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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An episode of extremely high PM concentrations over Central Europe causedby dust emitted over the southern Ukraine" *by* W. Birmili et al.

W. Birmili et al.

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Reply to the Anonymous Referee 1

First, thank you for your referee comments. We have addressed the points you raised individually below. Your comments are repeated in italics while our reply appears in normal typeface. All minor formatting issues have been been corrected for in the manuscript unless mentioned.

"Most important, the manuscript is quite long and could be easily shortened without loosing relevant information for the reader. This holds true as well for the very long

appendix B."

Wide parts of the manuscript have been reworked. The Sections 2, 3 and 4 have been condensed wherever this was appropriate based on all referees' comments. As suggested by Referee 1, also the appendix B has been condensed to its salient points.

"Partly, this can be easily corrected if some discussion is moved from Sections 3 and 4 to be presented in its entirety in Section 5."

According to your suggestion, elements of discussion have been removed to Section 5. Sections 3 and 4 are now more descriptive, and Section 5 a true discussion section.

"Section 1.3, page 12236, line 5. "Mass balance deficit" - please explain."

By "Mass balance deficit" we meant the underprediction of PM_{10} concentrations by most chemical transport models due to the imperfect consideration of resuspended dust. This has now been explained in the text.

"Section 2.4, page 12240, 1st paragraph. State briefly how the inversion of the SMPS data was done for this instrument."

The inversion is performed by solving a set of linear equations linking the particle mobility and particle size spectra. This is now mentioned in the corresponding text in the Appendix. We also added the appropriate reference, Stratmann and Wiedensohler (1996).

"Section 2.4, page 12240, 2nd paragraph. Method of the Grimm EDM is described in many details. Why? Could be shortened."

There have been many discussions around whether an optical device like the Grimm

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EDM can deliver reliable mass concentrations. Therefore we felt that a broader description of the instrument was necessary. Since Grimm EDM data were used for mainly illustrative purposes here, and its concentrations agreed relatively well with gravimetric measurements, we took up your suggestion to shorten the description of the Grimm EDM in Section 2.4.

"But what I would rather like to know in this context: Has a size calibration been done for this instrument? Do the particle optical properties (refractive index) significantly effect the size calibration? I expect they do, in particular if the soil dust contains absorbing components. This should be taken into account."

A size calibration was done by the manufacturer before installation of the instrument in 2006. Like you, we also believe that the particle optical properties affect the size calibration, and therefore the resulting mass concentration. A shortcoming of the instrument is that currently, the user cannot interfere with the manufacturer's protocol that converts particle size into particle mass.

As mentioned in Section 4.4.1. "the mean mass concentrations for PM_{10} and $PM_{2.5}$ over three days were 50.7 and 32.0 μg m^{-3}, and thus in fair agreement with the corresponding gravimetric measurements of 49.1 and 32.8 μg m^{-3}, respectively." We do believe that the size calibration of the EDM is accurate enough to yield particle mass concentrations within an accuracy of 10 % for the average background aerosol at Melpitz. However, we acknowledge that its values might not be that accurate for strongly absorbing aerosols, such as during the dust event. We therefore added the following sentence to Section 2.4.: "As the response of the EDM is a function of the particle refractive index, we are aware of possible deviations from the true gravimetric mass in the case of strongly absorbing aerosols.".

"Section 2.6, page 12241. Which meteorological data fields did enter into the trajectory

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calculations? Explain "sigma surface"."

The trajectory simulations were based on the local scale model (LM-Lokalmodell) as well as the global scale model (GME-Globalmodell) of the German Weather Service (DWD). The LM has a horizontal grid resolution of 7 km in Central Europe. A "sigma surface" describes the fluctuations of meteorological parameters near the Earth's surface. We feel that this term is too specialised, and therefore dropped it in the new version of the manuscript. Also, Section 2.6 was strongly condensed and incorporated into the results Section 3.3. "Backtrajectories".

"Explain more clearly, how the vertical settling velocities were entering the trajectory calculations. I am not sure I understand what the authors have done here."

For any trajectory particle (not to be confused with aerosol particle), gravitational settling effects a constant wind speed vector directed towards the ground. This vector was added to the motion of the particles when integrating along the 3D wind fields delivered by the local scale model (LM-Lokalmodell).

"Next page, same paragraph, last sentence. I did not understand this. What is the relation between mean values of pressure etc. and the "local contact frequency"? Are results of this shown in the manuscript?"

We feel that our descriptions have been too specialised. Since they are not necessary for the interpretation and understanding of the results, they were dropped in the revised version of the manuscript.

"Section 3.2, page 12243, 2nd paragraph. "The low level dust cloud is pictured by yellow and green colors." - This is not clear. The dust plume has not yet been introduced at this point. The colors of the trajectories represent meteorological parameters, not

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dust."

This has now been clarified (see Section 3.3. ("Backtrajectories")).

"3rd paragraph. "Transport only possible if gravitational settling is taken into account" - What is the meaning of this statement? ..."

As you say, we wanted to express that the Saharan air could not enter the BL air over Europe and only a vertical subsiding motion due to gravitational settling could bring Saharan dust particles into the soil dust plume. The discussion of the option of Saharan dust has now completely been moved to a dedicated discussion part, Section 5.2.

"Section 3.3. Should this section not appear first within the entire Section 3? The observations by satellite, showing actually the dust plume forming over the Ukraine, set the motivation for all the transport considerations."

According to your suggestion, we placed the satellite observations now to the first place in Section 3.

"Section 3.3, page 12245, lines 7-8. This sounds speculative. Can it not be checked if there was precipitation or not?"

We checked, and there was indeed precipitation (5-8 mm daily sum) in the region south of the plume. This is now mentioned to the text.

"And in general on in this paragraph: Again, I would suggest moving this aspect of possible Saharan dust contribution to the discussion section. This aspect appears quite redundantly in the manuscript at various places."

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See above. The discussion of Saharan dust has now completely been moved to a dedicated discussion part, Section 5.2.

"Section 4.1.3, page 12248, last two paragraphs. The vertical distribution is crucial to the estimate of the total dust mass in the plume. Here I would like to see some more observational evidence illustrating the average 1400 m layer thickness."

We refrain from showing additional Figures with radiosounding profiles in this already very long paper. However, more concrete figures on boundary layer heights derived from individual radiosounding ascents are now given in Section 4.1.3. The source of the radiosounding data is http://weather.uwyo.edu/upperair/.

"More importantly, regarding uncertainties in the area covered by the plume (horizontally and vertically), and variability of observed PM10 data, an uncertainty range for the 60 Gg of dust should be presented."

These uncertainties have now been been quantified (60$\pm$20 Gg). Corresponding text was added at the end of Section 4.1.3.

"Page 12250, lines 10-11. But this is to be expected, isn't it? Surface wind speed is always lower than wind speed (air mass advection) aloft."

This statement is in fact trivial and has accordingly been removed.

"Page 12250, discussion in last paragraph. Comment on why mixing with FT air is not possible would be of interest here. Is that due to synoptical/meteorological conditions - or due to the dust plume itself and its possible influence on atmospheric stability?"

The prime reason is that the boundary layer is topped by warm air merging from the south. This causes a temperature inversion and thus, an enhanced vertical stability.

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This reason is now mentioned in Section 5.1.

"Page 12251, last two paragraphs. This part I find rather lengthy and I don't see the relevance. Should be more focused or skipped."

The last two paragraphs are essential in linking or distinguishing the dust plume from former observations (dust, biomass burning) in terms of optical parameters. We therefore prefer to keep these paragraphs. Referee 3 found these aspects particularly interesting, and urged us to elaborate the topic further.

"The usage of the term "non-volatile" with respect to the inlet lines being heated to "only" 50 C I find very misleading. Often much higher temperatures are used in aerosol research to describe the non-volatile particle fraction."

It is true that higher temperatures are used frequently. However, the volatile fraction at 50 degrees C (ammonium nitrate to a large extent) is a feature that is clearly different between the fine and coarse particles modes. For better clarification, the temperature of 50 degrees C is now mentioned more frequently in the text and the Figure caption.

"Section 4.4.1, page 12252, last sentence. "Grimm EDM provides correct mass concentrations" - but this can be only true if there is a dominating coarse particle mode!?"

See above. In Section 2.4. we acknowledge "As the response of the EDM is a function of the particle refractive index, we are aware of possible deviations from the true gravimetric mass in the case of strongly absorbing aerosols.".

"Section 4.4.2, last paragraph. Is this important for this study? Can be skipped in my opinion."

The discussion of the chemical sub-fractions is of relevance for the discussion of

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the source mechanisms (dust, biomass burning). In any case, the paragraph was reformulated to place the measurements in context with continental background concentrations.

"Section 4.4.3. and 4.4.4. Overall I find these two sections too lengthy..."

Done. Section 4.4.4. was reduced to one sentence and combined with 4.4.3. The new section has been greatly condensed.

"Section 5.2. Essentially, this is not really a discussion but a summary of what has been said earlier in the manuscript... Author's should decide where the discussion of this aspect should go. It should occur only once. (And, as mentioned, an uncertainty range should be given.)"

This is a good suggestion. Consequently, Section 5 has been reorganised and builds upon the measurements introduced in the results sections.

"Section 5.3. This is an interesting section, dealing with the important information on how exceptional the dust event actually was, but I am wondering why only the data of the Melpitz station have been addressed here. What about the data records of the many other ground stations used in this study. Do they confirm the conclusion?"

Melpitz is IfT's own site where also one of the longest data records exist. Another issue is that most of the PM10 data of the 360 stations are not publicly available in the required form. Most of the institutions involved provided the data from March 23-25 to us by courtesy (they gave us a favour). Preparing larger amounts of data spanning many years of observations would cost a lot of time for these institutions (most of them can probably not justify doing this as part of their legal mandate), left alone that they might require being paid. While we acknowledge that your question is absolutely

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interesting, we are aware that it cannot be answered within short time and the scope of this paper.

"Furthermore, if this event was so extremely exceptional, it kind of takes the ground away of the discussion following later on in Section 5.4 and 5.5 (desertification trends). How do the authors rate the relevance of such a phenomenon, if it is so unusual."

This paper focuses on an extreme event (in terms of extremely high concentrations), which occurred unexpectedly. The remarkable feature is the exceptional measurement values. The fact that such dust plumes from Ukraine seem to appear infrequently takes, in our view, no ground away from the speculation whether this (admittedly low) frequency might increase further. As is shown now in Section 5.1. many factors need to come together to observe the extreme concentrations over Europe. We have, however, also reworked 5.4. and the conclusions section, and presume that such dust plumes might occur about every 10 years.

"Section 5.3, page 12257, line 8. Unusual stability - that's interesting, but it was not explicitly explained in the manuscript. Are there data on this?"

The prime reason is that the boundary layer is topped by warm air merging from the south, now mentioned in Section 5.1.

"Sections 5.4 and 5.5 make interesting reading, but are not clearly connected to experimental data (or model predictions). Therefore, I feel this can be somewhat shortened as well."

Sections 5.4 and 5.5 were combined and condensed accordingly.

"Conclusions section in general. This is rather a summary or abstract and I don't find

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new conclusions here....”

Done as requested.

“Appendix A (and table). Can the sources of the data be stated in more detail? Are the data publicly available?”

Most of the PM10 data of the 360 stations are — unfortunately, not publicly available in the required form of half hour averages. Most of the institutions provided the data from March 23-25 to us by courtesy.

“Figure 1. Too small and hardly decipherable.”

We originally designed the Figures to comply with the final ACP style (A4 format, two columns). Unfortunately such Figures may turn out extremely tiny in the ACPD style file, which uses landscape paper format. This issue has been discussed with the editor of ACP and will be improved for any subsequent submissions. In any case, the Figures will be much better legible in the final ACP style.

“Figure 3. Too small. Is there an essential difference betwe

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