Aerosols over Continental Portugal (1978-1993): their sources and an impact on the regional climate

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Author's Response

TABLE OF CONTENT:

We are very thankful to Anonymous Referee #1 for being so supportive of our work presented in this paper. Here we provide some replies to his/her comments and suggestions
(please note that the revised text is in italic and specific differences to the original text are in bold):

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1. Page 31010, line 17. Abstract: The most significant finding is the decrease of the daily temperature (and diurnal temperature range) related to the decrease of sunshine duration observed during the summer periods of increased content of the absorbing aerosols in the atmosphere. Is this a new finding or has been observed by other authors elsewhere? Indicate the main implications of this finding.

Yes, similar relations were found in previous studies. As is mentioned in the 2nd paragraph of the Sec. 4.1 "This cooling trend coincides with epochs of frequent SDE events (high <AIpos> values) and is in an agreement both with most recent/precise measurement (Santos et al., 2008) and with modeling studies (Santos et al., 2013)." Also, this effect is discussed in del Rio et al. (2012). We inserted the following paragraph in the Sec. 4.1 (after 2nd paragraph there):
The relations between the temperatures over Iberian Peninsula and SshD during the second half of the 20th c. were earlier reported (see del Rio et al., 2012 and references therein). They were attributed, mainly, to the variations of the circulation patterns over the North Atlantic and consequent changes in the cloudiness. However, accordingly to the data of our analysis, the variations of the SshD can result also from the strong dust intrusions.
New reference is added to the Reference list:
del Río, S., Cano-Ortiz, A., Herrero, L., Penas, A.:Recent trends in mean maximum and minimum air temperatures over Spain (1961–2006), Theor. Appl. Climatol., 109, 605–626, DOI 10.1007/s00704-012-0593-2, 2012.

2. Page 31011, Line 26: "The detailed analysis of the properties and time variations of the Portuguese aerosols can be found in Pereira et al. (2005, 2008, 2011, 2012), Santos et al. (2008, 2013), Catry et al. (2009), Calvo et al. (2010), Obregón et al. (2012)." The study carried out by Calvo et al. (2010) is developed in Spain and not in Portugal.

The sentence is changed to:
The detailed analysis of the properties and time variations of the Portuguese aerosols can be found in Pereira et al. (2005, 2008, 2011, 2012), Santos et al. (2008, 2013), Catry et al. (2009), Obregón et al. (2012), Vicente et al. (2012, 2013), Evtyugina et al. (2013); the analysis of the radiative effect of the aerosols originated from wildfires for the close region in the north-western Spain is presented in Calvo et al. (2010).

3. Page 31012, Line 5: "The present paper is dedicated to understanding of the local aerosol sources and the role of the local aerosol content played in variations of the climate of the Continental Portugal region for the 1978–1993 period". Do authors considerer volcanic emissions as a local source? Furthermore, this sentence should be rewritten; it is not clear.

The sentence is changed to:
The present paper is dedicated to understanding of the ***local and global*** *aerosol sources and* ***the effect of the local aerosol content in climate variations*** *of the Continental Portugal region for the 1978-1993 period.*

4. Page 31012- 31013: from page 31013, line 5 to page 31013, line 16: this section should be shortened. Too much information is given here. For example, it is not necessary to mention all the data sources used, they have already been described in section 2.

These lines present "Introduction" into our paper. Here we put the main tasks and need to explain what we will investigate. We can't delete this information and shorten text. However the list of the aerosol sources is removed from the 1st paragraph of the Sec. 3 because they are already described in the previous section.

5. Acronyms should be described the first time they appeared. For example TOMS is mention by the first time in page 31012, line 11 and their significance in reported in page 31013, line 19.

Corrected. Now the TOMS acronym significance is explained in the Introduction (4th paragraph there) and the FFT acronym is explained in the new Sec. 2.3 “Atmospheric parameters”.

6. Authors describe the study zones in section 2.1. “Aerosol parameters”. However, I would recommend including a new section entitled “Study zones” with this information.

Now the Sec. 2 includes sub-section 2.1 “Studied locations” containing the information on the sites of the measurements of the aerosol index and atmospheric parameters:
We use the aerosol data over two locations in the Continental Portugal (see Fig. 1a) – the only available TOMS aerosol data for this region. The first one is the site ID 082 over Lisbon (38° 46′ N, 9° 8′ W, 105 m a.s.l.), the second one is the site ID 288 over Penhas Douradas (40° 25′ N, 7° 33′ W, 1380 m a.s.l.). In the first case the region around the site is one of the most urbanized and industrial sites in Portugal where the anthropogenic effects expected to be strong. The second site corresponds to a less populated mountain region affected by the anthropogenic pollution in a lower degree but frequently exposed to forest fire smokes and dust events (Pereira et al., 2005; Pereira et al., 2008; Obregón et al., 2012). Hereafter we use a term “urban” for the site ID 082 and a term “rural” for the site ID 288.
Consequently, we used climatic data measured by two meteorological observatories that are close to the AI sites. The first data set belongs the Geophysical Institute of University of Coimbra (hereafter, “IGUC series”). The second set belongs to the Geophysical Institute of Instituto Dom Luiz of University of Lisbon (hereafter, “IGIDL series”). Both locations are shown on the map in Fig. 1a (marked as "Coimbra" and "Lisbon", respectively).

7. Page 31014, line 13: Replace “For each of two sites and for each of the months: : :.” by “For each site and for each month: : :”

Corrected.

8. Page 31015, line 15: Replace “Not only spatial and temporal distributions of aerosols are very variable but also their origin as well” by “Not only spatial and temporal distributions of aerosols are very variable but also their origin”.

Corrected.

9. Page 31016, line 13: Bourassa and Robock (2012) should be Bourassa et al., (2012).

Corrected.

10. For SO2 concentration estimation, authors calculate a mean value from five EMEP stations. Since EMEP stations are mainly background stations, how can this fact influence the conclusions obtained?

In the Supplementary Material to the presented manuscript (now this part is inserted in the main text) it is shown that the smoothed monthly variations of SO_2 (obtained from background stations of Continental Portugal) well confirm the behaviour of smoothed aerosol indices <AIneg> (obtained by satellites data). The correlation coefficients are high enough – see Figure S1.7 (now Fig. 2g). This result allows us to conclude that SO_2 can be one of the important pollutants that influence the whole aerosol content over the Continental Portugal. Regrettably, the measured data for other pollutants (like NO_2) that are available from the same data base for the studied period are very fragmented, and can't be used to create a reliable composite series. This situation is discussed in the end of the Sec. 3.1.4. (p. 31020, l. 19-26).

11. Figure captions included in the manuscript and in the supplementary information should be checked. Sometimes the information in the figure caption is already indicated in the figure. For example Fig. 2: Figure caption should be shortened. The information: ": : :gray bars show data related to the site ID 082; red-white crossed bars show data related to the site ID 288." is already indicated in the figures.

Figures captions are corrected and repetitions are removed.

12. Page 31020, line 2: "The annual values of the SO2 content are shown in Fig. 2f. As one can see, there is a strong dependence between the variations of the <AIneg> (shown in Fig. 2b) and the SO2 content". Please, indicate the correlation coefficient in the text.

Now the correlation coefficient is mentioned in the text:
The annual values of the SO_2 content are shown in Fig. 2f. As one can see, there is a strong dependence between the variations of the <AIneg> (shown in Fig. 2b) and the SO_2 content. The anti-correlation ***(correlation coefficient r = -0.53, p value = 0.06)*** *between the curves reflects the increase of the scattering particles in the atmosphere (lower <AIneg> values) coinciding with the growth of the measured SO_2 concentration.*

13. Page 31022, line 20: write a comma between "SDEs" and "the wildfires"

Corrected

14. Page 31023, line 3: "To our mind, this is a result of the different pollution and circulation conditions over the sites." Please, rewrite this sentence trying to clarify what authors want to say.

The whole paragraph is rewritten now:
Here we present the analysis of the relations between the aerosol content and the atmospheric parameters described in Section 2. The analysis was done separately for two locations. The analysis of the climatic conditions between the Lisbon and Coimbra (see the Supplementary

Material, Part 1.2) showed their strong similarity. This similarity results from the relatively short distance between these locations and their proximity to the ocean. On the other hand, the measured AI monthly means, as was discussed in Sect. 3, are different for these two sites. To our mind, there are two main reasons for these differences: First reason is that the Lisbon area is much more polluted than the region around the rural site (ID 288); second reason is that the more north-eastern position of the site ID 288 provides this location is affected by the dust intrusions more frequently.

15. Page 31011, line 8 and page 31024, line 3: IPCC 2013, is not in the reference list

New reference is added in the Reference List and corresponding sentences are corrected accordingly:
Climate Change 2013: The Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, IPCC (Ed.), Cambridge University Press, 2014.

16. Page 31027, lines 6- 19: Conclusions. The first paragraph is an abstract of the study carried out, it is not a conclusion. Authors can include an introduction sentence, but not 15 lines.

The conclusion is shortened, the 1st paragraph is removed and consequent changes are made in other paragraphs.

17. Supplementary material should be shortened, and repetition with the main manuscript should be avoided. Thus, for example, the description of both studied zones or the description of the parameters analyzed is included in the main manuscript

Supplementary Material is shortened, repetitions are removed, two figures are moved to the main text (Fig. S1.1 is now Fig. 1e-f and Fig. S1.7 is now Fig. 2g), and the following paragraph from the previous version of the Supplementary Material is inserted in the preface of Sec. 3:
The AI series for both sites show annual cycle, mainly, due to the well established seasonal changes of the <AIneg> (see Fig. 1e) – more scattering aerosols are seen from October to March, due to the seasonal cycles of nitrate aerosols (see e.g. Calvo et al., 2013) and/or other anthropogenic pollutants. During the autumn-winter cold period there is an additional input of soot from the domestic heating and, probably, an increase of the local traffic due to the rainy weather conditions (Pereira et al., 2012, Querol et al., 1998). The <AIpos> shows a tendency to bimodal seasonal variations having higher values in July-August with a second (lower) maximum in February-March (Fig. 1f). This bimodality is in an agreement with the in-situ measurements made in Évora, Portugal (38.5º N, 7.9º W, 300 m a.s.l.) during the 2002-2008 time period (Pereira et al., 2008, 2011). The summer peak is related to the wildfire smokes and intensive SDE events, and the winter maximum is mostly due to the combined effect of local traffic and increased emission from heating sources.

18. Reference list: Sato et al., 1993. Write a point at the end of the reference

Corrected

19. You can realize that the most important studies regarding emissions from wildfires have been carried out by the research team of the University of Aveiro. So, you can also include some references from this team: Evtyugina et al., (2013), Vicente et al. (2012, 2013) Alves et al. (2011).

The references are inserted (see our reply to the comment #2). Thank you very much for pointing out these references.

List of other corrections:

1. In the revised manuscript we use additional terms "urban" for the site ID 082 and "rural" for the site ID 288

2. The following sentences are added now to the conclusion:
   Our results confirm the data from previous studies showing the important role of the anthropogenic pollution, wildfires and SDEs as drivers of the aerosol variation over the Continental Portugal.

3. In the revised version we inserted additional references to the Table 2 for reader's comfort

4. Following text is added to the Section 2.3 Atmospheric parameters (previously Sec. 2.2):
   The comparison of the IGUC and IGIDL series shows that the climatic conditions in Lisbon and Coimbra are quite similar (correlation coefficients in the range from 0.5 to 0.998 with low p-values and meta p-values) but not totally identical. Most important differences were found for the April and August series of precipitation and DTR (correlation coefficients are lower than 0.5). A whole set of correlation coefficients between the IGUC and IGIDL series can be found in the Supplemented Material, Part 1.2.

5. Abstract: The first sentence "Understanding of aerosol sources **which** affect climate is an important problem open in front of scientists as well as policymakers."
   Changed to:
   "*Understanding of aerosol sources **that** affect climate is an important problem open in front of scientists as well as policymakers*."

6. Introduction, 2nd paragraph: The sentence "One of the important **outcomes** of aerosols is their effect on the Earth's radiation budget. Aerosols affect it in two ways: as a direct and an indirect forcing."
   Changed to:
   "*One of the important **impacts** of aerosols is their effect on the Earth's radiation budget. Aerosols affect it in two ways: as a direct and an indirect forcing*."

7. Introduction, 4th paragraph: "**This approach allows us to minimize the effect of the well known spatial heterogeneity of the aerosol content.** Here we take into account a number of different types of aerosols, their local and global sources and their relations with variations of some local climatic parameters: sunshine duration, precipitation, pressure and temperature. The satellite-based TOMS (Total Ozone Mapping Spectrometer) atmospheric aerosol index helps us to obtain information about the aerosol content in the studied region."

Changed to:

"*Here we take into account a number of different types of aerosols, their local and global sources and their relations with variations of some local climatic parameters: sunshine duration, precipitation, pressure and temperature. The satellite-based TOMS (Total Ozone Mapping Spectrometer) atmospheric aerosol index helps us to obtain information about the aerosol content in the studied region.* ***This approach allows us to minimize the effect of the well known spatial heterogeneity of the aerosol content****.*"

8. Last sentence of the Section 2: "Since the data series contain significant gaps (14% of the whole data set length), and measurement time intervals are different for different stations **we used these data with the linearly interpolation of the gaps to calculate a single mean series**."

   Changed to:

   "*Since the data series contain significant gaps (14% of the whole data set length), and measurement time intervals are different for different stations* ***we applied linear interpolation to estimate the missing data and calculated a single mean series****.*"

9. Section 4.1, 2nd paragraph, grammatical correction:

   "In September the correlation coefficients have an opposite sign and **are** *statistically insignificant."*

10. Section 4.2, 2nd paragraph, grammatical correction:

    "*First of all, the biggest correlation coefficients are obtained for the <AIneg> but not for the <AIpos> as for another* ***the other*** *location."*

11. New references are added:
    1. *Alves C., Vicente A., Nunes T., Gonçalves C., Fernandes A.P., Mirante F., Tarelho L., Sanchez de la Campa A., Querol X., Caseiro A., Monteiro C., Evtyugina M., Pio C. (2011) Summer 2009 wildfires in Portugal: emission of trace gases and aerosol composition. Atmospheric Environment. 45, 641-649, 2012.*
    2. *Bližňák, V., Valente, M. A., & Bethke, J. Homogenization of time series from Portugal and its former colonies for the period from the late 19th to the early 21st century. Int. J. Climatol, doi: 10.1002/joc.4151, 2014.*
    3. *Evtyugina M., Calvo A., Nunes T., Alves C., Fernandes P., Tarelho L., Vicente A., Pio C. VOC emissions of smouldering combustion from Mediterranean wildfires in central Portugal. Atmospheric Environment. 64, 339-348, 2013.*
    4. *Morozova, A. L., and M.A. Valente. Homogenization of Portuguese long-term temperature data series: Lisbon, Coimbra and Porto. Earth Syst. Sci. Data, 4, 187-213, 2012.*
    5. *Querol, X., Alastuey, A., Puicercus, J.A., Mantilla, E., Miro, J.V., Lopez-Soler, A., Plana, F., Artiñano, B.: Seasonal evolution of suspended particles around a large coal-fired power station. particulate levels and sources, Atmospheric Environment, 32, 11, 1963-1978, 1998.*
    6. *Stickler, A., Brönnimann, S., Valente, M. A., Bethke, J., Sterin, A., Jourdain, S., Roucaute, E., Vasquez, M.V., Reyes, D.A., R. Allan, R., Dee, D. ERA-CLIM:*

historical surface and upper-air data for future reanalyses. Bull. Amer. Meteorol. Soc., 95(9), 1419-1430, doi: http://dx.doi.org/10.1175/BAMS-D-13-00147.1, 2014.

7. *Vicente A., Alves C., Calvo A.I., Fernandes A.P., Nunes T., Monteiro C., Almeida S.M., Pio C. Emission factors and detailed chemical composition of smoke particles from the 2010 wildfire season. Atmospheric Environment. 71, 295-303, 2013.*
8. *Vicente A., Alves C., Monteiro C., Nunes T., Mirante F., Cerqueira M., Calvo A., Pio C. Organic speciation of aerosols from wildfires in central Portugal during summer 2009. Atmospheric Environment, 57, 186-196, 2012.*

12. A number of stylistic and grammatical corrections unrelated to the Referees' comments were applied.

We thank the Anonymous Referee #2 for his/her positive appreciation of our work and useful comments. Here we provide some replies to his/her comments and suggestions
(please note that the revised text is in italic and specific differences to the original text are in bold):

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1. I recommend the authors to use "urban/regional" or the names of the stations rather than the codes since it is difficult to remember which code denotes which station.

Corrected. Now we use additionally terms "urban" for the site ID 082 and "rural" for the site ID 288.

2. Why are only these two stations used? More stations would represent a better source characterization. If it is a matter of data availability, this should be clearly mentioned in the text.

The choice of the AI measurement locations was justified both by the TOMS data spatial resolution and the availability of the climatic series:
For the AI series: the TOMS data base includes data for only two locations over the Continental Portugal: Lisbon – ID 082, and Penhas Douradas – ID 288.
As to the climatic data, in this study we used the climatic data that (partly) are result of the ERA-CLIM project devoted to the homogenization of historical climatic series. The series of Coimbra (IGUC series) and Lisbon (IGIDL series) are part of this homogenized data set. Since these series are now considered free of non-climatic breaks we decided to use them for our analysis. To fit the quality of already published series we did the homogeneity tests for the rest of the data set.
Now this information is presented in the description of the data sites in the new Sec. 2.1 "Studied locations":
We use the aerosol data over two locations in the Continental Portugal (see Fig. 1a) – ***the only available TOMS aerosol data for this region****. The first one is the site ID 082 over Lisbon (38° 46′ N, 9° 8′ W, 105 m a.s.l.), the second one is the site ID 288 over Penhas Douradas (40° 25′ N, 7° 33′ W, 1380 m a.s.l.). In the first case the region around the site is one of the most urbanized and industrial sites in Portugal where the anthropogenic effects expected to be strong. The second site corresponds to a less populated mountain region affected by the anthropogenic pollution in a lower degree but frequently exposed to forest fire smokes and dust events (Pereira et al., 2005; Pereira et al., 2008; Obregón et al., 2012). Hereafter we use a term "urban" for the site ID 082 and a term "rural" for the site ID 288.*
Consequently, we used climatic data measured by two meteorological observatories that are close to the AI sites. *The first data set belongs the Geophysical Institute of University of Coimbra (hereafter, "IGUC series"). The second set belongs to the Geophysical Institute of Instituto Dom Luiz of University of Lisbon (hereafter, "IGIDL series"). Both locations are shown on the map in Fig. 1a (marked as "Coimbra" and "Lisbon", respectively).*

3. Is there an agreement between the source contributions estimated from this study with earlier studies (modeling, source apportionment)?

It is difficult to make direct comparison between our analysis and other studies because we found no other published work which uses the same or similar methodology to estimate the effect of different aerosol sources on the aerosol content. Besides, the regional differences and temporal variations can affect results of such analyses.

As we mention in the Introduction (previously on p. 31011 l. 19 - p. 31012, l. 4, now slightly enlarged), the previous studies showed dependence of the Portuguese aerosol content on the sources we take into consideration: SDE, pollution and forest fires. The following sentences are added now to the conclusion as well:

Our results confirm the data from previous studies showing the important role of the anthropogenic pollution, wildfires and SDEs as drivers of the aerosol variation over the Continental Portugal.

4. Supplementary material is very long and has to be shortened. Some figures (e.g. S1.1, S1.5, S1.7) and explanations and references can be moved to the main text. There are also overlapping text that should be removed from the supplement. Part 1.3.3 of the supplement can also be moved to the mal text or at least should be summarized as dust is an important source in the area and therefore the detection of the dust event is important.

Supplementary Material is shortened, repetitions to the main text are removed, two figures are moved to the main text (Fig. S1.1 is now Fig. 1e-f and Fig. S1.7 is now Fig. 2g), and the following paragraph from the previous version of the Supplementary Material is inserted in the preface of Sec. 3:

The AI series for both sites show annual cycle, mainly, due to the well established seasonal changes of the <AIneg> (see Fig. 1e) – more scattering aerosols are seen from October to March, due to the seasonal cycles of nitrate aerosols (see e.g. Calvo et al., 2013) and/or other anthropogenic pollutants. During the autumn-winter cold period there is an additional input of soot from the domestic heating and, probably, an increase of the local traffic due to the rainy weather conditions (Pereira et al., 2012, Querol et al., 1998). The <AIpos> shows a tendency to bimodal seasonal variations having higher values in July-August with a second (lower) maximum in February-March (Fig. 1f). This bimodality is in an agreement with the in-situ measurements made in Évora, Portugal (38.5° N, 7.9° W, 300 m a.s.l.) during the 2002-2008 period (Pereira et al., 2008, 2011). The summer peak is related to the wildfire smokes and intensive SDE events, and the winter maximum is mostly due to the combined effect of local traffic and increased emission from heating sources.

Concerning the part of the Supplementary Material related to the definition of the dust events (including Fig. S1.5), we still believe that its transition to the main text is not justified. First, the detection of the SDE is not one of the main objectives of our study. Second, it will, to our mind, unnecessary increase the length of the paper and number of the figures damaging the paper's coherency and readability.

5. Page 10, line 21: Please provide the range of % variation explained by the model rather than the minimum

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Page 10, line 21: Please provide in parenthesis the % contributions of each source discussed.

\+

Page 10, line 25: Please provide the range of % variation explained by the model

Since these three comments are related to the same part of the manuscript (p. 31022 of the ACPD pdf file) and to the same subject, we prepared a single reply:
All the mentioned values (contribution of each of the regressors and per cent of explained variance) are shown in the Table 2. We assume that the comprehension of this set of numbers is much easier in the tabular form where the numbers can be compared at a glance.
As is shown in Table 2, the per cent of explained variance is:
for the annual series - 35% for ID 082 (minimum) and 49% for ID 288,
for the summer series - 88% for ID 082 and 60% for ID 288.

In the revised version we inserted additional references to the Table 2 for reader's comfort and hope that in the final printed version the Table 2 will be relatively close to this part of the text.

6. a) Page 10, lines 29-32: For a typical urban site, traffic can be a very dominant emission source and can be characterized by NOx rather than SO2.

There are several major groups of anthropogenic gaseous pollutants: sulfur dioxide (SO_2), oxides of nitrogen (NOx: NO, NO_2), carbon dioxide (CO_2) etc. Only sulfur dioxide and nitric oxide are primary pollutants that are emitted directly from their sources and could be of interest for our study. The main anthropogenic source for SO_2 is the fossil fuel combustion, and for NOx it is the road transportation. Unfortunately, the absence of reliable (preferably measured) data of NOx for the studied period does not allow us to include information on this pollutant. For the studied period the measured data for NO_2 that are available from the same data base are very fragmented, and can't be used to create a reliable composite series. This situation is discussed in the end of the Sec. 3.1.4. (p. 31020, l. 19-26).

b) Is there any reference for the case in Lisbon (emission studies, modeling etc.)?

There are papers (e.g. Borrego et al., 2003, 2004; Ferreira et al., 2012) that take into account different emissions and models, however their conclusions can not by compared to our results.

1. C. Borrego, O. Tchepel, A.M. Costa, J.H. Amorim, A.I. Miranda. Emission and dispersion modelling of Lisbon air quality at local scale , Atmospheric Environment 37 (2003) 5197–5205
2. C. Borrego , O. Tchepel , L. Salmim , J. H. Amorim , A. M., Costa & J. Janko (2004) Integrated modeling of road traffic emissions: application to lisbon air quality management, Cybernetics and Systems: An International Journal, 35:5-6, 535-548, DOI: 10.1080/0196972049051904
3. Ferreira, F., Gomes, P., Carvalho, A.C., Tente, H., Monjardino, J., Brás, H., Pereira, P. Evaluation of the Implementation of a Low Emission Zone in Lisbon, Journal of Environmental Protection, 2012, 3, 1188-1205, http://dx.doi.org/10.4236/jep.2012.329137 Published Online September 2012 (http://www.SciRP.org/journal/jep)

c) Can NOx be used as a proxy also and explain the remaining variability that is not explained by the model?

Of course, it is possible. If we can add more information (e.g. NOx variability), that means our model includes additional parameter, and that can improve its statistical predictability.

Unfortunately, as was mentioned earlier, we found no reliable measured NOx data for the studied period with at least annual time resolution.

7. Page 11, line 9: It would be valuable to briefly mention these climatic differences in the two sites.

Following text is added to the Section 2.3 Atmospheric parameters (previously Sec. 2.2):
The comparison of the IGUC and IGIDL series shows that the climatic conditions in Lisbon and Coimbra are quite similar (correlation coefficients in the range from 0.5 to 0.998 with low p-values and meta p-values) but not totally identical. Most important differences were found for the April and August series of precipitation and DTR (correlation coefficients are lower than 0.5). A whole set of correlation coefficients between the IGUC and IGIDL series can be found in the Supplemented Material, Part 1.2.

8. Page 2, line 12: Replace ”which” with ”that”

Corrected.

9. Page 2, line 10: Replace “outcomes* with “impacts”

Corrected.

10. Page 3, line 6: “…local aerosol content effecting the variations…”

The sentence is corrected accordingly to comments of both Referees:
The present paper is dedicated to understanding of the ***local and global*** *aerosol sources and* ***the effect of the local aerosol content in climate variations*** *of the Continental Portugal region for the 1978-1993 period.*

11. Page 3, lines 7,8: Move the sentence “This approach: : :” to before the sentence “This information about…” in line 12.

Corrected.

12. Page 6, line 8: Please clarify what FFT refers to.

FFT stays for “Fast Fourier Transform”. The acronym is now explained in the text (new Sec. 2.3 “Atmospheric parameters”).

13. Page 7, line 22: “…we applied linear interpolation to estimate the missing data and calculated a single mean series”

Corrected.

14. Page 11, line 30: “ .. sign and are statistically : : :.”

Corrected.

15. Page 13, line 14 ” as for the other location.”

Corrected.

List of other corrections:

1. A new paragraph inserted in the Sec. 4.1 (after 2nd paragraph there):

   The relations between the temperatures over Iberian Peninsula and SshD during the second half of the 20th c. were earlier reported (see del Rio et al., 2012 and references therein). They were attributed, mainly, to the variations of the circulation patterns over the North Atlantic and consequent changes in the cloudiness. However, accordingly to the data of our analysis, the variations of the SshD can result also from the strong dust intrusions.

   New reference is added to the Reference list:

   del Río, S., Cano-Ortiz, A., Herrero, L., Penas, A.:Recent trends in mean maximum and minimum air temperatures over Spain (1961–2006), Theor. Appl. Climatol., 109, 605–626, DOI 10.1007/s00704-012-0593-2, 2012.

2. Page 31011, Line 26: “The detailed analysis of the properties and time variations of the Portuguese aerosols can be found in Pereira et al. (2005, 2008, 2011, 2012), Santos et al. (2008, 2013), Catry et al. (2009), Calvo et al. (2010), Obregón et al. (2012).”

   Changed to

   The detailed analysis of the properties and time variations of the Portuguese aerosols can be found in Pereira et al. (2005, 2008, 2011, 2012), Santos et al. (2008, 2013), Catry et al. (2009), Obregón et al. (2012), Vicente et al. (2012, 2013), Evtyugina et al. (2013); the analysis of the radiative effect of the aerosols originated from wildfires for the close region in the north-western Spain is presented in Calvo et al. (2010).

3. The TOMS acronym significance is explained now in the Introduction (4th paragraph there), not in the Sec. 2 as previously.

4. The part of the sentence on p. 31014, line 13: “For each of two sites and for each of the months...” is replaced by “*For each site and for each month...*”

5. The sentence on p. 31015, line 15: “Not only spatial and temporal distributions of aerosols are very variable but also their origin as well” is replaced by “*Not only spatial and temporal distributions of aerosols are very variable but also their origin*”.

6. p. 31016, line 13: Bourassa and Robock (2012) is changed to *Bourassa et al., (2012).*

7. Figures captions are corrected and information already indicated in the figures themselves is removed.

8. p. 31020, line 2: “The annual values of the SO_2 content are shown in Fig. 2f. As one can see, there is a strong dependence between the variations of the <AIneg> (shown in Fig. 2b) and the SO_2 content. The anti-correlation between the curves reflects the increase of the scattering particles in the atmosphere (lower <AIneg> values) coinciding with the growth of the measured SO_2 concentration.”

   Changed to

The annual values of the SO_2 content are shown in Fig. 2f. As one can see, there is a strong dependence between the variations of the <AIneg> (shown in Fig. 2b) and the SO_2 content. The anti-correlation (correlation coefficient $r = -0.53$, p value $= 0.06$) between the curves reflects the increase of the scattering particles in the atmosphere (lower <AIneg> values) coinciding with the growth of the measured SO_2 concentration.

9. Preface to the Sec. 4 is rewritten:
   Here we present the analysis of the relations between the aerosol content and the atmospheric parameters described in Section 2. The analysis was done separately for two locations. The analysis of the climatic conditions between the Lisbon and Coimbra (see the Supplementary Material, Part 1.2) showed their strong similarity. This similarity results from the relatively short distance between these locations and their proximity to the ocean. On the other hand, the measured AI monthly means, as was discussed in Sect. 3, are different for these two sites. To our mind, there are two main reasons for these differences: First reason is that the Lisbon area is much more polluted than the region around the rural site (ID 288); second reason is that the more north-eastern position of the site ID 288 provides this location is affected by the dust intrusions more frequently.

10. The reference "IPCC 2013" is changed to "*Climate Change 2013*" and a new reference is added to the Reference List: *Climate Change 2013: The Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, IPCC (Ed.), Cambridge University Press, 2014.*

11. The conclusion is shortened, the 1st paragraph is removed and consequent changes are made in other paragraphs.

12. New references are added:
    1. *Alves C., Vicente A., Nunes T., Gonçalves C., Fernandes A.P., Mirante F., Tarelho L., Sanchez de la Campa A., Querol X., Caseiro A., Monteiro C., Evtyugina M., Pio C. (2011) Summer 2009 wildfires in Portugal: emission of trace gases and aerosol composition. Atmospheric Environment. 45, 641-649, 2012.*
    2. *Bližňák, V., Valente, M. A., & Bethke, J. Homogenization of time series from Portugal and its former colonies for the period from the late 19th to the early 21st century. Int. J. Climatol, doi: 10.1002/joc.4151, 2014.*
    3. *Evtyugina M., Calvo A., Nunes T., Alves C., Fernandes P., Tarelho L., Vicente A., Pio C. VOC emissions of smouldering combustion from Mediterranean wildfires in central Portugal. Atmospheric Environment. 64, 339-348, 2013.*
    4. *Morozova, A. L., and M.A. Valente. Homogenization of Portuguese long-term temperature data series: Lisbon, Coimbra and Porto. Earth Syst. Sci. Data, 4, 187-213, 2012.*
    5. *Querol, X., Alastuey, A., Puicercus, J.A., Mantilla, E., Miro, J.V., Lopez-Soler, A., Plana, F., Artiñano, B.: Seasonal evolution of suspended particles around a large coal-fired power station. particulate levels and sources, Atmospheric Environment, 32, 11, 1963-1978, 1998.*
    6. *Stickler, A., Brönnimann, S., Valente, M. A., Bethke, J., Sterin, A., Jourdain, S., Roucaute, E., Vasquez, M.V., Reyes, D.A., R. Allan, R., Dee, D. ERA-CLIM:*

historical surface and upper-air data for future reanalyses. Bull. Amer. Meteorol. Soc., 95(9), 1419-1430, doi: http://dx.doi.org/10.1175/BAMS-D-13-00147.1, 2014.

7. *Vicente A., Alves C., Calvo A.I., Fernandes A.P., Nunes T., Monteiro C., Almeida S.M., Pio C. Emission factors and detailed chemical composition of smoke particles from the 2010 wildfire season. Atmospheric Environment. 71, 295-303, 2013.*
8. *Vicente A., Alves C., Monteiro C., Nunes T., Mirante F., Cerqueira M., Calvo A., Pio C. Organic speciation of aerosols from wildfires in central Portugal during summer 2009. Atmospheric Environment, 57, 186-196, 2012.*

13. A number of stylistic and grammatical corrections unrelated to the Referees’ comments were applied.

Aerosols over Continental Portugal (1978-1993): their sources and an impact on the regional climate

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Abstract

Understanding of aerosol sources ~~which~~ that affect climate is an important problem open in front of scientists as well as policymakers. The role of aerosols in local climate variability depends on a balance between ~~aerosol~~ light absorbing and scattering particles as well as on variability of environmental conditions. In this paper we investigate variability of aerosol content (both absorbing and scattering ~~UV~~ ultraviolet radiation) over Continental Portugal in dependence on aerosol sources (volcanic eruptions, dust events, wildfires and anthropogenic pollution). The effect of the aerosol on the climate is studied analyzing their contribution to variations of temperature, pressure, sunshine duration and precipitation over Portuguese regions. The present analysis is based on a developed modern multiple regression technique allowing us to build the statistical correlation models to determine both the main local aerosol sources and aerosol's influence on the climate of ~~the~~ Continental Portugal during 1978-1993 time period. The analysis allows us to conclude that the main sources driving the variations of the aerosol content over studied locations are wildfires, mineral dust intrusions and anthropogenic pollution. The relations between the aerosol content variations and the atmospheric parameters depend on the level of urbanization of the studied region, the type of aerosol and the season. The most significant finding is the decrease of the daily maximum temperature (and diurnal temperature range) related to the decrease of sunshine duration

observed during the summer periods of increased content of the absorbing aerosols in the atmosphere.

1 Introduction

Aerosol particles scatter and absorb solar and terrestrial radiations depending on their microphysical and optical characteristics. These characteristics vary significantly due to various aerosol sources, both anthropogenic and natural. Natural sources include volcanic emissions, plant vapors and chemicals released by tiny sea creatures, as well as dust from deserts. From the beginning of the industrial period anthropogenic sources include not only farming and charcoal burning but also emissions from car exhausts, factories and power plants. Some aerosols like dust particles or sea spray are mostly from natural origin. The other aerosols like sulfates and black carbon come from both natural and anthropogenic sources.

One of the important ~~outcomes~~ impacts of aerosols is their effect on the Earth's radiation budget. Aerosols affect it in two ways: as a direct and an indirect forcing. The direct aerosol radiative forcing is due to changes in solar irradiance and the indirect one is through aerosol effects on clouds. The report of the Intergovernmental Panel on Climate Change (IPCC, ~~ClimateonIPCC 2013Change 2013~~ Climate Change 2013, Ch. 7 “Clouds and Aerosols”) indicates that aerosols give overall a cooling effect on the Earth since pre-industrial times. This effect masked some of the global warming from greenhouse gases that would have occurred in the aerosols absence. Aerosols affect not only global climate, but also, due to unevenly spatial distribution, local weather and climate, visibility and human health. The local aerosol influence on the regional weather conditions can ~~have~~ be much stronger ~~effect on the regional weather conditions~~ and lead rather to local climate changes than to global. Normally, aerosol scattering makes the Earth’s atmosphere more reflective and lead to cooling of the climate system, while aerosol absorption has the opposite effect and lead to warming. The balance between cooling and warming depends on microphysical and optical properties of aerosols.

Most of the previously published papers are dedicated to the analysis of the aerosol content variations over Portugal for relatively short time periods. These periods start mainly in 2002 when the facilities allowing the in-situ measurements of many aerosol parameters were established at the Évora Geophysics Centre Observatory, (38.57N, 7.91W, 293 m a.s.l) – see e.g. Pereira et al. (2011, 2012). These studies showed the strong dependence of the local aerosol content and its composition both on anthropogenic and natural sources. The latter include mineral dust intrusions and wildfires. The detailed analysis of the properties and time variations of the Portuguese aerosols can be found in Pereira et al. (2005, 2008, 2011, 2012), Santos et al. (2008, 2013), Catry et al. (2009)~~, Calvo et al. (2010)~~, Alves et al. (2012), Obregón et al. (2012), Vicente et al. (2012, 2013), Evtyugina et al. (2013); the analysis of the radiative effect of the aerosols originated from wildfires for the close region in the north-western Spain is presented in Calvo et al. (2010). The impact of the aerosol variations on the local climate variations was studied as well, both with in-situ measurements (e.g. Santos et al., 2008; Obregón et al., 2012) and modeling (e.g. Miranda et al., 2002; Santos et al., 2013). For example, a noticeable cooling at the surface level was observed due to aerosols consisting of desert mineral dust and forest fire products (Santos et al., 2008; Calvo et al., 2010).

The present paper is dedicated to understanding of the local and global aerosol sources and the ~~role~~ effect of the local aerosol content ~~played~~ in ~~variations of the~~ climate variations of the Continental Portugal region for the 1978-1993 time period. ~~This approach allows us to minimize the effect of the well known spatial heterogeneity of the aerosol content.~~ Here we take into account a number of different types of aerosols, their local and global sources and their relations with variations of some local climatic parameters: sunshine duration, precipitation, pressure and temperature. The satellite-based TOMS (Total Ozone Mapping Spectrometer) atmospheric aerosol index helps us to obtain information about the aerosol content in the studied region. This approach allows us to minimize the effect of the well known spatial heterogeneity of the aerosol content. The information about climatic parameters variability is received from the Geophysical Institute of University of Coimbra and the Geophysical Institute of Instituto Dom Luiz of University of Lisbon. As aerosol sources we considered volcanic eruptions, Saharan dust, forest fires and anthropogenic pollution. The length of the studied period (about 15 years) allows us to analyze long term variations of both aerosol content and resulting climatic effects. The use of two different locations helps us to estimate the differences in the aerosol variations and their climatic consequences between the relatively clean and the industrially polluted regions. The correlation analysis and the multiple regression technique used in our study allow us to build statistical correlation models (1) to specify the main local aerosol sources and their input into the variations of the local aerosol content; (2) to study aerosols' influence on some local climatic parameters. Concerning the data sources, the preference was given to directly measured data series that have sufficient quality, statistical homogeneity and temporal resolutions, and are available for the whole studied period. In some cases the available measured data series can be considered as well as proxies for those parameters which lack direct measurement during the studied period (such are the cases of SO_2 series used as a proxy for other anthropogenic pollutants or sunshine duration series used as a proxy for cloud amount).

The paper is organized as follows: Section 1 presents modern state of the art and briefly gives overview of the paper. Section 2 contains the description of the used data sets as well as their statistical properties. Section 3 describes variations of aerosol content over ~~continental~~ Continental Portugal during 1978-1993 years and their main sources. In Section 4 we show how aerosol variation affect local climate during the studied period. Section 5 gives summary on the obtained results. Here in Section 2 we present only a short description of some data sets used in the study. The detailed analysis of ~~all~~ these parameters can be found in the ~~Supplemented~~Supplementary Material. Part1 of the ~~Supplemented~~Supplementary Material presents~~ the detailed description of the data sets used in the paper:~~ detailed description of the aerosol data (Part 1.1), comparison of atmospheric parameters from different meteorological stations (Part 1.2) and ~~aerosol forcings (Part 1.3). Part 1.3.3 of the Supplementedy Material contains~~ a description of the method used to detect dust events ~~and~~ (Part 1.3).~~4 presents a short analysis of the similarity in the trends of aerosols and anthropogenic sulfates variations.~~ Part 2 of the ~~Supplemented~~Supplementary Material presents the correlation analysis between the sunshine duration series and other meteorological parameters.

2 Data sets

2.1 Studied locations

We used the aerosol data over two locations of~~in the~~ Continental Portugal (see Fig. 1a) – the only available TOMS aerosol data for this region. The first one is the site ID 082 over Lisbon (38° 46′ N, 9° 8′ W, 105 m a.s.l.), the second one is the site ID 288 over Penhas Douradas (40° 25′ N, 7° 33′ W, 1380 m a.s.l.). In the first case the region around the site is one of the most urbanized and industrial sites in Portugal where the anthropogenic effects expected to be strong. The second site corresponds to a less populated mountain region affected by the anthropogenic pollution in a lower degree but frequently exposed to forest fire smokes and dust events (Pereira et al., 2005; Pereira et al., 2008; Obregón et al., 2012). Hereafter we use a term “urban” for the site ID 082 and a term “rural” for the site ID 288.

Consequently, we used climatic data measured by two meteorological observatories that are close to the AI sites. The first data set belongs to the Geophysical Institute of University of Coimbra (hereafter, “IGUC series”). The second set belongs to the Geophysical Institute of Instituto Dom Luiz of University of Lisbon (hereafter, “IGIDL series”). Both locations are shown on the map in Fig. 1a (marked as "Coimbra" and "Lisbon", respectively).

~~2.1~~2.2 Aerosol parameters

The ~~Total Ozone Mapping Spectrometer (~~TOMS~~)~~ Aerosol Index (AI) data (*http://disc.sci.gsfc.nasa.gov/acdisc/TOMS*) from 01 November 1978 to 06 May 1993 were used to study the variations of the aerosol content over Portugal. Under most conditions the AI is positive for the ~~UV~~ ultraviolet absorbing aerosols (pure absorption) and negative for the ~~UV~~ ultraviolet non-absorbing aerosols (pure scattering) when two close wavelengths in ~~UV~~ ultraviolet region near 360 nm are considered (see e.g. Ginouz and Torres, 2003 and http://visibleearth.nasa.gov/view.php?id=1043). The TOMS aerosol index is calculated in a way that allowed us to separate days with prevailing of absorbing (e.g. mineral dust, smoke, volcanic ash) or scattering (e.g. sea-salt aerosols in the regions relatively close to ocean and sulfate aerosols in urban areas) particles (Herman et al., 1997; Torres et al., 1998). More details about the AI calculations can be found in the Supplementary Material, Part 1.1. ~~We use the data on AI variations over two locations in the Continental Portugal (see Fig. 1a). The first one is the site ID 082 over Lisbon (38° 46′ N, 9° 8′ W, 105 m a.s.l.), the second one is the site ID 288 over Penhas Douradas (40° 25′ N, 7° 33′ W, 1380 m a.s.l.). In the first case the region around the site is one of the most urbanized and industrial sites in Portugal where the anthropogenic effects expected to be strong. The second site corresponds to a less populated mountain region affected by the anthropogenic pollution in a lower degree but frequently exposed to forest fire smokes and dust events (Pereira et al., 2005; Pereira et al., 2008; Obregón et al., 2012).~~ In this study we used daily data only in one case: to indentify days with Saharan Dust Events (SDE) – see Sec. 2.4~~3~~. For other purposes the monthly, seasonal and annual means of AI (and ~~all~~ other parameters) have been calculated. For each site~~of two s~~ and for each month~~of the s~~ we calculated three series: <AI>, <AIpos> and <AIneg> taking into consideration all, only absorbing or only scattering aerosols, respectively. The seasonal and annual mean series were calculated using corresponding monthly mean series. In the paper these mean series are referenced as monthly, seasonal and annual series, correspondingly. Variations of these three indices for both sites are shown in Fig. 1b-d.

~~2.2~~2.3 Atmospheric parameters

~~Two sets of climatic data were used in this study. First one contains parameters measured by the Geophysical Institute of University of Coimbra (hereafter, “IGUC series”). Second one consists of the series measured by the Geophysical Institute of Instituto Dom Luiz of University of Lisbon (hereafter, “IGIDL series”). Both locations are shown on the map in Fig. 1a.~~ The sets of climatic parameters used in this study include monthly and annual means of the following daily variables

1. minimum (*Tmin*), maximum (*Tmax*) and average (*averT*) temperatures, and ~~daily~~ temperature range (*DTR*);
2. accumulated precipitation amount (*precip*);
3. mean atmospheric pressure at station level (*p*);
4. sunshine duration (*SshD*).

The temperature and pressure series are part of the historical data set recently presented to the scientific community after the homogenization procedure done in the frame of the ~~of the~~ FP7 project ERA-CLIM (Morozova and Valente, 2012; Bližňák et al., 2014; Stickler et al., 2014). Other series are still under analysis~~(see also short description in the Supplementary Material, Part 1.2).~~ The relations between aerosol variations and atmospheric conditions in the region under consideration (see Sec. 4) were studied separately for two sites: the IGIDL series were used in pair with AI data from the urban~~site ID 082~~ site (ID 082), and the IGUC series were used in pair with the AI series from the rural~~site ID 288~~ site (ID 288). The distance between the aerosol detection site and the meteorological station is about 5.5 km in case of the site ID 082 and about 74 km in case of the site ID 288. In the second case the distance between the places of measurements of the aerosols and climatic parameters is quite large. Nevertheless, we used the IGUC series because the other data sets available for this region are of insufficient quality and time resolution (see also a discussion in the

~~Supplemented~~Supplementary Material, Part 1.2). The comparison of the IGUC and IGIDL series shows that the climatic conditions in Lisbon and Coimbra are quite similar (correlation coefficients in the range from 0.5 to 0.998 with low *p* values and meta *p* values) but not totally identical. Most important differences were found for the April and August series of the precipitation and DTR (correlation coefficients are lower than 0.5). A whole set of correlation coefficients between the IGUC and IGIDL series can be found in the Supplementary Material, Part 1.2.

Please also note that all significances (*p* values and meta *p* values) for correlation coefficients presented in this paper are calculated using 10,000 of the Monte-Carlo simulations with the random-phase Fast Fourier Transform as a randomizing procedure (Ebisuzaki, 1997). *P* value shows the probability for any specific correlation coefficient of the singular comparison to be obtained by chance. In cases when 12 separate monthly plus an annual series were analyzed simultaneously, the multiple comparisons significances (meta *p* values) were calculated as well.

~~2.3~~2.4 Aerosol sources

Not only spatial and temporal distributions of aerosols are very variable but also their origin ~~as well~~. In this study we take into consideration only main sources responsible for the aerosol content variations over the Continental ~~Portuguese~~ Portugal region. These sources are mineral dust from Sahara and Sahel regions, wildfires (Alves et al., 2012; Vicente et al., 2012, 2013; Evtyugina et al., 2013), anthropogenic pollution (Pereira et al. 2005, 2008, 2009, 2011, 2012; Santos et al., 2008; Calvo et al., 2010) and volcanic aerosols. Some other locally important aerosol sources (like sea-salt aerosols or anthropogenic aerosols other~~s~~ than SO_2) remain~~s~~ outside the frames of our study due to the absence of reliable (preferably measured) data on their variations for the studied period. Nevertheless, the regression models discussed in Sec. 3.2 (see also Table 2) show that even this limited set of aerosol sources allows us to reconstruct the aerosol content variations with a good accuracy.

~~Please also note that all significances (*p* value) for correlation coefficients presented in this paper are calculated using 10,000 of the Monte-Carlo simulations with the random-phase FFT as a randomizing procedure (Ebisuzaki, 1997). *P* value shows the probability for any specific correlation coefficient of the singular comparison to be obtained by chance. In cases when 12~~

~~separate monthly plus an annual series were analyzed simultaneously, the multiple comparisons significances (meta *p* value) were calculated as well.~~

Volcanoes. In this study we use the GISS climate simulation (http://data.giss.nasa.gov/modelforce/strataer) data on variability of the stratospheric aerosol optical thickness (AOT) at 550 nm for Northern Hemisphere as a proxy for the volcanic aerosol content changes. This data set has monthly resolution from October 1850 to December 2010. The data set is described in Sato et al. (1993) and Bourassa ~~and Robock~~et al. (2012). During the studied period four volcanic eruptions with the volcanic emissivity index VEI > 4 took places (numbered in Fig. 2c).

1. 1980 March – Mt. St. Helens, tropospheric eruption
2. 1982 March-April – El Chichon, stratospheric eruption
3. 1991 June & August – Pinatubo, stratospheric eruption
4. 1991 August-October – Mt. Hudson, stratospheric eruption

These eruptions, except the first one, deposited a significant load of sulfate aerosols to stratosphere over the globe. However, during the years following the eruptions the amount of the absorbing particles in a zone around 40° N latitude did not increased as dramatically as in regions around the equator (see e.g. Fig. 3 in Torres et al., 2002).

Saharan Dust Events (SDE). Saharan dust events are well known sources of the dust in the Mediterranean region (Pereira et al., 2008; Obregón et al., 2012). The maximum number of the SDE in the western Mediterranean is observed in summer period, especially in July-August (Moulin et al., 1998; Fig. 4 in Torres et al., 2002; Rogora et al., 2004; Fiol et al., 2005). These events are characterized by the high amount of the absorbing dust particles in the atmosphere coming from the Sahara and Sahel regions. In this work we identified SDE days using the method fully described in Barkan et al. (2005) and Varga et al. (2013). The main idea is to select days when standardized AI ~~are~~ is higher than a threshold value (see Supplementary Material for a brief description of the method and comparison to other published data~~)~~. The short analysis of the dust events frequency for both AI sites is also presented in the ~~Supplemented~~Supplementary Material, ~~(~~Part 1.3~~.2~~). The variations of the monthly mean <AIpos> index averaged over two Portuguese locations increase together with the total monthly number of dust events ~~(as shown in the Supplementedary Material)~~. <AIneg> variations on contrary have no relations to the SDEs.

Forest fires. Among all southern European countries Portugal shows the highest density of wildfire ignitions (Catry et al., 2009). Almost all fires occur in summer months (from June to September) due to the dry and hot weather that is common for the region at this time of a year (Pereira et al., 2008; Obregón et al., 2012). Portuguese Institute for the Conservation of the Nature and Forests (Instituto da Conservação da Natureza e das Florestas, ICNF, http://www.icnf.pt) provides the data on the number of fire occurrences and a total burned area (BA), organized by districts, from 1980 to 2011. In this study we used only the BA data because the fire occurrence series seem to be less reliable (Pereira et al., 2005). The BA data from the Coimbra, Guarda and Castelo Branco districts (marked by numbers 1-3 on the map in Fig. 1a) were used to compare with the AI series from the ~~site ID 288~~rural site (ID 288), and the BA data from the Santarém, Lisboa and Setúbal districts (marked by numbers 4-6 on the map in Fig. 1a) were used to compare with the AI series from the ~~site ID 082~~urban site (ID 082).

Pollution. Anthropogenic aerosols affect the radiation balance in the atmosphere both through the absorption and the scattering processes (Wang, 2013). We assume that the actual measurements of the air composition give more precise information about the aerosol content than the estimated production of the anthropogenic sulfates and/or nitrates. Therefore, in this work we used the data from the European Monitoring and Evaluation Programme (EMEP) database (http://www.emep.int), specifically EBAS database (http://ebas.nilu.no) which contains the monthly mean values of SO_2 (in μg S/m^3) for the five Portuguese stations for the period from August 1979 to December 2009:

1. Braganca (41° 49′ N, 6° 46′ W, 690 m a.s.l.)
2. Viana do Castelo (41° 42′ N, 8° 48′ W, 16 m a.s.l.)
3. Monte Velho (38° 05′ N, 8° 48′ W, 43 m a.s.l.)
4. Foia (37° 19′ N, 8° 54′ W, 902 m a.s.l.)
5. Faro (37° 01′ N, 7° 58′ W, 8 m a.s.l.).

~~. These stations are listed in the Supplemented Material (Part 1.3.4).~~ Since the data series contain significant gaps (14% of the whole data set length), and measurement time intervals are different for different stations we applied linear interpolation to estimate the missing data and calculated a single mean series.~~we used these data with the linearly interpolation of the gaps to calculate a single mean series~~

3 Variations of aerosol content and their sources

The <AIpos> and <AIneg> series (Fig. 1c-d) for the same sites do not correlate with each other: the correlations coefficients between the <AIpos> and <AIneg> monthly series are 0.14 (*p* value = 0.05) for the site ID 082 and 0.22 (*p* value < 0.001) for the site ID 288. The spatial correlation for all the types of aerosol indices is more or less strong, which is quite expected for the sites at a distance of about 200 km apart (see Fig. 1b-d). The correlation coefficients for the separate monthly and annual series are ~~also~~ shown the ~~Supplemented~~Supplementary Material. The analysis of the standard statistical parameters of the <AIpos> and ~~/~~<AIneg> series shows that the absorbing aerosols play more significant role over the ~~site ID 288~~rural site (ID 288) than over the ~~site ID 082~~urban site (ID 082). The <AIpos> series ~~for the site ID 288~~ has higher values of the mean, standard deviation and maximum values in case of the site ID 288 than ~~the similar series for~~in case of the site ID 082. On contrary, the same statistical parameters for the <AIneg> series are practically equal for these two sites.

The AI series for both sites show annual cycle~~,~~ mainly~~,~~ due to the well established seasonal changes of the <AIneg> (see Fig. 1e) – more scattering aerosols are seen from October to March~~,~~ due to the seasonal cycles of nitrate aerosols (see e.g. Calvo et al., 2013) and/or other anthropogenic pollutants. During the autumn-winter cold period there is an additional input of soot from the domestic heating and, probably, an increase of the local traffic due to the rainy weather conditions (Pereira et al., 2012, Querol et al., 1998). The <AIpos> shows a tendency to bimodal seasonal variations having higher values in July-August with a second (lower) maximum in February-March (Fig. 1f). This bimodality is in an agreement with the in-situ measurements made in Évora, Portugal (38.5º N, 7.9º W, 300 m a.s.l.) during the 2002-2008 time period (Pereira et al., 2008, 2011). The summer peak is related to the wildfire smokes and intensive SDE events, and the winter maximum is mostly due to the combined effect of local traffic and increased emission from heating sources.

3.1 Main aerosol sources

~~In this study we tried to take into account a number of main aerosol sources identifying their effect on the observed AI variations. These sources are volcanic eruptions, dust from Sahara~~

~~desert, smoke and soot from the forest fires, and anthropogenic pollutions (sulfate aerosols series is considered as a proxy for most of the anthropogenic pollutant).~~

Volcanoes. The annual variations of the <AIpos> measured over two Portuguese sites (Fig. 2a) show some increase after the eruptions of 1982 and 1991 (Fig. 2c), but these peaks could also be related (at least, partly) to other phenomena, like e.g. Saharan dust events. On the other hand the correlation analysis (see Table 1) shows (1) a weak but statistically significant dependence of the annual series of the <AI> and <AIpos> on the AOT variations and (2) no dependence between the <AIneg> and AOT annual series.

Saharan Dust Events (SDE). Since most of the SDEs take place in summer we compared not only annual values of aerosol indices and SDE number but also values calculated for the local summer season (from June to September). The correlation coefficients presented in Table 1 ~~(~~as well as comparison of Figs. 2a-b and 2d~~)~~ clearly show that the high values of the <AIpos> in 1982-1983 (at least, partly) and in 1988 are caused by the intensive Saharan dust intrusions. The <AIneg> series show no connection to the SDEs, as it has to be expected.

Forest fires. Figure 2e shows variations of the total burned area for the both groups of districts. The correlation coefficients between the <AI> and BA series (Table 1) are quite low. The reason, probably, is the stronger effect of other forcings (like SDEs) on the aerosol content variations. However, the multiple regression models, which will be discussed later in Section 3.2, detect the “forest fire” forcing as a regressor required to explain the AI series variations.

Pollution. The annual values of the SO_2 content are shown in Fig. 2f. As one can see, there is a strong dependence between the variations of the <AIneg> (shown in Fig. 2b) and the SO_2 content. The anti-correlation (correlation coefficient $r = -0.53$, p value $= 0.06$) between the curves reflects the increase of the scattering particles in the atmosphere (lower <AIneg> values) coinciding with the growth of the measured SO_2 concentration. Unsurprisingly, the <AIneg> variations over a highly populated location (ID 082 – Lisbon) show stronger dependence on the SO_2 content (see Table 1). The correlations between the <AIneg> and SO_2 variations became even much stronger when trends of these two parameters are studied. For example, the comparison of the monthly series of the SO_2 and the <AIneg> smoothed by the running averaging procedure (window of 36 months) shows that the satellite measured

<AIneg> series follows the ground measured sulfate content data with probably a lag of about 5-10 months – see Fig. 2g~~. (see Supplemented Materials, Part 1.3.4)~~.

Relatively high correlations between the <AIpos> over the ~~site ID 082~~urban site (ID 082) and the SO_2 values (Table 1) probably caused by the similarities in the variations of different pollution gases/aerosols. Since the anthropogenic sulfates are almost totally scattering aerosols, they can not affect the satellite-measured <AIpos> values. However the amount of other types of aerosols (like light absorbing black carbon) can follow the changes of the SO_2 content due to the same source of origin (e.g. fossil fuel combustion). Unfortunately, we found no measurements of other anthropogenic aerosols/gases for the studied period with an accepted time resolution and data quality to confirm this suggestion. On the other side, since the pollutants of different types are originated from the same sources (like traffic, coal and biomass burning, industrial activities etc., see e.g. Calvo et al., 2013) their temporal variations are more or less similar, and the SO_2 series can be considered in the frame of our study and to a certain degree as a proxy for most of anthropogenic pollutants.

3.2 Multiple regression models of aerosol variations

The analysis of the individual correlations between the AI and a number of natural and anthropogenic aerosol forcings allowed us to find the main sources of the aerosol content variations for this region. Those forcings are the Saharan dust events, the wildfires, the anthropogenic pollution and the volcanic eruptions. Some of these forcings affect both the absorbing and the scattering aerosols (e.g. anthropogenic pollution and forest fires). Other forcings influence only the absorbing part of the aerosol content (e.g. SDE). Linear multiple regression models (MRM) have been constructed to statistically connect the observed variations of the <AI>, <AIpos> and <AIneg> due to the changes of the above mentioned forcings.

The models were constructed using a “best subset” technique that finds a subset of regressors (aerosol forcings, in our case) that predict as much of the variations of the dependent parameter (AI, in our case) as possible. The quality of the MRM is defined by r and $r_{adj.}^2$ parameters. The first one is a correlation coefficient between the modeled and the original series, and its square multiplied by 100 defines the percent of explained variations. The second parameter is the so called “adjusted r^2”. The adjustment is done using differences between the model and the original data comparing to the original data variance and taking into account the number of degrees of freedom. The $r_{adj.}^{2}$ was used as a criterion to compare the MRMs with different subsets of regressors: the subset that gives a bigger $r_{adj.}^{2}$ value is the "best subset". The role of each of the regressors is estimated by a β coefficient that quantifies how strongly each regressor influences the dependent variable. The β is measured in units of standard deviation σ: ~~The~~ the regressors with highest (absolute) β values have greater impact on the dependent parameter. All parameters for the different MRMs are shown in Table 2 for the annual and summer (June-September) AI series. The MRMs for the annual and summer AI series together with the corresponding original AI data are also presented in Fig. 3. All discussed above forcings are used as regressors for the <AIpos> and <AI> series, and only the wildfires and the pollutions are used to model the <AIneg> variations.

The obtained results prove that chosen forcings represent a good set of regressors to explain the <AIpos> variations (Fig. 3b, e, h, k). The correlation coefficients between the MRMs and the original AI series are greater than 0.7 (see fourth column of Table 2), and the models explain (taking into account the number of degrees of freedom) at least 35% of the variations of the original <AIpos> series (see sixth column of Table 2). Unsurprisingly, Saharan dust events have a greatest contribution (columns ~~seventh to tenth~~eighth of Table 2) to the summer <AIpos> variations for both locations and the SO_2 content is an important regressor for the <AIpos> series measured above the more populated area (ID 082) – see tenth column of Table 2.

Concerning the <AIneg> series (Fig. 3c, f, i, l), it is clear that the used regressors can not sufficiently explain observed variations of the scattering aerosols, especially when the number of degrees of freedom is taken into account. However, the MRMs for the <AIneg> show similar to the original series trends: these trends follow the growth of the pollutant (SO_2 content in our case). The discrepancies between the models and the observations can result from the absence in the list of the MRM regressors of some important aerosol sources like, for example, sea-salts or others than SO_2 pollutants (e.g. NOx). Unfortunately, there is no reliable corresponding data series that can be used in the frame of our study.

Finally, the MRMs for the <AI> series (Fig. 3a, d, g, j) are well correlated with the original series for both locations and explain 33-55% of the <AI> variations. These results show that the SDEs, the wildfires and possibly the volcanic eruptions significantly affect the aerosol content over the low-populated location (ID 288), while the anthropogenic pollution plays an important role in the variations of the AI over the high-populated location (ID 082).

4 Regional climate variations in relation to aerosol content changes

Here we present the analysis of the relations between the aerosol content and the atmospheric parameters described in Section 2. The analysis was done separately for two locations. ~~For the comparison of the climatic conditions in Lisbon and Coimbra see the Supplemented Material, Part 1.2.~~ The analysis of the climatic conditions between the Lisbon and Coimbra (see the Supplementary Material, Part 1.2) showed their strong ~~On contrary to the~~ similarity. This similarity results from the relatively short distance between these locations and their proximity to the ocean.~~ of the climatic conditions for the studied locations~~ On the other hand, the measured AI monthly means, as was discussed in Sect. 3, are different for these two sites.~~, as was discussed in Sect. 3.~~ To our mind, there are two main reasons for these differences~~:~~. First ~~reason is that~~of all,~~this is a result of the different pollution and circulation conditions~~ the Lisbon area is much more polluted than the region around the rural site (ID 288).)~~ over the sites; second~~ Secondly, ~~reason is that~~ the more north-eastern position of the rural site ID 288 provides ~~this~~ the stronger and more frequent effect ~~location is affected by~~of the dust intrusions ~~more frequently~~.

4.1 ~~Site~~ Rural site ID 288

The results of correlation analysis for the pair “AI ID 288 vs IGUC series” are shown in Fig. 4 (b, d, f). As one can see, the relations between the climatic parameters and the aerosols of absorbing (<AIpos>) and scattering (<AIneg>) types strongly depend on the season. The effect of the absorbing aerosols is more prominent during the summer-autumn (Fig. 4d), but the relation between the climatic parameters and the scattering aerosols are stronger during the first half of a year (Fig. 4f). It has to be mentioned that the summer-early autumn period of a year is a dry season in ~~the~~ Continental Portugal, whereas late autumn, winter and spring seasons often are wet because of the influence of the North Atlantic cyclones~~.~~ (Miranda et al., 2002).

First we examine relations between the <AIpos> and the climatic parameters. One of the most important features is the anti-correlation between the <AIpos> and parameters like the *SshD*, *Tmax* and *DTR* during July. Similar relations take also places in June, August and October-November but the magnitude and the statistical significances of correlation coefficients *r* are smaller. This cooling trend coincides with epochs of frequent SDE events (high <AIpos> values) and is in an agreement both with most recent/~~-~~precise measurement (Santos et al., 2008) and with modeling studies (Santos et al., 2013). In September the correlation coefficients have an opposite sign and are statistically insignificant. As it is shown in Fig. 4d, the increase of the absorbing aerosol amount is accompanied by the decrease of the sunshine duration. The decrease of the *SshD* leads to the decrease of the amount of the solar radiation reaching the ground, which in turn affects *Tmax* (a parameter that can be considered as a measure of the day-time temperature): *Tmax* is decreasing. Consequently, the decrease of the *Tmax* affects the daily temperature ranges: the *DTR* decreases also.

The relations between the temperatures over Iberian Peninsula and *SshD* during the second half of the 20th c. were earlier reported (see del Rio et al., 2012 and references therein). They were attributed, mainly, to the variations of the circulation patterns over the North Atlantic and consequent changes in the cloudiness. However, accordingly to the data of our analysis, the variations of the *SshD* can result also from the strong dust intrusions.

This kind of relations between the *SshD* and temperatures usually is related to the cloud effect on the radiation distribution in the lower atmosphere (Climate C~~c~~hange 2007; Climate;~~,~~ ~~IPCC 2013~~Climate Change 2013, Ch. 7). ~~As it is shown in the Supplemented~~ary ~~Materials (Part 2), t~~The *Tmax* and *DTR* correlate very well with the *SshD* during almost a whole year (the whole set of correlation coefficients can be found in the Supplementary Material, Part 2). The precipitation amount anti-correlates with *SshD*, therefore, this can be considered as a confirmation of the existence of the clouds that block solar irradiance. However, there is a possibility that such relations between the *SshD* and <AIpos> during the dry summer period, at least partly, are due to the direct aerosol effect. We assume that the change of the radiation balance is also a reason for the correlation between the amount of the absorbing aerosols and the *Tmin* (a parameter that can be considered as a measure of the night-time temperature) found for the February series. The aerosol particles may play a role of high-level clouds reflecting some of the outgoing~~ IR ~~ infrared radiation back to the ground. The relation between the precipitation amount and the <AIpos> found for this site reflects also a process that can be identified as an indirect effect of the aerosols on the cloud formation (see e.g. Climate ~~change~~ Change 2007~~ (IPCC AR4)~~, Ch. 7.5 "Aerosol Particles and the Climate System"). The increase of the <AIpos> coincides with the higher amount of the precipitation (Fig. 4d). This effect is more pronounced in April, June and November-December. The aerosol particles may act as seeds for the cloud droplet in a relatively dry summer (and sometimes winter) air.

The effect of the scattering aerosols (described by the <AIneg> index) on some of the atmospheric parameters in the region is similar to the observed for the <AIpos>. The increase of the aerosol loading coincides with the decrease of the *SshD*, *DTR*, *Tmax* and *averT* in July (and in a weaker form in June and August) – Fig. 4f. The opposite relations take place in early spring season (February-April) when <AIneg> variations correlate with changes of the *Tmin*, *Tmax*, *averT* (and *DTR* in April). On the other hand, the precipitation has an opposite dependence on the <AIneg> variation compared with obtained for the <AIpos>. The precipitation amount decreases when the scattering aerosol loading in the atmosphere increases. This effect can be related to decrease of the cloud droplet size in the polluted air which increases the cloud lifetime and decrease precipitation (see e.g. Ch. 7.5 in Climate ~~change~~ Change 2007). The only exception is January: during this month the <AIneg> is correlated with the precipitation.

4.2 Urban s~~S~~ite ID 082

The results of correlation analysis for the pair "AI ID 082 vs IGIDL series" are shown in Fig. 4(a, c, e). As one can see there are significant differences in the relation between the variations of the AI and climatic parameters over Lisbon comparing to the Coimbra/Penhas Douradas region. The differences between these two sites can result from different aerosol sources in the more polluted Lisbon area. Besides, the Coimbra/Penhas Douradas area is stronger affected by the absorbing aerosols originated from wildfires and Saharan dust. As it was mentioned in Choobari et al. (2014), the combination of dust and soot particles increase absorption properties of the aerosols whereas the combination of dust and sulfates from pollution decrease absorption properties.

First of all, the biggest correlation coefficients are obtained for the <AIneg> but not for the <AIpos> as for ~~another~~ the other location. As one can see from the comparison of the Figs. 4(a, c, e) and 4(b, d, f), the similarities in the relations between the AI indices and the climatic parameters exist, mostly, for temperature parameters and <AIpos> during January-March, June and August-September periods. The significant seasonal differences are seen only in the <AIneg> variations. All temperature parameters tend to anti-correlate with <AIneg> amount in April and November (months of the transient seasons). On contrary for the May, August and September months (hot dry season) there is a tendency to correlation between the <AIneg> values and the temperatures. As a rule, temperature parameters tend to correlate with the AI (the more aerosol particles of both types, the higher the temperature) with just a number of exceptions (June for <AIpos>, and April and November for <AIneg>). The relations between the *SshD* and aerosol content are weak and sporadic. There is just a small number of significant anti-correlations between the <AIpos> and <AIneg> and the *SshD* series. These are June series for <AIpos> (Fig. 4c) and May series for <AIneg> (Fig. 4e). In the first case the relations are similar to ones obtain for the Coimbra/Penhas Douradas site: the increased amount of the aerosol loading coincides with shorter periods of sunshine duration.

4.3 Multiple regression models of sunshine duration variations

To study further the role played by the aerosols in the climatic variations of ~~the studied~~this region we constructed the multiple regression models that explain sunshine duration variations depending on the following parameters: precipitation and pressure (proxies for the cloud amount/clear sky conditions), and <AIpos> and <AIneg>. The choice of the parameters is defined by their high correlation coefficients with the *SshD* series (see e.g. Figs. 4c-f and the ~~Supplemented~~Supplementary Materials, Part 2). The MRMs are calculated separately for both sites for the monthly and annual means using the “best subset” technique and parameters described in Section 3.2. The results are shown in Figs. 5a-f.

Altogether, the selected regressors allowed us to construct quite good regression models for the *SshD* series. Figures 5a and 5b show examples of the MRM predictions (for the annual *SshD* series for IGIDL and IGUC, correspondingly). The correlation coefficients between the MRMs and the measured data are higher than 0.6 for the IGUC *SshD* series and higher than

0.4 for the IGIDL *SshD* series (see Figs. 5d and 5c, correspondingly). The explained variance taking into account the number of degrees of freedom ($r_{adj.}^2$) for the IGUC series changes from 25-31% in August-September to 70-83% in January-February and May-July; and for the IGIDL series from 14-25% in August-September to 70-82% in January and May-July. Overall, the MRMs for the IGUC series have better prediction quality than for the IGIDL series.

The role played by each of the regressors is shown in Figs. 5e-f using the β coefficients. As expected, the precipitation and the pressure series are included in the MRMs for almost all of the months throughout a year. The highest β coefficients (in absolute values) are mostly for the wet season (autumn-to-spring). The AI series are included in the "best subset" of regressors for many of the monthly (and annual) series but with quite low β values. The exceptions are dry summer months between June and September. For these MRMs the <AIpos> is an important regressor (see Fig. 5f). On contrary, the <AIneg> series are more often included as the regressors into the MRMs for the wet autumn-to-spring season. Thus, the results of the regression analysis confirm the importance of the aerosol loading to explain observed climatic variations.

5 Conclusions

~~The analysis of the aerosol content changes over two sites in the Continental Portugal, their sources and their role in the local climate variations were studied using both ground and satellite measurements for the 1978-1993 period. One site is an urban region around Lisbon; another is located in a less populated mountain region. The aerosol content was obtained from the data of TOMS instrument on board of Nimbus-7 satellite. Absorbing and scattering aerosols were studied separately. Four main aerosol sources were considered: volcanic aerosols, Saharan dust events (SDE), wildfires and anthropogenic pollution. The effect of the aerosol on the atmospheric conditions was studied using the direct ground measurements by two meteorological stations close to the aerosol measurement sites. The set of atmospheric parameters includes four temperature parameters (minimum, maximum and average daily temperatures, and daily temperature range – *DTR*), atmospheric pressure, precipitation amount and sunshine durations. All data are of monthly and annual time resolution (except wildfire burned area series which are of annual resolution only).~~

The results of the presented analysis show that the aerosol sources chosen in this study (volcanic aerosols, Saharan dust events (SDE), wildfires and anthropogenic pollution) play an important role in the ~~local~~ aerosol content variations over two Portuguese locations (an urban region around Lisbon and a less populated mountain region). Unfortunately, it is impossible to fully separate the effect of the volcanic eruptions, the wildfires and the SDEs. Nevertheless, the regression analysis confirms the relations between the periods of high aerosol content and the periods of more frequently observed wildfires and SDEs. The anthropogenic pollutants also found to affect local aerosol content, especially in the urban region around Lisbon. It was also found that aerosol series averaged over four summer months (from June to September) have stronger relation with the SDEs and wildfires than monthly or annual data. Our results confirm the data from previous studies showing the important role of the anthropogenic pollution, wildfires and SDEs as drivers of the aerosol variation over ~~the~~ Continental Portugal.

The variations of aerosol content were found to be in relations with the changes of atmospheric parameters (temperatures, atmospheric pressure, precipitation amount and sunshine durations). These relations depend on the parameters in questions and change throughout the year. The strongest effect is found for the less urbanized and industrial mountain site. The most significant (both in amplitude and statistically) results were found for the relations between the maximum daily temperature (*Tmax*) ~~(~~and~~, consequently,~~ daily temperature range (*DTR*), and absorbing aerosol content during summer months. These temperature and aerosols variations are also in an agreement with sunshine duration changes. The increase of the content of the absorbing aerosols coincides with the decrease of sunshine duration and, consequently, with the decrease of the *Tmax* and *DTR*. This can be related both to the direct (cooling due to the decrease of the solar radiation flux) and indirect (higher cloudiness amount) effect. The response of the atmospheric parameters to aerosol variations is found to be weaker for the more urbanized region.

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Table 1. Correlation coefficients between the variations of the AI and different forcing parameters: *annual* series in case of volcanic and pollution, and *summer* series in case of SDE and wildfire forcings. Values in brackets are *p* values (only *p* values ≤ 0.2 are shown).

Forcing	Sites	AI series		
		<AI>	<AIpos>	<AIneg>
volcanic	ID 082	0.66 (<0.01)	0.31	0.15
	ID 288	0.62 (<0.01)	0.43 (<0.01)	0.27 (0.03)
SDE	ID 082	0.53 (0.05)	0.75 (<0.01)	-0.11
	ID 288	0.55 (0.05)	0.80 (<0.01)	<0.1
wildfires	ID 082	0.2	0.31	-0.44 (0.12)
	ID 288	<0.1	-0.39 (0.19)	0.27
pollution	ID 082	<0.1	0.51 (0.08)	-0.53 (0.06)
	ID 288	<0.1	-0.17	-0.37 (0.20)

Table 2. Parameters of multiple regression models of AI *annual* and *summer* series (see Section 3.2 for parameter descriptions). The correlation coefficients r greater than 0.67 and $r_{adj.}^2$ equal or greater than 0.45 are in bold, and "x" marks the parameters that were excluded from a particular "best subset".

time period	AI type	site ID	r	p value	$r_{adj.}^2$	β			
						volcanic AOT	SDE number	area burned by wildfires	SO_2 content
annual	<AI>	082	**0.67**	0.014	0.34	0.69	x	x	0.12
		288	**0.81**	< 0.01	**0.55**	0.5	0.73	0.53	x
	<AIpos>	082	**0.72**	< 0.01	0.35	0.41	x	0.34	0.57
		288	**0.76**	< 0.01	**0.49**	x	0.98	0.43	x
	<AIneg>	082	**0.71**	< 0.01	0.40			-0.47	-0.5
		288	0.37	0.21	0.06			x	-0.37
summer (Jun-Sep)	<AI>	082	**0.79**	< 0.01	**0.50**	0.28	0.41	x	0.59
		288	**0.67**	0.012	0.33	x	0.97	0.68	x
	<AIpos>	082	**0.95**	< 0.01	**0.88**	x	0.57	x	0.63
		288	**0.82**	< 0.01	**0.60**	x	1.05	0.37	x
	<AIneg>	082	0.44	0.12	0.12			-0.44	x
		288	**0.68**	0.012	0.41			x	0.68

Figure captions

Figure 1. *a* - Map of Continental Portugal with the locations of the satellite AI observation points (ID 082 and ID 288) and the meteorological stations (Coimbra, Lisbon and Penhas Douradas). Districts used for calculation of the wildfire burned area: 1 – Coimbra, 2 – Guarda, 3 – Castelo Branco, 4 – Santarém, 5 – Lisboa, 6 – Setúbal. *b-d* - Monthly mean <AI> (*b*), <AIneg> (*c*) and <AIpos> (*d*) series for two satellite locations: ID 082 ~~(black lines filled dots)~~ and ID 288. ~~(red lines open dots)~~ *e-f* - Annual cycle of <AIneg> (*e*) and <AIpos> (*f*) for the sites ID 082 and ID 288 for 1979-1992 time period. Please note the inverted Y ~~axis~~ axes in *c* and *e*. Correlation coefficients *r* for the AI series from different sites are shown with *p* values in brackets.

Figure 2. (*a-b*) – Annual variations of aerosol indices <AIpos> (*a*) and <AIneg> (*b*) for two locations: ID 082 ~~(black lines filled dots)~~ and ID 288. ~~(red lines open dots)~~ (*c-f*) – Annual values of parameters describing aerosol forcings: volcanic aerosols (*c* – AOT, annual means), Saharan dust events (*d* – annual sums for two AI locations), wildfires (*e* – total annual burned area close to AI locations) and anthropogenic sulfates (*f* – SO_2, annual means). ~~White bars show global (AOT, mean values for the Northern Hemisphere) or averaged for a number of locations over whole Portugal (SO_2, means for five measurement locations) data; gray bars show data related to the site ID 082; red-white crossed bars show data related to the site ID 288.~~ Four most significant volcanic eruptions are marked in (*c*) by vertical lines: 1 – Mt. St. Helens, 2 – El Chichon, 3 – Pinatubo and 4 – Mt. Hudson. ~~Please note the inverted Y axis in *b*.~~ (*g*) – Monthly variations of SO_2 and aerosol indices <AIneg> for two sites (ID 082 and ID 288) smoothed by the 36-months running averaging. Correlation coefficients (*r*) are between the AI and the sulfate series (*p* values are shown in brackets). Please note the inverted Y axes in *b* and *g* (left).

Figure 3. Multiple regression models (MRM) of the annual (*a-f*) and summer (June-September, *g-l*) AI series: *a, d, g, j* – <AI> series; *b, e, h, k* – <AIpos> series; *c, f, i, l* – <AIneg> series (please note the inverted Y axis). ~~Original AI series are shown by black lines~~

~~with dots; MRMs are shown by red lines with open dots.~~ MRMs are calculated for two locations: ID 288 (*a-c* and *g-i*) and ID 082 (*d-f* and *j-l*). The sets of regressors are shown for MRMs of <AI>, <AIpos> and <AIneg>.

Figure 4. Correlation coefficients (*r*) between the AI indices (<AI> – *a, b*; <AIpos> – *c, d*; <AIneg> – *e, f*) for sites ID 082 (*a, c, e*) and ID 288 (*b, d, f*) and atmospheric parameters measured in Lisbon (IGIDL series – *a, c,* e) and Coimbra (IGUC series – *b, d, f*). Only correlation coefficients $|r| \geq 0.3$ are shown. The statistical significances for singular (*p* values) and multiple (meta *p* values) comparisons are shown by shading. All correlation coefficients are calculated in a way that in case of simultaneous increase/decrease of the amount of any type of aerosols (changes of an absolute value of any AI index) and an atmospheric parameter value the correlation coefficient is positive.

Figure 5. Multiple regression model (MRM) predictions and parameters for the annual and monthly sunshine duration from the IGIDL (*a, c, e*) and IGUC (*b, d, f*) series. *a-b* – the measured annual *SshD* series ~~(black lines filled dots)~~ vs the corresponding MRM predictions. ~~(red lines, open circles)~~ *c-d* – correlation coefficients *r* ~~(black lines)~~ between the MRM and the original series for the monthly data, and the variance explained by the MRMs ($r_{adj.}^2$, in per cent~~, dotted lines~~). *e-f* – *β* coefficients (in parts of standard deviation σ) for each of the regressors for the monthly MRMs.