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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The role of long-range transport and domestic emissions in determining atmospheric secondary inorganic particle concentrations across the UK" *by* M. Vieno et al.

Anonymous Referee #3

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This paper applies the EMEP4UK regional atmospheric chemistry transport model to evaluate the roles of long-range transport and domestic emissions during the 2001-2010 decade in determining the secondary inorganic particle concentrations across the UK. Modeled surface concentrations are evaluated with the long-term observational data from four representative sites of the UK AGANet monitoring network, which provided simultaneous measurements of SO2, NOx, NH3 gases and particulate NO3-, SO4=, and NH4+. Modeled monthly average particulate concentrations are in general agreement with the observed values for the decade. Model analysis of 2003 suggested that 40-60% of the monthly average total inorganic particulate mass at the UK sites was

due to long-range transport from continental Europe. The paper is generally well written, but the analysis seems a bit hurried and lacks the depth necessary to figure out the roles of different mechanisms that govern the inorganic aerosol concentrations. The manuscript can be recommended for publication in ACP after the following issues are carefully addressed.

Specific Comments:

1) Authors refer to Simpson et al (2012) for the description of the EMEP model and state that the gas-particle partitioning is done by EQSAM formulation (Metzger et al., 2002a,b). After reading these two papers, I find that EMEP simulates aerosols with two size modes (designed to calculate PM10 and PM2.5) while EQSAM is essentially a bulk aerosol equilibrium solver that does not treat size-dependent gas-particle partitioning. This is a gross oversimplification that is prone to large errors, especially in the modeled long range transport of semi-volatile species such as NO3-, Cl-, and NH4+ as they interact with sea-salt during transit from continental Europe to the UK. It has been shown that a fully dynamic mass transfer treatment is needed to accurately perform gas-particle partitioning to fine and coarse mode particles (Hu et al., 2008) and a considerable amount of effort has been put into developing such schemes (e.g., Jacobson, 2005; Zhang and Wexler, 2006; Zaveri et al., 2008). Also, the fundamental basis of the EQSAM thermodynamics model formulation itself appears to be fraught with serious issues (e.g., see interactive discussions for Metzger and Lelieveld, 2007; Xu et al., 2009; Metzger et al., 2010). The authors therefore need to clearly defend their choice of thermodynamic model as well as the bulk equilibrium approach, especially since the focus of this paper is on the role of long-range transport vs. domestic sources of inorganic aerosols, of which NO3- and NH4+ are shown to be major components based on observations.

2) Model predictions for both gas and particle phase observations of the three inorganic species (sulfate, nitrate, and ammonium) should be evaluated to ascertain the accuracy of the simulation.

3) Since EMEP simulates two size modes, it would be very useful to show the relative importance of the modeled inorganic species in the two modes (broken down by aerosol types - primary dust, primary sea-salt, and secondary) to the overall contribution.

4) The model perturbation study (turning off UK emissions) should be expanded to the entire decade to examine the variability in the relative contributions from domestic and long-range transport sources of inorganic aerosol.

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