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Advection pathways at the Mt. Cimone WMO-GAW station: Seasonality, trends, and influence on atmospheric composition

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1 **Advection pathways at the Mt. Cimone WMO-GAW station: seasonality,**
2 **trends, and influence on atmospheric composition.**

3
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14
15 **Keywords:** back-trajectories; teleconnection; trends; aerosol; atmospheric radiotracers.

16 **Highlights:**

- 17 • Characterisation of primary advection pathways to the Mt. Cimone baseline station
18 • Characterisation of atmospheric constituents by advection pathway to Mt. Cimone
19 • Trend analysis of atmospheric pathways and atmospheric composition at Mt. Cimone
20 • Associations between pathways, atmospheric composition and teleconnection indices
21

22 Abstract

23 Relationships are analysed between advection pathways and atmospheric composition at the
24 high-mountain station of Mt. Cimone (Italy), between 1999 and 2006. Back-trajectory cluster
25 analysis identifies eight main advection pathways. A connection is demonstrated between the
26 seasonality of air mass transport and atmospheric composition. Temporal trends and correlation
27 of variables, flow types and teleconnection indices show, among other, decreasing trends of ^{210}Pb
28 (a radionuclide of crustal origin; $-0.008 \text{ mBq m}^{-3} \text{ year}^{-1}$) as well as PM_{10} ($-0.15 \mu\text{g m}^{-3} \text{ year}^{-1}$),
29 indicating that previously observed downward PM_{10} trends in Europe may actually be
30 attributable to a combination of meteorological factors and decreasing anthropogenic emissions.
31 The detection of a positive (negative) correlation of these tracers with Western (Arctic) air
32 masses, showing significant downward (upward) trends at the study site, further confirms our
33 findings. Lastly, relationships between teleconnection indices and atmospheric transport
34 types/atmospheric variables are further analysed, focusing on large-scale atmospheric circulation
35 indices and regional low-frequency atmospheric circulation pathways, the Mediterranean
36 Oscillation and the Western Mediterranean Oscillation. The analysis reveals the important
37 influence of such regional indices on the advection pathways.

38 1 Introduction

39 There is a pressing need to improve understanding of processes contributing the seasonal
40 variability of background/baseline (i.e. well-mixed tropospheric) atmospheric composition in the
41 central north Mediterranean region, a hotspot of air pollution and climate change. In fact, due to
42 the sunny, hot and dry weather typical of this region especially during summer, together with the
43 convergence of long-range transport over the basin, air pollution in the form of reactive
44 compounds is often higher than in most European inland regions (Dulac et al., 2016). In addition,
45 climate change will significantly impact air quality with numerous two-way interactions not
46 always well understood.

47 Air pollution in the Mediterranean basin is primarily in the form of particulate matter and ozone
48 and nitrogen deposition (Ochoa-Hueso et al., 2017).

49 In this framework, clustering of backward trajectories has been used to study the influence of the
50 origin and pathway of air masses on composition change (for a review see Fleming et al., 2012).
51 The investigation of vertical motions in the atmosphere may take advantage of using ^7Be and
52 ^{210}Pb radiotracers, because of their naturally contrasting origin: in fact, ^7Be (half-life 53.3 days)
53 is produced by cosmic ray spallation reactions with nitrogen and oxygen in the stratosphere
54 (about 75%) and in the upper troposphere (Usoskin and Kovaltsov, 2008), while ^{210}Pb (half-life
55 22 years) is a tracer of continental air masses (Balkanski et al., 1993), being emitted as decay
56 product of ^{222}Rn (half-life 3.8 days) deriving from crustal rocks and soils (Turekian et al., 1977).
57 Once produced, both radionuclides attach to submicron-sized aerosol particles peaking in the
58 accumulation mode (e.g., Gaffney et al., 2004). Thereafter, the main removal mechanisms of ^7Be
59 and ^{210}Pb from the atmosphere are wet and dry scavenging of the carrier aerosol (Feely et al.,
60 1989; Kulan et al., 2006). For this reason, simultaneous measurements of ^7Be and ^{210}Pb , and
61 analysis of their ratio, can provide useful information about the vertical motion of air masses as
62 well as on convective activity in the troposphere (e.g., Koch et al., 1996; Lee et al., 2007).

63 The use of air mass classification together with atmospheric radiotracers is not common, but
64 has been the subject of some studies (e.g. Arimoto et al., 1999; Hernández et al., 2008; Dueñas
65 et al., 2011; Lozano et al., 2012; Chambers et al., 2013, 2014, 2016 a,b; Grossi et al., 2016;
66 Hernández-Ceballos et al., 2016). However, most of the previous studies of this kind in the

67 Mediterranean region focused on relatively short time series, and focused on understanding the
68 variability of atmospheric radiotracers without a clear connection to other atmospheric
69 compounds. Moreover, while the relation between natural radionuclides and teleconnection
70 indices has been the subject of recent studies (Grossi et al., 2016; Sarvan et al., 2017), the
71 variability in the occurrence of each trajectory group and the assessment of trends in association
72 with large-scale atmospheric circulation indices, such as the North Atlantic Oscillation index
73 (NAOi), is less common (Orza et al., 2013). Even less studied is the association in the
74 occurrence of advection pathways with the remaining modes of atmospheric circulation over
75 Europe, such as the Eastern Atlantic (EA) pattern, Eastern Atlantic/Western Russia (EA/WR),
76 and the Scandinavian (SCA) pattern. Together with NAO, these indices represent the most
77 important mid-latitude modes for the Mediterranean climate at the monthly time scale (Trigo et
78 al., 2006).

79 In this context, long-term measurements at the high-elevation WMO-GAW baseline station
80 of Mt. Cimone (Italy; 44°11' N, 10°42' E, 2165 m asl) are of paramount importance. In
81 particular, they are useful for the identification of dominant advection pathways, assessing
82 associations between pathways and atmospheric composition, and investigating links between
83 flow pathways and circulation modes in the Mediterranean region on seasonal and interannual
84 time scales. In addition, the occurrence of trends and the relationships of trends in atmospheric
85 composition with those in advection pathways and teleconnection indices in the monthly time
86 series are also explored, focusing not only on large-scale indices but considering two additional
87 regional low-frequency atmospheric circulation pathways, namely the Mediterranean Oscillation
88 (MO) and the Western Mediterranean Oscillation (WeMO). It should be emphasized here that,
89 historically, the investigation of atmospheric circulation model influences (both large-scale and
90 regional) typically focused on precipitation and temperature pathways. To date, there has been
91 limited exploration of the relationship between these modes and advection pathways/atmospheric
92 composition.

93 This work is organized as follows. We first describe the measurement techniques and the
94 statistical methods used. We then present and discuss our results on: 1) the description of the
95 main advection pathways found by the cluster analysis of back trajectories; 2) the analysis of the
96 relationships between advection pathways and meteorological parameters/other atmospheric
97 components; 3) the temporal analysis of the monthly time series, including trends; 4) the
98 associations of air flow types with teleconnection indices and meteorological/atmospheric
99 variables. We finally summarize our main conclusions.

100 **2 Materials and Methods**

101 **2.1 Sampling site**

102 Mt. Cimone, the highest peak of the Italian northern Apennines, hosts a global station of the
103 Global Atmosphere Watch (GAW) programme of the World Meteorological Organization
104 (WMO) constituted by a meteorological observatory by the Italian Air Force (active since 1941)
105 and a research facility managed by the Institute of Atmospheric Sciences and Climate (ISAC) of
106 the National Research Council of Italy (CNR), active since 1996. The site is located far away
107 from large industrialized and urban areas, has a 360° free horizon experiencing both regional and
108 long-range transport of air masses (Bonasoni et al., 1999, 2000b; Cristofanelli et al., 2006,
109 2009a, b, 2013; Cristofanelli and Bonasoni, 2009; Tositti et al., 2013). The elevation of the site
110 (2165 m asl) is such that the station lies above the planetary boundary layer (PBL) during most
111 of the year, even if an influence of the innermost layer is evident during warm months due to the

112 increased mixing height and the influence of the mountain/valley breeze regimes (Fischer et al.,
113 2003; Cristofanelli et al., 2007; Griffiths et al., 2014). For these reasons, under specific conditions
114 (i.e. usually during cold months and during stable summer nights when regional anti-cyclonic
115 conditions dominate), the measurements of atmospheric compounds and meteorological
116 parameters at this site can be considered representative of the well-mixed southern European-
117 Mediterranean basin free troposphere (Bonasoni et al., 2000a; Fischer et al., 2003; Cristofanelli et
118 al., 2007, 2018), a region which is recognized as a hot-spot both in terms of climate change and
119 air quality.

120 121 **2.2 Measurements**

122 As a WMO-GAW station, several atmospheric compounds have been measured at Mt.
123 Cimone for many years (Cristofanelli et al., 2018): CO₂ (since 1979) (Ciattaglia, 1983, 1986;
124 Colombo et al., 2000), tropospheric O₃ (since 1991) (Cristofanelli et al., 2015, 2018),
125 concentration and size distribution of particles with optical diameter between 0.30 and 20 µm
126 (since 2000) (Marinoni et al., 2008), black carbon (July 2005) (Marinoni et al., 2008), and CO
127 (since 2007) (Cristofanelli et al., 2009b).

128 ⁷Be, ²¹⁰Pb and aerosol mass loading in the form of PM₁₀ (airborne particulate matter with a
129 mean aerodynamic diameter less than 10 µm) were measured regularly in the period 1998-2011
130 with a Thermo-Environmental PM₁₀ high-volume sampler (average flow rate of 1.13 m³ min⁻¹ at
131 standard temperature and pressure conditions) (Lee et al., 2007; Tositti et al., 2012, 2013, 2014).

132 After retrieval, the observations of the various atmospheric parameters previously mentioned,
133 as well as of meteorological parameters such as temperature, pressure, relative humidity, and
134 wind speed, were averaged to the same time resolution of PM₁₀ and atmospheric radionuclides
135 for statistical homogenization of data. In fact, as the PM₁₀ filters at the station are manually
136 changed, sampling time is not uniform. Anyway as most of the samples were collected over 48
137 hours (sampling approximately 3250 m³ of air), in order to safely apply statistical techniques,
138 data have been firstly homogenized by selecting only those samples which collected a volume
139 between 2700 and 3700 m³.

140 We also included tropopause heights in the analysis, calculated from the radiosoundings in
141 San Pietro Capofiume (44°39'N, 11°37'E, 10m a.s.l.), a regional meteorological station located
142 in the Po Valley to the North-East of Mt. Cimone, available since 1987 from the University of
143 Wyoming website (<http://weather.uwyo.edu/upperair/sounding.html>).

144 145 **2.3 Teleconnection indices**

146 As reported in the Introduction, here we investigate the connection of atmospheric composition
147 and advection pathways with teleconnection indices, considering both large-scale and regional
148 scale teleconnections, i.e. NAO, EA, EA/WR, SCA, MO, and WeMO. Table 1 presents an
149 overview of the teleconnections investigated in this paper.

150 The NAO, a redistribution of atmospheric mass between the Arctic and the subtropical
151 Atlantic (Hurrell, 1995), has been identified as the dominant mode of variability of the surface
152 atmospheric circulation across the Atlantic (Barnston and Livezey, 1997). The NAO is
153 determined by the position and strength of the Icelandic low and the Bermuda-Azores High.
154 Oscillations between high and low NAO phases modulate the westerly jet stream and cause large
155 changes in the heat and moisture transport between the Atlantic and the neighbouring continents
156 (e.g., Hurrell, 1995, 1996), affecting the intensity and number of storms (Hurrell et al., 2003).
157 These changes influence air pollutant transport and dispersion, impacting, for instance, the

158 transport of Saharan dust into the Mediterranean and Atlantic in winter (Moulin et al., 1997), the
159 export and import pathways of pollution to the Mediterranean basin (Hurrell, 1995), influencing
160 local-to-regional scale pollutant concentrations (e.g., Cuevas et al., 2013; Cristofanelli et al.,
161 2015), and modifying the transport of pollutants from North America to Europe (Li et al., 2002).
162 A relation between the NAO phase and Stratosphere-to-Troposphere Transport (STT) variability
163 has also been pointed out (James et al., 2003; Cristofanelli et al., 2006, 2015). A common
164 measure of the NAO phase is the so-called NAO index (NAOi) which is commonly defined as
165 the difference in normalized sea level pressure (SLP) anomalies between either Lisbon (Portugal)
166 or Ponte Delgada (Azores), and Stykkisholmur/Reykjavik (Iceland) (Hurrell, 1995). Alternative
167 definitions of NAOi have been introduced, including one based on the empirical orthogonal
168 function (EOF) analysis of the SLP field. The NAOi in this case is identified as the leading
169 eigenvector (the first Principal Component, PC1) computed from the time variation of the SLP
170 field (e.g., Hurrell et al., 2003). The advantage of using EOF analysis of the SLP field is that the
171 PC1 provides a more accurate representation of the NAO pattern considering the shifting of the
172 NAO centres of action throughout the year (Pausata et al., 2012). This index appears to be less
173 noisy than the station-based indices. Both monthly indices present a significant correlation
174 coefficient equal to 0.80 and 0.76 over the periods 1998-2011 and 1999-2006 used in this work,
175 respectively. Another alternative NAO index, the CRU station-based NAOi, is calculated as the
176 difference between the normalised SLP over Gibraltar and the normalised SLP over southwest
177 Iceland (Jones et al., 1997). CRU station-based index presents a significant correlation
178 coefficient with the previous ones (equal to 0.79 with Hurrell station-based NAOi, and equal to
179 0.77 with Hurrell principal components-based NAOi).

180 The EA pattern is the second prominent mode of low-frequency variability over the
181 North Atlantic, and was first described by Wallace and Gutzler (1981) as anomalously high 500
182 mb height anomalies over the subtropical North Atlantic and eastern Europe when in positive
183 mode. It consists of a north-south dipole of anomaly centers spanning the North Atlantic from
184 east to west. The positive phase of the EA pattern is associated with above-average surface
185 temperatures in Europe in all months and it has been suggested to play a role in positioning the
186 primary North Atlantic storm track (e.g., Seierstad et al., 2007) and in modulating the location
187 and strength of the NAO dipole (Hurrell and Deser, 2009).

188 The EA/WR pattern (Lim, 2015) affects Eurasia throughout the year and consists of four
189 main anomaly centers. The positive phase is associated with positive height anomalies located
190 over Europe and northern China, and negative height anomalies located over the central North
191 Atlantic and north of the Caspian Sea.

192 The SCA pattern (Bueh and Nakamura, 2007) consists of a primary circulation center
193 over Scandinavia, with weaker centers of opposite sign over western Europe and eastern Russia/
194 western Mongolia. The positive phase of this pattern is associated with positive height
195 anomalies, sometimes reflecting major blocking anticyclones over Scandinavia and western
196 Russia, while the negative phase of the pattern is associated with negative height anomalies in
197 these regions.

198 The MO is a low-frequency variability pattern producing opposing barometric, thermal
199 and pluviometric anomalies between the western and eastern borders of the Mediterranean basin.
200 The MO was originally defined as the difference of standardised geopotential height anomalies
201 at Algiers (Alger) and Cairo (Egypt) (Conte et al., 1989), while similar indices have been defined
202 in terms of the difference of standardised pressure anomalies at Gibraltar (Spain) and Lod
203 (Israel) (Palutikof, 2003) or at Marseilles (France) and Jerusalem (Israel) (Brunetti et al., 2002).

204 It has a significant influence over rainfall in the Mediterranean basin (e.g., Martin-Vide and
205 Lopez-Bustins, 2006; Angulo-Martínez and Beguería, 2012). The WeMO was defined within the
206 synoptic framework of the Western Mediterranean basin and its vicinities (Martin-Vide and
207 Lopez-Bustins, 2006). It is defined as the difference between the standardized surface pressure
208 values in Padua (Italy), and San Fernando (Cadiz, Spain): while the north of Italy is an area with
209 a relatively high barometric variability due to the influence of the central European anticyclone
210 and the Liguria low, the gulf of Cadiz is often influenced by the Azores anticyclone. Similar to
211 MO, WeMO has an important effect on precipitation in the Mediterranean, and especially in the
212 eastern Iberian Peninsula (e.g., Martin-Vide and Lopez-Bustins, 2006; Angulo-Martínez and
213 Beguería, 2012; Izquierdo et al., 2013), where NAO is weakly correlated with precipitation.

214 215 **2.4 Clusters of back trajectories, significant differences and trends**

216 In order to analyse the origin of air masses arriving at the measurement site, 96-hour 3D
217 kinematic back-trajectories starting four times a day (00, 06, 12, 18 UTC) at three heights (1400,
218 2200 and 3000 m asl) were calculated with the HYbrid Single-Particle Lagrangian Integrated
219 Trajectory (HYSPLIT) model version 4.8 (Draxler and Hess, 1997, 1998; Draxler, et al., 2018).
220 A 96-hour time length was considered representative for long-range transport to the receptor site
221 and to better control the uncertainty of back-trajectories. Sensitivity tests with a 6-day time
222 length were also performed; however, their results indicate a reduced number of clusters with
223 lower significant differences in atmospheric parameters and constituents, suggesting that these
224 longer trajectories may lose part of their specific features.

225 The first issue faced in the calculation of the back-trajectories was the choice of the
226 meteorological fields used as input, linked to the strongest source of errors (Stohl et al., 2001)
227 when calculating back-trajectories, and eventually influencing the outcome of the trajectory
228 clustering (Cabello et al., 2008a).

229 As previously observed in Brattich et al. (2017a, b), the coarse resolution of the terrain model
230 included in the meteorological databases is generally not able to adequately resolve the
231 topography of Mt. Cimone. In this work we used the National Center for Environmental
232 Prediction (NCEP)/National Center for Atmospheric Research (NCAR) reanalysis with a 2.5°
233 latitude-longitude resolution, 17 pressure levels from 1000 to 10 hPa, and 6 hourly data, which
234 was the best compromise available at the time we started the back-trajectories calculation. Such a
235 coarse resolution, too large to resolve mesoscale subsynoptic processes, is still acceptable for our
236 study since we are most interested in the large-scale flow pattern more meaningful in a long-term
237 quasi-climatological approach. The vertical motion of the air parcels was calculated from the
238 vertical velocity fields. As a rule and due to the methodology applied to compute the back-
239 trajectories (the computation uses the horizontal gradient of the field, calculated as a “centered
240 difference” with meteorological data on a subgrid that follows the trajectory), it is recommended
241 that trajectories at three heights are calculated simultaneously (see
242 <https://www.arl.noaa.gov/hysplit/hysplit-frequently-asked-questions-faqs/faq-hg23/>). In this
243 case, trajectories were calculated at 2200 m asl (just above the monitoring site), at 3000 m asl
244 (800 m higher, at the edge of the free troposphere) and at 1400 m asl (above the terrain’s height
245 for the measurement site in the model, to better consider meteorology below the study site). In
246 the following, results are reported and discussed only for the height corresponding to that of the
247 receptor site. The analysis at 3000 m, useful to compare the study site with the free troposphere,
248 is included in Supporting Information (SI).

249 As for the association between trajectories and samples, each aerosol sample with its
250 physico-chemical properties was associated with a specific advection pattern only if at least 60%
251 of the calculated trajectories ending at the site during the sampling corresponded to that
252 advection pattern. However, samples out of the outlined conditions were also analysed in depth
253 with emphasis on flows characterized by fast and high-frequency variability often associated
254 with singular though relevant trajectories (i.e. cutoff lows or Saharan dust incursions).

255 Clusters of back-trajectories were calculated following a clustering procedure based on
256 the k-means algorithm, with specific features like the use of great-circle distances and
257 determination of the number of clusters from the evaluation of the classification into k clusters
258 (considering a large number of replicates), with k running from 15 to 3, (see for example Cabello
259 et al. (2008b), Dueñas et al. (2011), Perrone et al. (2013), Brattich et al. (2016)). Significant
260 differences in the analysed meteorological and atmospheric parameters according to the
261 identified clusters were analysed using the Kruskal-Wallis test, without any “a priori”
262 assumption of their distribution (Brankov et al., 1998). Whenever significant differences among
263 the groups were found, pairwise Mann-Whitney tests were performed to identify the significantly
264 different pairs. Conservatively, p-values in the latter were compared against adjusted
265 significance levels α using the Dunn-Sidak correction for multiple comparisons $\alpha = 1 -$
266 $(1 - \alpha_t)^{1/n}$, where $n = k(k - 1)/2$ is the number of pair-wise comparisons done between k
267 categories, with overall significance $\alpha_t = 0.05$.

268 Composite synoptic charts of 700, 850 and 1000 hPa geopotential height, computed with
269 data from NCEP/NCAR re-analysis project database (Kalnay et al., 1996), available from the
270 Earth System Research Laboratory, Physical Sciences Division, of the USA National Oceanic
271 and Atmospheric Administration (NOAA) at <http://www.esrl.noaa.gov/psd/> were used to analyse
272 the meteorology of individual situations.

273 The presence of trends in the monthly time series over the study period considered in this
274 work was examined through a number of nonparametric statistical methods, mainly based on the
275 Mann-Kendall (M-K) tau test to assess the significance of monotonic trends and the Theil-Sen
276 (T-S) slope estimate for trend magnitude. In particular, considering that the significance of a
277 trend is affected by the presence of serial correlation, and, conversely, the estimate of the serial
278 correlation is also altered by the presence of a trend, the correlation coefficients at different lags
279 were first estimated by computing the sample autocorrelation function (ACF) for each time
280 series. The results indicated that, in general, the analysed time series present some degree of
281 serial correlation, together with seasonality; as a consequence, two methods of trend analysis
282 have been used with the aim of removing, or reducing, the influence of seasonality and lag-1
283 autocorrelation in the monthly data:

284 (1) The seasonal Kendall test (Hirsch et al., 1982), which applies the M-K trend test
285 separately for each month and then combines the results. (2) The trend-free pre-whitening
286 (TFPW) procedure (Yue et al., 2002) applied to the seasonally adjusted monthly time series, to
287 remove the influence of the month-to-month correlations in the significance of the trends. The
288 TFPW procedure comprises several steps, including the linear detrending of the time series using
289 the T-S slope, the removal of the serial autocorrelation of the residuals and the add-back of the
290 discarded linear trend to the remaining time series, before the M-K test is applied. Seasonal-trend
291 decomposition of the time series was used to obtain the de-seasonalized time series, which were
292 subsequently analysed by the TFPW procedure. The decomposition technique used in this work
293 (STL decomposition hereafter) is based on LOESS (locally weighted low-degree polynomial
294 regression), a nonparametric regression technique recursively applied to the seasonal and trend

295 components (Cleveland et al., 1990). Additionally, the resulting (nonlinear) trend component has
 296 been used for the visual assessment of the long-term behaviour of the time series.

297 The association between the frequency of each advection pattern and observations at the
 298 sampling site, as well as with the NAOi and other large- and regional-scale teleconnections, has
 299 been examined for the de-trended monthly time series and for the seasonal means via least-
 300 square regression analysis with statistical significance evaluated by a two-tailed *t*-test. Since
 301 relationships are not necessarily linear, the nonparametric Kendall rank test has also been used to
 302 identify any statistically significant association without any “a priori” assumption of their form.
 303 Spearman correlation coefficients have been computed for the cases with significant association.

304 **3 Results and discussion**

305 **3.1 Characteristics of the main advection pathways**

306 Figure 1 shows the centroids (representative trajectories) of the 8 clusters obtained at 2200 m asl
 307 and the relative percentage frequency of each flow pattern over the whole 1998-2011 period,
 308 together with the mean height evolution over time and the monthly variation of the frequency of
 309 the air flow pathways reaching the receptor site.

310 Cluster names were chosen based on their region of provenance. Most of the trajectories
 311 correspond to westerly flows; in particular, westerly trajectories are classified into Northern
 312 Atlantic (N Atl), North America (N Am), Atlantic (Atl), Western (W), and North-Western
 313 Europe (NW-Eu) flows, together representing more than 60% of the flows. The remaining
 314 trajectories are classified into Arctic (A), Eastern (E), and Mediterranean-Africa (Me-Af).

315 As from the mean height evolution over time of the representative trajectories reported in
 316 Figure 1, the Arctic and North-American trajectories descend from the most elevated heights
 317 while approaching the site, and eventually rise again and cross over the Alps. North Western-
 318 Europe and Eastern flows do not considerably change their height during their transport, whereas
 319 Western, Atlantic and (more specifically) Mediterranean-Africa trajectories generally reach the
 320 observatory from very low levels.

321 Figure 2 shows the box plots of meteorological parameters, i.e., pressure, temperature,
 322 relative humidity, precipitation, tropopause height, wind speed, and mixing height by advection
 323 pattern. Similar to Figure 2, Figure 3 depicts box plots for the atmospheric species, such as O₃,
 324 CO₂, BC, CO, fine and coarse particles, PM₁₀, ⁷Be, and ²¹⁰Pb, associated with each flow pattern
 325 at 2200 m and at 3000 m asl. Additionally, the analysis extends over nuclidic and mass ratios
 326 such as ⁷Be/²¹⁰Pb, ⁷Be/PM₁₀, ²¹⁰Pb/PM₁₀, used to gain insights into the vertical motions of air
 327 masses as well as on convective activity in the troposphere (e.g., Koch et al., 1996), are also
 328 analysed. The summary statistics together with significant differences of each variable by
 329 advection pattern is reported in the SI. In order to better characterize the flow pathways, both
 330 boxplots and summary statistics refer to the “pure cases”, i.e., the samples attributed to only one
 331 advection pathway (when at least 60% of the trajectories ending at Mt. Cimone during one single
 332 sampling period belong to the same advection pathway).

333 Below we summarize the major characteristics of the identified advection pathways in
 334 terms of seasonal variability, mean height, meteorological variables and atmospheric
 335 composition, as indicated in Figures 2 and 3.

- 336 • A: advection of fast and elevated (mean height of the cluster equal to 3113 m) air masses
 337 originating in the Arctic/polar regions. This trajectory type is more frequent in autumn
 338 and winter. This subsiding air flow is associated with low temperatures, low relative
 339 humidity, low wind speeds, relatively low values of the tropopause height (probably due

340 to the fact that these air masses are not vertically thick compared to other air masses), and
341 moderate mixing heights. Such air masses can be linked to the presence of lows or cut-off
342 lows characterized by low tropopause or tropopause folding and in fact are also
343 associated with rather low pressure systems. Due to their subsiding nature, travelling at
344 high altitudes over remote regions, these air masses are generally moderately clean, i.e.
345 associated with low values of O_3 , black carbon, CO, PM_{10} , and ^{210}Pb . It is also associated
346 with high 7Be and therefore with high $^7Be/^{210}Pb$ and $^7Be/PM_{10}$. This kind of transport is
347 in fact frequently associated with STE, in agreement with previous observations of
348 stratospheric intrusions at Mt Cimone (Bonasoni et al., 1999, 2000a, 2000b; Brattich et
349 al., 2017a). In particular, the high 7Be , $^7Be/^{210}Pb$ and $^7Be/PM_{10}$ can be attributed to the
350 high production rate of 7Be in the stratospheric air at high latitudes (Beer et al., 2012), ,
351 even though the reduced ozone concentration points out a connection with subsidence
352 from the upper troposphere, a region connected with increases in 7Be but not in O_3 .

- 353 • E: advection of relatively slow and low (mean height equal to 2190 m) air masses from
354 East. This flow type is more frequent in April, May and September, and groups the 13%
355 of the trajectories. These air flows are associated with low tropopause height, while
356 pressure, wind speeds, humidity, and mixing height take intermediate values. This flow
357 type brings lower concentrations of PM_{10} than the Western advection, but is associated
358 with higher loadings of fine than coarse particles, in agreement with observations by
359 Tositti et al. (2013). This flow type is also associated with moderately high black carbon
360 and high ^{210}Pb , and to the lowest values of $^7Be/^{210}Pb$ activity ratio; overall this flow can
361 be labelled as “continental polluted”.
- 362 • Me-Af: relatively short and low (mean height equal to 2154 m) Mediterranean and North-
363 African air masses. These trajectories, grouping the 18% of the total, are active all-year
364 round but mainly in spring and autumn. These flows cross over the Mediterranean at low
365 altitude and correspondingly are warm and humid, and are associated with low wind
366 speeds and intermediate mixing height (close to 1400 m, which is below the typical mean
367 height reported for Saharan dust transport, between 1500 and 4000 m asl; see Jorba et al.,
368 2004 and Papayannis et al., 2008). These air masses bring substantial PM_{10} loadings as
369 linked to Saharan Dust transport (in agreement with Duchi et al., 2016), associated with
370 increases in both fine and coarse sized particles. It contributes also to BC and to high
371 ^{210}Pb and 7Be concentrations, and thus to rather low $^7Be/^{210}Pb$, $^7Be/PM_{10}$ and $^{210}Pb/PM_{10}$
372 ratios. Overall, this flow may be labelled as “African convective”, including the often-
373 observed biomass burning tracers. In particular, similar to what Dueñas et al. (2011) and
374 Brattich et al. (2017a) reported, Mediterranean-Africa air masses are linked to high
375 activities of both 7Be and ^{210}Pb , due to the combination of downward transport from the
376 upper troposphere and African dust uplifting.
- 377 • W: advection of relatively slow and low-level (mean height equal to 1915 m) air masses
378 from West, which are active all year round but are more frequent in July, August, and
379 October. This flow pattern groups the 15% of the trajectories. These air masses are
380 associated with high pressures, high temperatures, low relative humidity, high tropopause
381 height, moderate wind speeds, and moderate-to-high mixing heights. This advection
382 pattern carries elevated values of O_3 , PM_{10} , fine and coarse particles; it contributes to a
383 large degree also to black carbon, a tracer of combustion. This is likely related to of the
384 entrainment of aged, polluted air masses into this flow type when crossing coastal areas
385 in the western Mediterranean. It contributes also to high ^{210}Pb and 7Be concentrations

- 386 (low ${}^7\text{Be}/\text{PM}_{10}$, ${}^{210}\text{Pb}/\text{PM}_{10}$ and ${}^7\text{Be}/{}^{210}\text{Pb}$ ratios) suggesting associated convective
387 pathways.
- 388 • Atl: relatively fast and low-level (mean height equal to 1974 m) air masses coming from
389 the Atlantic Ocean. This advection pattern is most common from October to April. It
390 groups only the 8% of the trajectories. These air masses are moderately warm and humid,
391 present low pressure levels, moderate to high wind speeds, and low mixing heights. This
392 advection pattern shows low contributions of O_3 , black carbon, PM_{10} , ${}^7\text{Be}$ and ${}^{210}\text{Pb}$, as a
393 consequence of the renewal of air masses by these strong mid latitude maritime flows and
394 the relatively high wind speeds recorded at the site.
 - 395 • N-Am: polar fast and upper level (mean height of the cluster equal to 2965 m) air masses
396 that originate as continental air over North America. This air mass type is negligible in
397 summer months, mostly occurring from October to April. This advection pattern is the
398 least frequent among those emerged for Mt. Cimone (5% of the trajectories arriving at the
399 receptor site). Similar to the Arctic type, these are cold, dry subsiding flows. They are
400 related to the lowest temperatures (even lower than the Arctic ones), lowest pressure
401 levels, low relative humidity, low tropopause heights and mixing heights, and moderate
402 wind speeds at the study site. The polar-front jet stream is present at upper levels. North
403 American air masses are usually very clean (low in O_3 , black carbon, CO , PM_{10} as well
404 as in both fine and coarse particles), with the lowest mean and median values of these
405 species. The cleanliness of these flows derives from their subsiding nature originating
406 quite high above the North American region, and reaching Mt. Cimone (i.e. southern
407 Europe) at moderately high wind speeds. This results in the replacement of air masses
408 with cleaner, fresh air, as previously observed also at other southern Mediterranean sites
409 in Spain and Italy (e.g., Cabello et al., 2008b; Perrone et al., 2013, 2014). Also the
410 atmospheric radiotracers ${}^7\text{Be}$ and ${}^{210}\text{Pb}$ present low concentrations within this flow type,
411 probably due to the low concentration of suspended fine particles and relatively younger
412 upper level air masses.
 - 413 • N-Atl: relatively fast, but not very high (mean height of the cluster equal to 2562 m) air
414 masses coming from the Northern-Atlantic Ocean. Active throughout the year with
415 highest frequency in July. This group of trajectories comprehends the 14% of the total.
416 These air masses are moderately warm, very humid, and connected to slow wind speeds
417 and high mixing height. This flow pattern shows elevated contributions of O_3 and fine
418 particles, but low values of black carbon, carbon monoxide and ${}^{210}\text{Pb}$, while it
419 contributes moderately to PM_{10} and ${}^7\text{Be}$ (low ${}^7\text{Be}/\text{PM}_{10}$, ${}^{210}\text{Pb}/\text{PM}_{10}$ and ${}^7\text{Be}/{}^{210}\text{Pb}$ ratios),
420 probably due to contribution of aged pollutants from Western Europe where they travel
421 after their residence over the North-Atlantic Ocean, in agreement with Brattich et al.
422 (2016).
 - 423 • NW-Eu: slow and not very high (mean height equal to 2321 m) continental air masses
424 coming from North Western-Europe. This flow pattern, with mean height equal to 2321
425 m, is more frequent in summer months and groups the 19% of the trajectories. These air
426 flows present the lowest wind speeds and high pressures, frequently related to blocking
427 situations in the summertime; they are also related to high temperatures, relative humidity
428 and mixing height. Similar to the Eastern advection, it brings lower concentrations of
429 particulate matter with respect to the Western flow type, but associated with higher
430 loadings of fine particles than coarse ones. These flows contribute moderately, together

431 with Eastern air masses, to black carbon and to high ^{210}Pb (low $^7\text{Be}/\text{PM}_{10}$, $^{210}\text{Pb}/\text{PM}_{10}$ and
432 $^7\text{Be}/^{210}\text{Pb}$ ratios).
433

434 3.2 Atmospheric parameters by advection pattern

435 The large-scale advection pathways found at Mt. Cimone have been described in terms of
436 meteorological variables and atmospheric composition in the previous subsection. Summarizing,
437 North Atlantic and NW Europe advections, both passing over the British Isles and France,
438 present the highest O_3 levels. In turn, Atlantic as well as North America and Arctic air flows are
439 associated with low O_3 values, which points out the influence of precursor levels. CO_2 , a long-
440 lived greenhouse gas, is well-mixed in the free troposphere and not much affected by the
441 boundary layer dynamics, with values homogeneously distributed over all the flow types. While
442 Mediterranean Africa and Western air masses are associated with high number of both fine and
443 coarse particles, as related to the transport of marine and desert particles together with
444 anthropogenic pollution, North Atlantic advections are high only in fine particles related to the
445 transport of polluted particles from anthropogenic origin.

446 Low values of the ^{210}Pb crustal tracer are observed when air masses arrive from the ocean
447 (Atlantic, North Atlantic and Northern America) as expected, while ^{210}Pb maxima are linked to
448 flows with an explicit continental origin such as Mediterranean-Africa, Western, Eastern and
449 North Western-Europe. This behaviour is of course due to ^{210}Pb continental origin, as ^{222}Rn flux
450 from the oceans into the atmosphere is negligible due to its low marine source (low radon
451 emission) (Balkanski et al., 1993; Baskaran, 2011). ^7Be low values are connected to Atlantic and
452 Northern American air masses, while Western flows are related to the highest values, likely
453 connected to Gulf of Genoa and Gulf of Lion cyclogenesis, which have been long recognized as
454 associated with STE (e.g., Stohl et al., 2000; Aebischer and Schär, 1998).

455 Here it is worth noting that due to the coarse resolution of the meteorological field we are
456 using, our methodology is not able to resolve mesoscale and subsynoptic processes. However,
457 such processes may have important effects on the variability of the atmospheric species we are
458 considering. In particular, some of the identified advection pathways (the local and regional
459 transports) can be associated with favourable “stagnation” conditions (mostly during the summer
460 months), such as the increase in height of the regional PBL and/or mountain/valley breeze
461 regimes, as previously investigated by Cristofanelli et al. (2013, 2016).

462 Figures 4 and 5 analyse the connection of the seasonality of advection pathways with that
463 of radiotracers and PM_{10} , respectively. The seasonality of variables was analysed considering
464 monthly medians as the distributions of PM_{10} and of atmospheric radiotracers are decidedly non-
465 Gaussian (Tositti et al., 2013, 2014), and in this case the median should be preferred over the
466 arithmetic mean as a more robust indicator (e.g., Wilks, 2011).

467 As shown in Figure 4 and as previously highlighted (Tositti et al., 2014; Brattich et al.,
468 2016, 2017a), the seasonal behaviour of ^{210}Pb is characterized by the presence of one summer
469 maximum mainly due to higher mixing height and enhanced uplift from the boundary layer.
470 Conversely, ^7Be seasonal variations are more complex, being characterized by two relative
471 maxima, one during the cold season (March) associated with an increased frequency of STE
472 (James et al., 2003; Stohl et al., 2003; Brattich et al., 2017a) and one in the warm season mainly
473 (but not exclusively) associated with tropospheric subsidence balancing low tropospheric air
474 masses uplift generated by the convective circulation produced by the intense solar heating and
475 the higher tropopause height increase of this season (Ioannidou et al., 2014), occasionally
476 accompanied by STE (Cristofanelli et al., 2009a; Tositti et al., 2014). Figure 5 highlights,

477 however, that the seasonality of radionuclides can also be connected to the seasonality of air
478 mass transport at the site, as previously pointed out by Brattich et al. (2017b) by means of model
479 simulations with a Chemistry and Transport Model. In fact, while ^7Be March maximum seems to
480 be related to the seasonal pattern of Arctic air masses (as Atlantic and North American air
481 masses, presenting also a simultaneous winter peak, are associated with lower ^7Be values in the
482 boxplots of Figure 3), the ^7Be summer maximum seems to correspond to that presented by
483 Mediterranean-Africa, Western and North Atlantic air masses. ^{210}Pb summer maximum seems
484 instead to be well related with the seasonality of Western and North Western-Europe flows.
485 However the monthly analysis is not capable of resolving the contributions of advection
486 pathways occurring in the same month and therefore to uniquely determine a clear connection
487 between advection pattern and concentration.

488 Figure 5 provides similar analyses for the PM_{10} seasonal pattern, which, like ^{210}Pb , show
489 minimum values during the cold season and maxima during summer months, when it is uplifted
490 from the regional boundary layer due to thermal convection and increased mixing height (Tositti
491 et al., 2013). The seasonal pattern of PM_{10} might be, however, influenced by the seasonal pattern
492 of advection pathways bringing about elevated mass loads of particles, such as Mediterranean-
493 Africa, Western, North Atlantic and North Western-Europe air masses. In particular, while the
494 seasonal maximum frequency of Mediterranean-Africa in June contributes to the first PM_{10}
495 increase observed during this month, July values are related to the contribution of North Atlantic
496 flows, and August elevated values are linked to the seasonal pattern of Western and North
497 Western-Europe advections. Figure 6 also shows that the magnitude of the peaks is determined
498 by both the source of trajectories and the concentration over source regions, as indicated by the
499 analysis of the time spent by trajectories over North Africa together with the aerosol optical
500 depth (AOD) over Africa from MODIS Aqua 5.1 collection (Deep Blue AOD at 550 nm). We
501 have found that trajectories spend more time over northern Africa in November than in May, but
502 AOD in Africa is lower in November (mean equal to 0.125 in November vs. 0.396 in May), an
503 observation consistent with the low PM_{10} concentration in November. Similarly, the AOD at 550
504 nm (Land and Ocean) along the western Mediterranean shows higher values for May than for
505 November (0.27 vs. 0.14).

506 Though the seasonal frequency of events accounts for most of the variability, a detailed
507 analysis shows that singular events may make important contributions to some of the parameters
508 observed. For this reason, Figure 6 reports boxplots of the median $^7\text{Be}/^{210}\text{Pb}$ contribution per
509 number of episodes for each season. Figure 8 highlights that both summer Arctic as well as
510 summer North-American flows, though being infrequent, can contribute to increases in ^7Be (and
511 not in ^{210}Pb). Their average contribution to high $^7\text{Be}/^{210}\text{Pb}$ during summertime is higher than
512 during winter when they are more frequent. Figure 8 also emphasizes Arctic, North Atlantic,
513 North-American and Western flows as the main contributors to winter $^7\text{Be}/^{210}\text{Pb}$ increases;
514 Mediterranean-Africa flows are instead associated with expectedly large contributions of ^{210}Pb
515 and PM_{10} , while less obvious, but in agreement with previous studies (Hernández et al., 2008;
516 Menut et al., 2009; Dueñas et al., 2011; Gordo et al., 2015), is the inherently high contribution in
517 ^7Be . The high ^7Be of this flow type can be connected to the intense convection generated by the
518 extremely high temperature of the ground and the very dry conditions in the Sahara desert
519 together with the mineral dust size spectrum including also a large fraction of submicron
520 particles to which ^7Be attaches (Brattich et al., 2017a). This confirms how, given the suitable
521 dynamical framework, the $^7\text{Be}/^{210}\text{Pb}$ ratio is a pragmatic and efficient proxy of vertical motion.

522 523 **3.3 Trend analysis of transport pathways, teleconnection indices and atmospheric composition**

524 The assessment of the existence of temporal trends in the time series of the monthly frequencies
525 of the air flow types, as well as of monthly medians of the variables and of teleconnection
526 indices has considered the presence of seasonality and serial correlations in the time series (see
527 SI). Indeed, as previously reported in the Methodology section, the analysis of the pattern of the
528 ACF (AutoCorrelation Function) can reveal the presence of seasonality in the time series. Here,
529 the previously described seasonal nature of the advection pathways, as well as of the analysed
530 atmospheric variables, is also evidenced by the periodic behaviour of the ACF of their monthly
531 frequencies of occurrence (in the case of advection types) and monthly medians (in the case of
532 atmospheric species), with maxima and minima beyond bounds of significance (95% confidence)
533 and a full cycle of 12 months. Examples of ACF are reported in the SI. In all cases the use of the
534 STL (Seasonal and Trend decomposition using Loess) decomposition allowed the estimation of
535 the relative contributions of the seasonal, trend and residual components, and the subsequent
536 removal of the periodic structure (connected with the seasonal component) in the ACF for further
537 analysis.

538 Since significant data loss occurred for technical reasons in 2007, the trend analysis was
539 restricted only to the 1999-2006 time window. Although this time period is too short to provide
540 definitive trend assessment, this analysis can provide useful hints as to the role of specific
541 processes (e.g. meteorology vs anthropogenic emissions vs atmospheric transport) in modulating
542 the variability of the atmospheric species. In addition, considering the decreasing number of
543 samples towards the end of the time series and the consequent decrease in the number of
544 trajectories, for the analysis of trends we considered the corresponding fraction of each trajectory
545 type with respect to the total number of trajectories in a month. At this step of the research we
546 also included in the analysis the tropopause height data obtained from the Aqua AIRS
547 satellite, available since August 2002 from the NASA Goddard Earth Sciences Data and
548 Information Services Center (<http://mirador.gsfc.nasa.gov/>). The comparison of the tropopause
549 height from radiosoundings and from satellite observations yields a strong significant correlation
550 ($R^2 = 0.83$ for the monthly means and $R^2 = 0.71$ for the monthly medians).

551 Trend analysis on air flow pathways reveals some significant tendencies though of
552 limited extent and also with some differences according to the different approaches applied, in
553 particular seasonal Kendall test and trend-free pre-whitening methods (see Table 2). Both
554 methods consistently detect significant trends in a number of cases, in particular indicating an
555 increasing trend for Arctic flows and a decreasing one for Western flows. The seasonal absence
556 of both Arctic and North-American flows strongly biases the Theil-Sen slope to zero. However,
557 the seasonal Kendall tests suggest the presence of a significant trend in the Arctic time series,
558 and deseasonalization provides a better estimate of the upward trend. As for the variables, the
559 results indicate a strong upward trend for CO_2 , in agreement with the long-term CO_2 behaviour at
560 the global scale (Machta, 1972; Thoning et al., 1989; Randerson et al., 1997; WMO-GAW,
561 2017), and a significant decreasing trend for both the monthly medians of ^{210}Pb and PM_{10}
562 measured at the station in the period 1999-2006 (Figure 7). The mean annual change of the
563 original monthly time series is equal to $+1.80 \text{ ppm year}^{-1}$, $-0.008 \text{ mBq m}^{-3} \text{ year}^{-1}$ and $-0.15 \mu\text{g m}^{-3} \text{ year}^{-1}$,
564 for CO_2 , ^{210}Pb and PM_{10} respectively, while for the de-seasonalized monthly series it is
565 equal to $+1.90 \text{ ppm year}^{-1}$, $-0.01 \text{ mBq m}^{-3} \text{ year}^{-1}$ and $-0.30 \mu\text{g m}^{-3} \text{ year}^{-1}$.

566 The detection of contemporary decreasing trends of ^{210}Pb and PM_{10} is particularly
567 important in light of the decreasing trend of PM_{10} in the period late 90's-2010 observed in many
568 stations in Europe, especially at regional background stations (Pérez et al., 2008; Barmpadimos
569 et al., 2011; Colette et al., 2011; Barmpadimos et al., 2012). Generally, this PM_{10} drop is

570 attributed both to a decrease in anthropogenic emissions, as a result of to the mitigation strategies
571 adopted, as well as to different meteorological processes or cycles, such as the frequency and
572 intensity of Saharan dust episodes (Pérez et al., 2008). Both Colette et al. (2011) and
573 Barmadimos et al. (2012) showed that the decrease in anthropogenic emissions seems to be
574 more important than meteorology as a driving factor for the observed decrease. However, in our
575 case the observation of a contemporary decreasing trend of the ^{210}Pb radionuclide at this remote
576 background site, which cannot be ascribed to a decrease in anthropogenic emissions due to the
577 crustal natural origin of this nuclide, highlights the important role played by meteorology in
578 these decreases.

579 A visual inspection of the time series and their trend components obtained from the
580 seasonal-trend decomposition analysis (Figure 7) suggests that the upward trend of Arctic flows
581 was significant from 2002 on, while Western flows downward trend was almost constant over
582 the 1999-2006 study period. While CO_2 presents an upward trend over the time period, in
583 agreement with, e.g., WMO-GAW global analyses (2017), for ^{210}Pb and PM_{10} the decreasing
584 trend is stronger after 2001. Besides parameters characterised by the existence of significant
585 trends, we also reported results for the two NAO indices with the aim of illustrating the result for
586 this well-known and often studied teleconnection index, showing a slightly decreasing non-
587 significant trend.

588 The increase in ^{210}Pb activity from 2002 to 2003 might be due to the extremely high
589 temperature recorded in the whole European region, especially during the summer months
590 (Cristofanelli et al., 2009a; Pace et al., 2005), and connected also to anomalously high ozone
591 concentrations at Mt. Cimone (Cristofanelli et al., 2007) and to augmented radon exhalation
592 during the 2003 summer heat wave in Europe. In PM_{10} this increase is masked by the 2004
593 maximum connected to an exceptional Saharan dust episode previously described (Beine et al.,
594 2005) which resulted in an event concentration reaching $80 \mu\text{g m}^{-3}$ and characterized by a
595 significant increase in the coarse fraction and a reduced, though not negligible, increase in the
596 fine fraction (to which radionuclides attach), and with a less substantial increase in ^{210}Pb than in
597 PM_{10} (Tositti et al., 2013; Brattich et al., 2015a, b).

598 The analysis of the two tropopause height datasets shows no trends at Mt. Cimone,
599 contrarily to the increasing trend observed globally and suggested as an alternative detection
600 variable of climate change (e.g., Añel et al., 2006), connected to the increase in atmospheric CO_2
601 leading to tropospheric warming and stratospheric cooling, and to anthropogenically induced
602 depletion of stratospheric ozone, also inducing stratospheric cooling (e.g., see Chapter 5 of
603 WMO, 2007; Myhre et al., 2013; Santer et al., 2013). The absence of trends in our case is
604 possibly due to the different and short time window we use.

605 Both of the NAOi time series (the station-based and the Principal Components-based) do
606 not show any significant trends according to the tests, despite presenting a negative T-S slope.
607 For the sake of completeness, we also investigated the results for the CRU station-based NAOi
608 which indicate the absence of statistically significant trends during the analysed period. The only
609 teleconnection index presenting a significant trend during the study period is the WeMOi, with a
610 downward trend constant over the study period. The downward trend is particularly evident in
611 2005 and 2006 in correspondence with very large negative indices indicative of the presence of
612 strong lows in the Gulf of Cádiz and anticyclones in central Europe and associated with an
613 increase of humid airflows travelling over the Mediterranean Sea and a reduction of westerly-
614 northwesterly flows.

615 The analysis of the magnitude of the seasonal and trend components of the time series
616 revealed that the seasonal component dominates over the trend component and the small-time
617 scale variations in almost all the measured atmospheric variables, weighting about twice the
618 trend component. In turn, the small-scale variations dominate most of the teleconnection indices
619 with the exception of MOi, and the frequencies of the different advection pathways.

621 **3.4 Association among air flow types, meteorological/atmospheric parameters and teleconnection** 622 **indices**

623 In this work, the degree of association among air flow types, meteorological/atmospheric
624 parameters and teleconnection indices is assessed by analyzing the Spearman correlation
625 coefficient, considering both the complete yearly time series and separately by season.

626 Figure 8 shows the linear Spearman correlation coefficients between teleconnection
627 indices and flow types, while comprehensive tables with all the correlation coefficients are
628 reported in the SI.

629 The NAO is related to North-American flows (especially in winter), and weakly related
630 to Mediterranean-Africa (during summer and all-yearlong) and North-Atlantic pathways. It is
631 recognized that the positive NAO phase corresponding to a stronger than usual subtropical high-
632 pressure centre and deeper than normal Icelandic low results in more and stronger winter storms
633 crossing the Atlantic Ocean on a more northerly track, while the negative phase is connected to
634 fewer and weaker winter storms crossing on a more west-east pathway (Barnston and Livezey,
635 1987). An anti-correlation of westerly flows reaching three Mediterranean sites (Lecce, Elche
636 and Malaga) with NAOi was previously observed by Orza et al. (2013): this observation is
637 connected with the fact that the position of the subtropical high at lower latitudes during the
638 negative phase of the NAO promotes the access of westerlies (W)/southwesterlies (Me-AF) to
639 the Mediterranean. This is shown in Figure 9, presenting for each spatial grid cell the ratio
640 between the residence time of the air parcels reaching Mt. Cimone during the positive and the
641 negative phases of NAO (NAOi higher than +0.5 and lower than -0.5, respectively) for the
642 extended winter period (DJFM), calculated as the number of trajectory endpoints falling within
643 each area of interest divided by the total number of trajectory endpoints for the entire set of
644 trajectories in the considered time period.

645 Figure 9 reveals that south-westerlies and slower westerlies from the westernmost part of
646 Northern Africa and southern Spain are more frequent during the negative phase of the NAO,
647 while flows from Libya and surrounding regions (also belonging to the Mediterranean-Africa
648 cluster) occur preferentially during the positive NAO phase. Moreover, trajectories coming from
649 North-America are more frequent during the positive phase of NAO, as indicated by the
650 significant high correlation between North-American flows and NAO. Finally, North-Eastern
651 flows seem to be more usually observed during the positive NAO phase, even if this was not
652 readily observed from the correlation analysis.

653 The frequencies of Atlantic, North-Atlantic, North-American and Western flows are
654 related to the WeMOi. In particular, while Atlantic, North-American and Western flows are
655 related to the WeMOi both all-yearlong and during separate seasons specific for each advection
656 pattern, the frequency of North-Atlantic trajectories is strongly connected with WeMOi in
657 summer, and less so considering the whole time series. It is also weakly negatively related to the
658 Eastern advection pattern. These correlations can be easily understood considering that this index
659 measures the difference between the standardized surface pressure values measured in Padua

660 (Italy), and San Fernando (Cadiz, Spain). Therefore, these results suggest a likely connection of
661 the downward trends of Western flows with WeMOi as previously observed.

662 MOi presents weak relations with Western and North-Western Europe pathways, and
663 with North-American flows during winter. Also these relations can be easily understood from the
664 MOi construction as the difference of standardised geopotential height anomalies at Algiers
665 (Alger) and Cairo (Egypt).

666 EA, consisting of a north-south dipole of anomaly centres displaced South-Eastern with
667 respect to the NAO ones, appears negatively related to Arctic flows (especially in autumn, and
668 secondarily in winter and all-yearlong), and positively associated with Western (mostly in
669 winter) and Atlantic flows (not in spring). Its relation with Me-AF has only a winter nature.

670 EA/WR and SCA indices present less relations with air mass pathways, probably due to
671 their limited influence in central Europe; in fact, the EA/WR presents an association with
672 Western flows only during autumn, while the SCA pattern is negatively correlated with Western,
673 Atlantic, North-American and North-Atlantic pathways during winter, while positive
674 associations with North-American and Western flows are observed during summer and autumn,
675 respectively.

676 Figure 10 similarly reports the correlation coefficients between teleconnection indices
677 and the monthly medians of and atmospheric composition variables.

678 The positive correlation of NAO during the transition seasons with particles
679 concentration, even though not statistically significant, is linked to the fact that the positive NAO
680 phases are associated with drier weather conditions in the Mediterranean area which generate
681 intense uplift of particles from the ground; on the contrary, the negative correlation between the
682 station-based NAOi and coarse particles is linked to their transport from Western and
683 Mediterranean-Africa, and to a lesser extent from North Western-Europe flows, and to the
684 association of the negative NAO phase with more westerlies/south-westerlies entering the
685 Mediterranean. The association of the PC-based NAOi to O₃ during summer is in agreement with
686 the results of Pausata et al. (2012) and is linked to the drier conditions in the Mediterranean area
687 associated with the positive NAO phase resulting in the build-up of O₃ because of photochemical
688 processes; however, the transport of O₃ enriched air masses from the Atlantic Ocean, where O₃
689 build-up is linked to the low dispersion capacity of precursors, increase of the photochemical
690 yield and of kinetic reactions due to the high temperature, cannot be completely ruled out. In
691 particular, Pausata et al. (2012) associated the O₃ increase in south-western Europe to transport
692 of air masses from continental Europe, favoured by the presence of a more extended Azores
693 anticyclone. In the case of Mt. Cimone, we observed an increase of Me-AF transport linked with
694 the positive NAO phase. As such, the positive correlation of O₃ with NAOi could be linked to
695 the role of mesoscale circulations (enhanced vertical transport and photochemistry under
696 anticyclonic conditions) which are not resolved in our study due to the coarse resolution of the
697 meteorological field we are using. The index showing the highest number of significant
698 correlations with the variables is the MOi (both indices). In the MO positive phase, when higher
699 pressures are found over the western and central Mediterranean, the storm track is displaced
700 northward and the orientation of the westerly airflows is modified. This causes dry conditions in
701 the Mediterranean basin, with low precipitation and relative humidity. Conversely, low pressures
702 and in particular cyclogenesis over the western/central Mediterranean, which are linked to the
703 negative MO phase, are associated with precipitation and high ⁷Be/²¹⁰Pb and ⁷Be/PM₁₀ ratios.

704 Figure 11 reports the associations between monthly medians of the variables and
705 frequencies of air flow types during different seasons as well as throughout the year. Most of

706 these associations agrees with Figure 3. Amongst the correlations observed, it appears
707 particularly important and interesting to discuss those likely to be connected with the ^{210}Pb and
708 PM_{10} negative trends observed in the previous section: Arctic flows, presenting an upward trend,
709 are negatively related with ^{210}Pb and PM_{10} (all year-long, even though a positive relation during
710 winter season also appears), while Western flows, presenting a downward trend, are positively
711 associated with ^{210}Pb and PM_{10} . The anti-correlation of Arctic flows with ^{210}Pb and PM_{10} is
712 mainly related to the continental origin of ^{210}Pb and PM_{10} , in agreement with Brattich et al.
713 (2016, 2017b). In the Supplementary Material, tables reporting all significant correlation
714 coefficients between teleconnection indices/advection pathways and variables are reported.
715

716 **4 Summary and conclusions**

717 This work focused on finding relationships between the advection pathways and atmospheric
718 composition observed in a long time series of essential climate variables (ECVs) observed at the
719 WMO-GAW station of Mt. Cimone (Italy). Advection pathways were identified by a cluster
720 analysis of back trajectories starting at Mt. Cimone at three different heights; the cluster analysis
721 identified 8 groups at the initiation height of 2200 m, approximately at the height of the station.
722 The results reflect strong seasonal pathways with prevalence of westerlies as typical of mid-
723 latitude Northern Hemisphere sites. The main features of these flow pathways, both from the
724 meteorological and from the atmospheric composition point of view, were analyzed. The results
725 indicate that North-American air masses, associated with subsiding flows originating at high
726 altitudes, are related to low pressures and tropopause heights, cold, and dry air masses, and
727 linked to high wind speeds. These flows are negligible during summertime, besides being related
728 to low concentrations of atmospheric pollutants such as BC, CO, O₃, PM_{10} , but also of
729 atmospheric radionuclides ^7Be and ^{210}Pb . Arctic flows are typically cold (though less than North
730 American ones) and more frequent in the cold season. Being subsiding flows and travelling at
731 high altitudes over remote ocean and ice, they are also connected to low values of atmospheric
732 pollutants such as CO, O₃, BC, but also of particulate matter and ^{210}Pb . On the contrary, but for
733 the same reason, this flow type is associated with high ^7Be and seems connected to SI events.
734 Continental flows from North-Western Europe, Eastern Europe, Western and Mediterranean-
735 Africa are generally linked to higher values of atmospheric components; in particular, NW-
736 Europe, Western and Eastern flows are related to “pollution” events, being associated with high
737 levels of CO, BC, O₃ and fine particles number densities, leading to corresponding increases in
738 PM_{10} . In those cases, the relatively “short” back-trajectories suggest the occurrence of
739 meteorological conditions characterised by low ventilation that, especially during warm months,
740 can also promote the diurnal-scale transport of PBL air-masses to the receptor site (Cristofanelli
741 et al., 2017). Because of their continental origin, these flows are also associated with high ^{210}Pb
742 levels. Mediterranean-Africa flows associated with Saharan Dust events bring about high PM_{10}
743 values, both in the fine and coarse fraction of particles. Interestingly, this flow type was not only
744 associated with high ^{210}Pb values, but also with high ^7Be , which could be connected to the
745 combination of African dust uplifting and subsidence from the upper troposphere.

746 The association of the seasonality of air mass transports with the seasonality of
747 radionuclides and particulate matter has also been studied. In fact, while ^7Be winter maximum is
748 related to the seasonal behaviour of Arctic and North-Atlantic air masses that reach Mt. Cimone
749 after traversing the Alps, ^7Be summer maximum can be connected to the seasonal pattern of
750 Mediterranean-Africa, Western and North Atlantic air masses. ^{210}Pb summer maximum is well

751 related with the seasonality of Western and North Western-Europe flows, whereas the seasonal
752 pattern of PM₁₀ might be, however, influenced by the seasonal pattern of advection types
753 bringing about elevated mass loads of particles, such as Mediterranean-Africa, Western, North
754 Atlantic and North Western-Europe flows.

755 Temporal trends were detected by means of non-parametric techniques applied on the
756 monthly frequencies of flow types and on monthly medians: over the period 1999-2006, an
757 upward trend for Arctic flows and a downward trend for Western flows reaching Mt. Cimone at
758 2200 m was detected. In addition, a downward trend for the monthly medians of ²¹⁰Pb and PM₁₀
759 measured at the station, and a contemporary downward trend for WeMOi during the study
760 period, possibly connected to the decreasing trend of Western flows, were also detected. The
761 simultaneous decreasing trends of both ²¹⁰Pb and PM₁₀ cannot be ascribed exclusively to a
762 decrease in anthropogenic emissions, highlighting the potential influence exerted by
763 meteorology, and suggesting further investigations. In particular, the observation of a positive
764 correlation of ²¹⁰Pb and PM₁₀ with Western air masses, showing a decreasing trend, and a negative
765 correlation with Arctic flows, presenting an increasing trend, seems to largely explain the PM₁₀ and
766 ²¹⁰Pb trends observed in the time series. Significant upward temporal trends were detected for
767 CO₂, in agreement with longer time records. The analysis of the magnitude of the seasonal and
768 trend components of the monthly time series revealed that the largest variabilities in almost all
769 the studied atmospheric variables are associated with the seasonal components, with a reduced
770 weight of the trend component for all the series.

771 The association of teleconnection indices with advection pathways and atmospheric
772 variables was also examined. In particular, positive associations of NAOi with the frequency of
773 North-American, Atlantic and North-Atlantic flows, and between WeMOi and Western, Atlantic
774 North-American and North-Atlantic flow types, were observed. The relationship between
775 teleconnection indices and atmospheric variables highlight the significant influence of regional
776 short scale modes of variability, like MO, over synoptic conditions and atmospheric conditions at
777 the sampling site.

778 The results of this work highlight the role of flow pathways and teleconnections as
779 factors that can have a deep influence in the variations in atmospheric composition at a site
780 located in the central Mediterranean. This was possible since the time series of data acquired at
781 the station was long enough to characterize a sort of short-term climatology of the site. The
782 results are therefore of paramount importance to better understand processes controlling the
783 variability of atmospheric composition in a region recognized as a hotspot of air pollution and
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1158 **Tables**

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1160 **Table 1.** Analysed teleconnection with associated location of centers of action including the sign
 1161 of geopotential height (or pressure) anomalies for their positive phases.
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TELECONNECTION	ABBREVIATION	CENTERS OF ACTION
NORTH ATLANTIC OSCILLATION	NAO	Greenland (-), Azores (+)
EAST ATLANTIC	EA	North Atlantic (-), Subtropical North Atlantic and Mediterranean (+)
EAST ATLANTIC/WESTERN RUSSIA	EA/WR	NW Europe (+), Western Russia (-), NE China (+)
SCANDINAVIA	SCA	SW Europe (-), Scandinavia (+), Kazakhstan/Mongolia (-)
MEDITERRANEAN OSCILLATION	MO	Algiers (+), Cairo (-), Gibraltar (+), Israel (-)
WESTERN MEDITERRANEAN OSCILLATION	WeMO	Po Valley (-), Gulf of Cadiz (+)

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1187 **Table 2.** Results of the seasonal Kendall test for the monthly time series and the trend-free pre-
 1188 whitening Yue-Pilon (Y-P) procedure on the de-seasonalized monthly series for the detection of
 1189 monotonic trends applied on the 1999-2006 time series. For each case, the p (significance) value
 1190 and the mean change per year from the Theil-Sen slope are presented. In bold when significant at
 1191 the 0.05 level, in italic when the trend is only weakly significant, i.e., at the 0.1 level.
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MONTHLY FREQUENCIES				
INDEX	Seasonal Kendall		Deseasonalized Y-P	
	p value	mean change per year	p value	mean change per year
Hurrell_Stat_NAOi	0.6935	-0.07	0.3220	-0.08
Hurrell_PC_NAOi	0.2840	-0.05	0.3645	-0.05
CPC_Stat_NAOi	0.2110	-0.07	0.1307	-0.06
CRU_Stat_NAOi	0.4320	-0.07	0.4215	+0.04
WeMOi	0.0000	-0.15	0.0000	-0.17
MOi1	0.2530	+0.01	0.2497	+0.01
MOi2	0.6171	-0.01	0.1668	-0.01
EA	0.7206	+0.02	0.9028	+0.01
EA/WR	1.0000	-0.03	0.3612	-0.03
SCA	1.0000	+0.01	0.8875	+0.01
FLOW TYPE	Seasonal Kendall		Deseasonalized Y-P	
	p value	mean change per year	p value	mean change per year
A	0.0383	0.000	0.0462	+0.007
E	0.0760	+0.003	0.1358	+0.007
Me-AF	0.8284	+0.003	0.3924	+0.001
W	0.0376	-0.010	0.0274	-0.011
Atl	0.1061	-0.006	0.0349	-0.007
N-Am	0.0689	0.000	0.2872	-0.003
N-Atl	0.1605	-0.004	0.3462	-0.004
NW-Eu	0.7203	+0.003	0.9232	0.000
Monthly medians				
VARIABLE	Seasonal Kendall		Deseasonalized Y-P	
	p value	mean change per year	p value	mean change per year
p (mbar)	0.1237	+0.317	0.2317	+0.133
T (°C)	0.1855	+0.300	0.1024	+0.168
RH (%)	0.1234	+0.263	0.4996	-0.336
TH (m)	0.7690	+19.296	0.9919	-10.414
ws (m s ⁻¹)	0.1336	+0.054	0.2292	+0.117
Prec (mm)	0.6408	+0.000	0.7777	+0.000
MixHeight (mm)	0.9083	+4.822	0.6875	-3.568
O ₃ (ppbv)	0.1320	+0.279	0.1806	+0.292

CO₂ (ppm)	0.0000	+1.804	0.0000	+1.900
⁷Be (mBq m⁻³)	0.2840	-0.079	0.1984	-0.085
²¹⁰Pb (mBq m⁻³)	0.0450	-0.008	0.0135	-0.011
PM₁₀ (μg m⁻³)	0.0053	-0.154	0.0083	-0.296
⁷Be/PM₁₀ (mBq μg⁻¹)	0.1851	+0.007	0.1616	+0.012
²¹⁰Pb /PM₁₀ (mBq μg⁻¹)	0.7921	0.000	0.9839	+0.000
⁷Be/²¹⁰Pb	0.6678	0.000	0.3612	+0.083

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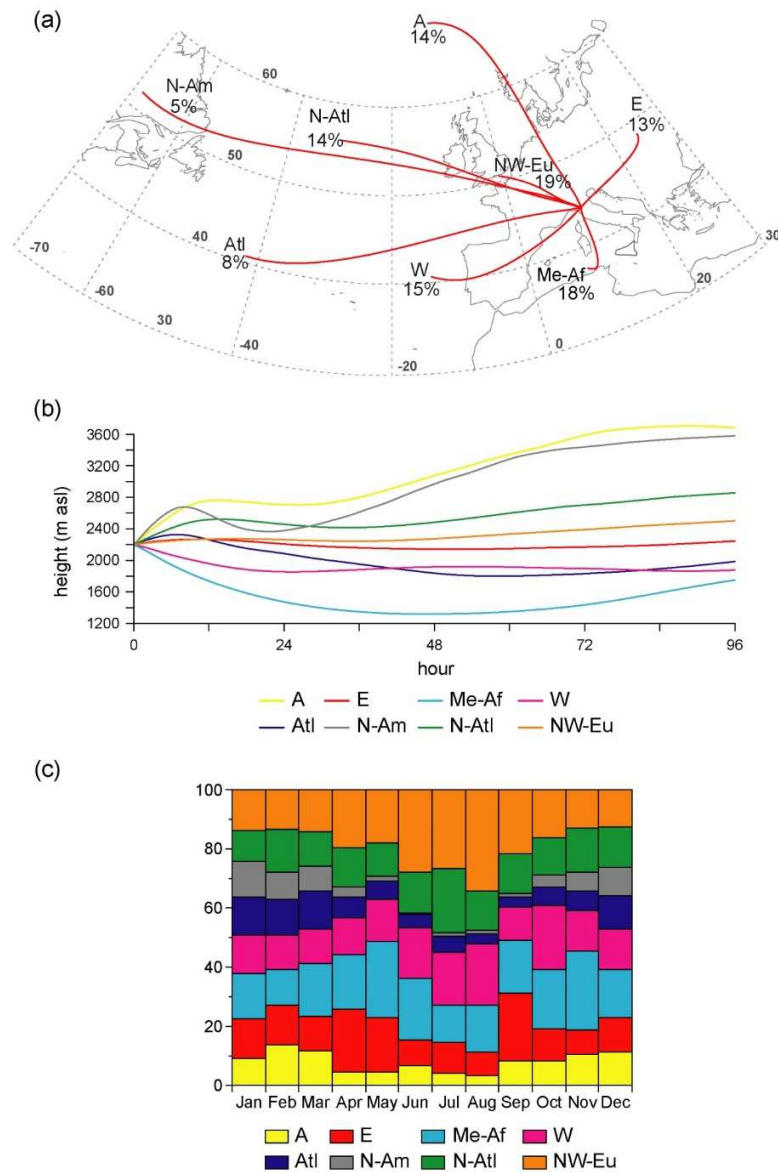
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1215 **Figures**

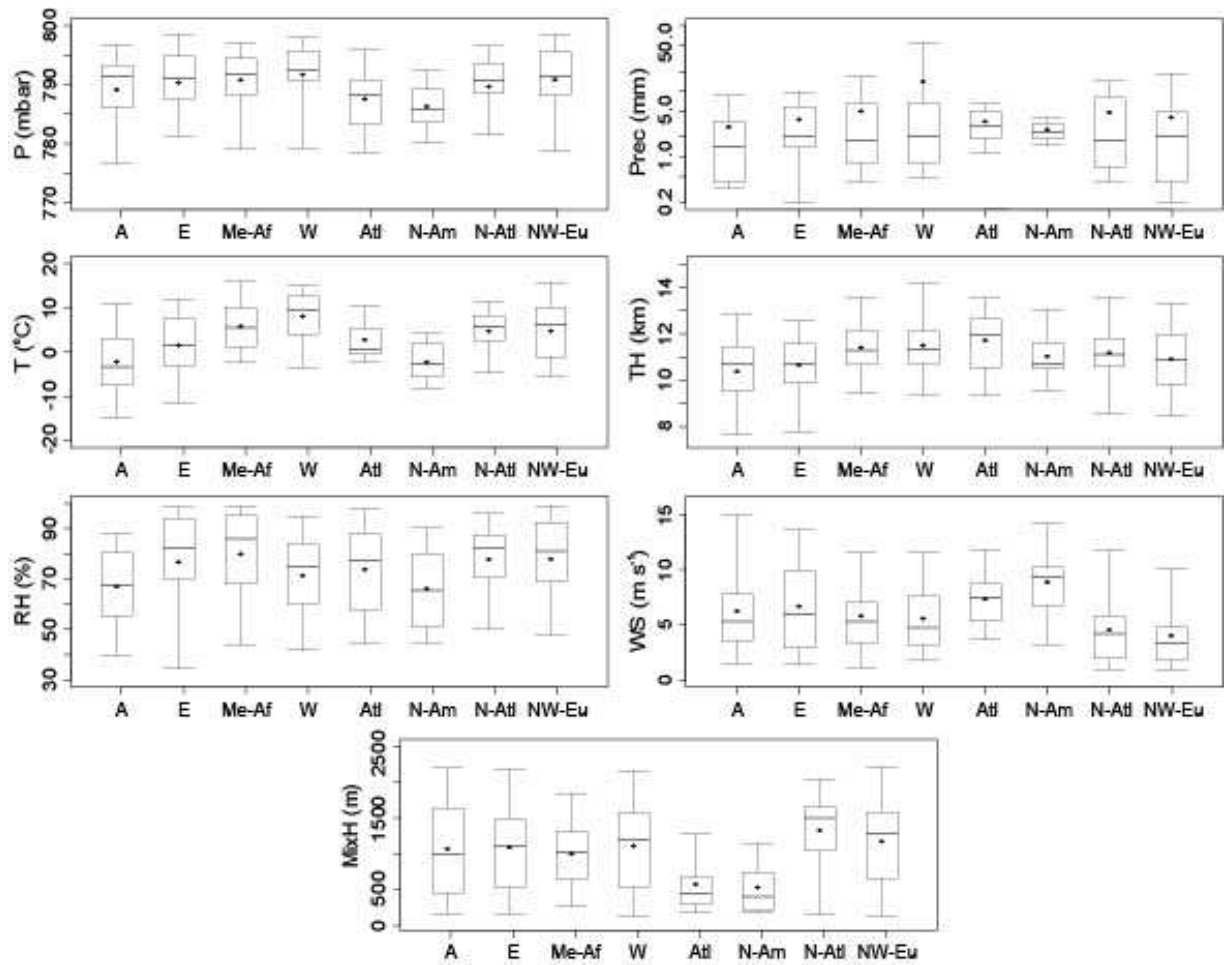
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1217 **Figure 1.** (a) Centroids of the trajectory clusters identified for 96-h back-trajectories arriving at
 1218 2200 m asl for the 12-year study period. The flow pathways are identified as follows: Arctic (A),
 1219 Eastern (E), Mediterranean-Africa (Me-AF), Atlantic (Atl), Northern Atlantic (N-Atl), North
 1220 America (N Am), North Western-Europe (NW-Eu). The percentage is for the frequency of
 1221 occurrence of each flow pattern in the whole 1998-2011 period. (b) Heights above mean sea
 1222 level of the representative back-trajectories vs. end-point time. (c) Monthly variation in
 1223 percentage frequency of the identified advection pathways.

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1229 **Figure 2.** Box plots of meteorological variables measured at Mt. Cimone (P = pressure, T =
1230 temperature, RH = relative humidity, Prec = precipitation, TH = tropopause height, WS = wind
1231 speed, MixH = mixing height) versus air flows arriving at the receptor site. The horizontal bold
1232 line in each box represents the 50th percentile (median), the circle represents the mean value,
1233 lower and upper boundaries locate the 5th and 95th percentile of the values and whiskers locate
1234 the minimum and maximum values.
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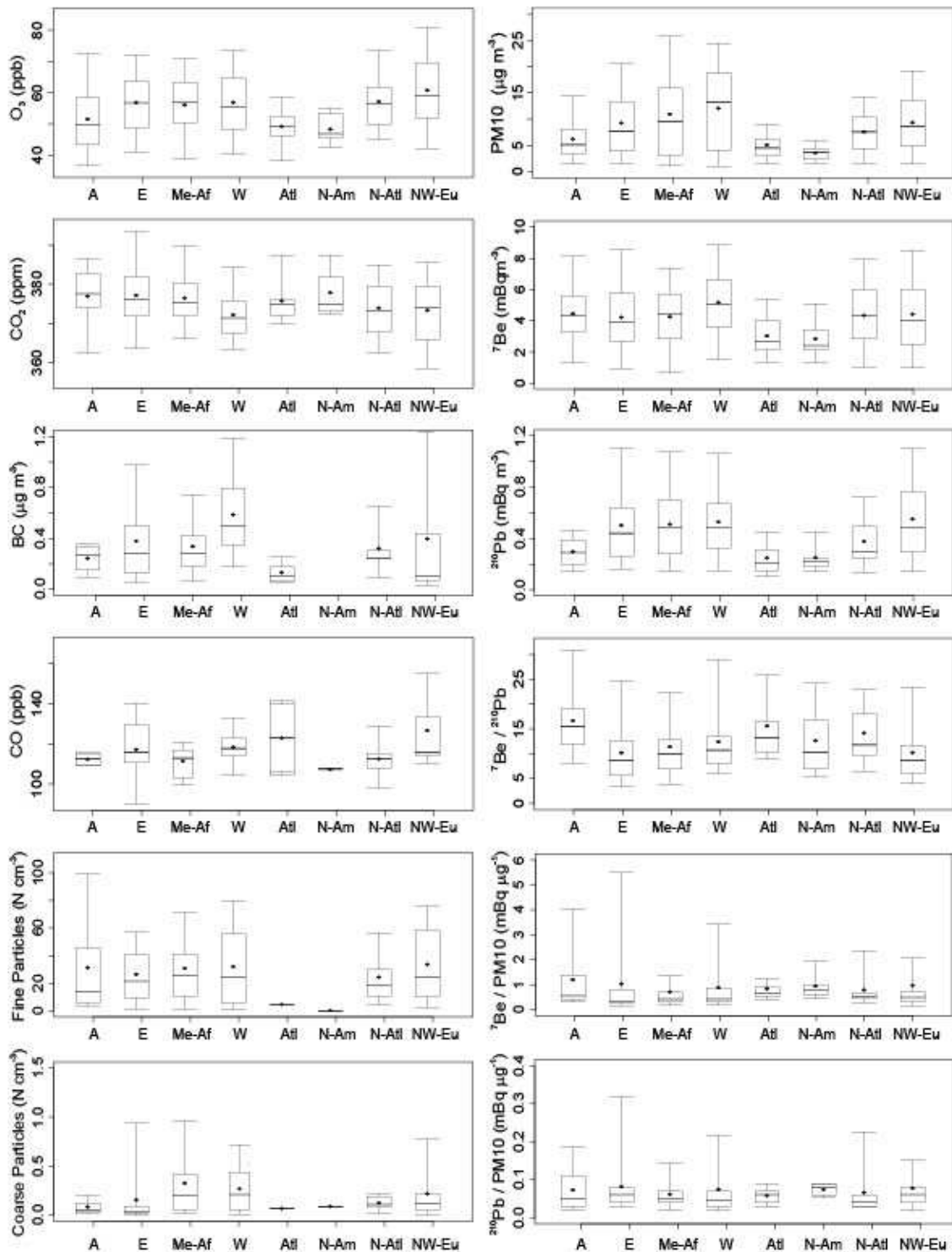
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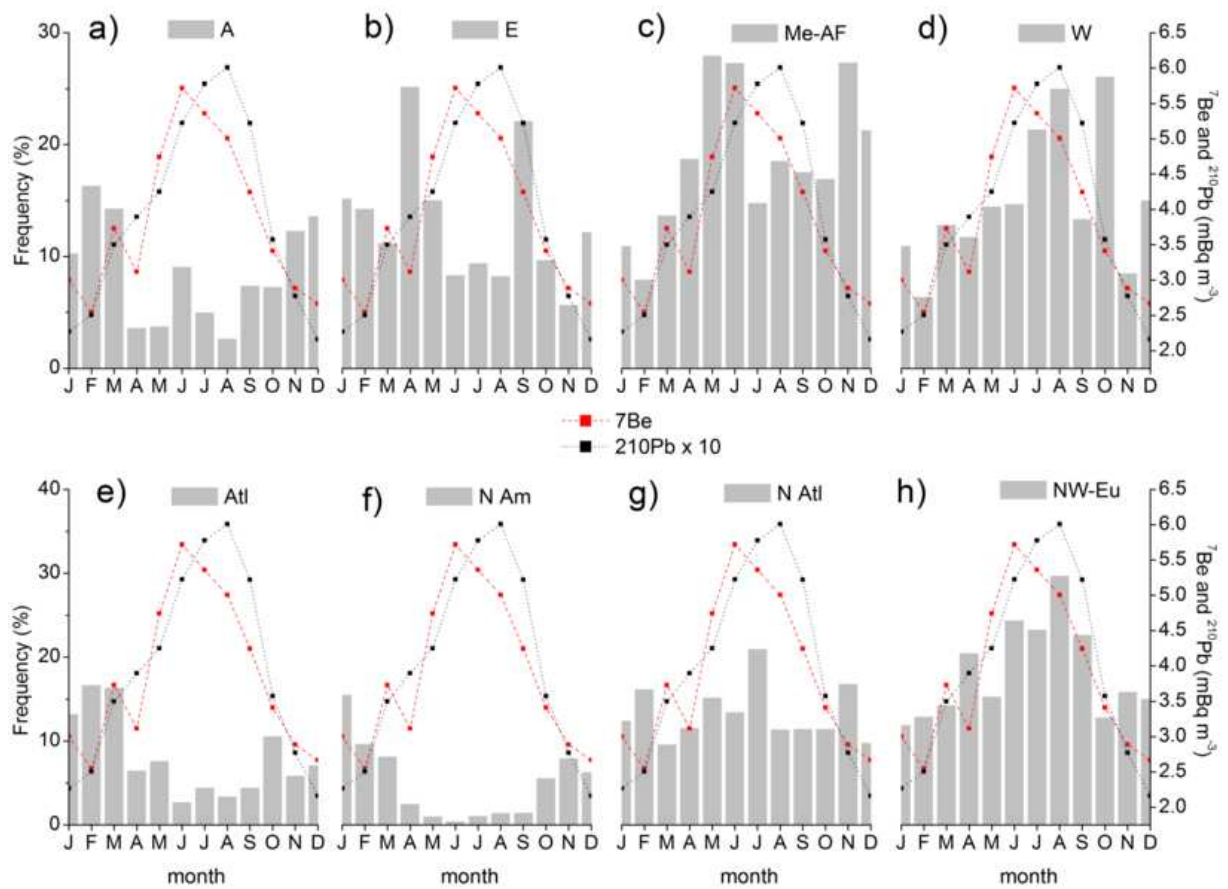
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1246 **Figure 3.** Same as Figure 2, but for atmospheric gases (O_3 , CO_2 , CO), black carbon (BC), fine
 1247 and coarse particles number density, PM_{10} , atmospheric radiotracers 7Be and ${}^{210}Pb$, ratio
 1248 ${}^7Be/{}^{210}Pb$, ratio ${}^7Be/PM_{10}$, ratio ${}^{210}Pb/PM_{10}$, versus air flows arriving at the receptor site.
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1251 **Figure 4.** Monthly median activities of ^7Be (right scale, red line) and ^{210}Pb (right scale, black
 1252 line) and their relationship with the monthly frequency of air flows (left scale, grey bar) at Mt.
 1253 Cimone from: a) Arctic; b) East; c) Mediterranean-Africa; d) West; e) Atlantic; f) North
 1254 America; g) North Atlantic; h) North Western-Europe

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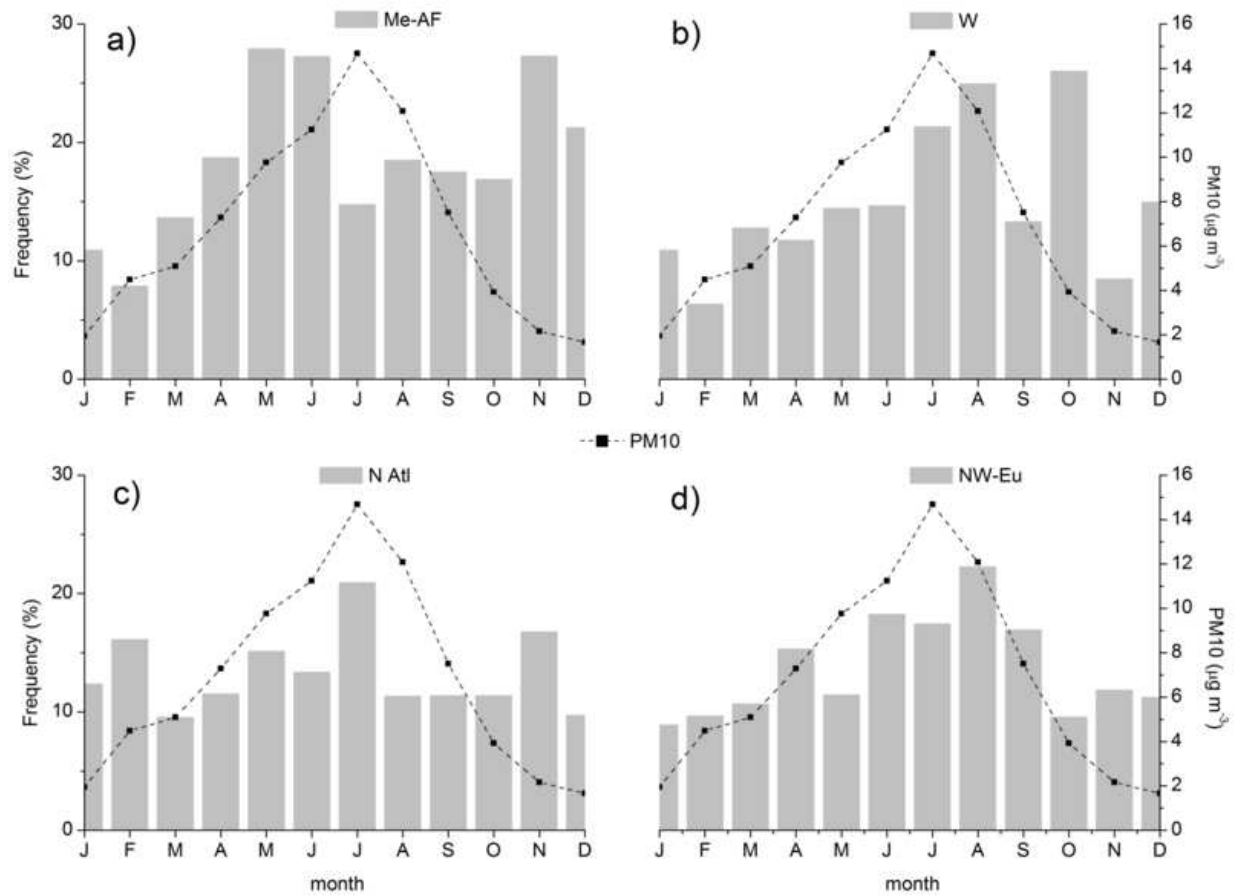
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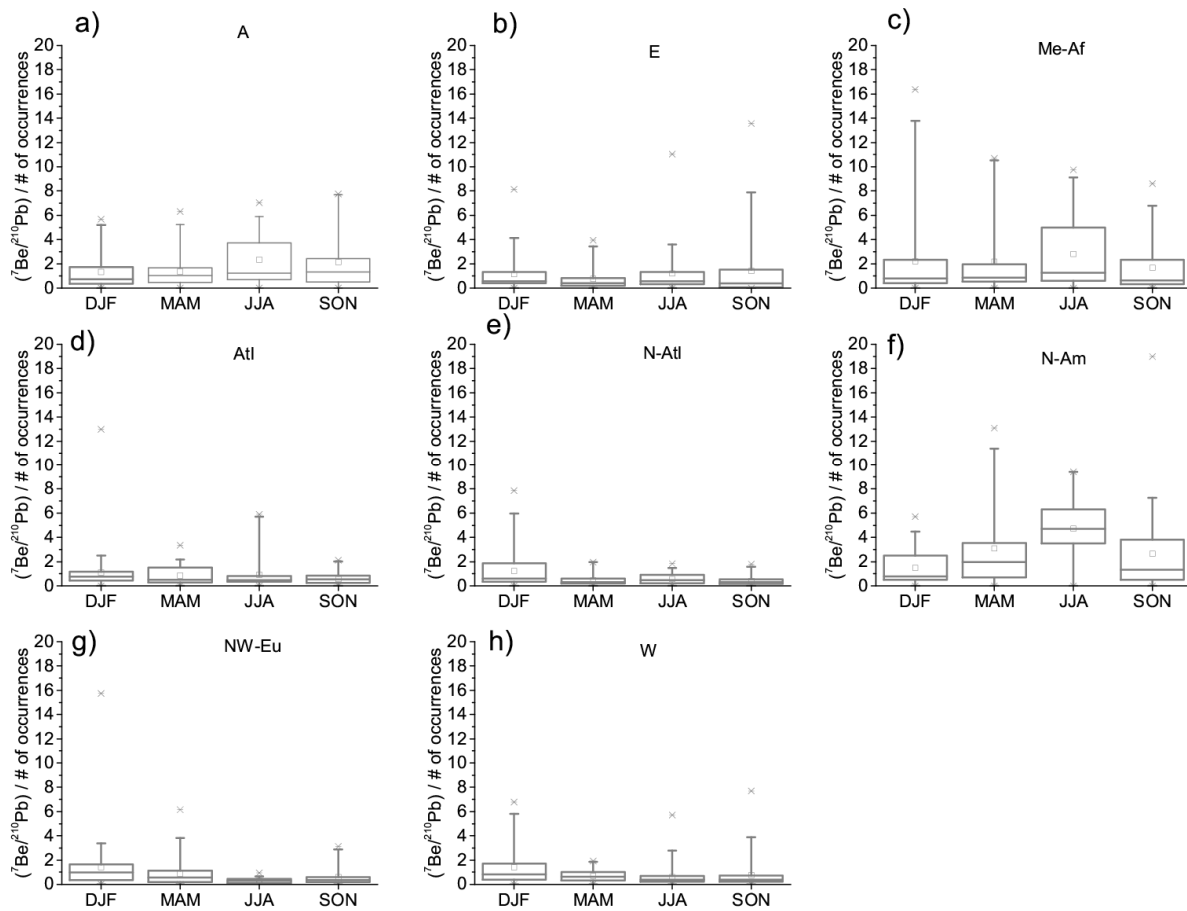


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1270 **Figure 5.** Monthly median concentrations of PM₁₀ (right scale, black dashed line) and
1271 relationship with the monthly frequency of air flows (left scale, grey column) at Mt. Cimone
1272 from: a) Mediterranean-Africa; b) West; c) North Atlantic; d) North Western-Europe.
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1287 **Figure 6.** Seasonal (DJF = December-January-February; MAM = March-April-May; JJA =
 1288 June-July-August; SON = September-October-November; i.e., winter, spring, summer and
 1289 autumn seasons in the Northern Hemisphere) boxplots showing the contribution to ${}^7\text{Be}/{}^{210}\text{Pb}$ per
 1290 number of events of each flow type: a) Arctic; b) Eastern; c) Mediterranean-Africa; d) Atlantic;
 1291 e) North-Atlantic; f) North-America; g) North Western-Europe; h) Western. The horizontal bold
 1292 line in each box represents the 50th percentile (median), the square represents the mean value,
 1293 lower and upper boundaries locate the 25th and 75th percentile of the values and whiskers locate
 1294 the 5th and 95th percentile values. Crosses and horizontal lines outside the boxes further indicate
 1295 1st and 99th percentile and minimum and maximum values, respectively.

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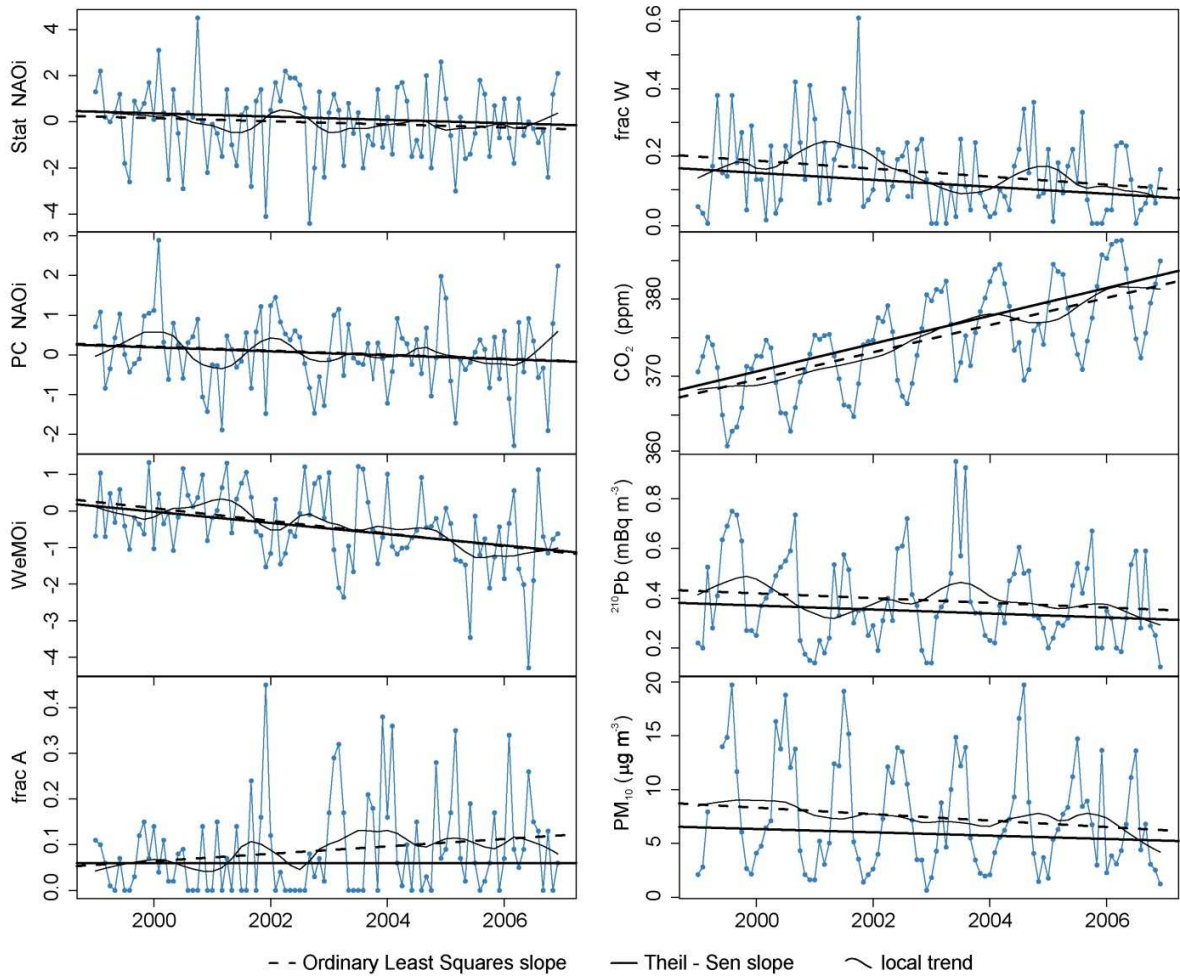
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1306 **Figure 7.** Evolution of the monthly frequency of occurrence of the Hurrell station- and principal
 1307 components-based NAO, WeMO indices, of the Arctic and Western flow types, and of the
 1308 monthly medians of variables which show significant trends, over the period 1999-2006 (CO_2 ,
 1309 ^{210}Pb and PM_{10}). Dashed lines are the linear regressions, solid lines are the Theil-Sen slope
 1310 estimates, and black solid curved lines are the local trends from the seasonal-trend
 1311 decomposition analysis.

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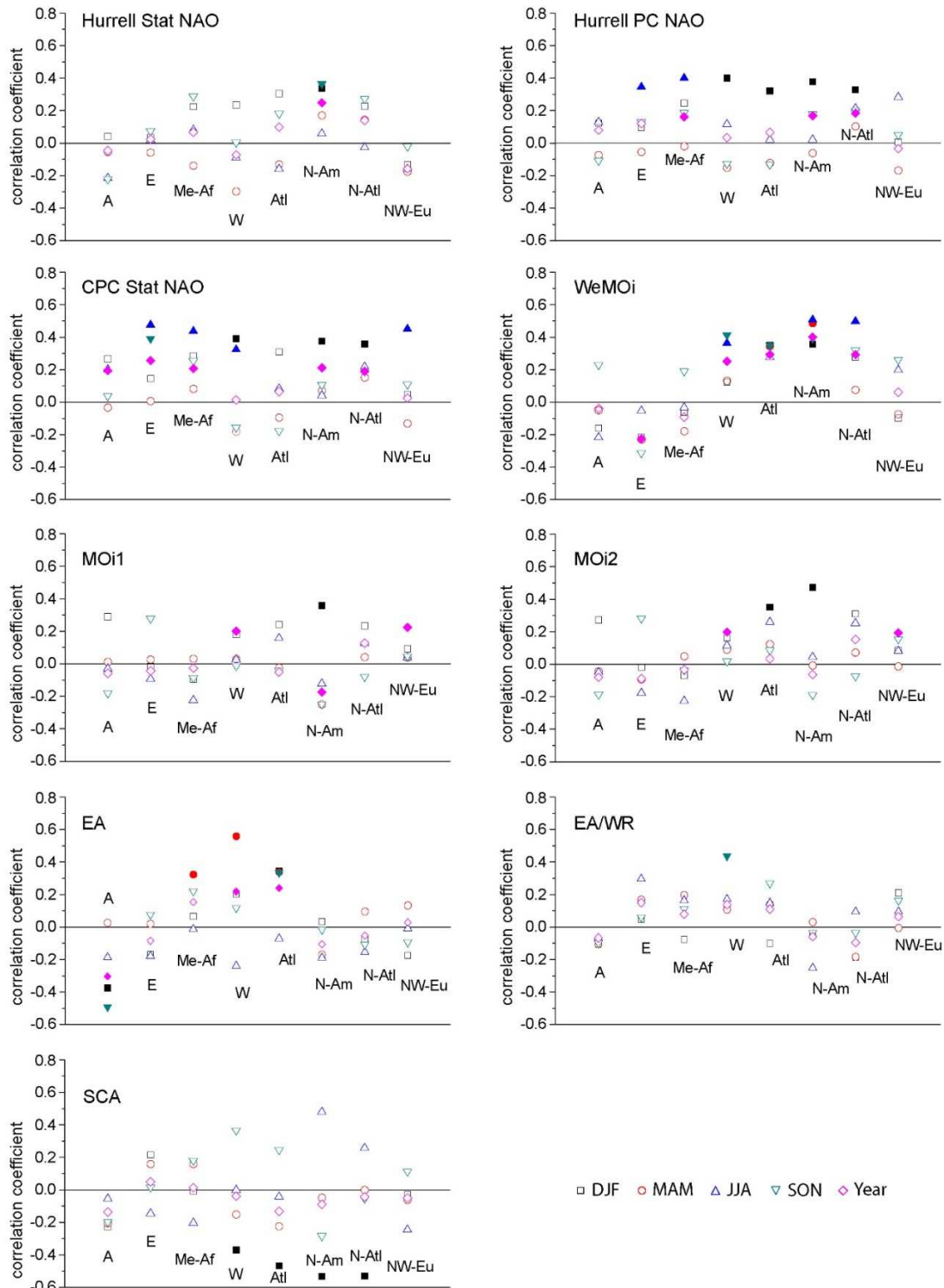
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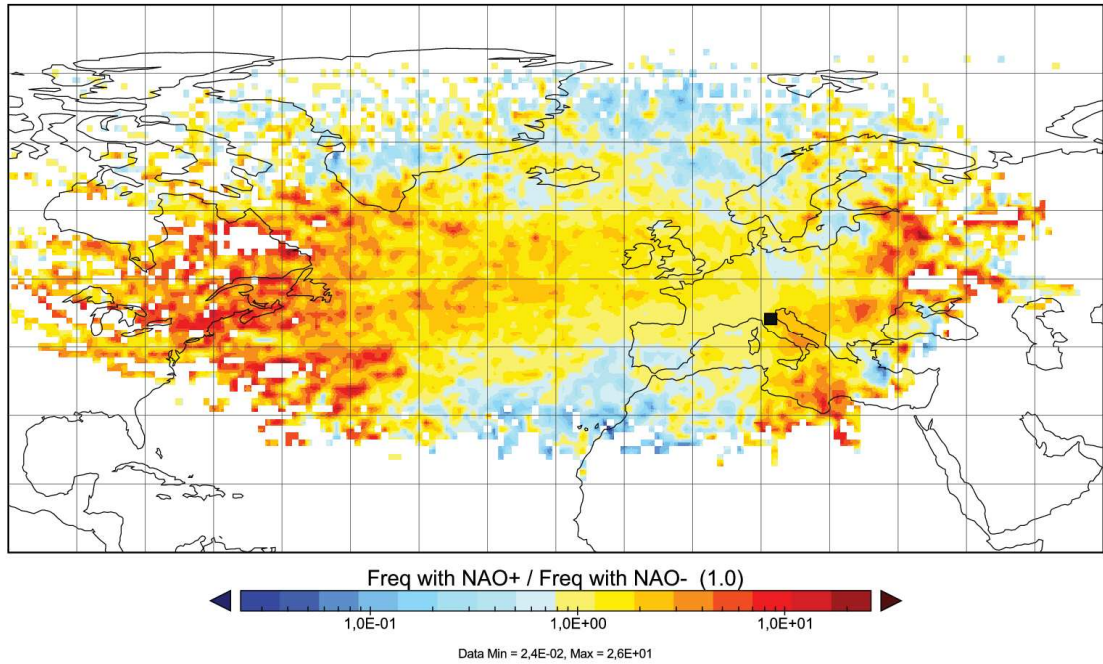
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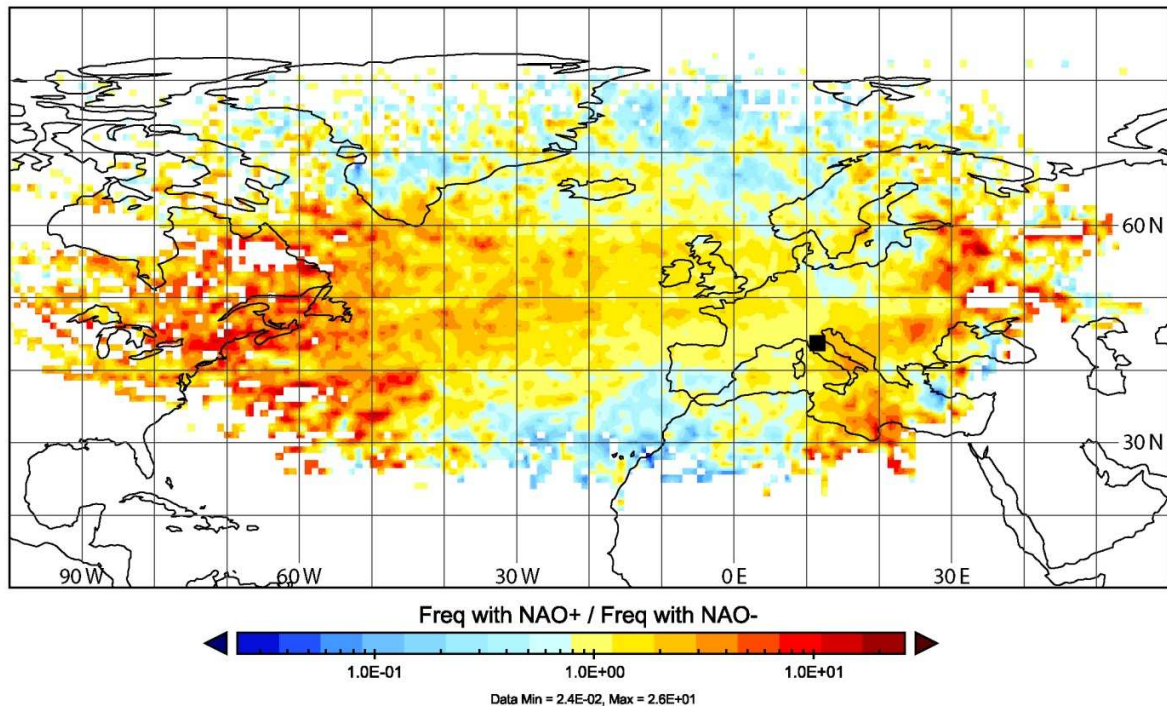


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1323 **Figure 8.** Spearman correlation coefficients between the frequency of occurrence of the different
 1324 teleconnection indices and air flow types by season and for the full year. Filled symbols indicate
 1325 significant correlations ($p < 0.01$ significance level).
 1326



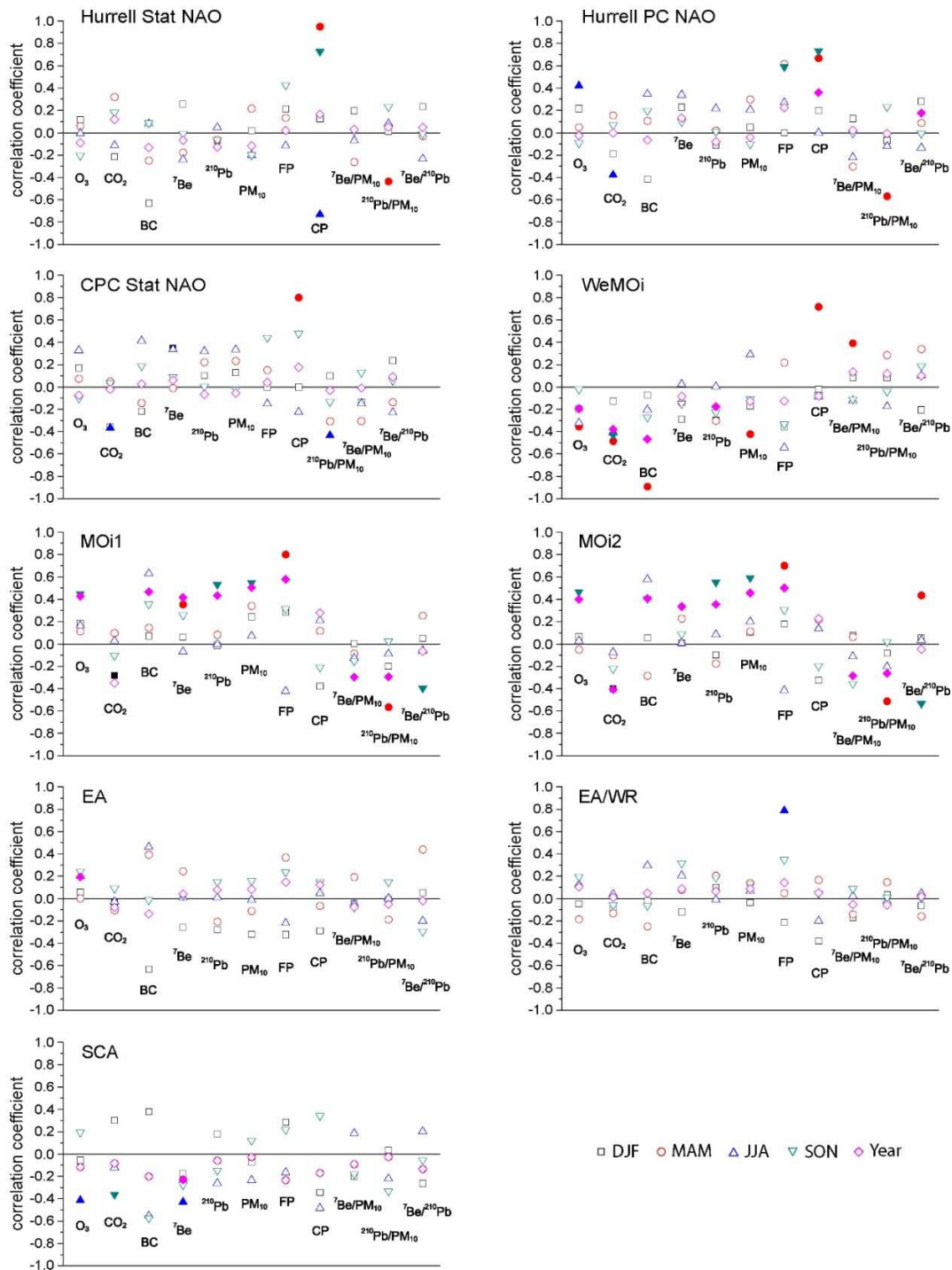
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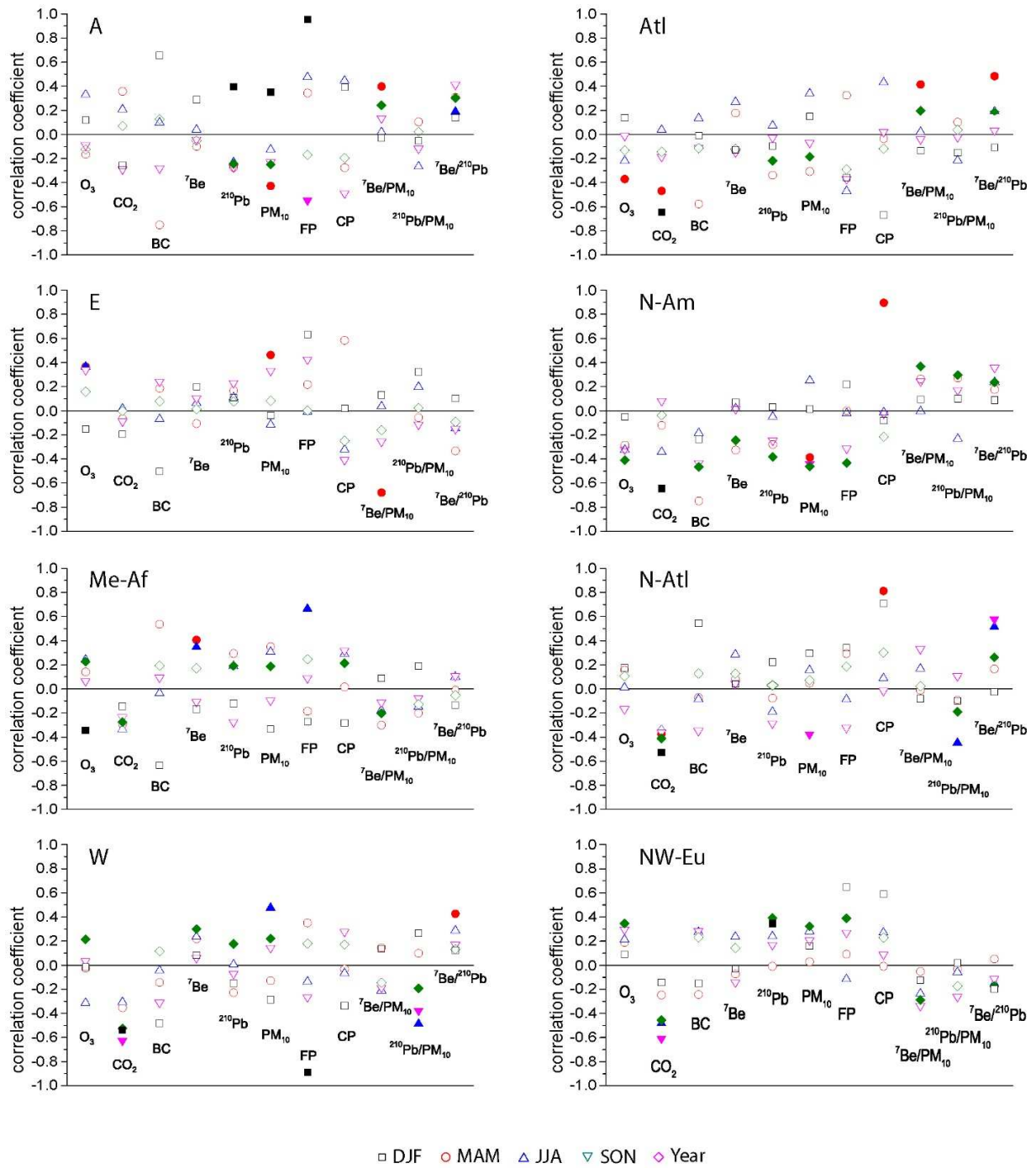
1329 **Figure 9.** Ratio of residence time of air parcels reaching Mt. Cimone in the positive and negative
 1330 phase of Hurrell Stat NAO (NAOi higher than +0.5 and lower than -0.5, respectively) in the
 1331 extended winter DJFM period. The black dot indicates the position of Mt. Cimone.
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1334

1335 **Figure 10.** Spearman correlation coefficients between the teleconnection indices and the
 1336 monthly medians of variables by season and for the full year. Filled symbols indicate significant
 1337 correlations ($p < 0.01$ significance level) letters indicate the variables: O_3 = ozone, CO_2 = carbon
 1338 dioxide, BC = black-carbon, ${}^7\text{Be}$, ${}^{210}\text{Pb}$, PM_{10} , FP = fine particles, CP = coarse particles,
 1339 ${}^7\text{Be}/PM_{10}$, ${}^{210}\text{Pb}/PM_{10}$, ${}^7\text{Be}/{}^{210}\text{Pb}$.
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1342 **Figure 11.** Same as Figure 10 but for the correlation between the frequency of occurrence of air
1343 flow types and the monthly medians of variables.
1344
1345

Journal Pre-proof

Author contributions

Use this form to specify the contribution of each author of your manuscript. A distinction is made between five types of contributions: Conceived and designed the analysis; Collected the data; Contributed data or analysis tools; Performed the analysis; Wrote the paper.

For each author of your manuscript, please indicate the types of contributions the author has made. An author may have made more than one type of contribution. Optionally, for each contribution type, you may specify the contribution of an author in more detail by providing a one-sentence statement in which the contribution is summarized. In the case of an author who contributed to performing the analysis, the author's contribution for instance could be specified in more detail as 'Performed the computer simulations', 'Performed the statistical analysis', or 'Performed the text mining analysis'.

If an author has made a contribution that is not covered by the five pre-defined contribution types, then please choose 'Other contribution' and provide a one-sentence statement summarizing the author's contribution.

Manuscript title: Advection pathways at the Mt. Cimone WMO-GAW station: seasonality, trends, and influence on atmospheric composition

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