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Atmospheric Chemistry and Physics Discussions

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

1 General comments

1. This paper details an interesting way to assess the information content in lidar measurements of aerosol backscatter and extinction with respect to model as-

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similation. It also demonstrates how this knowledge may be used to optimize the incorporation of lidar measurements in the model. This is a very interesting and relevant topic. Assimilation of lidar data into models is a field that is still developing rapidly, with a few different groups using very different techniques; therefore, well designed research into how best to use lidar data is very valuable. It is also potentially informative to the lidar community, since work must begin soon to design the next satellite lidar instruments if the lidar record is to continue. The choice of which measurements and which wavelengths to include has a large bearing on cost and technological difficulty, so having quantitative information about which measurements are most useful for improving models is critical. To that end, I would like to suggest some additional cases for Table 1, please see the specific comments below.
We thank the reviewer for the considerably thorough and supportive review, which will help us to improve various aspects of the manuscript. Our detailed response to the review comments follows.

2. The paper is well written with very nice clarity. However, the overall organization is somewhat difficult. The current organization consists of a very streamlined and easy-to-read main text with five very technically dense appendices. While the main text is pleasantly easy to read on the first pass, there is too much information missing. While it's appropriate to include extra, more detailed information in appendices, the main text still needs to be able to stand on its own, and in my opinion, it doesn't quite. I would suggest that the main equations and brief explanations should also be included in the main text, including all the equations that a reader would need to apply to calculate the kinds of results presented in this work. The appendices also include a lot of pedagogical development; this is the kind of information that I think rightly belongs in the appendix for readers who want more details. Since the appendices are 5 different topics, I also suggest that each appendix should be exist as a separate entity, with all variables defined, so

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that a reader can read Appendix D to learn about the application of constraints or Appendix E for the "practical aspects" without a close reading of Appendix A,B, and C, to find the definitions of the variables.

The organisation of the paper is indeed a delicate issue that was also brought up by other reviewers. Our main goal is to make this paper accessible to a broad community, including lidar instrument developers, remote sensing groups, and data assimilation researchers. For this reason, we prefer to include most of the theoretical developments in the appendix. However, we agree that this creates a significant problem by removing essential information from the main body of the paper. In the revised paper we will follow the reviewer's suggestion and re-state the most essential theoretical results from the appendix in the main text. This will make the paper more readable and self-contained, while avoiding the risk of making it too technical, which could narrow down the readership of this work.

3. The results and conclusions are also a little too abbreviated. Some key aspects are missing, like how was the specific weighting chosen and how do we know this is the best weighting? Also, as pointed out by another reviewer, the assessment (section 3.2) is really more of a demonstration. That is, although the theoretical development is compelling, the application/assessment section isn't sufficient to convince readers that this is a better way to assimilate lidar data than another way. This paper clearly reflects a lot of research on the part of the authors and I think the missing information probably exists but was left out in the effort to streamline the manuscript. I think adding this additional information should be fairly straightforward and would improve the usefulness of this research for the modeling and lidar communities without adding too much complication to the nice flow of the paper.

   This is also an important point, which was brought up by several reviewers. We will perform additional computations using the unconstrained assimilation algorithm and compare the constrained to the unconstrained analysis. The hypothesis

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is that the constrained analysis should be less noisy, because the unconstrained analysis is at risk of assimilating noise. Also, we will eliminate all instances of "numerical experiment" and replace it by a more appropriate term, e.g., "numerical test", "demonstration", or "illustration". Finally, we will add more explanations to Sect. 2.4 about the construction of the covariance matrix in the constraint term.

2 Specific comments

1. Lines 151-158: Here is an example where I think some important things are missing from the main text which only appear in the appendices. These eight short lines are the methodology section for the key calculations that are the novel part of your research and are critically important for a reader to understand. I suggest that a way to decide what should also be included here would be to target the subset of equations that a reader would need to apply to calculate results like yours, but without their derivations. Also include enough supporting explanation to describe what the equations say and how to use them.
   We agree, and we will make changes following the more detailed suggestions given in the following comments.

2. L152-153: Specifically here, Eq C6 and C?16 should be included in the text, since they are required to understand the meaning of the sentence. Later, at L159-160 where readers are directed to the appendix for more background information, I think that's fine.
   OK, we will revise the text and include the equations for the observation operator, the observability matrix, and the singular-value decomposition thereof.

3. L155-157: The equations for signal degrees of freedom and Shannon information content should also be included in the text.
   OK, this will be added with accompanying text.

4. L165: "a numerical experiment". In fact, it's more of a demonstration than an experiment. It's useful as a demonstration of the results of the technique, but there's nothing in the demonstration that addresses a hypotheses. Sharing more of the background work would make the paper more compelling. For example, as another reviewer suggested, comparing to a control experiment would be necessary for convincing readers that this technique is useful. For another example, a pair of runs with different weightings in the assimilation would help answer the question of why the weighting that was ultimately chosen was the best one.
   We will replace "numerical experiment" everywhere in the paper, as mentioned previously. Next, we will show a control run with the unconstrained assimilation system. The hypothesis is that the constrained analysis should be less noisy than the unconstrained analysis. We will revise the Figures and show both the unconstrained and the constrained analysis in the same plot. Also, we will add an extra figure to show both analysis results for different aerosol species in different size bins, as these are even more sensitive than size-integrated total mass mixing ratios. Finally, the case we picked in the original manuscript was not particularly challenging, since the background state was fairly close to the reference state. In the revised paper, we will pick a more challenging case in order to make the differences between both analysis runs as clear as possible. As for the different weightings, our tests, so far, indicate that the different approaches result in rather similar analysis results. So, the constrained analysis is not as strongly dependent on the weighting as one may expect. We will clarify this point by adding a discussion to Sect. 2.4.

5. L177: Depolarization is not included in the studied parameters, yet lidar studies have shown that depolarization measurements contain some information about aerosol composition (for example, Omar et al. 2009 as referenced in the introduction, but there are many others). Do the authors have any comment on depolarization and why it isn't included in this study?

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There are two major problems. The obvious practical problem is that the forward model would need to be based on nonspherical particles (as spherical particles do not depolarise). However, our simpler optics model is entirely based on spherical particles, while our newer optics model only accounts for the nonsphericity of bare black carbon, but not for that of mineral dust or dry sea salt. Thus our capabilities of modelling depolarisation are presently limited. The second problem is that the observation error for depolarisation may be very high, even though the measurement error is very low. This is because the forward-model error is likely to be quite high, since even slight variations in particle geometry (e.g. Kahnert et al. (2012)) or inhomogeneity (e.g. Kahnert (2015)) can result in large variations in the depolarisation ratio. If the forward-model error is, indeed, high, then the prospects of using depolarisation for constraining CTM model results are likely to be low. However, this question is open and will be investigated in future studies. But in order to do so, one would first need to obtain estimates of the forward-model error (e.g, by computing depolarisation ratios while varying particle morphology).

6. Table 1 and related discussion: From a lidar standpoint, some combinations of channels are more technologically affordable than others, so the discussion of which channels add significant information content is very interesting. However, the utility for the lidar community would be maximized if the combinations were ordered such that they roughly increase in technological difficulty. Also, some combinations don't really make sense from a technological standpoint. There is no lidar that measures extinction but not backscatter at the same channel (although modelers may use only the extinction). On the other hand, backscatter (actually attenuated backscatter) without a direct measurement of extinction is common. Also, since CALIPSO, CATS, EarthCARE and the $3\beta + 2\alpha$ combination of airborne HSRL2 are mentioned in the introduction and motivation sections, it would be useful if the combinations relevant to those instruments were

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included. CALIPSO = CATS =$\beta(\lambda 1)+\beta(\lambda 2)$. EarthCARE = $\beta(\lambda 3)+k(\lambda 3)$. HSRL2 =$\beta(\lambda 1)+\beta(\lambda 2)+\beta(\lambda 3)+k(\lambda 2)+k(\lambda 3)$. I would suggest these combinations of backscatter and extinction would be most interesting and useful to the lidar community: $\beta(\lambda 3)$
$\beta(\lambda 1)+\beta(\lambda 2)$
$\beta(\lambda 1)+\beta(\lambda 2)+\beta(\lambda 3)$
$\beta(\lambda 3)+k(\lambda 3)$
$\beta(\lambda 1)+\beta(\lambda 2)+k(\lambda 2)$
$\beta(\lambda 1)+\beta(\lambda 2)+\beta(\lambda 3)+k(\lambda 2)+k(\lambda 3)$
For these experiments, it appears that the observation error was always assumed to be the same in every channel. I think it's a reasonable assumption, to first approximation, that the measurement error would be similar in every channel, but as pointed out at L78-79, some lidar retrievals include additional non-random errors that can be much larger. This could and should affect the choice of channels to assimilate. For example, the Raman, HSRL, and transmittance techniques are fairly direct measures of extinction, but techniques that require an inferred lidar ratio to convert backscatter to extinction have relatively little additional measurement information content in the extinction.
We welcome the reviewer's suggestion to take technical realisations of lidar systems into account, and we will revise Tables 1 and 2 according to the reviewer's specific suggestions. We will also add a comment on the observation errors of lidar measurements, specifically on the fact that the observation errors may be different for different channels/parameters.

7. L 197-201. Here also the discussion of incorporating soft constraints and the specifics of the three weighting schemes should be in the main text of the paper and not just the appendix, since it is discussed here in the results section. This section is not understandable without the equations from the appendix and most of section D3.

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We will remove this discussion here. Instead, we will briefly discuss the construction of the constraint covariance matrix in Sect. 2.4.

8. L 203-204. Discussion of observation error vs. measurement error. This is interesting and useful, but could be clarified as to whether the forward model error (due to poor assumptions) is considered part of the observation error or is another separate source of error. If it is part of the observation error, how are the forward model errors represented and how are they transformed into the space of the measurement vector?
We will extend the text to clarify that the observation error is given by $\boldsymbol{\epsilon}_o = \boldsymbol{\epsilon}_m + \boldsymbol{\epsilon}_f$, where $\boldsymbol{\epsilon}_f$ denotes the forward-model error. We will also add a citation to the paper by Rabier et al. (2002) with a hint to their Eq. (1), which explains this terminology. A way to determine the forward-model errors theoretically is to perform light-scattering calculations while varying various parameters, such as particle morphology, refractive index, and size distribution within typical uncertainty ranges. This can provide us with an estimate of $\boldsymbol{\epsilon}_f$. To the best of our knowledge, it would be very difficult to determine $\boldsymbol{\epsilon}_f$ with experimental methods.

   We are not sure if we understand the last question. $\boldsymbol{\epsilon}_f$ enters into the definition of the observation error covariance matrix, i.e. $\mathbf{R} = \langle \boldsymbol{\epsilon}_o \cdot \boldsymbol{\epsilon}_o^T \rangle$, which is a matrix in the space of the measurement vector. No further transformation is necessary.

9. L 207 While there may be retrieval errors in the lidar backscatter and extinction due to assumptions, assumptions on particle shape and size distribution are not among the assumptions used in lidar retrievals. These examples belong only to the optics model (forward model). So, perhaps delete “also”. Poor assumptions in the optics model or in lidar retrievals would presumably lead to bias errors, whereas measurement errors would more typically be random. Does this make a difference in the analysis?
OK, we will delete “also”. We would generally not be sure if assumptions in the optics model necessarily (mainly) lead to biases. For instance, model errors may

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be dependent on size and morphology of the actual particles. The errors would, correspondingly, fluctuate over time. The amplitude of this fluctuation may well be larger than any possible biases. However, in case that the forward-model does introduce a large bias, than this would, indeed, be a problem, since analysis algorithms are typically based on the assumption that the errors are unbiased.

10. L 219. I strongly agree that estimating the uncertainties in the optics model is very important. Some discussion here seems warranted about how that can be done. Later I see that this is discussed in the summary (L281 – 292) but I think it would be better if it comes up first here in the discussion section.
    Agreed. We will add an explanation, but we will also mention it again in the conclusion section.

11. L 256 and caption to Fig 4. In both places, it would be kind to remind readers that the delta notation in $\delta x'$ means this is the difference between the value and the background value.
    It is not so simple. δx in physical space is the difference between the value and the background, while $\delta x'$ is obtained from δx by applying the transformation $\delta\vec{x}' = \mathbf{V}_R^T \cdot \mathbf{B}^{-1/2} \cdot \delta\vec{x}$. We will repeat this definition in the text with a reference to the definition (which is now found both in the main text and the appendix), and we will add a reference to the defining equation both at this point in the text and in the caption to the figure. But we think it would be a bit overdone to repeat the equation in the figure caption.

12. L 259-263. The choice of D21 with its sharp drop-off in weighting appears to mean that only one transformed variable is allowed to change in a meaningful way, although the measurement scenario chosen has nearly the maximum amount of information content available, close to DOF=4. Why was D21 chosen instead of D18, which would allow the measurements to play a bigger role? The only discussion of this choice is the rather vague comment in the Appendix “it is a

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matter of experience to test different approaches and select the one that proves to be most suited". How and why was this approach determined to be the most suited?

We have done some additional tests and found, in fact, that the analysis is less sensitive to the choice of weighting than we expected. We will explain this in the revised paper in Sect. 2.4. Also, we will do the following changes to Fig. 4. First, we will show $\delta x'$ for both the constrained and the unconstrained analysis. Thus the whole discussion of the figure will shift from a mere description of the behaviour of the constrained analysis to a comparative discussion. This will make it much clearer what kind of effects the weak constraints have on the analysis increments. Second, following a suggestion by reviewer 4, we will not show all 20 panels, but only a subset of panels sufficient to illustrate the different behaviour of signal- and noise-related (phase-space) model variables. Third, as mentioned earlier, we will pick a more challenging case in which the reference and background results differ more strongly than in the case we originally picked. So this figure will be changed considerably, and the accompanying discussion will become a lot more informative.

13. Comparison of Figure 3 and Figure 2, if I understand right, underscores the fact that there is a significant null space, not controlled by the measurements, since essentially the same measurements in Fig 3 correspond to both the black and red lines in Fig 2. What is not clear to me is what happens in a standard assimilation to the variables that are not well controlled by the measurements? Do they remain close to the background values, or do they vary wildly and arbitrarily? If the former, then the exercise of determining the singular values wouldn't help the assimilation very much (but would still be useful in terms of building knowledge about what we can and can't actually measure). On the other hand, if a standard assimilation arbitrarily varies state variables in the null space, then this is a very important motivation for this technique (and maybe that motivation could be

emphasized a little bit more in the introduction and conclusions). Not being very familiar with the field of model assimilation, I guess but don't actually know that there must be other "regularization" techniques in use to prevent an assimilation from arbitrarily varying parameters that are mostly in the null space of the observations, although I imagine existing techniques may be more ad hoc than the method presented here. Can you comment on other methods and demonstrate how this method performs better than other methods?

The reviewer's comment about the null space and the behaviour of the unconstrained (standard) assimilation raises an important issue. As mentioned earlier, we have now run an additional unconstrained assimilation, and we will show a comparison of both methods. Figure 2 will be replaced by two figures. The first figure will, similarly to the old figure 2, show the total mass concentration of different aerosol species, but now for both the constrained and the unconstrained analysis. The second figure will show a similar comparison of a selection of aerosol species in specific size bins. We anticipate that this comparison will illustrate that the unconstrained analysis yields more erratically varying vertical profiles (i.e., results that vary more wildly in the null-space).

As for ad hoc methods, we did review previously reported approaches in the introduction, such as the one by Benedetti et al. (2009) (L 53-54) based on constraining the total aerosol mass mixing ratio, and the one by Saide et al. (2013) (L 55-56) based on constraining the mass mixing ratio per size bin. One obvious disadvantage is that these approaches are quite inflexible. The number of constraints is fixed in these methods, so one cannot easily adapt the number of constraints to the number of independent measurements to be assimilated, as we can in our approach. (In fact, our method automatises this process.) Also, the available information may not be optimally exploited by these methods (L 57-59). We have not tested such methods, so we cannot comment on their performance. However, we also believe that the burden of proof for such a demonstration does not lie with us. We are employing a mathematically well-founded approach based

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on information theory. If other groups choose to not follow us, but continue to use ad hoc methods (which, admittedly, may be quite attractive owing to their simplicity), then it is up to them to demonstrate that such ad hoc methods yield sufficiently accurate results while exploiting the available measurement information. Owing to the ad hoc nature of these methods, such a demonstration would have to be repeated for any new set of measurements to be assimilated. Our method can serve as a reference for such tests.

14. L 298-299. “It also appeared”. This result is disappointingly empirical for such a well-founded theoretical study. This observation that SIC was most faithfully retrieved was made in a single case– would you expect this result to be general for all cases, and why? Answering the question is complicated since the singular variables are defined only in the transformed space and therefore the information about what variables are or are not constrained by the measurements is only in this transformed space, not the state space. Yet this statement highlights that it’s desirable to have information about which chemical species and size bins are constrained by the measurements. Is there any way to provide information about this quantitatively? For example, since each state variable is a linear combination of the transformed variables, would showing the linear coefficients in a table make it more obvious which state variables are most closely related to the most significant transformed variables? Perhaps there is a way to use the coefficients to calculate a “fractional significance” that would indicate that x% of the variability in a given state parameter is orthogonal with significant transformed variables while (1-x)% is orthogonal with insignificant variables?
This is a very good suggestion. We will add an extra figure with accompanying discussion and show the magnitude of the linear coefficients for the signal-related control variables. However, the coefficients will depend on the B- and R-matrices, which vary spatially. So, we do not anticipate that we can draw very general conclusions from a single test case. But we do think that such a discussion can help

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us understand why the analysis behaves the way it does in our specific case.

3 Minor comments

1. L37: Muller et al. 1999 and Veselovskii et al. 2002 and related papers (there are many) would be more relevant references here since they detail retrievals of refractive index, etc., from lidar. (Mishchenko et al. 2007 is an introduction to the Glory satellite and was about retrievals from a polarimeter.)
   Müller, D., U. Wandinger, and A. Ansmann (1999), Microphysical particle parameters from extinction and backscatter lidar data by inversion with regularization: theory, Appl Optics, 38(12), 2346-2357, doi: 10.1364/AO.38.002346.
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   Agreed. The references will be replaced.

2. L99: I infer that the ratios in the different size bins are fixed, or else there would be much more than 20 total variables. Is there a way to concisely clarify this in the sentence?
   There is no way to say this in a simple sentence, because it is not quite as simple as the reviewer suspects. We have gridded emission data, which means that the ratios among size bins can vary from one grid cell to the next. Although the mass-transport model does not account for microphysical processes (such as condensation, which would result in a dynamic evolution of the size distribution), this ratio can still dynamically evolve in each grid cell owing to transport processes and mixing of air masses originating from different emission sources.

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3. L109: maybe replace “in the present setup” with “currently in that version”. “The present setup” seems to refer to “the setup used in the present study” but that is misleading, since the present study uses the 20-variable version of the model.
   Agreed.

4. L134: “an” should be “and”
   Yes.

5. L142: “Error correlations ::: are not assumed to be separable”. I’m not sure what this means. What is (or is not) separable from what?
   Vertical and horizontal correlations are often assumed to be separable. We do not make such assumptions, because vertical correlations are often stronger on larger horizontal length scales. In our spectral model (where the horizontal correlations are Fourier-transformed) this means that vertical correlations are larger for smaller horizontal wavenumbers. Since this is not so essential in the context of this study (and potentially confusing), we will remove this text in L 142.

6. L153: “see Eq. D16”. Should this be C16?
   Yes. However, following earlier suggestions by the reviewer, this text will now be revised and supplied with the main equations from the appendix. So the text in its present form will be replaced.

7. L162-164: Should this sentence perhaps be part of section 2.4, as part of the description of the new technique? The rest of this paragraph (L164-174) is more about the demonstration of the new technique and so seems like a somewhat distinct topic.
   Agreed, we will move this text.

8. Figure 1: The caption says “note the nonlinear colour scale” Actually, the scale is hardly visible. Please expand the axis labels so they are a similar text size to the caption text.

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Actually, we think that this figure is not particularly relevant in the context of our study, since we do not consider aspects of regional modelling or horizontal information spreading in the analysis. It merely shows one out of many model variables in a single model layer, which does not convey much useful information. Also, since we consider a single profile, the analysis impacts the mass mixing ratio only at and around the observation site, which is difficult to see in a regional plot. We therefore suggest to remove this figure in the revised manuscript.

9. Figure 2: The axis labels' and inset box labels' font size should also be increased here.
OK, we will increase the font sizes in all figures wherever necessary.

10. L 391. The variable n is not defined. Possibly this is the only case, but I would also request that variables be re-defined frequently when used in key equations. If a reader is directed from another part of the paper to Equation D18 or C12, for example, then it would be nice if all the information relevant to understanding that equation is given immediately after that equation, rather than having to scroll through 8 or 10 pages to relocate the definitions of key variables.
Agreed, we will add the definition of n. Also, the problem with directing the reader to equations in the appendix will be significantly alleviated in the revised versions, since we will re-state the key equations in the main body of the paper (see our response to an earlier comment).

11. L563. The symbol lambda is used for wavelength elsewhere in the text. You might consider using a different symbol here.
OK, we will replace it by mu.

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