Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-914-AC1, 2017

Atmospheric Chemistry and Physics Discussions
Open Access
EGU

Interactive comment on "How much information do extinction and backscattering measurements contain about the chemical composition of atmospheric aerosol?" *by* Michael Kahnert and Emma Andersson

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Received and published: 2 February 2017

Below the reviewer comments are marked in blue, our response is marked in black.

The ACPD paper by Kahnert and Andersson deals with the assimilation of lidar observations into a chemical transport model. They investigate how much information about the chemical composition can be extracted from backscatter and extinction measurements and how this information is best assimilated into a chemical transport model.

Overall the paper is very well written and should be published as it is an interesting and important contribution to aerosol research. I only have a few minor comments which

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Discussion paper

the authors may consider for their final paper. I have to say that my experience lies more on the lidar and aerosol optics side than on the information theory / mathematical side, thus I was not really able to review all theory details described in the appendix.
We very much appreciate receiving comments from the lidar and aerosol optics community. The parts that deal with theoretical developments and chemical data assimilation have been very well covered by reviewers 2 and 4. We thank the reviewer for his supportive review and helpful comments!

Comments:

1. It may be beneficial to say a few words about the refractive index and the size bins of the individual species of MATCH. I suggest to add a table with the refractive index of these species at the lidar wavelengths.
   We will add a table providing the refractive indices, and an itemised list of the size bins and the corresponding size ranges.

2. Line 105-109: The description of the MATCH aerosol microphysics module could be shortened as it is not used in this paper.
   It is difficult to shorten these 5 lines. We could only remove them. Then again, we would like the reader to understand that there do exist more realistic optics models, but they are not so straight forward to test, owing to their nonlinearity. Thus the present study is meant as a first step in a larger project, which we will, hopefully, be able to follow up with an investigation of information content based on a more sophisticated description of aerosol optics.

3. Line 118: What about the emissions of the other species? Are they also from EMEP?
   Yes. But EMEP does not deliver gridded emission for black carbon and elemental carbon, only for total primary particulate matter. The sentence in question was

meant to explain how we converted these into gridded emission data for black carbon and elemental carbon. We will reformulate this to make it less ambiguous.

4. Line 134: "an" -> "and"
   Yes.

5. Line 147 "we constrain to better than observation error": It is not clear to me what this means.
   This formulation was also criticised by another reviewer. We will reformulate this part as follows: “Suppose we have an n dimensional model space. Given m observations (e.g., m_1 different parameters at m_2 different wavelengths, so that $m_1 \cdot m_2 = m$), how many independent model variables $N \leq n$ can we constrain with the observations?”

6. Line 151: Remove "the".
   OK. However, we will reformulate this entire section to accommodate the comments by reviewer 2.

7. Line 177: "To be specific" could be removed.
   Agreed.

8. Line 177: Do the results (N_s and H) presented in this section depend on the order of the parameters? If yes, are the changes significant?
   We do not quite understand this question, especially not what the reviewer means by “order”. Is the reviewer inquiring about the *ordering* and *grouping*, or about the *magnitude*? In the latter case, the answer is no, because N_s and H are computed from the scaled Jacobian of the observation operator, which does not depend on the magnitude of the parameters. In the former case, the results do depend on which parameters are being measured, but, of course, not on the ordering.

9. Line 185: "around 7.4 for a single wavelength to around 10-12 for two wavelengths" would be more precise.

OK. This text is likely to change significantly in the revised version, owing to comments by reviewer 2, who asked us to consider different and technically more realistic combinations of observables in table 1.

10. Line 203: I was not aware about the difference between "observation error" and "measurement error". Is this generally accepted terminology? Maybe you can add a reference here so that the reader not familiar with this terminology can see that is used also elsewhere or was introduced by someone (maybe Rodgers?).
We will add the formal definition for the observation error as $\epsilon_o = \epsilon_f + \epsilon_m$ and a reference to Rabier et al. (2002). They use the same terminology as we do, and they also denote the forward model error by ϵ_f. However, they use the symbol ϵ_o for the measurement error, which is potentially confusing. We find it less confusing to denote the measurement error by ϵ_m, and to reserve the symbol ϵ_o for the observation error. We will also mention that there can be other contributions to the observation error, such as representativity error. These concepts are well understood both in the data assimilation and in the satellite remote sensing/retrieval community, but not necessarily among instrument developers, who tend to identify ϵ_o with ϵ_m, while forgetting about ϵ_f. This can be a serious mistake in cases where $\epsilon_f \gg \epsilon_m$, as is the case, e.g., in lidar depolarisation measurements. We find this point sufficiently significant to repeat it, in rephrased form, in appendix B.

11. Fig. 1: The difference between the middle and the right sub-plot is hardly visible. Perhaps you find a better way to visualize it.
We will remove this figure. The regional model is merely used to generate a test case, but we do not address questions of regional modelling or horizontal information spreading in 3DVAR. Therefore this figure conveys no useful information for this study.

12. Line 229 (and at other places): You use β_{sca} and β_{bak} for the backscatter coeffi-

cient. Please use only a single symbol throughout the paper.
Yes, we will correct this and consistently use β_{sca}.

13. Line 241 "the secondary inorganic aerosol (SIA) species are almost completely restored by the 3DVAR": Is it understandable why exactly SIA is restored? Because of the refractive index? Or does it have something to do with the order (index number) of the species in the model?
    This question has also been brought up by other reviewers. We will add a figure in which we show the linear coefficients in the transformation of the control variables in Eq. (C16). Based on this extra figure we will discuss which aerosol components in model space make the dominant contribution to the signal-related variables in the transformed space. This will facilitate the interpretation of the analysis results.

14. Line 274 "there appeared ...": This was not really shown in the paper, so you might remove this sentence or write it in a different way.
    OK, we will strike this sentence.

15. Fig. 3: In this figure the difference between "observations" and "analysis" is much smaller than 10 % (the assumed "observation error"). As this is somewhat unexpected (but understandable as an optimization is applied) you may add a brief discussion about the effect a "measurement error" (noise) would have. Because of the assumed linearity this probably is not very difficult to explain.
    OK, we will add the following text. “In fact, the difference between the observation-equivalent analysis and the observations deviate by even less than 10 %. However, our tests confirmed that an increase in the observation error eventually results in analysis results of which the observation-equivalent increasingly deviates from the observations (not shown).”

16. Fig. 4: Could it be of interest to see which aerosol species (size bins) the individual variables represent? What would be the effect of changing the order of the

species?
We will add an extra figure that will show at least a selection of aerosol species in specific size bins. In response to reviewer 2 and 4, we will even show a comparison with an unconstrained analysis. This will likely make it clearer that the constrained analysis reduces the noisiness of the analysis, since it is being constrained to assimilating signal rather than measurement noise.

We do not understand the last question about changing the “order of the species”.

17. Line 277: "to be sure" could be removed.
Agreed.

18. Line 314: I think some aerosol species exist for which assuming externally mixed spheres is not that wrong.
It is unclear what kind of species the reviewer refers to. Certainly not dust or black carbon (BC). Sea salt is either mixed with water, or else it is nonspherical. Organic carbon (OC) and secondary inorganic aerosols (SIA) are rarely found in pure form. They are often mixed with each other, with water, NaCl, and even BC and dust. Even nucleation-mode particles are often the result of at least binary nucleation involving more than one species. In our more realistic aerosol microphysics model there is not a single size bin in which liquid-phase (i.e., spherical) aerosols consist of a single compound. We therefore prefer to keep the text in its present form.

References

Rabier, F., Fourrié, N., Chafaï, D., and Prunet, P.: Channel selection methods for infrared atmospheric sounding interferometer radiances, Q. J. R. Meteorol. Soc., 128, 1011–1027, 2002.

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