

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

**M. Ménégóz et al.**

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First of all, we want to express our sincere thanks to the second anonymous referee who has read our manuscript very carefully. We will revise our manuscript according to his/her comments. Scientific comments are exposed through 5 different points. Our response presented in the following exposes 5 parts discussing these points, and a last part is devoted to the technical comments.

1) Representation of the different types of aerosols in the model

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

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some aerosols are not considered in our model. In particular, the representation of sea-salt and organic aerosol is not yet 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). Only 20% of non sea-salt sulphate is internally mixed with other aerosols, the rest evolving independently from the other aerosols (Liu et al, 2005). In contrast with sulphate aerosol, black-carbon and organic carbon are generally mixed with other chemical species. In the future, modelling these aerosols should necessitate to represent the mixing between the different species, which can strongly affect their lifetime.

## 2) Improving the sulphate representation in the model.

In the introduction of the first version of our manuscript, we wrote "The first step when studying the impact of aerosols on climate - and how it may change due to human activities - is to describe the distribution of natural and anthropogenic aerosols over the globe." The second referee has suggested to clarify the impact of a better representation of sinks and sources of sulphate on the evaluation of the direct and indirect aerosol forcing with models. The following was added in the new version of our manuscript : "Sulphate is an aerosol which strongly affects the radiative balance of the atmosphere (eg. Twomey et al., 1977, Charlson et al., 1992). The sulphate forcing is strongly dependent on sulphate burden and distribution (Koch et al., 1999). According to the work of Kasibhatla et al. (1999), it is strongly linked with the seasonal variation of the atmospheric column sulphate burden. It is also dependent on the vertical distribution of this aerosol (IPCC, 2007). Studying the sinks and sources of sulphate helps to better estimate the changes in time of the spatial distribution of this aerosol, and finally helps to estimate the sulphate forcing". In the future, such a study could be used to make a simple parameterisation in a climate model describing the aerosol concentration and then the aerosol forcing. It could be also used as a basis to estimate the forcing of other aerosols on the radiative balance of the atmosphere.

### 3) Description of the "bins representation" for dust, BC and sulphate.

The AEROCOM inventory used in our study describes both the quantity of aerosols and precursors gases emitted in the atmosphere and the geometry of aerosols. This inventory also 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  $\mu\text{m}$  for dust, and 4 bins from 0.001  $\mu\text{m}$  to 10  $\mu\text{m}$  for sulphate and BC). 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.

### 4) Aerosol chemistry in our model

BC and dust are chemically inert in the atmosphere (IPCC, 2007). For this reason, these aerosols are directly emitted in the atmosphere, without being involved in chemical reactions in our model. This point is added in our new manuscript (paragraph 2.2).

### 5) Dust emissions

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).

### 6) Technical comments :

- The contribution of sedimentation to the total dry deposition of sulphate is negli-

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ble. Consequently, we do not detail the sedimentation process in the new version of our manuscript, and equation 1 is omitted in the new version of our manuscript. - As recommended by the second referee, the statement "We have to keep in mind that..." (paragraph 2.4) is substituted by "We have to underline that..." - The sentence "We can see an important annual cycle" is replaced by a clearer statement : "The sulphate burden shows a significant annual cycle". - The statement "There are two areas in Europe where the AEROCOM emissions of sulphur compounds are important" (page 4390, lines 13-14) is replaced in the new manuscript by "The AEROCOM inventory is characterised by strong emissions over two regions: Eastern Europe and Southern Europe". - As explained in our response to the first referee, we have improved the quality of different figures (figure 6b-6c-6d) in the new manuscript that we have submitted.

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