Review of Christiansen et al.
Differences in fine particle chemical composition on clean and cloudy days

This manuscript looks at differences in aerosol data from the IMPROVE network on clear and cloudy days. The difference is important because satellite optical depth measurements only work on clear days and could therefore be biased if aerosol concentrations are systematically different on cloudy days. Since satellites measure aerosol at ambient RH, such a bias could arise both from differences in the amount of dry aerosol and in its water uptake.

The concept of this manuscript is an excellent one – a simple study that fills a gap in the literature and has relevance to satellite measurements. It absolutely should be published in some form. The manuscript, however, could be much, much better if the logical flow were better and the figures were better organized.

Specifics:

Lines 118-119. I disagree with not including the water uptake by organic aerosol. Yes, it is not known as well as for sulfate and nitrate. But ignoring it will give a worse answer than putting in a reasonable value. It is known that the kappa for organics in relevant rural aerosol is often something like 0.2 or 0.3 (as just one example reference Chang et al., 10.5194/acp-10-5047-2010). I looked up the Jathar and Metzger references and they do not say that the OC water uptake is not known well enough to put in a best estimate.

Lines 212-220 (and elsewhere for species other than TOC): This paragraph follows a frequent bias in the literature by talking more about sources than sinks. Aerosol concentrations are also higher during clear sky periods because removal by precipitation is more frequent in cloudy sky periods (e.g. Grandet et al. doi:10.5194/acp-13-3177-2013; and later paper by Gryspeerdt). On the source side mentioning both photochemistry and stagnation events is good. I would also suspect that fire frequency is important for differences between clear and cloudy periods, maybe especially in the eastern US where there is small-scale agricultural burning.

My two big comments are about the figures and the logical ordering of the manuscript. Although it is there in the text if you read really carefully, the overall manuscript doesn't really present in a logical order but instead jumps far too quickly to aerosol liquid water (ALW). I kept wanting to see the differences in concentration shown before the next step of computing ALW. Most of the figures for the concentration differences mysteriously omit organics, one of the most abundant species. Finally, the relevant quantity for comparing to satellites is not ALW. It is the wet aerosol (dry plus liquid water). No existing satellite can measure aerosol water content – so why choose this as the basis for your analysis when the motivation for the entire project is biases in satellite retrievals?

You will have a much better paper if you organize the results by first showing the differences in concentrations, then showing hygroscopicity and how that translates to wet aerosol. A good

example of the poor organization is that Figure 1 is about ALW, which is a derived quantity that uses the information in Figure 2. Any reader just skimming the paper would get confused seeing Figure 1 come first. And even within figures information is poorly organized. In Figure 1, why isn't "mixed" in between clear and cloudy? In Figure 2, why is RH in between PM2.5 and its constituents?

In summary, start with the concentration differences as measured by IMPROVE, then extend those differences to ambient RH, as measured by satellite retrievals.

Another part of the paper that really needs work is the figures. Some figures simultaneously have too many panels and don't convey enough information. Figures 3 to 7 are almost illegible.

If it stays, Figure 1 could be done as one panel with grouped bars (just to be clear what I mean, googling "grouped bar chart" will show what I mean – I'm sure you use them all the time). Except you don't really need Figure 1 – it could be combined as a last column in Figure 2, more like Figure 8.

Figure 2 could be done in 4 panels, not 16. Except it should be 6 panels – it should include TOC and wet aerosol. The columns should be organized logically One good way to organize would be left-to-right to show SO4, NO3, and TOC, then dry PM2.5 (and label it "dry" for clarity), then RH, then wet aerosol. Figure 8 should follow FIgure 2.

Figures 3 to 7 are extremely hard to read, even at high magnification on the screen. And the color coding is a poor choice for quantitative information. There is research showing that bar and line graphs are read more accurately than are color codes or pie graphs. Readers can correctly discern quantitative changes in bar and line graphs that they can't accurately judge in other formats. Contour plots are good, too. Also, using five repetitive figures (3 to 7) doesn't really work very well. I'd try very, very hard to make some sort of bar graphs that are tied to a map and to put more than one species on the same plot. For ideas look at the way the IMPROVE data were plotted with bar graphs tied to a map in Hand et al. 2012 (doi:10.1029/2011JD017122) or line graphs superimposed on a map in Murphy et al. 2008 (www.atmos-chem-phys.net/8/2729/2008/).

I can't say I can see exactly how you should do the map plots. Please understand as a reviewer one sees figures that work well and figures that don't work. Your figures 3 to 7 don't work well.

There is distracting use of color: for there is no reason why the column headings in Figure 2 should be in colored fonts, and no particular reason why the "clear" should be brownish and the "cloud" should be blue.

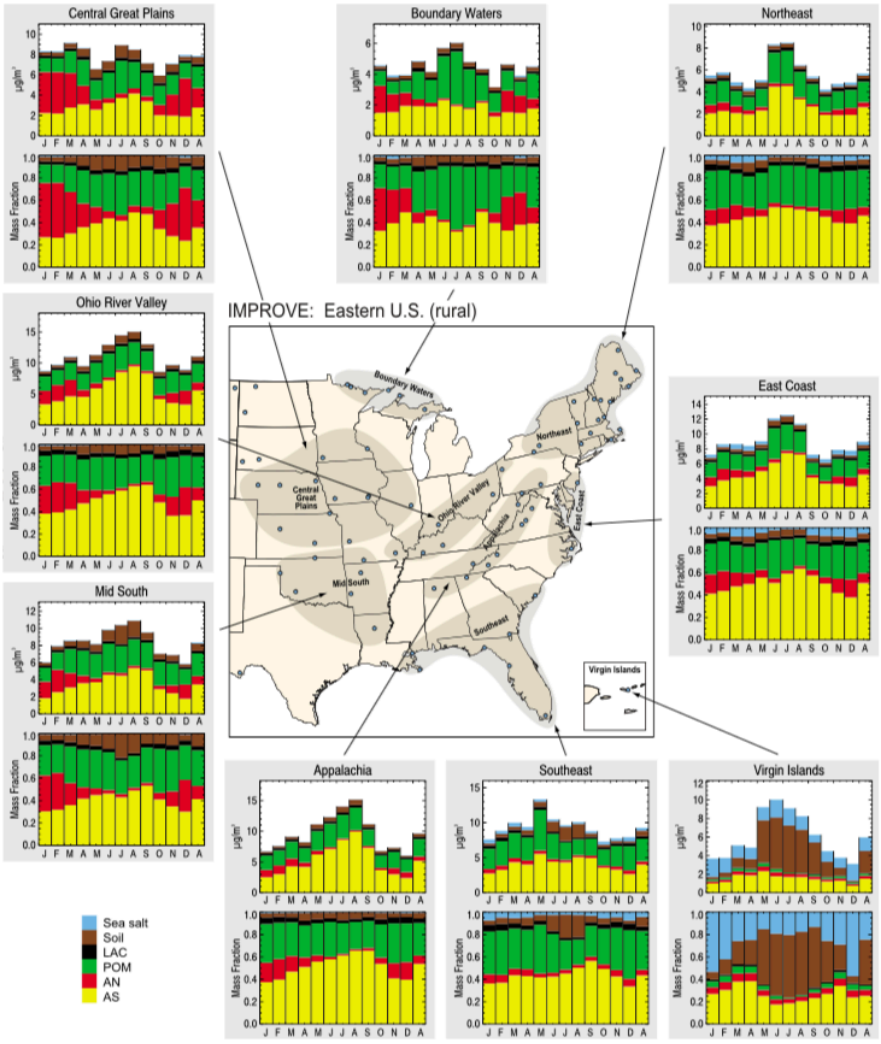

Figure 2a. IMPROVE 2005–2008 regional monthly mean $PM_{2.5}$ mass concentrations ($\mu g\ m^{-3}$) surrounded by $PM_{2.5}$ reconstructed fine mass fractions for the eastern United States, including the Virgin Islands region. The letters on the *x* axis correspond to the month, and "A" corresponds to "annual" mean. Ammonium sulfate (AS) in yellow, ammonium nitrate (AN) in red, particulate organic matter (POM) in green, light-absorbing carbon (LAC) in black, soil in brown, and sea salt in blue. The shaded area corresponds to the regions that comprise the sites used in the analysis, shown as dots.

Including this here isn't about the content in this figure from Hand et al. Instead, I'm putting this in as an example of how you might try to plot your data to be more legible than Figures 3 to 7 in the manuscript. Instead of the bar plots of composition by month you could have clear/cloud data for major constituents or something like that.