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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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In our manuscript we claim that we have measured a fluorescence signal from atmospheric aerosol that we think is created by organic material. According to referee #2 our argumentation was not convincing despite the fact that we presented data of very good quality. It is not quite clear to us whether the referee doubts the feasibility of the detection of fluorescence from aerosol in the atmosphere by a lidar or whether he doubts that the fluorescence is due to organic matter in the aerosol. Probably, we were not precise enough in our statements: We have evidence for inelastic scattering from aerosol plumes detected by a lidar. We interpret this as laser induced fluorescence from organic matter because this explanation is the most plausible one. We do not

claim the ability to determine the type of particles that induces the fluorescence. Instead we present different examples from the literature that can explain our finding like laser induced fluorescence of PAH on soot and the fluorescence of bioaerosol. One more interesting possibility, processed organic compounds in sulphuric acid droplets, was presented by Krieger in a short comment. All these examples have in common that the fluorescence is created by organic matter. We therefore conclude that we have detected fluorescence from biogenic aerosol in the atmosphere by a lidar.

Referee #2 is criticising the little space we have devoted on the description of the lidar techniques, in particular on the calibration of water vapour measurements by the Raman technique. We agree that a more precise description of the method will be helpful to support our conclusion. Concerning the retrieval of aerosol optical depth from nitrogen Raman signals and the retrieval of effective size from the colour index, we prefer to refer the reader to the publications cited in the manuscript, where these methods are explained in all the details. However, we will explain the calibration technique for water vapour retrieval more precisely in a revised version of the manuscript. We will demonstrate that the calibration accounts for an error in the water vapour retrieval of a few percent. The peak in the raw signals measured with the 407 nm channel presented in fig. 3 at 13 km altitude is about 2 orders of magnitude higher than what one would expect from Raman scattering by the water vapour that is present in the lowermost stratosphere. This demonstrates, that the calibration issue is not crucial for the evidence we provided concerning the significance of the inelastic signal received from the aerosol plume.

We then argue that fluorescence is the most likely physical mechanism that caused the inelastic signals that we measured. We demonstrate, that our system does not detected Raman scattering from ice or water particles and therefore we do not see any other explanation for our finding but fluorescence from a different type of particles. This conclusion is supported by the following research results:

1. It is well known (and numerous citations are listed for that) that numerous organic

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compounds including i.e. PAH and bioparticles do fluoresce and moreover, most of these particles may fluoresce in a way that is in principle detectible by our lidar system 2. These agents occur in the atmosphere and are particularly likely to occur in plumes of forest fires. 3. It is quantitatively possible, that we detected fluorescence from an aerosol plumes. This consideration in section 4.2 is based on a rough estimate because quantitative data on the fluorescence properties of atmospheric particles that is useful for the interpretation of lidar measurements are scarce and we don't see the possibility to produce better data in short term ourselves. Even though the uncertainties of this consideration are high, it shows that it can not be ruled out from a quantitative point of view, that we detected fluorescence from atmospheric particles. 4. In both cases where we detect inelastic scattering from aerosol layers, there is a strong link to forest fires: In early June 2003 large parts of the northern hemisphere was affected by Siberian forest fire plumes. On 5 August 2003 backward trajectories demonstrate that the strong plumes in the troposphere origin from Portuguese forest fires.

Based on these facts we argue that fluorescence from biogenic particles which are present in forest fire plumes is the most likely mechanism to explain our experimental results because this interpretation yields a consistent picture of the entire observation. Even if we can not offer a final proof for the existence of fluorescence, we think the indications that we are on the right track are rather strong. Other hypothesis that we have tested were by far not as convincing including experimental artefacts, interference from clouds or other physical effects that might occur on atmospheric particles like surface enhanced Raman scattering (SERS). Of course we can not rule out that another explanation exists, however, neither the presentation of these results in a wider circle of colleagues at workshops, nor in the ACP discussion section have so far brought up one.

It needs to be clarified again that we do not claim the ability to determine what exactly causes the fluorescence of the aerosol. If our statement at the end of §4.2 - saying that the precedent consideration suggests that PAH might be more important than biopar-

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ticles - is interpreted in this way it is justifiably criticised by anonymous referee #2. In fact, that sentence was written to counteract the impression that we are trying to prove that we can detect bioparticles in the atmosphere and to remind the reader, that PAH or other organic compounds on soot might be a lot more important. But it is not possible to discriminate theses agents from our observational data. As a matter of fact the use the fluorescence of aerosol to identify its type - or the chemical compound on its surface that fluoresce - is difficult even under laboratory conditions. The possibility to measure a full fluorescence spectrum helps a lot but still yields in some cases ambiguous results (see. Hill et al., 1999).

The efficiency of fluorescence in condensed matter depends on numerous variables including pressure and temperature but also the nature, concentration and location of fluorescing compounds in or on the particle. This information is hardly accessible. Therefore, we think that a theoretical treatment of fluorescence of atmospheric aerosols would be difficult to undertake and would hardly yield the inelastic scattering coefficients in sufficient accuracy to be useful for the interpretation of our data. Precise laboratory data is needed to interpret our results, but is currently not available.

We agree with referee #2 that the timing issue of fluorescence should be mentioned. However, it is of no importance: Fluorescence signals of PAH for example typically decay on a time scale of some 10-100 ns (see i.e. Niessner et al., 1991). The time resolution of the detection system of our lidar is 50 ns corresponding to a vertical resolution of 7.5 m. The finite duration of fluorescence leads therefore to a blurring of the vertical structure of a fluorescing aerosol plume in the range of some single altitude bins at the most and is therefore not detectable with our system because it is too close to its resolution. Moreover, the natural borders of aerosol plumes are usually not well enough defined to allow for the detection of such a small time delay.

We thank referee #2 for the detailed and helpful specific comment and we will try to improve the manuscript accordingly in a revised version.

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Interactive Comment