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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of Sulfate Aerosols over Austria: A Case Study" *by* Camelia Talianu and Petra Seibert

Anonymous Referee #2

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General comments: the manuscript investigates potential non-local sources of sufate aerosols influencing a background air quality monitoring station in Austria, on a case study basis. It integrates different aerosol data platforms and transport models: in situ measurements, Lidar measurements, assimilated satellite-based remote sensing data (CAMS products), and particle dispersion modelling (FLEXPART). This makes the manuscript interesting, although the integration of different data platforms is not new. The results are somehow qualitative, and the manuscript would gain significance if the discussion could be improved to provide some quantitative results. For example, estimating percent contribution of sulfate source regions. In addition, aerosol aging and dilution along the transport could be explored. Uncertainties and limitations of the method could also be better discussed. In my opinion, the discussion should be

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Discussion paper

improved, according to the specific comments below.

Specific comments:

1) Page 2, Lines 1-2 (Introduction): worldwide in situ observations of refractory PM1 chemical composition have shown that the sulfate contribution may reach more than 50% of aerosol mass, depending on the location. See, for example, Zhang et al., 2007.

2) Page 2, Lines 11-14 (Introduction): a recent and important reference on SO2 sources worldwide, and also on sulfate radiative effects, is Yang et al., 2017.

3) Page 2, Lines 18-20 (Introduction): I recommend that you add a phrase or two to improve the description of the sulfate radiative effects, both direct and indirect. Also, you must include some key references for that.

4) Page 2, Lines 27-29 (Introduction): Do you know of previous studies that promoted integration of data from in situ observations, remote sensing measurements and atmospheric transport modelling? I recommend that you provide an outlook of what has been done before, concerning to data integration from different platforms.

5) Page 2, Line 29 (Introduction): please include a reference for NATALI aerosol typing model.

6) Page 3, Line 15 (Methods): You must give more details about the ground based air quality monitoring site and surroundings. Are there local air pollution sources affecting the site? How is the topography of the surroundings? What are the typical aspects of atmospheric circulation? Are there other air quality monitoring stations nearby?

7) Page 3, Line 15 (Methods): I suggest that you include a description of the general aspects of climate and atmospheric synoptic scale circulation for the study region and season.

8) Page 3, Line 25-28 (Methods): It is very important to include a map showing the location of all stations explored in this manuscript. That will improve understandability

for the readers that are not familiar with EARLINET and with general aspects of Europe geography.

9) Page 5, Line 2 (Methods): I suggest that you briefly explain (2-3 phrases) how a source-receptor model works. What do you need as input? Are there iterations required to tune the model parameters, in order to match model results and observations?

10) Page 5, Line 12-14 (Methods): the term “pure aerosol” usually refers to homogeneous particles made of a single chemical compound. This is not the case of aerosol classes like “continental”. Please find another term.

11) Page 5, Lines 28-32 (Methods): it seems that there is a circular reasoning here: you aim to determine aerosol types from Lidar observations (Tables 3 and 6), but, at the same time, you have to assume aerosol types based on NATALI to make use of part of the Lidar observations. Please comment on that.

12) Page 6, Lines 1-2 (Methods): Please include a brief description (1-2 phrases) of the gradient method for detecting aerosol layers, and include more references for that. It is important to state the criteria used to identify an aerosol layer, to provide reproducibility of results. Also, please clarify that you applied the gradient method both for Lidar and CAMS profiles.

13) Page 6, Lines 11-15 (Methods): the analysis of air quality timeseries were performed for how many years of data? How frequent were events like the one you described in the manuscript, in April 2014? What is the objective criteria for “significant excess”?

14) Page 7, Line 14 (Methods): “The release is set to the location of the in situ station”. The word “release” is confusing in this context, because it gives the impression that pollutants were set to be released at the in situ station. Maybe “target” would work better here.

15) Page 7, Line 17 (Methods): did you also consider SO2 biogenic sources, like oxidation from DMS? If not, how does it influence your results?

16) Figure 1: SO2 lifetime in the troposphere is typically in the order of hours. Therefore, the SO2 observed at the ground based station may have a contribution of local sources, and possibly cannot be attributed to the regional transport (1-2 days) described in the case study. Please comment on that.

17) Figures 2 and 3: could you convert “model levels” to altitude, to improve understandability of the plots?

18) Page 8, Lines 15-17 (Results): if the diurnal evolution of sulfate and dust layers are correlated with SO2 and PM2.5, and dust is correlated with PM10, can I conclude that all variables are correlated? It would be interesting to see the diurnal evolution of ground based measurements and CAMS. In addition, is this correlation between CAMS and ground based measurements valid for all layers, or just for the lowermost layer?

19) Figure 4: the legend is illegible, text must be enlarged. To better interpret this Figure, it is important to know which profiles correspond to daytime and nighttime (i.e., local time of each plot). Information on the typical planetary boundary layer height at Pillersdorf would also help. It would be interesting to point out whether and when there was an input of aerosols from upper layers to the boundary layer, affecting air quality at Pillersdorf.

20) Figure 5: there are too many lines (altitudes) in the lower plots of sub-figures, it is difficult to interpret. There must be a compromise between completeness and understandability. I suggest that you keep only 3-4 representative altitudes (low, medium, high).

21) Figures 6, 10c, 11c, 12d: you must indicate the locations of the monitoring stations in the maps.

22) Figure 10c: the model calculates SO2 < 1 ug/m3 for layer 1, which is inside the boundary layer, all over Europe. How does it compare to your ground based measurements?

23) Table 3: can you see changes on aerosol properties as they are transported? For example, layers 1 and 2 at Leipzig and Pillersdorf are associated (Table 1). How does aerosol intrinsic and extrinsic properties change along this ∼24h transport?

24) Page 9, Lines 19-24: since you did not discuss April 4 in detail, I recommend moving it to the supplementary material, as well as the corresponding figures and tables.

25) Page 10, Lines 10-28: the discussion of trajectories and source regions is rather qualitative. Terms like “medium to smaller contributions” are vague. Could you estimate percent contributions? Also, it is important to recognize limitations and uncertainties of the method.

26) Page 10: meteorological maps for the case study period would help to support the conclusions on aerosol transport. Particularly, trajectories calculated below 2000 m are more prone to uncertainties.

27) Figure 16: it does not contribute significantly to the discussion. I suggest to exclude this figure, or to move it to the supplementary material.

28) Page 11 (conclusion): what are the main advantages of the method you used for this case study, compared to previous database-integration studies? What are the main limitations? How can the method be improved?

Technical corrections:

1) Page 2, Line 17: what do you mean by “key properties”? Optical? Physical? Be more specific.

2) Page 5, Line 11: omit the word “Ref.” before citing a reference. That occurs all

through the manuscript, please check.

3) Page 6, Line 6: “The values of the CAMS quantities”: please use a more specific term, instead of “quantities”.

4) Figure 7: scales are illegible.

5) Figures 7 and 8: could be merged into a single figure. I recommend reformulation of the lidar plots adopting a standard pattern for the contourplots.

6) Figure 9: what are the units for the color map? In addition, the units of longitude should be “degrees”, and not “degrees E”.

7) Table 3: Please define abbreviations in the table caption, to facilitate interpretation.

References:

Yang, Y., Wang, H., Smith, S. J., Easter, R., Ma, P., Qian, Y., Yu, H., Li, C. and Rasch, P. J.: Global source attribution of sulfate concentration and direct and indirect radiative forcing, Atmos. Chem. Phys., 17, 8903–8922, doi:10.5194/acp-17-8903-2017, 2017.

Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Allan, J. D., Coe, H., Ulbrich, I., Alfarra, M. R., Takami, A., Middlebrook, A. M., Sun, Y. L., Dzepina, K., Dunlea, E., Docherty, K., DeCarlo, P. F., Salcedo, D., Onasch, T., Jayne, J. T., Miyoshi, T., Shimono, A., Hatakeyama, S., Takegawa, N., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Williams, P., Bower, K., Bahreini, R., Cottrell, L., Griffin, R. J., Rautiainen, J., Sun, J. Y., Zhang, Y. M. and Worsnop, D. R.: Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes, Geophys. Res. Lett., 34(13), 1–6, doi:10.1029/2007GL029979, 2007.

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