Takuro Michibata Research Institute for Applied Mechanics Kyushu University 6-1 Kasuga-Park, Kasuga, Fukuoka 816-8580, Japan phone & fax: +81-92-583-7772 e-mail: michibata@riam.kyushu-u.ac.jp

September 19, 2014

Dear the editor of Atmospheric Chemistry and Physics,

Enclosed please find our revised manuscript (acp-2014-213) submitted to Atmospheric Chemistry and Physics. It has been revised according to the editor and referees' comments.

We wish this revised manuscript deserves to publication.

Sincerely,

Takuro Michibata

Response to Editor of acp-2014-213

Dear Editor,

Thank you very much for handling our manuscript. We revised our manuscript according to your comments. The authors' replies on individual issues are below. I hope that the manuscript is now acceptable for publication in *ACP*.

> **[EC]**: Editor comment <u>in *Italic*</u> [AC]: Author comment

<u>General issues:</u>

[EC] The manuscript draws some strong conclusions about the attribution of cloud macro and microphysical differences to aerosol effects, which is entirely based on the comparison of different regions that show different levels of aerosol optical depth. However, the analysed regions differ in a large number of factors, including land / sea fractions (the polluted region is predominantly over land, the clean regions predominantly over sea), that are also expected to affect cloud macro- and microphysical properties. I therefore concur with the reviewers that related statements and conclusions about aerosol effects should be less unambiguous and clearly stating assumptions and uncertainties. Alternatively please extend the analysis to support the attribution to specific effects. This relates to:

[EC1] Page 6, line 97: "This suggests the occurrence of the second indirect effect (Albrecht, 1989)".

[AC1] We used "may" in this sentence as follows: "This may suggest the occurrence of the second indirect effect (Albrecht, 1989)".

[EC2] Page 7, line 12 "We interpret this characteristic of CFODDs as a second indirect aerosol effect around the cloud base (large part of ICOD) over the Industrial area."

[AC2] We indeed think that to emphasize this interpretation may a little insufficient at this stage. Although the profile of aerosol concentration can be obtained from CALIPSO for example, this cannot measure reliable vertical aerosol information when optically thick cloud exists as in our study. In fact, we have been conducting further analysis to reveal this mechanism, based on sensitivity experiment using numerical simulation. We changed this sentence as follows: "We would like to interpret this characteristic of CFODDs as a result of suppression of precipitation due to high concentrated aerosols

around the cloud base (large part of ICOD) over the Industrial area.".

[EC3] Page 7, line 15: "over mid-latitudes in the Northern Hemisphere, and the stable and high LTSS environment results in a high aerosol concentration near the surface. Therefore, an aerosol-cloud interaction may occur that results in weaker radar reflectivity in the larger ICOD region."

[AC3] We added the following sentence after the above description: "This may be one of the hypotheses, and further analysis (e.g., sensitivity experiments using numerical modeling) is required in order to enhance the credibility.".

[EC4] Page 7, line 79: "In addition, we found a difference in "contoured frequency by optical-depth diagram" (CFODD) between the pristine oceanic area and the polluted land area, reflecting the aerosol–cloud interaction."

[AC4] This sentence was changed as follows: "In addition, we found a difference in "contoured frequency by optical-depth diagram" (CFODD) between the pristine oceanic area and the polluted land area, implying the aerosol-cloud interaction.".

In addition, we added a sentence after this (according to the referee #2): "However, we cannot completely exclude the possibility that other meteorological factors may be responsible for the differences between land and ocean."

Please see also the author reply to referee #2 –AC8 (below).

[RC8] Conclusions, p7.88. I think 'suggesting aerosol-cloud interaction' (or 'consistent with...') would be preferable to 'reflecting aerosol-cloud interaction'. I would also suggest adding a sentence after this, along the lines of "However, we cannot completely exclude the possibility that other meteorological factors may be responsible for the differences between land and ocean."

[AC8] We adopted your suggestion, and revised/added these parts as follows:

p7.82-83. "reflecting the aerosol-cloud interaction" to "implying the aerosol-cloud interaction"

p7.83~. added "However, we cannot completely exclude the possibility that other meteorological factors may be responsible for the differences between land and ocean."

[EC] Clouds in this manuscript are characterised by some of their macro- and microphysical properties. However, the actual cloud regimes may be very different for different seasons and regions. This should be clear.

Specific issues:

[EC5] Page 4, line 38"The results suggested that the precipitation occurrence is most strongly related to LWP, except for the Industrial area."

This is not directly obvious from the table. Please add more explanation that supports your conclusion.

[AC5] We modified this sentence in p4.40 to make it clear: "The results suggest that the precipitation occurrence is related to LWP (e.g., North Pacific 1 where higher LWP region is accompanied with high '[%] with rain'; Table 1), except for the Industrial area (i.e., high LWP but lower '[%] with rain', and vice versa).".

[EC6] Page 4, line 43"more cumulative cloud"
Presumably you mean convective or cumulus cloud?
[AC6] We have changed it to "convective or cumulus cloud".

[EC7] *Fig. 3: the quality plot should be improved.*

[AC7] We strongly agree with your comment "the actual cloud regimes may be very different for different seasons and regions". We added the information of mean values of LWP and LTSS for each diagram of Fig. 3, as follows (next page). The examination of cloud macro- and microphysics associate with atmospheric stability is one of the main points in our study. This improvement would be helpful for readers. In addition, the following explanation was added:

(p.5.12-13)

"(i.e., from no precipitation to rain<mark>, with a monotonous increase in LWP and a slight decrease in LTSS</mark>)".

(p5.18–)

"However, in the Industrial area where air pollution by anthropogenic aerosols is severe, the transition pattern is not as clear as over the ocean, and the variations of LWP are relatively small."



Fig. 3. Fractional occurrences of cloud optical thickness (COT) and cloud droplet effective radius (CDR) for each rain category: [A] no precipitation ($Z_{max} < -15$), [B] drizzle ($-15 < Z_{max} < 0$), and [C] rain ($0 < Z_{max}$). (**a**-**c**) are for the Industrial area in JJA, (**d**-**f**) for the Industrial area in DJF, (**g**-**i**) for the North Pacific 3 area in JJA, and (**j**-**l**) for the North Pacific 3 area in DJF. Averaged LWP and LTSS are shown in each diagram.

[EC8] Fig. 4 and 5: Define "Transition pattern of drizzle intensity" and explain this a bit more. Make captions entirely self-explaining. The arrows are not entirely self explaining and it needs to be entirely clear how they have been derived.

[AC8] These parts of captions (Fig. 4 and 5) were slightly changed from "Transition pattern of drizzle intensity" to "The distribution of maximum radar reflectivity Z_{max} as a function of LWP and *N*c". In addition, the explanation of black arrows was also added as follows: "The black arrows on (d) indicate one possible interpretation of growing process from cloud droplet to drizzle and raindrop (see text for details)."

Please see the author reply to referee #2 - AC4 as well (below).

[RC4] Results, p5.106-p6.2. This is one possible explanation. May it not also be possible that V-shape over the Japan region may arise due to land vs ocean differences that are independent of aerosol effects? After all, the Japan region has a mixture of land and ocean.

[AC4] We see your point. We cannot emphasize this detailed mechanism, because such V-shape transition pattern is just one possible explanation, and the lack of dataset for time-series in current observational techniques. However, to provide a possible interpretation of such the characteristic V-shape pattern, which may suggest a cloud growing process (from cloud droplet to drizzle and raindrop), would be valuable for readers.

[EC9] Page 7, line 12 "We interpret this characteristic of CFODDs as a second indirect aerosol effect around the cloud base (large part of ICOD) over the Industrial area."

It is not entirely clear what you mean by an "aerosol effect around the cloud base", in particular as it is unclear what cloud types this relates to.

[AC9] We changed this sentence, as mentioned in [AC2] above.

Single-layered lower cloud (only liquid phase) is the analysis object in our study. Therefore, we do not consider convective cloud in this study.

Thank you very much for handling and reviewing our manuscript.

Sincerely yours,

Takuro Michibata

Response to Reviewer #2 of acp-2014-213

Dear Reviewer,

Thank you very much for taking your time to review our paper again. I am returning herewith a manuscript revised according to your comments. I hope that the manuscript is now acceptable for publication in *ACP*.

> [RC]: Referee comment <u>in *Italic*</u> [AC]: Author comment

[RC] The authors have improved the quality of their manuscript. As long as they give due consideration to the minor comments below, I recommend publication in ACP.

[AC] We have tried to revise our manuscript so as to answer to your comments. Our reply and corrections on individual issues are below.

******Specific comments (minor)******

[RC1] Abstract, p2.22-24: I'm still not fully convinced that the results necessarily show that "aerosol-cloud interaction mainly occurs around the cloud base in polluted land areas during the winter season." I would recommend that you communicate some uncertainty by re-writing this sentence e.g. "our results suggest that", "our results are consistent with", or just include "may" somewhere in the sentence.

[AC1] According to your comment, this sentence was a little modified as follows:

"Our results suggest that aerosol-cloud interaction mainly occurs around the cloud base in polluted land areas during the winter season.".

[RC2] Introduction, p2.30-36: The first sentence refers to aerosol-cloud interactions, the second to the aerosol direct effect, and the third to aerosol-cloud interactions again. The logic of these sentences could be rearranged to make them less confusing.

[AC2] We reconsidered the logical structure in the initial part of the introduction, and rearranged for readers to understand more clearly.

(former version)

Aerosol particles play an important role in the climate system by serving as cloud condensation nuclei. The radiation budget is affected by their scattering and absorption properties, which are referred to as aerosol-radiation interactions. In addition, aerosol-cloud interactions affect cloud optical thickness (COT) and cloud particle size (e.g., Twomey 1977) as well as cloud lifetime (e.g., Albrecht 1989). (revised version)

The Earth's radiation budget is some affected by the scattering and absorption properties of aerosol, which are referred to as aerosol-radiation interactions. In addition, aerosol particles play an important role in the climate system by serving as cloud condensation nuclei (aerosol-cloud interaction). This affects cloud optical thickness (COT) and cloud particle size (e.g., Twomey 1977) as well as cloud lifetime (e.g., Albrecht 1989).

[RC3] Data and methodology. This section has been improved by the inclusion of further details. However, the structure/flow could be improved further. In particular, the discussion of derived-Nc could be moved to a section on the MODIS data.

[AC3] The discussion and derivation of *Nc* and LWP, which were retrieved from MODIS data (matched along the CloudSat footprint), were moved to the subsection 2.1 on the MODIS data. (i.e., p4.4–p4.26 was move to the last part of the subsection 2.1)

[RC4] Results, p5.106-p6.2. This is one possible explanation. May it not also be possible that V-shape over the Japan region may arise due to land vs ocean differences that are independent of aerosol effects? After all, the Japan region has a mixture of land and ocean.

[AC4] We see your point. We cannot emphasize this detailed mechanism, because such V-shape transition pattern is just one possible explanation, and the lack of dataset for time-series in current observational techniques. However, to provide a possible interpretation of such the characteristic V-shape pattern, which may suggest a cloud growing process (from cloud droplet to drizzle and raindrop), would be valuable for readers.

[RC5] Results, p6.32-35. In light of the modified discussion about the nature of the cloud geometrical thickness measurements (p6.15-24), it might be helpful acknowledge that the strong correlation between between cloud geometrical thickness and Zmax may be driven by precipitation causing an increase in cloud hydrometeor thickness (i.e. the direction of causality is not clear).

[AC5] Even the active sensor CloudSat cannot measure exact cloud geometrical thickness, therefore this note (p6.15-24) has been added. Thus we also cannot extend to examine the correlation direction of causality.

[RC6] Results, p7.22. Consider adding 'or other meteorological factors' after the Nakajima et al. reference.

[AC6] According to your comment, we added ", or other meteorological factors as well" in the revised manuscript.

[RC7] Results, p7.42-52. I appreciate that you have added this paragraph following my earlier comments. However, the paragraph contains two different ideas: 1. the fact that genuine aerosol-cloud interactions may behave differently under different meteorological conditions; and 2. the fact that meteorology may drive aerosol-cloud relationships (even in the absence of any aerosol-cloud interactions). Some readers may be confused by this. It would be worth considering how to make this distinction clearer to readers. (In light of this distinction, you may also want to consider modifying the final sentence of the conclusions at p7.92.)

[AC7] We think both of above ideas ("1. the fact that genuine aerosol-cloud interactions may behave differently under different meteorological conditions; and 2. the fact that meteorology may drive aerosol-cloud relationships (even in the absence of any aerosol-cloud interactions)") are important for our future work. The following sentences were added in subsection 3.4, to state these problems clearer with important relevant reference. We are currently satisfied with the sufficient description in the section of conclusions.

(Results, p7.44)

We must be careful about following two ideas: one is the fact that genuine aerosol-cloud interactions may behave differently under different meteorological conditions; and the other is the fact that meteorology may drive aerosol-cloud relationships (even in the absence of any aerosol-cloud interactions). Such the meteorological gradients sometimes cause spurious correlations (Grandey and Stier, 2010).

[RC8] Conclusions, p7.88. I think 'suggesting aerosol-cloud interaction' (or 'consistent with...') would be preferable to 'reflecting aerosol-cloud interaction'. I would also suggest adding a sentence after this, along the lines of "However, we cannot completely exclude the possibility that other meteorological factors may be responsible for the differences between land and ocean."

[AC8] We adopted your suggestion, and revised/added these parts as follows:

p7.82-83. "reflecting the aerosol-cloud interaction" to "implying the aerosol-cloud interaction"

p7.83~. added "However, we cannot completely exclude the possibility that other meteorological factors may be responsible for the differences between land and ocean."

******Technical corrections/suggestions******

[RC9] *p2.47-p3.30:* This updated paragraph is very long. Breaking it into two or more shorter paragraphs would improve readability.

[AC9] We divided this paragraph, according to your suggestion.

[RC10] *p4.13: incorrect use of semi-colon.*

[AC10] semi-colon was replaced by colon, in the revised version.

[RC11] *p4.38: 'suggested' to 'suggest'.*

[AC11] We changed 'suggested' to 'suggest'.

[RC12] *p4.43* and *p4.45*: should 'cumulative' be 'cumulus'?

[AC12] p4.43: We changed 'more cumulative cloud' to 'convective or cumulus cloud'. p4.45: We changed 'cumulative' to 'cumulus'.

[RC13] p4.50: using parentheses (brackets) like this is confusing. The easiest way to remedy this would be to include just one logical flow (i.e. delete the three words in brackets).

[AC13] This part (p4.50-54) was modified in the revised manuscript.

[RC14] *p4.90: I'm still confused by the use of 'the typical'. Does this have a technical meaning? If not, then consider replacing this with 'are commonly considered cloud physical variables' or similar.*

[AC14] We corrected this sentence, according to your comment.

[RC15] *p7.54-95: long paragraph that could be broken down to improve readability.* **[AC15]** We corrected.

[RC16] Fig. 1: In the response to reviewer 2 (AC24), you mentioned that Fig 1 had been modified as per my suggestion to indicate the regions in the top figure. However, the updated draft does not appear to incorporate this change. (To clarify, I do not expect you to make this change. I am merely pointing it out in case you accidentally submitted an outdated version of the figure. The other figure changes are looking good.)

[AC16] I hope that top panel of Fig. 1 was provided with the region lines in the revised paper. Thank you for your reminder.

References

Grandey, B. S. and Stier, P.: A critical look at spatial scale choices in satellite-based aerosol indirect effect studies, Atmos. Chem. Phys., 10, 11459–11470, doi:10.5194/acp-10-11459-2010, 2010.

We would like to thank the referee for helpful comments and support of revising our paper.

Sincerely yours,

Takuro Michibata