

Answers to Patricia Castellanos comments, received and published on 12 October 2013, on the manuscript:

“Evaluation of GEOS-5 sulfur dioxide simulations during the Frostburg, MD 2010 field campaign”

We thank the reviewers for providing comments that helped to improve the quality of the paper. The detailed responses to comments are listed below (text in black shows comments from the reviewers, and the text in blue is our answer):

The manuscript presents an evaluation of SO₂ and sulfate concentrations from the GEOS-5 model with surface and aircraft observations. The authors found that correcting the injection height of power plant SO₂ emissions improved the comparison of model SO₂ concentrations to EPA ground-based measurements. However, there continued to be a positive bias in surface SO₂ and sulfate concentration, which leads the authors to conclude that the loss of sulfate may be underestimated in the model.

Overall the manuscript is clear, concise, and appropriate for publication in ACP.

The main comment is regarding the conclusion that sulfate aerosol losses may be underestimated. It's difficult to see this from the data presented because the 2005 emissions used in the model are likely too high for 2010. This is supported by the high bias apparent in Fig 5, and the authors state this on page 21773 line 1. Could the high bias in sulfate also be attributed to the overestimated emissions - despite the high bias in SO₂ lifetime? Would it be possible to scale the 2005 emissions to 2010 using CEM data? Or compare 2005 surface observations to a model simulation using 2005 meteorology? If sulfate is still overestimated then I think you will have a stronger argument.

Following P. Castellanos and the reviewer comments, a new comparison has been performed for the year 2005 between ground-based EPA sulfate measurements and GEOS-5 simulated sulfate. A positive bias remains in the comparison but lower than the one observed for the year 2010. The positive bias in sulfate might also be attributed to the overestimated SO₂ emissions for 2010. Following this new analysis the text in section 3.2 “Sulfate aerosol” p 21773 has been updated.

As a minor comment, the section describing the comparison to MF-DOAS observations could use some more description. For example, it's not clear how the air mass factor is calculated to get the vertical column, what is the estimated spatial (horizontal and vertical) footprint of the observation, and what is the temporal resolution. All important factors when trying to reconcile model and measured concentrations.

The section 4.2 has been extended to include more details: “Analysis of the measured spectra is done using the DOAS technique which is based on the Beer–Lambert law (BLL). Full details of MFDOAS instrument as well as the DOAS analysis of SO₂ used in this study can be found in Spinei et al. (2010). DOAS analysis consists of two steps: (1) calculation of differential slant column density (Δ SCD) along the average photon path relative to the reference spectrum using BLL and (2) conversion of Δ SCD to vertical column density (VCD) using air mass factors (AMF). In this study we present only total

vertical columns from direct sun irradiance measurements (DS). SO₂ ΔSCD were derived from 307 – 327 nm wavelength window by simultaneous fitting of the following molecular absorption cross sections: O₃ (228 and 243K, Daumont et al 1992, Brion et al 1993, Malicet et al 1995), SO₂ (298K, Vandaele et al., 2009), NO₂ (270K, Vandaele et al., 1998). In addition, 3rd order polynomial was fitted to remove broadband extinction due to aerosol and molecular absorption and scattering. Direct sun reference spectrum used in DOAS fitting was measured by MFDOAS around local noon on 11-Nov-2010 (30 min average). DS AMF were calculated based on geometrical estimation (see Spinei et al. 2010) and approximately equal to 1/cos(solar zenith angle (SZA)). DS AMF has very low sensitivity to the species profile at solar zenith angles < 80°. Since SO₂ ΔSCDs were determined using ground-based reference spectrum, estimation of SO₂ amount in it is needed to convert to VCD. This is done by using minimum Langley extrapolation method (Cede et al., 2006, Herman et al., 2009), where only the smallest SO₂ ΔSCDs in AMF bins are plotted against DS AMF to extrapolate to AMF equal zero (extraterrestrial). SO₂ VCDs are then calculated by adding SCD in the reference spectrum to the ΔSCDs and dividing by DS AMF. Footprint of the measurements is determined by the solar position (zenith and azimuth angles) and PBL height where most of SO₂ is located. According to backscatter LIDAR at 355 nm, PBL heights during DS measurements were on average 900±95 m and SZAs ranged from 54° to 80°. This translates to horizontal footprint of about 1.2 km during high sun and 2.8 km during low sun hours following the sun in azimuthal direction from about 120° to 240° (from North).”