Atmos. Chem. Phys. Discuss., 4, S2544–S2548, 2004
www.atmos-chem-phys.org/acpd/4/S2544/

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.

Anonymous Referee #2

Received and published: 15 November 2004

Major comments

This paper describes a very interesting series of lidar measurements of a stratospheric aerosol layer observed in the lowermost stratosphere in the spring of 2003. The measurements shown appear to be of very good quality and the complementarity between the various parameters observed (aerosol backscatter ratio at various wavelengths, water vapour mixing ratio) give a good overview of this interesting event. However the argumentation for showing that these observations indeed correspond to anthropogenic fluorescing aerosol lifted up to the stratosphere is rather messy and as such not very convincing. In particular section 4, which focuses on the fluorescing character of the observed particles, provides a very rough analysis of the type of particles likely to

Full Screen / Esc
Print Version
Interactive Discussion
Discussion Paper

induce the signal observed on the water vapour Raman channel, with very little theoretical background on laser induced fluorescence in order to support the argumentation.

The article refers to various lidar techniques (elastic scattering, Raman scattering and laser induced fluorescence), so section 2 or an appendix should specify briefly for the non lidar specialists the formula used for the retrieved parameters (including the water vapour parameters and the effective radius) described in section 3 (Observations) and section 4 (fluorescence lidar). Such a paragraph would help understand why in section 3 the optical depth at 532 nm is retrieved from the nitrogen Raman channels, what colour index is used in that case and which scattering theory (most probably the Mie theory) is used to infer the effective radius from the measurements. It would also improve the understanding of section 4 which attempts to demonstrate that the signal observed is due to laser induced fluorescence. Moreover, the methodology for deriving water mixing ratio from the measurements should be described more thoroughly, in particular the calibration technique, since radiosonde measurements are both used to calibrate the measurement and to show possible biases in the water vapour mixing ratio due to anthropogenic aerosols.

A large part of the paper is devoted to the demonstration that there is no bias between the water vapour lidar measurements and radiosoundings performed on the same site. This argumentation is based on one measurement only, which is not very convincing. In that respect, the difference between lidar water vapour measurements and radiosondes should be shown in the whole measurement altitude range during the period of interest (beginning of June 2003). It is rather intriguing that the authors use the radiosonde data to calibrate the lidar data for these measurements while they argue in section 4.1 that biases between lidar and radiosonde measurements are due to anthropogenic aerosol.

In section 4.2, the suggestion that PAH is the most probable agent creating the observed signal is made only by default, using a very rough estimate of fluorescence by bacteria and pollen. More arguments are needed to support the authors' suggestion.

ACPD

4, S2544–S2548, 2004

Interactive Comment

Besides, the argumentation should also include considerations on the timing of the LIF signal and its detection by the lidar.

The title should be more precise about the location of the detected aerosols. The particles are indeed detected in the lowermost stratosphere, merely in the tropopause region as shown in Figure 2. The fate of such aerosol layer is uncertain and we don't know whether it will remain in the stratosphere or return to the troposphere.

Specific comments

Introduction

Page 5832, line 24-25 : The formulation is not clear : what is the process that suppresses tropospheric-stratospheric transport?

Page 5834, line 7-10: Here also the formulation is not precise enough: what aerosol distribution satellite or ground sensors are able to provide : vertical? Size? Why are such sensors enable to provide these informations?

Method

The authors should specify already in this paragraph the site where the measurements are performed and its coordinates.

Page 5834, line 17: how the same pulse energy is achieved 355 and 532 nm, using frequency doubling and tripling techniques?

Page 5834, line 28: Specify aerosol backscatter coefficient, or is it total backscatter coefficient?

Observations

Page 5835, line 25: specify the lower lapse rate tropopause definition.

Page 5836, line 8: in order to demonstrate that the aerosol layer is in the lowermost stratosphere, figure 2 should include the evolution of the altitude of the 380 K isentropic

ACPD

4, S2544–S2548, 2004

level.

Page 5836, line 18: specify the lidar ratio measured for standard background aerosol. Is such a high lidar ratio close to what has been observed for soot particles in the troposphere? The authors could give some examples found in the literature.

Page 5836, line 23: Indicate the value of the assumed refractive index.

Page 5837, line 2: Figure 3 should indicate the standard deviation derived from the counting statistics of the count rate at 407 nm.

Page 5837, line 3: The 407nm signal seems to be wider than the backscatter profile (especially on June 2). Do the authors have an interpretation for this?

Page 5837, line 8: to what parameter corresponds the figure 109?

Page 5837, line 11: the authors mention for the first time Figure 5 without describing it (it is done later). What does it demonstrate here ?

Discussion

Page 5838, line 14: the date seems to be wrong here.

Page 5838, line 15: Figure 5 should be in the same format as Figure 6 with the measurement error bars.

Page 5838, line 26: More information should be given on the backward trajectories shown in Figure 7. How where they calculated? From which meteorological analyses?

Page 5839, line 22: the acronym PAH should be introduced here.

Page 5840, line 15: explain why the backscatter coefficient (Raman supposedly) is used to estimate the sensitivity of the instrument regarding the detection of fluorescence.

Page 5840, line 18: the authors have indicated earlier that possible bias between lidar and radiosonde water vapour measurements could be due to fluorescence on aerosol

ACPD

4, S2544–S2548, 2004

particle. Why is the influence from fluorescence certainly negligible in the lower ranges in the beginning of June 2003?

Page 5840, line 25: specify the type of scattering cross section and the wavelength used.

Conclusion

Page 5842: the discussion on the origin of the aerosol layer should be included in section 4.2. Due to the length of the trajectories (10 days), this discussion should be based on clusters of trajectories. If the trajectories experienced convection, how accurate are they in determining the origin of the observed aerosol layer?

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