Background & Motivation

The effect aerosols have on climate and air quality is a function of their chemical composition, concentration and spatial distribution. These parameters are controlled by emissions, heterogeneous and homogeneous chemistry, where thermodynamics plays a key role, transport, which includes stratospheric-tropospheric exchange, and depositions sink. In this work we demonstrate the effect of some of these processes on the SO2-NOx-NH3 system using the GISS ModelE2 Global Circulation Model (GCM).

- Motivation: NOx aerosol is poorly constrained throughout the troposphere, especially above surface level.
- Mission: Bridge this knowledge gap with a collection of surface and airborne data and model results.
- Relevant studies: Bauer et al., 2007, Bellouin et al., 2011, Aan de Brugh et al., 2012, Hauglustaine et al., 2014, Paulot et al., 2015

Objective: evaluate the GISS ModelE2 aerosol schemes and pin point key process either included or missing in the model

How does NOx aerosol form?

1. GAS PHASE
SO2 + H2O \rightarrow SO2-H2O + O2

2. Dissociation into ions
H2SO4 \rightarrow H^+ + SO4^-2

3. Dissociation into ions
NH3 \rightarrow NH4^+ + H^-

4. Ionic solution – salt equilibrium
H2SO4 \rightarrow NH4SO4 + H2O

The phase is controlled by:
- Precursor abundance
- Ambient Relative Humidity (HR)
- Temperature

What are the major sources of nitrate and sulfate precursors?

- Fountoukis, Bauer et al. (2007a), Fully interactive trop and str chemistry
- RCP4.5 (2005 onwards)
- RCP8.5 (2005 onwards)
- CMIP – RCP4.5 (2005 onwards)
- Biosphere burning: GFD3
- Agricultural NH3 imposed seasonality according to zenith angle

GISS Aerosol Schemes

- MATRIX: microphysics model, tracks mixing stage
- OMA: bulk aerosol, includes heterogeneous uptake on dust
- ESDAM: parameterized nitrate absorptive
- ISOROPA: calculates the thermodynamics

Sensitivity runs: (Result 3) we test the sensitivity of NH3, NH4, HNO3, NO2 doubling and fivefold increase in agricultural NH3 emissions

Data and Methods

Monthly mean surface data of SO2, NH3, NH4, NO2 measured via the IMPROVE (USA) and EMEP-NL EU (networks during 2000-2010 is used to compare against the simulations. From the climatological mean (Figure 4) we adopt a regional approach (black frames in Figure 3). The flights were predominantly during spring and summer time and deployed the AMS instrument. Within a regional approach we parse out transect for flights and for flights within the Arctic, USA, WIGA regions we use the data within the regional boundaries to calculate a campaign mean per model layer. We sample our simulations according to the flight location.

References


Results

Is there a spatial pattern (Figure 4)? Surface concentration shows high concentrations in EUSA, EU and low concentrations in WUSA.

The statistics shows (Figure 5):
- Performance is controlled by region more than aerosol scheme
- Systematic underestimation of aerosols in EUSA, EU
- Big differences for SO2 with microphysics (MATRIX VS OMA)
- Overall good performance by the GCM (R=0.5)

NO2 annual cycle (Figure 6):
- Seasonality is reproduced in EUSA, EU
- Summer underestimations in all regions

Figure 5: (below) Surface regional statistics (2000-2010), correlation coefficient (R) and normalized mean bias (NMB) for the three simulations.

Take away:
- Missing aerosol mass, especially above the surface, could have important implications to aerosol radiative forcing
- Good correlations (R>0.5) at surface in regions where seasonality is reproduced
- HNO3 is sensitive to the heterogeneous sink – an important process to include in models
- HNO3 and NO2 partitioning is strongly dependent on NH3 partitioning
- Need for more assessments: few campaigns measured NH3 (CALNEX, TexAQ), no winter campaigns