Influence of convection and aerosol pollution on ice cloud particle effective radius

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Received: 28 August 2010 – Published in Atmos. Chem. Phys. Discuss.: 7 October 2010
Revised: 30 December 2010 – Accepted: 10 January 2011 – Published: 17 January 2011

Abstract. Satellite observations show that ice cloud effective radius ($r_e$) increases with ice water content (IWC) but decreases with aerosol optical thickness (AOT). Using least-squares fitting to the observed data, we obtain an analytical formula to describe the variations of $r_e$ with IWC and AOT for several regions with distinct characteristics of $r_e$-IWC-AOT relationships. As IWC directly relates to convective strength and AOT represents aerosol loading, our empirical formula provides a means to quantify the relative roles of dynamics and aerosols in controlling $r_e$ in different geographical regions, and to establish a framework for parameterization of aerosol effects on $r_e$ in climate models.

1 Introduction

It is well known that aerosols can cause changes in cloud number concentration and therefore alter cloud particle size, which can result in changes of cloud lifetime, reflectance, and precipitation (e.g. Twomey, 1977; Albrecht, 1989; Lohmann and Feichter, 2005; Penner et al., 2006). To date, quantification of the indirect effects of aerosols on climate remains a challenging problem. One difficulty is to separate dynamic effects, for example, the convective updraft strength, from aerosol microphysical effects. Inadequate understanding of the relationship between microphysical and dynamical processes contributes to large uncertainties in model simulations of aerosol effects on clouds, precipitation and climate.

Recent studies (e.g. Jiang et al., 2008, 2009; Su et al., 2011) using collocated Aura satellite’s Microwave Limb Sounder (MLS) and Aqua satellite’s Moderate-resolution Imaging Spectroradiometer (MODIS) observations showed reduction of upper tropospheric (UT) ice cloud effective radius ($r_e$) in polluted clouds, defined by collocated carbon monoxide (CO) measurements higher than a certain threshold value. The enhanced CO values are positively correlated with aerosol optical thickness (AOT) for a number of geographical regions and seasons. Jiang et al. (2008, 2009) also showed that $r_e$ generally increases with convective intensity indicated by ice water content (IWC), as stronger convection produces more condensates and larger cloud particles. For different regions and seasons, the variations of $r_e$ with IWC and CO (AOT) are different. Characterizing the regional differences of dynamic and aerosol effects quantitatively is needed for models to accurately simulate aerosol-cloud interactions.

Traditionally, the ice cloud particle size distribution is parameterized as a function of IWC following a gamma distribution (e.g. Matrosov et al., 1994; Evans and Stephens, 1995) or a bimodal distribution (e.g. Mitchell et al., 1996; Platt, 1997; McFarquhar and Heymsfield, 1997), neither of which takes into account the effect of aerosol on $r_e$ (i.e. the first indirect effect of aerosols on clouds). In this paper, we use least squares fitting to the observations and derive an analytical formula to describe the $r_e$ variations with convection and aerosol intensities in different geographical regions and seasons. It expands the previous $r_e$ – IWC relation to include the dependency of $r_e$ on AOT as well. This formula intends to provide a framework to quantify the relative effects of dynamics and aerosols on $r_e$, and to parameterize the first indirect effects of aerosols for use in climate models.
2 Data and approach

For this study, the MODIS $r_e$ and AOT measurements are from Aqua MODIS MYD08-D3 and MYD04-L2 data sets, respectively (Platnick et al., 2003; Remer et al., 2005). The MLS IWC is from MLS Version 2.2 Level 2 data (Wu et al., 2008). The MODIS and MLS measurements are collocated by averaging the MODIS data onto the MLS footprints (Jiang et al., 2009). Although the MODIS AOT data are usually missing in the cloudy regions, the MODIS measurements have much higher horizontal resolution ($\sim 10 \text{ km} \times 10 \text{ km}$) than the MLS measurements ($\sim 300 \text{ km}$ along track and $\sim 7 \text{ km}$ cross track), thus the AOT data are available in about half of the data in which MLS detects cloud (i.e when IWC > 0).

The MODIS $r_e$ measurement pertains to the top of cirrus clouds. Platnick (2000) studied weighting or probability distribution function (PDF) of the penetration distribution in optical depth units from water cloud-top. The same methods in the Platnick (2000) have been used for ice clouds by Zhang et al. (2010), which estimated the MODIS $r_e$ retrieval is mostly sensitive to $\sim 0.1$ to 0.2 optical depth from cloud top (approximate $< 1$ km, depending on how dense the clouds are). Different from MODIS, MLS retrieves height-resolved ice clouds above 215 hPa. MLS IWC measurements are vertically distributed at the pressure levels between 215 and 68 hPa, with a 3–4 km vertical resolution. Given that only the cloud top $r_e$ is available from MODIS, we limit the analysis to those $r_e$ measurements when IWC at 215 hPa is detectable by MLS, where clouds are likely associated with deep convection. We examine the cloud top $r_e$ relationship as a function of the column aerosol loading (indicted by AOT) and convective strength (indicated by MLS IWC at 215 hPa) under the premise that the influence of lower level aerosols on clouds is propagated to deep convective cloud tops (e.g. Sherwood, 2002). For parameterization of $r_e$ in climate models, it would be desirable to describe $r_e$ as a function of aerosol concentration and cloud water content (CWC) at each vertical height. However, due to the limitation of the data, the current study aims to provide a heuristic framework for future height-resolved analysis. The bulk relationships obtained here are insightful for understanding of the dynamics and aerosol effects on convective cloud particle sizes.

We also use the outgoing longwave radiation (OLR) data from GEOS-5 (Goddard Earth Observation Systems, Version 5) analysis to validate that MLS IWC at 215 hPa is a good index for convection. The GEOS-5 analyses are “snapshots” of the atmospheric state produced four times daily (at 00:00 Z, 06:00 Z, 12:00 Z, and 18:00 Z) using optimal combinations of model forecasts and many observations (Rienecker et al., 2008) via the Gridpoint Statistical Interpolation (GSI) technique of Wu et al. (2002). For this study, the GEOS-5 OLR data, which have a $0.5^\circ \times 0.67^\circ$ latitude-longitude resolution, are sampled onto the MLS footprints (Jiang et al., 2010) to match the observations.

The level of convective detrainment is approximately $\sim 200 \text{ hPa}$ (e.g. Kitzmiller et al., 2004; Devasthale and Fueglistaler, 2010), and the amount of ice water at this level is proportional to convective intensity. MLS IWC has been found to be indicative of convective strength. Liu (2007) showed in detail that MLS detected ice clouds are geographically and seasonally correlated with deep convection as inferred from the TRMM brightness temperature $< 210 \text{ K}$. He used MLS IWC at 100 hPa and 147 hPa in particular. Our analysis (Jiang et al., 2007, 2010; Su et al., 2006) showed 215 hPa IWC correlates with convective intensity. Figure 1 shows the relationship between the MLS IWC measurements at 215 hPa and the GEOS-5 OLR for the tropical region (30$^\circ$S to 30$^\circ$N). Consistent with previous studies, the 215 hPa IWC and the OLR have good correlations on instantaneous measurements and grid means, and thus can be used as an index for convective strength, approximately.

The reason we use IWC, not OLR, in the empirical fitting (described later in Sect. 4) follows conventional parameterization for $r_e$, which is directly related to the mass density of ice particles. Previous work such as McFarquhar and Heymsfield (1997), explicitly employed the dependence of $r_e$ on IWC, which inspired us to expand that relationship to include aerosol dependence. Using IWC, rather than OLR, to fit $r_e$ also helps to extend our empirical fitting framework from cloud-top $r_e$ to height-resolved $r_e$ in the future, when both vertically resolved $r_e$ and IWC are available.

![Fig. 1](https://example.com/fig1.png)
3 Observed relationships

We define the convective index (CONV) as
\[ \text{CONV}_i = \frac{\text{IWC}_i}{\text{IWC}} \tag{1} \]
where IWC$_i$ represents each single MLS 215 hPa IWC measurement and IWC is the mean of all the 215 hPa IWC samples in the region.

Next, we examine the relationships among the MLS CONV index and MODIS measurements of ice cloud $r_c$ and AOT from August 2004 to July 2008 between 50° S and 50° N. Figure 2a shows the MODIS $r_c$ binned according to the CONV index. The thick black curve is the average $r_c$ vs. CONV, and it includes all values of AOT. It can be seen that when CONV is relatively small, $r_c$ increases approximately linearly with IWC. When CONV is greater than $\sim 1$, the increase of $r_c$ with CONV becomes slower or saturated as the maximum $r_c$ may be limited by the settling velocities of the ice particles. The influence of aerosol on $r_c$ is illustrated by grouping the $r_c$ data for polluted clouds with AOT $> 0.4$ (thin black curve) and for clean clouds having AOT $< 0.05$ (blue curve). The $r_c$ for the polluted clouds is about 2–4 µm smaller than the $r_c$ for the clean clouds. Note that the clean cloud $r_c$ increases faster with CONV than the polluted cloud $r_c$.

Figure 2b shows the relation of $r_c$ to AOT. For $0 < \text{AOT} < 0.1$, $r_c$ quickly increases with AOT. When AOT $> 0.1$, $r_c$ decreases at a rate of about 0.5 µm per 0.1 increase of AOT until AOT $\sim 0.5$, then the $r_c$ decreasing rate becomes slower and stays approximately constant.

The dependence of $r_c$ on CONV and AOT is further illustrated by the Bi-Variate Composite (BVC) plot of the $r_c$-CONV-AOT relation (Fig. 2c), in which the MODIS $r_c$ data are binned according both to the MLS CONV and the MODIS AOT values. About 150,000 collocated MODIS and MLS measurements from August 2004 to July 2008 between 50° S to 50° N are used for the plot. The dotted lines show the occurrence frequency for each CONV-AOT pair. The BVC figure shows that $r_c$ increases with CONV but decreases with AOT; the largest $r_c$ resides where the convection (CONV) is largest and pollution (AOT) is lowest, i.e. strong convection in a clean environment. At constant CONV, the $r_c$ decreases with increasing AOT; suggesting an influence of aerosol loading on the cloud particle size. At constant AOT, $r_c$ increases with CONV until CONV approaches $\sim 1$. At the large CONV values (CONV $\gg 1$), $r_c$ becomes approximately constant when AOT is $\ll 0.3$, but decreases with IWC in high polluted cases (AOT $> 0.3$).

4 Empirical fitting

In order to obtain a simple mathematical expression for the above observed $r_c$-CONV-AOT relation, we assume that the dependence of $r_c$ on CONV and AOT is decoupled, i.e., $r_c = r_{aot} \cdot r_{conv}$, where $r_{conv}$ denotes how cloud particle effective radius changes with CONV and $r_{aot}$ represents the effect of aerosols on $r_c$. This assumption is chosen for its simplicity to capture the observed $r_c$-CONV-AOT relations, although we recognize that AOT and CONV are in fact not independent. We write
\[ r_{conv} = [1 - \exp(-\text{CONV}/\alpha)] \cdot \exp(-\beta \text{CONV}), \tag{2} \]
where $\alpha$ and $\beta$ are the parameters that control how fast $r_c$ changes with CONV. The reduction of $r_c$ by aerosol effect is assumed to follow a power-law:
\[ r_{aot} = \varepsilon \cdot \text{AOT}^\eta, \tag{3} \]
where $\eta$ is a parameter that determines how strong the aerosol effect is, and $\varepsilon$ is a scaling constant. A superposition of Eqs. (2) and (3) yields
\[ r_c = \varepsilon \cdot \text{AOT}^\eta \cdot [1 - \exp(-\text{CONV}/\alpha)] \cdot \exp(-\beta \text{CONV}). \tag{4} \]

We then perform two dimensional (2-D) least-squares fitting to the observed data with the empirical formula (4), i.e., to determine the parameters $\alpha$, $\beta$, $\eta$, and $\varepsilon$, through minimizing the following cost function
\[ \text{cost} = \sum_{i,j} \left[ r_{c,i,j} - r_{c}(\text{CONV}_i, \text{AOT}_j, \alpha, \beta, \eta, \varepsilon) \right]^2 \tag{5} \]
where $r_{c,i,j}$ (in µm) is the mean $r_c$ corresponding to the $i$-th CONV-bin and $j$-th AOT-bin. The CONV$_i$ is the mean CONV in the $i$-th CONV-bin and AOT$_j$ is the mean AOT in the $j$-th AOT-bin. Figure 2c shows an example of such $r_c$, CONV, and AOT relationship to be fitted. For our least-squares fitting computation, there are 24 bins of AOT between 0.0 and 1, and the CONV are computed using Eq. (1) from 24 bins of IWC between 0.01 and 100 mg m$^{-3}$.

The least-square fittings are computed by using four years of MLS IWC and MODIS $r_c$ and AOT measurements. The results of the fitted parameters for global (LON: 0°–360°; LAT: 50° S–50° N) and five different regions are given in Table 1. The choice of the five regions is based on the previous study of Jiang et al. (2009), where ice cloud particle sizes in these regions were shown to correlate with aerosol loadings. These regions are: South America (LON: 270°–340°; LAT: 40° S–10° N); Southern Africa (LON: 0°–55°; LAT: 35° S–0°); Northern Africa (LON: -10°–45°; LAT: 0° S–25° N); South Asia (LON: 70°–120°; LAT: 10° S–15° N); and East Asia (LON: 90°–180°; LAT: 20°–45° N).

Substituting the fitted parameters into Eq. (4), we obtain $r_c$ (µm) as functions of CONV and AOT for all of the above mentioned geographical regions. These empirical model results are plotted in Fig. 3 along with the observations. The $\chi^2$/DOF (degrees of freedom) listed in Table 1 illustrates the convergence of the fitting for different regions. The closer is the value of ($\chi^2$/DOF) to 1 (or less), the better is the fitting. We found that although all fittings converge after $\sim 20$...
iterations, the best fittings are those for the Global, South America, Southern and Northern African regions. The empirical models for South and East Asia do not fit with the observations as well as the other four regions. The regional differences are discussed below.

5 Analyses of regional characteristics

5.1 Qualitative differences: the BVC plots

We display $r_e$ as a function of CONV and AOT in Fig. 3 for a number of regions using both observations and the empirical fittings. Comparing with the observations (Fig. 3 left-column), the empirical models (Fig. 3 right-column) approximately capture the broad features of observed $r_e$-CONV-AOT relationships. For example, $r_e$ in general, increases with CONV but decreases with AOT, so that the largest $r_e$ occurs when convection (CONV) is strong and pollution (AOT) is low.

At the global scale (Fig. 3a, b), increasing aerosol pollution tends to reduce cloud particle size, while increasing convection tends to enlarge cloud particle size. Strong convection, however, also increases the distributions of aerosols into the UT. Thus, for the same columnar AOT, the UT ice clouds may be more affected by aerosols in regions of stronger CONV, indicated by a steeper gradient of $r_e$ with increasing AOT, than in regions of weaker CONV.

For South America (Fig. 3c, d), the observed $r_e$ increases with increasing convective strength, but varies less monotonically with AOT than in the global case. Large $r_e$ is broadly distributed in low AOT regions, but the largest $r_e$ is not collocated with the cleanest air. One reason may be that cloud particle size can increase with AOT in regions where moisture is abundant and cloud particles continue growing with increasing number concentration of cloud condensation nuclei (e.g. Ackerman et al., 2004). However, such a non-monotonic behavior is not captured by the empirical fitting, because the AOT dependence is chosen to be a monotonically decreasing function. In regions where CONV is larger than 1, the observed increase of $r_e$ with CONV is not as sharp as in the global case, while the empirical model appears to produce larger $r_e$ than the observation. Southern Africa (Fig. 3e, f) and Northern Africa (Fig. 3g, h) regions are very similar to each other in that the maximum $r_e$ is much smaller than the global case, resulting in weak $r_e$ gradients for all values of CONV and AOT. It appears that $r_e$ decreases with AOT more rapidly in Southern Africa than in Northern Africa.

South Asia (Fig. 3i, j) is very different from the rest of the geographical regions. The reduction of $r_e$ with AOT is more pronounced both at the low ($<\sim 0.8$) and high ($>\sim 2$) ends of

Table 1. Values of the empirical fitting parameters in Eq. (4) for various regions. The least-square fits are computed using IWC (mg m$^{-3}$) measured by MLS, and $r_e$ (µm) and AOT measured by MODIS from August 2004 to July 2008. The convergence of each fit is indicated by $\chi^2$/DOF.

<table>
<thead>
<tr>
<th>Region</th>
<th>$\alpha$</th>
<th>$\beta$</th>
<th>$\eta$</th>
<th>$\epsilon$</th>
<th>$\chi^2$/DOF</th>
</tr>
</thead>
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<tr>
<td>Global</td>
<td>0.133</td>
<td>0.00543</td>
<td>-0.0661</td>
<td>24.039</td>
<td>1.7</td>
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<tr>
<td>South America</td>
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<td>0.00308</td>
<td>-0.0632</td>
<td>23.795</td>
<td>1.9</td>
</tr>
<tr>
<td>Southern Africa</td>
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<td>0.00716</td>
<td>-0.0740</td>
<td>22.684</td>
<td>1.8</td>
</tr>
<tr>
<td>Northern Africa</td>
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<td>0.00678</td>
<td>-0.0589</td>
<td>23.703</td>
<td>1.7</td>
</tr>
<tr>
<td>South Asia</td>
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<td>0.0258</td>
<td>-0.0838</td>
<td>25.919</td>
<td>4.6</td>
</tr>
<tr>
<td>East Asia</td>
<td>0.144</td>
<td>0.0172</td>
<td>-0.0426</td>
<td>24.709</td>
<td>7.2</td>
</tr>
</tbody>
</table>

Fig. 2. (a): MODIS measured ice cloud $r_e$ binned according to CONV, based on MLS IWC at 215 hPa. The thick black curve is the average $r_e$ as a function of CONV using all values of AOT. The blue curve is for data with AOT < 0.05 and the thin black curve is for data in heavily polluted environment with AOT > 0.4. (b): MODIS $r_e$ binned according to the MODIS AOT measurements. (c): A Bi-Variate Composite (BVC) plot of the $r_e$-CONV-AOT relation, in which the MODIS $r_e$ data are binned according to both MLS IWC and MODIS AOT. The dotted-lines are the occurrence frequency for each CONV-AOT bin. All colocated MODIS and MLS measurements from August 2004 to July 2008 between 50° S to 50° N are used for the plot. The error bars in (a) and (b) denote the standard errors ($\sigma$/\sqrt{N}) of the bin average.
the CONV values. In the medium CONV values between 0.8 and 2, the reduction of \( r_e \) with AOT is slower than in the low and high CONV ranges, resulting in ice clouds having large (>27 µm) \( r_e \), extending into the high AOT (>0.4) range. This dependence of aerosol effects on convective regimes may indicate complicated coupling between aerosol forcing and meteorological conditions.

East Asia (Figs. 3k, l) has very irregular \( r_e \) changes with CONV and AOT. First, it seems to share a similar feature to South Asia in the medium CONV range, where the reduction of \( r_e \) with AOT is slow. Second, \( r_e \) is observed to rapidly increase with AOT in regions where AOT is less than ∼0.2 to 0.3 (Fig. 3k). Increases of both ice and liquid cloud \( r_e \) with AOT in the Southeast Asian regions have been previously reported (e.g. Chylek et al., 2006; Yuan et al., 2008). The first feature is simulated by the empirical model (Fig. 3l), while the second feature is not captured, which is the main reason for the large \( \chi^2/\text{DOF} \) value and poor fitting in this region.

5.2 Quantitative differences: each term in the empirical model

The empirical model has several different terms (see Eq. 4). The first term, \([1 - \exp(-\alpha/\text{CONV}^\mu)]\), represents the growth of \( r_e \) with respect to CONV (Fig. 4a): When there is no convection (CONV → 0), this term approaches 0. As CONV increases, it grows quickly. For CONV > 1, this term approaches the maximum of 1. This increasing effect of convection on \( r_e \) is the strongest in Northern Africa and South Asia, indicated by the largest values of \( \alpha \), around 0.23–0.26. The global case yields the smallest \( \alpha \), 0.13.

The second term, \([1/\exp(\beta \text{CONV})]\), is formulated to model the decrease of \( r_e \) with CONV (Fig. 4b), especially at large CONV values. This decreasing effect of CONV on \( r_e \) is the strongest in South and East Asia, corresponding to the largest \( \beta \). Figure 4c shows the combination of these two terms, \( \text{CONV}_{\text{conv}} = [1 - \exp(-\alpha/\text{CONV}^\mu)]/\exp(\beta \text{CONV}) \), which captures the initial growth of \( r_{\text{conv}} \) with CONV, reaching a maximum around CONV = 1, and decreasing \( r_{\text{conv}} \) when CONV further increases. For CONV > 1, the decrease of \( r_{\text{conv}} \) is most noticeable in South and East Asia. Possible causes for the reduction in \( r_{\text{conv}} \) with increasing CONV include: (1) Increased convection distributes more aerosols to the high altitudes in the UT thus affecting ice cloud formation there. For the same columnar AOT, stronger convection may result in increased aerosol effects (reduction of \( r_e \)) on ice clouds; (2) The cloud top is higher in stronger convection thus the MODIS \( r_e \) is measured at higher altitude with lower cloud top temperature. It is known that ice cloud formed in colder temperature has smaller particle size (e.g. McFarquhar and Heymsfield, 1997).

The third term, \( r_{\text{aot}} = \text{AOT}^\eta \), represents the modulation of AOT on \( r_e \) (Fig. 4d): increasing aerosol loading increases cloud number density and reduces \( r_e \). The differences among the curves for South America, Southern African and Northern Africa are relatively small, suggesting similar aerosol effects in these three regions. The slope of this term is the largest in South Asia and smallest in East Asia as characterized by the largest and smallest values of \((-\eta)\), respectively. The steep slope for South Asia indicates a strong aerosol influence, while the flatness of the curve for East Asia suggests that increasing aerosols in this region have a relatively small impact on the cloud particle size. On the other hand, the larger value of \((-\eta)\), for the same AOT, \( r_{\text{aot}} = \text{AOT}^\eta \) yields larger \( r_e \). In this case, \( r_e \) is larger for South Asia than East Asia for the same AOT, which may be due to more moisture in the South Asian monsoon region. South America
We thank the NASA ACMAP and IDS programs for support. The work was conducted at the Jet Propulsion Laboratory, California Institute of Technology, under contract with NASA.

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Edited by: B. N. Duncan
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