Reflectivity variations off the Peru Coast: Evidence for indirect effect of anthropogenic sulfate aerosols on clouds

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Abstract.
Using reflectivity measurements from the Total Ozone Mapping Spectrometer (TOMS), we show that over the months when stratocumulus clouds are prominent off the Peru Coast, the ultraviolet (UV) reflectance of two marine sites is consistently higher than that of the surroundings. The regions of reflectivity enhancement coincide with large anthropogenic sulfate aerosol emission sources, and the magnitude of the enhancement has a strong seasonal dependence that is related to the seasonal cloud movement. We propose the indirect aerosol effect as a plausible explanation for the reflectivity observations.

Introduction

Anthropogenic sulfate aerosols may impose radiative perturbations on climate by directly scattering the solar radiation (the direct effect) and by modifying the clouds (the indirect effect) [Charlson et al., 1992]. Serving as cloud condensation nuclei, sulfate aerosols may increase the number of cloud droplets while reducing the droplet size. This would increase the cloud reflectivity [Twomey, 1974; Charlson et al., 1987], and may also inhibit precipitation and increase the cloud lifetime [Albrecht, 1989]. Because of its potential for producing an important climate forcing, the indirect aerosol effect has drawn much attention, yet remains highly uncertain. Further constraining this effect is thought to be essential for predicting climate change [Charlson et al., 1992; Jones et al., 1994]. In this letter, we show some interesting reflectivity observations off the Peru Coast that may be related to the indirect effect of anthropogenic sulfate aerosols.

Data

The reflectivity data used in this study are the Nimbus-7 Total Ozone Mapping Spectrometer (TOMS) monthly reflectivity data (version 7) [McPeters et al., 1996]. The TOMS reflectivity measurements were taken in the ultraviolet (UV) at local noon. The Nimbus-7 TOMS

reflectivity is available for the years 1979-1992 in a 1.25° longitude × 1° latitude grid [McPeters et al., 1996].

Results and Discussion

Reflectivity off the Peru Coast is shown in Figure 1 for selected months. The reflectivity for each month is constructed by averaging over the years from 1979 to 1992. In February, the high reflectivity region, associated with the marine stratocumulus clouds, lies south of the Peru Coast. In May, the high reflectivity region moves north to the Peru Coast, and two reflectivity maxima become visible. Movement of the high reflectivity region is probably associated with changes in the distribution of subsidence associated with seasonal changes in the Walker circulation, the Hadley Cell or more localized circulations. In August, the high reflectivity region moves even closer to the Peru Coast, two strong reflectivity maxima are observed. In November, the high reflectivity region leaves the Peru Coast, moving south, while the two maxima are still observable. Data of intermediate months show similar results. Additional data for the years 1993-1994 and 1996-1999 from Meteor-3 TOMS and Earth Probe TOMS continue to show the described features.

The UV reflectivity is used here as a proxy for the top of atmosphere (TOA) broadband visible albedo. TOA broadband visible albedo measurements were made by the Earth Radiation Budget Experiment (ERBE) [Barkstrom, 1984], although their grid (2.5° × 2.5°) is too coarse for them to be directly used in this study. We compare in Figure 2 the monthly ERBE visible and TOMS UV measurements over the ocean areas off the Peru Coast (90°-70°W, 25°-5°S). Data from February, 1985 to January, 1989 are used. The data that we use have been modified by the National Center for Atmospheric Research (NCAR) [Hurrell and Campbell, 1992]. Both data are regridded into a 5° longitude × 5° latitude resolution for the comparison. A linear relation is found (r=0.94), with a slope close to unity (0.95±0.06). This is expected given that albedo variations of these regions are mainly due to clouds and cloud albedo is insensitive to wavelength in the short wavelength limit. There is an offset of 5.8±1.5% as the clear sky albedo of the ocean areas off the Peru Coast is higher in visible (~10%, Harrison et al. [1993], Figure 2) than in the TOMS UV measurements (~4%, Herman and Celarier [1997], Plate 1).
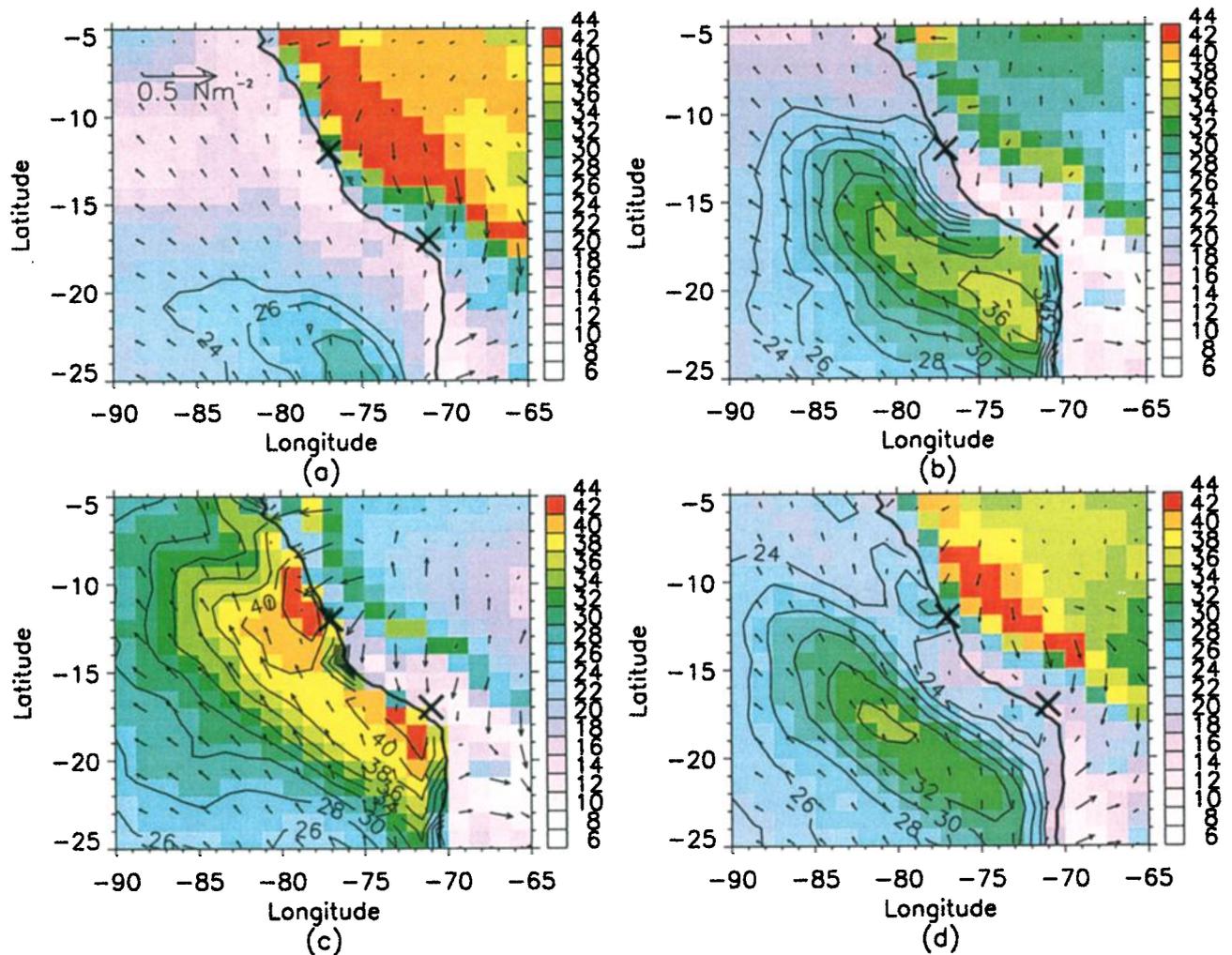
Figure 1. The climatological UV reflectivity for each month is constructed by averaging the TOMS measurements over the years 1979 to 1992. The reflectivity maps for February (a), May (b), August (c), and November (d) are shown for the Peru Coast. The coastline is over-plotted on the maps. In this work, we focus on the marine regions, i.e. the regions west of the coastline. Two major anthropogenic SO_2 emission sources, Lima (77°W, 12°S) and Ilo (71°W, 17°S) are marked by the × symbol. Contours are over-plotted on the maps from 24% to 42%, in steps of 2%, for the marine regions. The monthly surface wind stress averaged over the years 1979-1992 are overplotted for each month in arrows. The directions of the arrows have been reversed so that they are the same as the wind directions. The length of the arrow on the upper left corner of (a) represents a wind stress of 0.5 Nm^{-2}.

The consistency in locations of coastal maxima suggests that the reflectivity enhancement is related to sources fixed in geographic position. The fact that maxima are stronger when clouds are closer to the coast suggests that the enhancement is tied to the clouds.

Marked with cross symbols in Figure 1 are two large SO_2 emission sources, Lima (77°W, 12°S) and Ilo (71°W, 17°S). The SO_2 emission rate is $\sim$ 180,000 Ton/year around Lima and $\sim$ 275,000 Ton/year for Ilo, as documented by the Global Emissions Inventory Activity (GEIA) [*Benkovitz et al.*, 1996]. The large amount of SO_2 emission is related to copper smelting. Lima and Ilo stand out as two major emission sites, as other regions along the coast mostly have SO_2 emission rates less than 5000 Ton/year per 1° × 1° grid.

Large amounts of sulfate aerosols may form from the emitted SO_2 and contribute a large number of cloud condensation nuclei (CCN). Utilizing the spectral dependence of the radiance measured by the multi-channel TOMS instrument, an uncalibrated aerosol index has been derived using a residue technique [*Torres et al.*, 1998]. Negative aerosol index represents the presence of small non-absorbing sulfate aerosols that are typically of anthropogenic origin. Unfortunately, the amount of sulfate aerosol is not readily retrieved since the detection depends sensitively on the brightness of the underlying reflective surface. Over bright reflective surfaces (e.g. clouds, snow/ice), the non-absorbing aerosols become undetectable. Nevertheless, the so-derived aerosol index does show considerable negative values along the Peru Coast (not shown).

Monthly surface wind stress data, taken from the National Centers for Environmental Prediction (NCEP) / NCAR Reanalysis Project [*Kalnay et al.*, 1996], are av-
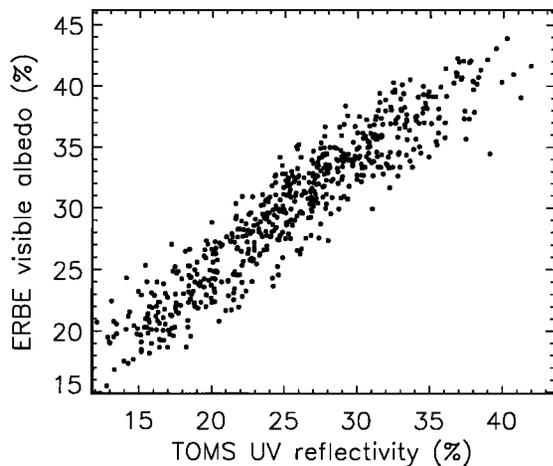
Figure 2. Comparison of the TOMS UV reflectivity and the ERBE visible albedo over the ocean areas off the Peru Coast (90°-70°W, 25°-5°S). Monthly data from February, 1985 to January, 1989 are used. Both data are regridded into a 5° longitude × 5° latitude resolution for the comparison. A linear relation is obtained (r=0.94) with A_{ERBE}=5.8(±1.5)%+0.95(±0.06)R_{TOMS} using least square fit.

eraged over the years 1979-1992 for the selected months and over-plotted in Figure 1. The directions of the arrows have been reversed so that they are the same as the wind directions. The length of the arrows represents the magnitude of the surface wind stress, which equals $C_d\rho V^2$. V is the wind speed and ρ is the air density. The drag coefficient C_d is $\sim 1.5\times10^{-3}$ over the oceans and may be several times larger over land [*Holton*, 1992]. Offshore winds of order a few m/s are generally observed along the Peru Coast. This would bring polluted continental air off the Peru Coast into the marine environment.

Clouds off the Peru Coast are mainly stratocumulus. These clouds are highly reflective and strongly affected by aerosols, both in the cloud reflectivity [*Charlson et al.*, 1987] and the cloud lifetime [*Albrecht*, 1989]. Many model studies predict a significant indirect aerosol effect off the Peru Coast [*Jones et al.*, 1994; *Boucher and Lohmann*, 1995; *Jones and Slingo*, 1997]. As seen in Figure 1, the reflectivity enhancement is strongly related to the marine stratocumulus close to the SO_2 emission sources. Stronger enhancement also coincides with stronger offshore winds that bring polluted air into the marine environment. Based on the above discussion, the indirect effect of anthropogenic sulfate aerosols appears to provide a physically plausible explanation for the reflectivity observations, although we note that without *in situ* measurements, the indirect aerosol effect remains only as a plausible explanation and no conclusive link can be drawn.

The indirect aerosol effect has been observed in, for example, "ship tracks" [*Coakley et al.*, 1987] and cloud modification by smoke particles [*Kaufman and Fraser*, 1997]. There are also indications of anthropogenic sul-

fate aerosols affecting the cloud albedo based on interhemispheric and transoceanic comparisons [*e.g. Kim and Cess, 1993*]. In these studies, the intrinsic variability of the cloud field presents the chief difficulty, as the meteorological conditions are highly variable on the cross-ocean-basin scale. Off Peru's coast, most of the aerosols are emitted from two point sources, which may serve as two strong plumes. The concentrated emission may produce large aerosol concentration gradients along the coast and allow observable indirect effect signals within relatively short distances. Therefore, the natural variations of the cloud field, chief difficulty in quantifying the indirect aerosol effect [*National Research Council*, 1996], would be considerably smaller as compared to the cross-ocean-basin scale studies [*Kim and Cess*, 1993].

If related to the indirect aerosol effect, the reflectivity observations described here suggest a stronger seasonal dependence than suggested by model studies. For instance, model studies by *Feichter et al.* [1997] found that the indirect sulfate aerosol forcing off the Peru Coast is around -4 to -5 Wm^{-2} for both January and July, with January forcing slightly more negative. Given the difference in the solar insolation off the Peru Coast (20°S-10°S) between January and July, their results imply that the effect of anthropogenic sulfate aerosols on clouds in July is about 1.5 times that in January. In Figure 1, however, we see that the reflectivity enhancement is totally absent in February when the main cloud region lies far away to the south of the SO_2 emission sources. The same is true for January. On the other hand, the reflectivity enhancement in August is on the order of 10%, corresponding to a forcing of roughly -35Wm^{-2} given a solar insolation of ~ 350 Wm^{-2} in August for the relevant latitudes. This implies a localized indirect aerosol effect that is strongly tied to the cloud field that is near the aerosol source region. Correct modeling of cloud migration would therefore be essential for estimating the indirect effect of sulfate aerosols.

Conclusion

Using TOMS reflectivity measurements off the Peru Coast, we find consistent reflectivity enhancement over regions near major aerosol emission sources when the marine stratocumulus clouds are prominent. The indirect effect of anthropogenic sulfate aerosols appears to be a plausible explanation. The observed reflectivity enhancement has a strong seasonal dependence that is related to the seasonal cloud movement. The localized reflectivity enhancement exists probably as a consequence of the point emission sources on the Peru Coast, as they may produce large sulfate aerosol concentration gradients and observable indirect aerosol effects within a short distance. If so, studies over this region would encounter less intrinsic variability of the cloud field and reduced difficulty in quantifying the indirect aerosol effect. We also note that the marine stratocumulus clouds

off the Peru Coast may play an active role in the tropical circulation [*Miller*, 1997]. Perturbations on these clouds might affect the global climate as well.

References

Albrecht, B. A., Aerosols, cloud microphysics, and fractional cloudiness, *Science 245*, 1227-1230, 1989.

Barkstrom, B. R., The Earth Radiation Budget Experiment (ERBE), *Bull. Amer. Meteor. Soc., 65*, 1170-1185, 1984.

Benkovitz, C. M., M. T. Scholtz, J. Pacyna, L. Tarrason, J. Dignon, E. C. Voldner, P. A. Spiro, J. A. Logan ,T. E. Graedel, Global gridded inventories of anthropogenic emissions of sulfur and nitrogen, *J. Geophys. Res. 101*, 29239-29253, 1996.

Boucher, O. and U. Lohmann, The sulfate-CCN-cloud albedo effect: A sensitivity study with two general circulation models, *Tellus 47B*, 281-300, 1995.

Charlson, R. J., J. E. Lovelock, M. O. Andreae, S. G. Warren, Oceanic phytoplankton, atmospheric sulfur, cloud albedo and climate, *Nature 326*, 655-661, 1987.

Charlson, R. J., S. E. Schwartz, J. M. Hales, R. D. Cess, J. A. Coakley, J. E. Hansen, D. J. Hofmann, Climate forcing by anthropogenic aerosols, *Science 255*, 423-430, 1992.

Coakley, J. A., R. L. Bernstein, P. A. Durkee, Effect of ship-track effluents on cloud reflectivity, *Science 273*, 1020-1022, 1987.

Feichter, J., U. Lohmann, I. Schult, The atmospheric sulfur cycle in ECHAM-4 and its impact on the shortwave radiation, *Clim. Dyn. 13*, 235-246, 1997.

Harrison, E. F., P. Minnis, B. R. Barkstrom and G. G. Gibson, Radiation budget at the top of the atmosphere in *Atlas of satellite observations related to global change*, edited by R. J. Gurney, J. L. Foster, and C. L. Parkinson, pp. 19-38, Cambridge University Press, New York, 1993.

Herman, J. R. and E. A. Celarier, Earth surface reflectivity climatology at 340-380 nm from TOMS data, *J. Geophys. Res., 102*, 28003-28011, 1997.

Holton, J. R., An introduction to dynamic meteorology, 3d ed., *Academic Press*, 511 pp., 1992.

Hurrell, J. W. and G. G. Campbell, Monthly Mean Global Data Sets Available in CCM History Tape Format, *NCAR Technical Note NCAR/TN-371+STR*, 94 pp., 1992.

Jones, A., D. L. Roberts, A. Slingo, A climate model study of indirect radiative forcing by anthropogenic sulfate aerosols, *Nature 370*, 450-453, 1994.

Jones, A. and A. Slingo, Climate model studies of sulphate aerosols and clouds, *Phil. Trans. R. Soc. Lond. B 352*, 221-228, 1997.

Kalnay, E., M. Kanamitsu, R. Kistler, and coauthors, The NCEP/NCAR 40-year reanalysis project, *Bull. Amer. Meteor. Soc. 77*, 437-471, 1996.

Kaufman, Y. J. and R. S. Fraser, The effect of smoke particles on clouds and climate forcing, *Science 277*, 1636-1639, 1997.

Kim, Y. and R. D. Cess, Effect of anthropogenic sulfate aerosols on low-level cloud albedo over oceans, *J. Geophys. Res. 98*, 14883-14885, 1993.

Lohmann, U. and J. Feichter, Impact of sulfate aerosols on albedo and lifetime of clouds: A sensitivity study with the ECHAM4 GCM, *J. Geophys. Res. 102*, 13685-13700, 1997.

McPeters, R. D., P. K. Bhartia and coauthors, Nimbus-7 Total Ozone Mapping Spectrometer (TOMS) Data Products User's Guide, *NASA Reference Publication*, 67pp., 1996.

Miller, R. L., Tropical thermostats and low cloud cover, *J. Climate 10*, 409-440, 1997.

National Research Council, Aerosol radiative forcing and climate change, *National Academy Press, Washington, DC* 1996.

Torres, O., P. K. Bhartia, J. R. Herman, Z. Ahmad, and J. Gleason, Derivation of aerosol properties from satellite measurements of backscattered ultraviolet radiation: Theoretical basis. *J. Geophys. Res. 103*, 17099-17110, 1998.

Twomey, S., Pollution and the planetary albedo. *Atmos. Env. 8*, 1251-1256, 1974.

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