

Interactive comment on “Dependence of solar radiative forcing of forest fire aerosol on ageing and state of mixture” by M. Fiebig et al.

M. Fiebig et al.

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Referee #1 gives helpful suggestions for clarifying the way data is displayed and improving explanations in the article. In order to include the suggestions, the following changes were made to the article:

1. Referee #1 refers to the point that it is necessary to assume the particles as spherical with a fixed refractive index to depict data of optical particle counters (OPCs) as size distribution. For externally mixed aerosol, this approach is not strictly correct but still much more illustrative than plotting a histogram of the scattering cross section actually measured by the OPC. However, when dealing with an externally mixed aerosol, size distribution parameters obtained this way have to be verified by calculating the aerosol's instrumental response for the respective OPC, i.e. the scattering cross section histogram generated in the OPC, and

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comparing it with the scattering cross section histogram actually measured. This analysis has been done for the LACE 98 forest fire aerosol (Fiebig et al., 2002), the present article builds on the results of that study. For all other analyses made, an external mixture of ammonium sulphate and soot was assumed. To clarify the the data treatment, the following changes have been made to the manuscript:

p. 1277, line 15: The following sentences have been added:

For this graph, the aerosol has been assumed to consist of ammonium sulphate and soot internally mixed with a refractive index of $1.6 + 0.03i$. To obtain the uncertainty in D , m was varied by $0.1 + 0.006i$.

p. 1278, line 27: The following sentences have been added:

This fact was taken into account throughout the reported analysis. The size distribution (Fig. 1) was validated by calculating the instrument responses of the OPCs on the externally mixed aerosol and comparison with the measured instrument responses (Fiebig et al., 2002).

2. The analysis proposed by referee #1 has indeed been done (Fiebig, 2001). Using the size distribution measured in the upper sublayer and assuming ammonium sulphate and soot as chemical components, several options are possible concerning the state of mixture. For a purely external mixture, 55% of soot and 45% of ammonium sulphate (all by volume) are necessary to reproduce the spectral lidar signal measured in the upper sublayer. A percentage of soot this high is unlikely even for a large forest fire. Other possible options are a purely internal mixture of 7% soot and 93% ammonium sulphate, or an external mixture of 20% pure soot particles and 80% internally mixed particles consisting of 6% soot and 94% ammonium sulphate. This result supports the article's conclusion that mixing between forest fire and background aerosol was stronger in the upper sub-layer than in the lower sub-layer, resulting in a more internal mixture between soot and ammonium sulphate in the upper plume parts as compared to the lower

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plume parts. However, without a measurement of the particle absorption coefficient, the exact state of mixture in the upper sub-layer remains too speculative to be used as argument. The state of mixture in the upper sub-layer was therefore not included in the present article's analysis, although the analysis quoted above supports the articles argument.

3. The statement “ultimately become internally mixed” refers to the finding that BC particles become internally mixed by coagulation or coating with condensable species (e.g. summary by Seinfeld and Pandis, 1998, page 708) - provided the air mass that contains BC particles mixes with background air masses that contain condensable species and / or scattering particles. Even a very dense and persistent forest fire plume will finally dissolve and mix with background air, facilitating internal mixing of the BC particles. However, referee #1 is right in pointing out that the corresponding statement is confusing. The corresponding sentence has been rephrased as follows:

p. 1286, lines 22 - 25: The sentence:

Since the absorbing forest fire particles are emitted externally mixed with purely scattering background particles, but ultimately become internally mixed, scenarios 2 and 3 are both treated as ideal internal as well as external mixtures to obtain bounding values for the corresponding solar radiative forcing.

has been rephrased to:

Since the absorbing forest fire particles are emitted externally mixed with purely scattering background particles, but become internally mixed when the respective part of the plume dissolves, scenarios 2 and 3 are both treated as ideal internal as well as external mixtures to obtain bounding values for the corresponding solar radiative forcing.

4. The statement “should be treated with caution” is indeed misleading and should be termed more precisely. The heating rate stated in the article is a maximum

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likelihood estimate for the plume age of 3 days, since it is based on the case and state of mixture (high initial particle concentration, no dilution, external mixture of absorbing and non-absorbing particles) that reproduces the size distribution in the LACE 98 lower sub-layer best. Higher heating rates are possible when treating the same case as internal mixture. However, this scenario is not plausible since dilution with background air is necessary to mix the absorbing particles internally with non-absorbing species. Lower heating rates down to zero heating are also possible when dispersion dissolves the plume.

In this light, it would be rather confusing for the reader if the possible range of heating rates is discussed besides a maximum likelihood estimate. Such a discussion would distract from the actual aim of stating the maximum likelihood heating rate. This aim is to give indication that the dynamics within the Canadian August 1998 forest fire plume and the 1991 Kuwait oil fire plume were similar.

Rather than adding a potentially confusing discussion of possible heating rates, the following changes were made to clarify the line of argument and the purpose of the stated heating rate:

page 1289, line 24:

The following sentence has been added:

This choice of parameters results in a maximum likelihood estimate for the heating rate at this plume age since this scenario reproduces the aerosol observed in the lower sub-layer.

lines 27-28:

The following sentence has been removed:

Due to the uncertainties in the temporal evolution of the microphysical properties in the LACE 98 forest fire plume, this heating rate has to be treated with caution.

page 1289, line 28 - page 1299, line 2:

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The sentence:

On the other hand, considering all information available on this plume, it appears possible that the plume developed into a radiative-convective mixed layer, feeding back onto its dilution, its microphysical properties and its radiative forcing.

has been replaced by:

Considering all information available on this plume, it appears possible that the plume developed into a radiative-convective mixed layer, feeding back onto its dilution, its microphysical properties and its radiative forcing.

5. Following this suggestion, the size distribution parameters of the LACE 98 forest fire aerosol (Table 1) were included in Table 2 two facilitate an easy comparison of all measurements. On the other hand, since Table 1 contains not only information on the accumulation modes, but also on Aitken and coarse mode, Table 1 was left unchanged.
6. The statement that the size distribution parameters obtained in different plume parts by Formenti et al. and during LACE 98 are not significantly different simply refers to the respective numbers and their standard deviation. Referee #1 is right in pointing out that taking into account additional transport time, the two two size ditributions could actually be different. On the other hand, since there is not enough information available to determine whether air parcels present over Lindenberg were transported to Greece, a connection between the these two aerosols remains highly speculative. To avoid confusion, the statement on the quantitative comparison between the LACE 98 and STAAARTE-MED forest fire aerosols was removed.

p. 1281, lines 18-19: The following sentence was removed: These value are not significantly different from the respective data obtained in the upper sublayer over Lindenberg (see Table 1).

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References

Fiebig (2001): Das troposphärische Aerosol in mittleren Breiten - Mikrophysik, Optik und Klimaantrieb am Beispiel der Feldstudie LACE 98 (The tropospheric aerosol at mid-latitudes - microphysics, optics, and radiative forcing illustrated by the LACE 98 field study), Ph.D. thesis at the Ludwig-Maximilians University Munich (in German).

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 1273, 2003.

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