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Comparisons of cirrus cloud microphysical properties between polluted and pristine air

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Cirrus clouds occur in the upper troposphere at altitudes where atmospheric radiative forcing is most sensitive to perturbations of water vapor concentration and water phase. The formation of cirrus clouds influences the distributions of water in both vapor and ice forms. The radiative properties of cirrus depend strongly on particle sizes. Currently it is still unclear how the formation of cirrus clouds and their microphysical properties are influenced by anthropogenic emissions (e.g., industrial emission and biomass burning). If anthropogenic emissions influence cirrus formation in a significant manner, then one should expect a systematic difference in cirrus properties between pristine (clean) air and polluted air. Because of the pollution contrasts between the Southern (SH) and Northern Hemispheres (NH), cirrus properties could have hemispheric differences as well.

Therefore, we study high-resolution ($\sim$200 m), in-situ observations from two global flight campaigns: 1) the HIAPER Pole-to-Pole Observations (HIPPO) global campaign in 2009-2011 funded by the US National Science Foundation (NSF), and 2) the Interhemispheric Differences In Cirrus Properties from Anthropogenic Emissions (INCA) campaign in 2000 funded by the European Union and participating research institutions.

To investigate the changes of cirrus clouds by anthropogenic emissions, we compare ice crystal distributions in polluted and pristine air, in terms of their frequency occurrence, number concentration (Nc) and mean diameter (i.e. effective-mean Deff and volume-mean Dc). Total aerosol concentration is used to represent the combined influence of natural and anthropogenic aerosols. In addition, measured carbon monoxide (CO) mixing ratio is used to discriminate between polluted and pristine air masses. All analyses are restricted to temperatures $\leq$ -40ºC to exclude mixed-phased clouds.

The HIPPO campaign observations were obtained over the North America continent and the central Pacific Ocean from 87ºN to 67ºS. Ice crystals are measured by a Fast-2DC probe, and the analyses are restricted to particles $\geq$ 87.5 μm to minimize the shattering effects and optical uncertainties. When analyzing ice crystals distribution in the HIPPO campaign, the occurrence frequency of in-cloud conditions increases with both total aerosol and CO concentrations. On the other hand, the changes of ice crystal sizes are not the same for increases of total aerosol and CO concentrations, that is, Dc increases with higher total aerosol concentration but decreases with higher CO concentration. These results suggest that ice crystal formation is likely facilitated when the air parcel is under influence of both natural and anthropogenic emissions, but the anthropogenic emission is likely to decrease the sizes of ice crystals.

During the INCA campaign, cirrus clouds were sampled with optical particle counters in the size range of about 1 to 800 μm at midlatitudes, mainly over the Pacific west of Punta Arenas and over the North Atlantic west of Great Britain. Simultaneous measurements of trace gases (CO, NO_x and O_3) and a suite of aerosols properties show that the INCA measurements in the SH occurred in air masses which were far cleaner than those measured in the NH. Previous INCA data analysis revealed differences between SH and NH cirrus: a lower Nc, a larger Deff, and a larger extinction in the cirrus in the SH compared to the NH (Gayet et al., JGR, 2004). We now recompiled the INCA data and performed a further analysis of the cirrus properties in correlation with simultaneous CO measurements. Based on in-situ sampling of ice crystals of different lower cut-off sizes ($\geq$1, 3 and 6 μm) from the INCA campaign, Nc is found to have weak positive correlation with CO concentration (r2 within a range of 0.2 to 0.6). The correlation appears to be significant (95% level) based on a limited set of tests with different data subsets. The correlation is strongest for the smallest ice particles. The correlation persists when restricting

the data to temperatures below -45°C. The data also reveal higher ice supersaturation in air masses with low CO concentration. The correlations suggest stronger ice nucleation in polluted air masses. Still, further measurements are desirably to exclude possible artifacts and to confirm these results.

Possibly due to the larger cutoff size ($\geq$ 87.5 μm), such correlations between Nc and CO are not captured in the HIPPO data. But the increasing Nc observed from INCA campaign is consistent with the decreasing Dc from the HIPPO campaign, since Nc and Dc are generally anti-correlated during ice crystal formation. The influence of dynamical conditions (e.g. nearby convection) and aerosol contents on the observed cirrus cloud perturbations has still to be investigated.

The comparison between data from SH and NH or from different pollution regions may be affected by sampling biases over different cirrus evolution phases. Diao et al. (GRL, 2013) suggested a method to identify the occurrence frequencies of five different phases of ice crystal evolution:
(1) Clear-sky ice supersaturated region, (2) Nucleation, (3) Early growth of ice crystals, (4) Late growth of ice crystals, (5) Sedimentation/sublimation.
"Nucleation" events in this analysis are partially cloudy segments in ice supersaturated air masses. The HIPPO and INCA data show different frequencies for these evolution phases. The full analysis of the data is still ongoing, but the INCA FSSP data (> 1 μm) seem to show more "clear-sky ice supersaturated region" and "nucleation" events to occur in the SH than in the NH.