Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-33-RC1, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Trends analysis of PM source contributions and chemical tracers in NE Spain during 2004–2014: A multi-exponential approach" *by* Marco Pandolfi et al.

Anonymous Referee #1

Received and published: 20 April 2016

Journal: ACP Title: Trends analysis of PM source contributions and chemical tracers in NE Spain during 2004 - 2014: A multi-exponential approach Author(s): Pandolfi M. et al. MS No.: acp-2016-33 MS Type: Research Article

Anonymous Referee #1 Comments.

General Comments.

In this paper a trend analysis of PMx concentrations recorded at two different sites in NE Spain during the period 2004-2014 has been performed. PM10 and PM2.5 chemical composition and PM10 source contributions have also been evaluated with the aim to obtain more accurately interpretations of the trends as well as of the effectiveness of

the pollution control measures implemented in this period by the administrations. Two different methodologies have been used. Namely, the Mann-Kendall test (MK) and the Multi-Exponential fit (ME). In brief, I think that the present work shows some significant and novel contributions to the global scientific community in relation with trend analysis of atmospheric pollutants issues. I think that the availability of time series of PM chemical composition and of estimations of PM source contributions from receptor models, such as PMF, is nowadays a key factor for establishing reliable source-receptor relationships and obtaining robust results from trend analysis and even epidemiological analysis. However, some significant changes must be performed to clarify the usefulness of the different methodologies employed, to reduce the excess of information provided in the manuscript that makes the reading very hard, and to justify the behaviour observed of the trends of some of the time series analyzed.

Specific Comments.

In spite of the fact that the main results of the work (the pollution control measures effectively produced a reduction of the contributions from some anthropogenic sources in such a way that the PMx levels decreased at the urban-background and the regional background sites) are rather consistent and well justified, it is not clear the advantage of using simultaneously both trend analysis approaches, MK and ME.

In general when the ME showed a linear fit, the MK also showed a statistically significant linear trend, but sometimes it is not statistically significant, as in the case of Zn and Na in PM10 for the Montseny site. How should we interpret these different behaviours?. In the case that the MK showed a highly significant linear reduction trend and the ME showed a double exponential fit, Cd in PM10 for the Barcelona site, what result must prevail, the MK or the ME one?. Please, try to clarify the best way to take advantage of using both methodologies.

Why did you decide to work with annual mean values instead of monthly mean values?. It strongly reduced the number of data for the trend analysis.

In my opinion, the MK results can be easily interpreted by the potential readers. I mean, the test provides a statistically significant downward or upward trends for a given confidence level and the value of an estimator of the trend (%variation/year). If the test provides a non-statistically significant trend, then it must be interpreted as the absence of trend. In the case of the ME results it is not clear the meaning of linear, single-exponential and double-exponential fit in relation with the trends of the pollutants. For the double-exponential fit cases, the values of T1 and T2 are sometimes quite different, even positive and negative. Despite some explanations of these values are included in the text, the interpretation remains somewhat obscure. Some more information should be included in the 2.5 section about the interpretation of the equations and coefficients representing linear, single-exponential and double-exponential fit. Is the multi-exponential fit statistically significant in all the cases?. I suppose that the fit parameters an and Tn are estimated by the program with statistically significance for a given confidence level. As in the case of the coefficients of a multilineal regression analysis. Is not it?. Please, try to clarify all these questions in the revised version of the manuscript.

Otherwise, the ME and MK methodologies provides for any case a lot of information. I mean, the trend estimation, p-value, type of fit,... These results are summarized in tables 1-5. However, the authors decided to include most of this information again in the text. As a consequence the reading is very hard and confusing. For the MK results it is not necessary to include the magnitude of the trend and the degree of statistical significance in all the cases. Please, try to highlight in the manuscript only the most important information and refer to the tables for details.

I would like to underline one important fact. Sometimes it can be read in the text statements as “non-statistically significant decreasing trend (-1.25 %/year” (page 10, lines 297-298). I completely disagree with this. If the test provides a non-statistically significant trend result, you cannot assure the existence of a trend, neither downward nor upward. That is the aim of performing statistical tests. Accepting or rejecting null

hypothesis with statistical significance. Hence, you can neither talk about increasing or decreasing trend nor show the value of the estimator of the trend in the non-statistically significant trend cases. I believe that you must rewrite the manuscript and the tables, excluding the values of the estimator of the trends for non-statistically significant cases.

The reason why you decide not to use the data of mineral matter and road traffic emissions from the Barcelona site before 2009, has been repeatedly mentioned across the manuscript.

The statement that “2002âĂŘ2014 represented the largest period of gravimetric PM2.5 measurements available at MSY station” has been also repeated unnecessarily.

In page 10 (lines 304-310) and page 13 (lines 402-403) you mentioned results from subsequent sections. I think it should be avoided for clarity.

In section 3.2 you attributed the differences observed in the magnitude of the trends for different time periods to meteorology variability. In section 3.3 (lines 509-511) you stated that “It is probable that variations in meteorological conditions from one year to another (i.e. intensity and frequency of Saharan dust outbreaks) might also explain the observed trend of mineral tracers at regional level”. In section 4.1 (lines 656-658) you also declared that “This decreasing trend could be due to a possible decrease of the emissions of anthropogenic mineral species from specific sources such as cement and concrete production and construction works”. These comments are highly speculative due to the fact that neither meteorological variables, nor information on “intensity and frequency of Saharan dust outbreaks”, nor information on “cement and concrete production and construction works” have been analyzed to support them. In my opinion it should be mandatory to carry out an analysis of this kind of data to confirm these hypotheses.

Some other minor comments and suggestions:

Did you achieve a PMF source contribution study with the PM2.5 data base?. It should

be interesting to compare the results of the trend analysis for PM10 and PM2.5 source contributions.

Page 6, lines 188-189. “(End user’s guide to multilinear engine applications from Pentti Paatero)”. What do you mean?. Is that a reference?.

Page 6, line 190. You declared that rotational ambiguity of the PMF solution was handled by means of the Fpeak parameter. However, some better tests to estimate rotational uncertainty than Fpeak are now available in the latest version of EPA PMF (V.5.0) such as the base model displacement error estimation and other rotational tools. Have you checked these new options?.

Page 7. Section 2.4. The description of the Mann-Kendall test is very short. Some more information should be included.

Page 8. Lines 230-232. What does Cbeg and Cend mean?.

Page 9. Lines 259-261. The description of the section was made before in the previous section. This paragraph can be omitted.

Page 9. Lines 263-264. “Note that the recommended annual data coverage for trend studies is typically 75%”. Where does it come from?. Can you include the source in the Reference section?.

Page 9. Lines 276-277. The results of the comparison between simultaneous PMx chemical speciated data collected at both BCN measurement sites can be showed as Supplementary Information.

Page 10. Line 317. What is the definition of the Residual Component (RC)?.How did you computed it?. This information should be included in Section 2.

Page 20. Line 633. Querol et al. 2007, is not included in the Reference section.

Technical corrections/Typing errors.

Figure 2 has very low quality. It is very hard to distinguish among the different symbols. The grey lines and symbols are very diffuse. This is also true for figures 3, 4, 6 and 7.

Table 4 is unnecessary. This information is showed in Figure 5.

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