Reply to anonymous referee #1

We would like to thank the referee for the thoughtful and insightful comments. We have addressed all of the comments. Our responses are itemized below.

General comments:

The most interesting aspect of the paper for me is that more realistic meteorological simulations (boundary layer heights of GEOS-4 and GEOS-5 compared to GEOS-3 and precipitation of GEOS-4 compared to GEOS-5) degrade the quality of model BC simulations. The ramifications of this result for applying model simulations to source attributions of BC or other pollutants are very significant. I hope that the authors can add more discussion on this aspect and state more clearly which model fractional attributions in this paper are reliable.

Discussions added.

For the reason described above, the least appealing section to me is 3.6 (I should say that I didn't find any error in that section). I personally find the large fractions of pollutants attributed to Asian sources across the Pacific uninteresting. One can make as good a guess simply by looking at the emission inventory.

Point well taken. Deleted.

There are three issues that I do think that the authors should think about and revise the paper accordingly. It is clear that the model underestimates BC in fire seasons, which should be stated explicitly in the abstract and conclusions.

Yes. Now stated.

The reason may be a large underestimation of fire emissions in the western U.S., which should be stated more directly in the paper if true.

Now stated.

A more detailed analysis should be done by separating biomass burning sources. There is no reason to lump fire emissions over the U.S. with the rest of the world (see Zeng et al., 2011). While the 50% increase of global biomass burning is large, the effect comes probably only from the emissions over the western US (and some from western Canada).

Analysis added.

I think it would be very reasonable to increase the regional emissions over the western North America by a factor of 2 and see if BC simulations are improved. If not, the model probably has another very serious problem not recognized by the community. I think that this aspect of the paper is very interesting and scientifically significant.

Point well taken. We already included a sensitivity simulation with an increase of a factor of 1.5 of the regional biomass burning emissions.

When comparing to high-altitude sites, the model results are probably sampled at the corresponding altitudes. I think that this is problematic. The IMPROVE sites are "surface" sites. In daytime, a large fraction of fire emissions at lower altitudes can be transported along the slope up to the sites. This is a very different process from boundary layer mixing and then transport. In fact, one issue with the model comparison may well be that boundary layer mixing is instantaneous such that the BC gradient from the surface to the mixed layer cannot be simulated in the model. The IMPROVE sites from 0-4 km all sample this "surface" layer, not the mixed layer. It is less an issue for anthropogenic BC since the sites are located away from the large anthropogenic sources.

Agreed. Discussions added.

I don't think that the horizontal resolution of the model is as important as the over the source vertical gradient. By comparing the 2x2.5 emissions in the model with the original GFED fire emission distributions, it would be useful to comment on how the spatial averaging of emissions may affect the BC simulation results.

Discussions added.