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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Estimation of aerosol particle number distribution with Kalman filtering – Part 2: Simultaneous use of DMPS, APS and nephelometer measurements" *by* T. Viskari et al.

Anonymous Referee #1

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Summary:

Firstly, it is not really possible in my opinion to review Part 2 of this paper without also reviewing (or otherwise validating) Part 1 to some extent so I have read both and whilst trying to keep this review relevant purely to Part 2, I will have to talk about Part 1 to some extent.

This study is an interesting analysis of the utility in applying extended Kalman filtering techniques to multiple aerosol sizing measurement datasets. Part 2 discusses simultaneous use of 3 quite different instruments whilst Part 1 lays out the method and validity for one (or rather 2 very similar DMPS instruments). Kalman filtering is a useful

technique in data assimilation and is applied to a range of atmospheric measurements where Gaussian measurement and error statistics can be assumed and a good forward model can be employed. However, I'm having a lot of trouble in reconciling that either Gaussian statistics or a relevant model can be employed here in the context of the measurement of ambient aerosol size distributions (and I stress "ambient" as opposed to closed airmasses, e.g. chamber data), especially in the coarse mode (super-micron), which I'll come back to further in the general comments below. This is without the subtleties of filtering over the multiple instruments' varying measured size spectra which I shall not go into in detail.

The general subject context of the paper is relevant to ACP and the paper is generally well-written and figures etc are of a good quality. However, the scientific conclusions (of both parts 1 and 2) are not satisfactorily justified, essentially incomplete and partially invalid in this reviewer's opinion. I will detail why below. There are many major (and subtle) reasons why I believe Kalman filtering cannot be applied to ambient point-sampled aerosol sizing measurements (especially for super-micron sizes). But having said this, I do believe there may be merit to the method under very specific ambient conditions and for a limited aerosol size range but this is highly contrived and to get this right in this publication would require major revisions. Without this, I worry that this method could be used inappropriately by others on already complex and noisy measurement datasets that could then become completely meaningless. I recommend major revisions (to both parts 1 and 2) which would better validate the applicability and limitations of this method under real ambient conditions with some guidance on when the method might be useful over simply using the measurements themselves and for what purposes. I don't think, in this case, that Kalman filtering can add anything useful to the measurement time series as the model in use is not designed for a changing background.

In my opinion, a complete rethink is required.

General comments:

1/ Airmass changes: Pivotal to the utility of Kalman filtering in general is the ability and applicability of the physical model and the Gaussian nature of the measurment. Here, the UHMA model is used – a well validated and proven aerosol box model. This is a great box model that is useful in calculating theoretical changes to aerosol size spectra through microphysical processes, coagulation, deposition etc. But it is only useful in a chamber (or analogous) or Lagrangian environment where good constraints on precursors, thermodynamics and deposition processes are well known. It is completely useless for ambient point sampling locations where the many controlling factors are unknown (as they are in this study) and/or when the airmass itself (in terms of background aerosol and precursors) is changing rapidly when compared to the timescale of the model processes (as they would be expected to over the stated 10-minute interval), i.e that the measurement itself is not at all Gaussian. The authors seem to have noted this themselves (I think, though the statement Is confusing) in part 1 (p.18856), stating that UHMA "...enables very detailed time evolution of aerosol processes but lacks the spatial aspects, similar to point-wise atmospheric aerosol measurements" but this limitation seems to be subsequently ignored.

In essence, the authors seem to be missing the point that ambient airmass background (at a point sampling location) can change much more quickly over ten minutes (the time interval used) than any microphysically-driven changes happening in a closed volume which has long since blow away over that time. The point measuremet is in the Eulerian frame of the atmosphere, whereas the UHMA model is appropriate only in the Lagrangian. This renders the utility of the UHMA model generally useless in this context, except for the most static of atmospheric conditions.

Where there is arguable utility in employing UHMA for ambient point measurements would be where the atmosphere is relatively static or where airmass composition is known to be otherwise invariant (e.g. remote locations far from sources) with special applicability to new nucleation events where UHMA comes into its own. But this is still only valid for the fine and ultrafine size modes and not for the coarse mode (see below).

2/ Instrument errors and coarse-mode error: The authors have correctly realised that DMPS and APS instruments cannot be intercompared easily and that significant systematic differences can exist in their overlapping size ranges. They have also spotted well-known electronic gain issues in the first channels of the APS and the problems with counting statistics in the largest sizing bins of the APS and they have taken reasonable steps to handle many of the issues. However, there is a subtle problem here in the way that measurement error is treated by the Kalman filter for the coarse mode in that errors on the low counting statistics of sizes generally larger than 1 micron become increasingly Poisson in nature and hence invalidate the Gaussian error assumption. This happens because of the finite sampling volume of the APS and the typically low concentrations of coarse-mode aerosol , meaning that point data are often 0 or 1. To overcome this would require smoothing over many data-points but we then run into the problem of non-Gausssian airmasses once more (see next point below). To overcome this problem and to allow matrix inversion stability, the authors have arbitrarily imposed a logic rule that the concentration in any size bin is either fixed to a minimum of 10ˆ-6 cmˆ-3 or that the concentration cannot decay by more than 90% of the previous measurement in any size channel between successive timesteps. This is completely invalid and would greatly amplify natural large spikes in coarse mode aerosol and introduce lags in the time-series and therefore potentially important positive biases in total aerosol mass. And despite this positively biasing constraint, we still see very large under-representation of the coarse mode in Fig 3 and 6 (see point 3 below).

3/ Further coarse-mode problems: Figures 3 and 6 appear to show a complete mismatch between the EKF estimate and the APS measurements above $\sim$5 microns with the APS being strongly positively biased. The authors have recognised this and have noted it on P. 18902 and vaguely conclude that "...we assume the method to be valid also over the majority of the APS measurement area.". I think a limit needs to be put forward explicitly here and I would recommend 1 micron from Figures 3 and 6. I would imagine that UHMA has a gravitational settlement term that is causing the EKF to decay to zero toward 10 microns. But as UHMA doesn't concern itself with the wind that

may be blowing in new coarse-mode aerosol or whipping up such aerosol from the ground local to the measurements site, it is simply not useful to use it and therefore to employ Kalman filtering on a measurement that will never be Gaussian due to both the instrument response and the ambient background change over the time interval considered.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 18889, 2012.