

## **Scholarly Paper for Master Degree of Meteorology**

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Since Twomey first proposed that cloud particle size is reduced by aerosols, there have been plentiful of evidences to support this so-called “Twomey effect”. Using NASA’s MODIS products, our study discovered an anti-Twomey phenomenon and a more general dependence of cloud particle size on aerosol. Cloud particle size may increase or decrease with aerosol depending on cloud regime and atmospheric conditions. For convective clouds developed over moist regions, the anti-Twomey effect prevails, while Twomey effect dominates for stratiform clouds over water limited regions. The dependence of particle size on aerosol varies over a large range with precipitable water. Precipitable water accounts for more than 70% of the variance. The anti-Twomey effect is attributed to droplet collision growth, while the Twomey effect is due to diffusion growth. Consideration of both effects in GCMs may significantly lower the estimation of global aerosol cooling effect and thus to agree better with observations. It may also resolve a controversial concerning if pollution suppress or enhance precipitation and thus improve our understanding of precipitation processes.

The general aerosol indirect effect (AIE) refers to any influence of aerosols on cloud microphysics, cloud duration, precipitation, etc.. Among the various types of AIE reported so far (Twomey 1977, Albrecht 1989, Ackerman et al., 2000?, Rosenfeld 2000, Ilan and Kaufman 2004, Andrea and Rosenfeld 2004), reduction in cloud particle size by aerosol (the Twomey effect) is most widely accepted with ample supporting evidence (Twomey et al. 1977, King et al. 1995, Feingold ? 2003, Kim et al. 2003, Penner et al. 2004). It is usually referred to the first type of AIE (AIE-I) or generally known as the “Twomey” effect. While the sign of the Twomey effect has never been disputed, its magnitude is very uncertain. More than a factor of 3 difference has been reported on the sensitivity of cloud microphysics to aerosol (Rosenfeld and Feingold 2003, Kim et al. 2003). Some of the differences may be artifacts resulting from the use of different methods and/or observation data. For example, the sensitivity retrieved from AVHRR (Nakajima et al. 2001) is systematically higher than that from POLDER (Breon et al., 2002). On the other hand, certain physical truth may be behind the scene, as a variable sensitivity was also found using the same data over the same region (Feingold et al., 2003). Unfortunately, we have little knowledge on the causes driving the change in AIE (Rosenfeld and Feingold 2003).

A lack of fundamental understanding has rendered a large range of uncertainties in model simulations of the global AIE (-1.1 to -4.4 Wm<sup>-2</sup>) (Rotstajn 1999, Ghan et al. 2001, Jones et al. 2001, Lohmann and Feichter 2001). In general, it is believed that the AIE has been overestimated (Rotstajn and Liu 2003). Using the global temperature records as a constraint, the AIE was estimated to lie between 0 to -1.2 Wm<sup>-2</sup>. While the overestimation could originate from inadequate treatment of different types of AIE (Nakajima et al. 2001, Han 2002, Coakley and Walsh), the study of Lohmann and Lesins (2002) pinpointed that a GCM overestimate the rate of decrease in cloud particle size with aerosol optical depth (AOT) relative to that obtained from satellite observations. So far, only one sound explanation was proposed by Liu and Daun (2002). They argued that aerosol could have a “warming effect” by increasing the relative dispersion of the cloud droplet spectrum. Adoption of this effect in a GCM helps reduces the magnitude of the AIE-I by 12 to 35% (Rotstajn and Liu 2003).

This study puts forth a new physical mechanism that could explain several paradoxes. Contrary to the Twomey effect, plentiful of evidence is presented here that cloud particle size can increase with AOD as well. We shall simply refer to it as an “anti-Twomey effect”. In general, we found that AIE-I varies from positive, negative and neutral with a large range of magnitude. Note the majority of previous studies were concentrated on stratus or stratocumulus clouds over relatively dry regions/seasons under stable atmospheric conditions that are unfavorable for moisture transport. By contrast, our study is focused on cumulus clouds in humid regions/seasons. The two opposite AIE-I effects exhibited under the two distinct atmospheric conditions and cloud regimes implies that the AIE-I effect must be understood in a broader context of atmospheric dynamics, thermodynamics and moisture conditions.

### **Approach**

In essence, AIE is a simplified measure of the relationship between an input (aerosol) and output (clouds or precipitation) of a complex system in which interactions among aerosol, cloud, dynamics and thermodynamics ultimately govern the relationship. As cloud and precipitation are realizations of various interaction/feedbacks taking place inside the system, isolation of the AIE is a very challenging but essential task to understand climate forcing, feedbacks and response (Hansen ?). Previous findings concerning the correlations between aerosols and clouds/precipitation were based either on a large ensemble of observations that effectively filter out the transitional atmospheric dynamic effects (Han et al. 1998, Wetzell and Stowe 1999, Nakajima et al. 2001, Liu et al. 2003); or on significant events for which aerosol effects exceed other factors, such as ship tracks, fire smoke and heavy air-pollution (Coakley et al. 2000, Kaufman and Fraser 1997, Rosenfeld 2000). Under ordinary conditions, the AIE is usually veiled by a large variation in dynamic and thermodynamic conditions. To isolate and quantify the AIE, other influential factors should be taken into consideration (Schwartz et al. 2002).

To this end, our working hypothesis is that AIE is contingent upon atmospheric dynamics which may be roughly delineated by cloud types; and upon the atmospheric environment for which water supply is a key factor. Stratus and stratocumulus clouds have weak vertical motion that may cut-off or weaken the transport of water vapor from the lower atmosphere. Such clouds therefore were often the targets for demonstrating the Twomey effect. Convective clouds developed in the summer over moist region have almost opposite dynamical and thermodynamic settings. The two converse regimes can thus provide good anchors to allow an examination of the full spectrum of AIE. An ideal scenario is shown in Fig. 1 around the Gulf coastal region in May 2003. The region is dominated by convective clouds in summer produced by the sea breeze that supplies a wealth of water vapor from the Gulf. Surface solar heating generates sufficient convection to form cumulus clouds over land, but no clouds over ocean. We have browsed a large quantity of MODIS images and found that the cloud and atmospheric conditions as shown in Fig. 1 are common which are often accompanied by the Bermuda High pressure system that blows moist air into the inland US over the Gulf region. So, our study was initiated over this region but expanded to much of inland US and a few other places around the world.

### **Data**

The study is based on analyses of the MODIS products of cloud, aerosol and water vapor. MODIS provides numerous cloud parameters such as cloud top, phase, droplet effective radius (DER), cloud optical depth (COD), liquid water path (LWP), etc. (King and ?, Platnick ?). From COD and DER, parameters, we derived cloud droplet number concentration (DNC) using the method of Han et al. (2002). As our focus is on convective water clouds, any cirrus contamination is removed. MODIS aerosol products also include many parameters. We employ

aerosol optical depth (AOD) as a proxy of cloud condensation nuclei (CCN). MODIS precipitable water vapor (PW) data derived from the infrared channel ( $\mu\text{m}$ ) are used and compared with those from the NCEP-NCAR.

MODIS cloud parameters are given at a nominal 1km resolution, while AOD is estimated from all clear pixels inside a 10 km grid. PW data are also estimated for clear pixels (ARE THEY GIVEN AS MEAN OVER 10KM?). Cloud parameters are retrieved for each cloudy pixel. To match the three types of data sets, cloud variables are averaged for all cloudy pixels within the 10km grids. So, the effective resolution of all the data used here is 10km. It is worth noting the cloud, aerosol and PW data are derived from different channels following different methods, and are thus independent data sets.

The MODIS quickview images helped us choose study regions from the granules obtained primarily in US on all days in July 2001. The study region is usually a portion of a MODIS granule of typically  $\sim 10^4 \text{ km}^2$  that is small enough to assure similar dynamic and thermodynamic conditions, but large enough to encompass variability in cloud and aerosol. A key criterion for the scene selection is the presence of broken clouds that allow retrieval of both cloud and aerosol parameters in vicinity. In addition to regions in US, we also analyzed a few samples acquired over other parts of the world to see how general our findings are.

## Results

The most straightforward means of evaluating if aerosol has any effect on cloud particles is to plot DER against AOD, as is shown in Fig. 2. Each panel represents the results for one study region. Note the figure is just a small subset of many cases (56?) we have examined, all of which are used in the following statistical analyses. Contrary to nearly all previous findings, the majority of the cases (>90%) examined here showed a positive dependence with a large range of variation. The correlation coefficient between the two quantities is higher than many previous studies (>0.6) as marked on the plots. The slope given in each figure denotes the change in DER ( $\mu\text{m}$ ) per unit change in AOD. For the cases shown, it ranges from -7.7 to 29.8  $\mu\text{m}$ . While some studies (Felgolder et al., Kim et al. 2003) also revealed a large range of variation, they are all in the negative territory, while ours are almost all positive except for a handful of cases showing flat or slightly negative. As those studies were for large stratus clouds while ours are for convective clouds, the sharp difference in the slope implies a fundamentally different physical process governing aerosol-cloud interaction.

To reveal the underlying physical factors driving the slope, we performed a range of statistical analyses. One factor was singled out to be the most influential factor: precipitable water (PW). Figure 2 plots the slopes against PW for all  $\sim 56$  cases under study. The correlation coefficient is 0.84, or 71% ( $R^2$ ) of the variance in the slope explained by changes in PW. The few negative and nearly neutral dependence occurred for PW less than 20 mm, whereas strong positive dependence correspondence to large PW. We also tried using NCEP-NCAR PW reanalysis. The correlation is also significant (0.7) but not as high due to the coarse resolution.

We examined several other potential factors such as cloud fraction, COD, LWP, and cloud updraft velocity. Vertical wind data were obtained from the NCEP-NCAR reanalysis data (Kalnay et al. 1996?). Cloud fraction was once suspected for driving both DER and AOD, leading to a false correlation between the two. DER could be biased high for larger cloud fraction with less edging effect that reduces DER. Meanwhile, AOT could also be overestimated due to cloud contamination that increases with cloud fraction. It turns out that both variables are independent of cloud fraction (correlation coefficient <0.1?). The same is true for their

dependence on COD. LWP has positive correlation with AOD, which has been known as the second type of AIE (REF?) [?], although other studies reveal no correlation [Nakajima ....

The relationship between the slope and AOD has a weak but discernible dependence on vertical wind (Fig. 3). As convective clouds are usually driven by updraft, the weak correlation may sound peculiar, but it is not unexpected. The reasons lies in the fact that all the cases selected here correspond to similar atmospheric dynamics. As a result, vertical wind data do not have sufficient dynamic range to anchor any meaningful statistical relation. Should we include stratiform clouds in the analysis, the correlation could be high, but the distinct dynamics would contaminate the correlation. Another cause for the low correlation lies in that the reanalysis data that have very poor spatial ( $2.5^\circ$ ) and temporal (?) resolution and the vertical wind is among the most difficult and uncertain variable to get. The influence of the low resolution is indicated by replacing the MODIS PW data with the reanalysis PW. The correlation coefficient between DER and AOD is reduced to 0.5?.

All our analyses attest that the high correlation between the slope of DER-AOD and PW is a physically true rather than a statistical artifact. So, the challenge is how to explain the positive dependence. Our hypothesis is that the dependence originates from enhancement of droplet coalescence by aerosols. By analyzing many previous field measurement data, Liu and Daun (2003) concluded that an increase in aerosol loading especially from anthropogenic sources tends to increase the relative dispersion of cloud droplet spectrum. Per conventional wisdom of cloud physics (Rogers and Yau 1989), large dispersion is favorable for droplet collision and coalescence. Another key factor igniting the collision/coalescence is droplet size. The critical size for collision to occur ranges from  $15\ \mu\text{m}$  (Rosenfeld ?) to  $20\ \mu\text{m}$  (Liu ?). From Fig. 2, it is clear that all the large slope cases meet this criterion. If this theory is valid, it differs from our conventional understanding that the AIE is linked to the diffusion growth of cloud droplets.

To put our hypothesis to test, cloud droplet number concentration is plotted against AOT for the same cases as shown in Fig. 2. The results shown in Fig. 4 revealed that DNC has an opposite correlation with AOT to the DER~AOT relation. For all the cases of positive correlation between DER and AOT, CDN is negatively correlated with AOT. As CDN was derived from a combination of COD and DER which are two independent variables, the anti-correlation seems not an artifact. The strong anti-correlation is markedly displayed in Fig. 5 that plots the slope of DER~AOT against the slope of DNC~AOT. The correlation coefficient between the two slopes is  $-0.8?$ . Note that we limit the data points to those slopes that were derived with a correlation coefficient larger than 0.2. Too large uncertainty exists in the derived slope if correlation coefficient is lower than this value. If the analysis is limited to more reliable estimates of the slopes by increasing the limit of the correlation coefficient, the magnitude of the correlation between the two slopes are even higher. QUOTE NUMBERS.

To examine if the above findings are generally true for the similar conditions as examined we did a few case studies in other places. Similar correlations have been found over Korea, Japan, Hong Kong, ????. Figure 6 shows the case in Hong Kong on ?DAY. Three study regions are selected that are located from the coast to inland China. It is very interesting to note that the slope changes from a strong positive value near the coast, to weak positive value in the middle and negative over the furthest inland region. This follows closely the trend of PW change.

## Implications

The findings of this study may have many significant implications. First, estimation of radiative forcing due to the AIE-I effect is likely to decrease by including both the cooling and warming effects induced by changes in DER. [Best explain how DER can have both a warming

and cooling effect: in the introduction you only referred to papers and never explained it]. This study indicates that the warming effect could be as significant and widespread as the cooling effects in light of the global distributions of stratiform and convective clouds and moisture.

Second, the findings may have a bearing on understanding of precipitation process. As enlarging cloud droplets by collision is a dominant mechanism for producing rainfall from warm precipitation process, our findings implies that aerosol could enhance or accelerate precipitation, rather than suppressing it (Rosenfeld ?). Note findings of rain suppression were primarily over relatively dry regions. Over humid coastal regions (e.g., Houston and Tokyo), enhanced precipitation has been reported (JAPANESE STUDIES, Shepherd?), although it has been attributed mainly to the dynamic influence of the urban heat island effect. This study may offer an explanation that is complimentary to the UHI effect by taking into account changes in cloud microphysics by aerosols as well. This is especially the case for hygroscopic particles serving as CCN. In this regard, the coast region of Gulf is the most ideal location to observe maximum rainfall enhancement where the sea-salt and sulfate aerosol from the dense distribution of plants that may produce ample of hygroscopic aerosols that are lifted and activated as CCN. Seabreeze brings plentiful of water to accelerate droplet growth by both diffusion and collision.

Likewise, the findings here could also be an alternative explanation to the “south drought and north flooding” trend in China, which has been explained by the direct effect of absorbing aerosol (Menon et al. 2003?). Per the theories proposed here, the general increasing trend of AOD in the region (Luo et al. 2002?, Li 2004) would have the same effects as explained above, i.e. reducing rainfall due to the Twomey effect in North China where PW is low, and increasing rainfall due to the anti-Twomey effect in the south because of high PW. The latter is likely fueled by UHI effect induced by more widespread urbanization in well-developed southern China than less developed northern China. Often, to verify any of the above findings requires a lot more investigations.

General Comment: I don't think the SCIENCE readership can be expected to be familiar with cloud formation processes. A little more explanation would be called for.

## **Notes and references**