We thank the reviewers for their supportive and thoughtful comments. Our responses to the comments are provided below, with the reviewers' comments italicized.

Review 1:

The paper of Wang et al. investigates the relative contribution of black carbon from anthropogenic pollution versus biomass burning to the arctic snow pack. This is done by combining information from the ARCTAS flights on the relative contributions of anthropogenic versus biomass burning black carbon in the atmosphere with GEOSChem/GEOS-5 simulations, then comparing results to snow samples.

Overall this is a readable paper which makes good use of the ARCTAS flights in amodeling framework. The paper describes what they did and I for one am pretty appreciative that I could mentally follow along. It lays out the problem and cites the previous literature extremely well. The topic they have chosen is considered a critical climate issue, and the paper is appropriate for ACP. There are many (including myself) who are starting to be persuaded that BC on arctic snow may have been a bit overblown. For the record, the cryosphere area is a bit out of my expertise, so my comments will be limited to the broad lines of biomass burning (as they use the FLAMBE source function which I authored 10 years ago), and the generally observability of the atmosphere/snow system that they model.

Speaking as someone who has studied aerosol and fire observability issues from many angles, I can attest that the problem they are attacking is very difficult. The uncertainties in any emission source function for black carbon is probably at best a factor of 2, and in some cases an order of magnitude. The authors have previous experience with the FLMABE product, using an ARCTAS version perturbed by Edward Hyer in our shop which included a full carbon budget. The present authors scaled FLAMBE emissions by $\frac{1}{2}$ *to match previously performed CO comparisons. In general however, it is our experience that FLAMBE underestimates particle emissions, likely by a factor of 2 or 3. So this correction based on CO many not be wholly appropriate. As CO is largely from smoldering combustion, and black carbon from flaming, nonlinearities in the source profile can be problematic-particularly in boreal or mid-latitude fires. That said, in their particular application the model appears to compare very well with their presented verification data. So well, in fact, it makes me a bit suspicious- not in that I think there is any impropriety. But clearly they did some tuning to source functions, which have high uncertainties of their own. I think this could be laid out a bit better in the paper.*

We now explain better in Section 2 that the BC and OA fire emission factors were in the end constrained by the ARCTAS observations: "We apply here the same reductions to BC and OA emissions **and further correct the emissions to fit the ARCTAS data**….

From there we use observations of the BC/OA concentration ratio in fire plumes to constrain the BC emission factor. ”

.It would have been beneficial if they could have run an ensemble of source functions to see what the underlying sensitivity of the system was. These problems are non-linear, and when operating at 2.5 degrees a lot of structure could get washed out (no pun intended). To their credit, in the paper the authors do mention this problem. Without such a sensitivity study, their results may be a bit difficult to interpret. For example, if they increased FLAMBE back to native, what does this do to their numbers? They mention in the paper that they intercomparred with GFED (which is another good way to bound the problem), but the results are not really elucidated. While I am up on biomass burning, I suspect that there are similar issues on the anthropogenic side. Fortunately, the ARCTAS data helps with the in-atmosphere partition. But again these measurements are not in Asia.

As pointed out in Section 2, the simulation is linear and this should address the concern of the reviewer. We now remind the reader of this linearity in Fig.7: “**Also shown in Fig.7 is the model source attribution using tagged tracers as described in Sect. 2. Since the model relationship between sources and concentrations is linear, the source contributions are additive and the sensitivity to source magnitudes can be readily inferred from the data shown here.**

In modeling I take tuning for granted. But here the authors are in a bit of a pickle. in the really interesting part of their finding, that is of BC deposition in Siberia, their reanalysis has no verification data-just a few data points from Sarah. This makes sensitivity work more crucial. They provide fractions of anthropogenic versus biomass burning to 2 significant figures, but the known uncertainties in sources and meteorology do not support this. If you tinker with emissions (like changing them by a factor of two in FLAMBE) and the scavenging efficiencies for BC and other aerosols (adjusting the scavenging parameters by up to an order of magnitude), you can get pretty much whatever result you want. This tuning is not necessarily wrong - just highly uncertain. Some of the ideas, e.g. only hydrophobic BC gets scavenged in cold clouds, are likely wrong. In any case, I think that their conclusion that snow BC is mostly anthropogenic (I notice that they apparently misspoke themselves in their conclusions since they say just the opposite, i.e., anthro only 43% in spring) is very uncertain and quite possibly wrong. This said, I don’t have a better answer (i.e. if we knew what we were doing, this would not be science).

Point well taken. We do think that the Doherty data provide important corroboration of the high BC deposition in the Siberian sector. To better make this point we have reworked the text in Section 4 to shorten it and tie it better to the constraints from the

Doherty data. We have also edited the Conclusions and Abstract accordingly, and now give just one significant figure there for the anthro vs. fire contributions.

The independent verification of BC emissions using observations in the mid-latitudes source regions and in fire plumes helps to constrain the wiggle room in the model. We have clarified this for fire plumes (see response to comment above) and have added a sentence in the abstract about the constraint from mid-latitude observations.

We have added a sentence in Section 2 to provide some support for our treatment of BC scavenging: **"Cozic et al. (2007) find that the BC fraction scavenged into cloud droplets decreases with decreasing temperature, from 60% at 0^{o}C to 10% at <-20^{o}C."** We recognize in the text that scavenging by cold clouds is highly uncertain.

We have corrected the mistake in the conclusion: "**Open fires in the model** account on average for 10% of BC content in the Arctic snow in Jan-Mar and 40% in Apr-May 2007-2009."

In concussion of my major comments, clarifying these issues in the paper is critical, and sensitivity runs would be preferable to understand what the bounds might be.

As pointed out above, sensitivity to emissions is linear and we now state this better in the text. Simulation of wet deposition is uncertain and non-linear but there are so many factors involved that it's not clear that sensitivity simulations would be useful. Instead we have added the following paragraph to the end of Section 2.1 recognizing the uncertainty and pointing out the constraints on the simulation:

Model transport of aerosol from northern mid-latitudes to the Arctic is highly sensitive to the representation of wet deposition (Liu et al., 2011). There are many associated uncertainties including model precipitation and its vertical distribution, the subgrid scale of precipitation coupled to transport, and the scavenging efficiencies from washout and rainout. The tropospheric lifetime of BC against deposition in our simulation is 6 days, not significantly different from the standard GEOS-Chem model (5-6 days), and within the range of 5-11days from current models (Koch et al., 2009b). The tropospheric lifetime of ^{210}Pb aerosol is 10.4 days (Amos et al., 2011), consistent with observational constraints (Liu et al., 2001). With regard to the Arctic, our successful simulation of observations combined with the relatively good constraints on emissions (see discussion below) gives some confidence to our scavenging parameterization. Fisher et al. (2011) previously showed that it allows a successful simulation of sulfate-ammonium aerosol in ARCTAS.

Other minor things: I am not so sure regressions are an appropriate metric in figure 6. To me it is more representative of a two state system (background versus polluted).

We don't understand the reviewer's point here.

I found the green versus blue dotted lines a bit hard to read in Figure 7.

We changed the green to brown.

For figure 10, it would be best if possible, to confine the data to 2007-2009 to match the subsequent plots

We now confine the data to 2007-2009.

Review 2:

General Comments

This work examines the sources and sinks of aerosols in the springtime Arctic during the

POLARCAT 2008 campaigns and augments the growing body of literature on this subject matter. Specifically, Wang et al. use the GEOS-Chem model, a global chemical transport model with a relatively long track record of development in the peer-reviewed literature, to model the emissions, transport, and removal of black carbon (BC) and organic aerosol (OA) in the Arctic. This study sets lofty goals to "understand the factors controlling the concentrations of carbonaceous aerosols in the Arctic, the deposition of BC to snow, and the implications for snow albedo and associated radiative forcing." While the authors address each of these topics in the paper, it is not immediately clear how significant the results actually are because they are based on a simulation. For example, the rationale behind the partitioning between wet and dry removal processes in the model (and comparison with other model partitioning and parameterizations) could be better explained in the paper and would increase the significance of the results presented. Furthermore, there is sometimes confusion in the paper whether the authors are referring to wet or dry deposition or both.

We tried to clarify when needed everytime we use the word "deposition" and added some statistics in Section 4 on the relative importance of wet and dry deposition: **"Wet processes in our model account for 91% of total BC deposition to the Arctic in spring"; "The relative contribution of dry deposition to total deposition is 15% in winter and 9% in spring, with little interannual variability. It is smallest over the Eurasian Arctic in spring where the deposition flux is highest."**

There are numerous studies of this general flavor in the literature but, to the authors'credit, this study is anchored by aircraft and ground-based observations. This paper is generally well written and comprehensively references the rapidly evolving research on Arctic aerosols. However, it would be nice to see some reference made to some of the early Arctic aerosol surveys in the 1980s (Schnell, 1984; Hansen and Rosen, 1984; Hansen and Novakov, 1989).

Done.

Since this paper has been submitted to the POLARCAT special session, I also suggest referring to and defining POLARCAT early in the paper.

Done.

The newer aspects of this work appear to be in the effort to model BC in snow and to simulate OA in the Arctic troposphere. I would recommend explicitly stating what is new in the paper in the introduction.

We did some editing of the Introduction to that end.

There is a good discussion in the paper on nucleation (in-cloud) and precipitation (below-cloud) scavenging parameterizations in the GEOS-Chem model. This could be further improved by relating the GEOS-Chem parameterizations with those from other global aerosol models.

We have expanded the discussion of uncertainties and constraints regarding aerosol wet deposition. See response to Reviewer 1. We already compared our lifetimes to the range from other global aerosol models (reference to Koch et al. 2009b)

In summary, this paper adds value to the literature on this topic and is worthy of publication in ACP subject to addressing these and the specific/technical comments below.

Specific comments:

p. 19397, lines 4-5, " . . . together with longer-term records in surface air and in snow.": This description could be better connected to the main subject of this sentence—using GEOS-Chem to interpret the aerosol measurements?

We replaced 'together with' by 'as well as'

p. 19399, lines 4-8: in addition to ARCTAS and ARCPAC, there were also single-particle measurements of BC during PAM-ARCMIP (Stone et al., JGR, 2010); this would be an appropriate place to cite this additional IPY study.

Done.

p. 19399, lines 20-22, "There has been far less attention to modeling OA over the Arctic,. . . ": This idea needs to be expanded since this is one of the newer topics in this paper.

We have added a new paragraph to expand on this: "**Attempts to model OA over the Arctic have been more limited. Open fires would be expected to be a dominant source on an annual average basis (Koch et al., 2007). There is a general tendency for models to underestimate observed OA concentrations in the remote atmosphere (Heald et al., 2005, 2011), and this has been attributed to poor representation of secondary organic aerosol (SOA) formation (Volkamer et al., 2006; Hodzic et al., 2010; Heald et al., 2011). "**

p. 19399, line 23 – p. 19400, line 4: I recommend revising this paragraph to state explicitly and succinctly what is new in this paper.

We have rewritten this paragraph accordingly.

p. 19400, line 24, " . . . standard resistance-in-series scheme . . . ": since dry deposition is a significant fraction of aerosol removal in the Arctic in winter and spring, this is a model process that is relevant to the subject of this paper; please describe the process in more descriptive terms; given the citation provided, a brief description should be adequate.

We have added brief description about dry deposition: "Dry deposition in GEOS-Chem follows a standard resistance-in-series scheme (Wesely, 1989) as implemented by Wang et al. (1998)**, with deposition velocities calculated locally using GEOS surface data for momentum and sensible heat fluxes, temperature, and solar radiation**."

p. 19400, line 25 – p. 19401, line 2, dry deposition in the Arctic is prescribed at 30% of the global amount in GEOS-Chem but what is the fractional contribution of dry deposition to total deposition in the Arctic in the model?; this should be mentioned in the context of the next section on wet deposition.

We now state in Section 4 that wet deposition in our model accounts for 91% of total deposition to the Arctic in spring.

p. 19401, lines 8-9, "Here we introduce such a distinction as well as other improvements to the scavenging scheme.": From what I can tell, this is a major change to the model and motivated the results presented in this work. As such, a brief summary of this improvement should be promoted to the introduction section.

We now address this improvement in the last paragraph in Introduction: "We show… **that the GEOS-Chem model with improved representation of wet deposition can successfully simulate these observations**."

p. 19401, line 16 and 19, "very fine" versus "nucleation" mode: if these are the same mode, use the same nomenclature; otherwise, please indicate the differences in the size ranges of the particles

We removed "very fine" from line 16 since that size range is negligible for mass scavenging in any case.

p. 19401, line 27, "Scavenging of accumulation-mode aerosols by snow is an order of magnitude more efficient . . . ": this does not directly follow from the values for the washout coefficients presented in lines 22-25; please clarify.

We have rewritten this sentence to: "Scavenging of accumulation-mode aerosols by snow **is 5-25 times more efficient than by rain for *P* in the range 0.01-1 mm h^{-1} because of the larger cross sectional area of snow crystals vs. rain drops (Murakami et al., 1983). The difference increases as P increases.**"

p. 19402, lines 2-13, mixed-phase clouds can exist over a wide range of temperatures, often below -15C (258 K); Stith et al., ACP, 2011 present a case in the northern midlatitude Pacific (a region relevant to this analysis) where BC concentrations were reduced in the region of mixed-phase clouds, possibly due to a scavenging process; this paragraph should be revised to take the wide range of mixed-phase cloud temperatures into account and more attention should be given to these important cloud types in the context of removal of BC and OA in the Arctic

There are large uncertainties in the BC scavenging mechanism that we now acknowledge better in the text (see response to Reviewer 1). In our opinion, a detailed treatment of scavenging from mixed-phase clouds would be overkill. We now justify better our T threshold on the basis of the data from Cozic et al. (2007) and have added the Stith et al. reference.

p. 19403, lines 7-9, "The global lifetime of carbonaceous aerosols in our simulation is 6 days . . . ": what was it previously, before changing the BC/OA removal schemes?

We have added, "The **tropospheric** lifetime of **BC against deposition** in our simulation is 6 days, **not significantly different from the standard GEOS-Chem model (5-6 days), and** within the range of 5-11days from current models (Koch et al., 2009b)".

p. 19403, lines 14-15, " . . . reducing deposition . . . ": wet deposition?

Now specify total deposition.

p. 19403, lines 16-20, "However, TRACE-P . . . coating for the back carbon particles (McNaughton et al., 2011)": This sentence is awkward; please rewrite for clarity.

Rewritten to: **However, the significant coating of BC particles indicated by aircraft observations of shell/core ratios (Kondo et al., 2011) and light absorption (McNaughton et al., 2011) in ARCTAS suggests that BC in the Arctic is mainly hydrophilic. In addition, TRACE-P aircraft observations in Asian outflow in March-April provide good constraints that the BC aging time scale is no more than 2 days (Park et al., 2005)**.

p. 19405, line 23 – p. 19406, line 1: what are the measured size ranges for the BC particles from the SP2 and OA particles from the AMS?

Ranges are now given.

p. 19406, line 3, " . . . ratio of 2.1 . . . ": please clarify.

We changed 'ratio' to 'scaling factor'.

p. 19406, line 10, " . . . smaller spatial domain around Fairbanks.": edit to correct, " . . . smaller spatial domain in the Alaskan Arctic."; most of the sampling during ARCPAC took place between the north coast of AK and 75N.

Done.

p. 19408, line 11, " . . . adding to the subsidence from the mid troposphere . . . ": The Arctic troposphere is very stratified in winter and early spring so I am not sure the concept of subsidence is correct here; rather, there may be episodic vertical mixing processes associated with frontal passages (rare in winter; more common in spring) or convective influences from open leads in the sea ice.

We deleted mention of subsidence.

p. 19410, line 24, "Deposition is mainly by wet processes (90%)." During what time of year? In the Arctic? This is a very important point in the paper and needs to be developed sooner (Section 2) and substantiated. How does the GEOS-Chem model results compare to other global aerosol model treatments in the Arctic that, e.g., have a 50-50 wet-dry partitioning?

We now elaborate further on this point: "**Wet processes in our model account for 91% of total BC deposition to the Arctic in spring. This is higher than in the previous model studies of Huang et al. (2010), who assumed a larger dry deposition velocity to snow and ice, and Liu et al. (2011), who assumed persistence of hydrophobic BC. We pointed out in Section 2 that our dry deposition velocities to snow and ice are constrained by observations, and that most of the BC in ARCTAS was likely**

hydrophilic. Spackman et al. (2010) inferred a dry deposition flux for BC of 100-5300 ng m^{-2} day^{-1} over snow/ice during ARCPAC on the basis of observed BC depletion in the boundary layer. Our computed dry deposition flux in the Western Arctic (mostly covered by snow/ice) is about 1500 ng m^{-2} day^{-1} in spring, consistent with that estimate. **Although the range is large, it implies that BC deposition in the Arctic must be dominated by wet processes**."

p. 19410, line 25: "Spackman et al. (2010) inferred a dry deposition flux for BC of 170-1700 ng m-2 day-1 over snow/ice during ARCPAC . . .": Spackman et al. reported 100-5300 ng m-2 day-1 in ACP. Please correct the text. If GEOS-Chem dry deposition is only 10% and comparable to the observed dry deposition in Spackman et al., how can the GEOS-Chem model results compare favorably to measurements of BC in snow from Hegg et al. and Doherty et al. (discussion at p. 19412, lines 3-14)? Spackman et al. showed their results compared favorably with these same measurements of BC in snow. This topic needs further discussion in the paper in order to put the GEOS-Chem results in context.

We now correct the dry deposition flux for BC inferred by Spackman et al. (2010): "Spackman et al. (2010) inferred a dry deposition flux for BC of **100-5300** ng m^{-2} day^{-1} over snow/ice …"

The estimation of BC content in snow in Spackman et al. depends on their snowfall of 5.3 cm, which corresponds to 5.3 kg m^{-2}. Although the total deposition is high in our work (e.g. 1500/10%=15000 ng m^{-2} day^{-1}), the snowfall in the Western Arctic in April 2008 in the model (from GMAO meteorological data) is also large, about 25 kg m^{-2} (even larger in Alaska, 20-50 kg m^{-2}). Consequently, the model generates similar BC content in snow as those in Spackman et al. and in Hegg et al.

p. 19414 – p. 19416: The conclusion section is largely a reiteration of what was already stated earlier in the paper. Please condense this section to summarize the major findings briefly.

We condensed some.

p. 19414, line 22, " . . . gave excessive consideration to fire plumes.": This was adequately explained for Matsui et al. (2011) but was not apparent for Warneke et al. (2010) earlier in the paper. For Warneke et al., by excessive consideration, are you referring to the analysis or the potential ARCPAC sampling biases? Please explain.

We deleted reference to Warneke et al. here (it's referenced elsewhere). The problem with that analysis has to do with ignoring BC deposition and dependence on the assumed background. This is not simply explained and doesn't seem worth the space.

p. 19414, line 29 – p. 19415, line 2, " . . . in surface air which is most relevant to BC deposition in snow . . . ": I do not think this can be so easily asserted here, especially in the context of the source attribution discrepancies between modeling studies and measurements of BC in snow. The role of open leads in the sea-ice on the stirring of the tropospheric column could be an important part of the mechanism that folds free tropospheric aerosol into the Arctic boundary layer.

Deleted.

p. 19415, line 16, " . . . dampens the seasonality at Barrow.": I think I see how the "stronger impact of fire emissions in spring" based on the discussion in Section 3.2 but this could use a little more clarification.

We deleted that phrase

p. 19415, lines 20-22, "total BC deposition": given the 90%-10% fractional contributions from wet and dry deposition in the model, how do you explain this factor of 2 difference (17 vs 30 Gg/mo) between winter and spring. Is this related to greater loadings in spring? Where? In the free troposphere or the Arctic boundary layer. Or is this due to increased wet removal in spring? In-cloud or below-cloud scavenging? These details are all important to understanding the treatment of BC in the model.

We added a sentence, "**Higher deposition fluxes in spring are due to fires.**"

p. 19415, line 24 – p. 19416, line 4, From what I can tell, this study does not resolve the dichotomy between the Hegg et al. results suggesting biomass burning influence in the snow and the model source attribution studies that keep the biomass burning aerosol aloft in the free troposphere. This discussion could be expanded to add any new perspectives provided by the GEOS-Chem modeling effort. It seems the GEOS-Chem model may get the total BC deposition correct (although I am not sure how) but the source attribution wrong. I think this seemingly contradictory result deserves more explanation.

We believe that we have a credible explanation to resolve the dichotomy and we have rewritten that text to make it clearer: "**Some of that difference can be reconciled by the biofuel source of BC, which they would diagnose as biomass burning but must be viewed as anthropogenic. In addition, the well-known mixing of anthropogenic and fire influences in Asian outflow could result in anthropogenic influences being correlated with biomass burning tracers.** "

Technical Comments

p. 19405, lines 6-7, " . . . network observations for 2008, and use therefore data for other years . . . ": Rephrase to " . . . network observations for 2008 and, therefore, use data for other years . . . "

Done.

p. 19405, line 23, "BC was measured with a SP2 . . . ": edit to replace "a" with "an", "BC was measured with an SP2 . . ."

Done.

p. 19406, line 15, " . . . and BC deposition . . . ": edit to clarify, " . . . and BC dry deposition . . . "

Done.

p. 19413, lines 19-20, " . . . BC concentrations in surface air and 2007-2009 pan-Arctic observations of BC snow content.": rephrase to, " . . . BC concentrations in surface air and in snow."

Done.

p. 19414, line 3, " . . . to improve the match to observations for BC, . . . ": edit to clarify, " . . . to improve agreement with BC observations, . . . "

Done.

p. 19414, line 16, " . . . that fires contribute . . . ": edit to add, " . . . that distant fires contribute . . . "

Not clear what "distant" would add.

p. 19414, line 17, strike "anthropogenic" from " . . . are mostly from of Asian anthropogenic origin, . . . "

Done.

p. 19415, line 8, "seasonal context": perhaps you mean "climatological context"?

replaced by '**broader** seasonal context'.

p. 19422, lines 18-21, Leibensperger et al. has been published in ACPD; update the reference accordingly.

Done.

p. 19423, line 13, McNaughton et al. is now published in ACP and should be updated accordingly.

Done.

p. 19427, line 5, change "2008" to "2008a".

Done.

Fig. 2 Caption, last sentence, edit to add, " . . . corresponding equation is given [in the] inset."

Done.

Fig. 8 Caption, " . . . model values sampled along the aircraft flight tracks . . . ": are the model values interpolated along the flight tracks?

They are not interpolated. We have modified the Caption to clear the confusion: "…Observations from the DC-8 aircraft (left panels) are compared to model values (right panels) sampled along the aircraft flight tracks **as described in the text**."

Fig. 10: The thin black lines are hard to see in the figure panels.

Fixed

Fig. 12: The purple circles are difficult to discern in the figure.

Fixed