

4.1 SPACE-BASED EVALUATION OF THE AEROSOL INDIRECT EFFECT IN THE ARCTIC

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1 Introduction

Arctic clouds modulate the surface radiation budget by emitting longwave radiation and reflecting incident shortwave radiation. The Arctic is characterized by high levels of anthropogenic pollution that commonly forms a haze in winter and spring, due to long-range transport of aerosols primarily from Eastern Europe and Russia (Sirois and Barrie, 1999). During the Arctic winter and spring strong stable stratification in the lower troposphere minimizes turbulence and wet deposition leading to long atmospheric lifetimes of aerosols (Law and Stohl, 2007).

Despite a long observational record of mid-latitude pollution in the Arctic, only recently have studies addressed potential indirect effects of aerosols on the surface energy budget through modification of cloud microstructures (Sirois and Barrie, 1999; Quinn et al., 2007). The primary effect appears to operate in the infrared. Polluted events correspond to clouds with higher concentrations of small droplets, higher long-wave emissivity and increased cloud insulation of the surface (Garrett et al., 2002; Garrett and Zhao, 2006; Lubin and Vogelmann, 2006).

Space-borne measurements make it possible to study cloud optical properties with the coin-

cident presence of aerosols, however spatially and temporally coincident cloud and aerosol information cannot be retrieved from the same satellite instrument. A commonly employed strategy pairs aerosol retrievals with clouds in adjacent airmasses. The assumption is that aerosol concentrations are horizontally uniform (Nakajima et al., 2001; Sekiguchi et al., 2003). Though this approach may work well in convective situations, it would be less suitable for studying stratiform clouds. Due to their large and uniform spatial coverage, horizontally and vertically adjacent masses of clear and cloudy air are more likely derived from different meteorological regimes. In order to ensure clouds and pollution are evaluated under the same meteorological circumstances, we use pollutant tracer fields produced by a chemical tracer transport model, which are co-located, spatially and temporally with satellite retrievals of cloud properties.

High latitude polar regions complicate satellite based studies because clouds are often similar to the underlying surface in terms of temperature and visible reflectance. Fortunately the A-train satellite formation provides a long term dataset of coincident measurements of multiple unique sensors. This study uses data from POLDER, CALIPSO and MODIS because the instruments work together synergistically where limitations from one instrument can be mitigated

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by the capabilities available from another.

2 Data

Instrument/Satellite	Parameters
MODIS/Aqua	Effective Radius, Phase
POLDER/Parasol	Cloud Height, Phase
FLEXPART Model	CO tracer, SO ₄

MODIS (MODerate resolution Imaging Spectroradiometer) AQUA Collection 5 Cloud droplet effective radius (r_{eff}) is used in this study to describe microphysical changes occurring in polluted clouds. MODIS airborne simulator (MAS) r_{eff} values in stratiform cloud agree well with in situ measurements of liquid clouds in the Arctic (Platnick et al., 2003). In this study we focus on differences and trends in MODIS r_{eff} , which minimizes potential bias errors.

POLDER (Polarization and Directionality of the Earth's Reflectance) is a wide field of view imaging radiometer on board the A-train satellite Parasol, which at present, is the only spaceborne instrument which provides combined spectral, directional and polarized measurements of reflected sunlight at a spatial resolution of 6 km (Fougnie et al., 2007).

Here, cloud height is determined from the POLDER cloud oxygen pressure (P_{O_2}) which is based on the differential absorption between the radiances measured at 763 and 765 nm, corresponding to the region of strong absorption by oxygen (Bréon and Colzy, 1999). Cloud multiple scattering places P_{O_2} more towards the center of a cloud rather than cloud top. However, compared to MODIS cloud top pressure retrievals, the P_{O_2} algorithm is unaffected by surface inversions. The P_{O_2} method performs poorly in tenuous cloud at high altitudes (Bréon and Colzy, 1999) so this study constrains cloud top heights to below 4 kilometers.

Polarization features of shortwave radiation reflected off clouds depend strongly on particle shape and size (Goloub et al., 2000). Also, in the infrared atmospheric window, there are strong differences in the spectral absorption

by ice and liquid water. Combined this information can be used to infer a highly accurate thermodynamic phase retrieval based on POLDER/PARASOL and MODIS/Aqua measurements. Coincident data from POLDER and MODIS are combined to produce a semi-continuous confidence index ranging from confident liquid to confident ice as opposed to the typical discrete classification of liquid, ice or mixed (Riédi et al., 2007). The new algorithm adds significant value to the study by better identifying mixed-phase clouds under conditions challenging for remote sensing.

Simulations of air pollution transport were made using the Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005). FLEXPART is purely a transport model and no removal processes were considered in the model calculations. The FLEXPART model was driven by the ECMWF forecasts and analyses at its 91 vertical model levels, with a global $0.5^\circ \times 0.5^\circ$ horizontal resolution. Data is output at 16 vertical tropospheric levels at intervals of 3 hours, corresponding to ECMWF forecast and analysis products. This study uses carbon monoxide (CO) as a tracer of anthropogenic combustion (biomass is not included), written as χ_{CO} to emphasize it's value as a tracer rather than real quantity. The CO tracer is entirely passive meaning that the concentrations are affected only by transport and mixing. As a built in component of the FLEXPART model, tracer particles are arbitrarily removed after twenty days of transport.

3 Methods

Compared to POLDER P_{O_2} cloud top heights, we have found a considerable bias in MODIS cloud top measurements for Arctic stratus clouds. CALIPSO Lidar, also in the A-train, provides accurate vertical profiles of the atmosphere, including vertical cloud placement, but with limited horizontal spatial coverage. On average MODIS has a 1.6 ± 0.5 km bias cloud top height compared with CALIPSO and POLDER

derived cloud heights for Arctic marine stratus clouds. An important implication of this bias is illustrated in Figure 1. MODIS cloud top heights correspond to CO tracer concentrations that are considerably different than the layer where the CALIPSO Lidar and POLDER cloud top height retrievals indicate the cloud actually lies.

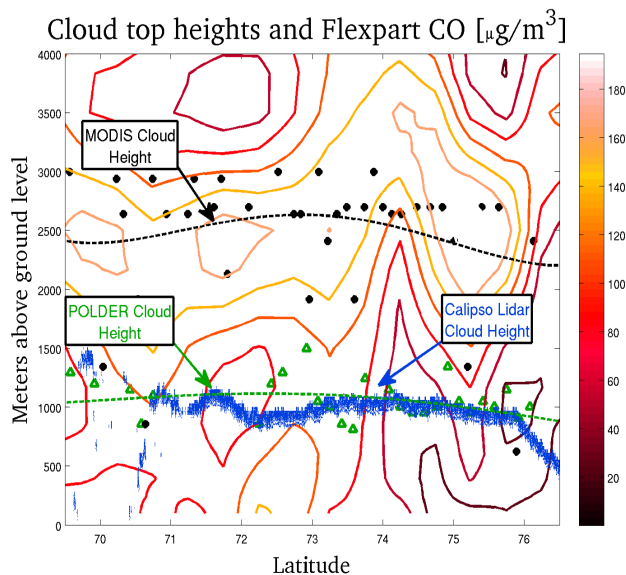


Figure 1: Comparison of MODIS, POLDER and CALIPSO cloud height retrievals co-located with FLEXPART CO tracer concentrations.

FLEXPART output is co-located temporally with the A-train overpass time, vertically with the POLDER derived P_{O_2} pressures and horizontally using geographical coordinates. For example, a 1520 UTC satellite granule is matched up temporally with the 1500 UTC FLEXPART output field. Satellite cloud-top pressures ranging between 950 to 850 hPa are collocated with the average from the FLEXPART concentrations at 1 km- and 1.5 km- level (Figure 2).

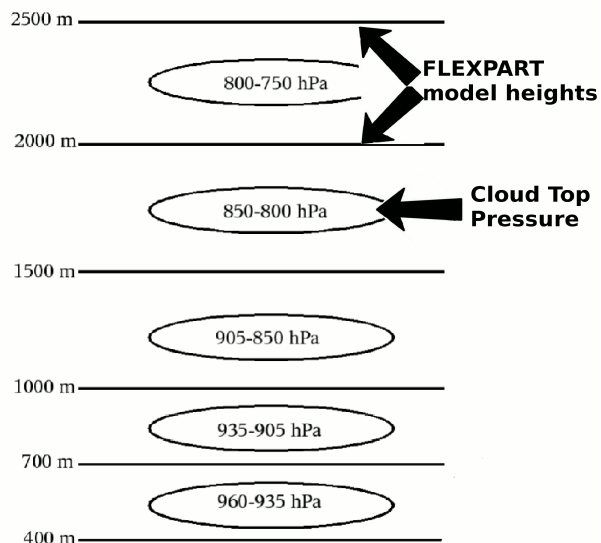


Figure 2: Vertical co-location method used to match cloud heights with pollution fields.

Cloudy pixels are filtered to be below 4 km and defined as “confident liquid” as defined by the MODIS-POLDER merged phase retrieval. Each 6 km x 6 km cloudy pixel is then stored with its corresponding pollution tracer value.

The relationship between anthropogenic pollution and Arctic clouds is quantified using an indirect effect parameter IE, which relates the relative change in r_{eff} for a relative change in some pollution quantity, which in our case is χ_{CO} .

$$IE = -\frac{d \ln r_{eff}}{d \ln \chi_{CO}}$$

FLEXPART χ_{CO} concentrations are merely a tracer of anthropogenic pollution, indicating only the extent to which CO sources have been diluted through atmospheric mixing. Calculating the IE parameter using χ_{CO} provides information about the sensitivity of arctic clouds to pollution, rather than the sensitivity of clouds to CCN concentrations. The two sensitivities will be equivalent if CO and CCN concentrations scale at sources, and if CCN concentrations are also affected only by atmospheric mixing.

4 Observations

During the month of April 2008, cloud data from 395 satellite overpasses was acquired and co-located with FLEXPART χ_{CO} fields. April was chosen in order to correspond to the transition from highly polluted winter to the less polluted summer, while providing adequate solar zenith angles to allow satellite cloud property retrievals. April 2008 also corresponds to the POLARCAT field project. The entire Arctic basin north of 67 latitude was analyzed to get a broad sense of the sensitivity of the Arctic to mid-latitude pollution. Overall, the majority of the data corresponds to un-polluted conditions but nonetheless shows a noisy but discernable tendency for smaller droplet sizes to be associated with higher value of χ_{CO} .

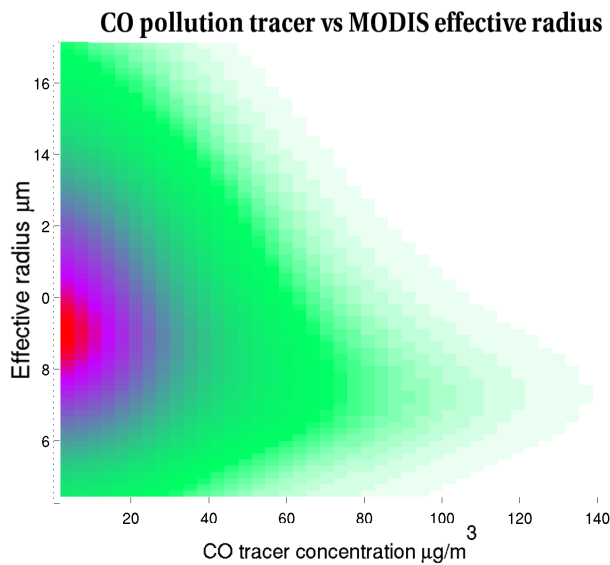


Figure 3: Distribution of FLEXPART χ_{CO} concentrations vs MODIS Effective Radius from April 2008 showing a tendency for smaller droplet sizes under higher value of χ_{CO} .

The IE parameter calculated for this data is 0.087 which is comparable to previous studies. Using a chemical mixing tracer, IE was calculated to be 0.09 for oceanic regions downwind of the Northeastern US Atlantic seaboard (Avey et al., 2007) and using aerosol scatter-

ing measurements at Barrow Alaska IE was found to range from 0.13-0.19 (Garrett et al., 2004). A statistical comparison of cloud effective radius under polluted and clean conditions is performed. Polluted is defined as the upper 10th percentile of pollution tracer data ($\chi_{CO} > 19.47 \mu\text{g}/\text{m}^3$) and clean is defined as the bottom quartile of pollution tracer data ($\chi_{CO} < 8.32 \mu\text{g}/\text{m}^3$). At the 95% confidence level, we test the hypothesis that the mean r_{eff} is smaller under polluted conditions, using a one sided Student's t-test. The hypothesis holds true where under clean conditions the mean r_{eff} is $10.1 \pm 2.5 \mu\text{m}$ and under polluted conditions the mean r_{eff} is $8.3 \pm 2.0 \mu\text{m}$. The correlation coefficient of the slope fitted to the data is $r^2 = .32$, suggesting that other factors also influence cloud droplet size. For example, wet scavenging of CCN en route to the Arctic will weaken the correlation between χ_{CO} and r_{eff} .

Conclusion

This study has explored the influence of mid-latitude pollution transported into the Arctic on cloud microphysical properties and the surface radiation budget. Using a novel space-based analysis technique, evidence is shown to suggest that elevated levels of anthropogenic pollution are weakly associated with smaller cloud droplet sizes. The analysis technique allows clouds and pollution to be compared. Future components of the study will evaluate the seasonality of pollution-cloud interactions.

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