheric transport patterns on the aerosol loading and the specific activities of both radionuclides.

In this paper we use the data for ^{7}Be and ^{210}Pb to investigate how the composition of aerosol particles varies in response to transport pathways and precipitation scavenging. The aims of this work were:

- To identify the main flows arriving at Málaga (southern Spain) at 3000, 1500 and 500 m height by means of cluster analysis of backtrajectories for a 7-year period.
- To obtain the relationships between the specific activities of ^{7}Be, ^{210}Pb and the aerosol mass concentration on a monthly scale and the airflow patterns in Málaga.
- To study the influence of precipitation on the activities of ^{7}Be and ^{210}Pb.

2. Material and methods

Airborne dust samples were collected at a height of 12 m above the ground to minimize the number of dust particles entering the sampler. The sampling station was located at the Faculty of Sciences of the University of Málaga (36° 43′ 40″ N; 4° 28′ 8″ W) in the north-west of the city. Málaga is the major coastal city of the Andalusia region, south Spain, with a population of 570,000 inhabitants. The sampling point was located approximately 5 km from the coastline, near the airport and surrounded by roads with traffic exhaust. Aerosol samples were collected weekly in cellulose membrane filters of 0.8 μm pore size, 5 cm diameter with an air flow rate of approximately 40 l min^{-1}. The air sample was lodged in an all-weather sampling station. Measurements by gamma-spectrometry were performed to determine the ^{210}Pb and ^{7}Be activities of the samples using a coaxial-type germanium detector (Canberra Industries Inc., USA). The gamma lines of 477.7 keV and 46.5 keV were used, respectively. A monthly composite sample containing 4–5 filters was formed (average volume of 1600 m^{3}) for the ^{210}Pb and ^{7}Be determination. Details of the low-background gamma-ray detection system used and the calibration of the resulting samples for gamma spectrometry have been previously described (Dueñas et al., 1999, 2002). The counting intervals ranged from 172,800 to 216,000 s. The errors reported are propagated errors arising from the one sigma counting uncertainty due to detector calibration and background correction. The concentrations were corrected for decay to the mid-collection period.

Dust content on the filters was determined by weighting the filters before and after exposure under the same conditions of temperature and humidity (Metler AJ100 analytical balance). The difference between the two weights gives the aerosol mass concentration in μg m^{-3}.

For the analysis of the origin of air masses arriving in the study site, 96-hour kinematic 3D backtrajectories starting at 0, 6, 12 and 18 UTC each day during the period 2000–2006 at three heights (3000, 1500 and 500 m a.s.l.) were computed with the HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT, version 4.8) model (Draxler and Rolph, 2003). FNL meteorological data from the FiNaL run in the series of NCEP operational model runs, available at the NOAA Air Resources Laboratory, was used.

Cluster analysis comprises a series of multivariate techniques for classifying a large data set into *non-predefined* dominant groups called clusters such that variance within each cluster is minimized and variance between clusters is maximized. The trajectory classification was performed by a k-means cluster analysis with hourly latitude and longitude as input variables and the Euclidean distance as similarity metrics. Beyond the selection of the type of clustering technique, the variables to cluster and the measure of similarity, the clustering algorithm involves some subjective nontrivial decisions. In the case of the k-means analysis, these are determining the best value for the number of clusters to retain and handling the sensitivity of the final result to the selection of centroids in the initialization stage.

The procedure described by Dorling et al. (1992) to reduce the subjectivity in the selection of the appropriate number of clusters was mainly followed: the algorithm was run for a range of cluster numbers between 30 and 2, and the percentage change in the total RMSD (i.e. the summation over the clusters of the root mean square deviation of each trajectory from its cluster mean) when the number of clusters is reduced from $k+1$ to k was used to find out the proper number of clusters. It was selected as the smallest k for which the percentage change in total RMSD is the smallest. The dependence of the final cluster solution on the selection of the initial centroids was handled by obtaining an ensemble of 50,000 solutions (from 50,000 k-means analysis, each one with initial centroids randomly chosen from the real trajectories) for each k; the solution with the smallest total RMSD (the clustering figure of merit) for each k was retained for the subsequent analysis of the proper number of clusters as mentioned above. This clustering methodology has been already used to relate air masses and aerosol size distributions at SE Spain (Cabello et al., 2008b). Details of the methodology and its comparison with the procedures of Dorling et al. (1992) and Mattis (2001) will be published elsewhere.

Composite synoptic charts of 700, 850 and 1000 hPa geopotential height for the days assigned to each of the identified clusters (not shown) were computed with data from the National Center for Environmental Prediction (NCEP)/ National Center for Atmospheric Research (NCAR) re-analysis project database (Kalnay et al., 1996). Synoptic average charts were useful to interpret the different atmospheric transport scenarios. The NCEP/NCAR re-analysis data are readily available from the Earth System Research Laboratory, Physical Sciences Division, of the USA National Oceanic and Atmospheric Administration at http://www.esrl.noaa.gov/psd/.

3. Results

3.1. Air-mass trajectory clusters

Trajectories are found to be clustered into 6 groups at each arriving height of 3000, 1500 and 500 m, according to the small increase in total RMSD observed when reducing from 7 to 6 clusters (not shown). Fig. 1 shows the backtrajectories assigned to each cluster and its representative trajectory (centroid). Most of the 3000 m trajectories correspond to westerly flows, identified as northwesterlies (NWs) of different speed (according to the length of the 96-hour trajectories in each cluster, NWfast, NWmod and NWslow), moderately short westerlies–southwesterlies (SW) and zonal (W) flows. The remaining trajectories are grouped into a cluster composed mainly by slow continental northerly–northeasterly flows (N–Eu). Additional types of flows are identified at 1500 and 500 m as eastern (Med) and western (WR) slow flows. At 1500 and 500 m there is an elevated occurrence of slow flows and recirculations associated with low pressure gradient situations.

In the Iberian Peninsula, the subtropical high pressure center over the Azores Islands and the Icelandic low control surface circulation at the synoptic scale. Their relative displacements and intensities allow a variety of circulations with distinct physical properties (Font, 2000). The Azores high is displaced to lower latitudes in winter and therefore the IP is affected by zonal (westerly) circulations combined with the perturbations that originated from the polar front. The Azores high may also expand eastwards to western Europe, inducing highly stable situations in the IP. In summer, the high pressure center expands towards higher latitudes, blocking the western/north western circulations over the IP, leading to frequent very weak synoptic forcing at the surface level that allows the development of the Iberian thermal low and of meso-scale circulations.

Fig. 2 shows the monthly variation of the occurrence of the identified air flow types arriving at 3000, 1500 and 500 m. A short description of the identified air masses and flows is given in the following:

NW_{fast}: Advection of polar maritime fast air masses that originate as continental air over North America. Due to the reduced passage time over the Atlantic relative to slower NW flows, NW_{fast} flows are less moistened. This cluster is identified only for trajectories arriving at 3000 and 1500 m. It is the smallest cluster in number of trajectories at 3000 m (6.7%) and 1500 m (8.1%). Almost non-existent in summertime, most of these flows occur from October to April at both 1500 and 3000 m.

NW_{mod}: Advection of polar maritime air masses with relatively lower speeds than NW_{fast}. It is less frequent in summertime and accounts for 16.5% of the trajectories arriving at 3000 m, 11.2% for 1500 m and 6.6% at 500 m.

NW_{slow}: Composed by slow NW advections and relatively slow flows passing over the British Isles and Western Europe, including polar and arctic air masses. It is distributed throughout the year with a maximum in April. 15.0% of the backtrajectories arriving at 3000 m, 12.4% at 1500 m and 12.4% at 500 m fall into this cluster.

W: Advection of Atlantic subtropical/tropical maritime air masses. It accounts for 19.1% of the trajectories arriving at 3000 m, 18.2% at 1500 and 15.0% at 500 m. It is distributed throughout the year.

SW: Air masses composed by moderately short W–SW–S trajectories associated to tropical (maritime and continental) air masses, many of them passing over the western coast of Morocco and the Straits of Gibraltar. This cluster is identified only for trajectories arriving at 3000 m, being the major cluster in a number of trajectories (32.3%). Their frequency is highest from March to September, although in winter SW flows account for up to 20% of the days.

Med: Composed by trajectories recirculating over the western Mediterranean, including slow advections from northern Africa and the southern part of western and central Europe (500 and 1500 m). It is the major cluster at

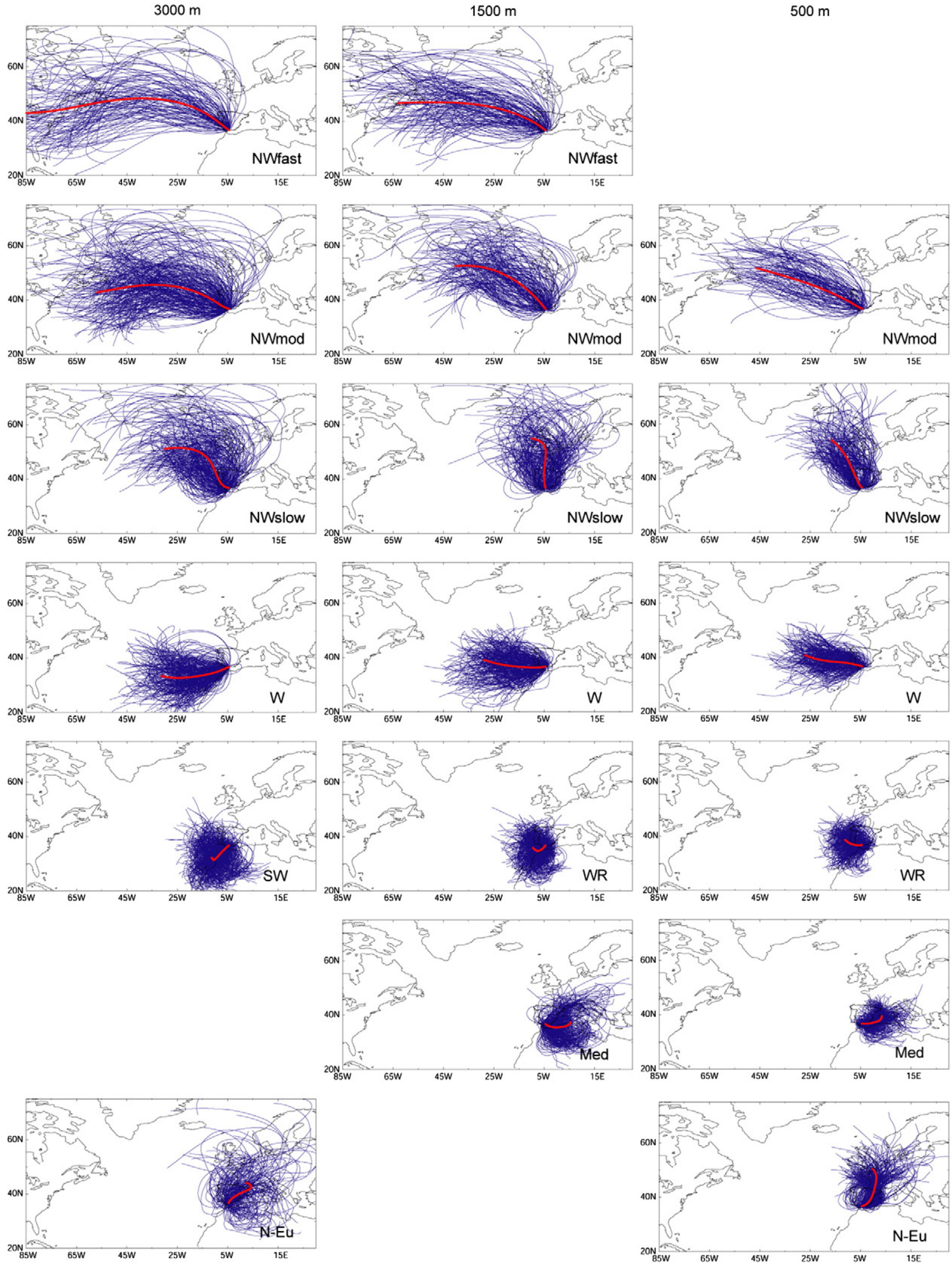

Fig. 1. Trajectory clusters identified for trajectories arriving at 3000 (left), 1500 (center) and 500 m (right) for the 7-year study period. A subset of the trajectories, those arriving at 12 UTC, is drawn (blue). The representative centroid of each cluster appears as a (red) thick line.

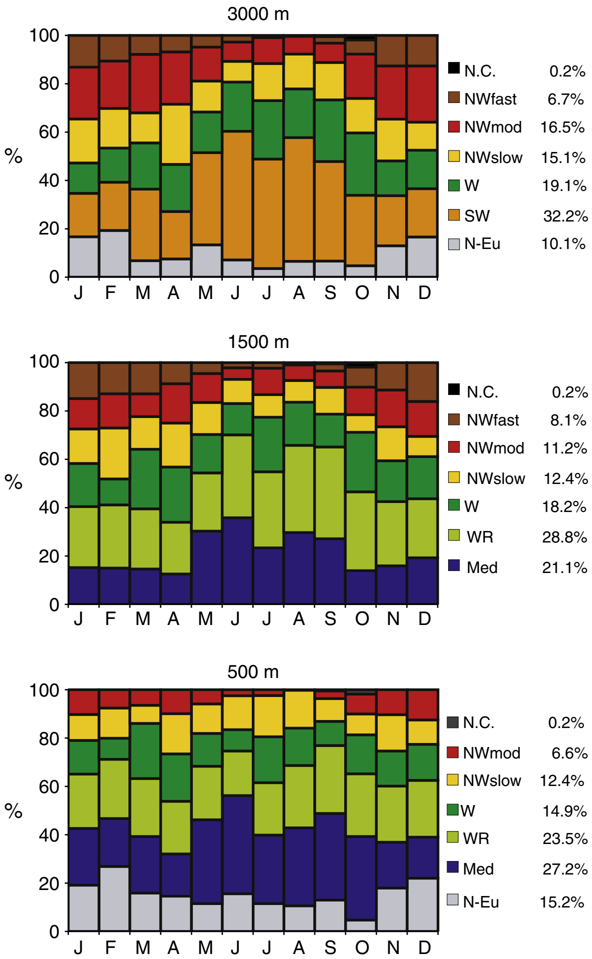

Fig. 2. Monthly variation of the frequency of the identified air mass types.

500 m (27.3%), and 21.1% of the trajectories arriving at 1500 m belong to this cluster. Med trajectories are most frequent from late spring to early autumn.

N–Eu: Composed by continental slow NE/N flows from Western Europe and easterly advections. This cluster is identified only for trajectories arriving at 3000 m (10.1%) and 500 m (15.2%), and it is less frequent in summertime.

WR: Similar to SW flows but lower in speed. Western slow flows and recirculations composed by aged air masses coming from the western Iberian Peninsula or from northern Morocco, associated to tropical maritime and continental air masses. This cluster is not identified for trajectories arriving at 3000 m. Its frequency is 23.5% at 500 m and it is the major cluster (28.8%) at 1500 m, common throughout the year.

3.2. Transport scenarios associated to African dust intrusions and precipitation

African dust is transported to the western Mediterranean basin mainly in layers at altitudes between 1500 and 4000 m (Rodríguez et al., 2001; Pérez et al., 2004). Dust outbreaks over the study area were registered on 23% of the days, according to the reports produced by the collaboration of the Spanish Environmental Ministry, the National Research Council (CSIC) and the Meteorology National Agency (AEMET). Trajectories arriving at Málaga at 3000 m were classified as SW on 61% of the days with African dust intrusion and 21.5% as W. The most frequent combinations under these events are SW_{3000m}–Med_{1500m}–Med_{500m} and SW_{3000m}–WR_{1500m}–Med_{500m}.

The average synoptic pattern corresponding to the arrival of SW flows at 3000 m explains such direction decoupling in the flows between lower and upper levels. An upstream deep trough to the west of the Straits of Gibraltar at mid and upper levels, between the North African and North Atlantic highs, induces SW flows over the study region. At the surface, the Azores high extends to central Europe although southern Iberia is under the influence of relative low-pressures due to a low located to the southwest of the IP and the North African thermal low that predominantly induces easterly flows at the surface level.

The arrival of W flows at 3000 m is associated with the following average synoptic features. The Azores anticyclone extends a ridge to central Europe and the Mediterranean Sea with a very low pressure gradient at surface level in all the western Mediterranean that leads either to WR or Med slow surface flows, while at upper levels an upstream trough is found southwest of the IP, between the North African and North Atlantic highs, that brings W flows over the area.

Precipitation in Málaga is highest from November to January while almost non-existent in July and August (from climatological averages for the 30-year period of 1971–2000). The amount of precipitation and the number of rainy days are predominantly associated with SW, W and NWs advections, in this order, arriving at 3000 m. The fraction of rainy days in these air masses is 15, 22 and 13%, respectively, ascertained from daily data for the period 2005 and 2006. Low-level easterly winds and SW/W flows aloft, frequent patterns previously described, favor higher amounts of precipitation.

Intensive Atlantic advection episodes (NWfast and NWmod flows) result in the renovation of air masses that decreases aerosol loading at the study area. In addition, the arrival of NWs flows at all levels, which is linked to the passage of cold fronts, contributes to precipitation mostly in the northwestern flank of the IP and therefore these air masses are clean when arriving to southeastern Spain.

Slow surface motions are in general associated to an increase in aerosol concentration levels due to the accumulation of aerosols within the area for several days.

In summary, North Atlantic maritime air masses (NWs and W) would contribute to marine aerosols while SW and N–Eu, as well as a fraction of the identified W trajectories, would contribute to continental aerosol loading. The W cluster would contribute to both types of aerosols. Precipitation is mainly associated with westerly circulation at mid and upper levels, common to tropical and polar maritime air masses (SW, W and NWs).

3.3. Frequency distributions and temporal variations of ^{7}Be and ^{210}Pb specific activities and aerosol mass concentration

The individual measurements of ^{7}Be and ^{210}Pb specific activities and aerosol mass concentration were analyzed to derive the statistical estimates characterizing the distributions. Summary statistics of the measurements including arithmetic mean (AM), geometric mean (GM), standard deviation (SD), dispersion factor of geometric mean (DF), range of values and coefficient of variation (CV) are shown in Table 1. These values are given in mBq m^{-3} for ^{7}Be and ^{210}Pb, and in μg m^{-3} for aerosol mass concentration. The monthly specific activities of ^{7}Be, ^{210}Pb and aerosol mass concentrations registered in Málaga are lognormally distributed. Assuming this type of distribution, the GM for ^{7}Be, ^{210}Pb and mass concentration should be used to characterize the average values: 4.6 mBq m^{-3} for the specific activity of ^{7}Be, 0.54 mBq m^{-3} for that of ^{210}Pb and 52.2 μg m^{-3} for the aerosol mass concentration.

The monthly ^{7}Be and ^{210}Pb activity concentrations from January 2000 to December 2006, as well as the aerosol mass concentration, are displayed in Fig. 3. Both ^{7}Be and ^{210}Pb are attached to sub-micron sized aerosol particles. These radionuclides laden aerosols are then scavenged from the atmosphere by dry and wet depositions. Moreover, Fig. 4 shows the box and whiskers diagrams by season of ^{7}Be, ^{210}Pb and aerosol mass concentrations. The activity of ^{7}Be presents seasonal variation with higher values in spring and summer months. The higher ^{7}Be concentrations in spring are normally assigned to the thinning of the tropopause resulting in air exchange between the stratosphere and troposphere during late winter and early spring. The activity of ^{210}Pb shows marked seasonal variations, with higher concentration values in summer months and lower values on other seasons. In addition, summertime in Málaga is a season without rainfall and the rising air temperature produces vertical mixing with the consequent increase of ^{7}Be concentration at ground level. Similarly, high ^{210}Pb concentrations in summer are assigned to prolonged cyclonic conditions with high air temperature and increased ^{222}Rn exhalation.

A regression relating each specific activity to the aerosol mass gives correlation coefficients of 0.53 for ^{7}Be and 0.52 for ^{210}Pb. This suggests that ^{7}Be and ^{210}Pb activities are associated to higher concentration of aerosol particles. Higher aerosol mass concentrations are reached in spring and summer coinciding with higher concentrations of these tracers, ^{7}Be and ^{210}Pb, when precipitation is very scarce, because rainfall is the main process of removal of the aerosols from the atmosphere.

3.4. Relationship of ^{7}Be and ^{210}Pb specific activities and aerosol mass concentrations to long-range transport scenarios

Tables 2–4 show the correlation coefficients between specific activities of ^{7}Be and ^{210}Pb and the monthly frequency of air flows arriving at 3000, 1500 and 500 m, respectively. A positive correlation coefficient indicates that an increase in carrier aerosol concentration is associated with the more frequent arrival of a particular air mass, while a negative correlation value points to a decrease in aerosol concentration with higher arrival frequency of a type of air flow. The probability p of obtaining each observed correlation coefficient by chance is also included in the tables. Correlations with $p > 0.05$ are not considered.

Table 1
Summary statistics of the activity concentrations of ^{7}Be and ^{210}Pb and the aerosol mass concentration.

	AM	GM	SD	DF	Range	CV (%)
^{7}Be	4.8	4.6	1.6	1.4	2.5–14.9	34.2
^{210}Pb	0.58	0.54	0.21	0.62	0.24–1.40	37.40
Aerosol mass	53.6	52.2	12.6	1.26	27.0–98.3	23.6

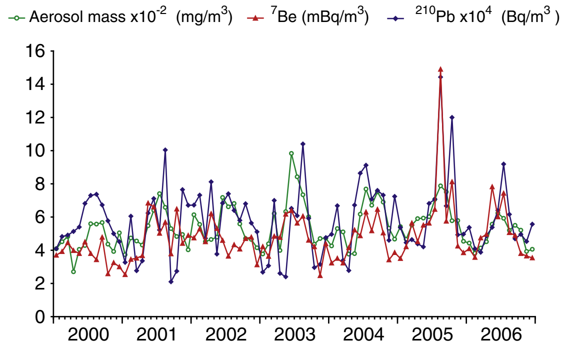

Fig. 3. Monthly-averaged specific activities of ^{7}Be and ^{210}Pb and aerosol mass concentration measured in Málaga during a 7-year period.

It can be appreciated from Tables 2–4 that correlation coefficients are highest for the trajectories arriving at 3000 m, while the lowest values are obtained for those arriving at 500 m. This means that ^{7}Be- and ^{210}Pb-bearing aerosols are predominantly transported by air masses arriving at Málaga at 3000 and 1500 m. In addition, the correlation coefficients between activities and mass concentration with the monthly frequency of the air flows increase and decrease simultaneously, showing the effect of the aerosol concentration on the specific activities of both radionuclides. Regardless of the different origin of ^{7}Be and ^{210}Pb activities, their corresponding correlation coefficients exhibit a similar behavior.

Carrier aerosol concentration increases when air masses pass over northern Africa, as inferred from the high and positive correlations found for SW flows arriving at 3000 m (SW_{3000m}), see Fig. 5 and Table 2, and their concurrent flows at lower altitudes Med_{1500m}, WR_{1500m}, and Med_{500m} (Tables 2–4). However, other continental air masses like those arriving from Europe (N–Eu) show negative correlations possibly due to the prevalence of NWs in the same time period. That reduces the correlation values found when looking at the whole set of flows passing over continental areas.

Conversely, low aerosol concentration is registered when Atlantic air masses (NWs and NWs + W flows) arrive at 3000 and 1500 m (Fig. 5, Tables 2 and 3), and also when NWmod arrive at 500 m (basically clean air masses with marine aerosols).

Continental regions are stronger sources of ^{210}Pb because the primary source of ^{210}Pb in the atmosphere is ^{222}Rn emanation from continental soils with global fluxes into the atmosphere estimated to be about (8–40) 10^{-3} Bq m^{-2} s^{-1} but merely 2 10^{-4} Bq m^{-2} s^{-1} from oceanic areas (Nazaro, 1992).

The highest and positive correlation coefficients 0.88 and 0.78 for specific activities of ^{7}Be and ^{210}Pb, respectively, and 0.93 for aerosol mass concentration, are obtained for trajectories at 3000 m and SW air flows (when air masses

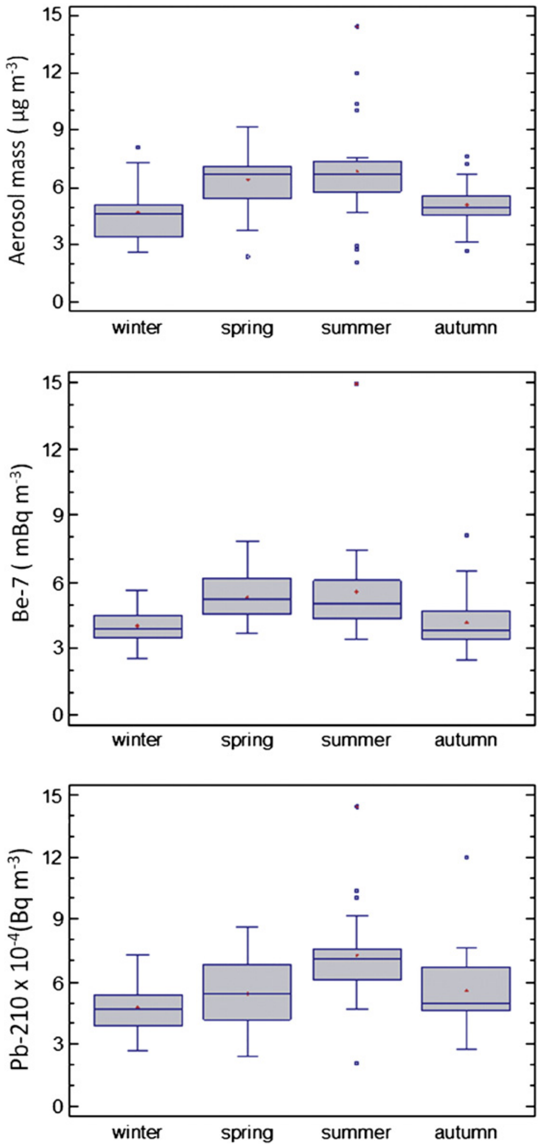

Fig. 4. Seasonal distribution of the aerosol mass concentration and ^{7}Be and ^{210}Pb activities.

pass over northern Africa), Fig. 5. African dust is transported to the western Mediterranean basin in layers mainly above the boundary layer at altitudes between 1500 and 4000 m

Table 2
Correlation coefficients and significance (in parentheses) of the monthly ^{7}Be and ^{210}Pb activities and the aerosol mass concentration to the frequency of air flows arriving at 3000 m.

	^{7}Be	^{210}Pb	Aerosol mass
SW	0.88 (<0.001)	0.78 (0.003)	0.93 (<0.001)
SW + N–Eu	0.78 (0.003)	0.73 (0.007)	0.84 (<0.001)
N–Eu	−0.63 (0.027)	−0.47 (0.119)	−0.64 (0.024)
$NW_{fast}+NW_{mod}+NW_{slow}$	−0.86 (<0.001)	−0.77 (0.003)	−0.86 (<0.001)
$NW_{fast}+NW_{mod}+NW_{slow}+W$	−0.79 (0.002)	−0.74 (0.006)	−0.84 (<0.001)

Table 3
Correlation coefficients and significance (in parentheses) of the monthly ^{7}Be and ^{210}Pb activities and the aerosol mass concentration to the frequency of air flows arriving at 1500 m.

	^{7}Be	^{210}Pb	Aerosol mass
Med	0.74 (0.006)	0.61 (0.036)	0.75 (0.005)
WR	0.56 (0.058)	0.75 (0.005)	0.83 (<0.001)
Med + WR	0.74 (0.005)	0.74 (0.06)	0.87 (<0.001)
NW_{slow}	−0.45 (0.143)	−0.60 (0.039)	−0.55 (0.066)
$NW_{fast}+NW_{mod}+NW_{slow}$	−0.84 (0.001)	−0.78 (0.003)	−0.90 (<0.001)
$NW_{fast}+NW_{mod}+NW_{slow}+W$	−0.75 (0.005)	−0.74 (0.006)	−0.87 (<0.001)

(Pérez et al., 2004). Trajectories arriving at Málaga at 3000 m were classified as SW on 61% of the days with African dust intrusion. Therefore, this good correlation may be related to the arrival of re-suspended aerosols from the African continent.

It should be noted that computed backtrajectories have limitations to represent subsynoptic scale flows. However, meso-scale processes are key to understand the downward transport of aerosols from the free troposphere into the atmospheric boundary layer (ABL), and subsequent ground-level concentrations. Colette et al. (2008) have demonstrated the injection of an African dust layer into the ABL in the Paris area by means of lidar measurements. Other authors point to subsidence and orographic enhancement (central Spain; Sánchez et al., 2007) or to the development of the ABL (southern Italy; Boselli et al., 2008). In the study area, the complex coastal terrain may be an additional relevant factor. A frequent summer situation is well documented for all the western Mediterranean (Millán et al., 1997; Pérez et al., 2004): besides the generalized subsidence due to the Azores anticyclone, there exists a well developed sea-breeze regime in all the region, reinforced in the coastal ranges by upslope winds that, in combination with the formation of the Iberian thermal low, leads to recirculation cells up to 3–5 km in height with their return flow aloft and large-scale compensatory subsidence over the surrounding coastal areas. These coupled meso-scale processes are responsible for the injection of pollutants from the ground, their recirculation and aging. Rodríguez et al. (2001) indicate that this strong convective activity leads to the abatement of the African air masses over the Iberian Peninsula.

The results suggest the existence of a weather pattern leading to a combination of African dust uplifting and downward movement from the upper troposphere. However a monthly analysis does not allow to properly identify single events of such combined situation. It should be noted that

Table 4
Correlation coefficients and significance (in parentheses) of the monthly ^{7}Be and ^{210}Pb activities and the aerosol mass concentration to the frequency of air flows arriving at 500 m.

	^{7}Be	^{210}Pb	Aerosol mass
Med	0.76 (0.004)	0.52 (0.087)	0.73 (0.007)
NW_{mod}	−0.84 (0.001)	−0.74 (0.006)	−0.92 (<0.001)
NW_{slow}	0.33 (0.287)	0.42 (0.172)	0.32 (0.306)
N–Eu	−0.70 (0.012)	−0.39 (0.209)	−0.46 (0.128)

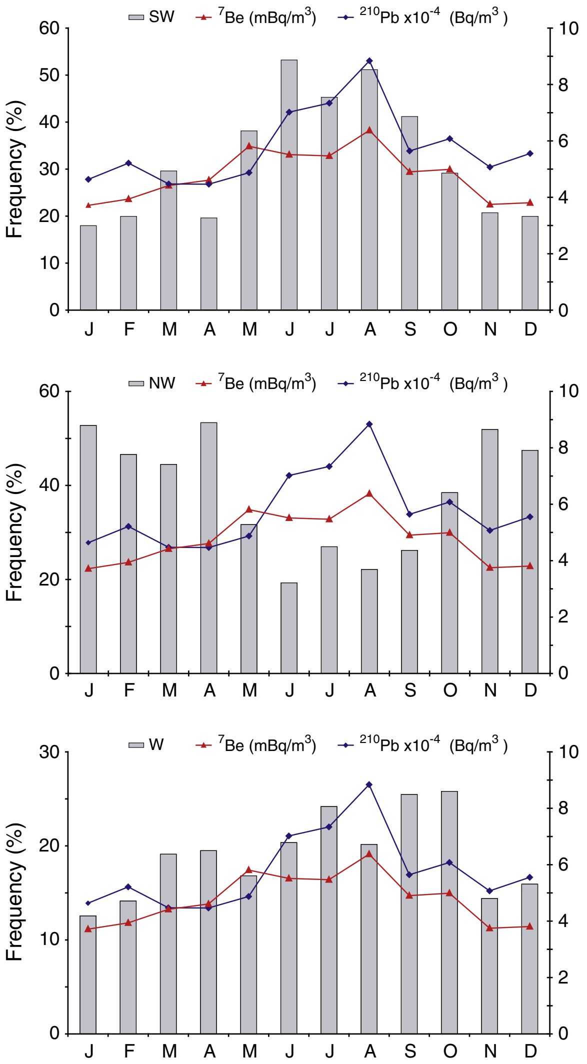

Fig. 5. Monthly specific activities for ^{7}Be and ^{210}Pb and their relationship to the monthly frequency of southwesterlies (top), northwesterlies (center) and westerly flows (bottom).

such situation is more common in summer than in spring at the study area, according to the higher ^{7}Be/^{210}Pb activities ratio found in spring (Fig. 4), and given that African dust outbreaks are more frequent in summer.

In turn, the highest and negative correlations, 0.84 and 0.78 for ^{7}Be and ^{210}Pb specific activities, respectively, and 0.97 for aerosol mass concentration, indicate that the polar maritime air masses (NW) are clean air flows with low aerosol levels (see also Cabello et al., 2008b). Such intense Atlantic advection episodes result in the renovation of air masses with the subsequent decrease in aerosol loading. Rain events are frequently associated with these Atlantic advections and may cause an additional decrease in aerosol concentration.

The mixed character of the W flows cluster, composed by purely zonal flows and trajectories that follow the northwestern coastline of Africa, explains in part that W flows are not associated with low aerosol concentration (Fig. 5) as polar maritime air masses (NWs) are. The prevalence of SW flows in summertime may also be relevant.

3.5. The influence of precipitation

There is a significant decrease in the concentration of both ^{7}Be and ^{210}Pb in air with increasing rainfall at the study site (Fig. 6). This is due to the cleaning of the ambient air leading to lower concentrations of both species during rainy periods, compared to periods of no (or very little) rainfall. Similar observations have been reported by a number of authors, Hotzl and Winkler (1987), Monaghan (1989), and Likuku (2006). Removal of both ^{210}Pb- and ^{7}Be-bearing aerosols by rainwater is supported by the results obtained by numerous workers who reported high correlations of ^{210}Pb and ^{7}Be depositional fluxes in rainwater with the amounts of precipitation: in Texas and Maryland, USA (Baskaran et al., 1993; Kim et al., 2000); in Geneva, Switzerland (Caillet et al., 2001); in northern Taiwan (Su et al., 2003); and in Málaga, Spain (Dueñas et al., 2002; 2003). Model studies by Koch et al. (1996) indicated that removal of ^{210}Pb and ^{7}Be from the troposphere is dominated by rainfall scavenging processes: approximately 88% of ^{210}Pb is removed by rainfall and 12% by dry deposition; whereas 68% of ^{7}Be is removed by precipitation and 3% by dry deposition.

The correlation coefficients between the monthly values of rainfall and the activities of both radionuclides, −0.74 and −0.54 for ^{7}Be and ^{210}Pb, respectively (see Fig. 6), suggest that rainfall at the study area may not be the only variable controlling the concentration of these isotopes in air. As shown in the previous subsection, correlations between the concentration of both nuclides and the occurrence of the different air masses are larger. The origin and the history of the air masses determine to a great deal both the concentrations of ^{210}Pb- and ^{7}Be-bearing aerosols, and the precipitation at the study area and along the route to the study area. In fact, only ^{7}Be- and ^{210}Pb-laden air masses arriving at the study area may present a clear anticorrelated behavior with rainfall: concentrations would be reduced if there is precipitation while they would remain high with no rainfall (e.g. single events of SWs). In turn, air masses coming from weak source areas of ^{7}Be and ^{210}Pb can hardly present such anticorrelation. Polar maritime air masses (NWs) present the largest negative correlation with both radionuclides, while only a low-to-moderate fraction of the trajectories corresponds to precipitation in the study area.

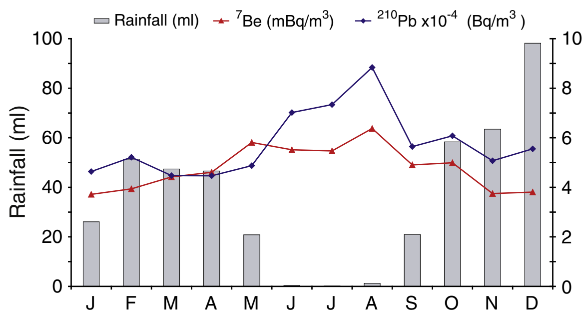

Fig. 6. Monthly rainfall and specific activities of Be-7 and Pb-210.

On a monthly basis, however, different situations are averaged. In the case of SW and W air flows, the transport of African dust and precipitation scavenging are competing processes that result in a concentration increase of both nuclides when such flows are more frequent.

It can be seen in Fig. 6 that monthly rainfall is much lower in January than in December, while the activity of both radionuclides is quite similar. The frequency of the different arriving air masses is also similar, thus activities in December should show a marked decrease. As depicted in Fig. 7, there is a large difference in the fraction of days with precipitation between December (40.0%) and January (29.4%) for the W flows. The number of African dust outbreaks in these months when W flows arrive is small, but they are more frequent in December than in January, which compensate the effect of precipitation scavenging during the last month of the year.

4. Conclusions

1. Six air flow patterns arriving at Málaga are identified by backtrajectory cluster analysis at each arrival height of 3000, 1500 and 500 m. A strong decoupling at the different altitudes is found to be frequent, with easterlies at low levels and westerlies at high levels as the most dominant situations. The association between the monthly frequency of each flow type and the surface level air concentrations of ^{7}Be and ^{210}Pb and aerosol mass concentration is highest for the trajectories arriving at 3000 m and lowest for those arriving at 500 m.
2. Surface level air specific activities of ^{7}Be and ^{210}Pb and aerosol mass concentration were measured in Málaga. Seven years of radionuclide data show that polar maritime air masses (NWs) are strongly associated to low activities of both species, while continental flows from northern Africa are strongly correlated with high specific concentrations. Such transport patterns are related, respectively, to low continental aerosol load and precipitation scavenging along the trajectory pathway before reaching the study area, and to the transport scenario leading to African dust intrusions. The latter scenario comprises SW flows that arrive from northern Africa at mid and upper levels whereas slow easterlies arrive from the Mediterranean Sea at lower levels, and is common to the western Mediterranean basin. Meso-scale coastal processes explain the abatement of these air masses and their impact at ground-level, but are not well represented by the computed backtrajectories.
3. The association between monthly activity concentrations of both radionuclides and the whole set of continental trajectory pathways is weaker than the one related to the advection of African dust to southwestern Europe. The statistical analysis relating radionuclide activity concentrations and air flow types (going beyond the difference between oceanic and continental trajectories) is scarce in the literature (Arimoto et al., 1999); the present work shows the interest to further investigate the relationship between the occurrence of African dust outbreaks and radionuclide activity concentrations in the European continent. In this sense, Menut et al. (2009) have recently reported an increase in ^{7}Be, ^{210}Pb and ^{137}Cs associated to one intense African dust event in southern France.
4. Higher aerosol mass concentrations are reached in spring/summer seasons coinciding with higher specific activity of the tracers, ^{7}Be and ^{210}Pb. There is a good correlation between specific activities of both radionuclides and aerosol mass concentration.
5. The association between rainfall at the study area and low ^{7}Be and ^{210}Pb specific activities is moderate indicating that the effect of washout by rainfall partly controls the surface level air concentration of the two isotopes to a similar degree. The arrival of SW and W flows at high levels contributes as a whole to both rainfall and the entrance of African dust. A daily analysis of such events over the study area allows further understanding of their opposite effects.

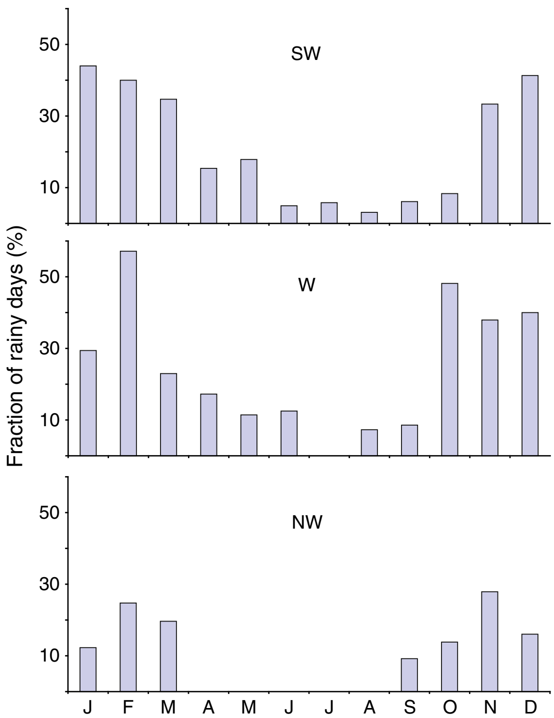

Fig. 7. Proportion of rainy days for the three precipitation-related air masses with respect to the number of air masses of each type.

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