

29 August 2011

Review of Wang et al., ACPD, "*Sources of carbonaceous aerosols and deposited black carbon in the Arctic in winter-spring: implications for radiative forcing*"

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.

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

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.

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.

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

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.

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.

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.

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

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.

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

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?

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

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?

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

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.

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.

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?

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.

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.

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.

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.

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.

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.

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.

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 . . . “

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

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

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.”

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

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

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

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

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

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

p. 19427, line 5, change “2008” to “2008a”.

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

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

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

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