Composition of the Asian Tropopause Aerosol Layer simulated with a coupled aerosol-chemistry-climate model: enhanced H_2SO_4-H_2O droplets, HNO_3-H_2SO_4-H_2O ternaries, organics, ice, or a mix?

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The Asian Tropopause Aerosol Layer (ATAL) is a confined region of enhanced aerosol associated with the Asian Summer Monsoon. Backscatter measurements by instruments such as the satellite-borne CALIOP or the balloon-borne COBALD reveal the ATAL in the altitude range between 90 and 130 hPa. However, the exact composition of the ATAL is not yet clear. Here we performed model simulations with the coupled aerosol-chemistry-climate model SOCOL-AER to investigate the sulfur budget of ATAL and its dependence on precursor emissions. The aerosol module AER comprises gaseous and aqueous sulfur chemistry and comprehensive microphysics with 40 size bins spanning radii from 0.39 nm to 3.2 µm, including size-dependent particle composition. In a "sulfur-only world" the model is capable of correctly reproducing the observed geographic position and altitude range of the ATAL. With about 0.25 ppbv of sulfuric acid (H_2SO_4) in the ATAL the amount of particles is about twice as high than at the same altitude outside of the ATAL. Compared to observations, however, the backscatter of the modeled sulfur-only ATAL is too low by a factor of 2 to 3. Balloon-soundings indicate that the ATAL aerosol appears exactly in the coldest region of the atmosphere, namely at most 1 K above the frost point. At such low temperatures, nitric acid (HNO_3) partitions from the gas phase to the particle phase, forming ternary HNO_3-H_2SO_4-H_2O solution droplets. Preliminary results of nitric oxide (NO) and total reactive nitrogen (NOy) measurements by the SIOUX instrument aboard the Geophysica research aircraft performed during the StratoClim campaign suggest mixing ratios of HNO_3 of about 1 to 3 ppbv. The uptake of HNO_3 results in a growth of the ATAL particles and an increase in the backscatter signal of the simulated ATAL. The HNO_3 uptake might be further enhanced in the presence of ammonia (NH_3). Furthermore, the model simulations suggest the transport of isoprene, which substantially contributes to global secondary organic aerosol (SOA) formation, into the ATAL. Finally, dispersed ice particles may also contribute to the observed backscatter signal. By disentangling the contribution of the above-mentioned factors to the backscatter signal of the simulated ATAL we aim for conclusions on the composition of the ATAL.

Key words: ATAL, Sulfate Aerosol, Nitric Acid, Chemistry-Climate Model