Atmos. Chem. Phys. Discuss., 4, S1462–S1465, 2004
www.atmos-chem-phys.org/acpd/4/S1462/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "A parameterisation of the soot aging for global climate models" *by* N. Riemer et al.

N. Riemer et al.

Received and published: 3 August 2004

First of all we would like to thank the reviewers for their very detailed comments and suggestions.

All three referees agree on the issue that the time scales derived in our study strongly depend on the chosen conditions and that they therefore will not be representative for the whole globe. Since at the present stage we cannot overcome the limitations of our current model formulation we reacted to this point by changing the scope of our paper: away from our original goal of developing a parameterisation for global models, towards a systematic discussion of the results of our case studies. Our new title ŞSoot Aging Time Scales in Polluted Regions During Day and NightŤ reflects this change. We rearranged section 4 (Results), included detailed discussions and removed section 5 (Parameterisation of  for global climate models).

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Detailed responses to the comments of Referee #1:

Fig. 7: The sulphate concentration in model is too low. The refereeŠs concern is that the lifetime of BC during night might be considerably shorter than calculated.

This figure is misleading. Shown in this figure are the concentrations of the modes ic+jc, that is the soot-containing modes. The overall aerosol concentrations are higher since the soot-free modes if+jf contain a considerable amount of the total mass. To clarify this issue we included a new figure (Fig. 5) that shows the total aerosol mass. It shows that in fact our calculated concentrations are in the range of observations for central Europe.

More sources contribute to BC than diesel cars alone. Primary organic carbon is not included.

The referee addresses an important point, that is the limited information about the emissions and the hereby introduced uncertainties in the model results. In our improved version we state clearly that we do not include other sources than diesel cars and that therefore a part of the real emissions is missing.

Aging by nitrate formation: Describe more clearly how the aging due to nitrate is treated.

Condensation of sulphuric acid on the surface of the soot particles and the subsequent formation of ammonium nitrate transfers soot into an internal mixture.

How is nitrate distributed over the modes?

A thermodynamic equilibrium of gas phase and aerosol phase is applied to calculate the concentrations of sulphate, ammonium, nitrate and water (Kim et al. 1993). The calculation of the thermodynamic equilibrium follows a bulk approach. This means that the aerosol concentrations summed over all modes enter the calculation. After the equilibrium concentrations are obtained the concentrations of ammonium, nitrate and water are distributed over the modes depending on the mass fraction of sulphate.

ACPD

4, S1462–S1465, 2004

Interactive Comment

Role of hydrolysis, aging by nitrate during the night.

The formation of ammonium nitrate also occurs during the night when the gas phase precursor HNO3 is formed due to the heterogeneous hydrolysis of N2O5 (Riemer et al., 2003a). However, since the formation and hence the condensation of H2SO4 ceases during the night due to the lack of sunlight, the soot particles that are emitted during the night cannot be coated with sulphate. Since in our current model formulation the total amount of nitrate is distributed over the modes according to the sulphate content the available nitrate ends up in the modes if, jf, ic and jc and not in mode s. This means that the aging of soot due to condensation is not effective during the night, both in winter and in summer. The new Figure 9 which displays a time height section of the soot transfer due to condensation (winter case only) shows this fact. We are aware of the fact that this treatment of aerosol chemistry represents a limitation of our model. With this assumption it is not possible to form ammonium nitrate on the soot particles without sulphate being involved. In reality this process might occur which means that we might overestimate the time scale for the situations where ammonium nitrate is present. This issue will be addressed in future work.

Parameterization is limited to areas where nitrate is present

Since we no longer aim for the global scale parameterisation this is not a problem any more.

Influence of clouds: discussion of cloud processes on the aging time scale should be included.

We included this in the new section 4.

Description of initialization and boundary conditions

We explain initialization and boundary conditions in section 2.

Detailed responses to the comments of Referee #2:

ACPD

4, S1462–S1465, 2004

How general are the results using only two scenarios? If they are cloud-free days how can they be representative of continental conditions?

We agree with the referee that we cannot generalize the results of our two scenarios for the whole globe. We therefore changed the focus of our paper and present the two scenarios as case studies to explore the variability of the soot aging and no longer derive a parameterisation for the global scale.

Is now sulphuric acid condensing on the particles during the night?

This issue is obviously a misunderstanding. Sulphuric acid is not condensing during the night because we do not allow it but because it is not formed in the first place due to the lack of OH. We tried to clarify this in section 4.

Can the authors suggest the maximum height up to which they expect that the parameterizations work? The authors neglect the height dependence in winter conditions and it is not clear why

We excluded the parameterisation from our new version of the paper.

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Detailed responses to the comments of Referee #3:

The referee is also mainly concerned about the applicability of our results to the global scale. In agreement with the other two referees we followed his suggestions and changed the scope of our paper. We also adopted the refereeŠs suggestion for the title of the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 2089, 2004.