Atmos. Chem. Phys. Discuss., 9, S737–S740, 2009
www.atmos-chem-phys-discuss.net/9/S737/2009/

Atmospheric Chemistry and Physics Discussions

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.

Anonymous Referee #1

Received and published: 11 March 2009

General:

This paper presents a six-year-long simulation of the global sulfur cycle using meteorological fields obtained from ECMWF operational analyses. The paper focuses on studying the sources and sinks of sulfate aerosols over Europe at different temporal scales (seasonal and year-to-year variability). The topic of this paper is very important, yet very few studies of this kind have been conducted earlier. However, in spite of its scientific importance and originality, I feel that he paper require some major improvements before I can recommend its acceptance of publication in ACP.

Major comments:

1. Representation of the aerosol size distribution and treatment of aerosol emissions in the model should be described in more detail. Are all three simulated aerosol types assumed to be expternally mixed? I suppose so but has not been mentioned in the text. What are the borders between size bins? Are they of equal "width" for each aerosol type, or do the try to mimic the general "modal" behavior of the ambient aerosol. Why the size of BC has been extended down to 1 nm, although very little BC exists below about 50 nm in the atmosphere? How the AEROCOM emissions have been distributed between different size bins in practice?

2. The model misses two major aerosol components, sea salt and organic aerosols. Both of them have been included in most of the current large-scale aerosol models. The authors should discuss the possible implications resulting from this omission.

3. If sulfate is treated as its own aerosol type, how is the formation of the sulfate aerosol treated in the model (see the above comment 1)? How is the sulfate produced in the gas phase or in clouds distributed over different size bins? Is a certain fraction of sulfur emissions assumed to be emitted as primary sulfate particles?

4. A few models studies have investigated the role/importance of aerosol microphysics when simulated global sulfate, including the mixing of sulfate with other aerosol types (e.g. Liu et al. JGR 110, D18206, 2005; Ma and von Salzen JGR 111, D08206, 2006; Trivitayanurak et al. ACP 8, 3149-3168, 2008). This issue should be discussed in the paper (see also comments 1 and 3 above).

5. Dust emissions depend crucially on wind speed and may therefore vary significantly from year to year. In this paper, no year-to-year variation is dust emissions has been taken into account when simulating the year-to-year variability of sulfate aerosols. This should be brought up explicitly in the paper. Since the model uses real-time wind fields, a mechanistic dust emissions scheme that depends on local wind speed could have been employed. Was there some reason not to use such scheme instead of prescribed emissions?

ACPD

9, S737–S740, 2009

Interactive Comment

6. The relation between sulfate burden and precipitation certainly depends on the averaging period (hourly, daily, monthly etc). This should be emphasized when discussing the relation between these two quantities (section 3.1). Visually, the relation would be observed more easily if the curves in Figure 3 and 4a were plotted in the same figure.

7. I am not very convinced on how much can be said about the performance of model based on Figure 12. Firstly, it seems that almost all measurement points are of the same color in the used color scale. Second, there are very few measurement points in the region where the model predicts largest gradients in sulfur burden (Eastern Europe).

Minor comments

Sedimentation gives very minor contribution to the dry deposition simulated aerosols (except the largest dust aerosols). As a result, I see no point of giving equation 1 explicitly in the paper.

Distribution of sulfate aerosol over Europe has been modeled also by Redington and Derwent (Atmos. Environ. 36, 4425-4439, 2002) and Schaap et al. (ACP 4, 857-874, 2004). These studies should be mentioned in the manuscript.

I do not understand the following statement on page 4390 (lines 13-14): "There are two areas in Europe where the AEROCOM emissions of sulphur compounds are important." I thought that all sulfur emission in the model originate from AEROCOM.

Please be careful when talking about 100% variability in sulfate column burden (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.

Technical comments:

Please explain how red lines in Fig. 2 have been obtained. They seem to be from equations 4 and 5, but how these equations have been obtained? A simple fit to experimental data?

ACPD

9, S737–S740, 2009

The authors use different units for the sulfur burden between section 3.1 and Figure 3. This is very inconvenient for the reader. Please either unify or give the conversion factor between the two units.

By looking at figures 4a and 5, I do not think that the reader can very easily see that precipitation and production by aqueous phase chemistry are correlated, as stated in the text (page 4389, lines 3-6).

The scales of figures 6b-d (especially 6c) has been chosen in such a way that it is difficult to see spatial differences. I would recommend using a different concentration scale. Roughly the same holds for Figure 11.

Figure 9 gives results from two winters (December-January). Since these two time periods are split between 4 years, it might be easier to call them winters 1 and 2 rather than using different years.

There are some grammatical problems throughout the text.

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