The Influence of Mixing on Evaluation of the Aerosol First Indirect Effect

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Abstract. The aerosol first indirect effect is known to cool the Earth radiatively. However, its magnitude is very uncertain. One of the difficulties in deriving this effect is caused by the coherent variation between aerosol abundance and meteorological conditions. In this study, we demonstrate that evaluation of the aerosol first indirect effect based on comparisons of clouds with different aerosol concentrations suffers an influence of the different degrees of mixing between clean and polluted clouds. By introducing a new method capable to remove this influence, we show that the strength of the aerosol first indirect effect is about half of that estimated by many previous investigators.

1. Introduction

Anthropogenic aerosols enhance cloud reflectance of solar radiation by increasing the number concentration of cloud droplets that form on aerosols. This process is referred to as the aerosol first indirect effect. There are two competing effects with opposite signs of the aerosol indirect radiative forcing. Twomey effect hypothesizes that an increase in aerosol concentration results in a large number of smaller cloud droplets, leading to a higher cloud albedo [Twomey, 1974]. The droplet dispersion compensating effect as proposed by Liu and Daum [2002] suggests that an increase in aerosol concentration broadens the spectrum of cloud droplets, which increases cloud effective radius and lowers cloud albedo. Although there is much evidence suggesting that the aerosol first indirect effect causes net cooling, its magnitude has been regarded as one of the greatest uncertainties in climate forcing. A currently accepted consensus for this effect ranges from 0 to -2 Wm$^{-2}$ as published by the Intergovernmental Panel on Climate Change report.
Recently, some studies indicate that the upper limit of this forcing may have been overestimated. For example, using global temperature records as a constraint, the estimate of this effect by Knutti et al. [2002] ranges between 0 to -1.2 Wm$^{-2}$. Similar result was obtained from model simulations using satellite observations as a constraint [Lohmann and Lesins, 2002]. However, large-scale observations have not been able to show consistent magnitude of this effect [Haywood and Boucher, 2000]. Discrepancies of more than a factor of 2 have been reported among various observational results [Feingold, 2003; Rosenfeld and Feingold, 2003]. The aforementioned droplet dispersion compensating effect is not big enough [Rotstayn and Liu, 2000] to explain the discrepancies alone. Narrowing this large range of uncertainty is pivotal in understanding how human activities influence the Earth's climate.

By definition, the aerosol first indirect effect is measured by the response of cloud effective radius $r_e$ to the change in aerosol number concentration $N_a$ for a given cloud, or, $-\partial \ln r_e / \partial \ln N_a$, where $\partial$ signifies that the difference is due to aerosol variation alone. If the change of aerosol number concentration doesn’t alter cloud liquid water content, it leads to $r_e \propto N_c^{-1/3}$, where $N_c$ is the cloud droplet number concentration. Therefore, $(\partial \ln N_c / \partial \ln N_a)/3$ equals to $-\partial \ln r_e / \partial \ln N_a$ [Twomey, 1977]. However, because it is impossible to have otherwise identical clouds under different environment of aerosol number concentrations, to compute the above derivatives from observational data, the differentials of the cloud and aerosol variables are actually calculated based on measurements of a group of clouds over different aerosol abundances. Currently, there are two major approaches to quantify the aerosol first indirect effect. One is to measure
the change of cloud droplet number concentrations due to the change of aerosol number concentrations [Twomey, 1977], i.e.,

\[
IE_1 = \frac{1}{3} \frac{\Delta \ln N_c}{\Delta \ln N_a},
\]

where \(\Delta\) means taking the difference of variables between clean and polluted clouds. The other is to measure the change in cloud radiative properties, such as the cloud effective radius or cloud optical depth, with respect to the change in aerosol number concentrations at constant liquid water path \(W\) [Feingold, et al., 2001], i.e.,

\[
IE_2 = \left( -\frac{\Delta \ln r_e}{\Delta \ln N_a} \right)_W.
\]

The first approach is often used in aircraft in situ measurements, while the second approach is more currently used in satellite observations where only vertically integrated quantities can be derived.

Since \(r_e \propto N_c^{-1/3}\) [Twomey, 1977], it has been thought that \(IE_1\) and \(IE_2\) represent the same measure of the aerosol first indirect effect, and should be equal. However, the results we compiled from recent observational studies, as summarized in Table 1, show that the observed magnitudes of \(IE_1\) are not in agreement with those of \(IE_2\). In fact, the values of \(IE_2\) are systematically lower (about half on average) than the values of \(IE_1\). To address the seemingly inconsistent results, some investigators [Rosenfeld and Feingold, 2003] attributed the discrepancies to the variation of the number of activated nuclei among different types of clouds, and some [Feingold, 2003] to the differences among the aerosol’s properties themselves. However, both explain the diversity of \(IE_1\) or \(IE_2\) rather than the systematic discrepancy between \(IE_1\) and \(IE_2\). In this study, we argue that the
systematic discrepancy between $IE_1$ and $IE_2$ is caused primarily by the differential loss of cloud liquid water between clean and polluted clouds as explained below.

2. Methodology

In nature, clouds are likely subjected to mixing with their ambient drier air and become sub-adiabatic. The degree of mixing may depend on meteorological conditions, and could differ among the group of clouds selected to compute $IE_1$ or $IE_2$. Let $N_c$ and $N'_c$ denote the cloud droplet number concentrations in an actual cloud and in an idealized adiabatic cloud, respectively. $N_c$ is always less than $N'_c$ under the same atmospheric condition because of evaporation of cloud droplets in the actual cloud. Assuming $N_c / N'_c = \beta$, we can breakdown $\Delta \ln N_c / \Delta \ln N_a$ into two parts,

$$\frac{1}{3} \Delta \ln N_c = \frac{1}{3} \Delta \ln N'_c + \frac{1}{3} \Delta \ln \beta. \quad (3)$$

Referring (1), the left side of (3) is $IE_1$. The first term on the right side of (3) indicates the change of cloud droplet number in response to the change of aerosol number concentration without the evaporation-induced reduction of cloud droplets. The second term on the right side of (3), however, is a term arising from the correlation between $\beta$ and $N_a$ within the group of clouds used for analysis. If $N_a$ and $\beta$ vary randomly or $\beta$ is uniform within the group of clouds, the averaged value of the second term should be zero. Otherwise, if $N_a$ and $\beta$ vary in a coherent fashion within this group of clouds, a nonzero value will be resulted for this term, which can be negative or positive depending on the spatial distributions of $\beta$ and $N_a$. Since the aerosol first indirect effect is meant to measure the change of cloud particle size due to aerosol variation without altering cloud liquid
water content [Twomey, 1977], it should be estimated within the clouds that have the same $\beta$. In other words, the aerosol first indirect effect proposed by Twomey [1974] should not be measured by $IE_1$, but by $\frac{1}{3} \Delta \ln N_c'/\Delta \ln N_a$. Therefore, based on (3), $IE_1$ overestimates the Twomey effect by a factor of

$$\frac{\Delta \ln N_c}{\Delta \ln N_a}/\frac{\Delta \ln N_c'}{\Delta \ln N_a} = \left(1 - \frac{\Delta \ln \beta}{\Delta \ln N_c}ight)^{-1}. \quad (4)$$

It has been observed, especially in stratus/stratocumulus clouds, that the mixing process tends to decrease cloud droplet number concentration, but not decrease the mean volume radius [Blyth and Latham, 1991; Brenguier, et al., 2000; Burnet and Brenguier, 2006]. This process is referred to as inhomogeneous mixing. During inhomogeneous mixing, the decrease of liquid water content ($L$) results solely from the reduction in cloud droplet number concentration ($N_c$) [Baker, et al., 1980], which gives rise to $N_c/N'_c \approx L/L' < 1$ [Blyth and Latham, 1991; Burnet and Brenguier, 2006]. Here the symbols with and without prime denote quantities in adiabatic and sub-adiabatic clouds, respectively. The relationship among $r_e$, $L$, and $N_c$ is given by [Brenguier et al., 2000; Martin et al., 1994; McFarquhar and Heymsfield, 2001; Pawlowska and Brenguier, 2000; Pontikis and Hicks, 1992; Wyser, 1998]

$$r_e \propto L^{1/3} (kN'_c)^{-1/3}, \quad (5)$$

where $k$ is defined by the cubic ratio of the volume radius to the effective radius [Liu and Daum, 2000; Martin, et al., 1994]. Based on the assumption of $N_c/N'_c \propto L/L'$ and the adiabatic relation between cloud liquid water content and cloud depth ($H$), i.e., $L' \propto H$ [Brenguier et al., 2000], we deduce the following relation from (5)

$$r_e \propto H^{1/3} (kN''_c)^{-1/3}. \quad (6)$$
From (5) and (6), we have

\[ \frac{1}{3} \frac{\Delta \ln N_c}{\Delta \ln N_a} = \frac{\Delta \ln A}{\Delta \ln N_a} - \frac{1}{3} \frac{\Delta \ln k}{\Delta \ln N_a}, \quad (7) \]

and

\[ \frac{1}{3} \frac{\Delta \ln N'_c}{\Delta \ln N_a} = \frac{\Delta \ln B}{\Delta \ln N_a} - \frac{1}{3} \frac{\Delta \ln k}{\Delta \ln N_a}, \quad (8) \]

where \( A = L^{1/3} r_e^{-1} \) and \( B = H^{1/3} r_e^{-1} \). Since \( k \) depicts the dispersion of cloud drop spectrum [Liu and Daum, 2000; Martin, et al., 1994], the last term in (7) or (8) is related to the droplet dispersion compensating effect [Liu and Daum, 2002]. Recalling that \( \frac{1}{3} \Delta \ln N'_c/\Delta \ln N_a \) represents the true Twomey effect (e.g., reducing cloud drop size solely due to the increased drop number concentration), we propose that the real strength of the aerosol first indirect effect (referred to as \( IE \) hereafter) should be the sum of the Twomey cooling effect and the compensating effect. Therefore, from (8) or (6), \( IE \) can be evaluated by

\[ IE = \frac{\Delta \ln B}{\Delta \ln N_a} \quad \text{or} \quad - \left( \frac{\Delta \ln r_e}{\Delta \ln N'_a} \right)_H. \quad (9) \]

Also, from (7) \( IE_1 \) can be evaluated by

\[ IE_1 = \frac{\Delta \ln A}{\Delta \ln N_a} \left/ \left( 1 + \frac{\Delta \ln k}{\Delta \ln N'_c} \right) \right. \right. \]

\[ \quad \text{(10)} \]

It should be noted that we assumed \( N_c \) being constant with height in deriving the above equations for simplicity, which may not well hold for all clouds because mixing could be stronger in upper part of the clouds. However, this simplification is acceptable because albedo is mainly determined by the top.
3. Data Analysis and Results

We evaluate $\Delta \ln A / \Delta \ln N_a$ and $\Delta \ln B / \Delta \ln N_a$ using data observed over the Northeast Pacific near the California coast (24°-36°N and 114°-150°W) during summer (June to August) of 2000. The dataset contains 3-monthly mean values of $W$, $r_e$, $H$ and $\tau_a$ (aerosol optical depth) in every 1° by 1° grid derived from data of the Tropical Rainfall Measurement Mission satellite, National Centers for Environmental Prediction / National Center for Atmospheric Research reanalysis, and the Moderate Resolution Imaging Spectroradiometer orbital aerosol optical depth product. This is the same dataset as used in Shao and Liu [2005] to study the $H - \tau_a$ coherent pattern problem, except that datum points with $WH^{2}$ less than 210 g m$^{-2}$ km$^{-2}$ are excluded here to further ensure observations being from uniform stratus/stratocumulus clouds. Considering liquid water content generally increases linearly with height for non-precipitating marine stratus [Brenguier et al., 2000; Martin et al., 1994], we calculate $L$ using $2WH^{1}$. Figure 1 shows the scatterplots of $A$ and $B$ versus $\tau_a$ on a logarithmic scale for this dataset. Because $\tau_a$ is generally proportional to $N_a$ [Nakajima et al., 2001], the slopes of the scatterplots are the values of the terms $\Delta \ln A / \Delta \ln N_a$ and $\Delta \ln B / \Delta \ln N_a$, respectively. From this figure, $\Delta \ln B / \Delta \ln N_a$ is ~0.1 and $\Delta \ln A / \Delta \ln N_a$ is ~0.26, which results in $\frac{1}{3}\Delta \ln \beta / \Delta \ln N_a \approx 0.16$.

To evaluate $IE_1$ from our observational data, $\Delta \ln k / \Delta \ln N_c$ is first estimated using observational data by previous investigators. Figure 2 shows the observed $k - N_c$ relation reproduced from Figure 1 of Liu and Daum [2002], which contains 37 observations from various investigators. The slope of the best fitting line indicates that the mean value of $\Delta \ln k / \Delta \ln N_c$ equals to -0.21, which results in $IE_1 \approx 0.33$ according to (10). Combining $\frac{1}{3}\Delta \ln \beta / \Delta \ln N_a \approx 0.16$ and $IE_1 \approx 0.33$, we have $\Delta \ln \beta / \Delta \ln N_c \approx 0.48$ for our dataset. It is
interesting to see that the value of 0.33 is comparable to the values of $IE_1$ listed in Table 1 and $\Delta \ln \beta / \Delta \ln N_c \approx 0.48$ translates to $IE_1$ being approximately twice larger than the Twomey effect according to (4). To demonstrate that $\Delta \ln \beta / \Delta \ln N_c \approx 0.5$ is more common than our studied case, Figure 3 shows the measurements of the relation between $L/H$ and $N_c$ based on data in Table 1 of Miles et al. [2000], in which they compiled observations of cloud properties by 15 investigators at various regions. In this plot, clouds with base higher than 750 m and with droplet number concentration larger than 360 cm$^{-3}$ are excluded for the following considerations: the interaction between aerosol and cloud properties may change its behavior when cloud base height is greater than 700-800 m [Durkee et al., 2000], and the aerosol first indirect effect may be saturated when aerosol loading becomes too heavy [Han et al., 2000]. As mentioned before, for stratocumulus clouds $L' \propto H$ and $N_c / N'_c \propto L / L'$, so that $\beta \propto L / H$. Therefore, the slope of the scatterplot in Figure 3 describes the dependence of $\beta$ on $N_c$. In spite of the points being scattered, the positive correlation between $\beta$ and $N_c$ is statistically significant. The mean value of the slope (0.51) agrees well with our result of $\Delta \ln \beta / \Delta \ln N_c \approx 0.48$. Since $\beta$ is the fraction of drop number concentration (or liquid water content) of the actual cloud with respect to the undiluted (adiabatic) cloud, the nonzero slope of the best fitting line (i.e., $\Delta \ln \beta / \Delta \ln N_c$) reflects the uneven degree of loss of cloud water for clouds with different drop number concentrations. The positive slope means that clean clouds fortuitously lose more liquid water than pollute clouds. The cause for this phenomenon is a very interesting topic, but it is beyond the scope of this study.

Using the same Northeast Pacific dataset, we also calculated $IE_2$ as defined in (2). Its probability distribution function is shown in Figure 4. The mean value of $IE_2$ for this
dataset is 0.086, which is comparable to the values given by previous investigators as listed in Table 1 and has the same order of magnitude as $\Delta \ln B/\Delta \ln N_a$ (i.e., $IE$) as well. This suggests that on average $IE_2$ yields similar meaningful results to $\Delta \ln B/\Delta \ln N_a$ to represent the aerosol first indirect effect for stratus/stratocumulus clouds although at a given $W$ bin the $IE_2$ value could be very different from the overall average.

4. Conclusions

Mixing has large influence on the evaluated magnitude of the aerosol first indirect effect. It has been found that clouds in clean area far from the coast tend to have higher tops and be less uniform [Bretherton and Wyant, 1997; Kaufman et al., 2005; Shao and Liu, 2005; Taylor and McHaffie, 1994], therefore, possibly are deviated more from adiabatic process than clouds in more polluted area near the coast [Brenguier et al., 2000; Taylor and McHaffie, 1994]. As a result, $\Delta \ln \beta/\Delta \ln N_a > 0$ and $IE_1 > IE$. Previous estimations of the aerosol indirect effect based on clouds with different aerosol concentrations has been biased because those clouds have different degrees of mixing, which corroborates the estimations of the mixing impact based on large eddy fine resolution simulation of clouds with full 3D radiative transfer [Chosson et al., 2006]. It is noted that the term of $\frac{1}{3} \Delta \ln \beta/\Delta \ln N_a$ may have had the same order of magnitude of $IE$, so that $IE_1$ is about twice of $IE$. The parameter of $IE_1$ used by many investigators is not suitable to quantify the aerosol first indirect effect because it includes an artifact. A physically more meaningful parameter is introduced in this study, using which the aerosol first indirect effect is assessed over the region of Northeast Pacific. It shows that the order of the aerosol first indirect effect measured by the new parameter is about half
of that estimated by many previous investigators. A final note is that $N_c/N'_c \propto L/L'$ within a group of clouds is used in deriving our equations related to the first indirect effect. This assumption, therefore, the strength of the estimated mixing artifact, is better suited to stratocumulus than to cumulus clouds [Burnet and Brenguier, 2006].

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References


### Table 1. Summary of the observed aerosol first indirect effects from various studies

<table>
<thead>
<tr>
<th>Source</th>
<th>$IE_1$</th>
<th>$IE_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chuang <em>et al.</em> 2000 (Fig. 9)</td>
<td>0.16</td>
<td></td>
</tr>
<tr>
<td>Feingold <em>et al.</em>, 2001 (Table 2)</td>
<td>0.18</td>
<td></td>
</tr>
<tr>
<td>Twohy <em>et al.</em> 2005 (Fig. 3)</td>
<td>0.18</td>
<td></td>
</tr>
<tr>
<td>Kaufman <em>et al.</em> 1991 (Eq. 14)</td>
<td>0.23</td>
<td></td>
</tr>
<tr>
<td>Conant <em>et al.</em> 2004 (Table 2)</td>
<td>0.28</td>
<td></td>
</tr>
<tr>
<td>Taylor <em>et al.</em> 1994 (Table 1)</td>
<td>0.29</td>
<td></td>
</tr>
<tr>
<td>Sekiguchi <em>et al.</em> 2003 (Table 1)(^a)</td>
<td>0.073</td>
<td></td>
</tr>
<tr>
<td>Nakajima <em>et al.</em> 2001 (Fig. 4)</td>
<td>0.080</td>
<td></td>
</tr>
<tr>
<td>Bréon <em>et al.</em> 2002</td>
<td>0.085</td>
<td></td>
</tr>
<tr>
<td>Feingold <em>et al.</em> 2003 (Table 2)</td>
<td>0.10</td>
<td></td>
</tr>
<tr>
<td>Kim <em>et al.</em> 2003</td>
<td>0.13</td>
<td></td>
</tr>
</tbody>
</table>

\(^a\) Average of over-ocean results for the global correlation slopes (0.100, 0.0689, 0.0498).
Figure Captions:

Figure 1. The dependence of parameters (a) A and (b) B on $\tau_a$. Red lines are the best fitting and blue curves represent the 95% confidence range. Shown in the legends are the slopes and the corresponding 95% confidence intervals.

Figure 2. The relation between $k$ and $N_c$ (cm$^{-3}$). Red line is the best fitting and blue curves represent the 95% confidence range. Shown in the legend are the slope and its corresponding 95% confidence interval.

Figure 3. The relation between $L/H$ (g m$^{-3}$ km$^{-1}$) and $N_c$ (cm$^{-3}$). Red line is the best fitting and blue curves represent the 95% confidence range. Shown in the legend are the slope and its corresponding 95% confidence interval.

Figure 4. The probability distribution function of $(\Delta \ln r_e/\Delta \ln r_a)_W$. Shown in the legend are the mean value and the 95% confidence interval.
The diagrams show the relationship between two variables, $A$ and $B$, with respect to $\tau_a$. The top diagram (a) shows the relationship between $A$ (in units of $g^{1/3} m^{-1} \mu m^{-1}$) and $\tau_a$, with a slope of $0.256 \pm 0.047$. The bottom diagram (b) shows the relationship between $B$ (in units of $km^{1/3} \mu m^{-1}$) and $\tau_a$, with a slope of $0.095 \pm 0.036$. The data points are represented by circles, with trend lines shown in blue and red, indicating the linear relationship.
The slope of the regression line is $-0.21 \pm 0.12$. The equation for the regression line is:

$$\ln(k) = \ln(N_c) \times \text{Slope} + \text{ Intercept}$$
\[ \ln\left(\frac{L}{H}\right) = \ln(N_c) \times 0.51 \pm 0.36 \]
Mean: $-0.086 \pm 0.015$