Aerosol indirect effect over the Indian Ocean

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Received 3 December 2005; revised 10 January 2006; accepted 8 February 2006; published 21 March 2006.

[1] We analyze the MODIS (Moderate Resolution Imaging Spectroradiometer) satellite data over the seas adjacent to the Indian sub-continent to investigate the effect of aerosols on the size distribution of cloud droplets and ice crystals (indirect aerosol effect). During the winter months of increased anthropogenic pollution we observe smaller sizes of cloud droplets in water clouds in agreement with the expected aerosol indirect effect. However, contrary to our expectations, we find that during episodes of increased pollution the effective radius of ice crystals is shifted toward the larger rather than smaller sizes. We propose a combination of natural seasonal variability of meteorological conditions and an "inverse aerosol indirect effect" caused by heterogeneous ice nucleation as a possible explanation of observed ice crystal growth. The ECHAM4 (European Center for Medium-Range Weather Forecast Hamburg version 4th generation GCM) results with heterogeneous ice nucleation reproduce the observed increase in ice crystal size during the enhanced pollution episodes. **Citation:** Chylek, P., M. K. Dubey, U. Lohmann, V. Ramanathan, Y. J. Kaufman, G. Lesins, J. Hudson, G. Altmann, and S. C. Olsen (2006), Aerosol indirect effect over the Indian Ocean, *Geophys. Res. Lett.*, *33*, L06806, doi:10.1029/2005GL025397.

1. Introduction

[2] Aerosols affect the Earth's radiation budget and climate through their direct interaction with solar radiation (aerosol direct effect) [*Chylek and Coakley*, 1974; *Charlson et al.*, 1992; *Penner et al.*, 1992; *Chylek and Wong*, 1995] and indirectly through their interaction with clouds [*Twomey*, 1977; *Lohmann and Lesins*, 2002]. Aerosols affect the cloud droplet size distribution, cloud life cycle, and distribution of heating between the atmosphere and the surface. Aerosols have a complicated non-uniform physical and chemical composition and highly variable temporal and spatial distributions. Anthropogenic aerosols produced over the Indian sub-continent are of special interest due to their potential effect on the Indian monsoon, on the hydrological cycle of sub-Saharan Africa, and on the sea surface temperature of the Indian Ocean.

[3] An increase in wintertime anthropogenic aerosols over India and adjacent seas has been well documented from the ground as well as from satellite observations [*Ramanathan et al.*, 2001; *Heymsfield and McFarquhar*, 2001]. Major components of the aerosol are sulfates, nitrates, sea salt, mineral dust, organic aerosols and black carbon. The spreading of anthropogenic aerosols over the Arabian Sea and the Bay of Bengal during the winter pollution episode is shown in Figure 1. The fraction of aerosol optical depth (AOD) due to fine aerosol mode increases as well. During the year this fraction (over the Arabian Sea and the Bay of Bengal) is typically in the 50 to 60% range of the total AOD, while during the winter pollution episodes it increases to 70 to 80%.

2. Data

[4] For our study we select the rectangular geographical area between 60°E and 95°E and from 15°S to 25°N. The selected region includes the Indian subcontinent as well as the adjacent Arabian Sea, Bay of Bengal and a part of the Indian Ocean. To quantify the selected microphysical parameters we divide the region into 56 square boxes of 5° by 5° as shown in Figure 2a. The background colors in Figure 2a correspond to aerosol optical depth due to fine mode fraction averaged from September 2000 to September 2004. The fine mode fraction characterizes the anthropogenic contribution (pollution) to the total aerosol optical depth.

[5] We use the MODIS [*Kaufman et al.*, 1997; *Tanre et al.*, 1997] level 3 monthly averaged products in our analysis. We average the MODIS retrieved effective radius of cloud droplets and ice crystals over the 5 × 5 degree geographical areas (Figure 2a) and over five Septembers from 2000 to 2004 (characterizing the time of clean air) and five Januarys from 2001 to 2005 (characterizing the time of anthropogenic pollution). Although the absolute errors in individual MODIS retrievals may be significant (satellite retrieved effective radii are generally larger than those measured in situ), it is reasonable to expect that errors in observed relative changes of effective radii are small [*Nakajima et al.*, 1991].

[6] As we move along the horizontal axis of Figure 2b we are moving from the lower left corner of Figure 2a (square No. 1) in the northward direction up toward the upper left corner (square No. 8). Each vertical line in Figure 2b represents the starting point of the next south to north column in Figure 2a. Thus the points 1, 9, 17, ...49 in Figure 2b represent the most southern row of 5x5

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0094-8276/06/2005GL025397

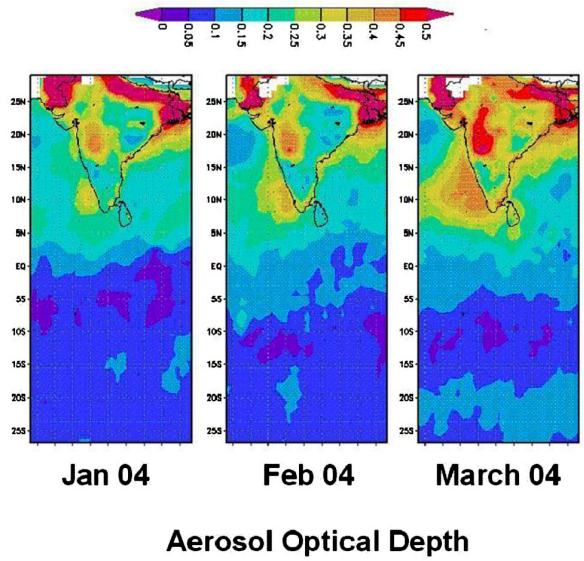

Figure 1. Aerosol optical depth for the months of January, February and March 2004. The color bar refers to the AOD.

degree squares as shown in Figure 2a (squares number 1, 9, 17, . . . 49).

3. Water Droplets

[7] The most prominent feature of the water droplet effective radii behavior (Figure 2b) is a decrease in effective radius as we move from the southern part of the region (boxes 1, 9, 17,. . ., 49) to the northern part of selected area (Figure 2). This trend persists at all longitudes during both September and January (enhanced pollution). The average MODIS retrieved effective radius is around 20 μm over the Indian Ocean compared to 12 μm over the northern parts of the Arabian Sea and Bay of Bengal. The major factor affecting the water droplet size distribution seems to be the number of available cloud condensation nuclei (CCN) which are much lower over the unpolluted Indian Ocean than the more heavily polluted Arabian Sea and Bay of Bengal (Figure 3). Over the northern Indian Ocean boundary layer CCN concentrations similar to or greater than the left-hand columns of Figure 3 (>1500/cm^3) were consistently observed on all 18 NCAR C-130 flights of the INDOEX field program (February–March 1999) [*Hudson and Yum*, 2002]. Low concentrations shown in the right-hand columns of Figure 3 (<200/cm^3) were consistently observed over the southern Indian Ocean on all three INDOEX flights that went south of the equator.

[8] Next we compare the MODIS retrieved effective radii of water cloud droplets during the months of September (clean air) and January (polluted air). We note that the absolute error in the MODIS retrieved cloud drop and ice crystal effective radii are high and yield results that are systematically higher than in situ measurements. However, our analysis focuses on relative changes in these satellite derived sizes which have much lower uncertainties [*Nakajima et al.*, 1991]. The largest differences are observed within the squares 40, 48 and 56 (Figure 2) corresponding to the land regions north of the Bay of Bengal. This is one of the most polluted regions of India. About 80% of the aerosol optical depth in these regions is due to fine mode aerosol (Figure 2). The regional air pollution supplies sufficient numbers of new cloud condensation nuclei to affect the cloud droplet size distribution significantly. The droplet effective radius in this region is decreased by up to 33% (from about 18 to 12 μm) during the winter months presumably due to air pollution produced in the north-western part of India and in Bangladesh. A decrease of droplet size with increasing pollution is also observed over the ocean with the largest reduction between September and January (from about 18 to 15 μm) observed over the northern part of the Arabian Sea.

4. Ice Crystals

[9] While water droplet radii are generally smaller in January (polluted air) than in September (clean air), the effective radii of cirrus ice crystals increased. During the clean month of September over the Arabian Sea and Bay of Bengal the ice crystal radii are in the 23 to 35 μm size range. The size range increases to about 40 to 50 μm during the polluted month of January. At the same time no increase in ice crystal size is observed over the land, over the Indian Ocean (Figure 2b) or in the tropical Pacific Ocean (not

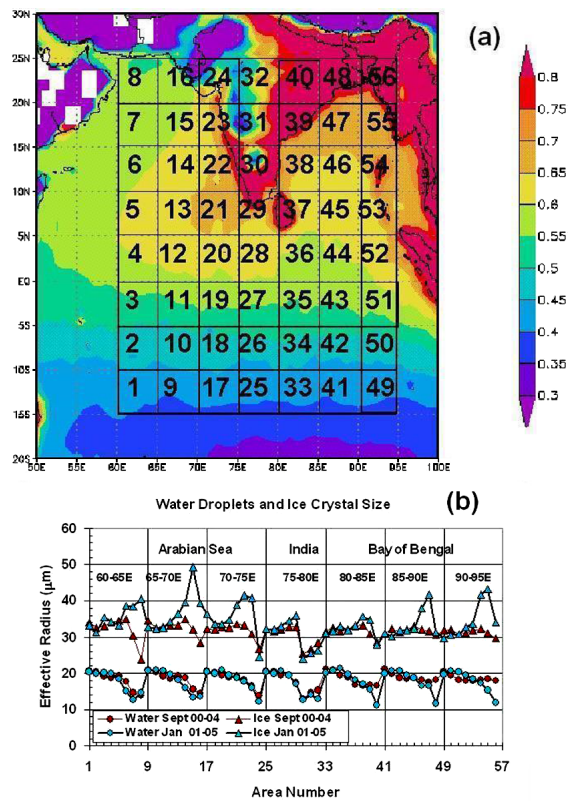

Figure 2. (a) Area selected for our study has been divided into 56 rectangular regions. The background colors show the 49 months average (Sept 2000 to Sept 2004) of the AOD fraction due to fine aerosol mode (see the color bar). The fine mode fraction characterizes an anthropogenic contribution to the total aerosol optical depth. (b) Effective radius of water droplets and ice crystals over the seas adjacent to India.

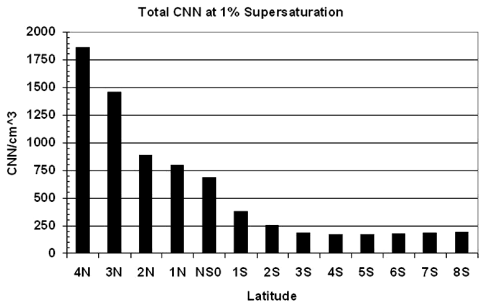

Figure 3. Cloud Condensation Nuclei concentrations over the Arabian Sea and Indian Ocean as a function of latitude (from 8°S to 4°N) as measured on March 4, 1999 during the INDOEX field program.

shown). The opposing changes in the effective radii of water droplets and ice crystals are also apparent from Figure 4.

[10] While it is relatively straightforward to understand the observed changes in the size of water cloud droplets as the first indirect aerosol effect (additional CCNs supplied by polluted air), the reason for the observed variation of ice crystal size over the Arabian Sea and Bay of Bengal cannot be clearly identified at this time. Both the seasonal variability of meteorological conditions and the effect of aerosols are expected to contribute.

[11] The larger ice crystals during the pollution episodes are counter intuitive considering the usual application of the aerosol first indirect effect (more but smaller droplets or ice crystals due to more CCN or ice nuclei). However, it has been pointed out [*Jensen and Toon*, 1997] that certain types of aerosols may lead to an inverted first indirect effect – a decrease in ice crystal numbers accompanied by growth into larger sizes. This "inversion" may occur when pollution aerosols supply heterogeneous ice nuclei (e.g., hydrophilic black carbon, mineral dust and organic aerosols) that initiate ice nucleation at supersaturations lower than those needed for homogeneous nucleation. This causes ice to form and grow on these heterogeneous ice nuclei first, thereby reducing the ambient water concentrations and supersaturation so that many of homogeneous ice nuclei are not activated, thereby decreasing the total number of ice crystal concentration.

[12] Most water-soluble aerosols (like sulfates and nitrates) present in Indian winter pollution serve as effective cloud condensation nuclei. However, they are not effective ice nucleation agents at low super-saturation. On the other hand organic aerosols and black carbon or mineral dust (once they become hydrophilic) can initiate heterogeneous ice nucleation at lower super-saturations than sulfates or nitrates.

[13] To explore the possibility of heterogeneous ice nucleation as a plausible mechanism for the observed growth of ice crystals during the months of enhanced anthropogenic pollution we have performed simulations with the ECHAM4 climate model [*Lohmann et al.*, 2004] with either homogeneous or heterogeneous ice nucleation. Homogeneous ice nucleation (freezing of sulfate aerosols) leads to smaller ice crystal sizes in January (during the polluted season) than in September (Figure 5b), which is contradicted by satellite observation (Figure 5a). Heterogeneous nucleation (immersion freezing of dust and soot in super-cooled liquid aerosol particles), on the other hand, leads to larger ice crystal sizes during the pollution episodes in qualitative agreement with MODIS satellite observations. A qualitative agreement between satellite observations and the ECHAM4 model simulations with heterogeneous ice nucleation suggests that heterogeneous ice nucleation may play an important role in polluted regions near the Indian sub-continent. The agreement could be further improved if both heterogeneous and homogeneous ice nucleation mechanisms were incorporated simultaneously in global models.

5. Summary and Conclusions

[14] We have investigated changes in water cloud droplet effective radius and cirrus cloud ice crystal effective radius over the seas adjacent to the Indian subcontinent (a) between the polluted winter season and the clean late summer and early fall seasons, and (b) between the clean Indian Ocean and the polluted northern part of the Indian Ocean (Arabian Sea and Bay of Bengal). We found that:

[15] i) The effective radii of cloud water droplets decrease as we move from the Indian Ocean northward toward the Arabian Sea and Bay of Bengal, in agreement with the expected aerosol indirect effect.

[16] ii) The effective radii of cloud water droplets over the Arabian Sea and the Bay of Bengal decrease from September (clean air) to January (polluted air). This decrease is again expected from the aerosol indirect effect, although seasonally changing meteorological conditions may also play a role.

[17] iii) The effective radii of cirrus cloud ice crystals over the Arabian Sea and the Bay of Bengal increase from September (clean air) to January (polluted air). This increase is opposite from that expected from the aerosol indirect

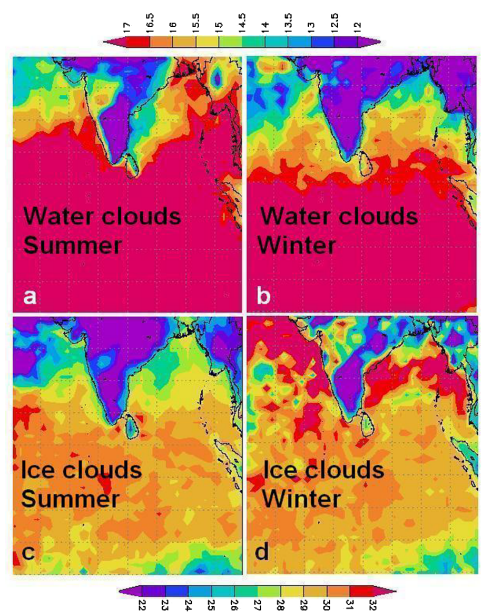

Figure 4. The average effective radius of water droplets and ice crystals during clean air summer months (June to September 2003) and polluted winter (November 2003 to March 2004). The corresponding scales are in μm.

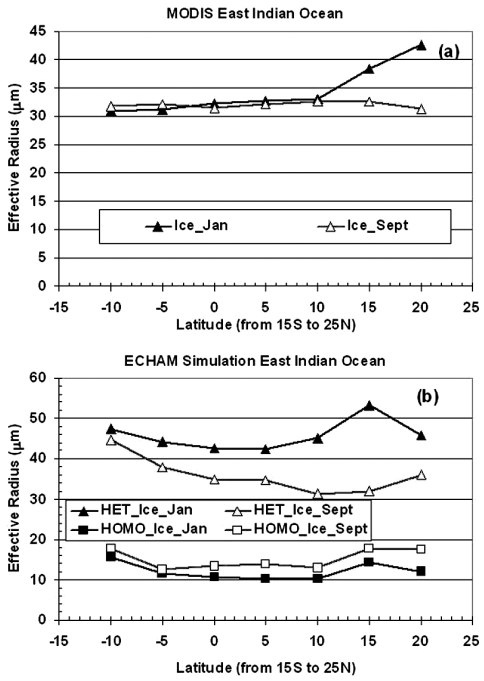

Figure 5. Ice crystal effective radii averaged over the months of September and January. Data points are plotted at the northern boundary of the 5° latitudinal bins: (a) MODIS retrieval averaged over indicated five year periods for the East Indian Ocean, and (b) results of the ECHAM4 climate model simulation with a heterogeneous and homogeneous ice nucleation.

effect. We propose that a combination of natural seasonal variability of meteorological conditions and aerosol induced heterogeneous ice nucleation is responsible for this unexpected behavior.

[18] The observed seasonal variation of water droplet and ice crystal effective radii will affect the radiative balance in the Indian subcontinent region. The effect of aerosols on water droplet size is included in the microphysics module in at least some climate models. However, the observed growth of ice crystals during the winter polluted season (either due to seasonal variability or due to aerosol induced heterogeneous ice nucleation) has not been included in any transient climate study. For the same ice water content the high ice clouds with larger and fewer ice crystals will reflect less solar radiation and let more outgoing thermal radiation escape to space.

[19] Although we are currently unable to separate the aerosol effect from the natural seasonal variation of ice crystal sizes, future field observations should be able to accomplish this task. Heterogeneous ice nucleation may also induce changes in ice crystal habits that could affect ice cloud radiative properties [*Fu et al.*, 1998; *Yang et al.*, 2005]. To determine the precise magnitude of the aerosol effect involved in the ice size and shape variations will require future in situ field measurements of ice clouds and anvils, including the size and shapes of ice crystals, measurements of black carbon and mineral dust within cloud droplets and ice particles as well as the interstitial cloud volume.

[20] **Acknowledgments.** The reported research was partially supported by Los Alamos National Laboratory's Directed Research and Development Project entitled "Resolving the Aerosol-Climate-Water Puzzle (20050014DR)" and by the IGPP "Mixed Phase Clouds" grant.

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