

# Answer to Editor comments on "Effects of long-range aerosol transport on the microphysical properties of low-level liquid clouds in the Arctic"

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March 18, 2016

Dear Graham,

We thank you for the different comments you made about the article. Please consider the following answers to your concerns.

- 1 *"By using PCI, you now introduce a rather strange concept which is the interaction between CO and clouds, which to my knowledge is not a real interaction. What is real is that CO correlates with aerosol and so can be used as a proxy for aerosol or CCN, especially in the absence of wet scavenging. When wet scavenging exists, it is not clear what the CO-cloud relationship means."*

There are two issues at hand: the sensitivity of clouds to aerosols as represented by the ACI, and the sensitivity of clouds to long-range transport of pollution plumes as represented by the PCI. These are not necessarily the same since aerosols may be scavenged en route. From a climate impacts standpoint, it is ultimately the PCI we are most interested in anyway – in the limiting case that all additional aerosols are immediately scavenged after emission, we would not care about anthropogenic aerosol indirect effects. The ACI would remain positive but more meaningfully the PCI would be zero since the aerosols are now gone. The PCI references changes in clouds to CO, which is inert but correlates with both pollution plumes and aerosols at their sources. The PCI and ACI are the same only provided aerosols are not scavenged, hence it's particular utility for estimating the impacts of long-range aerosol transport.

However, we appreciate that the term PCI is novel and do not wish to confuse. We volunteer as a substitute the terminology net ACI instead.  $ACI^{net}$  can be defined as

the ACI, as defined in previous papers less aerosol-scavenging interactions (ASI):

$$ACI^{net} = ACI - ASI \quad (1)$$

Thus,  $ACI^{net}$  represents the impact of aerosols assuming no dry or wet scavenging en route to the Arctic (e.g. the effect of pollution plumes). In the new version of the article we change PCI to  $ACI^{net}$ .

We change the text on pg. 7 line 2:" To account for scavenging, we employ the term  $ACI^{net}$  or the net aerosol-cloud interaction parameter.  $ACI^{net}$  is the same as the ACI while additionally accounting for any reduction of the ACI due to dry or wet scavenging of aerosols during transport.  $ACI^{net}$  can be interpreted as a measure of the sensitivity of a cloud at any given location to pollution plumes from distant sources. It allows for the passive components of a plume (e.g. CO) to remain while aerosols have been removed: (...)"

*"In a number of places the text becomes very awkward. For example, in the abstract: 'For a given set of meteorological conditions, we find that the liquid water path of arctic clouds does not respond strongly to pollution' Why should LWP respond to CO? What is the physical mechanism?"*

We refer here to the second indirect effect. Pollution plumes can contain high concentrations of CCN as well as CO provided the CCN are not previously scavenged. If CCN are present, then aerosol-cloud interactions are possible such that LWP would increase under conditions that sufficient q is available. Tietze et al. (2011) argued that observations suggest there exists an undetermined feedback processes that causes LWP to increase in response to pollution. Our results suggest that such a feedback is not as important as was initially thought, probably because Tietze et al. (2011) did not control sufficiently for meteorological variability. We change in the text:"Carefully controlling for meteorological conditions we find that the liquid water path of arctic clouds does not respond strongly to aerosols within pollution plumes."

*"or, 'If, however, aerosols are scavenged during long-range transport, then the cloud sensitivity to pollution is low even if the sensitivity to aerosols remains high.' what do you mean by cloud sensitivity to pollution? I think you mean to the particles for which pollution (CO) is a proxy, but this sounds very odd. And if the aerosols are scavenged, how can the sensitivity of the clouds to aerosols be high."*

We change the text to "But if CCN have been scavenged from pollution plumes, then the observed sensitivity of clouds to the pollution plumes should be expected to be low."

- 2 *"I am very confused by why the  $PCI_{r_e}$  is often quite different from the  $PCI_{\tau}$ . The authors seem to look for physical explanations, but if both are cover the same LWP range, and both use the same CO values, I do not see how large differences can follow, and any differences will likely be a result of methodology."*

Fig. 5 controls sensitivities for the specific humidity, LTS, cloud top temperature, and two ranges in LWP ( $0-40 \text{ g m}^{-2}$  and  $40-400 \text{ g m}^{-2}$ ). We observe in that figure that  $PCI_{\tau}$  and  $PCI_{r_e}$  have similar values. In Fig. 6 and Fig. 7 we do not control for LWP. The two PCIs are different because LWP can increase. Note that  $d \ln \tau = d \ln r_e + d \ln \text{LWP}$ .

*"If you are not stratifying by LWP, then please make this crystal clear (see questions below on Eqns). But since the LWP range is the same for both tau and re, why should there be such large differences in the two PCI values? This should also be true when you stratify by LTS or q since you would still include the same LWP values."*

When it is not mentioned in the text, we do not stratify by LWP (e.g. Fig. 6 and Fig. 7). We did not find any clear correlation between q and LWP (absolute value of correlation coefficient is 0.05) or between LTS and LWP (0.04). So when we control for q, it does not imply that the LWP is controlled for as well. For Fig. 6 LWP is only stratified by two bins (less than or greater than  $40 \text{ g m}^{-2}$ ) with the aim to distinguish whether cloud thermal radiative properties are determined by cloud brightness temperature or cloud droplet size (Garrett et al., 2006). The LWP range is the same for  $r_e$  and  $\tau$  but to some extent, increases in  $\tau$  may be due to increased LWP, hence the greater values of  $PCI_{\tau}$  compared to  $PCI_{r_e}$ . We add in the text at pg. 9 line 4: "In what follows, we examine the influence of LTS, specific humidity and pollution concentration on  $ACI^{net}$ . We do not stratify the data according to LWP."

- 5 *"On pg. 5, you state: 'The goal of this study is to use satellite, tracer transport model, and meteorological data sets to determine the effects of pollution on cloud microphysics due only to pollution itself and not to the meteorological state.' Again, pollution is suggested as influencing clouds. In addition, the end of the sentence implies that you are not interested in the role of meteorology, but that can't be the case since you bin by q and LTS."*

Pollution plumes can impact cloud microphysical properties because they are associated with different meteorological conditions and higher concentrations of aerosols. If we notice differences in cloud microphysical properties while controlling for LTS and q, than we can draw the conclusion that the observed differences are due to the aerosols. Further, we show in Figures 6 and 7 how meteorological state influences the magnitude of cloud-pollution interactions. This is a different question than studying the effect of meteorological parameters on cloud microphysical properties.

- 6 "*Eq. (3) should be formulated in terms of aerosol or CCN for it to be of any interest. As it stands it is quite obvious.*"

We changed in the text "droplet number concentration" by "CCN number concentration".

- 7 "*Eqns 4) and 5): are these partial derivatives at constant LWP or full derivatives. See question 2.*"

We change the derivatives to full derivatives to be coherent with the parameters used in this study.

- 8 "*Eq.6: What is being held constant for these partial derivatives?*"

See comments on point 7.

- 9 "*Eq. 7 seems inconsistent with Eqns 4 and 5 (partial vs. full derivatives).*"

See comments on point 7.

- 10 "*The methodology of placing less weight on outliers is an example of how methodology can influence results.*" The RMS method does not change the main conclusions of the article. We used this method to decrease the weight of isolated and calculate more precise results with limited contribution from outliers.

- 11 "*Pg. 9, line 12, I was looking for some explanation but never got one.*"

We explain it with Fig. 8: The polluted air parcels are associated with a higher cloud top potential temperature than clean air parcels. With warmer temperatures, precipitation and aerosol scavenging is more likely to occur along transport pathways, thereby decreasing  $ACI^{net}$ . Also, if the two types of air parcels come from different regions, their sources and chemistry might be different possibly influencing CCN efficiency.

These explanations are noted on pg. 10 line 21.

- 12 "*Pg. 9, line 19: is humidity a proxy of LWP? How well do they correlate?*"

LWP and specific humidity do not correlate significantly. The correlation coefficient is approximately 0.05. We include those results in the article on pg. 9 line 22: "The same applies for the specific humidity and the LWP. The correlation coefficient of the two parameters is 0.05."

- 13 "*Pg. 11, line 12: Higher LTS might mean clouds are more adiabatic, which would be consistent with Kim et al. 2008.*"

We state now the similarities on pg. 11 line 2: " This result is similar to results found by Kim et al. (2008) who found that aerosol-cloud interactions are strongest in clouds with adiabatic liquid water content profiles. Such clouds might be expected more frequently when LTS is high and there is reduced vertical mixing."

- 14 *"My personal opinion is that the Conclusions become too far reaching if one considers the uncertainties in quantification of the PCI or ACI values."*

We change the conclusion on pg. 11 line 21: "However, this study also suggests that any associated decrease in LTS could partially counter-act this effect"

- 15 *"I assume black is the highest density in Fig. 3. How does one make the decision to place less weight on the outliers? If one places too little weight on them then the slope is no longer defined (e.g., a fit through the black and green points might give -ve  $PCI_{re}$ .)"*

The fit method places less weight on outlier points. This is especially true for isolated points. In Fig. 3, the weight of points indicated by a red line is approximately similar to those from black and blue lines. We clarify this in the text on Pg. 7 line 29: "In Fig. 3, points indicated by the red line are weighted similarly to those indicated by black and blue lines. The slope retrieved by the linear fit, in Fig. 3, is  $-0.13 \pm 0.016$ . Referring to Eq. (5)  $ACI_{re}^{net}$  equals  $+0.13 \pm 0.016$ ."

- 16 *"Pg. 2, line 26: is orthogonal the correct word?"*

By orthogonal we mean that passive tracer and cloud properties are decoupled.

- 17 *"The tau, LWP, re relationship used is for a homogeneous cloud. Please state so."*

We add it on pg. 7 line 19

- 18 *"'15% of total space': Is this area, number of pts?"*

It refers to area. More precisely it refers to the range length of specific humidity and LTS. We specify it in the article on pg. 8 line 6.

- 19 *"In addition to Tietze et al. the use of CO as a CCN proxy has also been applied by Berg et al. (2011, GRL) and Lance et al. (2012, ACP)."*

We add both citations on pg. 2 line 20.