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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Space-based evaluation of interactions between pollution plumes and low-level Arctic clouds during the spring and summer of 2008" *by* K. Tietze et al.

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We are very appreciative to the reviewer for the careful read through of our manuscript, which enabled us to clarify some important points. We address the comments (bold) in a point-point fashion (normal) with associated changes made to the manuscript (italics).

This paper evaluates the relationship between cloud properties derived from satellite data and the concentration of a pollution tracer from a chemical transport model. This method provides some advantages to using satellites alone to constrain the aerosols and clouds. The authors succeed in presenting some interesting empirical relationships and the paper deserves publication based on

this contribution. However, the discussion succeeds only in proposing plausible explanations for the observations without demonstrating their validity.

This is an observational study, so we agree that there are limitations to the validity of the explanations in the absence of detailed modeling work, even if some of the mechanisms we discuss have been explored elsewhere by us and others. However, we feel that the paper would be poorer if we didn't highlight particular features in the results that might have plausible explanations. Our speculations may add to similar suggestions made by others in suggesting fruitful future avenues of research.

Major concerns: 1) The paper often appeals to cloud-top temperature and pressure values as tracers of air mass transport. This assumption is central to the methodology of the paper, where sorting of the data by cloud-top temperature and pressure is employed as a means of controlling for coincidental variations of aerosol and cloud that are not related to aerosol-cloud interactions (p. 29119, lines 16-18), and is also central to the interpretation of the results, where cloud-top temperature and pressure are taken as indicators of air mass age and origin (e.g. p.29130, lines 14-16). Presumably this approach is hampered by the fact that clouds coincide with mixing processes, which would act to dilute the properties of the original air mass. The strength of the methodology and the interpretation of the results seem to rely quite heavily on how well these measurable cloud-top properties represent traceable air mass properties. Can the authors cite a study or provide and argument for why cloud-top temperature and pressure can be interpreted as representative of air mass properties that are conserved during transport? Is figure 8 really showing the properties of different isentropic surfaces as suggested on P. 29128, line 19?

We make no argument for conservation during transport. Fig. 8 applies to an isentropic surface for the clouds that were sampled within the Arctic itself. Regarding mixing during transport, this is explicitly included within FLEXPART, as is cross-isentropic transport due to radiative cooling. We do cite a study (Stohl, 2006) that references these

effects. To make it more explicitly clear that the isentropic surfaces we refer to apply only to measurements within the Arctic, we have changed the conclusions and discussion as follows, including as well some reference to the potential for dry scavenging. In the conclusions

However, there is a pronounced decrease in the sensitivity of clouds to pollution plumes at Arctic temperatures that are both warmer and colder than freezing, or alternatively Arctic potential temperatures that are warmer or colder than about 286 K. We suggest that an explanation for this "inverted-U" phenomenon is the extent of time-integrated scavenging of aerosols along transport pathways from mid-latitudes. For Arctic air-masses with warmer temperatures, the decrease is due to more efficient wet scavenging of CCN in seasonally warm and moist air-masses. For Arctic air with colder temperatures, air rides closer to the surface and the transport time to the Arctic of air from mid-latitudes is prolonged, which increases potential exposure to precipitation and dry scavenging events..

In the discussion the text now reads

What is a bit surprising is that there appears to be lowered sensitivity of clouds to pollution plumes with locally cold temperatures below -6 C or below potential temperatures of about 278 K. It is unclear why this should be so given that wet scavenging is unlikely to be particularly efficient due to low precipitation rates. Perhaps one explanation is that locally cold air masses are also associated with longer transport times from mid-latitude pollution source regions (Stohl, 2006). It is the time-integral of precipitation rates that ultimately determines the extent of wet scavenging. Additionally, cold potential temperatures will tend to be more associated with surface air that has increased susceptibility to dry deposition (Spackman et al., 2010). It may be that the "inverted-U" shape in the IE signature appears due to two competing effects: where surfaces of potential temperature are cold in the Arctic, precipitation is low but dry deposition is high and transport times are long; conversely, where Arctic potential temperature surfaces are warm, transport times are short but precipitation is high. Values of IE are at a max-

imum for temperatures where there is a minimum in the time integral of precipitation and dry deposition along transport pathways.

2) A number of intriguing results are found when the dependence of the cloud optical thickness, cloud drop effective radius, and cloud liquid water path are evaluated for narrowly defined cloud-top pressure and cloud-top temperature bins. These include a reduced indirect effect at temperatures both less than and greater than 0 C, and a weaker IE for "graybody" clouds than "blackbody" clouds. Curiously, wet deposition of aerosols is invoked to explain the reduced IE for both high temperatures and low temperatures. Unfortunately, wet deposition is not explicitly evaluated in the study, either in the observations or in the FLEXPART modeling, leaving the explanation for the temperature effects rather unconvincing. Given that the mechanisms invoked in the discussion were not explicitly tested in the analysis, I think the authors should be more cautious in assigning specific reasons for their observed IE values. Can other possible explanations be excluded? Could not lower values of IE coincide with weaker updrafts or shallower clouds?

We think that some of the necessary caution is already explicitly stated, where the text states

A correlation between CO and LWP may also be expected for dynamical reasons. As a polluted mid-latitude and, thus, relatively warm air mass intrudes into the Arctic, it typically rises slantwise along a frontal surface above the colder Arctic air. A cloud formed in such an air mass may be expected to be deeper than an average Arctic cloud considered in our analysis. The impact on our results is minimized by controlling our analysis for both cloud top temperature and pressure and considering only stratiform clouds with cloud tops below 2 km, whose depth is clearly limited. Still, the effect may partly explain why IE_{LWP} values are larger than IE_{r_e} values.

Further explanation for the observed sensitivity of LWP to the pollution tracer CO war-

rants further investigation. Perhaps, sensitivity studies using LES (Large Eddy Simulation) type cloud models may provide better insight into interpreting our observations. Similarly, if precipitation observations could be coupled with our cloud property observations, a more precise understanding of pollution-cloud interactions could be achieved, since precipitation determines wet scavenging and is closely tied to cloud microphysical properties.

A "likely" is also changed to a "possibly" in the abstract.

3) I must be misunderstanding figures 6 and 9. It appears that in many cloud-top temperature bins the IE is greater for all samples regardless of LWP than it is for either samples with LWP<40 g mˆ-2 (labeled "graybody") or for samples with LWP>40 g mˆ-2 ("labeled blackbody"). Are there additional samples included in the "all LWP" sample that are excluded from either the graybody or blackbody samples? Likewise, in figure 9, the ratio of the IE for cloud optical thickness to the IE for cloud drop effective radius is substantially greater for the all LWP samples than for either of the other two groups of samples. How can this be if the graybody and blackbody samples are simply a division of the same data as the all LWP group?

A similar point was raised by Reviewer 1. Consider that in the limit of infinite subdivision, there could be no enhancement. That the magnitude of the enhancement is nearly identical for graybody and blackbody clouds suggests that, if a single mechanism is at play, it must apply over the full range of LWP. If so it would point more to drizzle-aerosol effects than cloud-radiative feedbacks, since the latter apply only to graybodies. To address this concern, we have added to the discussion, the following remark.

Note that any constraint on LWP is necessarily going to constrain the magnitude of any associated enhancement of IE_τ: the sum of the enhancement factors for graybody and blackbody clouds is equal to that for all clouds. What is implied, however, is that the magnitude of the enhancement is not specific to clouds of any particular thickness.

Thus, our results suggest that LWP in Arctic low-level liquid clouds is more sensitive to mid-latitude pollution plumes than is r_e. This is surprising, given that the most simple understanding of cloud physics is that values of LWP are determined primarily by thermodynamic constraints rather than aerosol concentrations. We cannot isolate an exact physical mechanism from the observations. One possibility, though, is that enhancement at low LWP may be indicative of a infrared radiative feedback process that accelerates cloud development when clouds are thin and polluted. More likely, given that the observed enhancement of IE_τ is not specific to a particular range of cloud LWP, is that suppression of warm rain and drizzle by pollution aerosol may lead to a long term thickening of liquid stratiform clouds.

Minor concerns: 4) A feedback process related to the LWP is mentioned in the abstract and elsewhere, without clearly explaining what that feedback process is. Is this process the cause of the IE_LWP? Or is the IE_LWP better explained by a suppression of rainfall, as is typically argued?

In the discussion we describe two possible feedback processes (see point 3 above), one being precipitation supression, and the other being radiative-dynamic feedbacks (see text above). We can't tell for certain which one it might be, which is why it is not specified in the abstract.

5) The success of the FLEXPART model in predicting CO concentrations is mentioned only qualitatively ("agreed well", P. 29123, line 5). Presumably the reference cited provides a quantitative assessment. Given that the explanation for the observed differences in IE is not explicitly tested in this study, one motivation for publishing the empirical results would be to provide a basis for a future comparison with model derived IE. But without some quantitative uncertainties, such as on the reliability of the FLEXPART CO, it will be difficult to evaluate how well a model calculation agrees with the observation. At the very least the paper should cite a quantitative uncertainty in the CO values from the literature. The results of this paper would be substantially more useful if the uncertainty in the

FLEXPART CO could be translated into an uncertainty in the IE attributable to FLEXPART errors.

The variability in the IE calculations is going to be overwhelmingly due to the dominant control of meteorology, and in any case, provided one makes the reasonable assumption that FLEXPART errors are due most to turbulence and therefore are random, they would not change the calculated value of the slope. We are not trying to be cavalier about this, because the reviewer raises a legitimate concern, but it is a difficult point to address with any great precision because the precise nature of FLEXPART errors is not extremely well characterized. Any validation method will have its own limitations. Nonetheless, we have amended the text in this section to be more explicit on this point. It now reads

Not only were pollution plume locations accurate, but, on average, predicted CO enhancements were within 30% of coincident airborne measurements (Warneke et al., 2010). Similar agreement has been seen in mid-latitude pollution plumes (Stohl et al., 2003, 2007). Some of this error is likely due to the limited sample volumes of aircraft measurements and would be lower for grid-cell averaged data. Finally, comparisons with space-based retrievals of CO for Arctic pollution plumes show a low bias of FLEXPART CO (which may also be due to retrieval errors and is not important for this study) but otherwise agreement of CO columns to within some 10% (see Fig. 8 in Sodemann et. al. (2010)).

6) It is stated that FLEXPART grid boxes with less than 50% cloud coverage are excluded from the analysis (P. 29125, line 23). How much of the total cloud cover in the sample set is left out because of this constraint?

We have rewritten the text to read

For the atmospheric heights below 800 hPa used in this study, clouds are generally stratiform so that within a typical FLEXPART grid box, the variability in cloud properties is relatively small. However, about 7% of FLEXPART grid boxes that were characterized

by liquid clouds had less than 50% cloud coverage. These cases were not incorporated in the assessment of pollution-cloud interactions.

7) Actual values of the probability densities are left out of figures 4 and 5. The qualitative points of the figures are conveyed, but they seem rather naked considering you went to the trouble of quantitatively processing a mountain of data.

Both plots are probability distribution functions, implying that the values are normalized to unity. Nonetheless the captions for these figures are now made more clear to indicate that the line spacing is linear.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 29113, 2010.