

## ***Interactive comment on “Aerosol-cloud interaction determined by both in situ and satellite data over a northern high-latitude site” by H. Lihavainen et al.***

**H. Lihavainen et al.**

heikki.lihavainen@fmi.fi

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We would like to thank the reviewers for their comments. We have replied to the questions, see below, and changed the manuscript according their suggestion, which we think made the paper stronger.

As a supplement material is the revised version. The track change has been on in the revised version to help find changes we have done.

Anonymous Referee #1 Received and published: 8 January 2010 General comments. This paper aims at quantifying the first aerosol indirect effect (AIE) from measurements taken at the ground and analysis of satellite data. There is however a general misunderstanding of what the first AIE is, mainly based on former publications. Following recent

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meetings and international assessments, the situation has now significantly evolved. I therefore recommend that the authors follow these recommendations and focus on the original part of their contribution to this subject. Underlying the aerosol indirect effects is the fact that the droplet number concentration in warm cloud depends (among others) on the concentration of cloud condensation nuclei, which depends on the size distribution and chemical composition of the aerosol particles. This was a very active field of research in the 1950th, referred to as “aerosol/cloud interactions”. The concept of AIE was introduced later when Twomey (and others) hypothesized in the 1970th, from aerosol/cloud interaction studies, that the increase of the droplet concentration should result in an increase of the cloud optical thickness (at constant LWP).

Formally the aerosol indirect effect expresses as  $d\ln\text{COT}/d\ln N_d$ .

Later on, and combining aerosol/cloud interactions and the Twomey hypothesis, attempts were made to connect the whole chain from aerosol to cloud optical thickness ( $d\ln\text{COT}/d\ln\tilde{A}_a$ ) using diverse proxy ( $\tilde{A}_a$ ) for the aerosol and assuming diverse relationships between that proxy and the droplet concentration as  $d\ln N_d/d\ln\tilde{A}_a$ . The rationale for such an approach is to develop simple parameterizations of the AIE in general circulation models where the aerosol/cloud interactions cannot be explicitly simulated, but information exists about the spatial distribution of chemical composition of the aerosol. This paper addresses the first part of the chain (aerosol/cloud interactions), i.e.  $d\ln N_d/d\ln\tilde{A}_a$ , discussing the best way to define  $\tilde{A}_a$  and the consequences for the simulation of the AIE in GCM. The most robust approach is to directly measure  $N_d$ , such as in Data set 2 and 3. When deriving the droplet radius from satellite measurements, the correlation generally decreases due to uncontrolled fluctuations of the LWC ( $N_d$  is proportional to the ratio of LWC to the power 3 of the droplet size). To tackle the first indirect effect is more difficult because the COT shall be measured in clouds with the same LWP. Considering that COT is roughly proportional to  $LWP \times N_d^{1/3}$ , its value is 3 times more sensitive to LWP fluctuations than to variations of  $N_d$ . “Fluctuation” is used here to describe the parameter that is not measured precisely (LWP), while “vari-

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ation” is for the parameter from which the susceptibility is derived ( $N_d$ ). This results in a reduction of the correlation in the derived ACI due to the broad range of LWP values accepted in the data base, i.e. from 100 to 200  $\text{gm}^{-2}$ . For the contributions of LWP fluctuations (a factor of 2) to be negligible compared to  $N_d$  variations, the measured  $N_d$  values should vary by a factor of  $2 \times 3 = 6$ . The changes in the quantification of the ACI and the reduction of the correlation factor is mainly due to the added noise when trying to address the whole chain without precisely constraining LWP.

ANSWER: We agree with the reviewer. In the revised version of the paper, a more detailed discussion on the problems and uncertainties associated with determination of the first indirect effect is presented (Second paragraph in Introduction).

I therefore recommend to focus the paper on the most reliable part of the study, i.e. “aerosol/cloud interactions” and discuss more in depth the results shown in Fig. 1 and 2 and the consequences when trying to connect the aerosol to the cloud optical thickness to quantify the AIE.

This part is original in the sense that it characterizes a regional aerosol burden and provides important information on which part of this aerosol is likely to be activated in boundary layer clouds. The approach based on statistical variation of the ACI with the lower bound diameter of the spectrum and the sudden decrease of the correlation seems very powerful to characterize this property on a statistical basis. Interpretation of these results could also be slightly developed, especially if there are additional information on the chemical composition of the aerosol. Another issue of interest would be if, within each data base, a few aerosol types can be identified, with very different activated fractions, depending on the air mass origin for instance, that could further be used to parameterize aerosol/cloud interaction in a GCM. Finally, it would also be useful to perform a simple calculation of aerosol activation using a parcel model and vertical velocities typical of the boundary layer clouds. I assume it is probably in the range from 0 to 2 m/s with a mode at 1, or slightly less than 1 m/s, as in typical BL clouds. This would help at understanding if the droplet concentration in these cases is

aerosol limited or dynamically limited.

The part of the study based on satellite retrievals does not deserve publication since it has the same weaknesses as previous analyses (unknown LWP), and a lower statistical significance compared to global studies.

ANSWER: The two other reviewers did not recommend removal of the satellite data from the paper, so we decided to keep them in. However, in the revised version we have put more emphasis on ground-based measurements, as well as on the combination of ground-based measurements and satellite retrievals. The remote sensing data were recalculated with a larger data set and more stringent constraints on LWP (section 3.2). Aerosol chemical composition and air updraft velocities are discussed briefly as well (section 3 prior to 3.1).

Detailed comments

Page 27470, line 22: “The first two partial derivatives must be calculated at the constant LWP or cloud liquid water content”. The framework of the first AIE hypothesis is constant LWP. Constant LWC is not sufficient unless its vertical integral is, i.e. the LWP.

ANSWER The requirement of a constant LWP is explicitly brought up in the revised manuscript.

Page 27471, line 4: where  $d_k$  is the. . .  $d_k$  cannot be mentioned here because it does not appear in the equation. It should rather be mentioned 4 lines below where the diameter range is discussed.

ANSWER The quantity  $d_k$  appears on the left hand side of the equation

Page 27472, line 7: in the range  $d_k=50-150$  nm (not 15)

ANSWER Corrected.

Page 27473, lines 5 to 9: No, the main reason for the decrease of the correlation and lower ACI values are the fluctuations of LWP.

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ANSWER In spite of dividing the data according to LWP, we see a similar behaviour. This is now discussed in section 3.2.

Fig. 1 and 2:  $d$ , on the X axis

ANSWER OK

J. Quaas (Referee)

Lihavainen et al. investigate the influence of aerosol concentrations on cloud microphysical properties. Since by these processes, anthropogenic aerosols may indirectly affect the Earth's energy balance, careful analysis and process understanding is essential for climate science. It is particularly valuable to assess aerosol-cloud interactions from observations, thus also providing evaluation metrics for climate model parameterisations. The study follows propositions in the literature to define a metric of the impact of aerosols on cloud microphysics formally as the partial derivative of the microphysical quantity with respect to the logarithm of aerosol concentration. The observation of both aerosol concentration is taken in this study from different kinds of observations at or in the region of one particular site in Finland. Specifically, the metric as derived from satellite remote sensing data is compared to in-situ data and a combination of the two data sources. The study thus addresses an important subject which is pertinent to the field covered by Atmos. Chem. Phys.. The applied methods are adequate and interesting. The English language is very good. There are a number of points on which in my opinion the study could be improved considerably.

Literature review I would suggest the authors discuss in their paper the article by Andreae (Atmos. Chem. Phys., 9, 543-556, 2009) which shows that cloud-base cloud condensation nuclei concentrations are very well correlated with AOD.

ANSWER The paper is now mentioned in section 3.3.

There is of course already a large body of in-situ results for the aerosol-cloud interaction metric, which is not discussed in the paper. An example is the study by Boucher

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and Lohmann (Tellus, 1995) who summarized aircraft results available at that time. Their results have been implemented as a parameterisation in many general circulation models. Their formula "D", e.g., yields  $ACI = 0.41 / 3$ . At least some review of the plenty of aircraft studies should be given.

ANSWER This study is now discussed in section 3.4.

Important studies of the scale-dependency of the metrics for aerosol-cloud interactions have been described by Sekiguchi et al. (J. Geophys. Res. 2003) and the paper by McComiskey et al. (J. Geophys. Res. 2009) already cited. More discussion of their results would be useful.

ANSWER This issue is mentioned in section 3.2 of the revised manuscript.

Main comments The study addresses only the first aerosol indirect effect (Twomey effect). A crucial element in defining this is that cloud liquid water path (LWP) is held constant. In this study, this is condition not always really satisfied. For the remote sensing data, LWP is only a confined to a very broad range ( $100 - 200 \text{ g m}^{-2}$ ), which, in addition, pre-selects a particular cloud type. One possibility to better address this issue would be to stronger constrain LWP. Another one would be to rely on the satellite-derived cloud droplet number concentration as, e.g., Quaas et al. (Atmos. Chem. Phys., 9, 8697-8717, 2009). It would be useful to directly compare the individual quantities (cloud droplet effective radius and aerosol concentration) of remote sensing vs. in-situ observations rather than only the regressions between the two.

ANSWER The remote sensing data were recalculated with a larger data set and more stringent constraints on LWP. In the revised version LWP is divided to  $10 \text{ g m}^{-2}$  bins.

Specific comments I23: not meteorological conditions, but just updraft velocity probability distribution (c.f. Köhler theory)

ANSWER Corrected.

I26: this is not quite true. If effective radius or cloud optical depth are used, it is a metric

for the first aerosol indirect effect only when cloud liquid water path is held constant.

ANSWER Corrected.

p27467 I16: Do I understand it correctly that at the site, aerosols are measured continuously, but cloud properties only during specific measurement campaigns? Please specify. Are clouds in all cases liquid water clouds?

ANSWER Aerosols are measured continuously, and lately also some cloud properties. We chose to use these data because they have the best quality assurance. All clouds studied here are liquid-phase clouds.

p27469 I11: Is this limitation on cloud top assuring that only entirely liquid-water clouds are looked at?

ANSWER The reason for this limitation is to look at low-level clouds only, so that ground-based aerosol measurements could be connected more reliably to cloud properties. This also limits cloud temperatures.

I16: This choice for LWP range would need to be justified. Does the aerosol-cloud interaction metric quantification depend on this choice? How large would the parameter be if the range was reduced, what if clouds with less LWP would be investigated? Is the LWP the one retrieved by MODIS? It would be very useful if a histogram of LWP would be shown. What is the LWP distribution for the in-situ cases?

ANSWER The range has been changed, see above. The LWP is the one retrieved from MODIS. There are only small fraction (<10 %) of LWP retrievals available for in situ cases, so it leaves does not serve it purpose. We decided to leave it out from this study.

I21: Should this read "doy"?

ANSWER Corrected.

I23: It would be very useful to add a comparison (joint histogram) of effective radius

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and aerosol concentration in-situ vs. remote sensing.

ANSWER Unfortunately, we did not get the point made by the referee

I25: What is actually used: Collection 4 (Remer et al.) or collection 5 (Levy et al.)?

ANSWER It is collection 5. We changed the wording a bit

p27470 I5: It would be useful to add another study in between 2.1.2 and 2.1.3, in which the same cloud microphysical data are used as in 2.1.2, but the AOD as in 2.1.3, in the 1\_x1\_ grid-box around the cloud retrieval. This would allow to assess to which extent the spatial averaging of the cloud properties is responsible for the decrease in the aerosol-cloud interaction metric.

ANSWER This would lead to very low number of data points, since there are only few (slightly >over 200) AOD retrievals altogether (Aqua and Terra) only from 50 km around Pallas over six years (2000-2005). And these retrievals are mostly on non-cloudy days. We decided to leave this kind, though useful, study out.

I9: Please specify that this definition is only true for constant liquid water path.

ANSWER Corrected.

I17: (eq. 1) It is formally a partial derivative.

ANSWER Corrected.

p27471

I3: Some words on how this equation is evaluated numerically are needed. Is it the slope of a linear regression?

ANSWER Text; “ Equations 1 and 2 are calculated as a slope of linear regression of  $\ln \tau_{\text{a}}$  as a function of  $\ln \tau_{\text{a}}$ , or  $\ln \text{COT}$  as a function of  $\ln \tau_{\text{a}}$  etc.” Was added to the end of chapter

I9: I had difficulties in understanding this definition. A potentially clearer re-formulation

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would be: "...we calculate different  $ACI(d_k)$  for varying cut-off aerosol diameter  $d_k$  by..."

ANSWER The text around equation 2 was modified to make it more understandable.

I18: Could you report here some information about aerosol chemical composition and characteristics of the updraft probability distribution?

ANSWER A brief discussion on these issues is included in the revised manuscript.

I21:  $ACI=0.3$  means  $dnNd / d \ln \bar{A}_a = 0.9$ , right?

ANSWER Yes, this is correct. Different ways of calculating  $ACI$  in equations 1 and 2 are now presented in a clearer way.

I4: Is most interesting that this seems to be true even for the lowest size bin.

ANSWER We added some text about the low probability of new particle formation, which affects the >lowest sizes of the particle number size distribution.

I6: It would be sufficient if the CCN concentration scaled with the aerosol number concentration.

ANSWER We agree on this.

I7: Which  $dk$  range is actually meant? To me, it looks more like 20 – 300 nm.

ANSWER There was a typo in the text that was corrected. The whole paragraph refers to particle sizes between 50 and 150 nm.

I9: drop "3"

ANSWER Done.

p27473 I10: A reason for this could be variations in liquid water content. LWC scales with cloud droplet effective radii scale to the third power, while it scales only linearly with optical depth.

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ANSWER With our new approach, the difference decreased and became in practice negligible (see figure 3a and the revised text in section 3.2.

l21: To be more clear on this, you mean, the local aerosol concentration is not necessarily representative for the entire 5\_x5\_ region, right?

ANSWER No. When combining in situ aerosol measurement with cloud retrievals, only retrievals in >an area with a radius of 20 km around Sammaltunturi was used.

p27481 Fig.1: Please label in nm rather than m (10nm – 100nm – 1 $\mu$ m). Maybe reformulate "as a function of cut-off size. (c)"

ANSWER We rather stick with SI units here.

p27483 Fig. 3: What is the meaning of the error bars? Could you show a histogram of AOD?

ANSWER Figure was removed

Anonymous Referee #3 Received and published: 1 February 2010

#### General comments

Lihavainen et al. investigate aerosol-cloud interactions based on in-situ observations, a combination of in-situ and satellite cloud observations and just only satellite retrievals of cloud microphysical/optical and aerosol optical properties. A major part of our poor understanding of the role aerosols has on global climate change is due to the large uncertainties that are associated with aerosols influences on clouds. Since satellite sensors, constructed for investigations of cloud and aerosol optical and microphysical properties, have produced data for nearly 10 years now there is a good opportunity to use the retrieved products in aerosol-cloud interaction studies. Thus, aerosol trend could be studied but more important for the present study the aerosols influence on clouds could be observed based on a large and robust data set. Even so, both the aerosol and cloud products need to be validated. For aerosol optical thickness Levi et

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al. 2007 have shown that the MODIS Collection 5 product agrees well with AERONET sun-photometer measurements. Considering the cloud products no such observation global network is however available, which mean that the results from these retrievals may be associated with substantial uncertainties. On the other hand, by comparing the aerosol cloud interaction (ACI) by using the three different approaches in the study by Lihavainen et al gives an opportunity to actually compare the satellite retrievals with probably more reliable observations obtained from the ground. The results in Figure 3 are also very promising. The inconsistency in ACI, estimated based on three different approaches, is discussed in the present study, but the assumed reasons for the discrepancies should, however, be more clearly explained. It is important that the investigations of ACI, based on the three different approaches, uses same time periods and is nearly consistent concerning the spatial resolution and finally be close to the definition of ACI as well as. The latter means that the authors should more properly take the LWP into account when ACI is estimated (see comment 1 below). The consequence of the poor horizontal resolution in the data used for the satellite cloud and aerosol optical retrievals are used, compared to two other methods, to estimate ACI should be more properly discussed (see comment 2 below). The study by Lihavainen et al is relevant for ACP, although there are a number of points that have to be taken in consideration before the work can be accepted for publication in ACP.

1) In an attempt to obtain consistent conditions for the three different methods, used to estimate ACI, include also data set 4 in the comparisons between satellite and ground based data. For the same reason extend the time period in the study, when only satellite aerosol and cloud retrievals are used to estimate ACI, so that it cover the years 2000 to 2007. This will increase the data set and give the opportunity to estimate ACI with a subdivided range of LWP values to exclude or reduce dynamical effects. In addition, the authors should plot the ACI values as a function of LWP (all retrieved values) both for COT and effective radius. The LPW should be subdivided into smaller range (for example with increments of 20 g m<sup>-2</sup>) and the variability around each mean

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ANSWER In our revised version we took a different approach to combine the ground base aerosol data and satellite retrievals. In the new results the ACI is investigated in  $10 \text{ g m}^{-2} > \text{LWP}$  bins.

2) The present grid of  $1^\circ \times 1^\circ$  for the averaged aerosol and cloud properties imply an area approximately something like 40 km, thus, approximately 10 times larger than for the in-situ measurements. Additionally, to get probably enough of aerosol optical data in the present study a very large area ( $65^\circ$  to  $70^\circ$  N,  $20^\circ$  to  $30^\circ$  E) is used as an investigation area in the present study. This area is also very close to Kola Peninsula, a region associated with high human activities. However, the local emissions from this area are not expected to influence the large investigation area homogeneously. Since the authors do not take the air mass transports in to consideration in the analyses of the investigation area it seems unfair to compare the result obtained only with the satellite retrievals with the two other approaches. Although the authors mention air updraft velocity as a possible factor for affecting the variability in ACI between the three different approaches, considering also that the present cloud in-situ observations have been performed at a high-altitude station (Sammaltunturi). This means that higher updraft is expected compared to the surrounding area, which indeed is not representative for the large area the satellite aerosol and cloud retrievals have been analyzed for.

ANSWER We did study where we restricted the area to  $65^\circ$  N to  $70^\circ$  N and  $20^\circ$  E and  $26^\circ$  E. This did not have nothing meaningful effect to results except that the statistics got worse. We certainly agree that air updraft velocities for cloud measured at our high-altitude site are not necessary representative of those over a larger area around Pallas. The second paragraph of section 3.2 was rewritten to discuss these issues. Aerosol chemical composition and air updraft velocities are discussed briefly as well (section 3 prior to 3.1).

3) Considering the requirements for the satellite cloud retrievals the authors should also remove a) multilayered clouds b) too thin clouds ( $< 100 \text{ m}$ ), since those cannot be accurately retrieved from space c) ice clouds.

ANSWER OK, done

Page 27467, lines 20-21; the heights of the two stations should be given.

ANSWER OK

Page 27469, line 20; “sets 1-4” should be “sets 1-3”

ANSWER No, it should be sets 1-4, Cloud cases, The First PaCE, The Second PaCE and 2007 Data

Page 27469, line 25; Collection 4 or 5?

ANSWER Collection 5, added

Page 27469, lines 24-25; The spatial pixel resolution of MODIS AOT should be given. I suppose it is 10 km. Collection 4 or 5?

ANSWER Collection 5 and 10 km , added,

Page 27473, line 4; should be “: : ...(Fig. 1a).”

ANSWER In the revised version, Figures are referred to in a correct way.

Page 27473, lines 3 -10: Probably also due to large variability in LWP. Keep in mind that COT is more sensitive to dynamic effects, which means that the effective radius should be more reliable to estimating ACI, compared to COT, when the LWP is not known. The author write “This demonstrate that when aerosol and cloud properties are not exactly co-located, determination of aerosol-cloud interactions becomes challenging.” The in-situ aerosol and cloud observations are as well not exactly co-located. The question arise, could higher updraft velocity at the in-situ cloud site, which then is probably not representative for a larger area (see comments 2 above), explain some of the deviations in the results of ACI, obtained with the approaches 1 and 2? and of course when method 3 is also taken in to consideration.

ANSWER In the revised manuscript, remote sensing data was recalculated using a

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very tight constraints of the LWP (see comments to the two other reviewers). The difference between the approaches 1 and 2 remained, so variability in the LWP was not a reason for this difference.

Although our aerosol and cloud measurements are not exactly co-located, we have >previously shown that aerosol measurements at the two sites in the Pallas area are about equal. We certainly agree that air updraft velocities for cloud measured at our high-altitude site are not necessary representative of those over a larger area around Pallas. The second paragraph of section 3.2 was rewritten to discuss these issues.

Page 27474, lin4; estimated correlation coefficients according to a linear fit or another one? Additionally, the coefficient of determination ( $R^2$ ) should be estimated instead of correlation coefficient ( $R$ ), then consistent with the referred results and a correct statistical treatment of the problem. For the same reason also change Figures 1b and 2b so that  $R^2$  is presented instead of  $R$ .

ANSWER Correlation coefficient between  $\ln r_e$  and  $\ln N_i$ . Correlation coefficient is now presented as  $R^2$ .

Page 27474, lines 5-18; Emphasize that the high altitude in-situ station leads to updraft velocity that is probably high and not representative for the surrounding area (see also comment 2 above).

ANSWER In section 3.2, we have added a general discussion on how different type of averaging related to remote-sensing data might influence our results.

Page 27476, lines 12-14; should be “One influencing factor, probably also in the present study, is that aerosol and cloud properties are often not measured at the same place and time: : : : : :”

ANSWER Corrected.

Technical comments Page 27481, Figures 1d and 2d; y-labels are missing.

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ANSWER There was not room for labels, they same as 1c and 2c (now 1c and 3c).  
Explanation was added to figure caption

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/9/C12407/2010/acpd-9-C12407-2010-supplement.pdf>

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 27465, 2009.

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