Re-analysis of tropospheric aerosols for the period 1980-2005 using ECHAM5-HAMMOZ

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1.OBJECTIVES

- Understanding historical trends of trace gas and aerosol distributions in the troposphere is essential to evaluate the efficiency of existing strategies to reduce air pollution and to design more efficient future air quality and climate policies.
- We performed simulations for the period 1980–2005 using the aerosol-chemistry-climate model ECHAM5-HAMMOZ, to assess our understanding of long-term changes and inter-annual variability of the chemical composition of the tro-posphere.
- We separated the impact of the anthropogenic emissions and natural variability on atmospheric chemistry.

2.METHOD

- 2 simulations of the period 1980–2005 with ECHAM5-HAMMOZ:
  1. SREF: changing anthropogenic emissions
  2. SFIX: fixed anthropogenic emissions (year 1980).
- T42: ~ 2.8° x 2.8 °; 31 vertical levels (surface to10 hPa). Nudging, ERA-40 & IFS32r2 ECMWF.

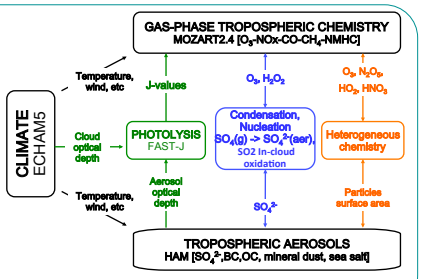

Fig. 1: ECHAM5-HAMMOZ model

3.EMISSIONS

- Anthropogenic emissions of gas species from RETRO (1980-2000). Trends derived from EMEP, USEPA, and REAS and applied for the period 2001-2005 over Europe (EU), North America (NA), East Asia (EA), and South Asia (SA). Fig. 2.
- Anthropogenic emissions of SO2, BC, and OC from AEROCOM (1980-2005). Fig. 2.
- Wildfire emissions from RETRO and van der Werf. Fig. 3f.
- On-line biogenic emissions (MEGAN), NOx lightning, DMS, sea salt, and mineral dust. Fig. 3.

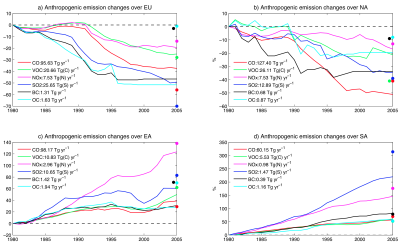

Fig. 2: Percentage changes in total anthropogenic emissions from 1980 to 2005 over **(a)** Europe, EU; **(b)** North America, NA; **(c)** East Asia, EA; **(d)** South Asia, SA.The total annual emissions are reported for the year 1980. The colored points represent the percentage changes between 1980 and 2005 in Lamarque et al. (2010).

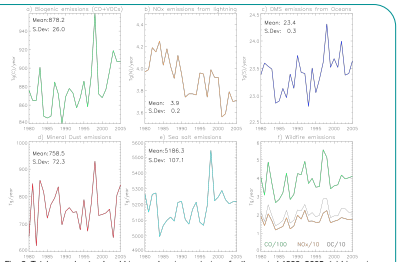

Fig. 3: Total annual natural and biomass burning emissions for the period 1980–2005: (a) biogenic CO and VOCs emissions from vegetation; (b) NOx emissions from lightning; (c) DMS emissions from oceans; (d) mineral dust aerosol emissions; (e) marine sea salt aerosol emissions; (f) CO, NOx, and OC aerosol biomass burning emissions.

4.GLOBAL AOD AND SULFATE BUDGET

- The global annual average AOD ranges between 0.151 and 0.167 during the period 1980– 2005 (SREF). Fig. 4a.
- The anthropogenic emissions decraese AOD over a large part of the Northern Hemisphere, -0.2 over Eastern Europe, and increase AOD over East and South Asia (+0.2). Fig. 4b.
- The monthly mean anomalies of surface sulfate concentrations are largely influenced by anthropogenic emissions, while anomalies of AOD are determined by variations of natural aerosol emissions, including biomass burning (Fig. 5).
- The variability of the global sulfate burden is largely determined by meteorology. The contrasting temporal trends of gas-phase and in-cloud sulfate production can be explained by the changes in the geographical distribution of the emissions (Fig. 6).

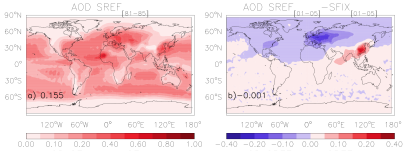

Fig. 4: Maps of total aerosol optical depth (AOD) and the changes due to anthropogenic emissions. We show (a) the 5-yr averages (1981–1985) of global AOD; (b) the effect of anthropogenic emission changes in the period 2001–2005 on AOD, calculated as the difference between SREF and SFIX simulations.

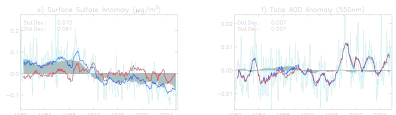

Fig. 5: Monthly mean anomalies for the period 1980–2005 of globally averaged fields for the SREF and SFIX ECHAM5-HAMMOZ simulations. Light blue lines are monthly mean anomalies for the SREF simulation, with overlaying dark blue giving the 12 month running averages. Red lines are the 12 month running averages of monthly mean anomalies for the SFIX simulation. The grey area represents the difference between SREF and SFIX. (e) sulfate surface concentrations (μg m−3); (f) total aerosol optical depth.

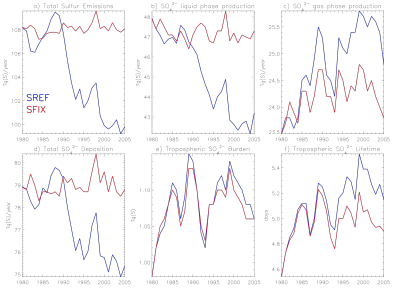

Fig. 6: Global tropospheric sulfate budget calculated for the period 1980–2005 for the SREF (blue) and SFIX (red) ECHAM5-HAMMOZ simulations: (a) total sulfur emissions; (b) sulfate liquid phase production; (c) sulfate gaseous phase production; (d) surface deposition; (e) sulfate burden; (f) lifetime.

5.REGIONAL SULFATE

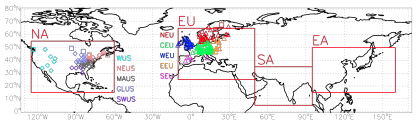

Fig. 7: Map of the selected regions for the analysis and measurement stations with long records of sulfate surface concentrations. North America (NA), Europe (EU), East Asia (EA), and South Asia (SA). Triangles show the location of EMEP stations, squares of WDCGG stations, and diamonds of CASTNET stations. The stations are grouped in sub-regions: Northern Europe (NEU); Central Europe (CEU); Western Europe (WEU); Eastern Europe (EEU); Southern Europe (SEU); Western US (WUS): North-Eastern US (NEUS); Mid-Atlantic US (MAUS); Great lakes US (GLUS); Southern US (SUS).

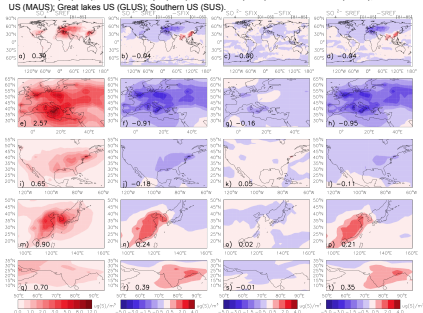

Fig. 8: Maps of surface sulfate concentrations and the changes due to anthropogenic emissions and natural variability. In the first column we show 5 yr averages (1981–1985) of surface sulfate concentrations over the selected regions, Europe, North America, East Asia, and South Asia. In the second column (b) we show the effect of anthropogenic emission changes in the period 2001–2005, calculated as the difference between SREF and SFIX simulations. In the third column (c) the natural variability of sulfate concentrations is shown, which is due to natural emissions and meteorology in the simulated 25 yr, calculated as the difference between the 5 yr average periods (2001–2005) and (1981–1985) in the SFIX simulation. The combined effect of anthropogenic emissions and natural variability is shown in column (d) and it is expressed as the difference between the 5 yr average period (2001–2005)–(1981–1985) in the SREF simulation.

- Europe: Sulfate concentrations decreased by ~ 35% due to sulfur emission controls. The natural impact is small but significant.
- North America: The emissions reductions of 35% reduced sulfate concentrations on average by 0.18 μg(S) m^{-3}, and up to 1 μg(S) m^{-3} over the Eastern US. Meteorological variability results in a small overall increase of 0.05 μg(S) m^{-3}.
- East Asia: Growing anthropogenic sulfur emissions (60%) produced an increase in regional annual mean concentrations of 37%. Natural variability is small.
- South Asia: sulfate concentrations increased by 56% due to increasing anthropogenic sulfur emissions (220%)
- Modeled and measured sulfate trends are in good agreement (Fig. 9), but a poor representation of the emissions seasonality contributes to the discrepancies of the sulfate trends.

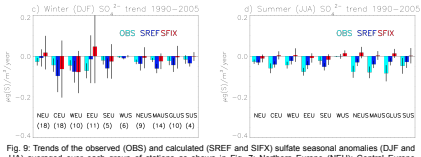

Fig. 9: Trends of the observed (OBS) and calculated (SREF and SIFX) sulfate seasonal anomalies (DJF and JJA) averaged over each group of stations as shown in Fig. 7: Northern Europe (NEU); Central Europe (CEU); Western Europe (WEU); Eastern Europe (EEU); Southern Europe (SEU); Western US (WUS): North-Eastern US (NEUS); Mid-Atlantic US (MAUS); Great lakes US (GLUS); Southern US (SUS). The vertical bars represent the 95 % confidence interval of the trends. The number of stations used to calculate the average seasonal anomalies for each subregion is shown in parentheses.

6.CONCLUSIONS

- Globally, anthropogenic OC emissions increased by ~10%, while sulfur emissions decreased by ~10 % from 1980 to 2005, but regionally changes are larger, 10–50 % in NA and EU, but increased between 40–220 % in EA and SA.
- Small global inter-annual variability for DMS emissions (1%) and sea salt aerosols (2%). A larger variability was found for mineral dust (10 %).
- The global inter-annual variability of surface sulfate (10%) is strongly determined by regional variations of emissions.
- Comparison of computed trends with measurements in Europe and North America showed in general good agreement.
- Despite a global decrease of sulfur emissions from 1980 to 2005, global sulfate burdens were not significantly changing, due to a southward shift of SO2 emissions, which determines a more efficient production.
- Globally AOD is more influenced by natural varibility. Regionally we found a decline of 28% for EU, and an increase of 19% and 26% for EA and SA.
- Further details: Pozzoli et al., Atmos. Chem. Phys., 11, 9563-9594, 2011