**Atmospheric
Chemistry
and Physics
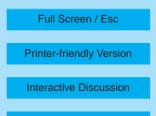Discussions**

Interactive
Comment

Interactive comment on "Equilibrium of sinks and sources of sulphate over Europe: comparison between a six-year simulation and EMEP observations" *by* M. Ménégoz et al.

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First of all, we want to express our sincere thanks to the first anonymous referee who has read our manuscript very carefully. We will revise our manuscript according to his/her comments. Major comments are exposed through 7 different points. The first 5 points concern the aerosol description in our model, which we agree should be improved in our paper. Our response presented in the following is divided into 4 parts: a first part details the aerosol representation in the model, a second part exposes our answers to the last two major comments, and a third part is devoted to the minor comments. The last part contains the responses to the technical comments.

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1. Aerosol representation on the model (response to the major comments 1 to 5)

The AEROCOM inventory used in our study describes the quantity of aerosols and precursors gases emitted in the atmosphere and provides a distribution for each type of aerosol. This distribution is a sum of log-normal distributions. Diameter, standard deviation and fraction number of these different modes are presented for each aerosol in the first column of table 1 of the new version of our manuscript. In our model, aerosol log-normal distributions are discretized into bins of different sizes, as described in Martet (2008). The chemical production of sulphate and the direct emissions of black-carbon and mineral dust are injected into the atmosphere following the bin distributions defined in the second column of table 1 (please, see table 1 in the new manuscript; 5 bins from 0.01 to 100 μm for dust, and 4 bins from 0.001 μm to 10 μm for sulphate and BC). Note that, as suggested by the AEROCOM project recommendations, a fraction of 2.5% of sulphur emissions is directly injected into the atmosphere under the form of primary sulphate particles. Bins are wide enough to cover entirely the log-normal distributions suggested by the AEROCOM project, both in term of number and mass fraction distributions. In addition, aerosols are assumed to be externally mixed, each one evolving independently from the others in our simulation.

This approach is very simplified, and can be criticized because it does not describe perfectly the real aerosol distribution and composition. For example, as the first anonymous referee pointed out, the size of BC should not be extended down to 1 nm, because very little BC exists below about 50 nm in the atmosphere. However, our approach allows to perform global simulations at a reasonable computational cost. We assume this description of aerosol distribution is reasonable to evaluate the aerosols fluxes in global and regional simulations at first order.

Our study focuses on the analysis of sulphate sinks and sources. Missing processes in our model probably causes biases in the simulation of sulphate. For example, the interactions between the different types of aerosols are not taken into account and some aerosols are not considered in our model. In particular, the representation of

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sea-salt and organic aerosol is not included in our model. Nevertheless, the interactions between the different types of aerosols are second-order processes compared to transport, emissions, wet deposition and chemistry, which need to be well represented to correctly simulate sulphate aerosol (Trivitayanurak et al., 2008). The major part of the atmospheric black-carbon is mixed with the other aerosols, especially with sulphate, which can strongly affect its lifetime. This is not the case of sulphate, because only 20% of non sea-salt sulphate is internally mixed with other aerosols, the rest evolving independently from the others aerosols (Liu et al, 2005). Ma, X., and K. von Salzen (2006) pointed out that coagulation and condensation can affect the mass size distribution of sulphate. The major sink for this aerosol is wet deposition, composed at 95% by in-cloud scavenging and at 5% by below-cloud scavenging (Boucher et al., 2002). In our model, the parameterisation of the in-cloud scavenging does not depend on the mass size distribution. It only depends on the physical and chemical properties of sulphate. Consequently, we assume that our global simulation of sulphate is weakly penalized by the fact that coagulation and condensation processes are not taken into account.

Dust emissions are not dependent on wind velocities in the version of the model that we used. Since we did not take into account the interactions between the different types of aerosol in our simulation, this can not have an impact on the sulphur cycle. A scheme describing wind-dependent dust emissions has been implemented in the MOCAGE model. It is presented and validated in Martet (2008).

2. Response to the other major comments

- Relationship between sulphate burden and precipitation (major comment number 6)

As the first anonymous referee explained, the relation between sulphate burden and precipitation depends on the averaging period (hourly, daily, monthly, etc...). In our new manuscript, in order to clarify the study of the link between the sulphur cycle and temperature and precipitation, we plotted all variables considering weekly moving

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averages along the six year simulation.

The variance of precipitation averaged over Europe is very strong at the daily time scale (not shown). It is significantly reduced considering weekly timescale. Sulphate burden integrated over Europe shows approximately the same variance considering daily or weekly timescale. There is no evident link between the sulphate burden and the precipitation at a weekly timescale (The Fig. 3 in the new manuscript shows the evolution of these variables at a weekly timescale). However, as we pointed out in our manuscript, both variables show a significant annual cycle. Nevertheless, the maximum of sulphate burden occurs about three month after the maximum of precipitation. As we said in the previous version of our manuscript (and also in the new manuscript), it is difficult to characterize the link between precipitation and sulphate : High precipitation implies large scavenging rates on one hand, but it implies also significant aqueous chemistry production of sulphate because of the high LWC of the atmosphere on the other hand.

- Comparison between sulphate surface concentration simulated by MOCAGE and observed in EMEP network (Major comment number 7).

According to a suggestion of the first anonymous referee, we improved the colour scale of the graph which compares the sulphate surface concentration modelled by MOCAGE with those observed at the EMEP network stations (Figure 12 on the old version of the manuscript). The new graph has been integrated to the new manuscript, simplifying the comparison between the model and the observations. Eastern Europe is the most polluted area in Europe, because of strong emissions of SO2. Unfortunately, there are few EMEP stations in this region, making it difficult to conclude whether the modelled pattern of the sulphate ground concentration is realistic or not there. Nevertheless, the couple of EMEP stations existing in this region help us to evaluate the performances of our model : The spatial agreement between the model and observations seems to be quite good over our domain, whereas the model has a general tendency to overestimate the sulphate ground concentration.

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3. Response to the minor comments

As the first anonymous referee pointed out, the contribution of sedimentation to the total dry deposition of sulphate is negligible. Consequently, we do not detail the sedimentation process in the new version of our manuscript.

The two studies of Redington and Derwent (Atmos. Environ. 36, 4425-4439, 2002) and Schaap et al. (ACP 4, 857-874,2004) present an analysis of aerosol simulations over Europe. They focus mainly on nitrate, but they also show a validation of the representation of sulphate in the models they used. As recommended by the first anonymous referee, we cite these two references in the introduction of our new manuscript.

The statement "There are two areas in Europe where the AEROCOM emissions of sulphur compounds are important" (page 4390, lines 13-14) does not mean that some emissions do not originate from AEROCOM in our simulation. It means that two regions are concerned by strong emissions in the AEROCOM emissions inventory (Southern and Eastern Europe). In the new manuscript, this statement is replaced by "The AEROCOM inventory is characterised by strong emissions over two regions : Eastern Europe and Southern Europe".

As pointed out the first referee, we have to be careful with the statement "Regionally, the year to year variability of the monthly mean aerosol burden can reach 100% because of different weather conditions" (page 4391, lines 7-8). A 100% would mean doubling the burden, while a 100% decrease would mean that the burden is zero in the latter case. Consequently, this sentence is substituted in the new manuscript by the statement "Regionally, the monthly mean aerosol burden can vary by a factor of 2 from one year to another, because of different weather conditions".

4. Response to the technical comments

The red lines in Fig. 2 have been obtained from a simple fit to experimental data presented in the work Kasper-Giebl (2000). The equations (4) and (5) were obtained

from this simple fit.

The sulphur burden curve of Fig. 3 is converted from Tg[S] to mg[S].m-2 in our new manuscript. This new unit is then in concordance with the text of section 3.1.

To facilitate the comparison between the sulphate burden (Fig. 3), the precipitation (Fig. 4a), the temperature (Fig. 4b) and all sulphur fluxes (Fig. 5), the curves describing these variables were all included in the same figure.

The colour scale of the figures 6 and 11 were modified to improve the visualisation of the spatial differences between the different graphs.

According to the suggestion of the first anonymous referee, the winters 2001-2002 and 2003-2004 were called winter 1 (W1) and winter 2 (W2), both in the text and in figure 9.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 4381, 2009.

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