The Abdus Salam International Centre for Theoretical Physics

2339-5

Workshop on Atmospheric Deposition: Processesand Environmental Impacts

21 - 25 May 2012

Global Modelling of Atmospheric Reactive reduced Nitrogen

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Joint Research Centre

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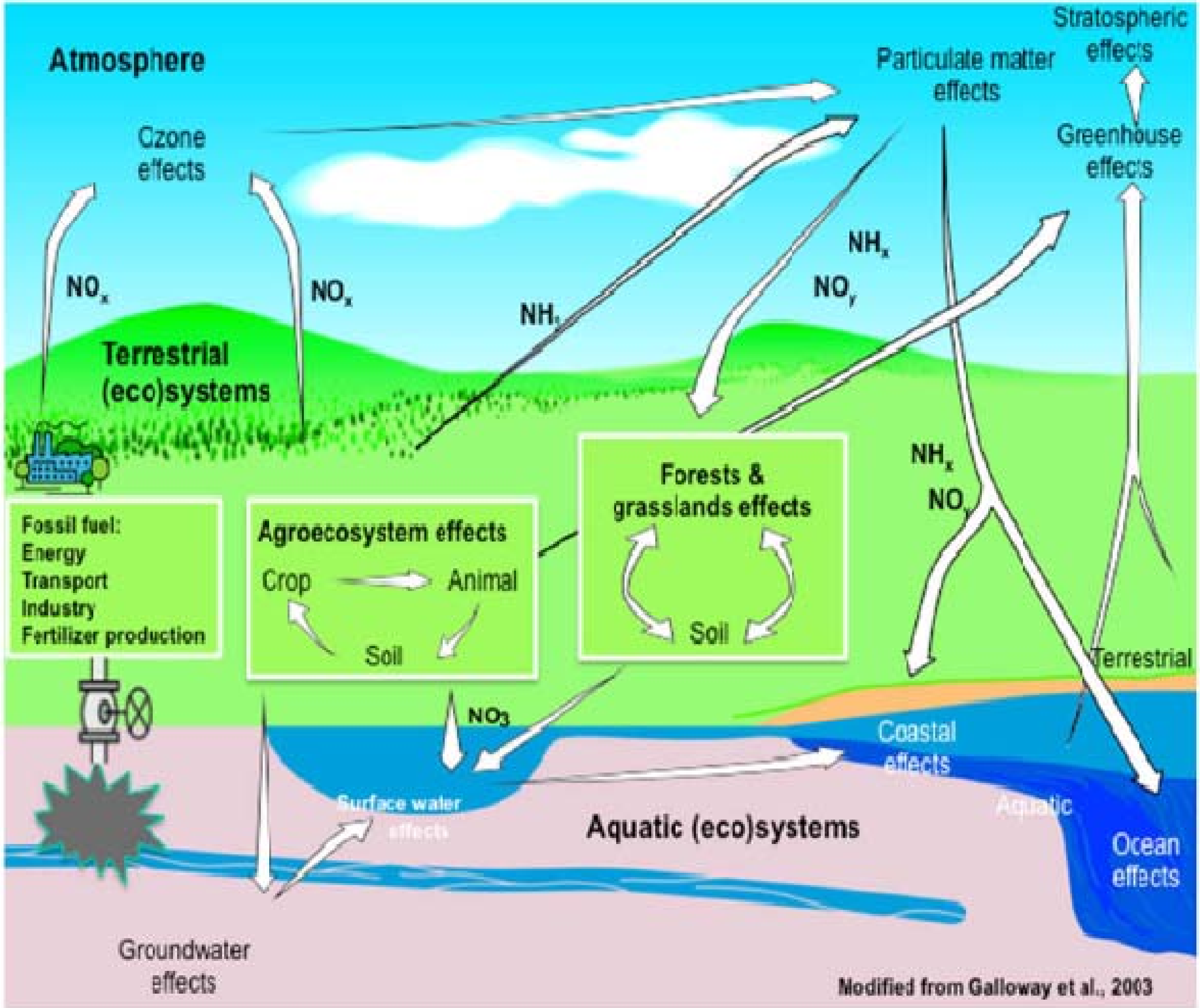


- •O3 and aerosol formation= air pollution
- •Oxidant chemistry and feedbacks on other pollutants
- •Eutrophication, acidification and ecosystem diversity
- •Terrestial and ocean carbon uptake, N2O formation

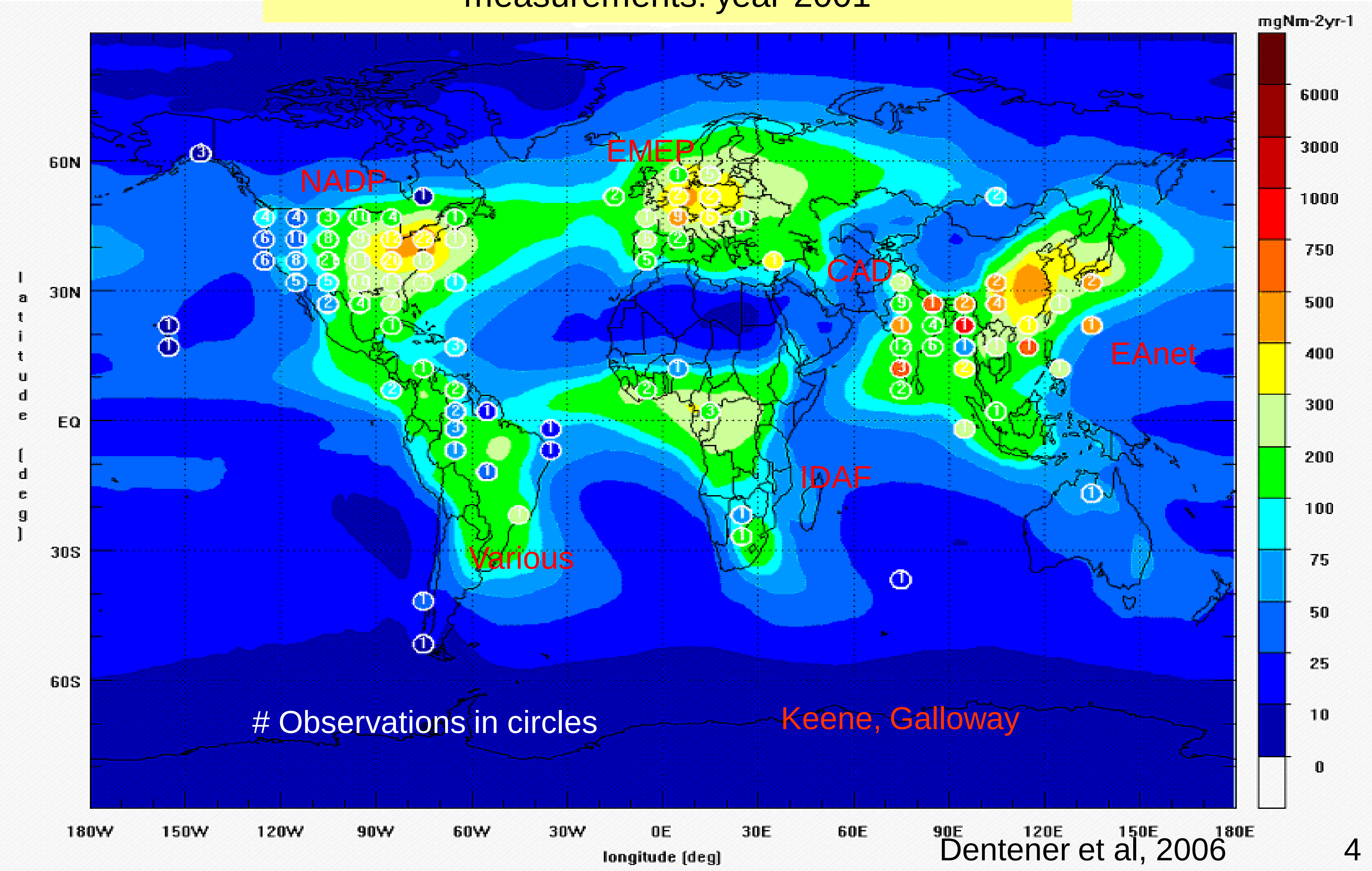
Erisman, 2011

- Oxidized NOy: NO, NO2, HNO3, and NO3p, HONO, organic nitrates (including PAN)
  more than 20 global models, however varying level of detail
  (e.g. hardly any model includes HONO; isoprene nitrates)

- NH3: Reduced NHx NH3 and NH4, amines
  ca. 5-6 globals models include NH3, but only few studies really focussed on NH3.

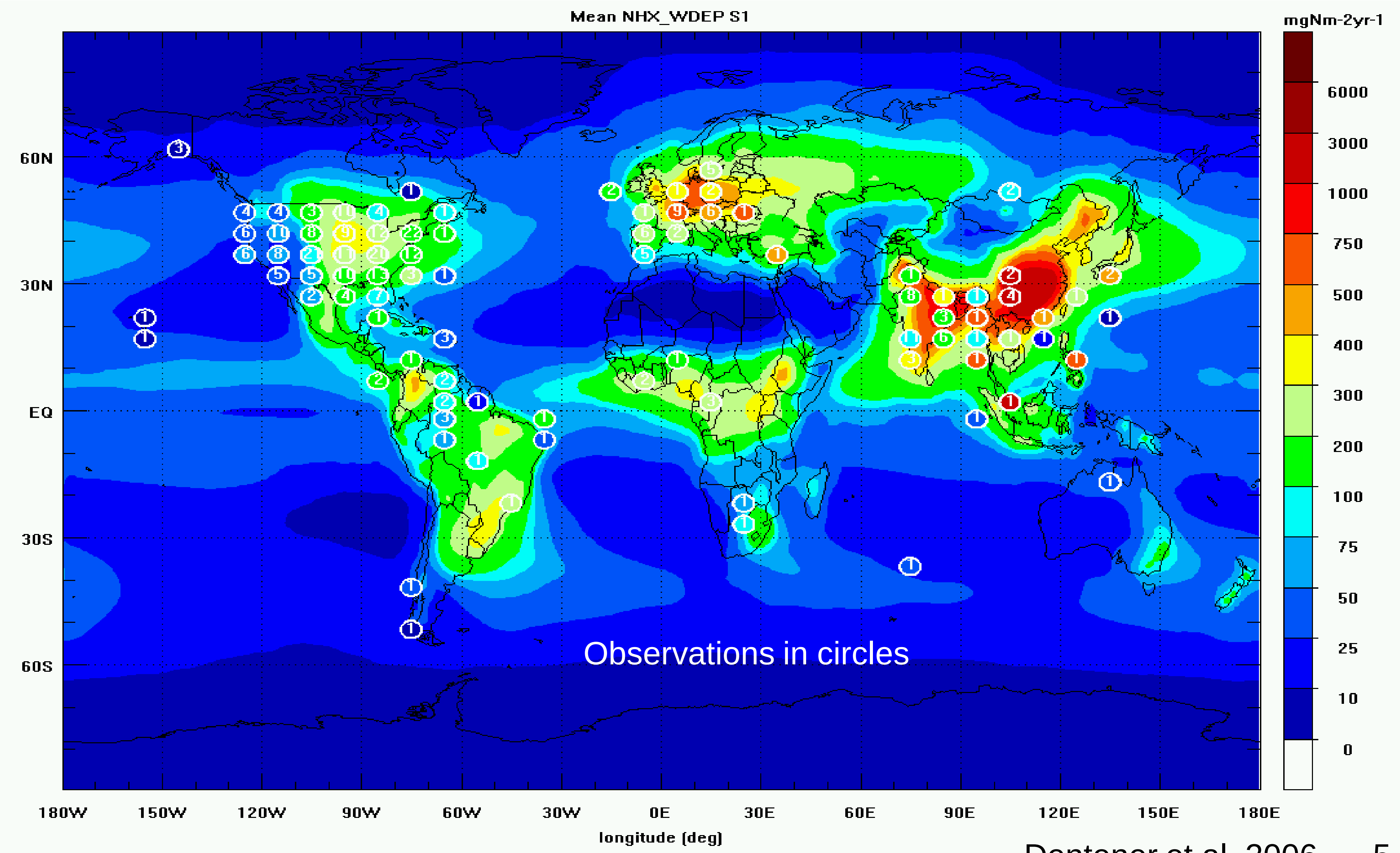
- N2O: atmospheric modelling mostly interesting for verification of budgets (and stratosphere).

HNO_3 and $NO3_p$ wet deposition: models and measurements: year 2001

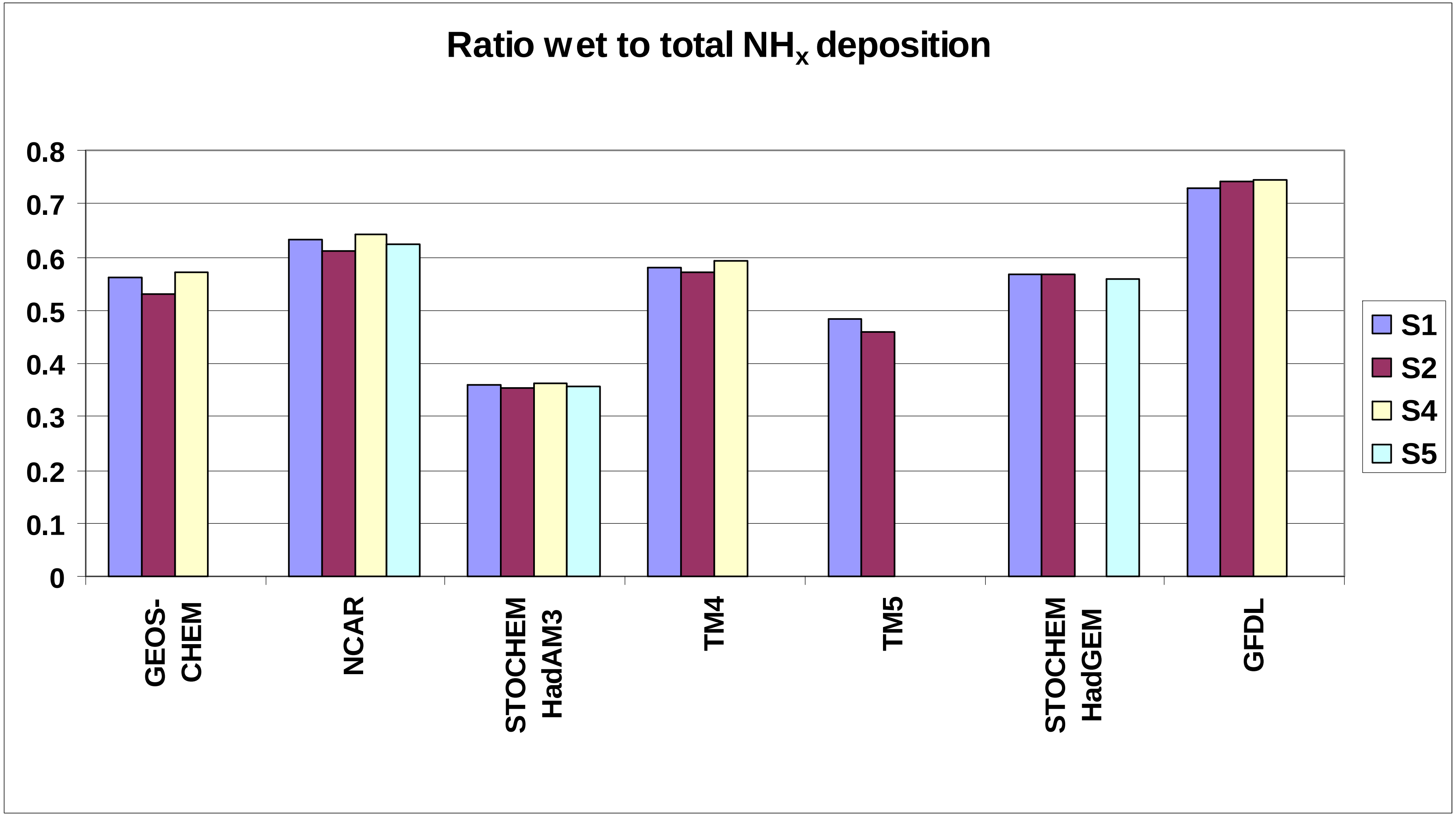


Dentener et al, 2006

Mean model NH_x wet dep



Dentener et al, 2006



Dentener et al, 2006

New developments:

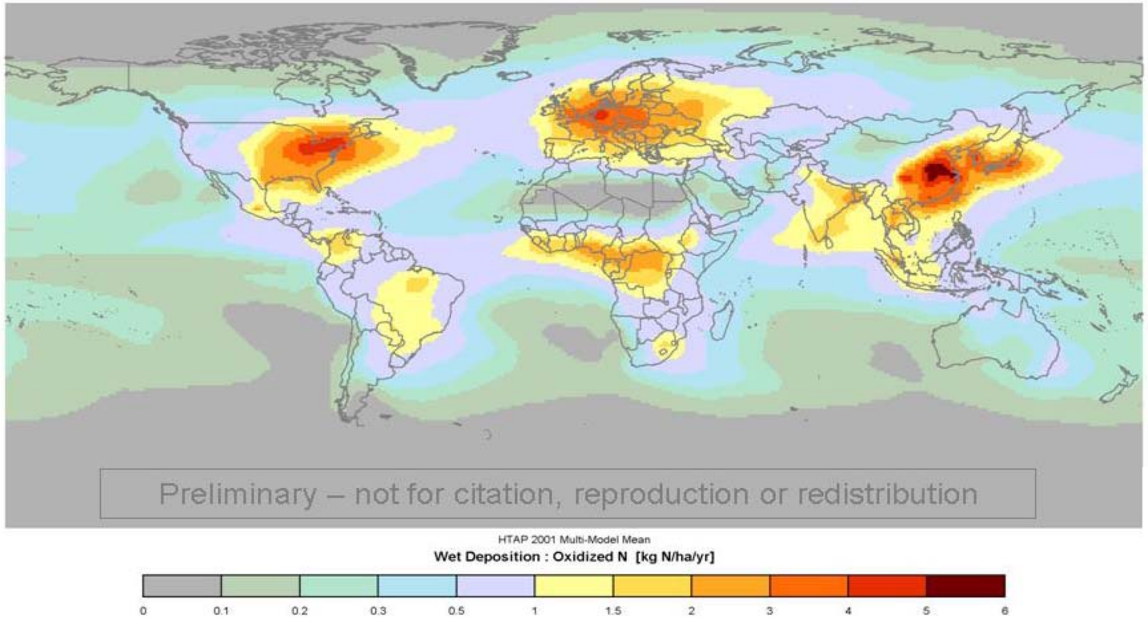
- WMO precipitation chemistry assessment, including reactive nitrogen, now scheduled for April 2012

- Historical and future emissions to inform the IPCC-AR5 report including NH_3 and NO emissions

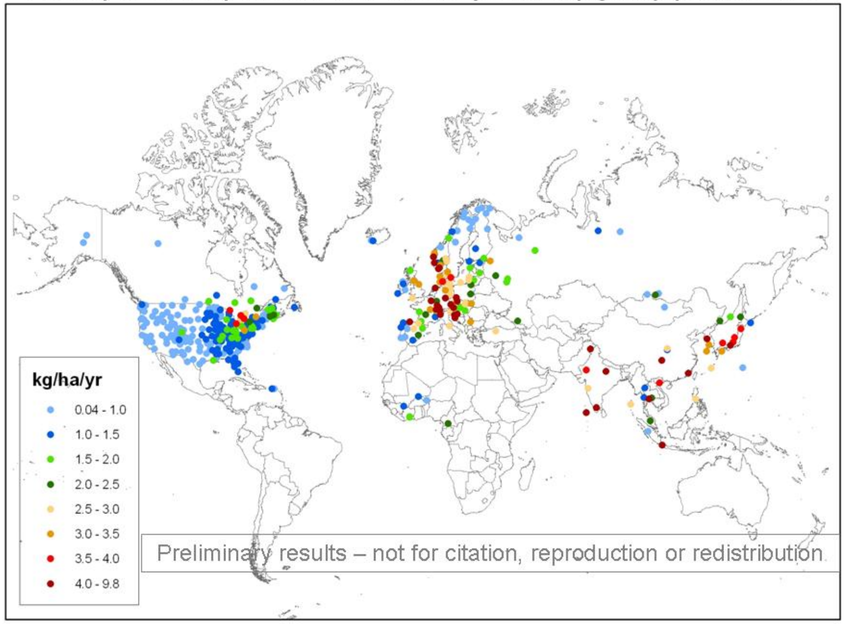
- HTAP: Source receptor relationships on the hemispheric scale

- ACCMIP: timeslice experiments including climate change and chemical sensitivity studies using ‘newest’ generation of global models

Wet deposition of N-NO_3^- (kg N/ha/yr): 2001 HTAP *model* ensemble mean (left) and 2000-2002 *measurements WMO*)



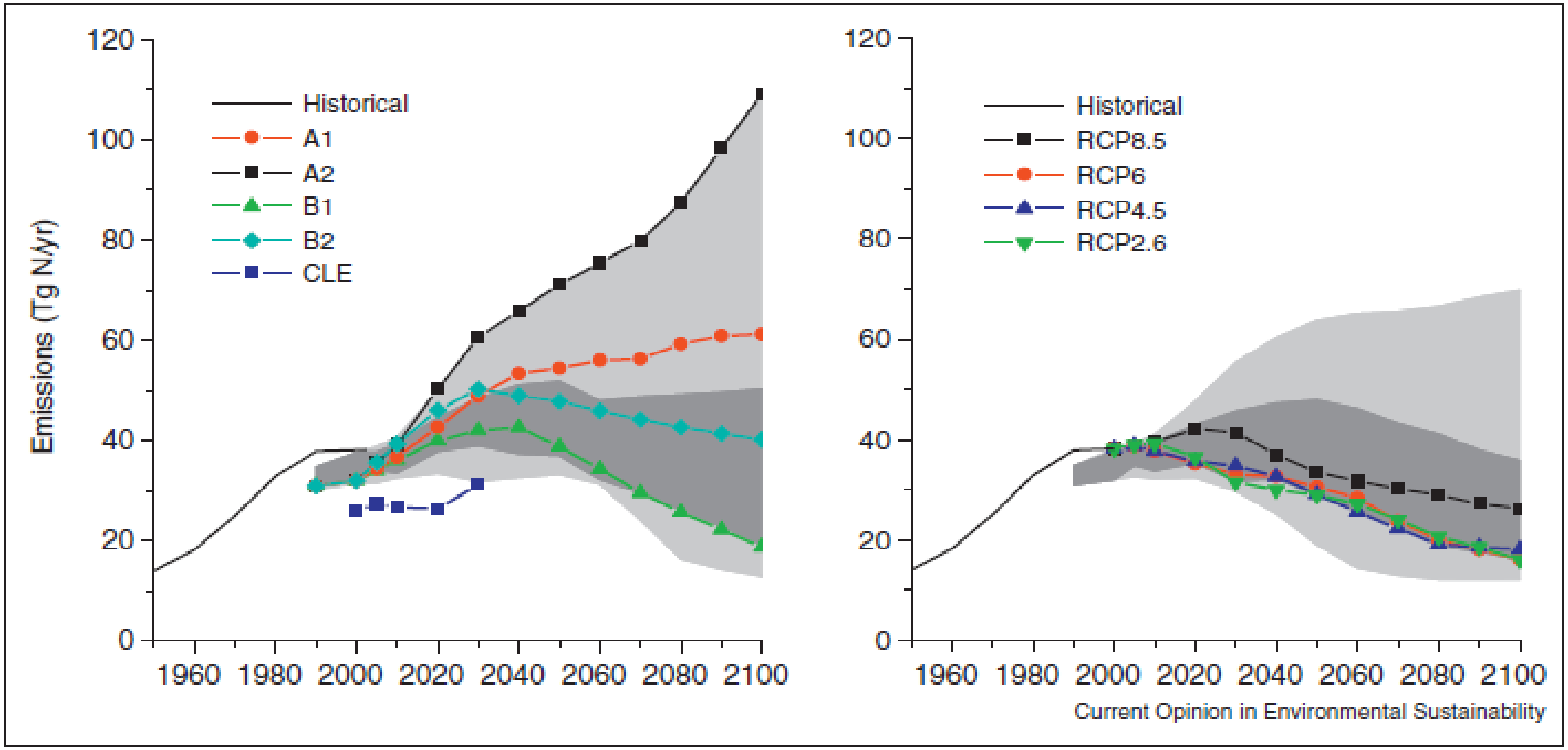
3-Year (2005-2007) Mean N-NO_3^- Wet Deposition (kg/ha/yr)-Rural Sites



RCP NOx emissions

Without climate policy

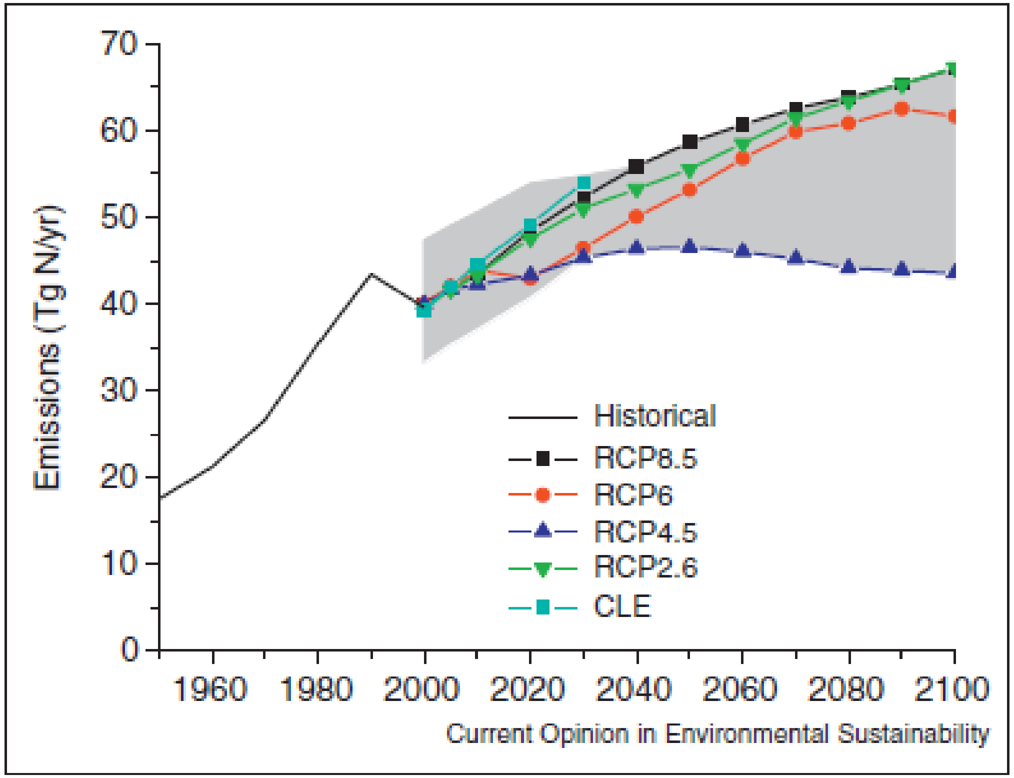
With climate policy



Emissions factors decrease by about 60% in the 2000-2050 period
Clear impact of climate policy via change in energy system

Van Vuuren et al 2011

RCP NH_3 emissions



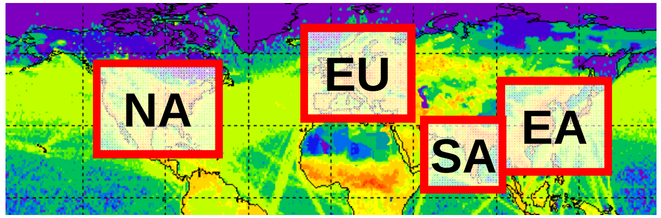
Dominant role for NH_3
Increasing

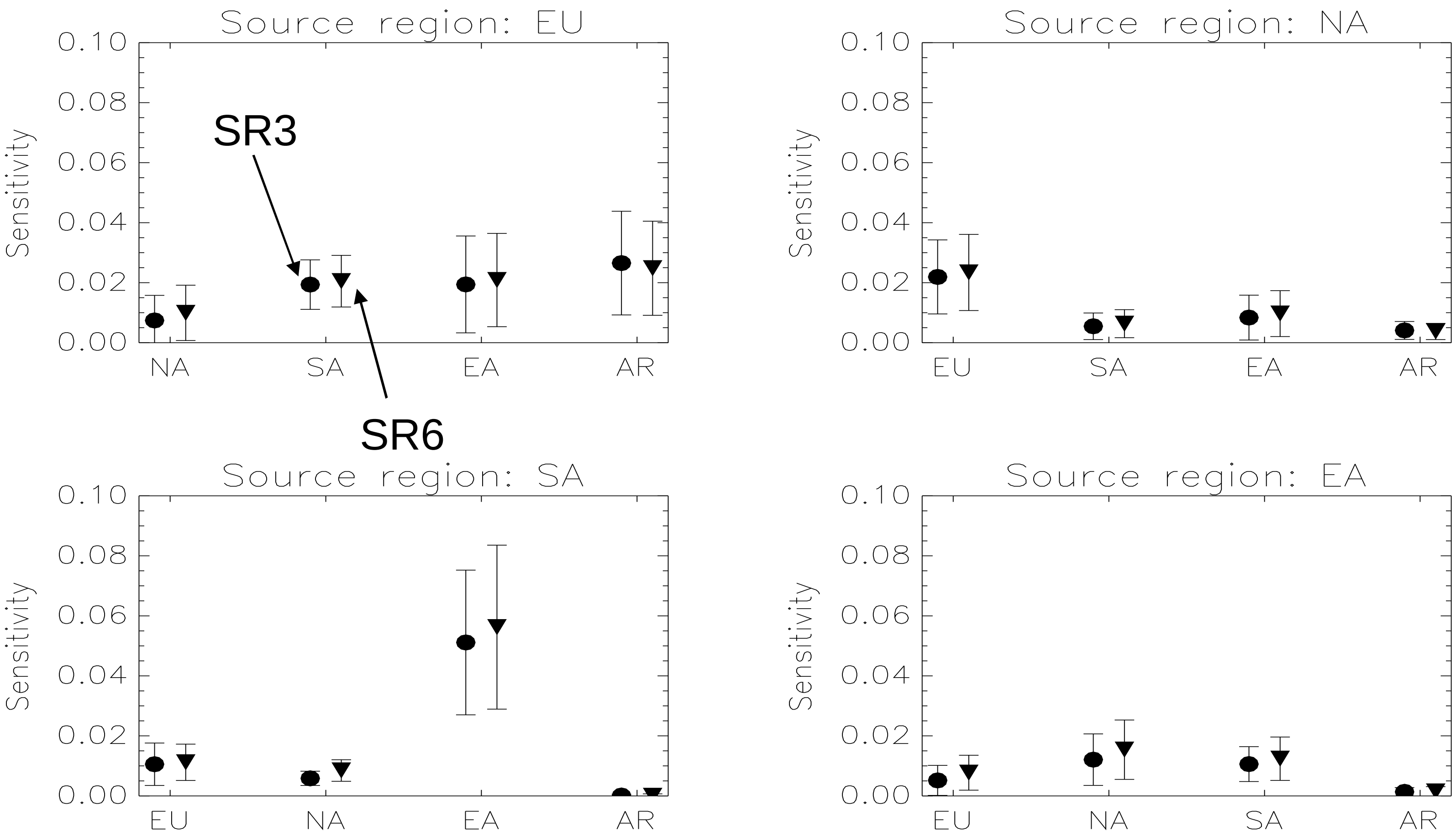
RCPs do not cover the full range of possible futures.

Designed for climate relevance. show “desirable” air pollution futures but do not include the non-compliance baselines.

Van Vuuren et al 2011

HTAP Source-Receptor Relationships
Annual NOy deposition



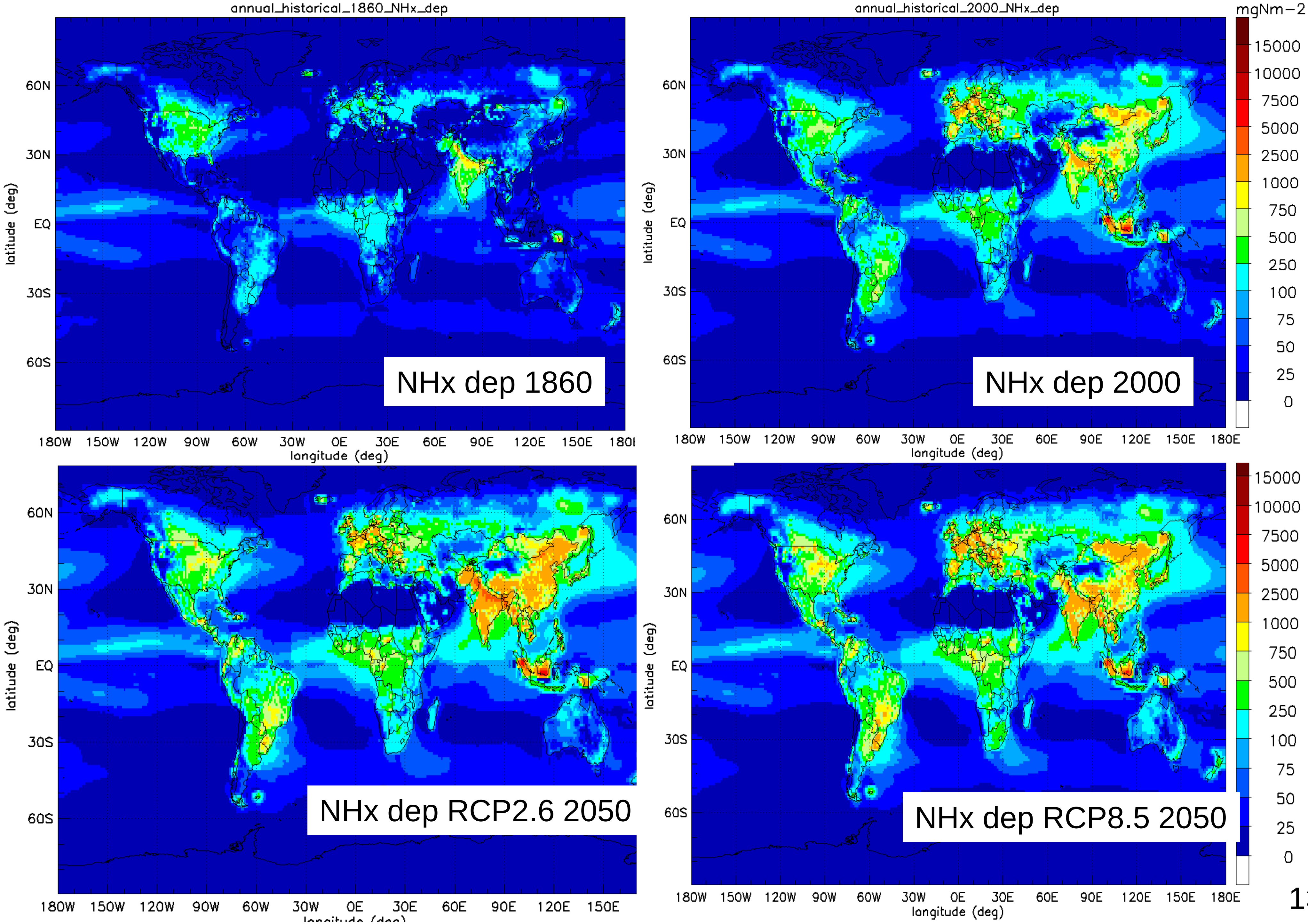


$$Sensitvity = \frac{dep(ctl, receptor) - dep(expt, receptor)}{emiss(ctl, source) - emiss(expt, source)}$$

Sanderson et al, 2008

IGAC ACCMIP

- Links to AMIP5 climate simulations (IPCC AR5)
  did not include a lot of chemistry
- Timeslice experiments evaluating sensitivity to
  climate and chemistry, 1860-2100
- prescribed decadal mean SST
- about 10 global models
- focus on changes in photochemistry
- also deposition output
- Higher resolution
- Increase in complexity of NOy, NHx deposition not expected


annual_historical_1860_NHx_dep
annual_historical_2000_NHx_dep
mgNm−2
15000
10000
7500
5000
2500
1000
750
500
250
100
75
50
25
0
NHx dep 1860
NHx dep 2000
NHx dep RCP2.6 2050
NHx dep RCP8.5 2050
latitude (deg)
longitude (deg)
60N
30N
EQ
30S
60S
180W
150W
120W
90W
60W
30W
0E
30E
60E
90E
120E
150E
180E

Some history of global atmospheric NH_3 modelling

Journal of Atmospheric Chemistry **19:** 331–369, 1994. 331
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A Three-Dimensional Model of the Global Ammonia Cycle

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(Received: 16 August 1993; in final form: 25 January 1994)

Abstract. Using a three-dimensional (3-D) transport model of the troposphere, we calculated the global distributions of ammonia (NH_3) and ammonium (NH_4^+), taking into account removal of NH_3 on acidic aerosols, in liquid water clouds and by reaction with OH. Our estimated global 10° × 10° NH_3 emission inventory of 45 Tg N–NH_3 yr^{-1} provides a reasonable agreement between calculated wet NH_4^+ deposition and measurements and of measured and modeled NH_4^+ in aerosols, although in Africa and Asia especially discrepancies exist.

- Moguntia 10x10 degrees; 10 vertical layers; climatological meteorological driver
- NH3 coupled to existing sulfur and photochemical scheme
- Previously there were some global NH3 emission estimates, but consistency with measurements unclear
- Global emissions 45 Tg N
- No equilibrium (but assumed maximum equilibration of (NH4)1.5) SO4; equilibration timescale ca. hours. (‘NH4NO3 globally not very important’)
- Canopy and ocean emission via compensation point: highly simplistic!
- Accounting for fast ‘subgrid’ deposition processes of NH3

TABLE I. Yearly NH_3 emissions [Tg N yr^{-1}]

Anthropogenic:	
dairy cattle	5.5
beef cattle/buffaloes	8.7
pigs	2.8
horses/mules/asses	1.2
sheep/goats	2.5
poultry	1.3
fertilizer	6.4
biomass burning	2.0
subtotal	30.4
Natural:	
wild animals	2.5
vegetation	5.1
ocean	7.0
subtotal	14.6
Total	45.0

Vegetation emissions:

•Canopy compensation point
apoplastic NH4=48 umol/m3; pH=6.8

•Exchange velocity scaled with vegetation amount;relationship with climate

•Ocean emission: scaled with DMS emissions

Surface level (few hundred meters average) NH_3 concentrations

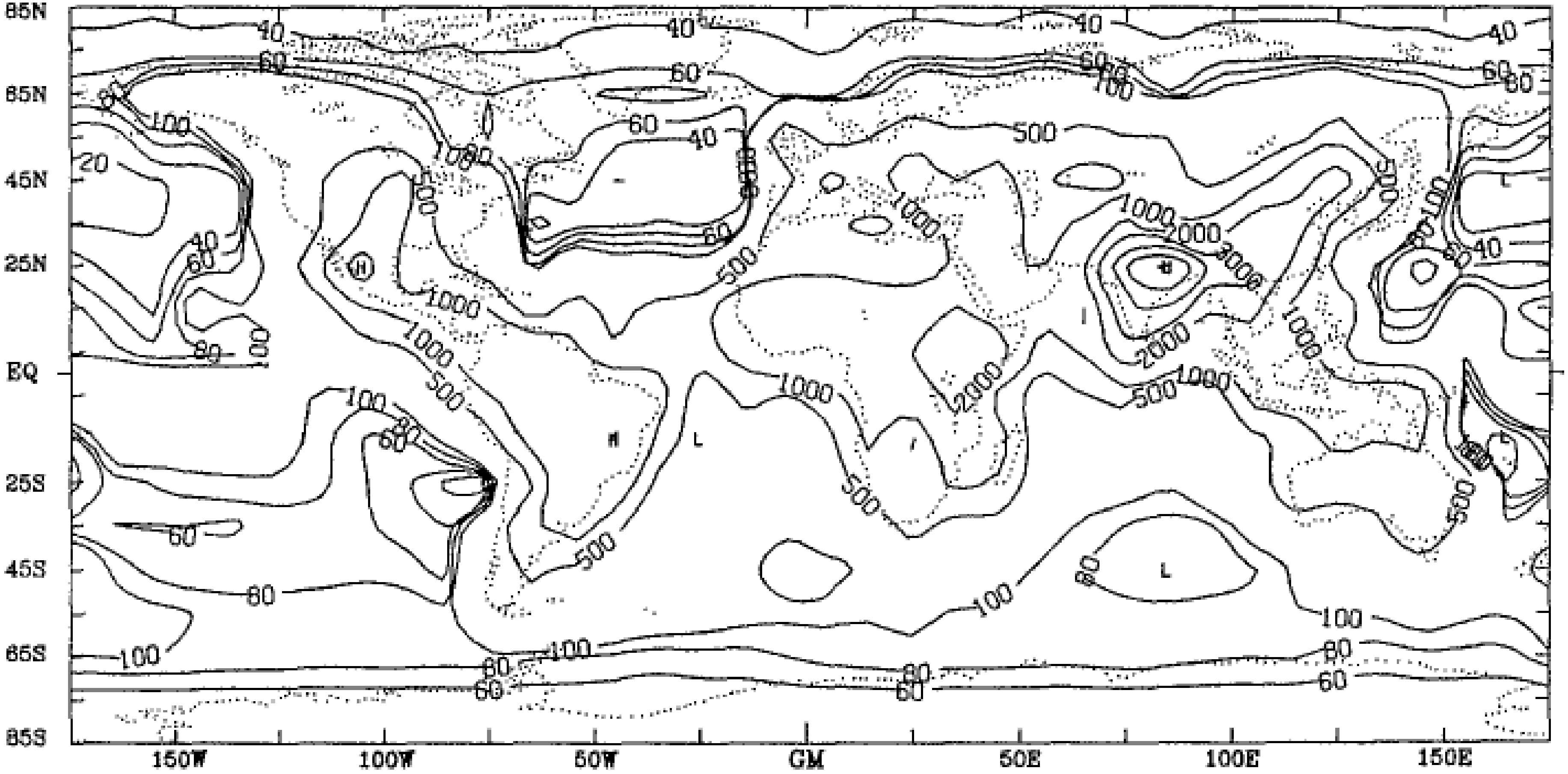


Fig. 2a.

Dentener and Crutzen, 1994

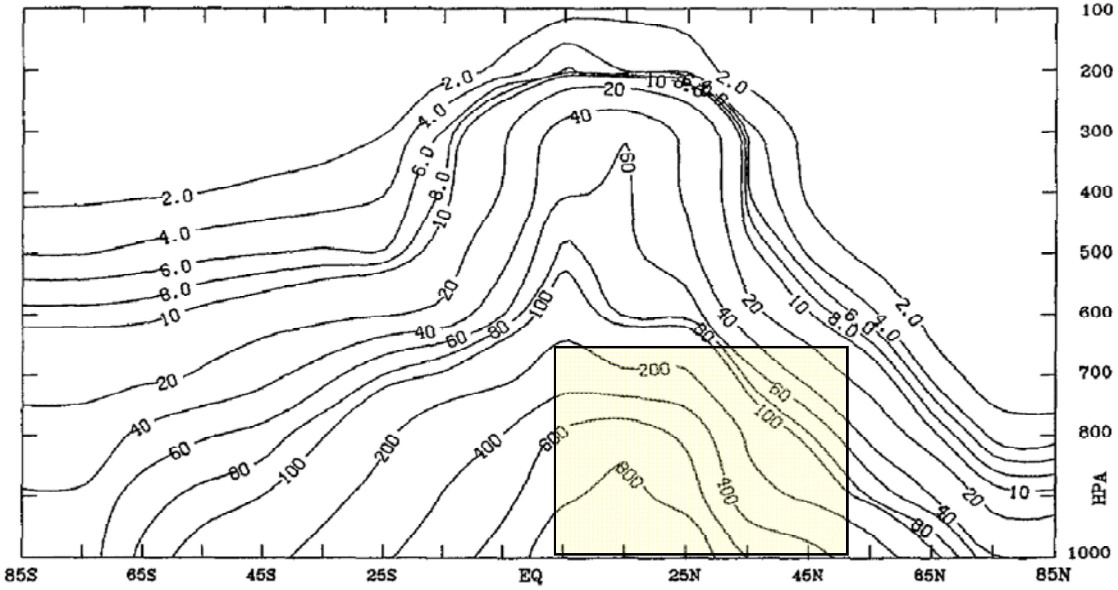
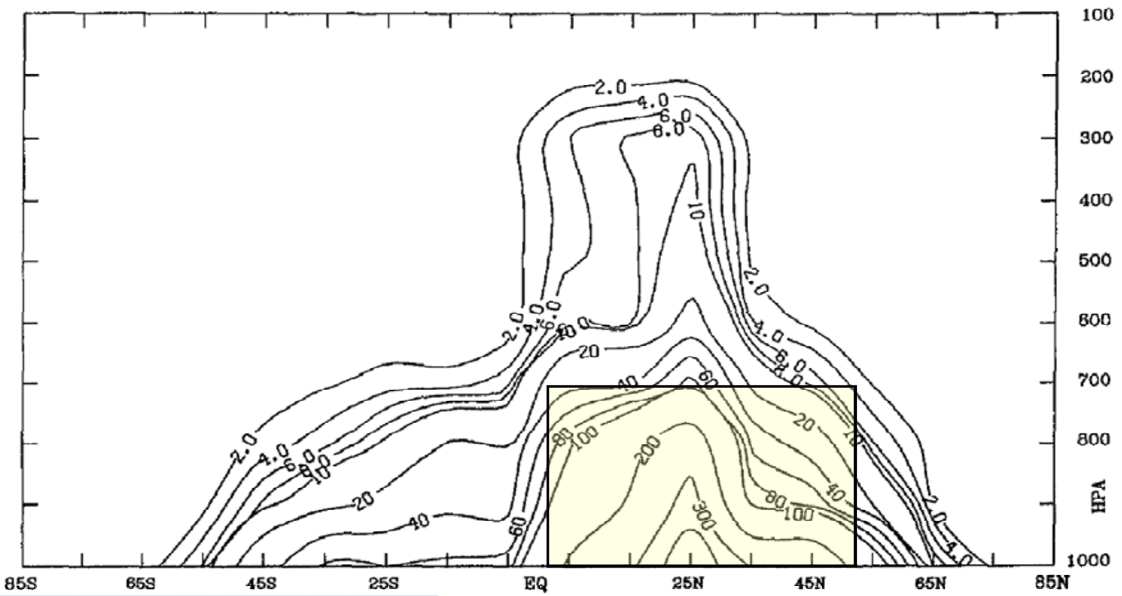


Fig. 2b.



Canopy compensation point
Ocean emission (scaled to DMS)

And without these natural emissions

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 104, NO. D11, PAGES 13,791–13,823, JUNE 20, 1999

Global concentrations of tropospheric sulfate, nitrate, and ammonium aerosol simulated in a general circulation model

Peter J. Adams and John H. Seinfeld
Department of Chemical Engineering, California Institute of Technology, Pasadena

Dorothy M. Koch
National Aeronautics and Space Administration Goddard Institute for Space Studies, New York

Abstract. Global sulfate aerosol composition is simulated c
Space Studies general circulation model II′ (GISS GCM II-
peroxide, gas phase ammonia, and particulate ammonium ar

Bouwman et al 97 inventory
No compensation point
Isorropia Thermodynamic Equilibrium
Better comparison to sfc observations

Table 2. Global Ammonia Emissions by Source [*Bouwman et al.*, 1997]

Source	Emission, Tg N yr^{-1}
Domesticated animals	21.6
Fertilizers	9.0
Oceans	8.2
Biomass burning	5.9
Crops	3.6
Humans	2.6
Soils under natural vegetation	2.4
Other	0.4
Total	53.6

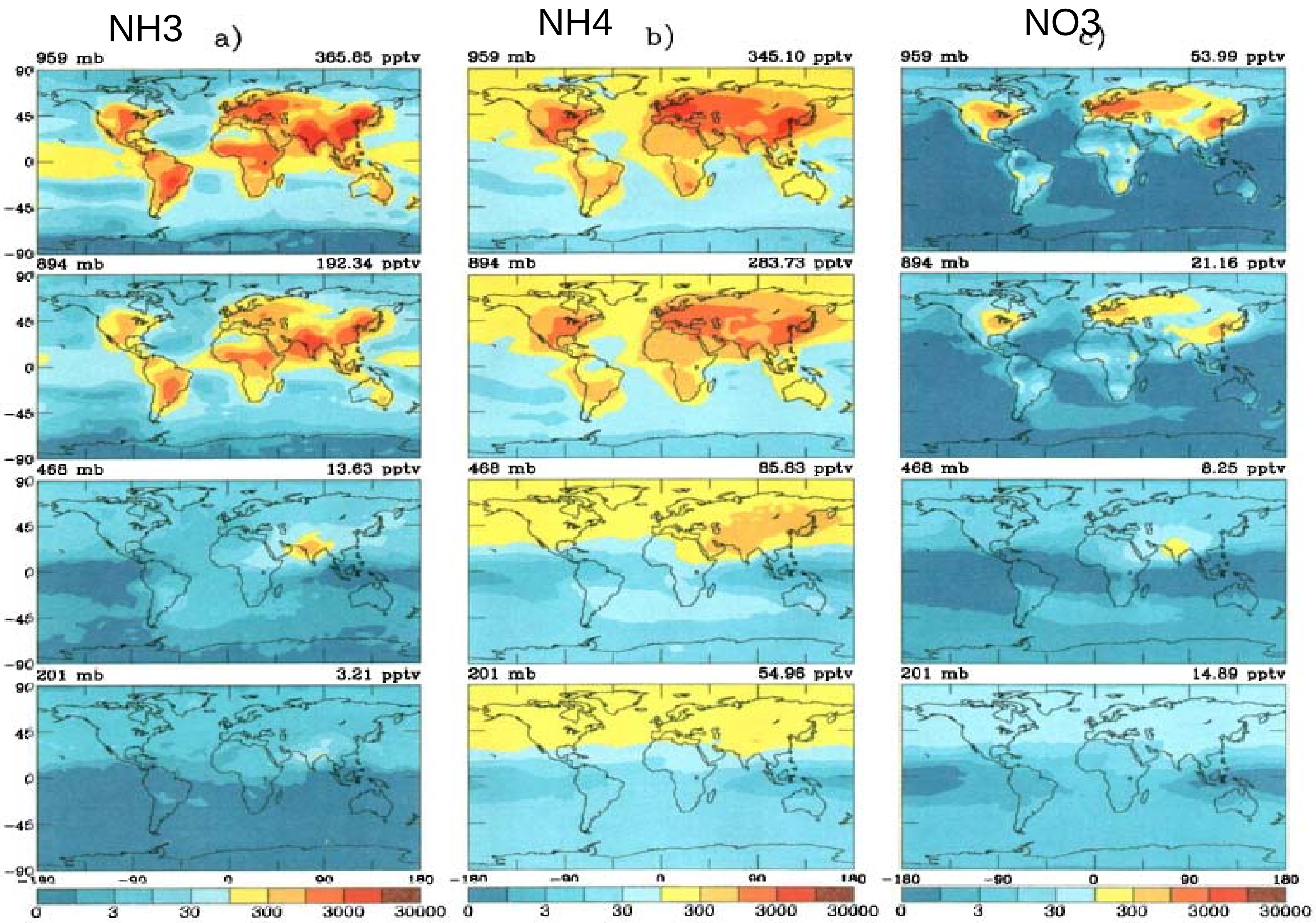
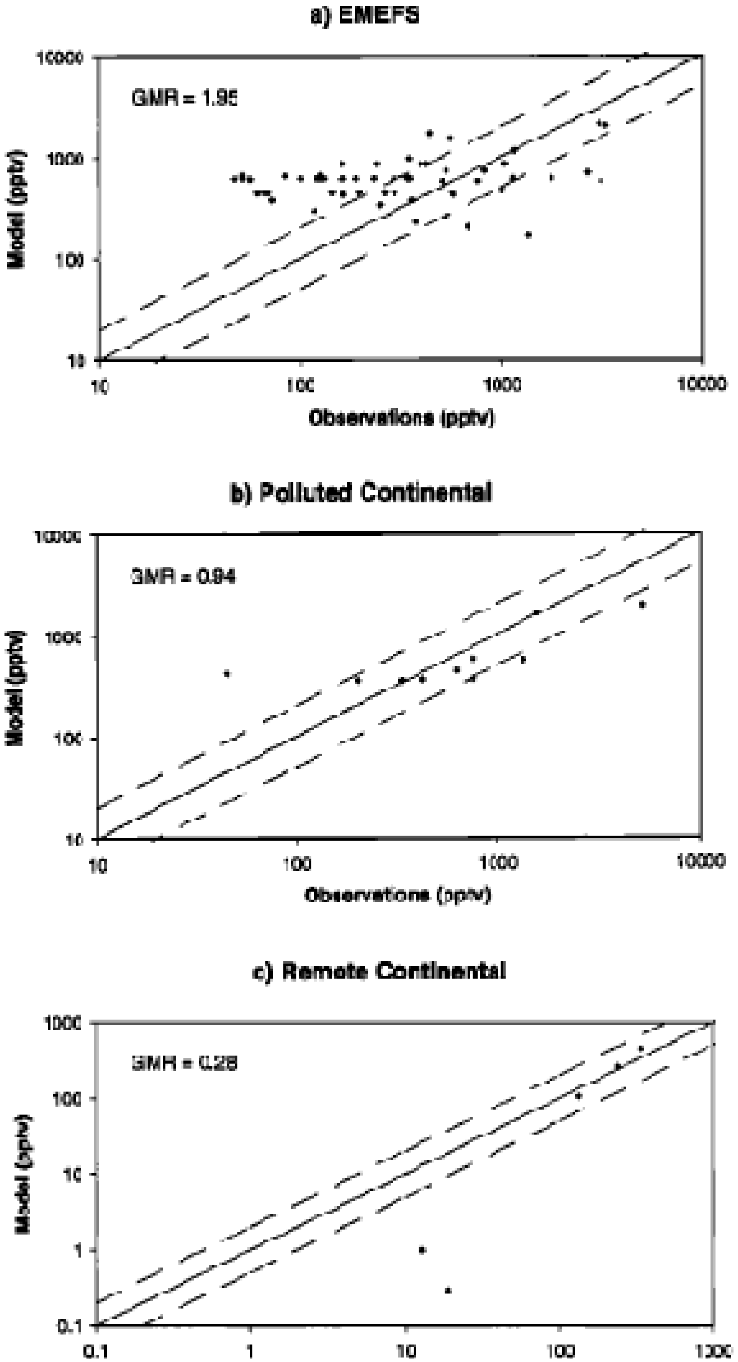


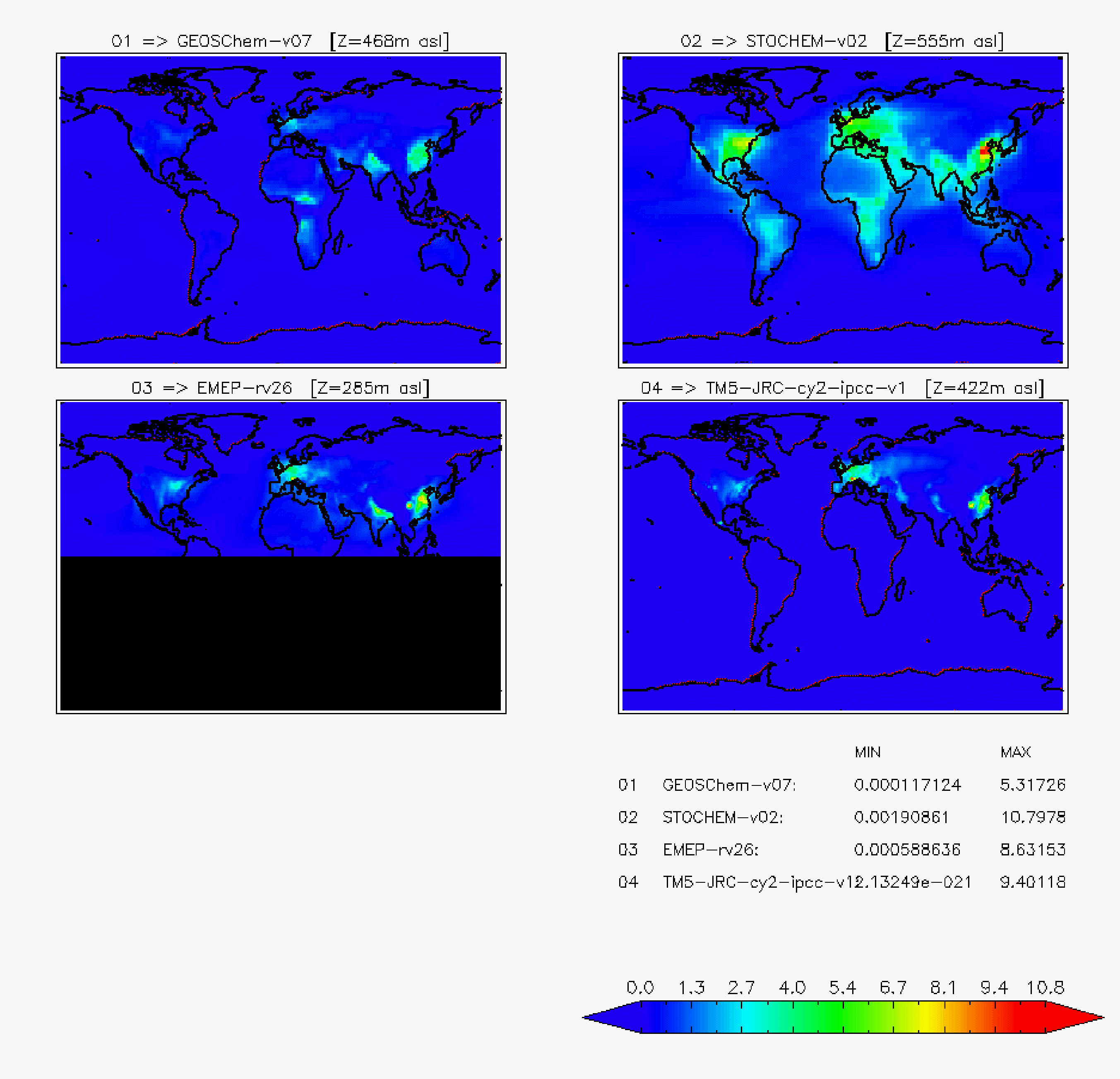
Plate 3. (a) Annual average ammonia mixing ratios. Note that 1 µg m^{-3} NH_3 = 1457 pptv NH_3 at 298 K and 1 bar. (b) Annual average ammonium mixing ratios. Note that 1 µg m^{-3} NH_4^+ = 1377 pptv NH_4^+ at 298 K and 1 bar. (c) Annual average nitrate mixing ratios. Note that 1 µg m^{-3} NO_3^- = 400 pptv NO_3^- at 298 K and 1 bar. Above each plot, the pressure level of the corresponding model layer is indicated, as is the average mixing ratio in that layer. Contour lines are 1, 3, 10, 30, 100, 300, 1000, 3000, 10,000, and 30,000 pptv.



Verification of surface NH_3

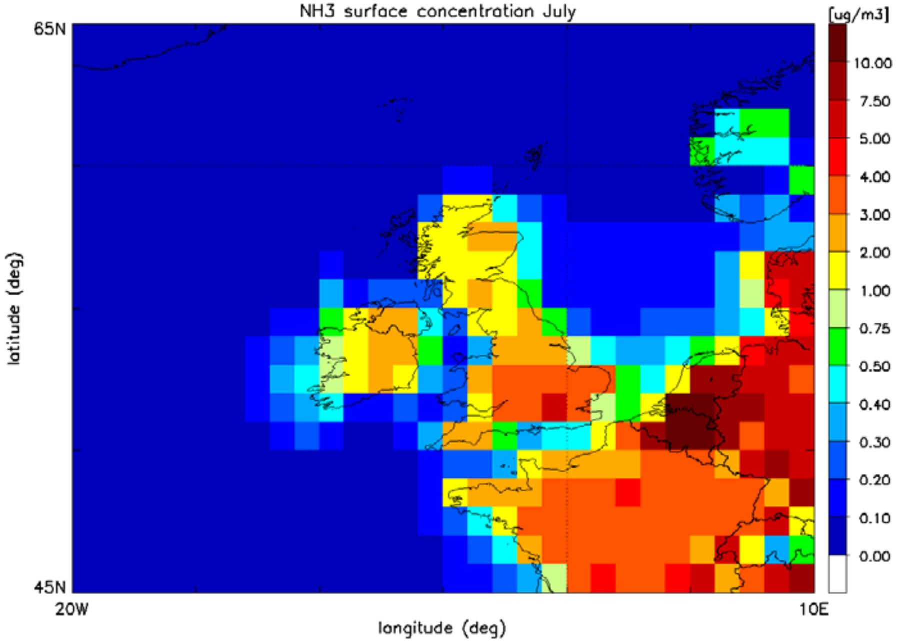
Adams et al, 1999

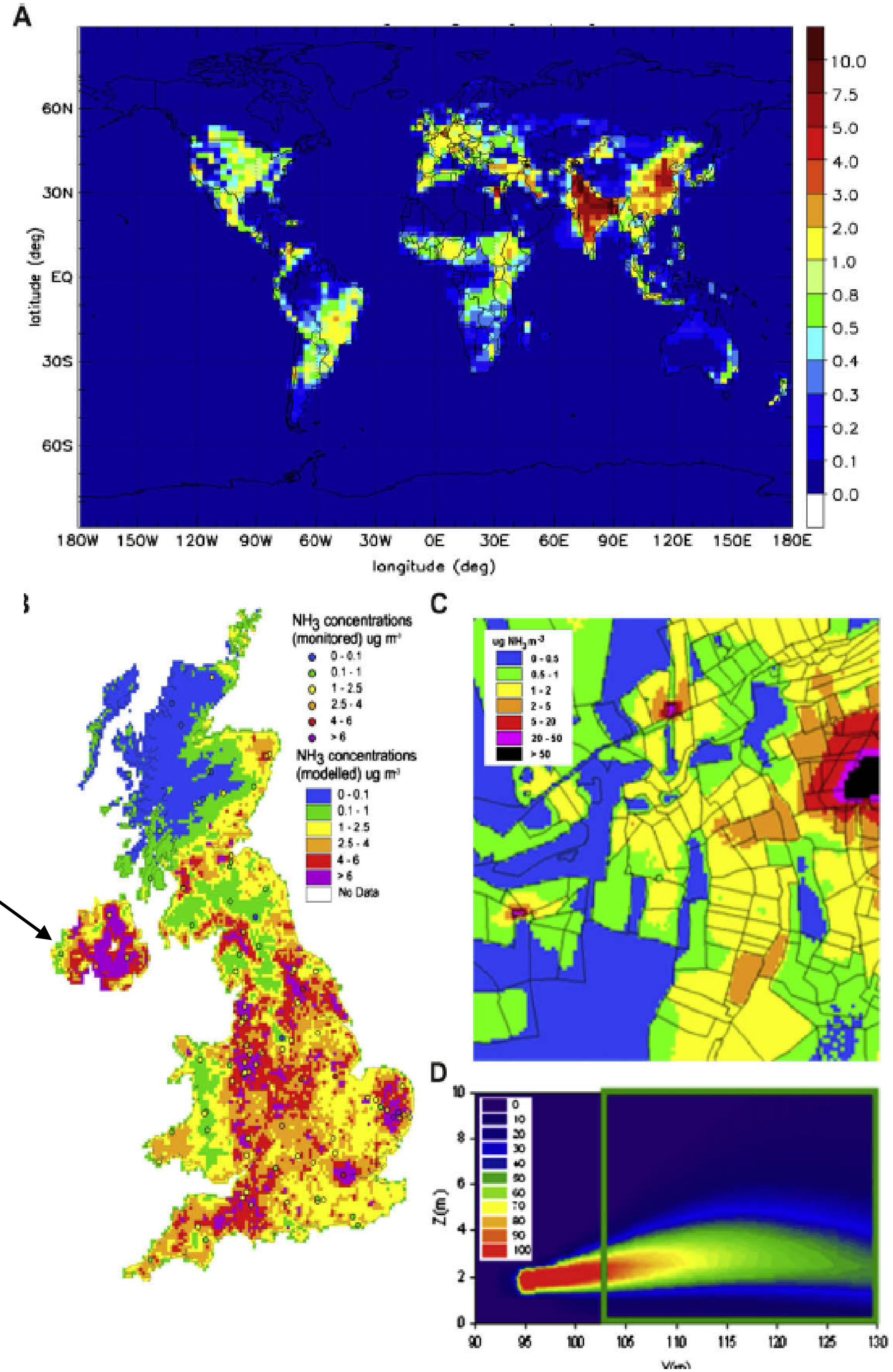
Aerosol NH_4NO_3 in HTAP comparison: annual average



TM5; 1x1 degree

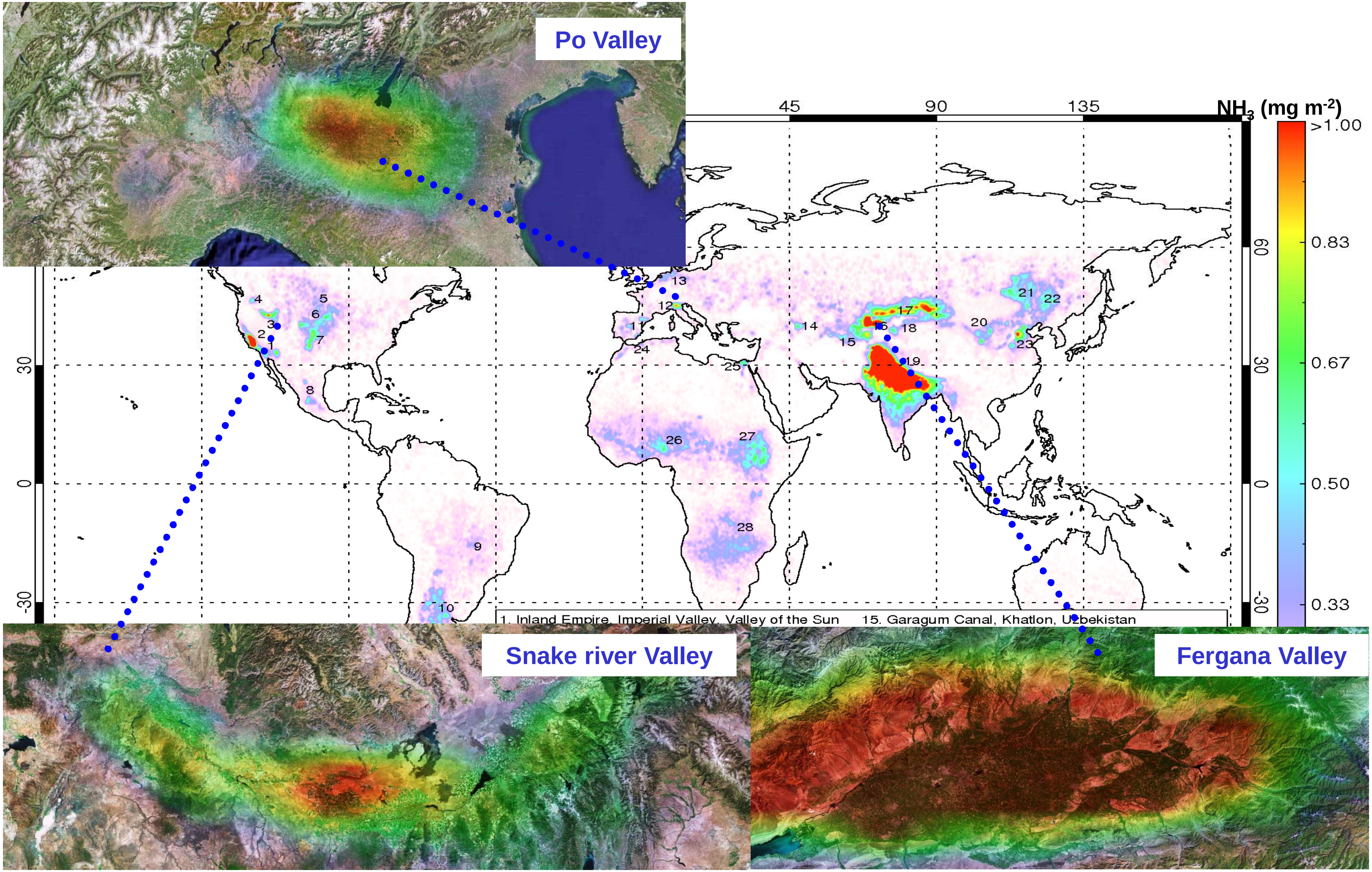
M.A. Sutton et al. / Environmental Pollution 156 (2008) 583–604





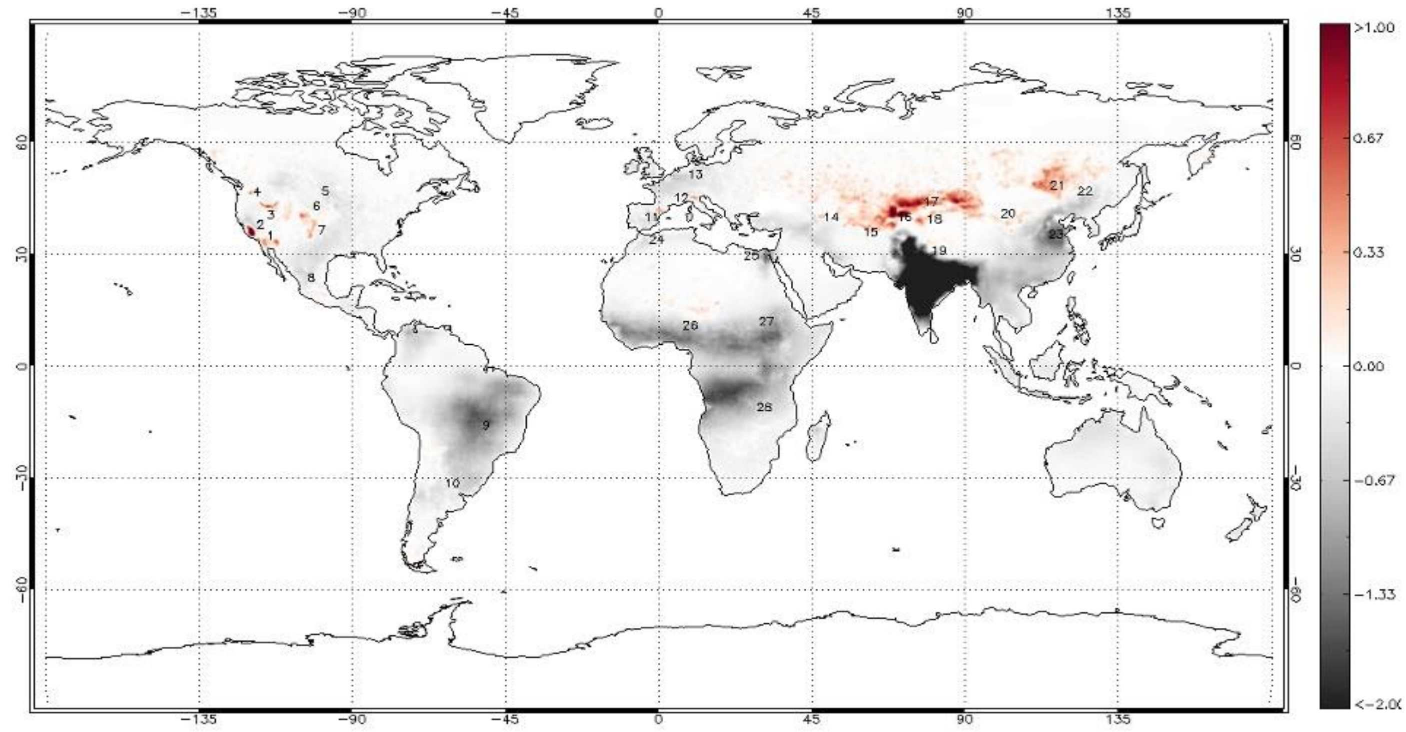
Comparison of NH_3 on global, country and urban scale

Sutton et al, 2008



Clarisse et al, [2009]

Difference of IASI and TM5:



IASI > TM5 → **Missing emissions**

IASI < TM5 → **Detection threshold (Thermal contrast!)**

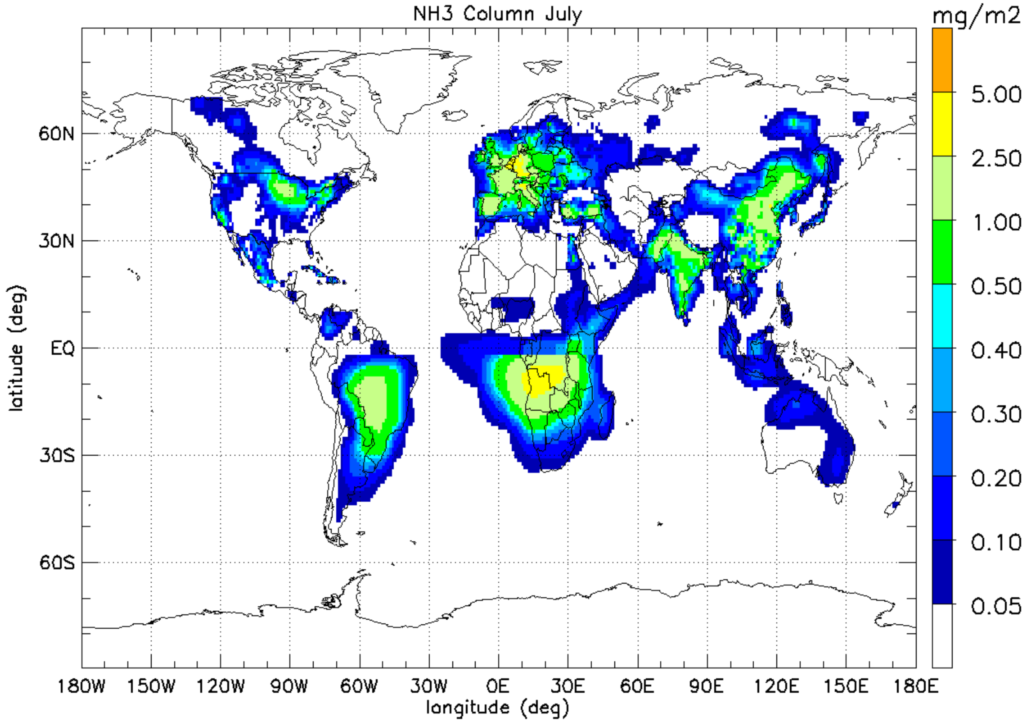
We expect these results to be a **lower bound**:

-Limited thermal contrast in colder parts of the world

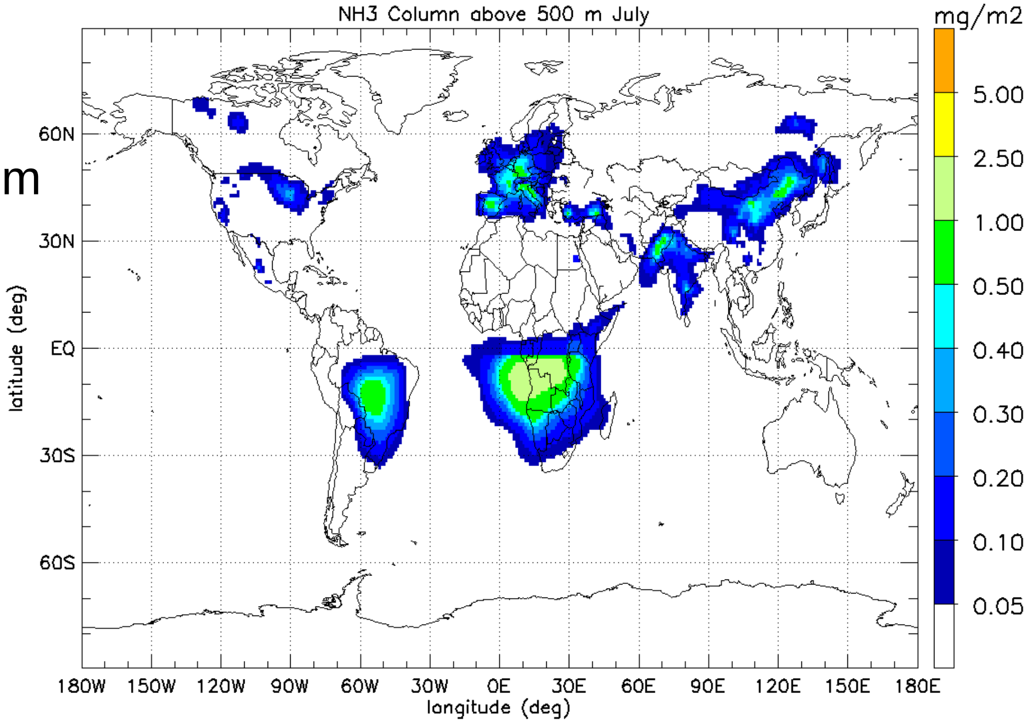
-Detection threshold: + Daily 0.2K → 3mg/m^2

+ Monthly 0.08K → 1.2mg/m^2

Everything lower disappears in the noise



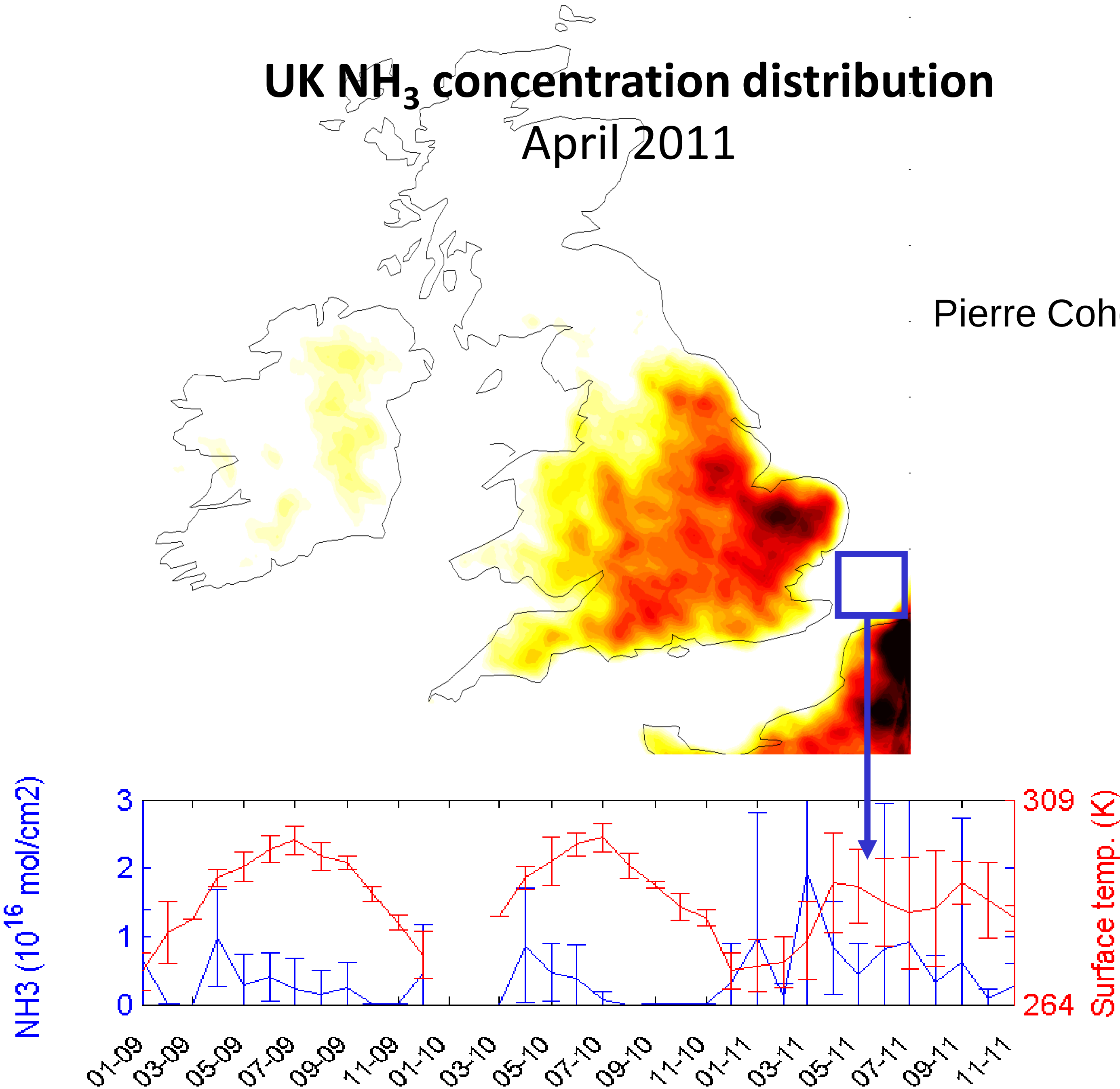
Column higher than 500 m

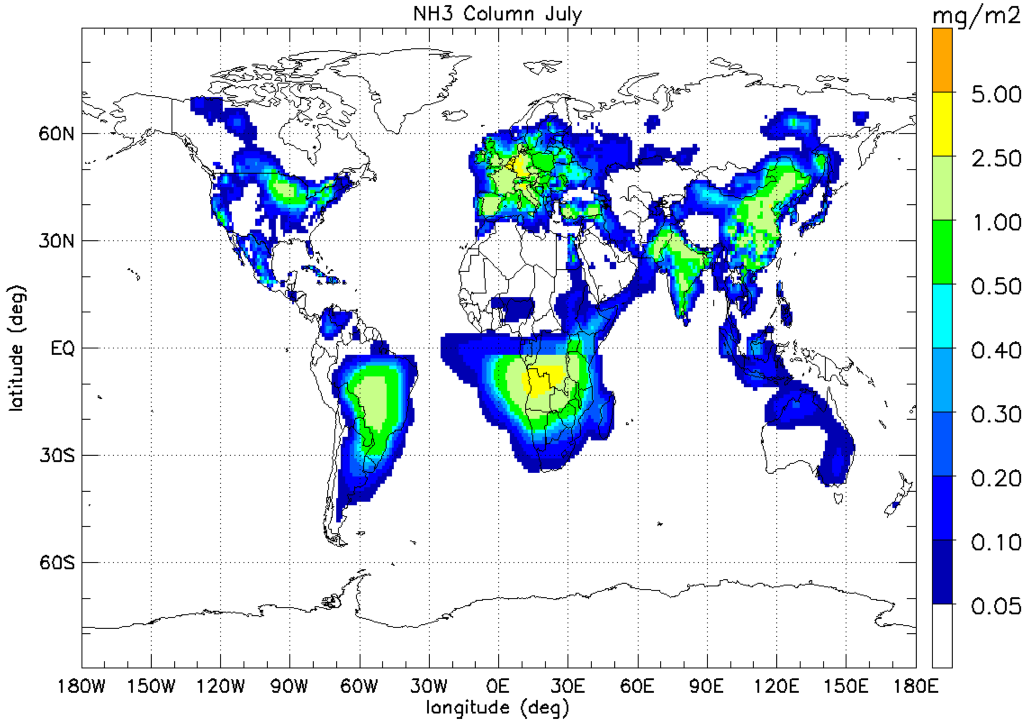


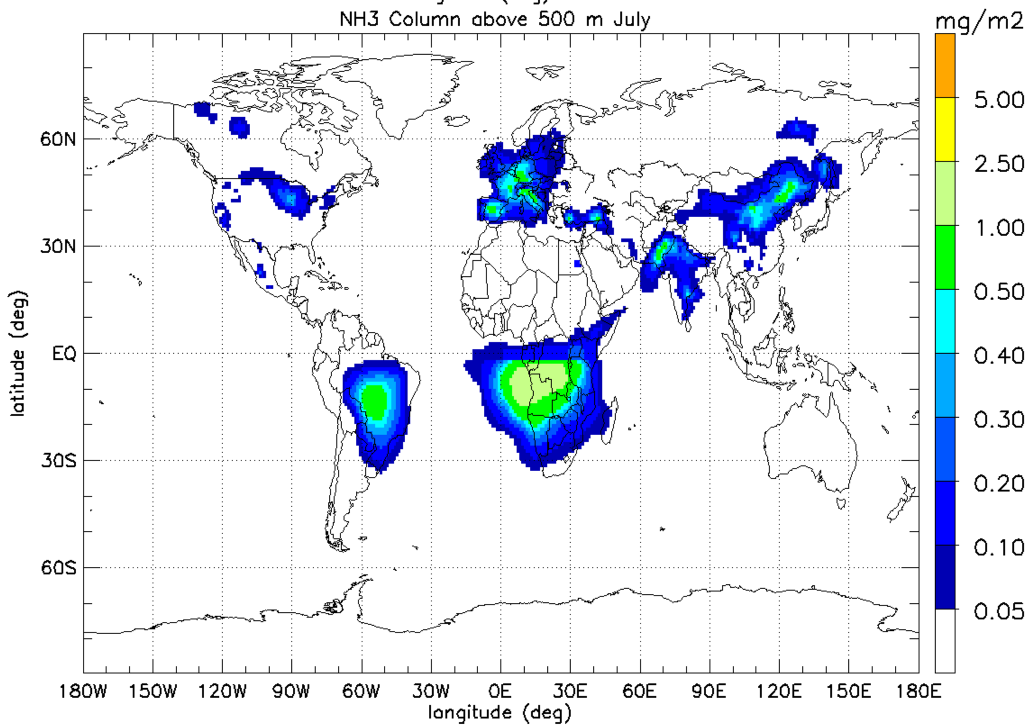
threshold

UK NH_3 concentration distribution
April 2011

Pierre Coheur and coworkers







Proper link of model and observations requires: sensitivity of instrument deep into the Boundary Layer Good description of the vertical mixing in the model (resolution !) Understanding of the time scale of reaction of NH3 Proper description of the vertical profile of the reactants (i.e. HNO3/SO4)

Emissions: uncertain!

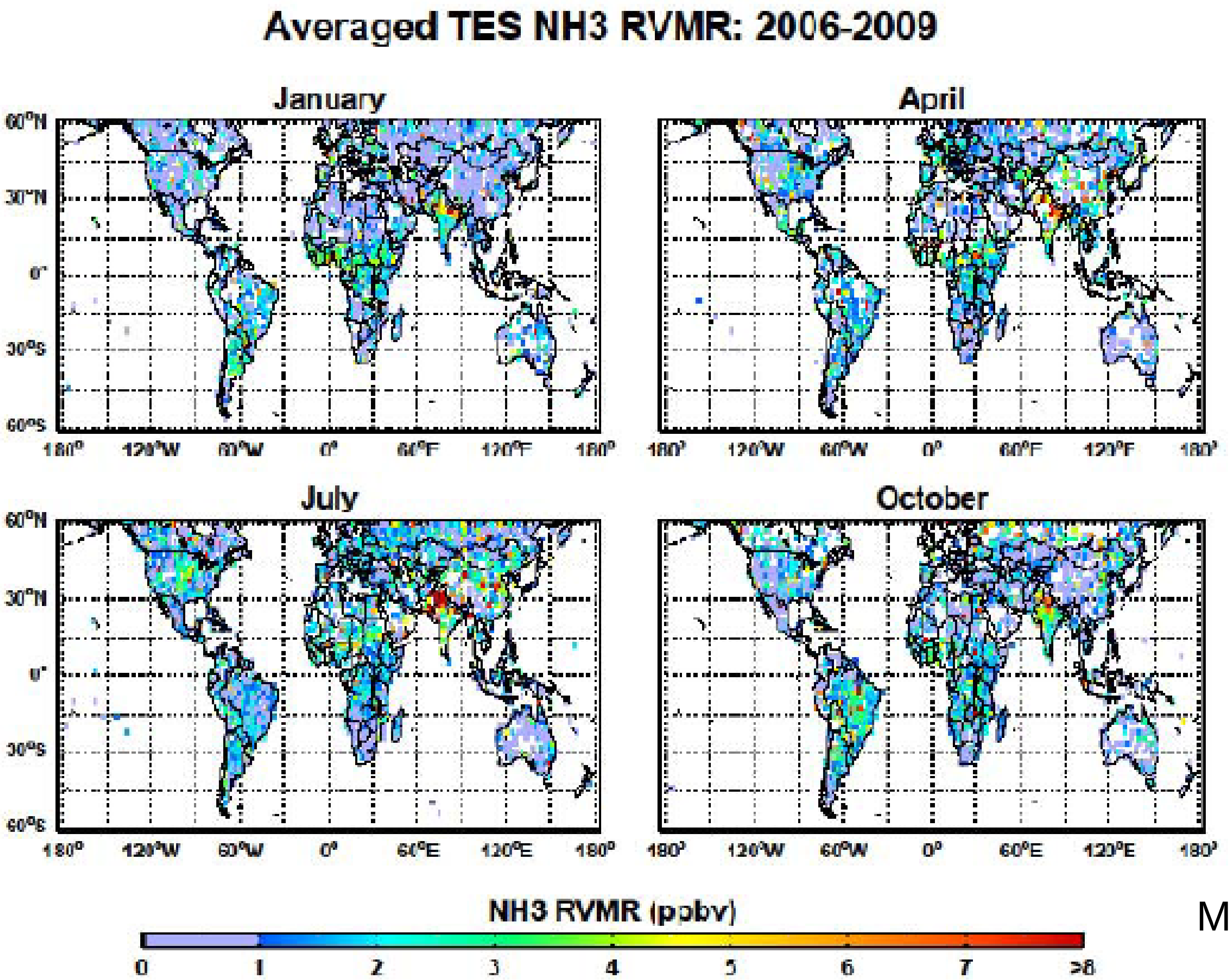
- Global emissions too low?
- agricultural practices
- lack of emission factors- or emission process knowledge (T dependency)= only implicitly included.
- temporal variations
- heterogeneous emissions (and removal) on farm scale
- natural cycle- compensation point (is it the soil or vegetation?)

Chemistry: relatively straightforward (?)

- reaction with SO_4 and NO_3 in aerosol and clouds
- Interplay with subgrid mixing, timescales

Removal: not so simple!

- dry deposition (compensation point? See emissions)
- Co-deposition, surface humidity
- Soil characteristic
- models predict dry deposition between 30-70 % of total deposition
- wet deposition, difficulties like for all aerosol components



A factor of two
higher than GEOSCHEM

Mark Shepard, ACP, 2011

Budgets?

Overall global budget depends on the emissions

Wet deposition: not constraining enough.
Aerosol measurements; strongly depends on NO3 and SO4
Models do wet and dry deposition very different

NH_3 at the surface hardly routinely measured (somewhat improving)

Point measurements very local- implication for large scales difficult to assess

Satellite data are very welcome addition- complex interpretation in model context

Earth system approach to couple atmosphere-biosphere-ocean

Thank you!