Atmos. Chem. Phys. Discuss., 4, S2672–S2674, 2004
www.atmos-chem-phys.org/acpd/4/S2672/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Fluorescence from atmospheric aerosol detected by a lidar indicates biogenic particles in the stratosphere" *by* F. Immler et al.

U. Krieger

ulrich.krieger@env.ethz.ch

Received and published: 22 November 2004

This is a very exciting report about a series of LIDAR measurements in the lowermost stratosphere showing a fluorescence signal in the water Raman channel of their instrument at a wavelength of 407 nm. The authors show very convincingly that this signal is due to laser induced fluorescence from aerosol particles. They attribute this signal to organic material released by biomass burning from Siberian forest fires lifted across the tropopause and transported around the globe.

An alternative explanation for the observation could be that the fluorescence stems from organic material processed in acidic sulfuric acid aerosol particles under dry conditions as observed in laboratory experiments [Hegglin et al., 2002]. Hegglin et al. showed that a wide range of different organic matter induces fluorescence in H_2SO_4/H_2O solutions. They suggested that the ubiquity of organic substances in the atmosphere, their relatively high abundance, and the insensitivity of the fluorescence with respect to their concentrations observed in the laboratory experiments will render most acidic natural aerosols subject to light absorption and fluorescence. The concomitant light absorption may be relatively weak, however.

If this explanation proves true it may also explain the observation that the observed aerosol layer showing the fluorescence also exhibts a significant depolarization with a clear diurnal cycle. If the aerosol particles are of a mixed phase type consisting of an organic (polymerised) phase in an inorganic aqueous acidic liquid they will show a depolarizing signal [Krieger et al., 2004] and its magnitude depends on the relative volume of both phases. The volume of the aqueous phase however, may show a diurnal cycle because the absorption of light in the organic phase during the day will induce a temperature change with a subsequent evaporation of part of the water until a new equilibrium is reached. During the night this process will be reversed.

Taking all this into consideration it could be that the reported florescence of atmospheric aerosol in the lowermost stratosphere is not a very interesting curiosity but a more common feature of acidic atmospheric aerosol containing organics when dry conditions and lifetime allow for enough chemical processing (polymerization) of the organics. We believe a dedicated LIDAR instrumentation (using a channel with considerable bandwidth since the fluorescence signal has a broad spectral feature) could prove or disprove this hypothesis in future measurements.

References:

ACPD

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