Evaluating Ammonium, Nitrate and Sulfate Aerosols in 3-Dimensions

National Aeronautics and Space Administration
Goddard Institute for Space Studies
New York, N.Y.

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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 depositional sinks. In this work we demonstrate the effect of some of these processes on the SO_4-NH_4-NO_3 system using the GISS ModelE2 Global Circulation Model (GCM).

- **Motivation:** NO_3 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 aerosol models.
- **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

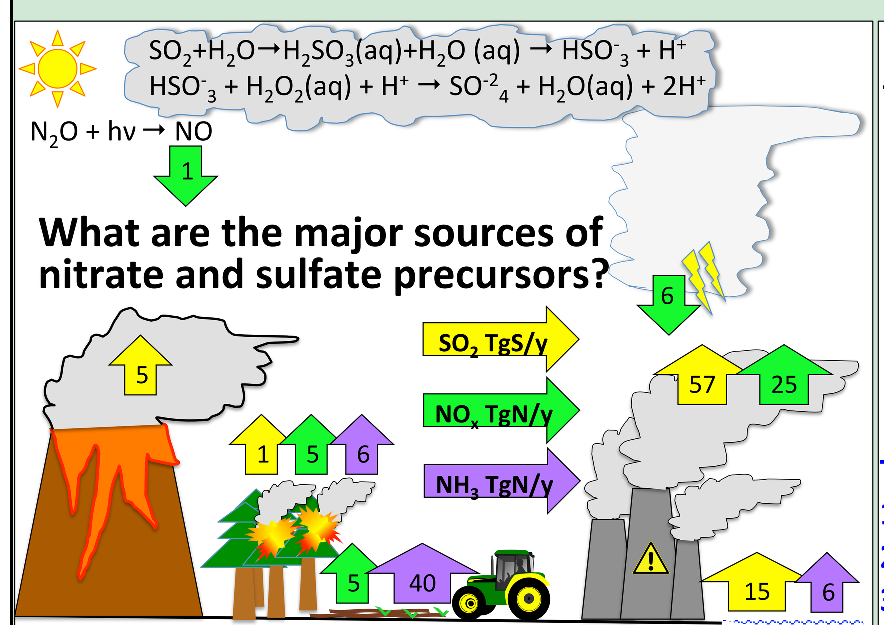


Figure 1: precursor sources and emissions (a compilation of references)

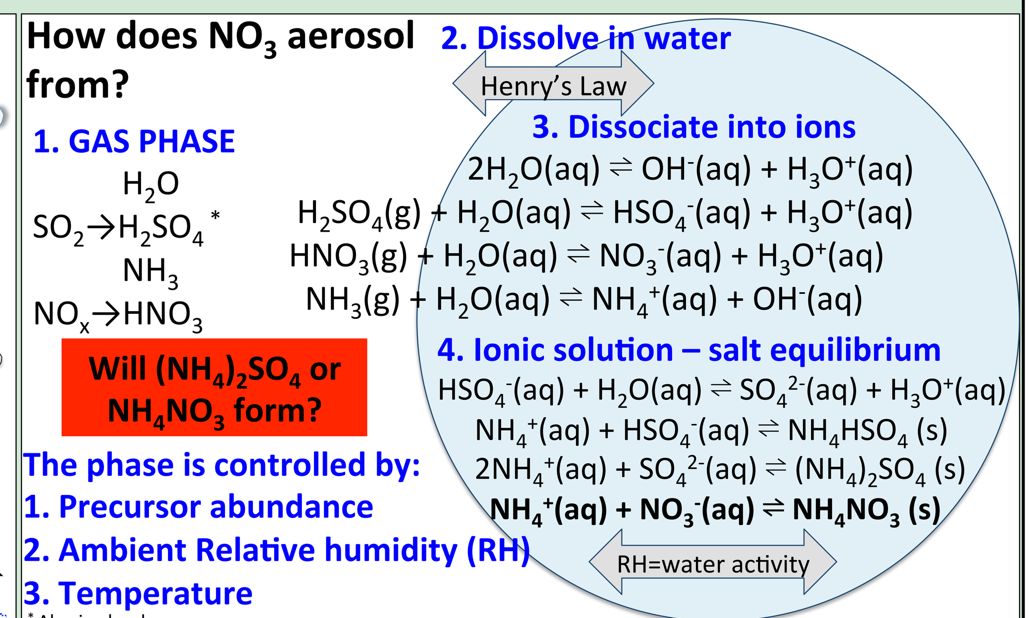


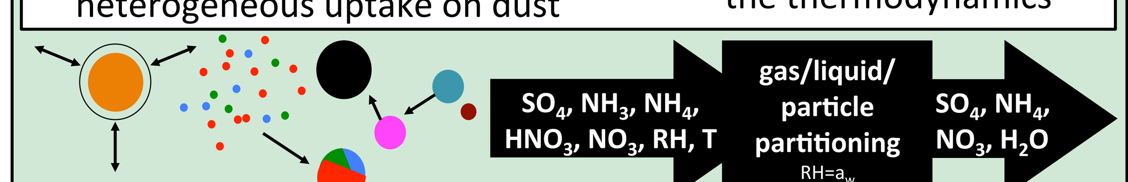
Figure 2: The processes controlling NO_3 formation

Setup:

- 2° x 2.5° resolution, 40 vertical layers
- Fully interactive trop and strat chemistry
- Horizontal wind nudged: 6-hourly NCEP
- SST and Ice Cover prescribed from obs
- Emissions:
  - CMIP5
  - RCP4.5 (2005 onwards)
  - Biomass burning: GFED3
  - Agricultural NH_3 imposed seasonality according to solar zenith angle

GISS Aerosol Schemes

- **MATRIX**: microphysics model, tracks mixing state
- **OMA**: bulk aerosol, includes heterogeneous uptake on dust
- **EQSAM**: parameterized thermodynamics
- **ISORROPIA II**: calculates the thermodynamics



Sensitivity runs: (Result 3) we test the sensitivity of NH_3, NH_4, HNO_3, NO_3 to doubling and fivefold increase in agricultural NH_3 emissions.

Data and Methods

Monthly mean **Surface** data of SO_4, NH_3, NH_4, NO_3 measured via the IMPROVE (USA) and EMEP-NILU (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 Figures 3,4), where the mean, standard deviation, normalized mean bias and correlation coefficients are calculated for the stations within a region along with their matching model grid boxes.

During 2001-2011, 14 **flight campaigns** took place in the NH and measured SO_4, NH_4, HNO_3, NO_3 (Figure 3). The flights were predominantly during spring and summer time and deployed the AMS instrument. With a regional approach we parse out transit flights and for flights within the ARC, EUSA, WUSA 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.

Arctic (ARC)	[55°-90°N, 60°-170°W]
Eastern USA (EUSA)	[30°-50°N, 60°-95°W]
Western USA (WUSA)	[30°-50°N, 114°-130°W]
European Union (EU)	[35°-70°N, 10°W-30°E]

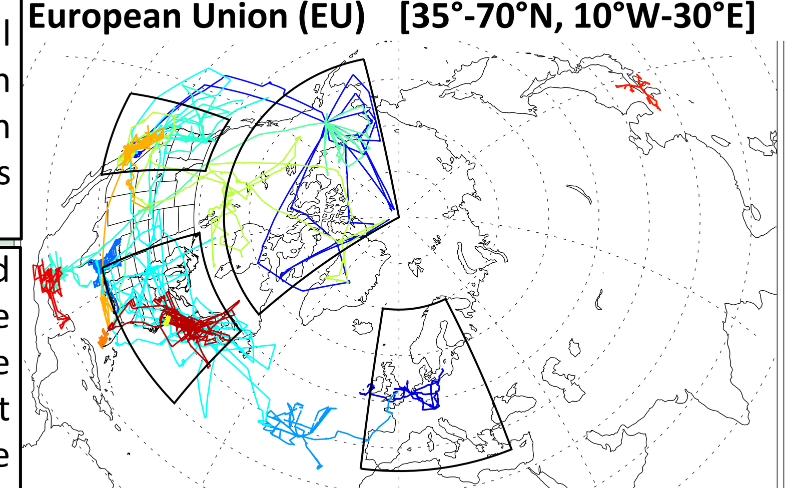


Figure 3: (above) Flight tracks of 14 flight campaigns used in this study

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Results

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

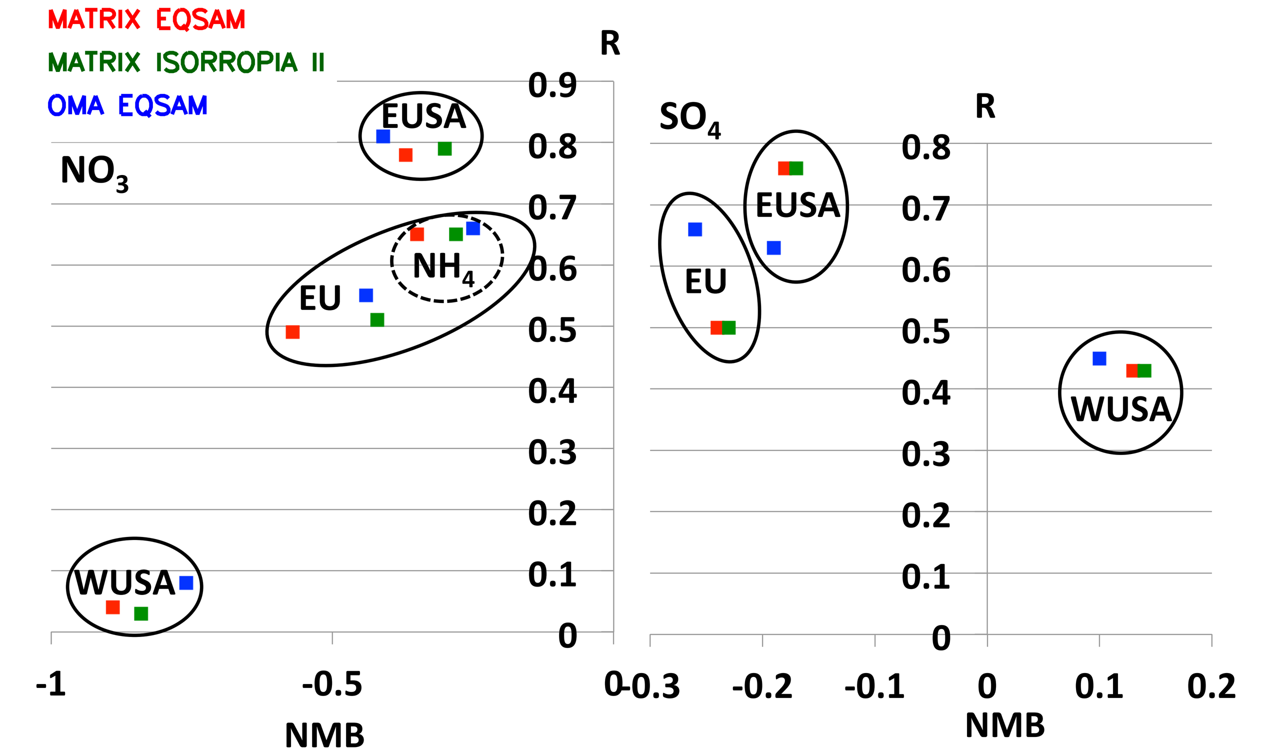
The statistics shows (Figure 5):

- Performance is controlled by region more than aerosol scheme
- Systematic underestimation of aerosols in EUSA, EU
- Big differences for SO_4 with microphysics (MATRIX VS OMA)
- Overall good performance by the GCM (R>0.5)

NO_3 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.



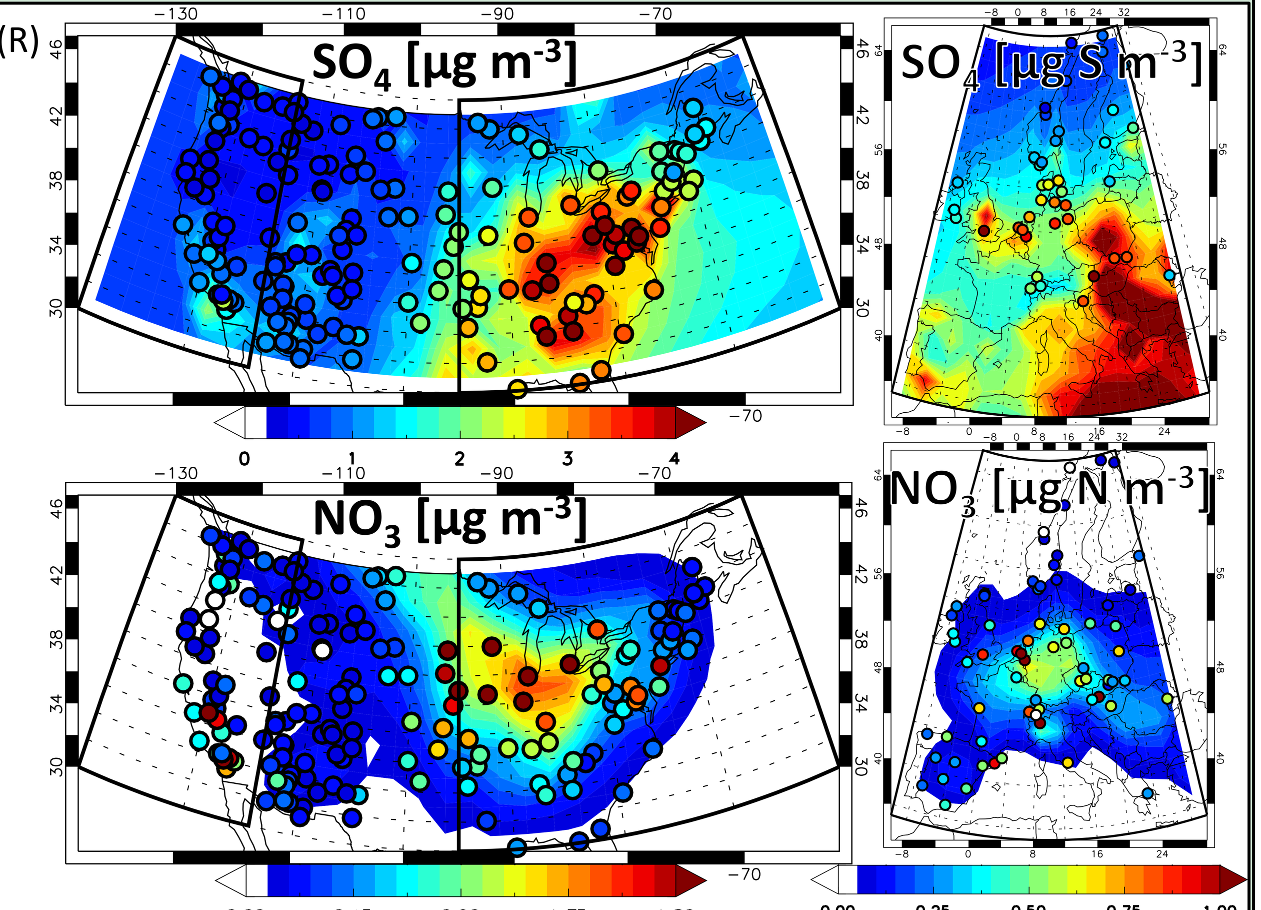
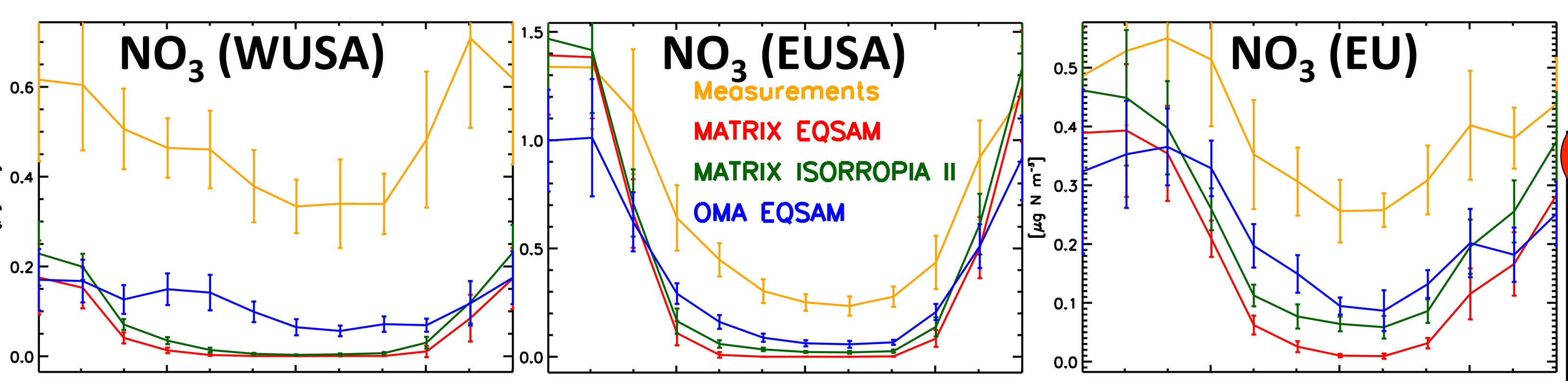


Figure 4: (above) Mean surface concentration (2000-2010) simulated by MATRIX-EQSAM overlaid by measurements

Figure 6: (right) 2000-2010 NO_3 mean annual cycle. Error bars represent ± one standard deviation.



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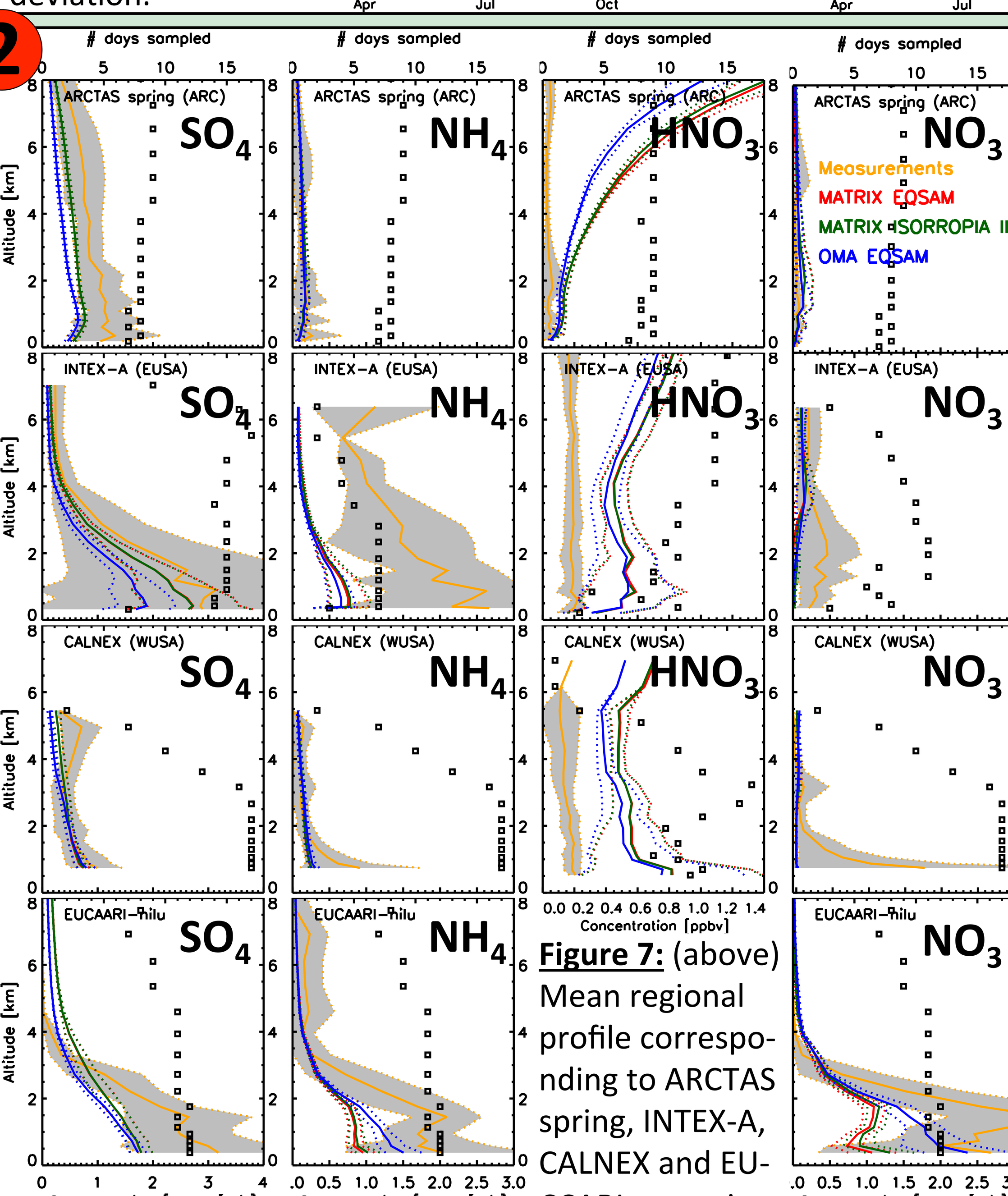

Figure 7: (above) Mean regional profile corresponding to ARCTAS spring, INTEX-A, CALNEX and EU-CCARI campaigns

Is the vertical consistent with the surface?

- Model underestimation of aerosol concentration, especially in the boundary layer, more for NH_4, NO_3
- SO_4: MATRIX simulations (microphysics included) show higher concentrations than the bulk scheme (OMA). Thermodynamic scheme makes a minor difference: green and red lines overlaping each other.
- The model overestimates upper tropospheric HNO_3: observed and attributed in previous studies to overestimation of strat-trop exchange.
- Missing processes? (1) Heterogeneous chemistry sink on dust surfaces included in OMA, yields lower concentrations. (2) Homogenous chemistry: (i) Crustal (Mg, K, Li) and sea salt (Na, Cl) ions are not part of the thermodynamics implemented in the model. (ii) Thermodynamics do not take into account the different time scales associated with particle size distribution. (iii) Organic nitrate formation, especially important in the summer over forested areas.
- Uncertainty in NH_3 emissions (go to 3).

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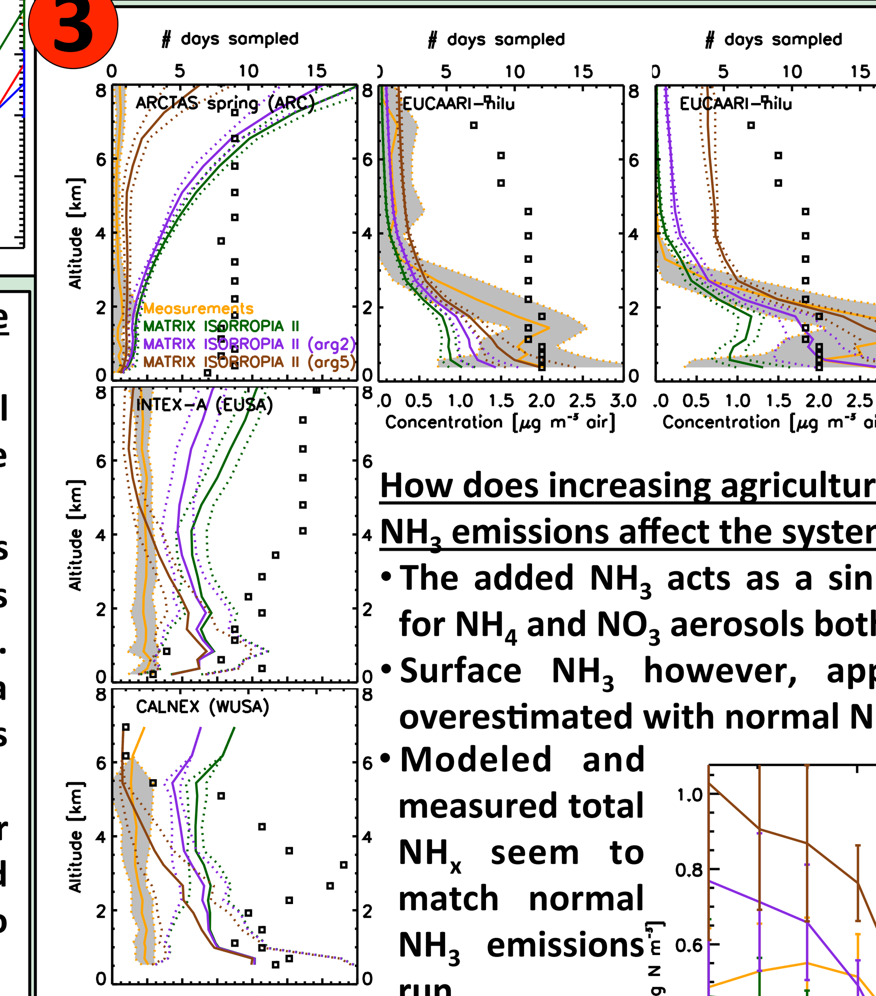
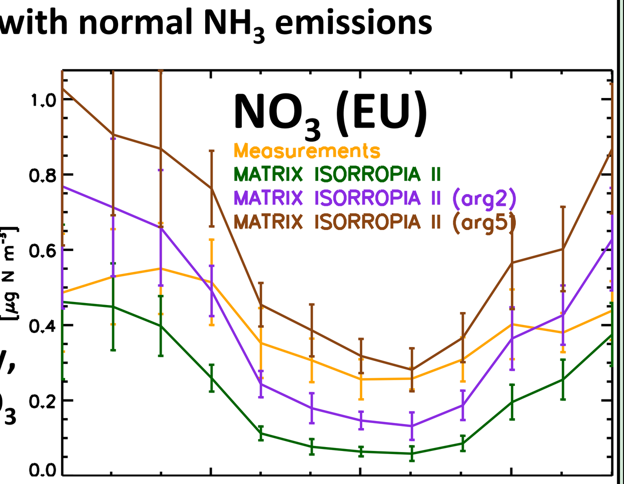


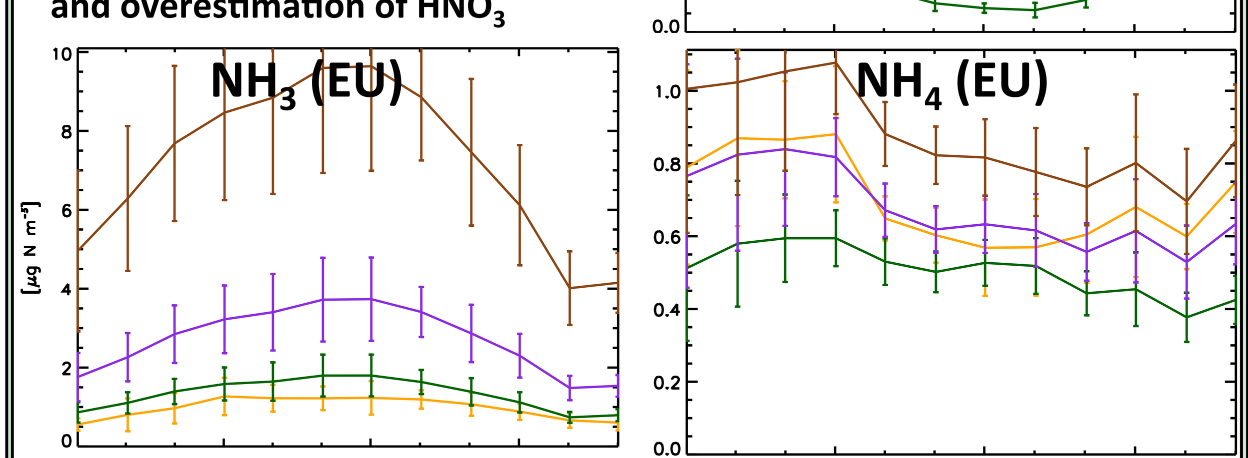
Figure 8: (left) Mean regional profile from ARCTAS spring, INTEX-A, CALNEX and EUCCARI campaigns plotted against NH_3 emissions sensitivity runs. **Figure 9:** (below) same as Figure 6, for EU with sensitivity runs

How does increasing agricultural NH_3 emissions affect the system?

- The added NH_3 acts as a sink to HNO_3 and source for NH_4 and NO_3 aerosols both at surface and above
- Surface NH_3 however, appears to be already overestimated with normal NH_3 emissions
- Modeled and measured total NH_x seem to match normal NH_3 emissions run.

➢ NH_x is not partitioning properly, leading to underestimation of NO_3 and overestimation of HNO_3





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
- ➢ HNO_3 is sensitive to the heterogeneous sink – an important process to include in models
- ➢ HNO_3 and NO_3 partitioning is strongly dependent on NH_x partitioning
- ➢ Need for more measurements: few campaigns measured NH_3 (CALNEX, TexAQ), no winter campaigns