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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

Camelia Talianu and Petra Seibert

camelia.talianu@gmail.com

Received and published: 25 March 2019

Dear referee,

Thank you very much for the comments to our paper.

Here are the answers to your comments. In the following: "RefC" is the comment from Referee, "AuthR" is the author's response and "AuthCM" represents the author's changes to the manuscript. Page and line number refer to the page and line number in the version submitted for discussion.

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Discussion paper

Specific Comments

Comment 1.

RefC: "Section 2 (Methodology) is rather lengthy and very detailed. Some parts could perhaps be moved into an Appendix Section."

AuthR: Due to the complexity of the synergy between remote sensing instruments, in situ monitors and modelling, we would prefer to provide the methodology in one section, in the article, even if it is indeed lengthy and very detailed. Splitting it in a short(er) version in the article and details in the Appendix could make it difficult to follow.

AuthCM: none

Comment 2.

RefC: "The authors should avoid to mention Trade Names or direct references to companies and commercial instruments, unless absolutely necessary for the understanding of the methods deployed."

AuthR: The trade names of the instruments were deleted from the text. Please note that PollyXT and RALI are the names of the instruments, as used by EARLINET to identify the instruments at the corresponding station, not trade names.

AuthCM: Page 3, Line 21 "a Thermo Scientific Model 43i SO_2 Analyzer" was changed to "a SO_2 analyzer"

Page 3, Line 22 - Line 23 "Optical Particle Counter GRIMM Dust Monitor Model EDM180," was changed to "optical particle counter"

Page 3, Line 23 - Line 24 "a Thermo Environmental Instruments Ozone Analyzer, model TEI 49C," was changed to "an ozone analyzer,"

Page 4, Line 4 "Jenoptik ceilometers CHM15kx" was changed to "ceilometers"

Comment 3.

RefC: "Since the trajectories in Fig.5 indicate sources processes from almost all over Europe (understandable, especially in the lower & mid levels), but also very distant sources (mostly in elevated layers), the authors should show the relevant meteorological maps for the study period (850, 700, 500, 300 & 200 or 250 hPa circulation) to provide physical evidence for 'conflicting' circulations in some of the layers, and especially for the 'outlying regions'. Of course, FLEXTRA ingests the upper air data from ECMWF, but a cross-verification with 'real meteorological data' will make the cases more convincing."

AuthR: There is no reason to not trust the FLEXTRA calculations. However, seeing weather maps may help to understand the prevailing synoptic pattern. Therefore, a few weather maps are now provided in the Supplement.

AuthCM: Added weather maps as supplement.

Comment 4. Part A

RefC: "P10 / L27 & 28: - 'No contributions from Europe are seen for these layers.' This may be true for the period in April, as there may not have been any deep convection. However, it would be interesting to also study a summer period with strong convective activity over Central Europe (obviously, in a separate paper !)."

AuthR: The summer periods for the years 2014 – 2017 are under study, and a paper is under preparation.

AuthCM: Page 11, Line 28 Added text: "The spring period studied in this paper is characterized by low, if any, deep convection. For the summer period, one expects however to have a strong convective activity over Central Europe. A study of the summer periods for the years 2014–2017 for the same region was also performed; the results will be presented in a separate paper."

Comment 4. Part B

RefC: "P10 / L27 & 28: "I am still a bit skeptical about the long-range transport of pollutants - there would be a significant dilution factor . . .! Unless there are major sources emitting ? An indication of such sources would make your findings more convincing."

AuthR: Flexpart simulates not only the transport due to the large-scale winds but also turbulent diffusion and mixing by subgrid-scale mesoscale motions (A. Stohl et al., 2005). Furthermore, it has implemented the treatment of all loss processes, including dry and wet deposition of gases or aerosols, gravitational settling of particles (S. Eckhardt et al., 2017). Flexpart also has implemented a deep convection scheme. Comprehensive validations of Flexpart were performed for intercontinental air pollution transport, see e.g. [A. Stohl et al Atmos. Environ., 32, 4245–4264, 1998], [A. Stohl and T.Trickl, Geophys. Res., 104, 30,445–30,462, 1999], [N.Kristiansen et al., Geophys. Re. Lett. 42, 588-596, doi 10.1002/2014GL062307, 2015]. Thus, there is no reason to doubt the results.

For the case study presented in this paper, the major sources of SO_2 are coal power plants and other industrial facilities (refineries, chemical industry, etc), present in the regions mentioned: Central Europe 'Black triangle', industrialized cities from Morocco, Eastern part of US (e.g. Ohio, New Jersey), Southeastern part of US (e.g. Louisiana,

Alabama). An exhaustive list of US sources is mentioned in the report "U.S. EPA 2014 NEI Version 1.0" [https://www.epa.gov/sites/production/files/2017-04/documents/2014neiv1_profile_final_april182017.pdf] A recent study on SO_2 sources worldwide is published in (Y. Yang et al, 2017), which was added to the references.

AuthCM: added references (A. Stohl et al., 2005) to Page 3, Line 4 and (S. Eckhardt et al., 2017) to Page 3, Line 2 for Flexpart and (Y. Yang et al, 2017) to Page 2, after Line 14 for SO_2 sources.

Technical corrections

Comment 5.

RefC: "Figs. 4 & 13 - Key for variables needs to be enlarged P8 / L20 - Height of layers 'amsl' or 'AGL' (also in tables)"

AuthR: The recommended corrections were done in the two figures. AGL was added to text and to the caption for the two figures. No AGL added to the tables, I think it is enough to mention in the text and to add to figure.

AuthCM: Page 3, Line 11: changed to "as ground-level altitudes (AGL)."; added AGL to the caption of the new Figs.4 & 13

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1155, 2018.

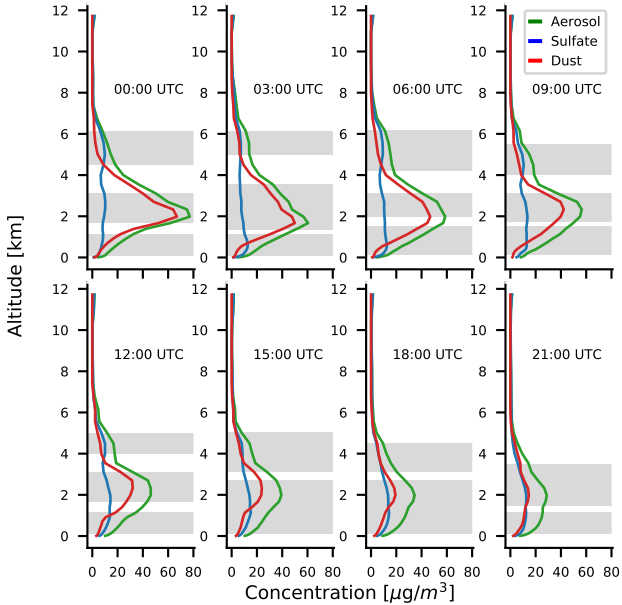

Fig. 1. CAMS total aerosol, sulfate and dust profiles for 02 April 2014, Pillersdorf. Grayed area represents the identified sulfate layers. Altitudes are given in km AGL. Local time is UTC+2

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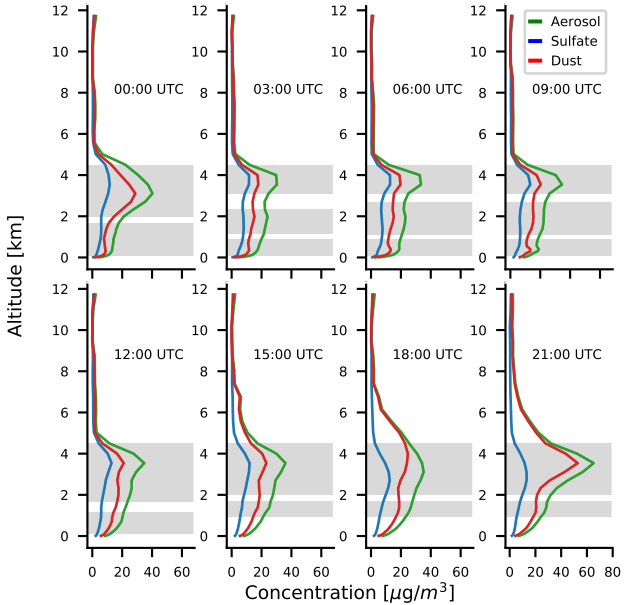

Fig. 2. CAMS total aerosol, sulfate and dust profiles for 04 April 2014, Pillersdorf. Grayed area represents the identified sulfate layers. Altitudes are given in km AGL. Local time is UTC+2

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Discussion paper

Specific Comments

Comment 1.

RefC: 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."

AuthR: This reference was added to the text and it was included in References list.

AuthCM: Page 2, Line 2: Added
"; worldwide in situ observations of refractory PM_1 chemical composition have shown that the sulfate contribution may reach more than 50% of aerosol mass, depending on the location (Zhang et al., 2007)."
Added reference (Zhang et al., 2007).

Comment 2.

RefC: 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."

AuthR: Added sentence to the text, referencing also the paper.

AuthCM: Added on Page 2, Line 14: “A recent review of SO_2 sources worldwide can be found in (Yang et al., 2017).”

Comment 3.

RefC: 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."

AuthR: Done, see changes below.

AuthCM: Page 2, Line 18: added references AEROCOM project, IPCC AR5 for cooling effects of sulfate aerosol
Page 2, Line 20: Added text:
“The direct radiative effects are strongly correlated to the emission sources, while the indirect effects are correlated to both emission sources and cloud cover (Déandreis et al., 2012) (Yang et al., 2017).”

Comment 4.

RefC: 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."

AuthR: In the last decade, the synergy of the in situ, remote sensing data and models was used in more atmospheric studies related to long-range transported aerosols and estimation of their potential sources (see for example A. Pappayanis et al. (Sci Total Environ. 2014;500-501:277-94. doi:10.1016/j.scitotenv.2014.08.101, 2014 - C.T. coauthor), D. Nicolae et al. (2013 - C.T. coauthor, Ansmann et al 2018, Eckhardt et al 2008 - P.S. coauthor, Cazacu et al 2012 - C.T. coauthor], [Sauvage et al 2017], [Chalbot et

al 2017],[D.G. Kaskaoutis et al., 2012]).

However, to our best knowledge, there have been no studies combining CAMS-based aerosol data with remote sensing and in situ measurements and transport models.

AuthCM: Added on Page 3, at the end of section “Introduction”:
“The synergy of the in situ, remote sensing data and models was used in more atmospheric studies related to long-range transported aerosols and estimation of their potential sources; see for example (Papayannis et al., 2014) for dust, (Nicolae et al., 2013) and (Ansmann et al., 2018) for fires, (Eckhardt et al., 2008) and (Cazacu et al., 2012) for volcanic ash, (Sauvage et al., 2017), (Chalbot et al., 2013) and (Kaskaoutis et al., 2012) for anthropogenic aerosols. However, to our best knowledge, there have been no studies combining CAMS-based aerosol data with remote sensing, in situ measurements and transport models. The assimilation of ground-based remote sensing measurements in CAMS is a long-term goal.”

Comment 5.

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

AuthR: Added reference for NATALI aerosol typing.

AuthCM: Page 2, Line 29 "... and NATALI aerosol-typing model, and atmospheric transport modeling." is replaced by "... and NATALI aerosol-typing model (Nicolae et al., 2018), and atmospheric transport modeling."

Comment 6.

RefC: 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?"

AuthR:

AuthCM: Added text to Page 3, Line 16
“Pillersdorf (315 m) is located in hilly terrain in the northeastern part of Austria, around 60 km north from Vienna. The station is a part of the national background monitoring network and an EMEP background monitoring station. The surroundings are mostly forests and agricultural areas far from strong anthropogenic sources. Austria belongs to the midlatitude climate belt, in the transition between maritime and continental climate, and the weather is dominated mostly by travelling highs and lows. The station provides:”

Comment 7.

RefC: 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."

AuthR: The description was added to the text for Comment 6.

AuthCM: none

Comment 8.

RefC: 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."

AuthR: OK.

AuthCM: A map has been added to the Supplement.

Comment 9.

RefC: 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?"

AuthR: A more detailed explanation has been introduced. FLEXPART is not tuned or iterated.

AuthCM: Page 5 Line 2: Added after "...or gridded sources"
"(Seibert and Frank, 2004). The model ingests ECMWF 3D meteorological fields and solves the equations for transport, turbulent diffusions and other relevant processes in a Lagrangian framework (Stohl et al., 1998) (Pisso et al., 2019). The sensitivity of a receptor concentration to potential sources is obtained directly as the model output in the case of a backward run (Seibert and Frank, 2004) (Eckhardt et al., 2017)."

Comment 10.

RefC: 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."

AuthR: Changed "pure aerosol" to "typical aerosol".

AuthCM: All occurrences of “pure aerosol” changed to “typical aerosol”.

Comment 11.

RefC: 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."

AuthR: The classes of typical aerosols in NATALI are defined based on the optical properties. If one of the properties is not measured, the type of the aerosol could still be identified based on the other measured properties, which are usually enough to constrain (approximately) the aerosol type. So, on Page 5, Line 28-32, the aerosol type is identified using particle depolarization ratio and AOD at different wavelengths. The lidar ratio is not measured, but it is assumed to be in the range attributed to that class. Please note that the validation of the NATALI model was performed on measurements having all properties measured. So, it is not a circular reasoning, but an estimation based on fully-characterized cases. There is no other way if the measurement is missing. As a cross-check, the type of the aerosol is constrained identifying the potential source with the transport model.

AuthCM: none

Comment 12.

RefC: 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."

AuthR: Done, see changes to manuscript.

AuthCM: Page 6, Lines 1-2 changed to
"The aerosol layers are identified from the lidar measurements with the gradient method, applied to the RCS profiles (Belegante et al., 2014) (Nicolae et al., 2018). The gradient method is based on the identification of the peaks/valleys from the first derivative applied to the vertical profiles. If two consecutive layers are very close (less than 100 m), these layers are merged into one layer. Also, if the signal to noise ratio in the layer is lower than a threshold (here set to 5), the layer is discarded."
Page 6, Lines 24-26 changed to:
"The layers for the event at the in situ site are then determined by applying the same gradient method as for lidar data processing, but applied to the altitude profiles of aerosol concentrations. The concentrations are computed by multiplying the CAMS mixing ratios and the air density."

Comment 13a.

RefC: 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?"

AuthR: The analysis of air quality time series were performed for the spring and summer periods for the years 2010–2014. Events as described in the manuscript occur typically 1–2 times per year, between March and April. Unfortunately there were no lidar data to analyze the other events (no trajectories over lidar stations, no high-enough quality data for the corresponding periods). April 2010 was also dominated by the Eyjafjallajökull volcanic eruption.

AuthCM: none

Comment 13b.

RefC: Page 6, Lines 11-15 (Methods): "What is the objective criteria for “significant excess”?"

AuthR: The criterion for "significant excess" is 50% above the averaged values for 30 days.

AuthCM: Page 6, Line 13: changed "...is identified, " to "...is identified (values exceed by 50% the averaged values for 30 days),"

Comment 14.

RefC: 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.""

AuthR: "release" was changed with "receptor".

AuthCM: "The receptor is set to the location"

Comment 15.

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

AuthR: yes, the Flexpart model considers all the sulfate sources, including SO_2 biogenic sources. Also, for the trajectories at high altitude over ocean, the influence of biogenic sources is small.

AuthCM: none

Comment 16.

RefC: 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."

AuthR: As we mentioned before (see answer to comment 6), the station Pillersdorf is a regional background site, with no significant local anthropogenic sources, especially of SO_2 (by the requirements of EMEP "background site" and also because the SO_2 emissions are generally very low in Austria). The SO_2 measured at Pillersdorf is the result of the sulfate transported together with dust.

AuthCM: none

Comment 17.

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

AuthR: we would prefer to keep the model levels in these plots because the value (as altitude) of each model level is variable, it depends on the meteorological conditions (temperature, humidity, etc) that are variable in time. For each case, the altitude can be computed using the geopotential heights.

AuthCM: none

Comment 18.

RefC: 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?"

AuthR: There is a correlation between sulfate and dust layers and values of SO_2, $PM_{2.5}$ and PM_{10} from in-situ measurements, but it is important to analyze each case separately to find the correlation factor between them. Also, the correlation between CAMS and ground based measurements is valid for the lowermost layer (in situ data are assimilated in CAMS). For the rest of the layers, not much can be said as CAMS does not assimilate yet ground-based lidar data.

AuthCM: none

Comment 19.

RefC: 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."

AuthR: We have improved the figure and added a sentence on boundary-layer heights. We also added in the caption of Fig. 4 the UTC–local time difference.

AuthCM: Changed fonts in the legend of Fig. 4; modified caption: changed “aerosol” to “total aerosol”; added “Local time is UTC+2.”.

Added on Page 6, Line 31:
“During the period under investigation, with low wind speeds and mostly clear skies, the boundary-layer height varied at Pillersdorf from less than 100 m at night to about 1500 m in the afternoon.”

Comment 20.

RefC: 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)."

AuthR: The figure was changed, keeping only the trajectories passing over the lidar stations involved in this analysis.

AuthCM: New Fig. 5

Comment 21.

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

AuthR: The figures were changed adding the location of the monitoring stations.

AuthCM: New figures 6, 10c, 11c, 12d

Comment 22.

RefC: 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?"

AuthR: There was a bug in superposing the concentration distribution on map for the zoomed distribution (only). The figures 10c, 11c and 12d were replaced with figures with correct data.

There is a good agreement between model and ground based measurements, as can be seen from Fig. 1 and Fig. 10c.

AuthCM: the figures 10c, 11c and 12c were replaced.

Comment 23.

RefC: 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 $\approx$ 24h transport?"

AuthR: Yes, changes on aerosol properties can be observed for the correlated layers. For layer 1, the aerosol transported from Leipzig to Pillersdorf is mixed with the aerosols accumulated inside the PBL (the trajectory is under 1500 m), leading to an increase of the sulfate fraction in aerosols. The aerosol size increase also. These are reflected by the decrease in the lidar ratio and AE, measured at Leipzig and computed at Pillersdorf.

For Layer 2 (above PBL), the depolarization ratio, sulfate fraction and AE decreases, due to chemical processes (aging) and aerosol removal processes. The lidar ratio slightly increases, due to the decrease of the sulfate fraction from aerosol.

AuthCM: none

Comment 24.

RefC: 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."

AuthR: We would prefer to keep the event in the article, as it is consistent with the event from Apr 02 and emphasize the main message of the paper (long-range transport of sulfate aerosols must be considered for local changes, as it can have non-negligible effects).

AuthCM: none

Comment 25.

RefC: 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."

AuthR: I could compute the contributions of the sources, but they would be obtained from the transport model, they would be a rough estimation, therefore I prefer to give only qualitative values. For confident quantitative results, I would need more measured data, time-dependent, not only monthly averaged sources, to be able to compute the central value and to compute the uncertainties.

AuthCM: none

Comment 26.

RefC: 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."

AuthR: The meteorological maps will be added to the Supplement of the paper.

AuthCM: meteorological maps in the supplement.

Comment 27.

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

AuthR: OK

AuthCM: Fig. 16 was moved to the Supplement of the paper.

Comment 28.

RefC: 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?"

AuthR: I think this is covered by the text added in Introduction as answer to Comment 4 and by the text from Page 11, lines 25 - 31.

AuthCM: none

Technical corrections

Comment 29.

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

AuthR: "key properties" means "optical, physical and chemical properties".

AuthCM: Page 2, Line 17
"The key properties" was changed to "The optical, physical and chemical properties"

Comment 30.

RefC: Page 5, Line 11: "omit the word “Ref.” before citing a reference. That occurs all through the manuscript, please check."

AuthR: OK.

AuthCM: removed “Ref. “

Comment 31.

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

AuthR: "the CAMS quantities” were specified in text at page 4, line 22 - line 28.

AuthCM: Page 6, Line 6: “... the CAMS quantities” was changed to “... the CAMS products (mixing ratios, temperature, specific humidity, etc)”

Comment 32.

RefC: Figure 7: "scales are illegible."

AuthCM: Figure 7 The fonts for scales were increased to be more legible.

Comment 33.

RefC: 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."

AuthR: The lidar plots are taken in the format available in the Earlinet database. I prefer to keep the format used by the Earlinet database for consistency.

AuthCM: none

Comment 34.

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

AuthR: The units for color map are seconds. The units of longitude and latitude were corrected.

AuthCM: the figures 9, 10, 11, 12 and 15 were replaced with the figures with units.

Comment 35.

RefC: Table 3: "Please define abbreviations in the table caption, to facilitate interpretation."

AuthR: The abbreviations are defined in the text.

AuthCM: none

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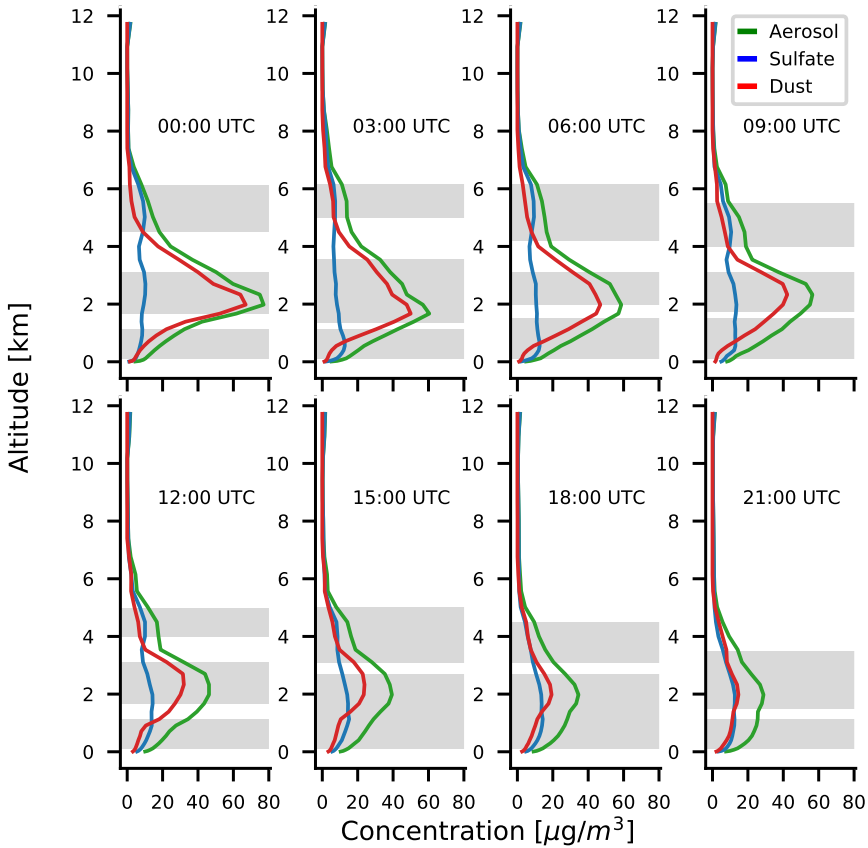

Fig. 1. CAMS total aerosol, sulfate and dust profiles for 02 April 2014, Pillersdorf. Grayed area represents the identified sulfate layers. Altitudes are given in km AGL. Local time is UTC+2.

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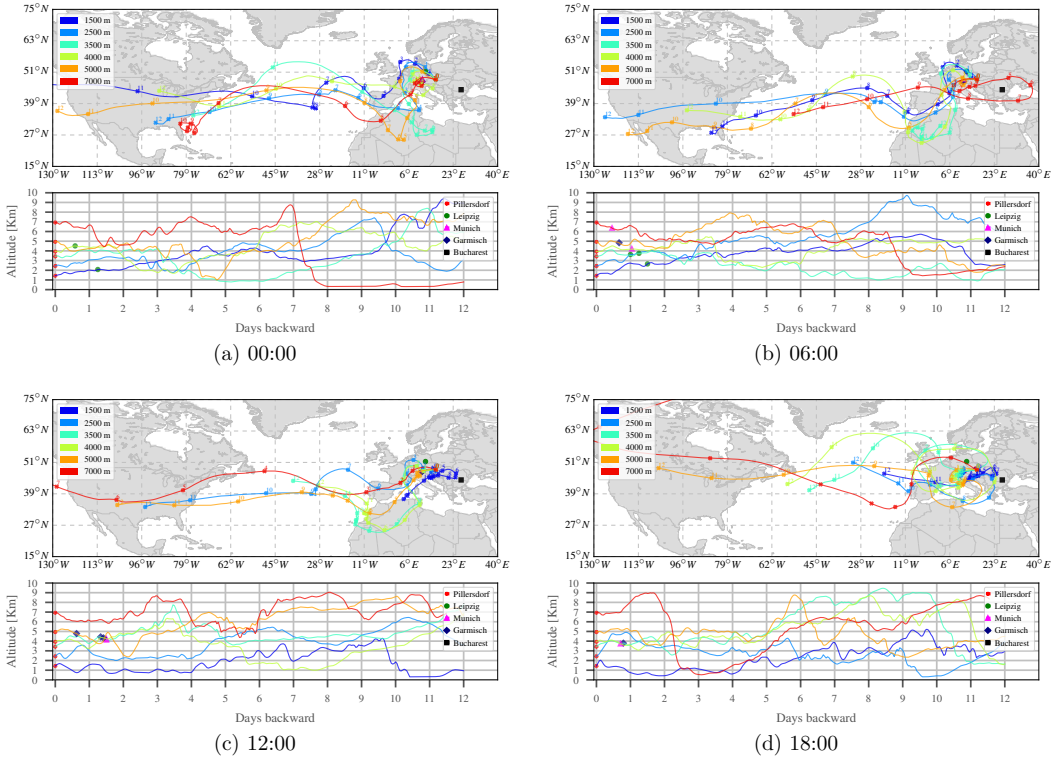

(a) 00:00 (b) 06:00

(c) 12:00 (d) 18:00

Fig. 2. Pattern of back-trajectories (upper plot of sub-figure) and their altitude profile, including overpassed lidar stations (lower plot of sub-figure) for Pillersdorf, 02 April 2014.

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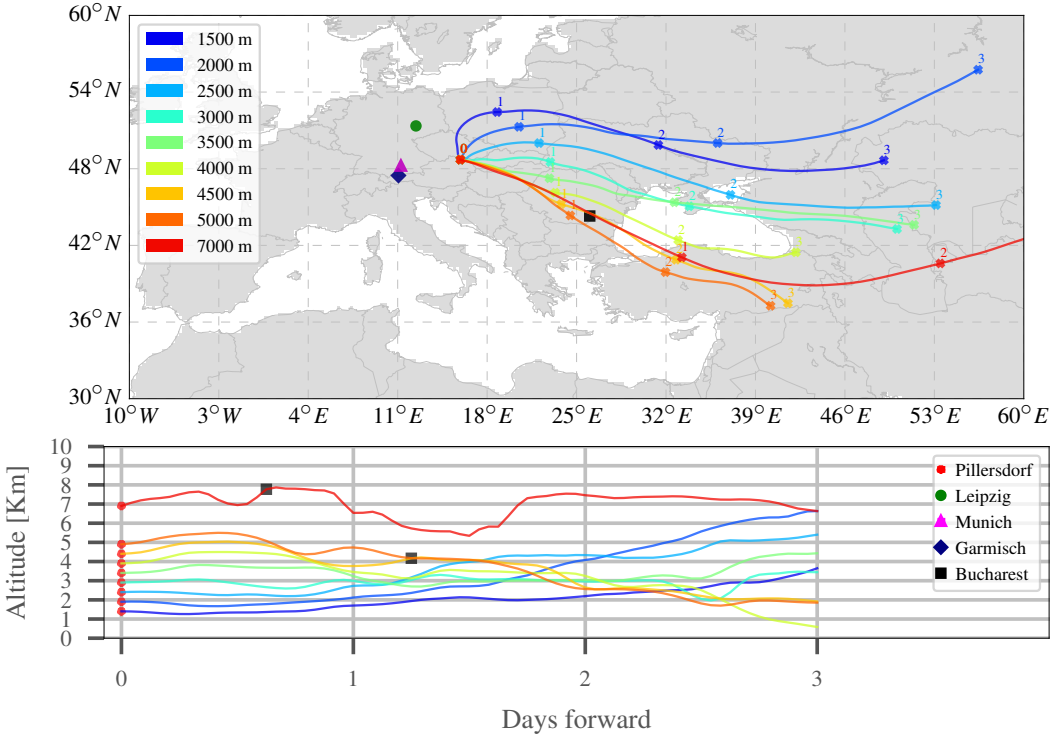

Fig. 3. Pattern of forward-trajectories (upper plot) and their altitude profile, including over-passed lidar stations (lower plot) for Pillersdorf, 02 April 2014, 06:00.

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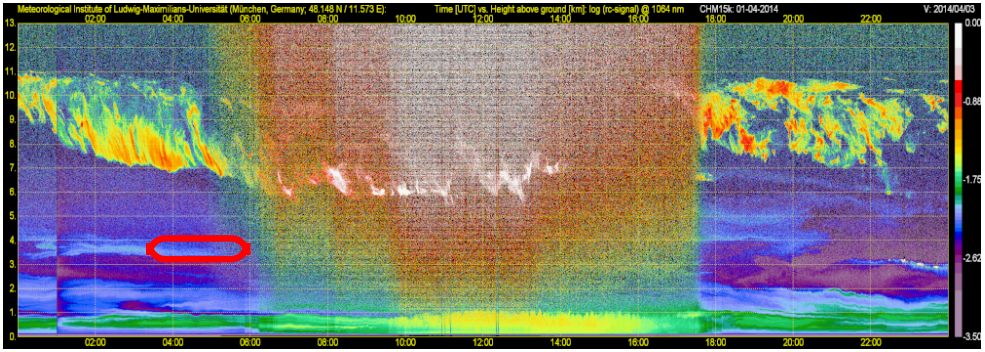
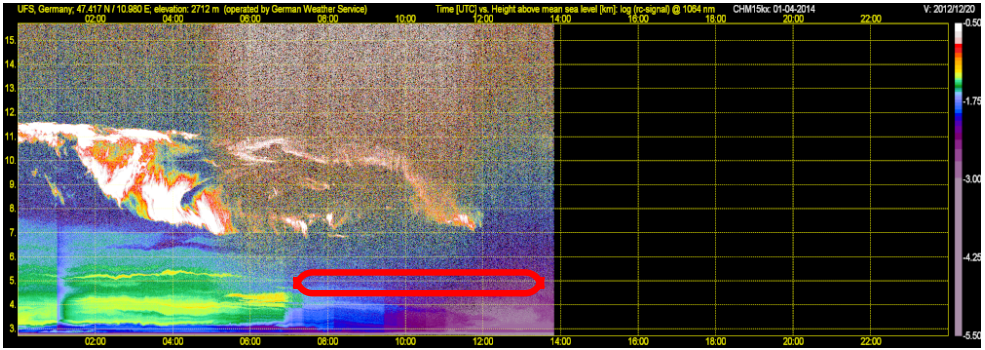


(a) Munich, 01 April 2014 Ceilometer YALIS



(b) Garmisch, 01 April 2014 Ceilometer

Fig. 4. Logarithm of the range corrected signal at 1064 nm, 24 h, for Munich (a) and Garmisch (b) stations. The red line boxes represent the identified layers.

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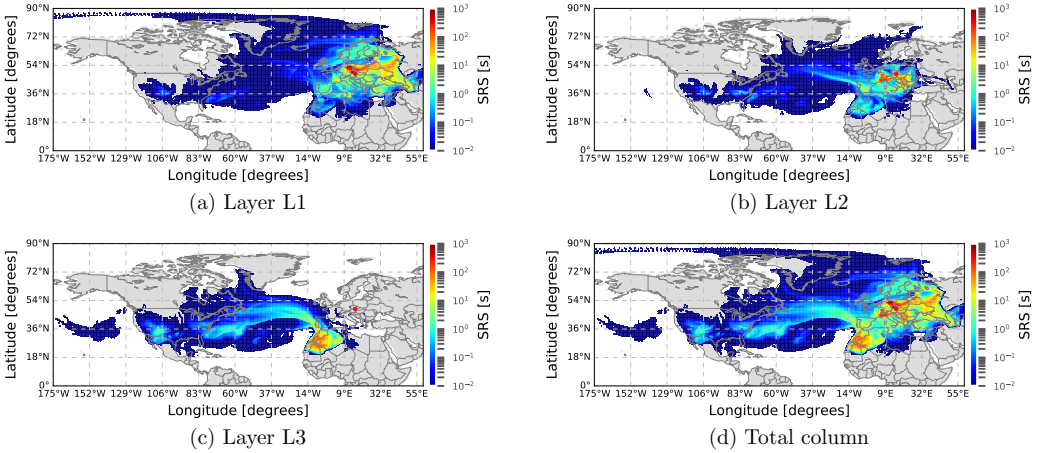
Discussion paper



(a) Layer L1 (b) Layer L2

(c) Layer L3 (d) Total column

Fig. 5. Source-receptor sensitivity for layer L1 (a), L2 (b) and L3 (c) and total column (d), Pillersdorf, 02 April, 6:00.

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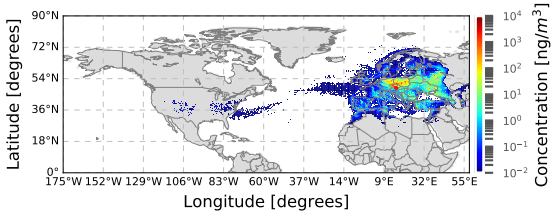
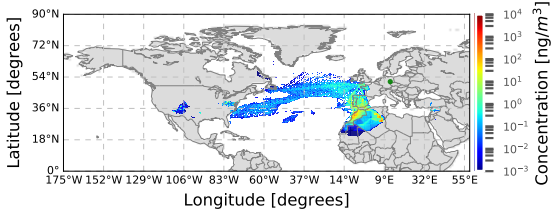
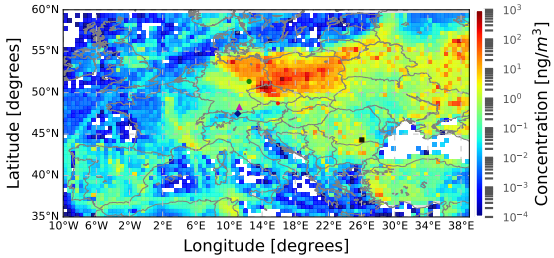
Discussion paper



(a) Pillersdorf, 02 April, 06:00, layer L1

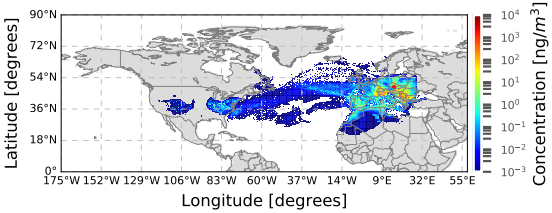


(b) Leipzig, 31 March, 18:00, layer corresponding to L1

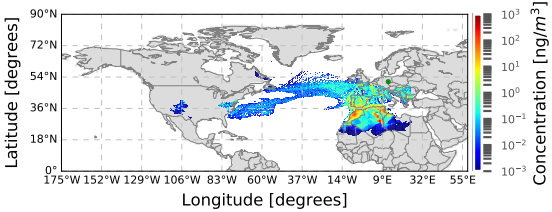


(c) Pillersdorf, 02 April, 06:00, layer L1, zoomed

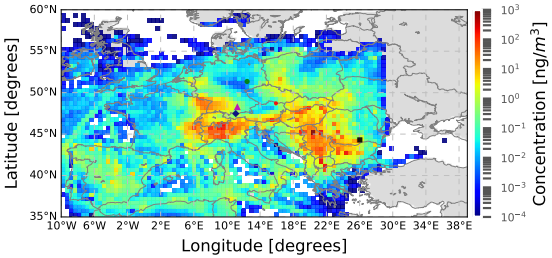
Fig. 6. Relative distributions of SO2 sources for Pillersdorf layer L1 (a), Leipzig (b); zoomed distribution for Pillersdorf layer L1 (c).



(a) Pillersdorf, 02 April, 06:00, layer L2

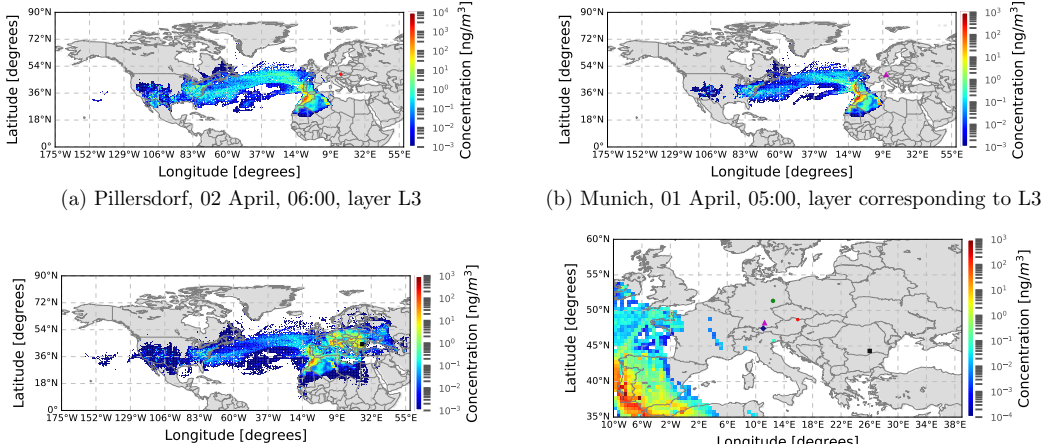


(b) Leipzig, 31 March, 23:00, layer corresponding to L2



(c) Pillersdorf, 02 April, 06:00, layer L2, zoomed

Fig. 7. Relative distributions of SO2 sources for Pillersdorf layer L2 (a), Leipzig (b); zoomed distribution for Pillersdorf layer L2 (c).



(a) Pillersdorf, 02 April, 06:00, layer L3

(b) Munich, 01 April, 05:00, layer corresponding to L3

(c) Bucharest, 03 April, 13:00, layer corresponding to L3

(d) Pillersdorf, 02 April, 06:00, layer L3, zoomed

Fig. 8. Relative distributions of SO2 sources for Pillersdorf layer L3 (a), Munich (b), Bucharest (c); zoomed distribution for Pillersdorf layer L3 (d).

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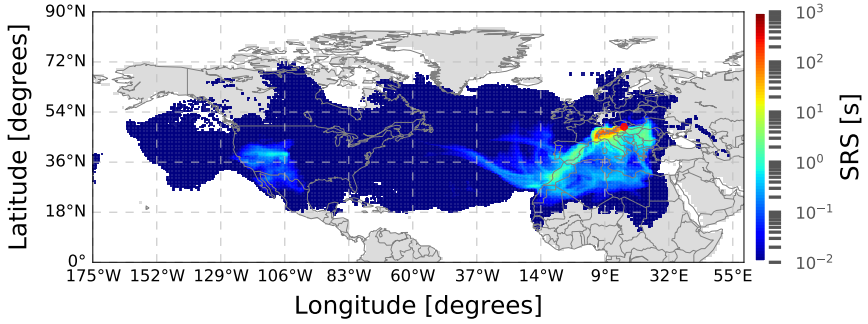
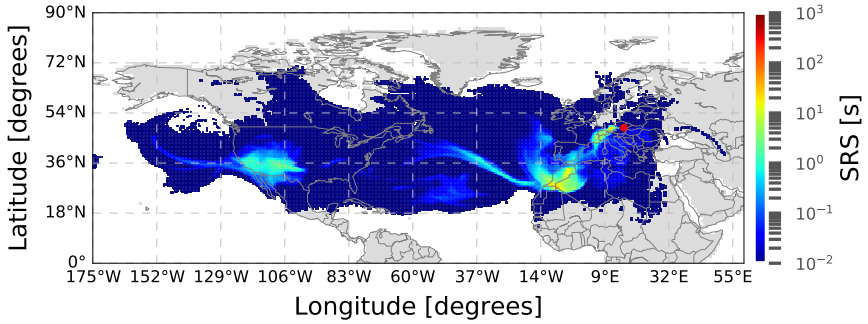


(a) Pillersdorf, 04 April, 12:00, layer L1



(b) Pillersdorf, 04 April, 12:00, layer L2

Fig. 9. Source-receptor sensitivity for layer L1 (a) and L2 (b), Pillersdorf, 04 April, 12:00

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Discussion paper

Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-1155-AC3, 2019

Atmospheric Chemistry and Physics Discussions

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

Camelia Talianu and Petra Seibert

camelia.talianu@gmail.com

Received and published: 25 March 2019

Dear editor,

Here are some technical corrections from the authors. Comment 1 to Comment 10 are minor language corrections. For maps in the Figures 5, 6, 9, 10, 11, 12 and 15, the color of the continents was changed from etopo color style to gray style (continents) and white (oceans) to make the maps more clear (Comment 11). Also included are the changes due to moving the GIOVANNI maps to the Supplement. In the following "AuthCM" represents the author's changes to the manuscript. Page and line number refer to the page and line number in the version submitted for discussion.

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Discussion paper

Technical corrections:

Comment 1.

AuthCM:
Page 2, Line 21
Changed in text: "The study was based on" to
"The study is based on"

Page 2, Line 22 - Line 25
Rephrased text to
“to assess the relation between the excess with respect to monthly averaged values observed in the in situ measurements of SO_2, O_3, $PM_{2.5}$ and PM_{10} at the Austrian air quality background station Pillersdorf at the beginning of Apr 2014 with aerosols layers observed in lidar measurements at the closest EARLINET stations around Austria and with tropospheric sulfate aerosols as found in Copernicus Atmosphere Monitoring Service (CAMS) products (CAMS, 2018)”

Page 2, Line 27
Changed in text: "to estimate the sulfate aerosols potential sources." to
"to estimate the potential sources of sulfate aerosols"

Comment 2.

AuthCM:
Page 3, Line 10
Changed in text: "in the format HH:mm, H being the hour and m the minutes" to
"in the format HH:mm, HH being the hour and mm the minutes"

Page 3, Line 13
Added: “In the plots, the stations are represented as: Pillersdorf (red circle), Leipzig (green circle), Munich (magenta triangle), Garmisch (blue rhombus), Bucharest (black square).”

Page 3, Line 17
Changed in text: "daily mean concentration and the maximum value per day of half-an hour averaged concentrations for" to
"daily mean concentration and the maximum half-hour mean value per day for"

Page 3, Line 19
"averaged" was changed with "mean"

Comment 3.

AuthCM:
Page 4, Line 30
Changed in text: "FLEXPART and FLEXTRA models were used in this paper for atmospheric transport modelling." to
"In this paper, the models FLEXPART and FLEXTRA were used for atmospheric transport modelling."

Comment 4.

AuthCM:
Page 5, Line 3 - Line 4
Changed in text: "parcels by mean winds, ignoring turbulence and convection, and do

not represent" to
"parcels by mean winds, ignoring turbulence and convection, and do not provide"

Page 5, Line 6
Changed “x" with “$\times$”

Comment 5.

AuthCM:
Page 8, Line 4
Changed in text: "“total aerosols" (sum of all species defined in CAMS data) (a), for sulfate (b) and for dust (c)." to
"“total aerosols" (sum of all species defined in CAMS data), for sulfate and for dust."

Page 8, Line 8
Changed in text: "for Munich (a), Leipzig (b) and Bucharest (c)." to
"for Munich, Leipzig and Bucharest."

Page 8, Line 21
Changed in text: "Fig. 5 for 00:00 (a), 06:00 (b), 12:00 (c) and 18:00 (d)." to
"Fig. 5 for 00:00, 06:00, 12:00 and 18:00."

Comment 6.

AuthCM:
Page 9, Line 2 - Line 3
Changed in text: "for the layers L1 (a), L2 (b), L3 (c) and total column (d)" to
"for the layers L1, L2, L3 and total column"

Page 9, Line 34
Changed in text: "(not shown in this paper)" to
"(not shown)"

Comment 7.

AuthCM:
Page 10, Line 11 - Line 17
Changed in text "transverse" to
"traverse"

Page 10, Line 19
Changed in text "at Pillersdorf: Southern and" to
"at Pillersdorf, namely Southern and"

Comment 8.

AuthCM:
Page 11, Line 7
Changed in text "Garmisch-Partenkirchen, Pillersdorf and Bucharest" to
"Garmisch-Partenkirchen, Pillersdorf and Bucharest, and"

Page 11, Line 8
Changed in text: "the adsorption of the SO_2 on the dust mineral oxides compounds." to
"the adsorption of the SO_2 on oxides contained in the mineral dust."

Comment 9.

AuthCM:
Page 22, Figure 7
Changed in caption: "log(range corrected signal)" to
"Logarithm of the range corrected signal"

Comment 10.

AuthCM:
Changed in text: "Apr" to "April" and "Mar" to "March"

Comment 11.

AuthCM:
Changed color of maps to gray (continents) and white (oceans) in the Figures 5, 6, 9, 10, 11, 12 and 15.

Technical corrections: content moved to Supplement

AuthCM:
Page 11, Line 10 - Line 13: moved to Supplement.

Changes in text, in the Supplement: "Monthly-averaged maps of column mass density for sulfate are available from EarthData NASA GIOVANNI (NASA,2018) online data system" to

"Time averaged maps of sulfate column mass density, monthly, are available from EarthData NASA GIOVANNI online data system (NASA,2018)."

Page 12, Line 8 - Line 10: acknowledgement to GIOVANNI moved to Supplement.

Moved Fig. 16 to Supplement.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1155, 2018.

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Discussion paper

Analysis of Sulfate Aerosols over Austria: A Case Study

Camelia Talianu[1, 2] and Petra Seibert[1]

[1]Institute of Meteorology, University of Natural Resources and Life Sciences, Vienna, Austria
[2]National Institute of R&D for Optoelectronics, Magurele, Romania

Correspondence: Camelia Talianu (camelia.talianu@boku.ac.at)

Dear editor,

Here are the changes between the version submitted for discussion and the revised version, updated as result of the referee comments and author supplementary comments. No other changes were done.

In the following: for “Changes in the paper", the page and line number refer to the page and line number in the latexdiff version; for “Paper supplement" (text added in the Supplement, added in the revised version) the page and line number refer to the page and line number in the supplement file.

The reason of each change is given in the format “ACx: Comment xx: AuthCM: ccc” where

- AC1: ’answer_acp-2018-1155-RC1’, Camelia Talianu, 25 Mar 2019 (link: answer to comments from Referee 1)
- AC2: ’answer_acp-2018-1155-RC2’, Camelia Talianu, 25 Mar 2019 (link: answer to comments from Referee 2)
- AC3: ’Author comments: acp-2018-1155’, Camelia Talianu, 25 Mar 2019 (link: author comments)

The RefC1 (RefC2) is a copy of RefC for referee 1 (2) from AC1 (AC2); AuthR and AuthCM are the corresponding author response and author changes in the manuscripts, also copies from AC1, AC2, AC3, respectively.

Changes in the paper

Complete list of changes for the paper, as shown in the latexdiff file.

1. Page 1, Line 1: change resulted from AC3: Comment 10.
   AuthCM: Changed in text: "Apr" to "April"

2. Page 2, Lines 2–3: change resulted from AC2: Comment 1.
   RefC2: 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."
   AuthR: This reference was added to the text and it was included in References list.
   AuthCM: Added "; worldwide in situ observations of refractory PM_1 chemical composition have shown that the sulfate contribution may reach more than 50% of aerosol mass, depending on the location (Zhang et al., 2007)."
   Added reference (Zhang et al., 2007).

3. Page 2, Line 17: change resulted from AC2: Comment 2.
   RefC2: 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."
   AuthR: Added sentence to the text, referencing also the paper.
   AuthCM: "A recent review of SO_2 sources worldwide can be found in (Yang et al., 2017)."

4. Page 2, Line 20: change resulted from AC2: Comment 29.
   RefC2: Page 2, Line 17: "what do you mean by "key properties"? Optical? Physical? Be more specific."
   AuthR: "key properties" means "optical and chemical properties".
   AuthCM: "The key properties" was changed to "The optical, physical and chemical properties"

5. Page 2, Lines 22–25: change resulted from AC2: Comment 3.
   RefC2: 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."
   AuthCM: added references AEROCOM project, IPCC AR5 for cooling effects of sulfate aerosol and added text: "The direct radiative effects are strongly correlated to the emission sources, while the indirect effects are correlated to both emission sources and cloud cover (Déandreis et al., 2012) (Yang et al., 2017)."

6. Page 2, Line 29: change resulted from AC3: Comment 10.
   AuthCM: Changed in text: "Apr" to "April"

7. Page 2, Lines 30–32: change resulted from AC3: Comment 1.
   AuthCM: Rephrased text: "to assess the relation between the excess with respect to monthly averaged values observed in the in situ measurements of SO_2, O_3, $PM_{2.5}$ and PM_{10} at the Austrian air quality background station Pillersdorf at the beginning of Apr 2014 with aerosols layers observed in lidar measurements at the closest EARLINET stations around Austria and with tropospheric sulfate aerosols as found in Copernicus Atmosphere Monitoring Service (CAMS) products (CAMS, 2018)"

8. Page 2, Line 33: change resulted from AC3: Comment 1.
   AuthCM: Changed in text: "to estimate the sulfate aerosols potential sources." to "to estimate the potential sources of sulfate aerosols"

9. Page 3, Line 1: change resulted from AC3: Comment 1.
   AuthCM: Changed in text: "The study was based on" to "The study is based on"

10. Page 3, Line 3: change resulted from AC2: Comment 5.
    RefC2: Page 2, Line 29 (Introduction): "please include a reference for NATALI aerosol typing model."
    AuthR: Added reference for NATALI aerosol typing.

AuthCM: "... and NATALI aerosol-typing model, and atmospheric transport modeling." is replaced by "... and NATALI aerosol-typing model (Nicolae et al., 2018), and atmospheric transport modeling."

11. Page 3, Lines 9–12: change resulted from AC1: Comment 4. Part B.

RefC1: "P10 / L27 & 28: "I am still a bit skeptical about the long-range transport of pollutants - there would be a significant dilution factor . . .! Unless there are major sources emitting ? An indication of such sources would make your findings more convincing."

AuthR: Flexpart simulates not only the transport due to the large-scale winds but also turbulent diffusion and mixing by subgrid-scale mesoscale motions (A. Stohl et al., 2005). Furthermore, it has implemented the treatment of all loss processes, including dry and wet deposition of gases or aerosols, gravitational settling of particles (S. Eckhardt et al., 2017). Flexpart also has implemented a deep convection scheme. Comprehensive validations of Flexpart were performed for intercontinental air pollution transport, see e.g. [A. Stohl et al Atmos. Environ., 32, 4245–4264, 1998], [A. Stohl and T.Trickl, Geophys. Res., 104, 30,445–30,462, 1999], [N.Kristiansen et al., Geophys. Re. Lett. 42, 588-596, doi 10.1002/2014GL062307, 2015]. Thus, there is no reason to doubt the results.

For the case study presented in this paper, the major sources of SO_2 are coal power plants and other industrial facilities (refineries, chemical industry, etc), present in the regions mentioned: Central Europe 'Black triangle', industrialized cities from Morocco, Eastern part of US (e.g. Ohio, New Jersey), Southeastern part of US (e.g. Louisiana, Alabama). An exhaustive list of US sources is mentioned in the report "U.S. EPA 2014 NEI Version 1.0" [https://www.epa.gov/sites/production/files/2017-04/documents/2014neiv1_profile_final_april182017.pdf] A recent study on SO_2 sources worldwide is published in (Y. Yang et al, 2017), which was added to the references.

AuthCM: added references (S. Eckhardt et al., 2017) and (A. Stohl et al., 2005) for Flexpart

12. Page 3, Lines 14–19: change resulted from AC2: Comment 4.

RefC2: 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."

AuthR: In the last decade, the synergy of the in situ, remote sensing data and models was used in more atmospheric studies related to long-range transported aerosols and estimation of their potential sources (see for example A. Pappayanis et al. (Sci Total Environ. 2014;500-501:277-94. doi:10.1016/j.scitotenv.2014.08.101, 2014 - C.T. coauthor), D. Nicolae et al. (2013 - C.T. coauthor, Ansmann et al 2018, Eckhardt et al 2008 - P.S. coauthor, Cazacu et al 2012 - C.T. coauthor], [Sauvage et al 2017], [Chalbot et al 2017],[D.G. Kaskaoutis et al., 2012]).

However, to our best knowledge, there have been no studies combining CAMS-based aerosol data with remote sensing and in situ measurements and transport models.

AuthCM: Added: "The synergy of the in situ, remote sensing data and models was used in more atmospheric studies related to long-range transported aerosols and estimation of their potential sources; see for example (Papayannis et al., 2014) for dust, (Nicolae et al., 2013) and (Ansmann et al., 2018) for fires, (Eckhardt et al., 2008) and (Cazacu et al.,

2012) for volcanic ash, (Sauvage et al., 2017), (Chalbot et al., 2013) and (Kaskaoutis et al., 2012) for anthropogenic aerosols. However, to our best knowledge, there have been no studies combining CAMS-based aerosol data with remote sensing, in situ measurements and transport models. The assimilation of ground-based remote sensing measurements in CAMS is a long-term goal."

13. Page 3, Line 23: change resulted from AC3: Comment 2.
    AuthCM: Changed in text: "in the format HH:mm, H being the hour and m the minutes" to
    "in the format HH:mm, HH being the hour and mm the minutes"

14. Page 3, Line 24: change resulted from AC1: Comment 5.
    RefC1: "Figs. 4 & 13 - Key for variables needs to be enlarged P8 / L20 - Height of layers 'amsl' or 'AGL' (also in tables)"
    AuthR: The recommended corrections were done in the two figures. AGL was added to text and to the caption for the two figures. No AGL added to the tables, I think it is enough to mention in the text and to add to figure.
    AuthCM: added "(AGL)."

15. Page 3, Lines 26–27: change resulted from AC3: Comment 2.
    AuthCM: Added: "In the plots, the stations are represented as: Pillersdorf (red circle), Leipzig (green circle), Munich (magenta triangle), Garmisch (blue rhombus), Bucharest (black square)."

16. Page 3, Lines 30–32, Page 4, Lines 1–2: change resulted from AC2: Comment 6.
    RefC2: 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?"
    AuthR:
    AuthCM: Added: "Pillersdorf (315 m) is located in hilly terrain in the northeastern part of Austria, around 60 km north from Vienna. The station is a part of the national background monitoring network and an EMEP background monitoring station. The surroundings are mostly forests and agricultural areas far from strong anthropogenic sources. Austria belongs to the midlatitude climate belt, in the transition between maritime and continental climate, and the weather is dominated mostly by travelling highs and lows. The station provides:"

17. Page 4, Line 3: change resulted from AC3: Comment 2.
    AuthCM: Changed in text: "daily mean concentration and the maximum value per day of half-an hour averaged concentrations for" to
    "daily mean concentration and the maximum half-hour mean value per day for"

18. Page 4, Line 6: change resulted from AC3: Comment 2.
    AuthCM: "averaged" was changed with "mean"

19. Page 4, Lines 8–11: change resulted from AC1: Comment 2.
RefC1: "The authors should avoid to mention Trade Names or direct references to companies and commercial instruments, unless absolutely necessary for the understanding of the methods deployed."
AuthR: The trade names of the instruments were deleted from the text. Please note that PollyXT and RALI are the names of the instruments, as used by EARLINET to identify the instruments at the corresponding station, not trade names.
AuthCM: "a Thermo Scientific Model 43i SO_2 Analyzer" was changed to "a SO_2 analyzer"
"Optical Particle Counter GRIMM Dust Monitor Model EDM180," was changed to "optical particle counter"
"a Thermo Environmental Instruments Ozone Analyzer, model TEI 49C," was changed to "an ozone analyzer,"

20. Page 4, Line 22: change resulted from AC1: Comment 2.
RefC1: "The authors should avoid to mention Trade Names or direct references to companies and commercial instruments, unless absolutely necessary for the understanding of the methods deployed."
AuthCM: "Jenoptik ceilometers CHM15kx" was changed to "ceilometers"

21. Page 5, Line 16: change resulted from AC3: Comment 3.
AuthCM: Changed in text: "FLEXPART and FLEXTRA models were used in this paper for atmospheric transport modelling." to
"In this paper, the models FLEXPART and FLEXTRA were used for atmospheric transport modelling."

22. Page 5, Lines 23–26: change resulted from AC2: Comment 9.
RefC2: 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?"
AuthR: A more detailed explanation has been introduced. FLEXPART is not tuned or iterated.
AuthCM: Added after "...or gridded sources"
"(Seibert and Frank, 2004). The model ingests ECMWF 3D meteorological fields and solves the equations for transport, turbulent diffusions and other relevant processes in a Lagrangian framework (Stohl et al., 1998) (Pisso et al., 2019). The sensitivity of a receptor concentration to potential sources is obtained directly as the model output in the case of a backward run (Seibert and Frank, 2004) (Eckhardt et al., 2017)."

23. Page 5, Line 28: change resulted from AC3: Comment 4.
AuthCM: Changed in text: "parcels by mean winds, ignoring turbulence and convection, and do not represent" to
"parcels by mean winds, ignoring turbulence and convection, and do not provide"

24. Page 5, Line 30: change resulted from AC3: Comment 4.
AuthCM: Changed "x" with "$\times$"

25. Page 6, Line 2: change resulted from AC2: Comment 30.
RefC2: Page 5, Line 11: "omit the word "Ref." before citing a reference. That occurs all through the manuscript, please

check."

AuthR: OK.

AuthCM: removed “Ref. “

26. Page 6, Lines 3–5: change resulted from AC2: Comment 10.

RefC2: 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."

AuthR: Changed "pure aerosol" to "typical aerosol".

AuthCM: All occurrences of “pure aerosol” changed to “typical aerosol”; added in text "(called "pure aerosol" in the reference)".

27. Page 6, Lines 25–29: change resulted from AC2: Comment 12.

RefC2: 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."

AuthR: Done, see changes to manuscript.

AuthCM: changed to “The aerosol layers are identified from the lidar measurements with the gradient method, applied to the RCS profiles (Belegante et al., 2014) (Nicolae et al., 2018). The gradient method is based on the identification of the peaks/valleys from the first derivative applied to the vertical profiles. If two consecutive layers are very close (less than 100 m), these layers are merged into one layer. Also, if the signal to noise ratio in the layer is lower than a threshold (here set to 5), the layer is discarded.”

28. Page 6, Line 30: change resulted from AC2: Comment 30.

RefC2: Page 5, Line 11: "omit the word “Ref.” before citing a reference. That occurs all through the manuscript, please check."

AuthR: OK.

AuthCM: removed “Ref. “

29. Page 7, Line 2: change resulted from AC2: Comment 31.

RefC2: Page 6, Line 6: "“The values of the CAMS quantities”: please use a more specific term, instead of “quantities”."

AuthR: "the CAMS quantities” were specified in text at page 4, line 22 - line 28.

AuthCM: “... the CAMS quantities” was changed to “... the CAMS products (mixing ratios, temperature, specific humidity, etc)”

30. Page 7, Line 9: change resulted from AC2: Comment 13b.

RefC2: Page 6, Lines 11–15 (Methods): "What is the objective criteria for “significant excess”?"

AuthR: The criterion for "significant excess" is 50% above the averaged values for 30 days.

AuthCM: changed "...is identified, " to "...is identified (values exceed by 50% the averaged values for 30 days),"

31. Page 7, Lines 11–12: change resulted from AC3: Comment 10.
    AuthCM: Changed in text: "Apr" to "April" and "Mar" to "March"

32. Page 7, Lines 21–23: change resulted from AC2: Comment 12.
    RefC2: Page 6, Lines 1–2 (Methods): " Also, please clarify that you applied the gradient method both for Lidar and CAMS profiles."
    AuthR: Done, see changes to manuscript.
    AuthCM: changed to: “The layers for the event at the in situ site are then determined by applying the same gradient method as for lidar data processing, but applied to the altitude profiles of aerosol concentrations. The concentrations are computed by multiplying the CAMS mixing ratios and the air density.”

33. Page 7, Lines 28–29: change resulted from AC2: Comment 19.
    RefC2: Figure 4: "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."
    AuthR: We added a sentence on boundary-layer heights.
    AuthCM: Added: “During the period under investigation, with low wind speeds and mostly clear skies, the boundary-layer height varied at Pillersdorf from less than 100 m at night to about 1500 m in the afternoon.”

34. Page 8, Line 5: change resulted from AC2: Comment 30.
    RefC2: Page 5, Line 11: "omit the word “Ref.” before citing a reference. That occurs all through the manuscript, please check."
    AuthR: OK.
    AuthCM: removed “Ref. “

35. Page 8, Line 11: change resulted from AC2: Comment 14.
    RefC2: 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.""
    AuthR: "release" was changed with "receptor".
    AuthCM: "The receptor is set to the location"

36. Page 8, Lines 27–29: change resulted from AC3: Comment 10.
    AuthCM: Changed in text: "Apr" to "April" and "Mar" to "March"

37. Page 8, Line 33: change resulted from AC3: Comment 5.
    AuthCM: Changed in text: "“total aerosols" (sum of all species defined in CAMS data) (a), for sulfate (b) and for dust (c)." to
    "“total aerosols" (sum of all species defined in CAMS data), for sulfate and for dust."

38. Page 9, Line 1: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

39. Page 9, Line 4: change resulted from AC3: Comment 5.
AuthCM: Changed in text: "for Munich (a), Leipzig (b) and Bucharest (c)." to "for Munich, Leipzig and Bucharest."

40. Page 9, Line 6: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

41. Page 9, Line 8: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

42. Page 9, Line 17: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

43. Page 9, Line 17: change resulted from AC3: Comment 5.
AuthCM: Changed in text: "Fig. 5 for 00:00 (a), 06:00 (b), 12:00 (c) and 18:00 (d)." to "Fig. 5 for 00:00, 06:00, 12:00 and 18:00."

44. Page 9, Line 22: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

45. Page 9, Line 24: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

46. Page 9, Line 29: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

47. Page 9, Lines 33–34: change resulted from AC3: Comment 6.
AuthCM: Changed in text: "for the layers L1 (a), L2 (b), L3 (c) and total column (d)" to "for the layers L1, L2, L3 and total column"

48. Page 9, Line 34: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

49. Page 10, Lines 2–3: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April" and "Mar" to "March"

50. Page 10, Lines 6–8: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April" and "Mar" to "March"

51. Page 10, Line 13: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

52. Page 10, Line 16: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

53. Page 10, Lines 26–27: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April" and "Mar" to "March"

54. Page 10, Line 30: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Mar" to "March"

55. Page 10, Line 32: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Mar" to "March"

56. Page 10, Line 32: change resulted from AC3: Comment 6.
AuthCM: Changed in text: "(not shown in this paper)" to
"(not shown)"

57. Page 11, Line 3: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

58. Page 11, Line 4: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

59. Page 11, Line 8: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

60. Page 11, Lines 9–15: change resulted from AC3: Comment 7.
AuthCM: Changed in text "transverse" to
"traverse"

61. Page 11, Line 18: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

62. Page 11, Lines 28–29: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

63. Page 12, Line 5: change resulted from AC3: Comment 8.
AuthCM: Changed in text "Garmisch-Partenkirchen, Pillersdorf and Bucharest" to
"Garmisch-Partenkirchen, Pillersdorf and Bucharest, and"

64. Page 12, Line 6: change resulted from AC3: Comment 8.
AuthCM: Changed in text: "the adsorption of the SO_2 on the dust mineral oxides compounds." to "the adsorption of the SO_2 on oxides contained in the mineral dust."

65. Page 12, Lines 9–12: change resulted from AC2: Comment 27.
RefC2: Figure 16: "it does not contribute significantly to the discussion. I suggest to exclude this figure, or to move it to the supplementary material."
AuthR: OK
AuthCM: Text was moved to the Supplement of the paper.

66. Page 12, Line 14: change resulted from AC3: Comment 10.
AuthCM: Changed in text: "Apr" to "April"

67. Page 12, Lines 27–30: change resulted from AC1: Comment 4 Part A.
RefC1: "P10 / L27 & 28: - 'No contributions from Europe are seen for these layers.' This may be true for the period in April, as there may not have been any deep convection. However, it would be interesting to also study a summer period with strong convective activity over Central Europe (obviously, in a separate paper !)."
AuthR: The summer periods for the years 2014–2017 are under study, and a paper is under preparation.
AuthCM: Added text: "The spring period studied in this paper is characterized by low, if any, deep convection. For the summer period, one expects however to have a strong convective activity over Central Europe. A study of the summer periods for the years 2014–2017 for the same region was also performed; the results will be presented in a separate paper."

68. Page 13, Lines 10–12: change resulted from AC2: Comment 27.
RefC2: Figure 16: "it does not contribute significantly to the discussion. I suggest to exclude this figure, or to move it to the supplementary material."
AuthR: OK
AuthCM: Text was moved to the Supplement of the paper.

69. Page 19, Figure 2: change resulted from AC3: Comment 10.
AuthCM: Changed in caption: "Apr" to "April" and "Mar" to "March"

70. Page 20, Figure 3: change resulted from AC3: Comment 10.
AuthCM: Changed in caption: "Apr" to "April" and "Mar" to "March"

71. Page 21, Figure 4: change resulted from AC3: Comment 10, from AC1: Comment 5 and from AC2: Comment 19.
AuthCM: Changed in caption: "Apr" to "April"
RefC1: "Figs. 4 & 13 - Key for variables needs to be enlarged P8 / L20 - Height of layers 'amsl' or 'AGL' (also in tables)"

AuthR: The recommended corrections were done in the two figures. AGL was added to text and to the caption for the two figures.

AuthCM: Changed in caption:"Altitude are given in km AGL."

RefC2: 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)."

AuthR: We have improved the figure. We also added in the caption of Fig. 4 the UTC–local time difference.

AuthCM: Changed fonts in the legend of Fig. 4; modified caption: changed “aerosol” to “total aerosol”; added “Local time is UTC+2.”.

72. Page 22, Figure 5: change resulted from AC3: Comment 10, from AC3: Comment 11 and from AC2: Comment 20.

AuthCM: Changed in caption: "Apr" to "April"

AuthCM: Changed color of maps to gray (continents) and white (oceans) in the figures.

RefC2: 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 un- derstandability. I suggest that you keep only 3-4 representative altitudes (low, medium, high)."

AuthR: The figure was changed, keeping only the trajectories passing over the lidar stations involved in this analysis.

AuthCM: New Figure 5

73. Page 23, Figure 6: change resulted from AC3: Comment 10, from AC3: Comment 11 and from AC2: Comment 21.

AuthCM: Changed in caption: "Apr" to "April"

AuthCM: Changed color of maps to gray (continents) and white (oceans) in the figure.

RefC2: "Figures 6, 10c, 11c, 12d: you must indicate the locations of the monitoring stations in the maps."

AuthR: The figures were changed adding the location of the monitoring stations.

AuthCM: New Figure 6

74. Page 24, Figure 7: change resulted from AC3: Comment 9 and from AC2: Comment 32.

AuthCM: Changed in caption: "log(range corrected signal)" to "Logarithm of the range corrected signal"

RefC2: Figure 7: "scales are illegible."

AuthCM: Figure 7 The fonts for scales were increased to be more legible.

75. Page 26, Figure 9: change resulted from AC3: Comment 10, from AC3: Comment 11 and AC2: Comment 34.

AuthCM: Changed in caption: "Apr" to "April"

AuthCM: Changed color of maps to gray (continents) and white (oceans) in the figures.

RefC2: Figure 9: "what are the units for the color map? In addition, the units of longitude should be “degrees”, and not “degrees E”."

AuthR: The units for color map are seconds. The units of longitude and latitude were corrected.

AuthCM: Added units for color map and changed units of longitude and latitude in the figures.

76. Pages 27–29, Figures 10, 11, 12: change resulted from AC3: Comment 11, from AC2: Comment 34, from AC2: Comment 21 and from AC2: Comment 22.

AuthCM: Changed color of maps to gray (continents) and white (oceans) in the figures.

RefC2: "In addition, the units of longitude should be “degrees”, and not “degrees E”.."

AuthR: The units of longitude and latitude were corrected.

AuthCM: Changed units of longitude and latitude in the figures.

RefC2: "Figures 6, 10c, 11c, 12d: you must indicate the locations of the monitoring stations in the maps."

AuthR: The figures were changed adding the location of the monitoring stations.

AuthCM: New figures 10c, 11c, 12d

RefC2: 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?"

AuthR: There was a bug in superposing the concentration distribution on map for the zoomed distribution (only). The figures 10c, 11c and 12d were replaced with figures with correct data. There is a good agreement between model and ground based measurements, as can be seen from Fig. 1 and Fig. 10c.

AuthCM: the figures 10c, 11c and 12c were replaced.

77. Page 30, Figure 13: change resulted from AC3: Comment 10 and from AC1: Comment 5.

AuthCM: Changed in caption: "Apr" to "April"

RefC1: "Figs. 4 & 13 - Key for variables needs to be enlarged P8 / L20 - Height of layers ‘amsl‘ or ‘AGL‘ (also in tables)"

AuthR: The recommended corrections were done in the two figures. AGL was added to text and to the caption for the two figures.

AuthCM: Changed in caption:"Altitude are given in km AGL."

78. Page 31, Figure 14: change resulted from AC3: Comment 10.

AuthCM: Changed in caption: "Apr" to "April"

79. Page 32, Figure 15: change resulted from AC3: Comment 10, from AC3: Comment 11 and from AC2: Comment 34.

AuthCM: Changed in caption: "Apr" to "April"

AuthCM: Changed color of maps to gray (continents) and white (oceans) in the figures.

RefC2: "what are the units for the color map? In addition, the units of longitude should be “degrees”, and not “degrees E”."

AuthR: The units for color map are seconds. The units of longitude and latitude were corrected.

AuthCM: Added units for color map and changed units of longitude and latitude in the figures.

80. Page 32, Figure 16: change resulted from AC3: Technical corrections: content moved to Supplement.
AuthCM: Moved Fig. 16 to Supplement.

81. Page 33, Table 1: change resulted from AC3: Comment 10.
AuthCM: Changed in caption: "Apr" to "April"

82. Page 34, Table 2: change resulted from AC3: Comment 10.
AuthCM: Changed in caption: "Apr" to "April"

83. Page 35, Table 3: change resulted from AC3: Comment 10.
AuthCM: Changed in caption: "Apr" to "April"

84. Page 36, Table 4: change resulted from AC3: Comment 10.
AuthCM: Changed in caption: "Apr" to "April"

85. Page 37, Table 5: change resulted from AC3: Comment 10.
AuthCM: Changed in caption: "Apr" to "April"

86. Page 38, Table 6: change resulted from AC3: Comment 10.
AuthCM: Changed in caption: "Apr" to "April"

Paper supplement

A paper supplement is added to the revised version. List of added text and figures in the supplement:

1. Page 1: added Figure S1 as resulted from AC2: Comment 8.
RefC2: 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."
AuthR: OK.
AuthCM: A map has been added to the Supplement.

2. Pages 2–4: added Figure S2, Figure S3, Figure S4 as resulted from AC1: Comment 3 and from AC2: Comment 26.
RefC1: "Since the trajectories in Fig.5 indicate sources processes from almost all over Europe (understandable, especially in the lower & mid levels), but also very distant sources (mostly in elevated layers), the authors should show the relevant meteorological maps for the study period (850, 700, 500, 300 & 200 or 250 hPa circulation) to provide physical evidence for 'conflicting' circulations in some of the layers, and especially for the 'outlying regions'. Of course, FLEXTRA ingests the upper air data from ECMWF, but a cross-verification with 'real meteorological data' will make the cases more convincing."

AuthR: There is no reason to not trust the FLEXTRA calculations. However, seeing weather maps may help to understand the prevailing synoptic pattern. Therefore, a few weather maps are now provided in the Supplement.

AuthCM: Added weather maps as supplement.

RefC2: 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."

AuthR: The meteorological maps will be added to the Supplement of the paper.

AuthCM: meteorological maps in the supplement.

3. Page 5: Moved Figure 16 (GIOVANNI maps) and the corresponding text and aknowledgement from paper to supplement as resulted from AC2: Comment 27.

RefC2: Figure 16: "it does not contribute significantly to the discussion. I suggest to exclude this figure, or to move it to the supplementary material."

AuthR: OK

AuthCM: Fig. 16 was moved to the Supplement of the paper.

Analysis of Sulfate Aerosols over Austria: A Case Study

Camelia Talianu[1, 2] and Petra Seibert[1]

[1]Institute of Meteorology, University of Natural Resources and Life Sciences, Vienna, Austria
[2]National Institute of R&D for Optoelectronics, Magurele, Romania

Correspondence: Camelia Talianu (camelia.talianu@boku.ac.at)

Abstract. An increase of the sulfate aerosols observed in the period 01 – 06 ~~Apr~~ April 2014 over Austria is analyzed using in situ measurements at an Austrian air quality background station, lidar measurements at the closest EARLINET stations around Austria, CAMS near-real-time data and particle dispersion modelling using FLEXPART, a Lagrangian transport model. In-situ measurement of SO_2, $PM_{2.5}$, PM_{10} and O_3 were performed at the air quality background station Pillersdorf, Austria (EMEP station AT30, 48°43'N, 15°55'E). A CAMS aerosol mixing ratios analysis for Pillersdorf and the lidar stations Leipzig, Munich, Garmisch, Bucharest indicates the presence of an event of aerosol transport, with sulfate and dust as principal components. For the sulfate layers identified at Pillersdorf from the CAMS analysis, backward and forward trajectory analyses were performed, associating lidar stations to the trajectories. The lidar measurements for the period corresponding to trajectory overpass of associated stations were analyzed, obtaining the aerosol layers, the optical properties and the aerosol types. The potential sources of transported aerosols were determined for Pillersdorf and the lidar stations using the source-receptor sensitivity computed with FLEXPART, combined with MACCity source inventory. A comparative analysis for Pillersdorf and the trajectory-associated lidar stations showed consistent aerosol layers, optical properties and types, and potential sources. A complex pattern of contributions to sulfate over Austria was found in this paper. For the lower layers (below 2000 m) of sulfate, it was found that the Central Europe was the main source of sulfate. Medium to smaller contributions come from sources in Eastern Europe, the Northwest Africa and Eastern US. For the middle-altitude layers (between 2000 m and 5000 m), sources from Central Europe (Northern Italy, Serbia, Hungary) contribute with similar emissions. Northwest Africa and Eastern US have also important contributions. For the high-altitude layers (above 5000 m), the main contributions come from Northwest Africa, but sources from Southern and Eastern US contribute also significantly. No contributions from Europe are seen for these layers. The methodology used in this paper can be used as a general tool to correlate measurements at in situ stations and EARLINET lidar stations around these in situ stations.

1 Introduction

Sulfate is one of the major aerosol components for particles with diameter smaller than 2.5 $\mu\mathrm{m}$ ($PM_{2.5}$), and for particles with diameter smaller than 10 $\mu\mathrm{m}$ (PM_{10}). Other components of the particulate matter (PM) are: organic carbon (OC), elemental

carbon (EC), nitrate, ammonia, mineral and sea salt. Sulfate normally accounts for about 10% to 30% of PM mass concentration (Stocker et al., 2013); worldwide in situ observations of refractory PM_1 chemical composition have shown that the sulfate contribution may reach more than 50% of aerosol mass, depending on the location (Zhang et al., 2007). More details about the mass concentration of these aerosol components from various rural and urban sites in Europe are given in the IPCC AR5 report (Stocker et al., 2013). The anthropogenic sulfate is produced mainly by oxidation of sulfur dioxide (SO_2), or produced by aqueous phase reactions, where O_3 and hydrogen peroxide act as important oxidants (Seinfeld and Pandis, 2006), or by adsorption of SO_2 on solid particles and subsequent reaction with adsorpted oxygen; the exact mechanism depends on several atmospheric factors (solar radiation, presence of catalysts, NO_x, temperature, relative humidity, etc). The adsorption is an important mechanism of sulfate production in urban atmosphere. Soot (elemental carbon) particles and semiconductor metal oxide particulates from mineral dust (e.g. Fe_2O_3, TiO_2) are potential surfaces for this process (Dupart et al., 2012). The primary precursor for sulfate in the troposphere is SO_2 emitted (Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, 2007) from:

- anthropogenic sources: major contribution from combustion of fossil fuel (about 72%) and small contribution from biomass burning (about 2%),
- natural sources: from dimethyl sulfide (DMS) emissions by marine phytoplankton (about 19%) and from volcano eruptions (about 7%).

A recent review of SO_2 sources worldwide can be found in (Yang et al., 2017).

Beside chemical processes, SO_2 is removed efficiently by dry deposition, while sulfate aerosol is removed from atmosphere by wet deposition (Seinfeld and Pandis, 2006). Tropospheric sulfate, mostly in the accumulation mode, has a lifetime estimated of one week (AeroCom, 2018). The ~~key~~ optical, physical and chemical properties of the sulfate are well defined (Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, 2007). Sulfate particles have a cooling effect by light scattering (AeroCom, 2018) (Stocker et al., 2013), they are very hygroscopic and therefore represent active cloud condensation nuclei, and they enhance absorption when deposited as a coating on elemental carbon. The direct radiative effects are strongly correlated to the emission sources, while the indirect effects are correlated to both emission sources and cloud cover (Déandreis et al., 2012) (Yang et al., 2017). As main component in the aerosols, sulfate can have an important contribution to the aerosol optical depth (AOD).

The purpose of this study is

- to assess the relation between the excess with respect to monthly averaged values observed in the in situ measurements of SO_2, O_3, $PM_{2.5}$ and PM_{10} at the Austrian air quality background station Pillersdorf ~~,~~ at the beginning of ~~Apr~~ April 2014 ~~, with tropospheric sulfate aerosols observed in Copernicus Atmosphere Monitoring Service (CAMS) products and aerosols~~ with aerosols layers observed in lidar measurements at the closest EARLINET stations around Austria and with tropospheric sulfate aerosols as found in Copernicus Atmosphere Monitoring Service (CAMS) products (CAMS, 2018)
- to estimate the ~~sulfate aerosols potential sources~~ potential sources of sulfate aerosols.

The study ~~was~~ is based on the synergy of the remote sensing instruments from European Aerosol Research Lidar Network (EARLINET) (Boesenberg et al., 2003), the ceilometer network of the German Meteorological Service (DWD) and in situ monitors, combined with CAMS products and NATALI aerosol-typing model (Nicolae et al., 2018), and atmospheric transport modeling. The ground-based remote sensing instruments and the CAMS products (assimilating satellite-based remote sensing data) are used to determine the properties of long-range transported aerosols and their vertical distribution. In-situ measurements of PM and trace gases provide local concentrations at the surface and at specific heights in the troposphere. Details about data collection are given in Sec. 2.1.

The back-trajectories analysis relates the aerosol mass loading changes at a receptor location to spatially-fixed sources, identifying the sources by a source-receptor matrix calculation (Seibert and Frank, 2004), (Eckhardt et al., 2017). In this paper, the analysis of the trajectories has been performed with FLEXTRA (Stohl et al., 1995) (FLEXTRA, 2018), while the estimation of the potential areas of aerosols' sources has been performed using the Lagrangian transport model FLEXPART (Stohl et al., 2005), (Stohl et al., 2010). A detailed description of the processing of the collected data and the subsequent analysis is given in Sec. 2.3, while the results and the discussion are presented in Sec. 3.

The synergy of the in situ, remote sensing data and models was used in more atmospheric studies related to long-range transported aerosols and estimation of their potential sources; see for example (Papayannis et al., 2014) for dust, (Nicolae et al., 2013) and (Ansmann et al., 2018) for fires, (Eckhardt et al., 2008) and (Cazacu et al., 2012) for volcanic ash, (Sauvage et al., 2017), (Chalbot et al., 2013) and (Kaskaoutis et al., 2012) for anthropogenic aerosols. However, to our best knowledge, there have been no studies combining CAMS-based aerosol data with remote sensing, in situ measurements and transport models. The assimilation of ground-based remote sensing measurements in CAMS is a long-term goal.

2 Methodology

The optical properties of the aerosol considered in this analysis are: backscatter coefficients, extinction coefficients, volume depolarization ratio, particle depolarization ratio (PDepR), lidar ratio (LR) and Ångström exponent (AE).

In this paper, all times are given as UTC times, in the format HH:mm, ~~H~~ HH being the hour and ~~m~~ mm the minutes. The altitudes are given as ground-level altitudes (AGL).

Whenever referring to measurements, the geographical name is used as indicator for the station location (e.g. Pillersdorf means Pillersdorf site, Leipzig means Leipzig lidar station). In the plots, the stations are represented as: Pillersdorf (red circle), Leipzig (green circle), Munich (magenta triangle), Garmisch (blue rhombus), Bucharest (black square).

2.1 Data collection

The in situ measurement of SO_2, $PM_{2.5}$, PM_{10} and O_3 were performed at the air quality background station Pillersdorf, Austria (EMEP station AT30, 48°43'N, 15°55'E) (Umweltbundesamt Austria, 2014)~~which~~. Pillersdorf (315 m) is located in hilly terrain in the northeastern part of Austria, around 60 km north from Vienna. The station is a part of the national background monitoring network and an EMEP background monitoring station. The surroundings are mostly forests and agricultural areas

far from strong anthropogenic sources. Austria belongs to the midlatitude climate belt, in the transition between maritime and continental climate, and the weather is dominated mostly by travelling highs and lows. The station provides:

- daily mean concentration and the maximum half-hour mean value per day ~~of half-an hour averaged concentrations~~ for SO_2
- daily mean concentration for $PM_{2.5}$ and PM_{10}
- maximum value per day of hourly ~~averaged~~ mean concentrations and maximum value per day of 8-hours ~~averaged~~ mean concentrations for O_3

The SO_2 measurements are performed with a ~~Thermo Scientific Model 43i~~ SO_2 ~~Analyzer~~analyzer, with a detection limit of 0.05 ppb, and a range up to 100 ppm. The $PM_{2.5}$ and PM_{10} measurements are performed with an ~~Optical Particle Counter GRIMM Dust Monitor Model EDM180,~~ optical particle counter with a precision of 0.1 $\mu g\,m^{-3}$. The O_3 measurements are performed with ~~a Thermo Environmental Instruments Ozone Analyzer, model TEI 49C,~~ an ozone analyzer, with a detection limit of 0.4 ppb and a range of 0.05 to 200 ppm.

The EARLINET lidar stations (Wandinger et al., 2016) used for this study are Garmisch-Partenkirchen (47.47°N, 11.06°E), Leipzig (51.35°N, 12.43°E) (both stations located in Germany), and Bucharest (44.35°N, 26.03°E, Romania). The two DWD ceilometer stations used are located in Munich (48.20°N, 11.45°E) and Schneefernerhaus (47.42 °N, 10.98°E). The following remote sensing devices are deployed:

- High spectral resolution lidar HSRL (Wandinger et al., 2016), located at Garmisch-Partenkirchen, Germany
- Portable Raman multispectral lidar system Polly^{XT} (Engelmann et al., 2016), having 8 channels including one water vapour channel and 2 depolarisation channels, located at Leipzig, Germany
- Raman multispectral lidar system RALI (Belegante et al., 2014), having 7 channels including one water vapour channel and one depolarization channel, located at Bucharest, Romania
- ~~Jenoptik ceilometersCHM15kx~~ceilometers (Wiegner and Geiß, 2012) at Munich and Schneefernerhaus, Germany

The measurements were done at the following wavelengths: 355 nm, 532 nm and 1064 nm for the elastic channels, 387 nm and 607 nm for the Raman channels, and 532 nm for the depolarization channel. For HSRL, the 313 nm channel was used. For ceilometers, the 1064 nm channel was used.

The lidar and the ceilometer measurements provide the vertical distributions of aerosols, retrieved from the range corrected signal (RCS, the preprocessed lidar/ceilometer signal corrected with squared range), and the vertical distributions of aerosol polarization, if the instrument is equiped with a polarization channel.

For the remote sensing sites Leipzig, Munich and Bucharest, the column-integrated AOD measurements for various wavelengths were taken from the AERONET sun/sky photometer measurements, the AERONET instruments being collocated with the lidar stations.

In this paper, products from CAMS, the Copernicus Atmosphere Monitoring Service (CAMS, 2018) of the European Earth Observation programme Copernicus were also used; it provides global reanalysis datasets for the period 2003 – 2012, and global near-real-time (NRT) datasets (Dee et al., 2011) for 2013 to present. These datasets were produced (Benedetti et al., 2009) using 4DVar data assimilation in CY42R1 of ECMWF's Integrated Forecast System (IFS), with 60 hybrid sigma/pressure (model) levels in the vertical, with the top level at 0.1 hPa. Atmospheric data are available on these levels and they are also interpolated to 25 pressure, 10 potential temperature and 1 potential vorticity level(s). "Surface or single level" data are also available.

For this analysis, the CAMS products for "Model levels" and "Surface level" from NRT "Atmospheric composition" dataset were selected for the times 00:00, 06:00, 12:00 and 18:00 for the analysis data and a step of 3 h for forecast data. The mixing ratios of dust, hydrophilic and hydrophobic black carbon, hydrophilic and hydrophobic organic matter and sulfate were retrieved from the lowest 31 model levels, which covers the tropospheric altitudes; temperature and specific humidity were also retrieved for the same model levels. The logarithm of surface pressure was retrieved from the lowest model level, while the geopotential and the aerosol optical depth (AOD) at 550 nm for total aerosol, black carbon, organic matter, dust and sulfate were retrieved from the surface level.

2.2 Aerosol and atmospheric transport modelling

In this paper, the models FLEXPART and FLEXTRA ~~models were used in this paper~~ were used for atmospheric transport modelling.

FLEXPART ("FLEXible PARTicle dispersion model") is a Lagrangian particle dispersion model designed for calculating the long-range and mesoscale transport, diffusion, dry and wet deposition, and radioactive decay of air pollutants from point, line area and volume sources. FLEXPART can be run in forward mode, simulating the transport and dispersion of emissions from given sources towards receptor points or producing gridded output concentration and deposition, or in backward mode from given receptors to produce source-receptor relationships with respect to a point source or gridded sources~~.~~ (Seibert and Frank, 2004). The model ingests ECMWF 3D meteorological fields and solves the equations for transport, turbulent diffusions and other relevant processes in a Lagrangian framework (Stohl et al., 1998) (Pisso et al., 2019). The sensitivity of a receptor concentration to potential sources is obtained directly as the model output in the case of a backward run (Seibert and Frank, 2004) (Eckhardt et al., 2017).

FLEXTRA is a kinematic trajectory model. It simulates only the transport of air parcels by mean winds, ignoring turbulence and convection, and do not ~~represent~~ provide concentrations, deposition, etc.

For both models the ECMWF (European Centre for Medium Range Weather Forecasts) Era Interim meteorological fields with a horizontal resolution of 0.5°~~x~~ × 0.5°, the lowest 61 vertical levels (corresponding to pressure levels from surface to 250 hPa) out of the 137 vertical levels, and a temporal resolution of 3 h were used. A sub-domain covering a part of North Hemisphere (175°W – 60°E, 0°N – 90°N), including Europe, a part of the Atlantic Ocean, North America and a part of Africa was extracted as "mother" domain.

For the determination of the aerosol optical properties for sites without lidar measurements, where the aerosol composition is determined from CAMS products, the aerosol model from ~~Ref.~~ (Nicolae et al., 2018) was used, called in the following NATALI aerosol model. Six classes of ~~pure aerosol~~ typical aerosol (called "pure aerosol" in the reference) were considered in this model: continental, continental polluted, dust, marine, smoke, and volcanic. In the model, the optical properties are computed for ~~pure~~ typical aerosols and for mixtures of two or three ~~pure~~ typical aerosols at fixed wavelengths 350 nm, 550 nm and 1000 nm with the T-Matrix method using light scattering on non-spherical particles (Mishchenko et al., 1996) for a log-normal distribution of homogeneous particles. The microphysical parameters (effective radius, standard deviation and complex refractive indices) of the components, needed as input in the model, were taken from the GADS database (Global Aerosol DataSet) (Koepke et al., 1997).

For the comparison with optical properties obtained from lidar measurements, the optical properties computed in the model are re-scaled to the lidar wavelengths (355 nm, 532 nm and 1064 nm) using an AE equal to one, as the values of model and lidar wavelenghts are very close.

2.3 Data processing and analysis

2.3.1 Lidar and ceilometer data processing

The vertical profiles of the backscatter coefficients were determined using the Fernald–Klett method (Fernald, 1984; Klett, 1981) for remote sensing instruments with only elastic channels. For instruments with elastic and Raman channels, the backscatter and the extinction coefficients were determined using the combined method (Ansmann et al., 1992). The PDepR was computed using the volume depolarization ratio and the backscatter coeficients (Freudenthaler, 2016). The AE is computed from the extinction coefficients for the wavelengths 532 nm and 355 nm.

The LR was computed as the ratio of the extinction coefficient to backscatter coefficient. For ceilometers, lidars with only elastic channels and lidar measurements during the day (when only backscatter coefficients can be retrieved), the value of the LR was taken from the NATALI aerosol model, which gives an estimate of the LR for 14 aerosol types. The values for 532 nm used in this paper are: 23 ± 10 sr for marine, 40 ± 8 sr for dust, 68 ± 6 sr for continental, 52 ± 2 sr for continental polluted, 53 ± 5 sr for polluted dust, 64 ± 8 sr for smoke and 46 ± 10 sr for mixed dust.

The aerosol layers are identified from the lidar measurements with the ~~methodology described in Ref. (Belegante et al., 2014)~~ gradient method, applied to the RCS profiles ~~.~~ (Belegante et al., 2014) (Nicolae et al., 2018). The gradient method is based on the identification of the peaks/valleys from the first derivative applied to the vertical profiles. If two consecutive layers are very close (less than 100 m), these layers are merged into one layer. Also, if the signal to noise ratio in the layer is lower than a threshold (here set to 5), the layer is discarded.

The aerosol type is determined from the lidar measurements using the NATALI typing algorithm, described in ~~Ref.~~ (Nicolae et al., 2018).

2.3.2 CAMS product processing

The values of the CAMS ~~quantities~~ products (mixing ratios, temperature, specific humidity, etc) for a given location were computed by interpolating the gridded CAMS values, using the inverse weighting distance interpolation.

The air density and the altitude specific to the model levels were computed according to CY42R1 from IFS documentation (Benedetti et al., 2009).

2.3.3 Data analysis

The concentrations of SO_2, $PM_{2.5}$, PM_{10} and O_3 measured in situ at the air quality background station Pillersdorf were analyzed for sliding periods of one month, to identify excesses with respect to the measured average values. If a significant excess is identified (values exceed by 50% the averaged values for 30 days), the corresponding period is analyzed in detail, using also CAMS products at the in situ station and measurements and CAMS products at the closest lidar stations around the in situ station. For Spring 2014, a period with a significant excess was identified in the time interval 15 ~~Mar~~ March – 14 ~~Apr~~April, which is presented in this paper.

The CAMS products are retrieved for the in situ site. The time series of mixing ratios of sulfate, dust, organic matter and total aerosols are then analyzed for the same period as the in situ data. If one of the aerosol components has no significant contribution to the aerosol concentration, this component can be neglected in the subsequent analysis of the aerosol. The time series are also retrieved for the lidar stations around the in situ site.

To assess if the excess is caused by a local event or long- or medium-range transported aerosol is involved, a qualitative analysis of the in situ concentration measurements, the time series of mixing ratios at the in situ station and at the lidar stations around the in situ station is done. If the event is present only at the in situ station, we can assume that it is a local event. If the event is seen at some of the lidar stations around the in situ site, the event has contributions from an aerosol transport event.

The layers for the event at the in situ site are then determined by ~~appplying the gradient method (Belegante et al., 2014) on~~ applying the same gradient method as for lidar data processing, but applied to the altitude profiles of aerosol concentrations. The concentrations are computed by multiplying the ~~mixing ratio~~ CAMS mixing ratios and the air density.

A statistical analysis of trajectories is then performed for each layer identified at the in situ site. Three-dimensional kinematic hourly trajectories are computed with the FLEXTRA model, run in backward mode for a transport time of 10 –20 days (typical for long-range transport) and in forward mode for few days for several receptor altitudes between 1500 m and 7000 m. Due to the turbulence in the planetary boundary layer, trajectories below 1500 m are usually not included in the analysis, being mostly local trajectories. During the period under investigation, with low wind speeds and mostly clear skies, the boundary-layer height varied at Pillersdorf from less than 100 m at night to about 1500 m in the afternoon.

A trajectory is associated with a lidar station if the projection of the trajectory on the Earth surface intersects a $0.5^\circ \times 0.5^\circ$ cell centered on the lidar station location. The altitude of the trajectory and the time the trajectory overpasses the lidar cell are the altitude and time of the FLEXTRA trajectory at the corresponding location.

If a trajectory overpasses a lidar station, the lidar measurements for the overpass time are analyzed. The aerosol layers are identified with the same method (Belegante et al., 2014) as for in situ station, applied to the RCS profiles. The optical properties are computed for each identified aerosol layer, as described in Sec. 2.3. The type of the aerosol is determined from the optical properties using the NATALI typing algorithm. The aerosol concentrations are also computed for each layer, using the method described in ~~Ref.~~ (Mamouri and Ansmann, 2017). For each layer, the sulfate fraction (SF) is computed as the ratio of sulfate concentration to total aerosol concentration.

The layers determined from lidar measurements are then compared with the altitude of the trajectories overpassing the lidar station. If the altitude matches a layer within a reasonable distance, the trajectory is associated with the layer. The matching distance is defined as $2\sigma_{\mathrm{lidar}}$, where σ_{lidar} is the effective spatial resolution of the lidar, tipically of the order of $\sim$60 m.

The source-receptor sensitivity (SRS) is then computed for each layer identified in the sulfate profile at the in situ station using FLEXPART with sulfate as passive tracer. The ~~release~~ receptor is set to the location of the in situ station, at the altitude determined for that layer and the corresponding event time interval. Sources are considered to be situated between 0 – 100 m. Wet and dry deposition are taken into account in the computation. Combining the source-receptor sensitivity with emission inventories, the relative distributions of $\mathrm{SO_2}$ sources for the corresponding sulfate layer are computed. In this study, the MACCity anthropogenic $\mathrm{SO_2}$ emission inventories from the Emissions of atmospheric Compounds & Compilation of Ancillary Data (ECCAD) emission database (Darras et al., 2018) was used.

A cross-check of sulfate concentrations from lidar measurements, CAMS sulfate products and FLEXPART is done for the layers at the lidar stations associated with the layers at the in situ station. One expects the values from the three methods to be in agreement.

The optical properties of the aerosol from each layer at the in situ station are then computed according to Sec. 2.2 and compared with the optical properties of the aerosol from the layers at the lidar stations associated with the layers at the in situ station. The optical properties determined at both sites have to be compatible, up to the changes due to the transport from one site to the other. The compatibility is also cross-checked for the type of aerosols at both stations, where the type is determined using the NATALI aerosol model at the in situ site and the NATALI typing algorithm at the lidar station.

3 Results and discussion

3.1 Results

The in situ measurements of $\mathrm{SO_2}$, $\mathrm{O_3}$, $\mathrm{PM_{2.5}}$ and $\mathrm{PM_{10}}$ concentrations recorded at Pillersdorf for the period 15 ~~Mar~~ March – 14 ~~Apr~~ April 2014 (Umweltbundesamt Austria, 2014) are shown in Fig. 1, together with the averaged values for this period (dotted line). An excess with respect to the averaged values is observed for all measurements in the period 01 – 06 ~~Apr~~April: 66% for $\mathrm{SO_2}$, 11% for $\mathrm{O_3}$, 90% for $\mathrm{PM_{2.5}}$ and $\mathrm{PM_{10}}$. If the excess period is excluded from the calculation of the average values, the excess increases to 100% for $\mathrm{SO_2}$, 14% for $\mathrm{O_3}$, 153% for $\mathrm{PM_{2.5}}$ and 143% for $\mathrm{PM_{10}}$.

The time series of aerosol mixing ratios from CAMS near-real-time data for Pillersdorf are shown for the same period in Fig. 2 for “total aerosols" (sum of all species defined in CAMS data)~~(a)~~, for sulfate ~~(b)~~ and for dust~~(c)~~. One observes a sulfate

increase with a peak on 02 ~~Apr~~April, and a second, less pronounced peak on 04 ~~Apr~~April. The aerosol mixture is dominated by dust and sulfate, as can be seen by comparing qualitatively the total, sulfate and dust distributions.

Similar distributions, retrieved from CAMS near-real-time data also, are observed for the lidar stations around Pillersdorf, as shown in Fig. 3 for Munich~~(a), Leipzig (b) and Bucharest(c)~~, Leipzig and Bucharest. From these distributions, one can infer the presence of an event of sulfate transport over Europe.

The vertical profiles of sulfate, dust and "total aerosol" concentrations are shown in Fig. 4 for Pillersdorf, 02 ~~Apr~~April. The sulfate layers, identified with the gradient method, are shown as grayed area in the same figure.

For 02 ~~Apr~~April, from 0:00 to 12:00, sulfate layers mixed with dust are well defined between 2 km and 3 km, and between 4 km and 6 km. During the day, the layers descend slowly and disperse, such that it mixes with dust and the aerosols from the planetary boundary layer. This can also be seen from the concentration profile of "total aerosol", which also shows a similar structure, indicating a common transport path of sulfate and dust as polluted dust nearby Pillersdorf. The evolution of the sulfate and dust layers during the day is correlated with the increase of SO_2 and $\mathrm{PM}_{2.5}$ concentrations measured in situ, while the evolution of the dust layers is correlated with the increase of the PM_{10} concentration.

For the layers identified above, the back-trajectories of the aerosols were computed with FLEXTRA, starting from Pillersdorf at the time corresponding to the aerosol profiles for a backward period of 12 days. As mentioned before, trajectories below 1500 m are not computed, due to turbulence in the planetary boundary layer.

For 02 ~~Apr~~April, they are shown in Fig. 5 for 00:00~~(a)~~, 06:00~~(b)~~, 12:00 ~~(c)~~ and 18:~~00 (d).~~ 00. From the trajectory analysis, the time and the altitude of the trajectories passing over the lidar stations were determined. The station, the time and the altitude are shown in the lower plots of each sub-figure.

The aerosol layers identified Pillersdorf were transported further. Some of the layers pass over the lidar station from Bucharest. Their trajectories were analyzed running FLEXTRA in forward mode for three days, starting from Pillersorf. Fig. 6 shows the forward-trajectories for 02 ~~Apr~~April, 06:00, which pass over Bucharest lidar station on 03 ~~Apr~~April.

The lidar measurements for the stations overpassed by the trajectories determined from the backward and forward analysis are presented as range corrected signal time series (RCS) in Fig. 7 and Fig. 8 and for the event on 02 ~~Apr~~ April in Pillersdorf.

Aerosol layers, their optical properties and the concentration were determined from the lidar measurements following the methodology described in Sec. 2. The layers identified are marked on the corresponding RCS plot.

The association of the layers identified from lidar measurements to the altitude of the backward or forward trajectories over the stations corresponding to the layers identified in Pillersdorf was performed for all eight concentration profiles measured (see Fig. 4 for 02 ~~Apr~~ April and Fig. 13 for 04 ~~Apr~~April). The association for trajectories from ~~Apr~~ April 02, 06:00 is presented in Table 1. The trajectory altitude (Traj. alt.) in the table represents the altitude of the trajectory when overpassing the lidar station. The corresponding layers are also marked in the RCS plots (red line box).

The source-receptor sensitivity was computed for each layer identified in the sulfate profiles at Pillersdorf; the column-integrated source-receptor sensitivity was also computed. Fig. 9 shows the corresponding distributions for the layers L1~~(a)~~, L2~~(b)~~, L3 ~~(c)~~ and total column ~~(d)~~ from 02 ~~Apr~~April, 06:00.

For each layer, the relative distribution of the SO_2 sources was computed from the source-receptor sensitivity and the source inventory MACCity. Fig. 10 (a) shows the distribution for layer L1 at Pillersdorf, 02 ~~Apr~~April, 06:00, while Fig. 10 (b) shows the distribution for the corresponding layer at Leipzig, 31 ~~Mar~~March, 18:00. To evaluate the local distribution of sources near Pillersdorf, a zoomed view of the SO_2 relative distribution in shown in Fig. 10 (c) for the sub-domain covering a part of the Europe, centred in Austria (10°W – 40°E, 35°N – 60°N). Similar distributions are shown for layer L2 in Pillersdorf in Fig. 11 (a), with a corresponding layer in Leipzig (31 ~~Mar~~March, 23:00), shown in Fig. 11 (b), and the zoomed view for Pillersdorf in Fig. 11 (c). For layer L3 at Pillersdorf, the distribution is shown in Fig. 12 (a), with associated layers in Munich (~~Apr~~ April 01, 05:00), Garmisch (~~Apr~~ April 01, 14:00 – not shown as very close to Munich) and Bucharest (~~Apr~~ April 03, 13:00) shown in Fig. 12 (b) and Fig. 12 (c), respectively, and a zoomed view for Pillersdorf in Fig. 12 (d).

For the lidar stations, a comparison of concentrations computed from the lidar measurements with the sulfate concentrations computed from CAMS values for the lidar station location and the concentrations computed from the modelled SRS are given in Table 2.

The optical properties, the sulfate fraction and the aerosol types for the aerosol layers identified for Pillersdorf, 02 ~~Apr~~April, 06:00 and the associated layers at the lidar stations are given in Table 3. For Leipzig and Bucharest, the optical properties are computed from the lidar measurements; for Pillersdorf, Garmisch and Munchen they are computed using the NATALI model.

The peak on 04 ~~Apr~~ April was also analyzed similarly to the peak on 02 ~~Apr~~April. The corresponding vertical profiles of sulfate, dust and "total aerosol" concentrations are shown in Fig. 13. From the backward and forward trajectory analysis, only one lidar station could be associated with a trajectory, for layer L2 at Pillersdorf, 12:00. The corresponding RCS at the lidar station is shown in Fig. 14. The SRS for the identified layers at Pillersdorf, 12:00, are presented in Fig. 15. Layers at Pillersdorf were associated to layers at the lidar stations; they are given in Table 4. The comparison of the aerosol concentrations at the lidar station over-passed is given in Table 5, and the optical properties are given in Table 6.

3.2 Discussion of the results

The daily variations of the in situ measurements of SO_2, O_3, $PM_{2.5}$ and PM_{10} concentrations depend on more factors, such as variations in source emissions, photochemical reactions, meteorological conditions, PBL heights and short-, medium- and long-range transport of aerosols.

Fig. 1 indicates a period between 27 ~~Mar~~ March and 6 ~~Apr~~ April in which in situ measurements of SO_2, O_3, $PM_{2.5}$, PM_{10} concentrations recorded at Pillersdorf exceed the averaged values for the period 15 ~~Mar~~ March – 14 ~~Apr~~ April 2014. A significant load of aerosols in the atmosphere in this period is also confirmed by the AOD values between 0.07 and 0.73 for Pillersdorf, retrieved from CAMS products, which are above the AOD threshold of 0.06 for clear atmosphere (Kaskaoutis et al., 2012). For the period 27 ~~Mar~~ March to 31 ~~Mar~~March, no significant load of aerosols is observed at the lidar stations around Pillersdorf, therefore no medium- or long range transport of aerosols is involved. The source-receptor sensitivity computed for 31 ~~Mar~~ March (not shown~~in this paper~~) points to a short-range transported event, of small duration and at low altitude, with sources in the South Eastern of Austria. This event is not described in this paper.

From a qualitative analysis of in situ concentrations for PM_{10}, $PM_{2.5}$ and SO_2 (Fig. 1) and the CAMS time series of mixing ratios for dust, sulfate and total aerosol at Pillersdorf (Fig. 2) and the lidar stations around Pillersdorf (Fig. 3), the presence of an event of sulfate transport over Europe can be inferred, with two peaks, on 02 ~~Apr~~ April and 04 ~~Apr~~April, respectively.

On 02 ~~Apr~~April, one observes from the concentration profiles (Fig. 4) that in the morning the dust was dominant in the layer between 0.55 km and 1.50 km and in the layer between 1.98 km and 3.11 km, while sulfate was dominant in the higher altitude layer, between 4.20 km and 6.15 km. In the afternoon, the sulfate concentration increases gradually in the lower layers, mixing with the dust, while the upper layer become thinner (layer range from 4.0 km to 5.0 km).

The back-trajectories for 02 ~~Apr~~ April (Fig. 5) show a consistent pattern. In the morning (00:00 and 06:00), the lower trajectories (below 2000 m) originate from Eastern and Southern United States (US), ~~transverse~~ traverse the North Atlantic Ocean and pass over Central Europe, spending $\sim$ 6 days in this region, arriving at Pillersdorf from the northwest direction. The middle-altitude trajectories (2000 m – 5000 m) originate from Southern US, ~~transverse~~ traverse the ocean and pass over Northwest Africa (spending $\sim$ 3 days), arriving in the Central Europe from southwest, then arriving along the Alps at Pillersdorf. The high-altitude trajectories (above 5000 m) ~~transverse~~ traverse the ocean, arriving at Pillersdorf from the west direction. In the afternoon (12:00 and 18:00), the lower trajectories originate from Eastern Europe, while the middle-altitude and high-altitude trajectories originate from Eastern US, ~~transverse~~ traverse the ocean and the Northwest Africa, arriving at Pillersdorf from the west direction.

The SRS patterns, shown in Fig. 9, and the relative distributions of SO_2, shown in Fig. 10, Fig. 11 and Fig. 12, indicate the influence of five source regions for the transport of the sulfate event recorded on day 02 ~~Apr~~ April at Pillersdorf: Southern and Eastern US, Northwest Africa, Central Europe and Eastern Europe.

For the lower layers, the Central Europe, including industrial centres from the "Black triangle" (Eastern Germany, Southwest Poland and Czech Republic) was the main source contributing to sulfate transported over Northern Austria, where Pillersdorf station is situated. Medium to smaller contributions come from sources in Eastern Europe, the Northwest Africa and Eastern US.

For the middle-altitude layers, sources from Central Europe (Northern Italy, Serbia, Hungary) contribute with similar emissions. Northwest Africa and Eastern US have also important contributions.

For the high-altitude layers, the main contributions come from Northwest Africa, but sources from Southern and Eastern US contribute also significantly. No contributions from Europe are seen for these layers.

For the peak on 04 ~~Apr~~April, having only one lidar station associated to aerosol trajectories, the analysis is more difficult. From the existing information, we can conclude that the pattern is similar with layer L2 and L3 from 02 ~~Apr~~April, with contributions from Northen Italy, Northwest Africa and Southern US.

The AEs for the event have values between 0.67 and 0.79, which correspond to a mixture of fine and coarse particles, with size distribution centered on 0.75 µm. For this size distribution, the sulfate (Ding et al., 2017) and the dust (accumulation mode) are the dominant aerosols. The LR is comparable for all sites, having values between 45 and 55 sr, while the linear PDepR has values between 0.07 to 0.22. These values correspond to low to medium absorbing aerosol with non-spherical shape (Nicolae et al., 2018).

The aerosol type is determined from the optical properties for the layers identified in this event, at the in situ station and the lidar stations. Consistent aerosol type was found between the in situ station and the lidar stations along the trajectories. The changes in the values of the aerosol LR, AE and linear PDepR along the trajectories can be explained by:

- the mixing of dust with secondary sulfate from anthropogenic sources during the transport paths to Leipzig, Munich, Garmisch-Partenkirchen, Pillersdorf and Bucharest, and
- the adsorption of the SO_2 on ~~the dust mineral oxides compounds~~oxides contained in the mineral dust. The sulfate particles are expected to be formed by SO_2 oxidising on dust surface due to mineral oxides compounds from dust (e.g. hematite).

~~Monthly-averaged maps of column mass density for sulfate are available from EarthData NASA GIOVANNI (NASA, 2018) online data system. For short-time events, they can be used only for a qualitative interpretation, being monthly averaged. The map of sulfate column mass density for Mar 2014, Fig. **??** (a), shows an increased density over South–Eastern and Eastern US and a reduced density over Central Europe. For Apr 2014, shown in Fig. **??** (b), the density increases over Central Europe.~~

4 Conclusions

The excess of SO_2, $PM_{2.5}$, PM_{10} and O_3 observed in the period 01 – 06 ~~Apr~~ April 2014 at the Austrian air quality background station Pillersdorf was analyzed using in situ data, lidar measurements at the closest EARLINET stations around the in situ site, CAMS near-real-time data, and aerosol and atmospheric transport modelling. This excess was associated with the transport of sulfate aerosols, mixed during the transport with dust. By correlating the local information with a trajectory analysis and an analysis of aerosol potential sources, a complex pattern of contributions to sulfate at the in situ station was found. The lower layers (below 2000 m) originated mainly from the Central Europe. Medium to smaller contributions came from sources in Eastern Europe, the Northwest Africa and Eastern US. For the middle-altitude layers (between 2000 m and 5000 m), sources from Central Europe (Northern Italy, Serbia, Hungary) contributed with similar emissions. Northwest Africa and Eastern US have also important contributions. The high-altitude layers (above 5000 m) originated from sources from Northwest Africa and from Southern and Eastern US, as transported secondary sulfate mixed with dust. The effect of medium- and long-range transport of aerosol is significant, and can not be neglected when analyzing the air quality at an in situ station. For a quantitative analysis and modelling of aerosol deposition, more measurements are needed, including precise vertical aerosol profiles at the in situ station.

The spring period studied in this paper is characterized by low, if any, deep convection. For the summer period, one expects however to have a strong convective activity over Central Europe. A study of the summer periods for the years 2014–2017 for the same region was also performed; the results will be presented in a separate paper.

The methodology developed in this paper allows to obtain a better understanding of the effects of aerosol transport on the in situ measurements. It can be used as a general tool to correlate measurements at in situ stations with ground-based remote

sensing stations located around these in situ stations. A dedicated paper for the methodology, extended to trace gases and other aerosols, with analysis of more case studies is under preparation.

Author contributions. CT collected and processed all data, developed the methodology and performed the data analysis. Both authors contributed to the optimization of the analysis and the interpretation of the results. PS provided the pre-release of FLEXPART version 10, with a better wet deposition and other improvements. The manuscript was prepared by CT with contributions from PS.

Competing interests. The authors declare that they have no conflict of interest.

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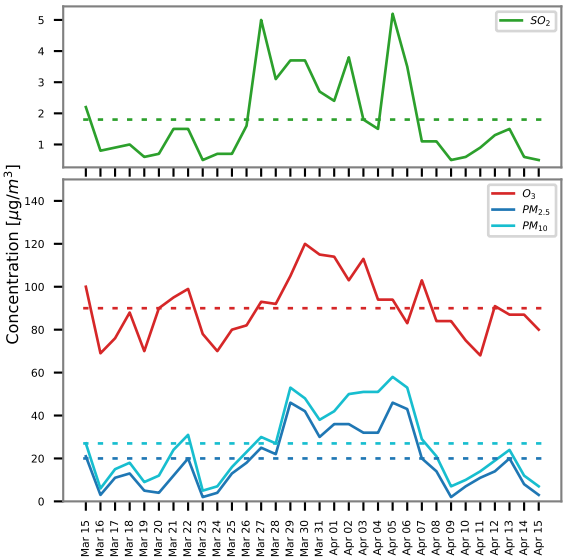
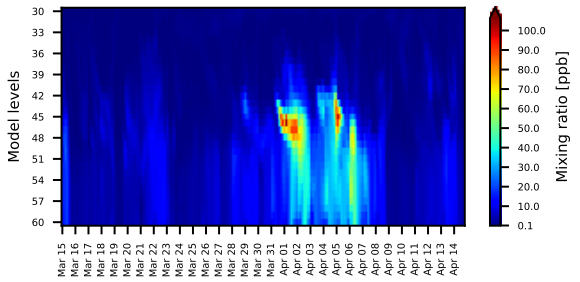
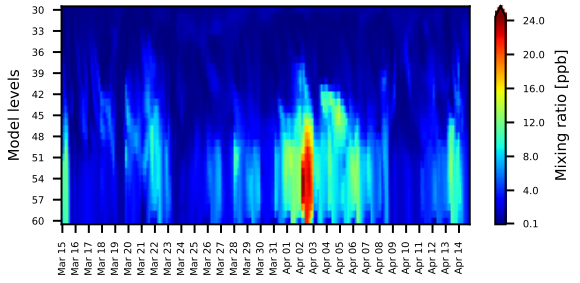


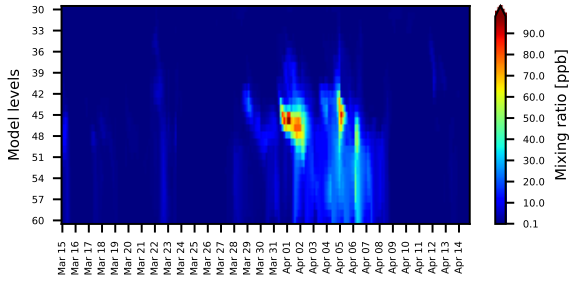
Figure 1. In-situ SO_2, O_3, $PM_{2.5}$ and PM_{10} concentrations measured at Pillersdorf, Austria (EMEP station AT30, 48°43'N, 15°55'E). The dotted lines represent the averaged values for the plotted period.



(a) Aerosol

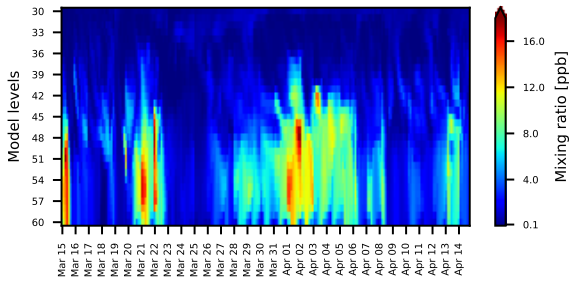


(b) Sulfate

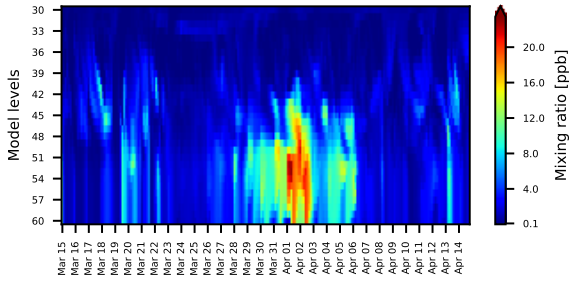


(c) Dust

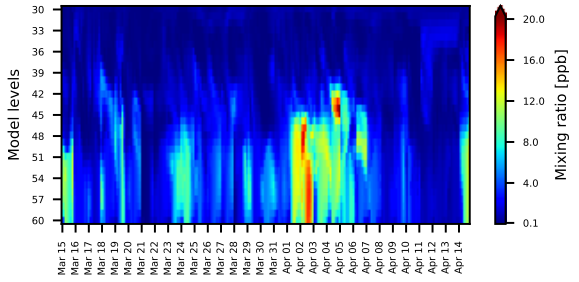
Figure 2. Time series of CAMS mixing ratios for total aerosol (a), sulfate (b) and dust (c), Pillersdorf, 15 ~~Mar~~ March – 14 ~~Apr~~ April 2014.



(a) Munich



(b) Leipzig



(c) Bucharest

Figure 3. Time series of CAMS mixing ratios for sulfate for Munich (a), Leipzig (b) and Bucharest (c), 15 ~~Mar~~ March – 14 ~~Apr~~ April 2014.

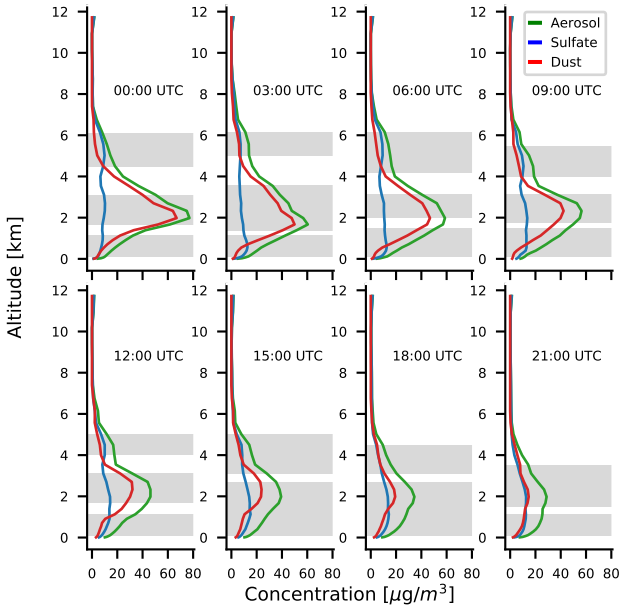


Figure 4. CAMS total aerosol, sulfate and dust profiles for 02 ~~Apr~~ April 2014, Pillersdorf. Grayed area represents the identified sulfate layers. Altitudes are given in km AGL. Local time is UTC+2.

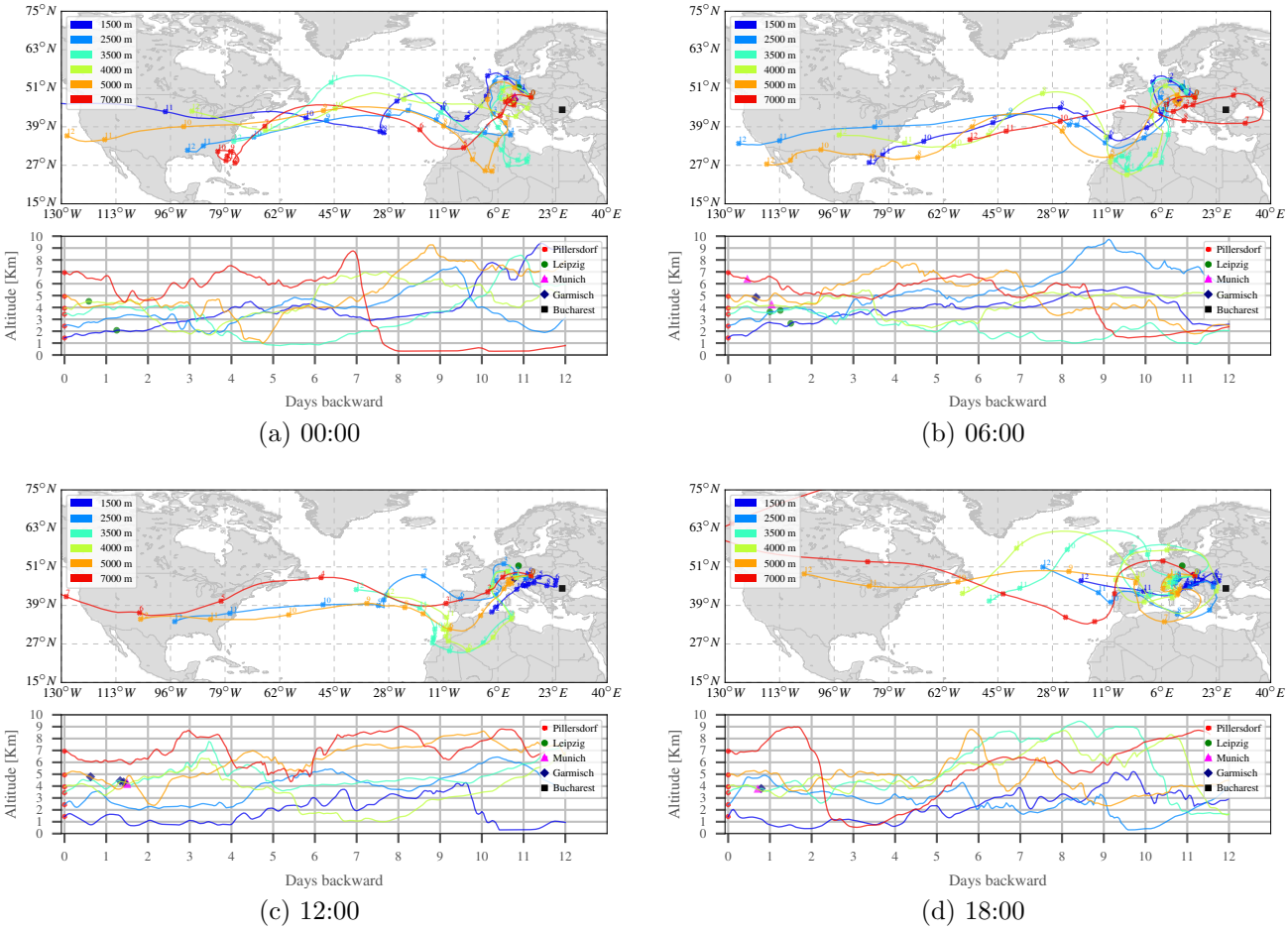


Figure 5. Pattern of back-trajectories (upper plot of sub-figure) and their altitude profile, including overpassed lidar stations (lower plot of sub-figure) for Pillersdorf, 02 ~~Apr~~ April 2014 at 00:00 (a), 06:00 (b), 12:00 (c), 18:00 (d).

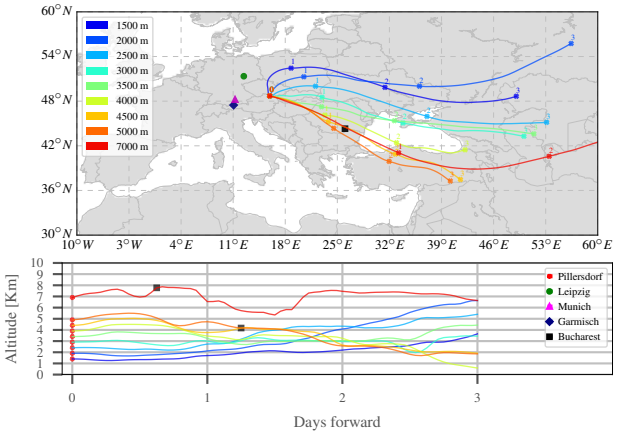
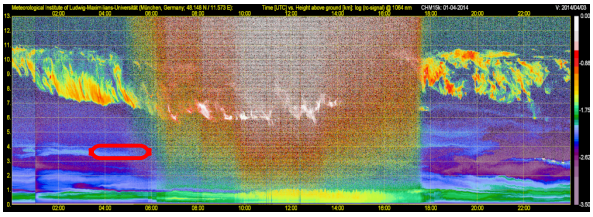
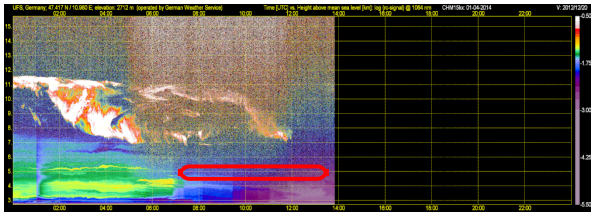


Figure 6. Pattern of forward-trajectories (upper plot) and their altitude profile, including overpassed lidar stations (lower plot) for Pillersdorf, 02 ~~Apr~~ April 2014, 06:00.

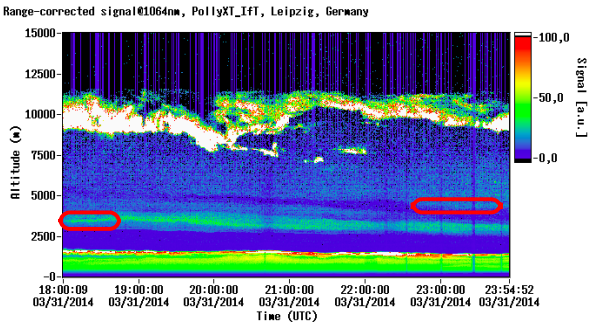


(a) Munich, 01 April 2014 Ceilometer YALIS

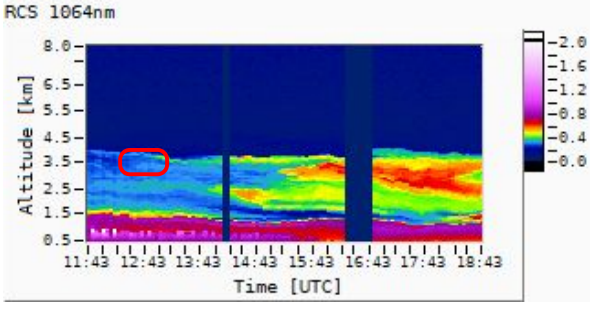


(b) Garmisch, 01 April 2014 Ceilometer

Figure 7. ~~log(~~Logarithm of the range corrected signal ~~)~~ at 1064 nm, 24 h, for Munich (a) and Garmisch (b) stations. The red line boxes represent the identified layers.

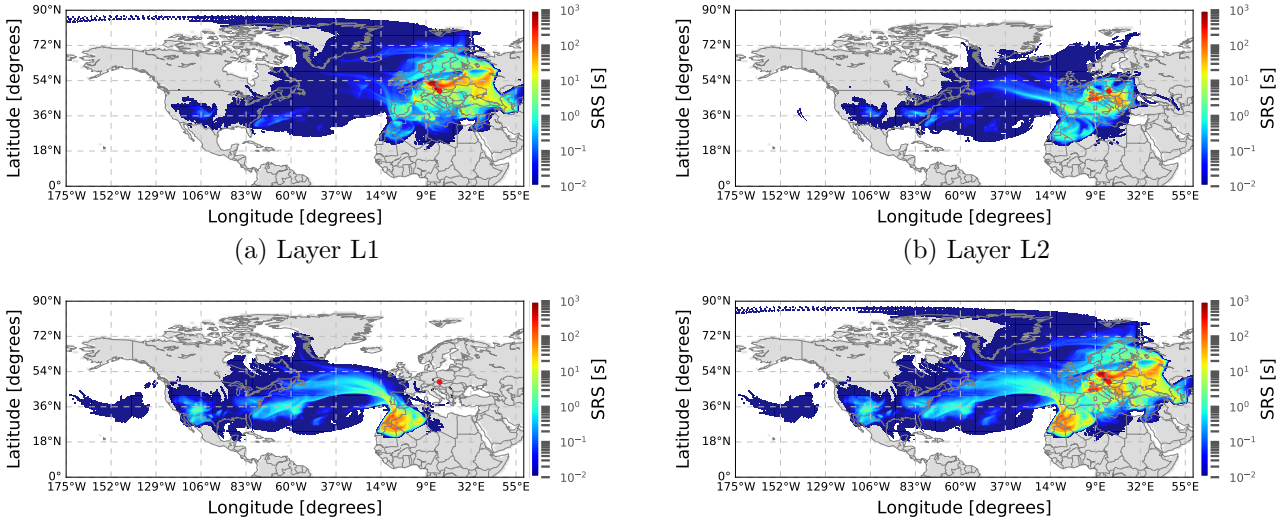


(a) Leipzig, 31 March 2014 PollyXT



(b) Bucharest, 03 April 2014 RALI

Figure 8. Range corrected signal at 1064 nm for Leipzig (a) and Bucharest (b) stations. The red line boxes represent the identified layers.



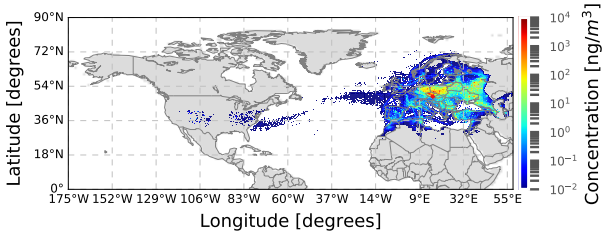
(a) Layer L1

(b) Layer L2

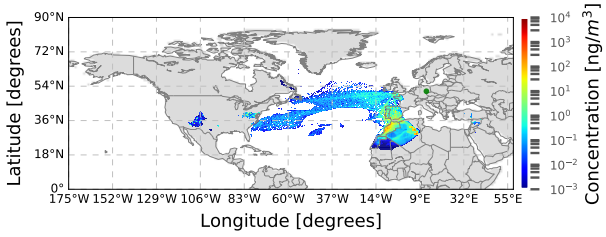
(c) Layer L3

(d) Total column

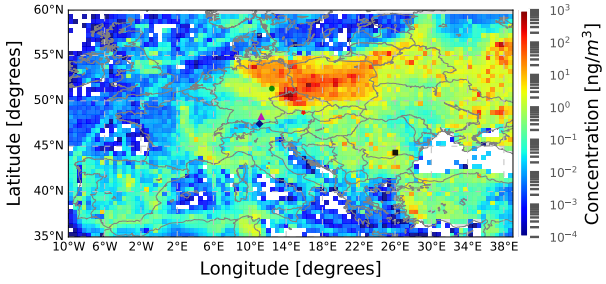
Figure 9. Source-receptor sensitivity for layer L1 (a), L2 (b) and L3 (c) and total column (d), Pillersdorf, 02 ~~Apr~~April, 6:00



(a) Pillersdorf, 02 April, 06:00, layer L1

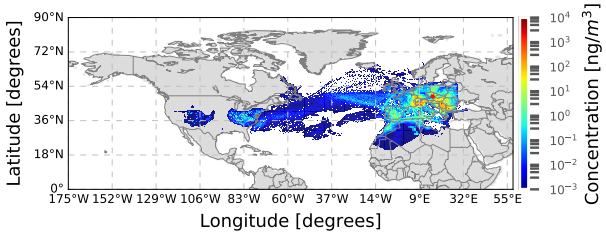


(b) Leipzig, 31 March, 18:00, layer corresponding to L1

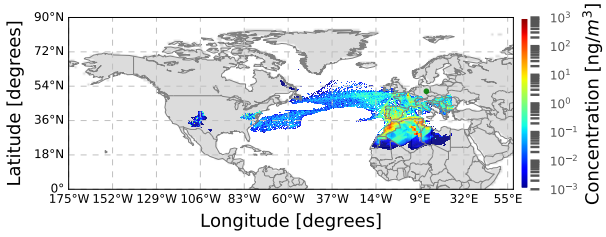


(c) Pillersdorf, 02 April, 06:00, layer L1, zoomed

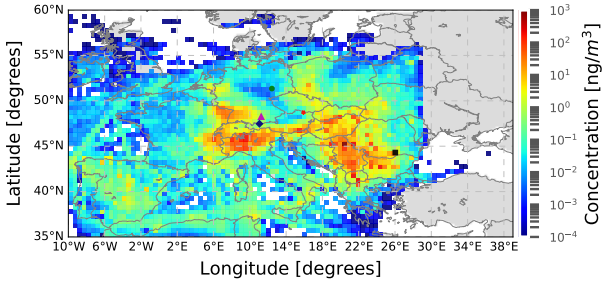
Figure 10. Relative distributions of SO_2 sources for Pillersdorf layer L1 (a), Leipzig (b); zoomed distribution for Pillersdorf layer L1 (c).



(a) Pillersdorf, 02 April, 06:00, layer L2

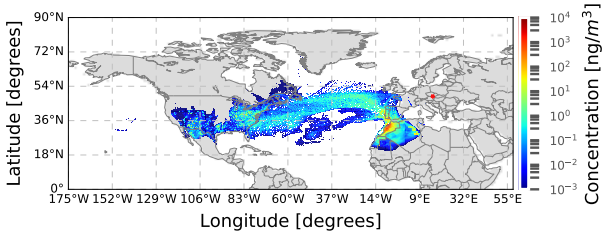


(b) Leipzig, 31 March, 23:00, layer corresponding to L2

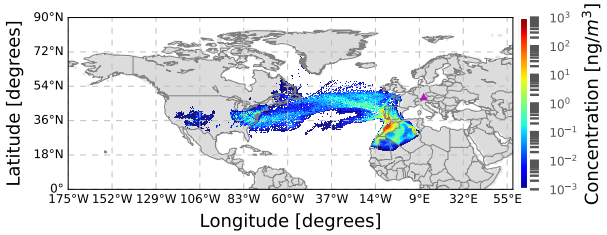


(c) Pillersdorf, 02 April, 06:00, layer L2, zoomed

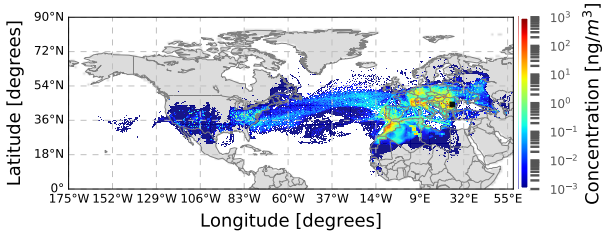
Figure 11. Relative distributions of SO_2 sources for Pillersdorf layer L2 (a), Leipzig (b); zoomed distribution for Pillersdorf layer L2 (c).



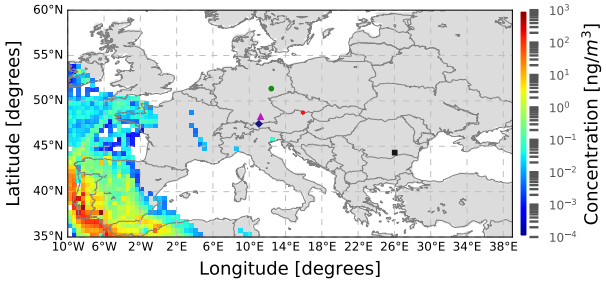
(a) Pillersdorf, 02 April, 06:00, layer L3



(b) Munich, 01 April, 05:00, layer corresponding to L3



(c) Bucharest, 03 April, 13:00, layer corresponding to L3



(d) Pillersdorf, 02 April, 06:00, layer L3, zoomed

Figure 12. Relative distributions of SO_2 sources for Pillersdorf layer L3 (a), Munich (b), Bucharest (c); zoomed distribution for Pillersdorf layer L3 (d).

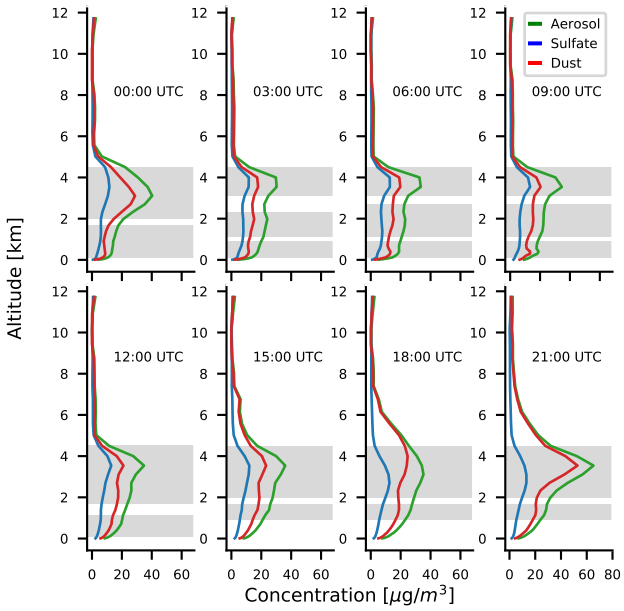


Figure 13. CAMS aerosol, sulfate and dust profiles for 04 ~~Apr~~ April 2014, Pillersdorf. Grayed area represents the identified sulfate layers. Altitudes are given in km AGL.

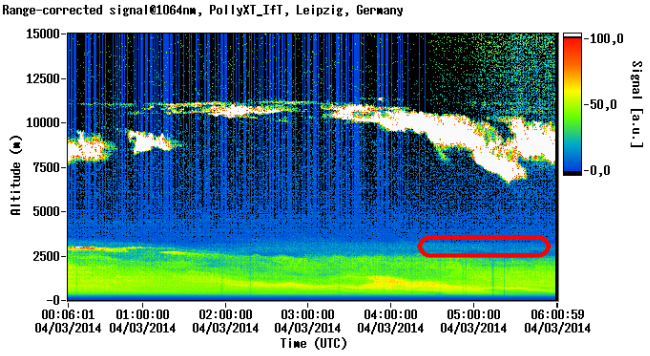
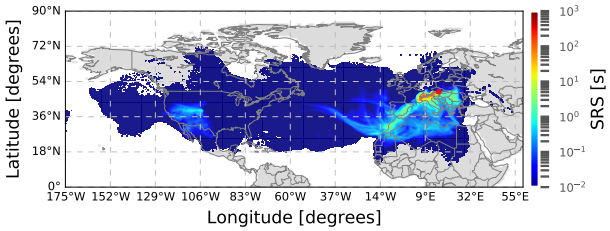
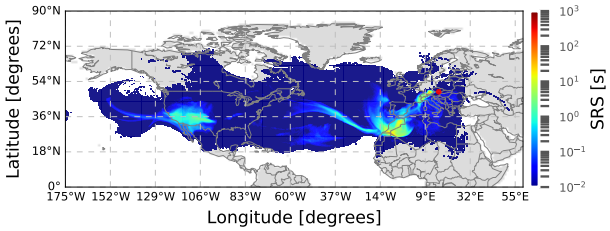


Figure 14. Range corrected signal at 1064 nm for Leipzig station, 03 ~~Apr~~ April 2014. The red line box represents the identified layer.



(a) Pillersdorf, 04 April, 12:00, layer L1



(b) Pillersdorf, 04 April, 12:00, layer L2

Figure 15. Source-receptor sensitivity for layer L1 (a) and L2 (b), Pillersdorf, 04 ~~Apr~~April, 12:00

~~GIOVANNI time averaged map of sulphate column mass density for Mar 2014 (a) and Apr 2014 (b).~~

Table 1. Association of layers from lidar measurements with layers and trajectories computed for Pillersdorf, 02 ~~Apr~~ April 2014, 06:00.

Pillersdorf	Lidar station, time	
	Traj. alt.	Lidar layer
L1: 0.55 – 1.50 km	Leipzig, Mar 31, 18:00	
	2.66 km	2.70 – 3.75 km
L2: 1.98 – 3.11 km	Leipzig, Mar 31, 23:00	
	3.75 km	3.85 – 4.20 km
L3: 4.20 – 6.15 km	Munich, Apr 01, 05:00	
	4.20 km	3.54 – 4.43 km
	Garmisch, Apr 01, 14:00	
	4.84 km	4.91 – 5.81 km
	Bucharest, Apr 03, 13:00	
	3.90 km	2.70 – 4.05 km

Table 2. Comparison of sulfate concentration computed from lidar measurements, CAMS products and FLEXPART for layers at lidar stations associated with layers from Pillersdorf, 02 ~~Apr~~ April 2014, 06:00.

Layer	C_{lidar} [µg m^{-3}]	C_{cams} [µg m^{-3}]	$C_{flexpart}$ [µg m^{-3}]
Leipzig, Mar 31, 18:00 2.70 – 3.75 km	14.61	12.52	12.94
Leipzig, Mar 31, 23:00 3.85 – 4.20 km	15.96	13.48	13.42
Bucharest, Apr 03, 13:00 2.70 – 4.05 km	15.24	11.95	13.26
Munich, Apr 01, 05:00 3.54 – 4.43 km	20.14	19.58	18.98
Garmisch, Apr 01, 14:00 4.91 – 5.81 km	17.93	16.76	15.39

Table 3. Optical properties, sulfate fraction and aerosol types for aerosol layers corresponding to Pillersdorf, 02 ~~Apr~~ April 2014, 06:00.

Layer	LR [sr]	PDepR	AE	SF	Type
Pillersdorf Apr 02, 06:00 0.55 – 1.50 km	51	0.22	0.67	0.49	Polluted dust
Pillersdorf Apr 02, 06:00 1.98 – 3.11 km	55	0.10	0.76	0.33	Mixed dust
Pillersdorf Apr 02, 06:00 4.20 – 6.15 km	54	0.07	0.74	0.62	Mixed dust
Leipzig Mar 31, 18:00 2.70 – 3.75 km	55	0.20	0.79	0.25	Polluted dust
Leipzig Mar 31, 23:00 3.85 – 4.20 km	54	0.17	0.79	0.44	Mixed dust
Bucharest Apr 03, 13:00 2.70 – 4.05 km	54	0.14	0.71	0.55	Mixed dust
Munich Apr 01, 05:00 3.54 – 4.43 km	47	0.18	0.75	0.40	Mixed dust
Garmisch Apr 01, 14:00 4.91 – 5.81 km	45	0.16	0.71	0.41	Mixed dust

Table 4. Association of layers from lidar measurements with layers and trajectories computed for Pillersdorf, 04 ~~Apr~~ April 2014, 12:00.

Pillersdorf	Lidar station, time	
	Traj. alt.	Lidar layer
L1: 1.98 – 4.50 km	Leipzig, Apr 03, 05:00	
	2.96 km	2.70 – 3.45 km

Table 5. Comparison of sulfate concentration computed from lidar measurements, CAMS data and FLEXPART for layers at lidar stations associated with layers from Pillersdorf, 04 ~~Apr~~ April 2014, 12:00.

Layer	C_{lidar} [µg m^{-3}]	C_{cams} [µg m^{-3}]	$C_{flexpart}$ [µg m^{-3}]
Leipzig, Apr 03, 12:00 2.70 – 3.45 km	8.38	6.75	7.99

Table 6. Optical properties, sulfate fraction and aerosol types for aerosol layers corresponding to Pillersdorf, 04 ~~Apr~~ April 2014, 12:00.

Layer	LR [sr]	PDepR	AE	SF	Type
Pillersdorf Apr 04, 12:00 0.55 – 1.50 km	54	0.07	0.75	0.25	Mixed dust
Pillersdorf Apr 04, 12:00 1.98 – 4.50 km	54	0.07	0.74	0.33	Mixed dust
Leipzig Apr 03, 05:00 2.70 – 3.45 km	55	0.11	0.76	0.74	Mixed dust