This paper describes the use of a four dimensional data assimilation technique to assess the uncertainties in black carbon emissions over California. Extensive mathematical relationships associated with the adjoint technique are presented and they focus on two cases associated with the ARCTAS-CARB measurements to improve estimates of BC emissions. In general, the text is well written and the science sound; however, some of the motivation and conclusions are not articulated very well. In addition, I have a number of comments that need to be addressed before the manuscript is suitable for publication.

Major Comments: 1. Level of Detail: Extensive details associated with the mathematical relationships associated with four dimensional data assimilation will likely reduce
the overall target audience. While the air quality community is interested in methods
to improve emission estimates, I would expect that much of those details will not be of
much interest to the same community. More importantly, it was not clear whether the
details on the adjoint are the same as those presented previously in the literature or
whether they are new. If the relationships are typical of those of previous adjoint pa-
pers, perhaps more of the details could be put into the appendix. The authors should
more clearly differentiate what is new and what is not new.

2. As described in a few of my specific comments, the discussion of what type of observ-
ations would be desirable to further constrain the adjoint technique and improve the
emission estimates. The aircraft flights that targeted the fires were obviously critical,
but would other flight paths be more useful? Or would more cases be useful? Were
there only two periods during ARCTAS-CARB that were useful, or were the number of
cases examined more limited by the computational expense of adjoint techniques?

3. The authors acknowledge that other meteorological factors will affect their analysis.
They also mention that mostly clear skies were observed over California, so that com-
plex cloud processes (i.e. wet removal) did not occur in this study. Near the end of the
paper they mention it would be useful to have even simpler meteorological conditions to
reduce uncertainties in meteorology. But it would seem very difficult to find such cases
and one would have to confront complex real-world conditions at some point anyway.

Specific Comments:

Title: ARCTAS is misspelled.

Page 1, line 7: Consider changing “multiple” to “three”. The next line lists 3 inventories.

Page 1, line 9: Change the use of “x” and through out the text to write out what they
actually mean in terms of a change. I find the usage in this particular sentence to be
confusing.

Page 3, lines 25-35: The motivation of why ARTCAS-CARB campaign is used for their
analysis should be improved. The way the paragraph is phrased, it basically just says they are going to use this particular campaign. But this could be changed to upfront state that the campaign had aircraft measurements characterizing both anthropogenic and biomass burning sources of BC and therefore would be useful to test their adjoint-based technique.

Page 4, line 17: Change “also turned off” to something else. Since it is not available, it is not possible to turn off that option. Also large fires could significantly affect meteorology by dramatically reducing incoming shortwave radiation, so some of the uncertainties in the adjoint technique will be due to this process that is neglected – in addition to the other meteorological processes they note.

Page 4, line 32: The grid spacing of 18 km is rather coarse, especially in resolving terrain-induced circulations in California. There have been numerous studies on this subject for California, and it would be useful to point that out. Although not explicitly stated here, the choice of coarse grid spacing is likely due to the computation cost of the adjoint-based technique.

Page 5, line 5: I could be wrong, but I thought the FINN emissions provided emissions per fire (i.e. point) that did not provide a spatial information on the size of the fire.

Page 5, line 13: I am skeptical of scaling the AOD based on a known high bias in GEOS-5. The bias is not necessarily linearly related to emissions. The problem in AOD in GEOS-5 could be a host of issues, such as representing the right mix of scattering and absorbing aerosols, water uptake of aerosols, and the treatment of aerosol optical properties.

Page 6, lines 5 – 9: Figure 2 shows the location of the MODIS fires, but the discussion is more about the shifts in the fires in the datasets. The text somehow implies Figure 2 illustrates those shifts, which it does not. Please change the text to clarify.

Page 14, line 9: the use of “swept out” should be changed. Implies the winds pushed
the aircraft over the ocean as opposed to a choice by the scientists or aircraft crew.

Page 14, lines 3 – 14: Suggest including the aircraft flight paths in Figure 2 to illustrate the discussion in this paragraph.

Page 14, line 18: Do you mean “Average” or “re-average”? They mention the 10-s data in the previous sentence, so perhaps that is a 10-s average? If that is the case, the text does not say so. The 90-s averaging is acknowledging the mismatch between the observations and the temporal resolution of the model. What I found missing was a discussion on the mismatch in spatial resolution, or does that even matter in the adjoint technique used? The model grid cell is 18 km, so it cannot resolve small-scale variation, so shouldn’t the data be averaged to 18 km intervals?

Page 15, line 18: The authors mention that their adjoint technique requires 600 more computational time than a single simulation. It is good that this is mentioned. What is not included how many man-months or man-years such an effort requires (and that does not need to be included). But I would like to see some discussion at the end of the paper to weigh in on the advantages and disadvantages of the additional computation cost. In other words, is it worth the effort?

Page 15, lines 22 – 26: In this section of the details of the adjoint technique, I found it difficult to understand why it is important to show the convergence properties. I found the answer a few lines down on line 26. This rationale is a bit buried. I found the other sections had similar problems in terms of why it was important to visit aspects of the adjoint technique. This is a reflection that the authors assume the audience has a detailed knowledge of data assimilation and the adjoint technique in particular. So I think the concepts could be better communicated to a larger audience.

Page 16, line 12: In relation to “overprediction seems to be less of a problem” to me is a result of the coarse grid spacing. Using dx =18 km, the model should underpredict the peak concentrations of the emissions are correct. Only when the grid spacing is finer will the model resolve details of the smoke plume and there will be periods in which the
concentrations could be higher than observed.

Page 16, line 35: A 3.8 factor of uncertainty is used for the emissions. Did the authors try using values larger than 3.8?

Page 18, lines 3 – 11: The increases in BC emissions in the Fresno area are remarkable, so that it seems to have higher emissions than the LA area (at least from the scale in the figure). The explanation for missing BC emission seems plausible, but would missing railroad emission produce values as high as the entire LA basin? Does this seem realistic?

Page 21, lines 17-18: Isn’t the first phrase in this sentence an obvious one that does not require this study to point out?

Page 22, lines 9 – 15: I am not sure what to make with the conclusion in the last sentence. It is true that in the cases examined in this study the aircraft flight paths and surface data might be saying different things about sources (since they are likely decoupled from one another). On other days that might not be the case when the aircraft flies in the boundary layer and will be more similar to the surface measurements. The concluding sentence seems to contradict the whole notion of using ARCTAS-CARB for their test of the adjoint technique. However, a large fraction of fires, as well as field campaign data to characterize fires are conducted over forest regions that are often located in areas of complex terrain. It seems to be a problem one has to confront.

Page 22: line 27: The authors state upfront and here that the paper focused on emissions only and not other factors that can affect BC concentrations. In the future, will they extend the analysis to meteorology to reduce those factors as well?

Conclusion: There are a few points I find missing or poorly articulated. The first is related to recommendation on future sampling. Here and there in the paper the authors mention or allude to changes in the sapling strategy that would help in better constraining the emission. It would be useful to include a summary of X, Y, and Z they
feel would be useful. Second, how many more cases would be needed to have more robust estimates of the emissions? Finally, another discussion I mentioned in an early comment, is the computational cost and effort of using an adjoint technique worth the cost? One could take an alternative approach is to simply perform a small number of sensitivity simulation that scale the emissions to get a better fit. This is of contrast to a brute force method, which is not as mathematically pleasing as the adjoint.

Figure 2: The colors used for the vegetation distribution make it difficult to see the red dots denoting the fires. It would be useful to include the aircraft flight paths.

Figure 7: Consider changing the color scale, it is very difficult to see changes from one figure to the next.

Figure 9: The caption denotes posterior BB, but it is not clear which lines in the figure are posterior based on the legend.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-573, 2016.