Aerosol Chemistry

Global modeling of nitrate and ammonium: Implications for atmospheric radiation, chemistry, and ecosystems

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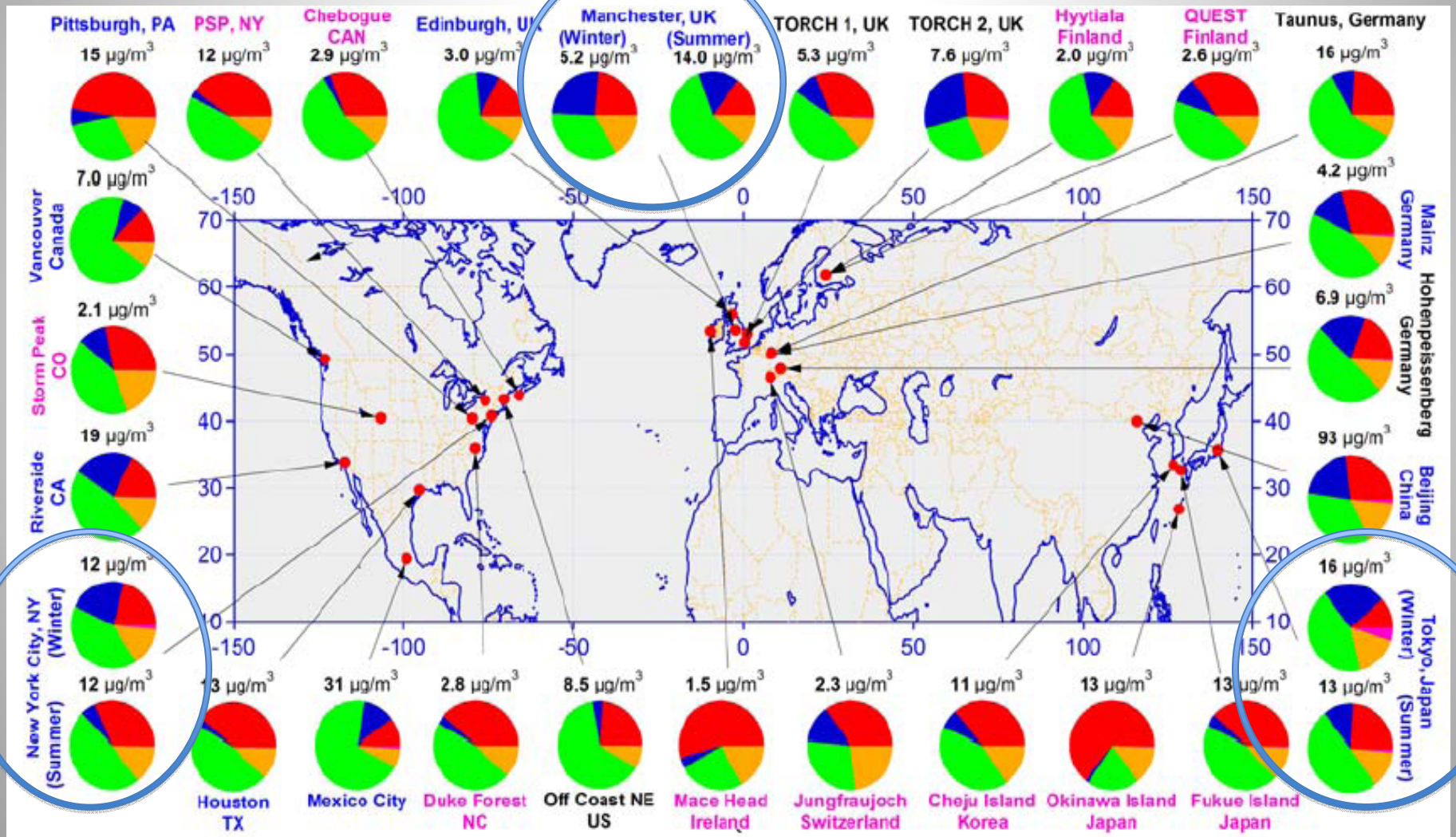
Motivation

1. Nitrate and ammonium are among the most abundant aerosol components in present atmosphere.
2. They have important roles in atmospheric radiation field, atmospheric chemistry and ecosystem.
3. IPCC reported “very low” level confidence in the study of their mass distributions and their environment impacts.

Measurement aerosols (PM1) at surface

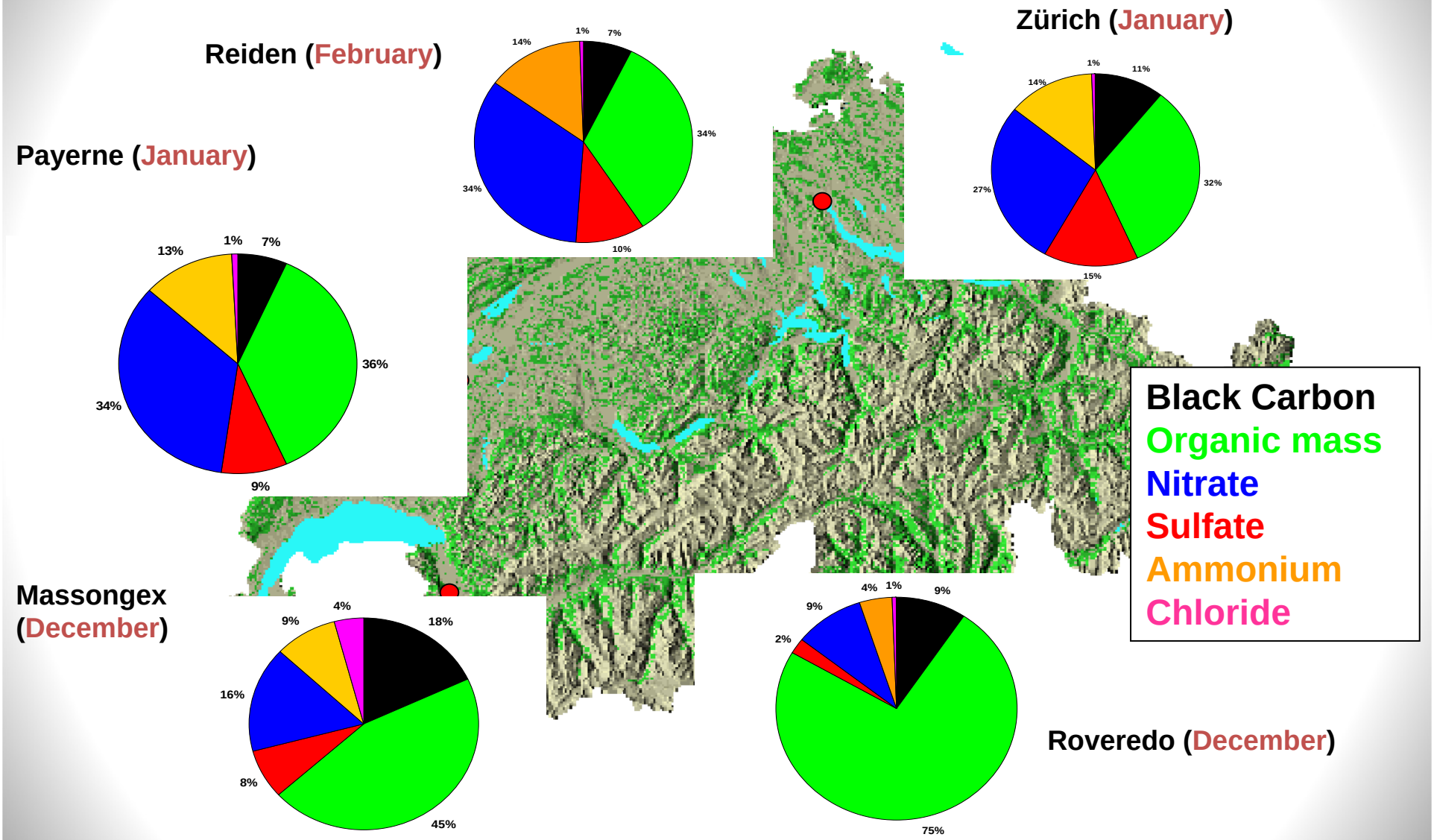
Organics sulfate nitrate ammonium chloride Zhang et al., 2007



Urban / Urban downwind / Rural remote

Field Measurements

Average compositions in winter



Urs Baltensperger et al., 2008

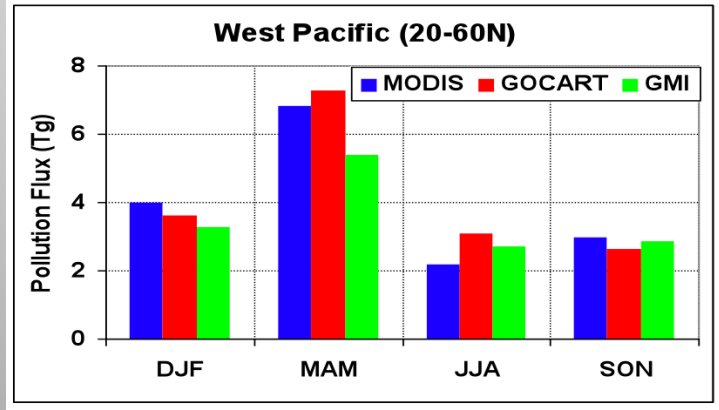
1. Simulate nitrate and ammonium in GEOS-5 online and offline systems

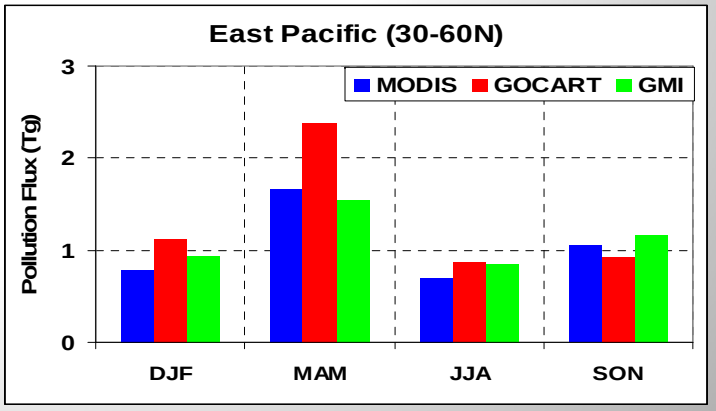
In order to calculation Nitrate, the model need to

1. simulate aerosol mass and AOT for sulfate, black carbon, organic carbon, dust, and sea salt (note: GOCART-like dust and sea-salt)

Aerosol mass

Yu et al., 2008

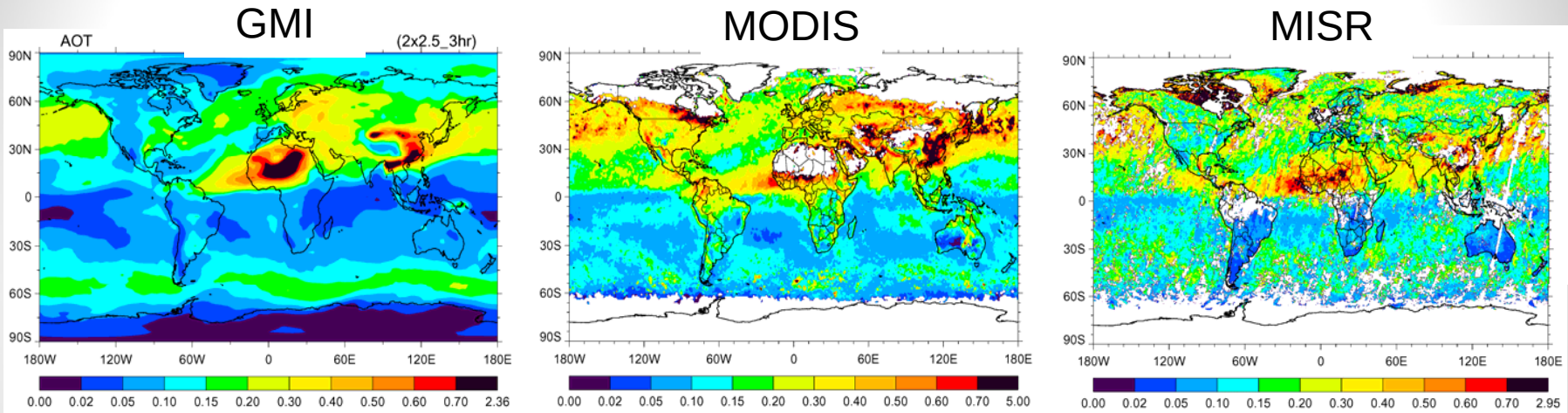




Example of current GMI aerosol simulations

AOT

Bian et al., 2009



2. have nitrogen chemistry (from Combo model)

In order to calculation Nitrate, the model also need to

3. have a coupled aerosol and gas phase chemistry (leverage with other proposals: Rodriguez, Colarco, and economic stimulus plan),

4. implement ammonia chemistry,

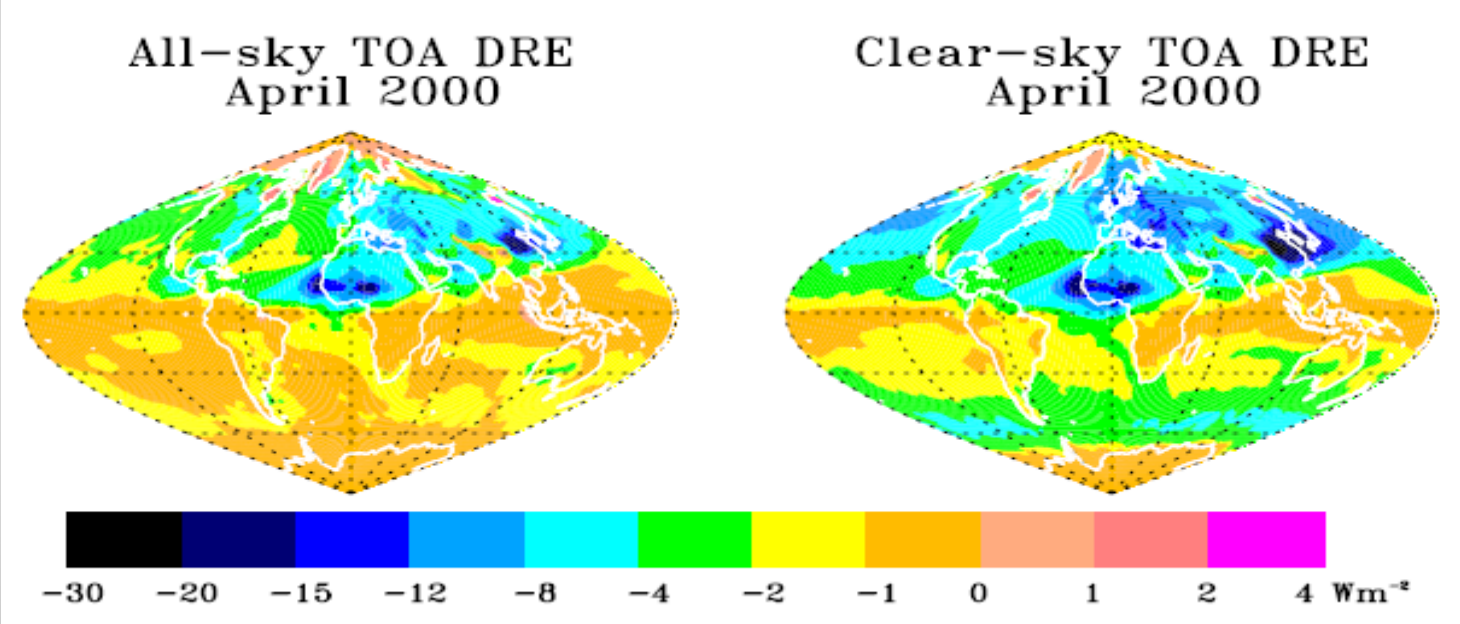
5. implement a thermodynamic equilibrium model (EQSAM3, by collaborating with Dr. Swen Metzger).

3-5 are proposed model developments

Nitrate simulation development benefits both GEOS5 online and offline models

2. Assess the effects of nitrate aerosols on atmospheric radiative field

GMI simulate aerosol forcing (w/o $NO3^-$)



A direct RF of -0.10± -0.10 W/m^2 is tentatively adopted for nitrate in IPCC 2007: few estimates and large uncertainty

Large uncertainty is mainly due to uncertainty in nitrate aerosol size distribution.

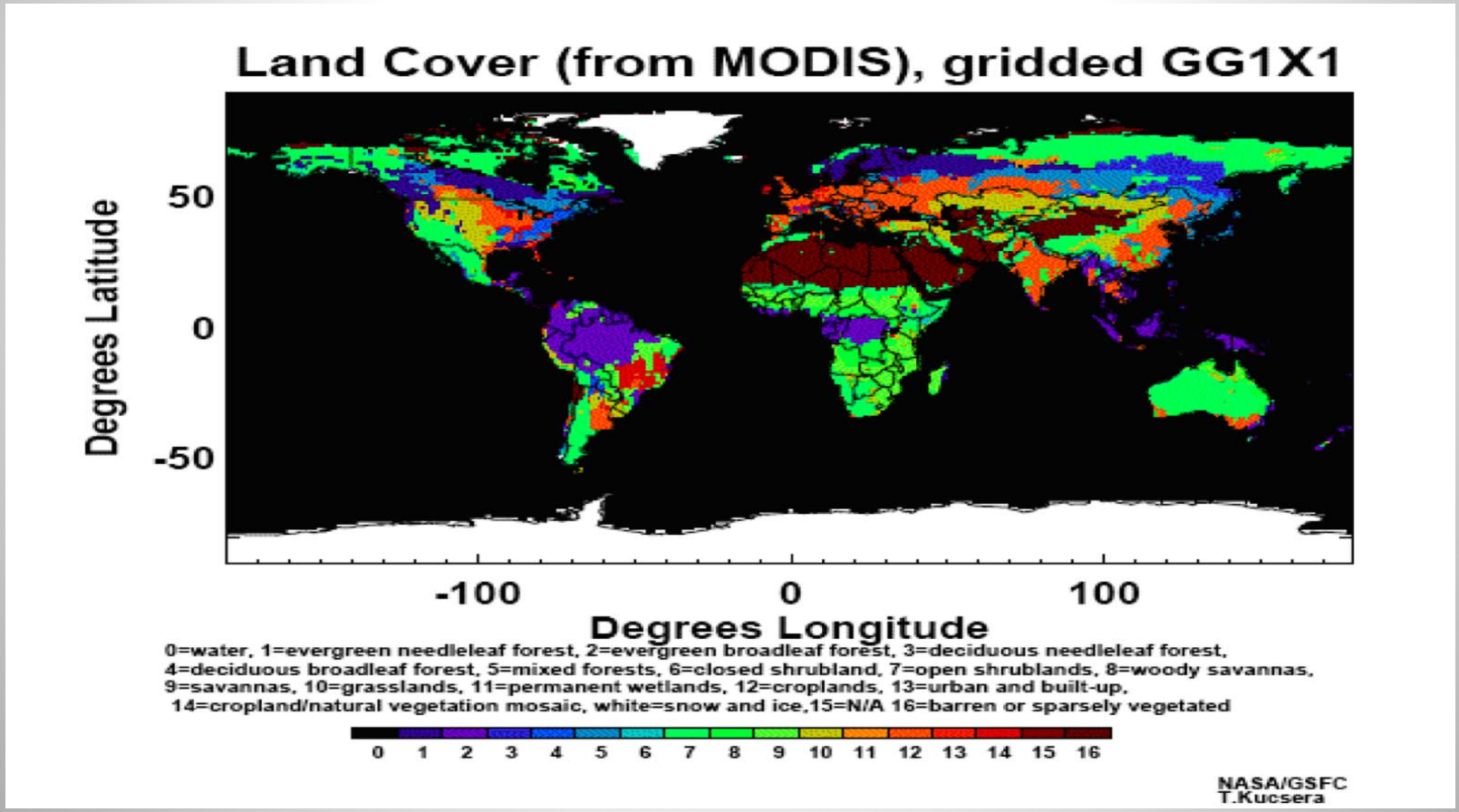
3. Examine the effect on atmospheric chemistry

Five simulations:

1. GMI runs with Combo nitrogen gas phase chemistry only,
2. GMI runs with an integrated package of nitrogen chemistry, ammonium chemistry, and EQSAM3. Photolysis rates and heterogeneous reactions are unchanged,
3. Similar to Exp2 but with an additional impact of the nitrate aerosols on photolysis rates,
4. Similar to Exp2 but with an additional impact of nitrate aerosols on heterogeneous reactions,
5. Similar to Exp2 but with in inclusion of the nitrate aerosol impact on both photolysis and heterogeneous reactions.

4. Estimate N deposition to different ecosystems

Approach 1: Estimate N deposition to each ecosystem and compare it with the corresponding N critical load



4. Estimate N deposition to different ecosystems

Approach 2: Estimate the fraction of global and regional area

(Europe, North America, and Asia) which receives N

deposition in excess of the “critical load” threshold of

1 gNm-2yr-1 [Dentener et al., 2006, Lamarque et al., 2005]

5. Estimate the impacts of future climate change and emission scenario on nitrates and their atmospheric impacts

Four simulations:

1. clim2000-emi2000: baseline simulation
2. clim2050-emi2000
3. clim2000-emi2050
4. clim2050-emi2050

One year (2050) met field from GEOS-5 (Steven Pawson) which will be driven by projected GH gases from the IPCC.

Future emission scenarios will be projected from the same storyline (Bryan Duncan)

Timeline

5.2. Schedule

Activity	*Year 1*	*Year 2*	*Year 3*	*Year 4*
Implementation of EQSAM into GMI	x			
Implementation of ammonia chemistry into GMI	x			
Simulate nitrate distribution at current day	x	x		
Model evaluation with observations	x	x	x	x
Analysis of nitrate impact on aerosol forcing	x	x	x	x
Analysis of nitrate on trop chem	x	x	x	x
Analysis of nitrate on ecosystem	x	x	x	x
Simulate nitrate at year 2050 and repeat above analyses			x	x

Proposed model developments

NH3 emissions

Source	NH3 (Tg N/yr)
Domesticated animals	21
Fertilizers	9
Human bodies	2.6
Biofuel	1.6
Industry	0.2
Fossil Fuel	0.1
Ocean	8.2
Biomass burning*	5.9
Crop	3.5
Soils	2.4
Wild animals	0.1

* for year 2004

Proposed model developments

Information for EQSAM3:

Thermodynamic and chemical equilibrium and internally mixed of aerosol particles

Partitioning depends on: T, RH, and various tracer concentrations

Unique of EQSAM: aerosol water mainly depends on ambient RH, aerosol mass and the type of solute, so that parameterization of single solute molalities and activity coefficients ban be defined only depending on the type of solute and RH. This leads the entire aerosol equilibrium composition can be solved analytically, i.e. non-iteratively, which considerably reduced computational time.

	DMS		SO_2		SO_4^{2-}		MSA	
Source	17.7		92.8		27.7		1.4	
	Emission	17.7	Anthropogenic	65.1	Gas-phase prod.	6.4	Chemical prod.	1.4
			Biomass buring	2.3	In-cloud prod.	21.3		
			Volcanoes	4.8				
			Ship emission	4.3				
			Photochemical	16.3				
Sink	17.7		92.8		27.7		1.4	
	Reaction with OH	11.6	Gas-phase loss	6.4	Dry dep.	3.9	Dry dep.	.1
	Reaction with NO_3^-	4.8	In-cloud loss	21.3	Wet dep.	23.8	Wet dep.	1.3
	Reaction with O_3	1.3	Dry dep.	38.7				
			Wet dep.	26.6				
Burden	0.055		0.20		0.46		0.021	
Lifetime	1.1		0.8		6.1		5.6	

Table 6.12: Global DMS, SO_2, sulfate, and MSA budgets. Units are Tg S yr^{-1} for source and sink, Tg S for burden, and days for lifetime.

Nitrate burden: 0.2-0.51 Tg N/yr
Ocean emission: ~ 14% of total

DMS contribute to so4: 19% [IPCC 2007]

Lifetime of HNO3 [Balkanski et al., 1993]: in PBL ~ 5 days, in UT ~ 2-3 weeks

RF:
Total direct aerosol RF: -0.5 ±0.4 w/m2 (midium-low level of scientific understanding.
The direct RF of the individual aerosol species is less certain than the total direct aerosol RF:
Sulfate: -0.4 ± 0.2 W/m2
Fossil fuel OC: -0.05 ± 0.05 w/m2
Fossil fuelBC: 0.2 ± 0.15 w/m2
Biomass burning 0.03 ±0.12 w/m2 (strongly influenced by aerosol overlying clouds)
Nitrate: -0.1 ± 0.1 w/m2
Mineral dust: -0.1 ±0.2 w/m2