## Comments on "Direct Radiative Forcing of Anthropogenic Aerosols over Oceans from Satellite Observation"

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### ABSTRACT

Previous observational studies have estimated anthropogenic aerosol direct radiative forcing over oceans without due consideration of cloudy-sky aerosols. However, when interaction between clouds and aerosols located below or above the cloud level is taken into account, the aerosol direct radiative forcing is larger by as much as 5 W m<sup>-2</sup> in most mid-latitude regions in the Northern Hemisphere.

Key words: cloudy-sky aerosol forcing, anthropogenic aerosol, aerosol optical depth, satellite observations

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#### 1. Introduction

Recently, Chen et al. (2011) estimated anthropogenic aerosol direct radiative forcing (ARF), defined as the perturbation of top-of-the-atmosphere radiative fluxes by anthropogenic aerosols, for the year 2005 for all sky conditions (i.e., cloudy-sky and clear-sky) within the global ocean of 60°S-60°N. To determine global ARF from the observation data, they used the Single Scanner Footprint dataset, measured from the Clouds and the Earth's Energy System (CERES), and the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the *Terra* satellite. Like many other studies regarding aerosol-cloud-radiation relationships, they assumed that aerosols in cloudy regions do not exert radiative forces that compromise observational limitations (Bellouin et al., 2005). According to this assumption, ARF for all sky conditions (ARF<sub>all,1</sub>) can be obtained by scaling the clear-sky ARF by the fraction of clear-sky region in a given grid (1 - f):

$$\operatorname{ARF}_{\operatorname{all},1}(i,j) = \operatorname{ARF}_{\operatorname{clr}}(i,j) \times [1 - f(i,j)], \quad (1)$$

where  $\text{ARF}_{clr}$  is the averaged ARF for the clear-sky region at each latitude (*i*) and longitude (*j*). In Eq. (1), it follows that  $\text{ARF}_{all,1}$  is identical to  $\text{ARF}_{clr}$  under the clear-sky condition (i.e., f = 0) and that  $\text{ARF}_{all,1}$  is zero under the overcast condition (i.e., f=1); However, in the real atmosphere, the latter is not true.

In fact, aerosol particles interact with clouds by scattering (and/or absorption) the radiation reflected from the clouds, continually exerting appreciable radiative forces (Keil and Haywood, 2003; Chand et al., 2009). In actuality, the ARF for overcast regions is not zero at all, but rather gives the combined effects of ARF<sub>clr</sub> with modification of ARF by clouds ( $\Delta$ ARF). For this reason, Chen et al. (2011) admitted errors that resulted from the lack of consideration of ARF in cloudy regions.

It is therefore worth questioning how the value of  $ARF_{all}$  changes when cloudy-sky aerosols are included. We addressed with question by modifying Eq. (1) used in Chen et al. (2011) from a climatological perspective by using monthly averages of satellite-retrieved radiative fluxes and clouds to avoid data sampling problems. For this purpose, we employed CERES flux data (FSW, Edition 2c) and MODIS/Terra cloud data (MOD08, collection 5) with a 1°×1° resolution for the year 2005.

As noted previously, for aerosols within fractional cloud coverage f in each 1°-grid, ARF can be expressed by ARF<sub>clr</sub> plus  $\Delta$ ARF. Thus ARF<sub>all</sub> can be determined using Eq. (2):

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In Eq. (2), the estimation of  $ARF_{all,2}$  requires calculations of  $ARF_{clr}$  and  $\Delta ARF$ . To calculate  $ARF_{clr}$ , we used shortwave flux values for the pristine (i.e., without aerosol and cloud) and clear-sky (i.e., with aerosol and without cloud) conditions, which were available in the CERES/FSW data. However, the challenge was the calculation of  $\Delta ARF$ . Because  $\Delta ARF$  is not observable, we had to simulate it using a radiative transfer model. We used the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998), the flux simulations of which showed a fairly good agreement with flight observation data (Valero et al., 2003). This scheme has been popularly used in many radiation studies. The shortwave fluxes were simulated under the equivalent spectral range (i.e.,  $0.3-5.0 \mu m$ ) of the CERES spectrometer. The prescribed shortwave insolation in the model was scaled to be comparable with the observed value from the FSW data.

To obtain  $\Delta ARF$ , we first built a lookup table (LUT) to describe  $\Delta ARF$ -cloud-aerosol relations using SBDART. To generate the LUT, the data regarding the vertical distribution of anthropogenic aerosol were needed. These data were available from the level-2 vertical feature mask observed by the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), which covers the field-of-view corresponding to MODIS/CERES data. The anthropogenic aerosol levels indicate the sum of polluted continental aerosols, polluted dust, and smoke aerosols, which were classified by the CALIOP. To maintain consistency among aerosol, cloud, and shortwave flux data, we used the vertical profile for the year 2005. However, the only global aerosol profile available was from the CALIOP after April 2006. According to the annualmean vertical profiles of anthropogenic aerosol over the global ocean from the CALIOP from 2007 to 2010, interannual variability in the aerosol profile was very small. Therefore, we used the four-year (2007–2010) mean anthropogenic aerosol profile over the global ocean observed by the CALIOP. Although it is not shown as a figure, the vertical profile indicated two maximal values in the lower troposphere below 1 km (i.e., 0.4 km and 1 km). Above 1 km altitude, aerosols gradually decreased with height. The optical depth of this aerosol profile was 0.2, which corresponded to the global average.

Notably, this aerosol vertical profile was used only to generate the LUT. The LUT was then matched with MODIS-observed grid-mean bulk properties of aerosols and clouds to determine  $\Delta ARF$  in each grid. In this process, therefore, regional characteristics of  $\Delta ARF$  were captured using regionally different aerosol and cloud properties from MODIS observations, even though a fixed vertical distribution was employed.

We then allowed various feasible single-layer overcast cloud amounts with spherical droplets and aerosol conditions such as cloud optical depth (0-20), cloud effective radius  $(0-30 \ \mu m)$ , surface temperature minus cloud top temperature (0-60 K)<sup>a</sup>, aerosol single scattering albedo (ASSA; 0.87–0.99), and aerosol asymmetry factor (AAF; 0.52–0.76). Notably, the relative altitude of cloud to aerosol was indicated by cloud-top temperature under the fixed aerosol vertical profile. In this study, a difference smaller than  $\sim 10$  K indicated a case of aerosol above the cloud layer. A few exceptional cases that were not included in the LUT were also taken into account by interpolation and/or extrapolation of  $\Delta ARF$  via a polynomial regression function between the values of  $\Delta ARF$  and cloud (or aerosol) properties in the LUT.

Reliable estimation of  $\Delta ARF$  should be based on regionally prevailing aerosol and cloud properties. We used MODIS observations to obtain these regional properties, and all the properties were averaged monthly before their use for matchup with the LUT. To confine our focus to "anthropogenic" aerosol optical depth (AOD<sub>anth</sub>), we used the methodology of Kaufman et al. (2005), in which natural coarse-mode (marine and dust) AOD is subtracted from the total AOD. This AOD<sub>anth</sub> was used to scale the  $\Delta ARF$ values from the LUT based on the other properties, using a factor of the ratio of  $[1-\exp(AOD_{anth})]$  to  $[1-\exp(AOD_{anth})]$  $\exp(AOD_{LUT})$ ] (Zhao et al., 2011). Next, ASSA and AAF were obtained using the methodology of Choi et al. (2009), in which a pair (ASSA, AAF) is retrieved for each grid using two layer ARF efficiencies (i.e., ARF<sub>clr</sub> per AOD<sub>anth</sub> at the top-of-atmosphere and at the surface) because ARF efficiency is a function of only two variables: ASSA and AAF. MODIS cloud properties used in this study were the grid-mean cloud fraction, cloud optical depth, cloud effective radius, and cloudtop temperature. Finally, MODIS surface temperature was defined as the maximum brightness temperature at 11  $\mu$ m over a month for each grid.

<sup>&</sup>lt;sup>a</sup>We used surface temperature minus cloud-top temperature in the LUT for comparison with MODIS thermal retrievals. Because  $\Delta$ ARF was not directly affected by thermal condition of clouds and the surface, this difference may have been inadequate as a control parameter of  $\Delta$ ARF. However, the use of cloud height or pressure had the same effect on the calculation of  $\Delta$ ARF because the relative altitude of cloud to aerosol is much more important in determining  $\Delta$ ARF than the absolute altitude of cloud.

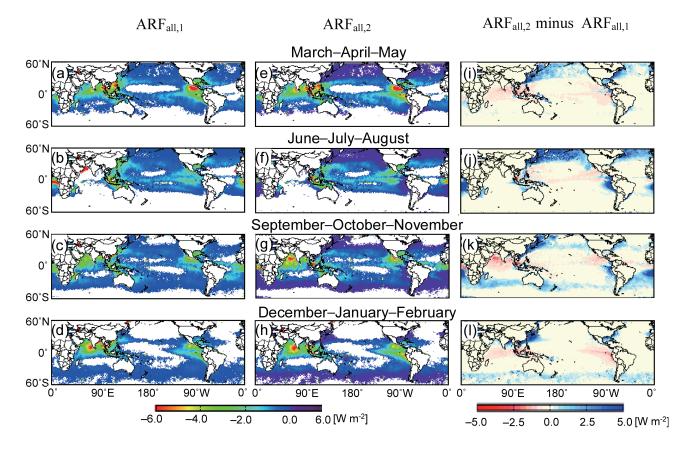


Fig. 1. Global distributions of anthropogenic aerosol radiative forcing (ARF) for all-sky by Chen et al. (2011) (ARF<sub>all,1</sub>; a–d) and the present study (ARF<sub>all,2</sub>; e–h) and their difference (ARF<sub>all,2</sub> minus ARF<sub>all,1</sub>; i–l) for four seasons of the year 2005.

# 2. Distribution of aerosol direct radiative forcings

The seasonal averages of ARF<sub>all</sub> [ARF<sub>all,1</sub> from Eq. (1) and ARF<sub>all.2</sub> from Eq. (2)] and their differences are presented in Fig. 1. The values of ARF<sub>all,1</sub> (Figs. 1a–d) mostly coincided with results from Chen et al. (2011). Relatively large cooling effects ( $\leq -3 \text{ W m}^{-2}$ ) occurred in the neighboring coastal seas of the Middle East, India, East Asia, Mexico, and Colombia. The annual average  $\pm$  standard deviation of ARF<sub>all,1</sub> was  $-0.42\pm0.40$  W m<sup>-2</sup>, which was slightly smaller than  $-0.34\pm0.16$  W m<sup>-2</sup> (reported by Chen et al., 2011). The minor underestimation of ARF<sub>all.1</sub> in this study may be explained by two experimental differences. First, we used a flux for pristine condition in each grid from the CERES/FSW, whereas Chen et al. (2011) calculated it using the linear extrapolation to zero of aerosol optical depth (AOD) with respect to solar zenith angle and latitude. Second, ARF<sub>all,1</sub> in this study was a monthly arithmetical average of instantaneous values; however, for Chen et al.

(2011) ARF<sub>all,1</sub> was the daily average<sup>b</sup>. Consequently, the methodology used in this study, though somewhat different from that of Chen et al. (2011), provided a fair comparison between ARF<sub>all,1</sub> and ARF<sub>all,2</sub>.

After the influence of cloud amount on ARF in cloudy regions was considered, the spatial patterns of ARF<sub>all,2</sub> were plotted (Figs. 1e–h). The overall patterns of a dimming effect by anthropogenic aerosols and its seasonal variation are similar to those in Figs. 1a–d. However, the cooling effect by the aerosols was fairly diminished across the whole season, which is remarkable over the mid-latitude ocean. The annual mean  $\pm$  standard deviation of ARF<sub>all,2</sub> was  $-0.12\pm0.82$  W m<sup>-2</sup>. The smaller ARF<sub>all,2</sub> compared to ARF<sub>all,1</sub> shows that cloud amount, especially an optically thick and large cloud amount, can interrupt the radiation reflected back to space by aerosols and can modify the direct radiative effect of aerosols (Chand et al., 2009).

Figures 1i–l display the difference between  $ARF_{all,2}$ and  $ARF_{all,1}$ , which indicates cloud impact on  $ARF_{all}$ . Notably, the sign of  $ARF_{all,2}$  minus  $ARF_{all,1}$  was pos-

<sup>&</sup>lt;sup>b</sup>Daily average is a 24-h mean value by applying diurnal variation of shortwave flux assuming constant aerosol optical properties.

itive over the whole ocean because of cloud influence, which blocked out the scattering by anthropogenic aerosols. The annual mean  $\pm$  standard deviation of ARF<sub>all,2</sub> minus ARF<sub>all,1</sub> was  $0.53\pm0.63$  W m<sup>-2</sup>. The difference was small ( $\leq 0.2$  W m<sup>-2</sup>) in low latitudes and large (>2 W m<sup>-2</sup>) in mid-latitudes, particularly during summer in the Northern Hemisphere. This result implies that cloud effect on ARF<sub>all,2</sub> is more significant over mid-latitudes than over low latitudes.

### 3. Discussion and summary

Chen et al. (2011) concluded that the annual average  $\pm$  standard deviation of ARF<sub>all.1</sub> was  $-0.34\pm0.16$ W m<sup>-2</sup> over the ocean between 60°S and 60°N. However, the  $ARF_{all,1}$  proposed by Chen et al. (2011) may not have provided a complete explanation of the direct effect of aerosols in all sky conditions because they overlooked the influence of anthropogenic aerosols for cloudy regions. Although this simplified assumption is commonly used in aerosol-cloud-radiation relationships, it may cause uncertainty in ARF<sub>all</sub>. Therefore, this study reproduced ARF<sub>all,1</sub> using a slightly different method from Chen et al. (2011). We additionally calculated ARF<sub>all.2</sub> using the same method, except we added a new consideration of the cloud impact on ARF for cloudy regions. The annual mean of  $ARF_{all,2}$  minus  ${\rm ARF}_{\rm all,1}$  was 0.53 $\pm$ 0.63 W m $^{-2}$  over the global ocean, indicating the influence of clouds that suppress scattering by anthropogenic aerosols. Particularly, ARF<sub>all,2</sub> minus  $ARF_{all,1}$  was >2 W m<sup>-2</sup> over the oceanic areas such as East Asia and the eastern United States due to a large cloud amount and cloud optical depth, which led to positive ARF<sub>all,2</sub> values.

There may also be a discrepancy between shortwave fluxes from RTM and from CERES. To compare RTM data with CERES-based ARF<sub>clr</sub> employed in this study, we first simulated  $ARF_{clr}$  shortwave fluxes for pristine and clear conditions with respect to each latitude and solar zenith angle. In clear conditions, the shortwave flux was determined using preestimated  $AOD_{anth}$ , ASSA, and AAF given in each  $1^{\circ} \times 1^{\circ}$  grid. The results show that the spatial pattern of RTM-based ARF<sub>clr</sub> was in good agreement with CERES-based  $ARF_{clr}$  (correlation coefficient = 0.94) overall. However, RTM-based  $ARF_{clr}$  was slightly lower, by  $0.13 \text{ W m}^{-2}$  on the average, than CERESbased  $ARF_{clr}$ , which resulted from the relatively low absorptance in the model (Valero et al., 2003). Applying RTM-based  $ARF_{clr}$  to Eqs. (1) and (2), respectively, ARFs for all sky conditions (i.e., ARF<sub>all,1</sub> and  $ARF_{all,2}$ ) were then calculated. Compared with the ARFs obtained from the CERES-based  $ARF_{clr}$ , the RTM-based ARF<sub>all,1</sub> and ARF<sub>all,2</sub> had negative biases of 0.05 W m<sup>-2</sup> (7.04%) and 0.01 W m<sup>-2</sup> (7.69%), respectively.

Our estimates of  $ARF_{all,2}$  were derived from the simplified conditions of a fixed aerosol profile and a single-layer cloud due to the insufficient observations. However, cloud effects on  $ARF_{all,2}$  increased when high-level aerosols (above clouds) increased. They decreased when high-level clouds (above aerosols) increased. When the aerosols were located between two cloud layers, the cloud effect on  $ARF_{all,2}$  was mostly subject to the upper-level clouds. Our simplification limited the accuracy of  $ARF_{all,2}$ , inducing an uncertainty of approximately  $\pm 0.1$  W m<sup>-2</sup> on the global average.

The present results reveal that there was a significant cloud contribution to ARF in cloudy regions with respect to seasons or regions in comparison to the approach from the Chen et al. (2011). Therefore, we suggest that cloud impact on  $ARF_{all}$  and interaction with aerosol should be properly regarded with caution for cloudy regions when evaluating and interpreting  $ARF_{all}$ .

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