Response to Dr Flanner:

We would like to thank Dr Flanner for his comments; our responses to the major issues raised are outlined below. To maintain clarity the original review comments are in italic, our responses in plain text and suggested text changes or additions to the paper in bold.

1) One general concern relates to the simplicity of some of the model treatments and how this simplicity may impact the sensitivity studies and conclusions. One example of this is that there does not seem to be any consideration of mixed-phase clouds, which are known to exist over a range of temperatures and which may be prevalent in the Arctic, at least in certain seasons and locations (e.g., Curry et al, 2000, BAMS). Riming may occur frequently in these clouds and there is strong evidence that riming is an effective scavenging process (e.g., Hegg et al, 2011 Tellus). How might treatment of mixed-phase clouds influence the sensitivity studies?

The simplicity of the deposition treatment in GLOMAP is necessitated by the inability of a chemical transport model to simulate cloud phase. Mixed-phase clouds can occur over a range of temperatures and are particularly prevalent in the Arctic lower troposphere during spring (Hobbs & Rangno 1998, 2001) and are likely to have an efficiency between that of ice-nucleation scavenging and liquid-phase scavenging (Hegg et al 2011). Therefore, our treatment of mixed-phase clouds as liquid is likely to overestimate the scavenging rate in spring.

In the modelling study Bourgeious and Bey (2010), which used a model capable of simulating cloud phase, the scavenging efficiency of stratiform mixed-phase was decreased from 85 and 99% (soluble accumulation and coarse mode particles) to 6 and 75%. Ice-phase cloud scavenging efficiency was decreased to 6% over the entire particle size range. This reduction in scavenging efficiency resulted in greater simulated concentrations of BC and SO_4 aerosol in the Arctic spring, although the model remained biased low against observations. Therefore, model information about mixed-phase clouds does not really enable a better or more physically based treatment of scavenging until the processes are better quantified. Our simple switch from liquid to ice-phase at -15°C is consistent with what is seen in observations (Garrett et al 2010).

We agree with the reviewer that this subject requires greater discussion in the paper and will include the following in section 5.1:

Page 3422, line 12: **However, observations have shown that in spring mixed-phase cloud is also present in the Arctic lower troposphere (Hobbs & Rangno 1998). Mixed-phase cloud scavenging can occur via nucleation of ice on individual particles (as discussed above) and riming of ice particles with water droplets and is likely to have an efficiency between that of ice-nucleation scavenging and liquid-phase scavenging (Hegg et al. 2011). Therefore, our treatment of mixed-phase clouds as liquid-phase is likely to overestimate the scavenging rate in spring.**

Mixed-phase cloud formation can occur over a wide temperature range. The scavenging efficiency of individual clouds is dependent on the ratio of ice-to-water, which is affected by physical processes such as the Bergeron-Findesien process and can be highly variable over limited temperature ranges (Smith et al. 2009). In models that include mixed-phase clouds (Bourgeious and Bey, 2010) assumptions still need to be made about scavenging rates that are not treated explicitly, and agreement with observations is not necessarily improved. Our switch from liquid to ice-phase at -15°C is consistent with what is seen in observations (Garrett et al., 2010), and is a reasonable assumption until the physical processes are resolved better. The implications of the exclusion of mixed-phase scavenging are discussed below (section 7.3.2)

The exclusion of mixed-phase scavenging is also likely to affect the results of our ice-phase temperature sensitivity studies. As discussed in section 7.3.2, in winter regional Arctic scavenging rates are predicted to be insensitive to the ice-phase temperature threshold because of the sharp meridional temperature gradient. However, in spring (when mixed-phase clouds have been observed (Hobbs and Rangno 1998)) the response to increases in the ice-phase temperature threshold is non-uniform at Arctic ground-sites. This effect could be sensitive to assumptions about mixed-phase scavenging. We suggest adding the following to section 7.3.2 to discuss this possibility.

Page 3435 line3: *at all other sites bias has changed negligibly….* **The non-uniform springtime response at different sites to increases in the ice-phase temperature threshold might change if mixed-phase scavenging processes were included. The scavenging efficiency of mixed-phase clouds is likely to be spatially variable due to variations in liquid-to-ice cloud ratios between regions (Smith et al., 2009). Thus, better treatment of mixed-phase cloud processes has the potential to improve the simulated spatial and temporal patterns of aerosol concentrations.**

It is possible that by suppressing (or overestimating) scavenging rates in mixed-phase clouds during the spring we delay (or accelerate) the simulated transition from the Arctic late-spring haze period to the `clean` summer boundary layer. However, this effect is not evident in the majority of our ground-station comparisons (Fig 7).

2) The ICE sensitivity studies assume that insoluble aerosols nucleate at temperatures below -15ºC, while soluble aerosols are not scavenged at all. It does not seem reasonable that only the insoluble aerosols should nucleate. Please explain the reasoning for this in more detail. Would it be more realistic to allow some ice nucleation of the soluble aerosols? It appears that this approach was intended to capture the lack of scavenging through collision and coalescence in ice clouds. Please elaborate on any limitations of this approach.

Insoluble aerosol particles in the model are assumed to act as IN in ice-phase clouds in the deposition mode. There is evidence to suggest that BC or soot is preferentially scavenged in ice-nucleation. This effect is due to numerous factors including the differing surface structure of soot which is more likely to be aggregated and so initiate ice nucleation (Gorbunov et al. 2000 J. Aerosol Sci.). However, the rate of scavenging is equivalent to that if collision and coalescence processes were still happening. Our treatment of IN scavevenging is likely to overestimate the scavenging rate of insoluble aerosol in ice-phase clouds (although this effect will occur equally in control simulations).

We have examined the impact of IN scavenging . The deposition of insoluble BC is relatively small (0.1% of total global deposition compared to 77.9% dry deposited and 22.0% impaction scavenged). This small contribution is because we assume a size threshold for scavenging, and the majority of insoluble aerosol particles are small (Aitken mode) and so not activated as IN. Therefore, concentrations of insoluble aerosol in the model are not significantly impacted by ice-phase scavenging.

We agree this reasoning is unclear in the text and suggest substituting the following text in section 5.1:

Page 3421, at line 17: **For insoluble aerosol, a cloud temperature threshold is used to determine scavenging rates. In cloudy regions below -15°C insoluble particles can act as ice nuclei (IN) (Fan, 2004) (see Figure 4). The prescribed scavenging rate of IN is equivalent to that of cloud droplets in a warm cloud, so it is possible that this process overestimates the deposition of insoluble aerosol from ice nucleation. However, the majority of insoluble particles are small and exist in the Aitken mode (freshly emitted BC and OC) and are therefore only weakly**

scavenged (0.1% of the total global insoluble BC mass deposited) compared to impaction scavenging (22%) and dry deposition (77.9%). Thus, model assumptions about IN scavenging of BC particles have only a small effect on the model results.

3) Another issue is that some of the results may be sensitive to the BC ageing processes that determine the relative portions of soluble and insoluble BC, and/or to the assumed fraction of emitted BC that is soluble. For example (p3430,6): "all three drizzle runs have similar median BC concentrations due to the predominance of insoluble BC particles (from BB sources)...". There is observational evidence that a large portion of BC emitted from biomass (BB) sources is soluble, or the BC becomes soluble within minutes of emission. Although differences between SO4 and BC results are sometimes discussed in the context of differences in hygroscopicity, the general importance of BC ageing for these results could be stated or discussed more generally.

In GLOMAP BC is emitted within the insoluble modes and aged into the soluble. BC particles are aged by the condensation of SO_2 and become hydrophilic when they have a sufficient coating of SO4, assumed in this study to be 10 monolayers (Mann et al., 2010). Thus, the ageing rate of BC is sensitive to SO2 emission and the size of the H_2SO_4 condensation sink. In late summer (particularly August) and early autumn the fraction of insoluble BC is particularly high in some regions (70-90% of the total BC load). However, it is unclear whether the overestimation of BC mass in the late summer (Fig 7) results from of an overestimation of the BC ageing time, underestimation of scavenging rates, inaccurate (or coarsely resolved) emissions, or a combination of these factors.

The effect of accelerating the ageing process (increasing the soluble fraction of BC) can be estimated from the sensitivity of SO_4 to drizzle rates as scavenging of soluble BC and SO_4 are equivalent. We agree with the reviewer that this subject requires greater discussion in the paper and suggest adding the following in section 7.4:

At line 20 page 3436 add text: **The dominance of insoluble Aitken mode BC in the late summer and early autumn over the Canadian and Siberian Arctic (70-90% of the total BC mass) is the result of the ageing scheme used in GLOMAP. Aerosol in GLOMAP is aged by condensation of SO_2. BC (and OC) particles become hydrophilic when they have a sufficient coating of SO_2, assumed here to be 10 monolayers of H_2SO_4. This is significantly higher than the previous threshold of 1 monolayer, and was altered to improve the model agreement with remote observations of BC (Mann et al., 2010). A similar decision was made by Liu et al. (2011) in their global model. However, in Liu et al. (2011) the required thickness of H_2SO_4 was explicitly calculated from the Köhler equations.**

The effect of assuming that BC rapidly becomes soluble can be estimated from the dependence of SO_4 on drizzle rates. In August, high latitude SO_4 concentrations fall by a factor of 600 between runs CTRL-MODIS and DRIZZ and by a factor of 200 between runs CTRL-MODIS and DRIZZx0.5, while BC falls by a factor of 1.6 and 1.3 respectively (Fig 8). Thus, assuming highly soluble BC would reduce BC by a factor of ~200, simulated August BC concentrations would be in better agreement with observations at Alert and Barrow (Fig 7). However, it is not known whether the overprediction of August high latitude BC is the result of inaccurate emissions or incorrect scavenging.

We also suggest the addition of the following to emphasise that a predominantly hydrophobic BC load is confined to August; page 3430 replace paragraph beginning: *Arctic surface BC concentrations.....*

Arctic surface BC concentrations are less sensitive to the drizzle rate than SO_4, with median summertime concentrations above 70°N in DRIZZx0.1 increasing by a factor of 3 compared

to DRIZZ (Fig 8b). The effect on the bias at Alert and Barrow is also negligible. In August all three drizzle runs have similar median BC concentration to the control. The negligible effect of drizzle scavenging is confined to August and stems from a high fraction of hydrophobic BC in the Arctic BC load. Therefore, we suggest that BC concentrations during this month are sourced from a biomass burning plume transported rapidly to the Arctic and unaffected by ageing processes (see section 7.4 for further discussion).

4) One methodological issue needs to be clarified: p3424,26 states: "drizzle rate ... was increased" How was it increased? Was the parametrization listed in Equation 3 altered just in a small region? Was the drizzle rate simply held constant year round? Please describe this modification in more detail.

For the European Arctic (as defined in table 3 as between 15E-150W longitude and 60-80N latitude) the drizzle rate calculated from observations via equation 3 was too low compared to precipitation measurements from the northern European EMEP measurement sites. Thus, when condensate lifetime was calculated (in equation 2) the drizzle rate in this region only was changed from 0.08kg/m^2/d to 0.42kg/m^2/d from March-May and Sep-Nov and from 0.44kg/m^2/d to 0.8kg/m^2/d from June-August (as shown in table 2).

The text was amended to read:

from page 3424, line 21: **Arctic drizzle rates calculated using Equation 3 (scaled with mean low cloud fraction of 80-90%) compare reasonably well to averaged precipitation measurements at Barter Island (70N, 143W) and Barrow of 0.6kg/m^2/d in summer and 0.08kg/m^2/d in spring (Curtis et al., 1998) assuming that the majority of measured precipitation is drizzle. However at northern European sites measured precipitation rates are higher than those calculated (0.3-1.0kg/m^2/d in spring and 1.7-2.0kg/m^2/d in summer). It is unlikely that all of the precipitation measured is drizzle. Thus, the spring and summer drizzle rate in the European Arctic (defined in Table 2) was changed to 0.42kg/m^2/d and 0.8kg/m^2/d respectively. The effect of this increase on CCN grid-box lifetime is shown in Figure 5.**

5) Finally, a previous study ("Importance of deposition processes in simulating the seasonality of the Arctic black carbon aerosol", JGR, doi:10.1029/2009JD013478) also evaluated, using some of the same observations, how changes in model deposition processes (in a different model) influence simulated Arctic aerosol concentrations, but this study was not cited. Please reference this study and explain key differences and similarities between it and the current study.

The study Huang et al. (2010) uses a chemical transport model (CAM) coupled with a meteorological module (GEM) to examine the effect of perturbations to the standard deposition processes in the CAM model on the seasonality of Arctic black carbon. These perturbations included: reducing the dry deposition velocity of all particles by 50%, replacing the prescribed scavenging efficiencies for stratiform and convective clouds with one explicitly calculated from the precipitation rate and assumed liquid water content, and replacing the below-cloud scavenging efficiency calculated from Stokes Law with the explicit analytical expression calculated from Feng (2007) for both water droplet and snow/graupel impaction scavenging.

Using the methodology of Feng (2007) to calculate the impaction scavenging flux was found to affect below-cloud scavenging efficiency only in strong precipitation events. Changes to dry deposition processes were found to increase Arctic surface BC but not affect the modelled seasonal cycle. Overall the seasonal cycle in BC at Arctic ground-stations was most sensitive to changes in below/in-cloud scavenging and perturbation of both processes was required for the model-to-observation ratio to fall within a factor of 2.

In this study: we focus entirely on the effect of nucleation scavenging (and the suppression of ice-phase scavenging) on both BC and sulphate. We have identified potential factors controlling the seasonal shift in Arctic aerosol concentration (cloud-phase) and scavenging patterns (low-cloud formation) and suggest that the late spring transition from Arctic haze to `clean` summer is the result of a combination of these factors. Huang et al. (2010) note that the seasonal cycle in BC is driven by the seasonal cycle in Arctic precipitation (with the new wet deposition processes increasing scavenging rates in mid-summer and decreasing scavenging rates in winter and early spring). This result is consistent with our findings and this reference has been added into our conclusions.

Minor issues:
6) Abstract, 14: "in a model" - please mention which model.

This has been be changed to ‘in GLOMAP’

7) 3411, 14-17: I don’t see much distinction between the "up to 2C" warming since the 1980s and the 1C warming between 1976 and 2007. Is the only difference between these numbers the time period over which they were averaged?

Yes, the 1°C warming refers to the temperature increase averaged from 1976 to present day and the 2°C warming refers to when the temperature anomaly is averaged from the late 80s excluding the period of cooling that occurred in the late 70s and early 80s. We agree with the reviewer that this is unclear and suggest modifying the text so that

Page 3411 from line 10 change paragraph to: **The rapid increase in Arctic surface temperatures since the mid-1970s is estimated to be between 1.2 and 1.8°C (Shindell and Faluvegi, 2009). The inverse modelling study of Shindell and Faluvegi (2009) suggests that less than one-third of this warming is attributable to changes in either greenhouse gas concentrations or the Arctic ozone budget. However, changes in anthropogenic aerosol, particularly the decrease in global sulphate emissions since the late 1980s, may account for up to 1°C of the observed warming trend between 1976 and 2008**.

8) 3412, 25: "decline in BC beginning in the 1970s" - McConnell’s ice core record actually shows a decline in BC beginning in the 1910s or 1920s!

The phrase beginning in the 1970s was used as the previous comparison referred to ground-station observations beginning in the 1970s. However, the Greenland ice-cores do cover a far longer time period. We suggest replacing the text from page 3412 line 24 with:
This decline in both BC and SO_4 is also evident in Greenland ice-cores (McConnell et al 2007).

9) 3412, 26: I suggest including a reference for "global decrease in aerosol concentrations", or changing "global" to "regional".

We have referenced the 2001 IPCC report here

10) 3413, 13-15: Please qualify this statement a bit more carefully. Specifically mean that global scavenging rates are insensitive to global temperature changes less than 5C? It doesn’t seem like you are making that assertion in your study.

A discussed in section 7.3.2 we suggest that the reduction (or suppression) of scavenging in the Arctic winter is relatively insensitive to temperature increases (up to 5°C) due to the sharp meridional temperature gradient over the pole. This has been amended in the text to:

Line 13, Page 3413, with: **Here, we suggest that wintertime Arctic scavenging rates are relatively insensitive to temperature increases of less than 5°C (see section 7.3.2).**

We will also include discussion of the sensitivity of both stratocumulus and ice phase scavenging in our conclusions
Line 5, Page 3442 replace paragraph with: **The possible ramifications of an Arctic drizzle 'filter' and cloud scavenging temperature threshold are worth further investigation in the context of climate change. In winter, when weak ice-phase scavenging controls aerosol transport, Arctic concentrations of SO4 and BC are relatively insensitive to temperature increases of less than 5C due to the sharp meridional temperature gradient. However, the weaker zonal temperature gradient in spring means that higher springtime temperatures are likely to increase scavenging, potentially reducing the lifetime of the Arctic haze. In summer, increased temperatures, reduced sea ice and higher humidity will cause more drizzle and reduced aerosol concentrations and deposition rates in the high Arctic. Thus, aerosol concentrations and deposition rates in the high Arctic are likely to fall as temperatures rise.**

11) 3415, 11: Please define Dp where it is first used

Dp has been changed to particle diameter in the text

12) 3416, 5-20: This passage refers to measurements that assume MAE values that vary by 3-fold. This would seem to translate into large uncertainty in measured BC values. Is most of this spread caused by different assumptions about the magnitude of non-BC light absorption at each site, in order to translate EBC to BC? If not, can you identify any other causes for this spread?

We agree with the reviewer that the uncertainty related to using EBC measurements requires more explicit discussion in the paper and we suggest including the two paragraphs below.

Line 21, page 3416: **At the Alert and Barrow sites different instrumentation is used to measure equivalent black carbon concentrations so the two datasets are not directly comparable. The major uncertainty in these measurements is the inclusion of secondary absorbing species in the particles, which overestimates refractory black carbon mass. Due to the seasonality of aerosol mass concentration at Arctic sites the use of uniform MAE (such as at Barrow) is likely to overestimate summer BC mass while underestimating winter BC mass. The inter-annual variability of Arctic aerosol is also high so it is likely that from year to year the agreement of EBC mass with actual BC varies considerably. In this study we focus on reproducing the seasonal cycle (and in particular the late-spring transition) of BC and for this purpose these observations are applicable. However, quantitative comparison of these observations with the model (below) will be affected by this uncertainty.**

13) 3419, 15: ISCCP was already defined.

Changed to ISCCP

14) 3423, 22: are -> our

Changed to our

15) 3423, 23: "upper limit for the given condensate lifetime." - What are the implications of this change being an upper limit? If it is important, you may want to return to this point in the discussion of the DRIZZ results and comparison with observations.

Evaporative processes during precipitation formation are neglected during stratocumulus, frontal and convective rainfall in GLOMAP. For stratocumulus precipitation, assuming that vertical transport within the Arctic summertime boundary layer is sufficiently rapid, evaporation processes were approximated in the drizzle sensitivity runs (Fig 9). Leon et al. (2008) suggest that 50% of drizzle droplets can evaporate before deposition. This reduction is equivalent to the drizzle rate in DRIZZx0.5 the results of which are discussed in section 7.2.2.

16) 3424, 22: What are the measured drizzle rates at Barter Island?

Due to their close proximity (with respect to model resolution) and similar cloud fraction the precipitation measurement at Barter island and Barrow was averaged and the values shown are that mean. This analysis is not clear in the text and has been changed.

17) 3425, 7: "higher low cloud fraction introduced north of 60N results in Arctic CCN life-times... equivalent to" - How does cloud fraction influence CCN lifetime? Equations 2 and 3 show that CCN lifetime depends on LWP, z, and Nd, but not cloud fraction.

Here, we mean the grid-box CCN lifetime which scales with the low-cloud fraction. The paper has now been amended to discuss in-cloud and grid-box aerosol lifetime.

18) 3429, 6: "... although the effects of drizzle scavenging are confined to the late spring and summer." - Why are the BC effects different from SO4 effects in this regard? Is it because much of the spring/summer BC is hydrophilic?

The majority of Arctic BC during the winter and spring is hydrophilic while, in the late summer BC is more likely (in some regions) to be hydrophobic. Therefore, in spring, we would expect the response of SO_4 and BC to drizzle scavenging to be the same. The delayed response of BC (and SO_4 at Alert) to drizzle (May rather than March) is the result of extremely low BC (and SO_4) concentrations (from Jan-April) in both the CTRL and DRIZZ simulations (Fig 7). The text here is unclear and has been amended.

19) 3431, 17: "This analysis suggests that Arctic drizzling low cloud acts as a filter, strongly suppressing the transport of aerosol between the marginal and high Arctic". - This only holds for aerosol that is transported into the lower altitudes of the Arctic. The sensitivity of results to the drizzle parameterization will depend on the amount of aerosol residing in the boundary layer or lower troposphere. As the authors discuss later, some of the transport mechanisms bring BC into the Arctic much above the drizzle zone. Some observational studies also show BC residing very high (up to 7 km) in the Arctic atmosphere (e.g., Brock et al, 2011, ACP). Please qualify this discussion accordingly.

Discussion has been modified at line 17 page 3431 paragraph replaced with: **This analysis suggests that Arctic drizzling low cloud acts as a filter, strongly suppressing the transport of aerosol within the lower troposphere between the marginal (60-70ºN) and high (70ºN) Arctic. In addition, it is likely that the cloud layer acts to filter air vertically mixed down from the upper troposphere, reducing the effect of high altitude aerosol plumes (Warneke et al., 2010) on boundary layer concentrations. The spatial extent and efficiency of this filter is dependent on the imposed drizzle rate and stratocumulus boundary in the model as well as the height and thickness of the simulated cloud layer.**

Although Arctic drizzle is an additional deposition process in the model, it is sufficiently rapid at the edge of the Arctic that net deposition of SO_4 and BC is enhanced by drizzle only between about 60 and 70ºN, but decreased at higher latitudes (Figure 9). This result suggests that the majority of aerosol transported into the Arctic boundary layer enters horizontally through the lower troposphere/boundary layer and is quickly deposited to the surface after vertical mixing into the cloud layer.

20) 3440, 25: "... means that the ice cloud scavenging effect becomes much less important in summer" - Do you mean "the suppression of ice cloud scavenging becomes less important"?

Reworded to: **The generally higher temperatures and the predominance of low-level transport means that the suppression of scavenging in ice-clouds becomes much less important in the summer**

21) Table 1: Please indicate the meaning of "X" and "-". Assuming "X" means "included", are the symbols under column "ice-cloud scav" reversed? You might also consider separating effects by insoluble and soluble aerosol in this table.

The symbol X indicates processes are included. This has now been included in the comments for Table 1. An x in the column ice-cloud scav indicates that a simulation includes the suppression of ice-phase cloud scavenging as discussed in section 6.

22) Table 2: "stratacumulus"

Has been corrected

23) Table 3: Why are the values under DRIZZICE italicized?

For emphasis

24) Fig 6: "SO4 (a,c) and BC (b,d)" seems to be mis-labelled. Also, legends would help this figure.

Figure has been relabelled and a legend added

25) Fig 9: Please describe the meaning of the gray shaded "Arctic SZ" regions of panels (a) and(b).

The grey shaded region indicates the latitudinal extent of the Arctic stratocumulus zone (this description has been added to the comments)

26) Fig 10: Larger axes labels would improve this figure.

This has been done

Additional references

Hegg, D. A., A.D. Clarke, S.J. Doherty and J. Ström, Measurements of black carbon aerosol washout ratio on Svalbard, 2011, *Tellus*, 63B, 5, 891-900, doi:10.1111/j.1600-0889.2011.00577.x

Smith, A. J., V.E. Larson, J. Niu, A. Kankiewicz and L. D. Carey, Processes that generate and deplete liquid water and snow in thin midlevel mixed-phase clouds, 2009, *J. Geophys, Res.*, 114, doi:10.1029/2008JD0111531

Liu, J. , S. Fan, L.W. Horowitz and H. Levy II, Evaluation of factors controlling long-range transport of black carbon to the Arctic, 2011, *J. Geophys. Res*., 11, D04307, doi:10.1029/2010JD015145

Huang, L., S.L. Gong, C. Q. Jia and D. Lavoue, Importance of deposition processes in simulating the seasonality of the Arctic black carbon aerosol, 2010, *J. Geophys. Res*., 115, D17207, doi:10.1029/2009JD013478},