JP1.7 EVALUATION OF THE AEROSOL INDIRECT EFFECT USING SATELLITE, CHEMICAL TRANSPORT MODEL, AND AIRCRAFT DATA DURING ICARTT

Lance Avey [1], Timothy J. Garrett*[1], Andreas Stohl[2]
[1]University of Utah, Salt Lake City, Utah
[2]Norsk instititutt for luftforskning, Kjeller, Norway

1 INTRODUCTION

Anthropogenic pollution is known to increase the albedo of shallow layer clouds by increasing concentrations of droplets and decreasing droplet radii (Twomey, 1974). Smaller droplets are less effective at producing precipitation sized drops through the collision-coalescence process, so that pollution may also lead to increased cloud lifetime and cloud cover (Albrecht, 1989). Conversely, the absorption of solar radiation by haze may reduce fractional cloudiness (Ackerman et al. 2000).

The magnitudes of these aerosol "indirect effects" have substantial uncertainties (International Panel on Climate Change (IPCC) 2001). Recently, numerous studies have used satellite data to investigate aerosol indirect effects on both regional and global scales (e.g. Nakajima et al., 2001; Sekiguchi et al. 2003, Quaas et al., 2004; Matheson et al., 2005; Kaufman et al., 2005). However, a drawback in each of these studies is that passive satellite methods are unable to measure aerosols exactly collocated with clouds, requiring instead aerosol retrievals be made in regions directly adjacent to clouds. An assumption is then made that aerosol concentrations are homogeneous throughout the cloudy and cloud-free regions (e.g. Quaas et al., 2006).

A possible concern with this approach is that because cloud properties depend foremost on meteorology, and almost by definition, the cloud and aerosol retrievals are derived from different meteorological regimes.

An alternate strategy for satellite-based assessment of the indirect effect is to use high temporal and spatial resolution output from the FLEXPART chemical tracer transport model to collocate anthropogenic pollution with low-level clouds. FLEXPART uses European Centre for Medium-Range Weather Forecasts (ECMWF) analysis and forecasts to describe the transport of particulate and gasous tracers. Here, we use anthropogenic sulfur dioxide (SO_2) and carbon monoxide (CO) tracers since their concentrations in the FLEXPART model are tied to mixing, and not on wet scavenging or oxidation (i.e. they are treated as passive tracers). The advantage here is that pollution and clouds can be treated as independent quanities.

Our procedure is to examine Aqua and Terra Moderate Resolution Imaging Spectroradiometer (MODIS) cloud properties and FLEXPART regional SO_2 and CO tracer fields. Data is collected into three 4° x 4° boxes along the line of prevailing winds downwind of the Northeast U.S. megacity for the time period of the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) 2004 summer campaign. Aircraft measurements of cloud properties and trace gases from ICARTT permit similar comparisons in-situ.

2 DATA

2.1 MODIS

The Aqua and Terra satellites offer daily morning and afternoon overpasses of the northwest Atlantic. MODIS retrieves cloud top temperature and pressure at 5 km spatial resolution and cloud top effective radius (r_e), cloud optical depth (τ_c), and liquid water path (LWP) at 1 spatial km resolution (King et al. 2003; Platnick et al. 2003).

* *Corresponding author address:* Timothy J. Garrett, 135 S 1460 E, Room 821 Salt Lake City, UT 84112-0110; e-mail: tgarrett@met.utah.edu

2.2 Chemical transport model

The Lagrangian particle dispersion model FLEXPART version 6.2 is designed for atmospheric transport modeling and analysis (Stohl et al. 2005). FLEXPART is driven by the ECMWF at a resolution of 1/4° x 1/3° over the northwest Atlantic. Recent studies of intercontinental air pollution transport have been used to validate the model (e.g. Stohl et al. 2005) showing good agreement between model-predicted pollution concentrations and measured concentrations from observational data. For the ICARTT campaign, FLEXPART predicted fields of SO_2 and CO tracers every 2 hours

2.3 Aircraft

During ICARTT, the Forward Scattering Spectrometer Probe (FSSP- 100) and Optical Array Probe (OAP-2DC) were integrated onto the NOAA WP-3D aircraft. The distributions of droplets between 2- and 1500-μm diameter were obtained from these instruments, from which number concentration (N), liquid water content (LWC), and r_e can be derived. Mixing ratios of CO and SO_2 were measured by the vacuum ultraviolet (VUV) fluorescence instrument aboard the WP-3D.

3 ANALYSIS METHOD

Fig. 1 illustrates our co-location technique. The FLEXPART output is co-located temporally with MODIS overpass times and vertically with MODIS derived cloud-top pressures. The MODIS low-level cloud properties are then gridded with FLEXPART SO_2 and CO concentrations into a 0.5° x 0.5° cells and composited into three 4° x 4° boxes located downwind of the northeast U.S. urban corridor during ICARTT. While the MODIS and FLEXPART resolution is finer than 0.5° x 0.5°, a coarser grid is chosen to compensate for sources of two possible location errors in concentrations of FLEXPART: (1) strong gradients in the pollution tracers close to the source regions and (2) advection errors, which are proportional to about 10% of the distance from the pollution source.

4 OBSERVATIONS

Comparisons between FLEXPART and MODIS were performed for 20 satellite overpasses when low-level clouds and pollution were present over the northwest Atlantic during the ICARTT period (figure 2). The data represent a range of cloud-top pressures between 800-900 hPa, chosen to constrain the relationship between cloud r_e and pressure. The CO and SO_2 bins were chosen such that each bin contains the same number of measurements: 21, 23, 27 measurements per bin for analysis boxes 1, 2, and 3 respectively.

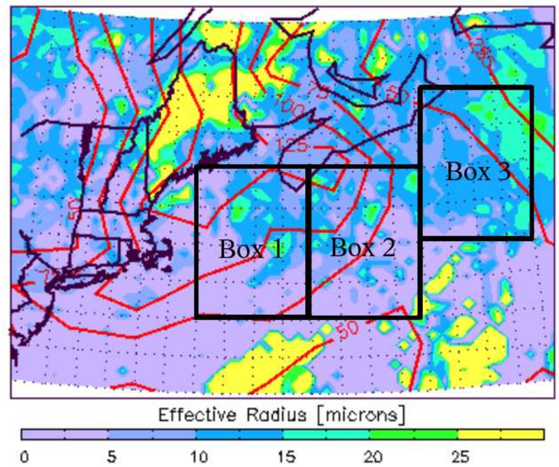

Figure 1: MODIS cloud-top effective radius and FLEXPART regional CO within analysis boxes. Red contours are CO concentrations ($\mu g/m^3$) at 1 km altitude.

The slope, obtained from linear least square fits, indicates that r_e decreases, with increasing concentrations of both CO and SO_2 in each of the analysis boxes . However, the variability, expressed in terms of standard deviation, is quite large, especially in analysis box 1. To see if the decrease in r_e from clean to polluted clouds is statistically significant, retrievals of cloud r_e are sorted to upper (polluted) and lower (non-polluted) quartiles in CO and SO_2, from which one-sided Student t-tests are performed. The Student t-tests show, to within 95% confidence, that the mean value of r_e is smaller for polluted clouds than non-polluted clouds for all analysis boxes.

The results for both CO and SO_2 compared well. In each case, analysis box 1 shows the weakest trend in r_e with increasing pollution and the highest variability, while the strongest trend occurs in analysis box 2. The high variability in analysis box 1 may be attributed to problems with col-location associated with the gradient errors discussed in section 3 since box 1 is located nearest to the sources of pollution. The greater slopes found in boxes 2 and 3 may be the result of aged pollutants becoming more efficient

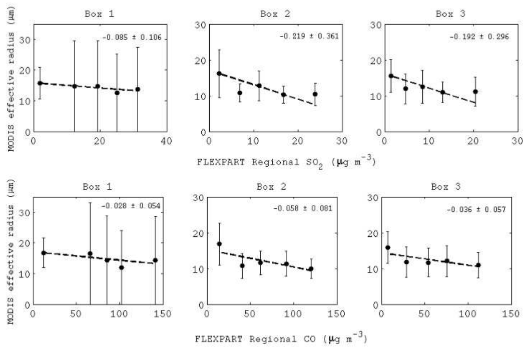

Figure 2: MODIS r_e - FLEXPART anthropogenic pollution relationships for each analysis box over the northwest Atlantic Ocean during ICARTT. Error bars show $\pm$ 1 standard deviation within each bin. Dashed line is linear fit to the bin means. Also, the mean and the estimate of their uncertainties are given for the slope of the linear fit.

per unit mass at changing cloud properties.

Results from aircraft measurements are shown in figure 3. The analysis is for LWC binned between 0.2 t0 0.3 g m^{-3} with each dot representing 10-second averages for CO. The slopes are derived from linear least square fits. Only box 1 is analyzed since no low-level cloud measurements were made in boxes 2 and 3 during ICARTT. Cloud droplet number concentration (N) increases while r_e slightly decreases with increasing CO mixing ratios, consistent with the hypothesized first indirect effect. The increase in N with CO from aircraft measurments indicates that FLEXPART anthropogenic CO is a good indicator of anthropogenic aerosols, as this is a central assumption for our study.

The slope, dr_e/dCO, is slightly smaller for the in-situ derived value (Fig. 3) than the FLEXPART derived value (Fig. 2) in analysis box 1, -0.018 $\pm$ 0.006 μm $ppbv^{-1}$ and -0.028 $\pm$ 0.054 μm μg^{-1} m^3 respectively. Note that FLEXPART gives CO in actual concentrations while the mixing ratio is given from the WP-3D data. There is no correlation between N, r_e, and SO_2 (not shown), suggesting wet removal of SO_2 during transport.

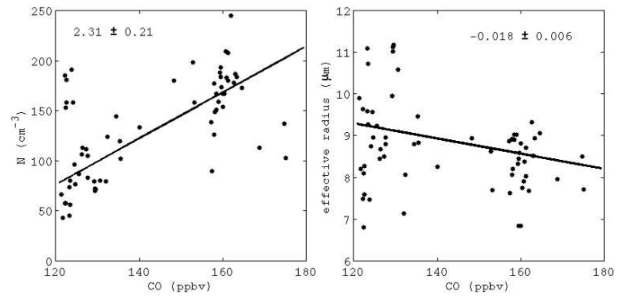

Figure 3: N - CO and r_e - CO relationships from aircraft measurements for low-level clouds in analysis box 1. Dots represent 10-second averages for CO, N, and r_e. The solid line gives the linear fit to the data. Data is binned for LWC values between 0.2 to 0.3 g m^{-3}.

5 DISCUSSION

Combined FLEXPART and MODIS data permit objective evaluation of the aerosol indirect effect. Colocation of FLEXPART anthropogenic tracers and MODIS cloud retrievals during ICARTT illustrate how pollution from the U.S. northeastern seaboard results in smaller droplets in low-level clouds downwind over the northwest Atlantic. However, the effect is weak as shown by the considerable ambient variability. Also, the relatively small sample of FLEXPART, satellite, and aircraft data could lead to misleading results that may not be seen if more samples were examined. Therefore, we are cautious to attribute our results to the aerosol indirect effect.

With this in mind, the use of the FLEXPART model and MODIS data will be extended to periods and locations beyond the ICARTT campaign to test their capability of quantifying the indirect effects. With an extended database, a more comprehensive analysis of the aerosol indirect effect (e.g. changes in τ_c and fractional cloudiness) may also be performed.

6 ACKNOWLEDGMENTS

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