



Volcanic influence on background sulfurous and carbonaceous aerosol in the Lowermost Stratosphere

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Previous measurements in the upper troposphere (UT) and the lowermost stratosphere (LS) have indicated the presence of a carbonaceous component in the aerosol (Murphy et al., 1998; Nguyen et al., 2008; Martinsson et al., 2009). Here the occurrence of carbonaceous and sulfurous particles around the tropopause is investigated. The data were taken from the CARIBIC (Civil Aircraft for Regular Investigation of the atmosphere Based on an Instrument Container) platform, where instruments onboard a Lufthansa passenger aircraft on inter-continental flights are used for examination of the atmospheric composition in the UT/LS at 8-12 km altitude (Brenninkmeijer et al., 2007). CARIBIC undertakes aerosol sampling for chemical characterization, as well as measurements of particle number concentrations and mixing ratios of a large number of trace gases including O₃, CO, NO/NO_y, Hg, water (gaseous and condensed), greenhouse gases and halogenated hydrocarbons. The CARIBIC dataset also contains data on meteorological conditions.

500 aerosol samples were collected during 150 flights with a sampling time of 100 minutes by an impaction technique (Nguyen et al., 2006). Specimens are then analyzed by quantitative multi-elemental analysis by PIXE (Particle-Induced X-ray Emission) and PESA (Particle Elastic Scattering Analysis) to obtain elemental concentrations for sulfur, iron, titanium, potassium, hydrogen, carbon, nitrogen and oxygen among others (Nguyen and Martinsson, 2007). The present study is based on samples collected in the LS from May 2005- August 2008.

Concentrations of particulate carbon and sulfur in the LS is shown to follow seasonal cycles, correlated with ozone concentrations, with increasing concentrations from the tropopause through the LS. This indicates downward transport from the so-called stratospheric over-world (SOV) as an important source for these species. Sulfuric acid particles are formed in the stratosphere from carbonyl sulfide (OCS) via photochemical reactions and from SO₂ injected during volcanic eruptions. Chemical characterization of volcanic aerosol injected to the LS from the Kasatochi eruption in August 2008 (Martinsson et al. 2009) has shown large amounts of carbon present in the particulate fraction of volcanic aerosol.

Vernier et al. (2009) show strong volcanic impact on aerosol scattering in the Northern Hemisphere (NH) midlatitude during 2006 -2008 caused by eruptions in the tropics. The present study shows high particulate carbon and sulfur concentrations on samples collected during November and December flights, when downward transport from the SOV maximizes, indicating transportation of volcanic aerosol from the SOV. Concentrations of particulate carbon and sulfur increase in the midlatitude NH from 2006 - 2008. This is explained by injection of volcanic aerosol to the stratosphere via eruptions in the tropics, and transportation to midlatitude by the Brewer-Dobson circulation, where air is transported downward to the LS. The composition of the carbonaceous aerosol is not known. A stoichiometric oxygen-to-carbon ratio of 0.2 indicates organic matter.

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