

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):

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 main 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 <AI_{neg}> (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 <AI_{pos}> 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.

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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 NO_x rather than SO₂.

There are several major groups of anthropogenic gaseous pollutants: sulfur dioxide (SO₂), oxides of nitrogen (NO_x: NO, NO₂), carbon dioxide (CO₂) 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 SO₂ is the fossil fuel combustion, and for NO_x it is the road transportation. Unfortunately, the absence of reliable (preferably measured) data of NO_x for the studied period does not allow us to include information on this pollutant. For the studied period the measured data for NO₂ 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 be 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 NO_x 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. NO_x 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 NO_x 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), Evtuygina 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 SO₂ content are shown in Fig. 2f. As one can see, there is a strong dependence between the variations of the <AI_{neg}> (shown in Fig. 2b) and the SO₂ content. The anti-correlation between the curves reflects the increase of the scattering particles in the atmosphere (lower <AI_{neg}> values) coinciding with the growth of the measured SO₂ concentration."
Changed to

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 <AI_{neg}> (shown in Fig. 2b) and the SO₂ 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 <AI_{neg}> values) coinciding with the growth of the measured SO₂ 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., Evtugina 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. *Evtugina 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., Puigercus, 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.