

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

F. Immler et al.

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The discussion of our manuscript "Fluorescence from atmospheric aerosol detected by a lidar indicates biogenic particles in the stratosphere" has brought up some interesting comments: U. Krieger pointed to fluorescence observed at levitated sulphuric acid droplets caused by a contamination with organic compounds. The intensity of the fluorescence was 2-3 orders of magnitude larger than the Raman scattering of the droplet. We have included these results (published by Hegglin et al.) in a revised version of our manuscript and show that the fluorescence created by this type of particle could be the cause of the inelastic signal that we have detected from aerosols with our Raman lidar.

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This example adds to a number of examples mentioned in our paper of atmospheric particles that can fluoresce when excited with a UV laser. We claim that we have detected such a fluorescence with our water vapour Raman lidar without being specific on the exact type of particle. This result is based on the observation of a significant deviation of the Raman lidar water vapour profile from radiosonde measurements. Anonymous referee #2 expressed doubts whether the reported deviation were significant and argued that the calibration of the Raman lidar could introduce large errors. Following the suggestion of referee #2 we have added a more detailed description of the Raman method for water vapour measurement to the revised version of our manuscript. Also, we have changed some figures to demonstrate more clearly the significance of the detected deviations. In fact, there are always differences between lidar and radiosonde data. Some of which are due to difference in time and space of the measurements, some are due to differences in vertical resolution, some could be due to inaccuracies in both lidar and radiosonde measurements. As demonstrated in figure 2 of the revised version these differences are on the order of 0.1 g/kg and are certainly worth being discussed. However, in our manuscript we focus on a much larger discrepancy between lidar and radiosonde data of the order of 1 g/kg that we observed during the presence of strong aerosol layers. We argue that such a large deviation could not be caused by the reasons mentioned above nor could they be due to problems with the calibration.

We conclude that the observed deviation are most likely caused by fluorescence from the aerosol. This conclusion has two important implications: Fluorescence of aerosol could be used as a tool to identify the aerosol's source. To our knowledge the fluorescence is caused only by organic compounds. Therefore we conclude that the stratospheric aerosol layer is of biogenic origin and is a remnant from Siberian forest fires which were abundant across the northern hemisphere at that particular time. As emphasized by referee #1 our finding also implies that fluorescence from aerosol could be a source of error for water vapour profiling by Raman lidars. This problem was widely ignored by the community and should receive larger attention. Some numbers that allow an assessment of the strength of this error source depending on the aerosol load

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and type are provided in our manuscript. However, we think that a more detailed investigation requires more information from field and laboratory experiments than currently available. An additional inelastic detection channel - off line the water vapour and nitrogen Raman lines - would certainly be a very helpful tool to detect fluorescence from aerosol and widely eliminate its effect on the Raman measurement. It would also allow to detect biogenic aerosol layers without the need to compare lidar to radiosonde data.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 5831, 2004.

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