Analysis for AeroCom III nitrate experiment

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AeroCom 2016

Three objectives:

(1) address the diversity of nitrate simulations by the AeroCom models and understand the reasons for the intermodel differences,

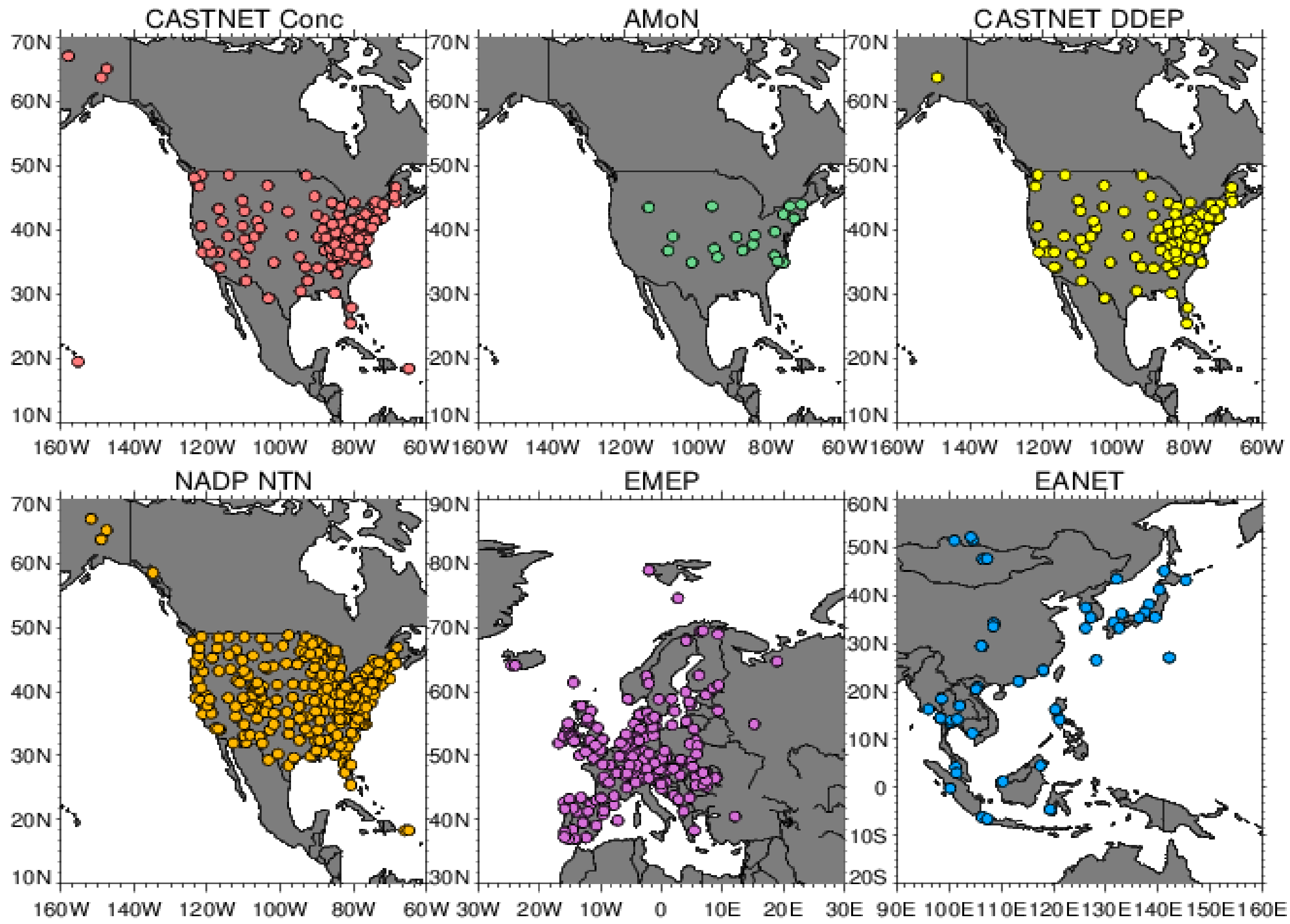
(2) compare model simulated nitrate with measurements from ground networks, aircraft campaigns, and satellite retrievals,

(3) investigate how nitrate formation changes in different models in response the perturbation of precursor emissions and meteorological conditions.

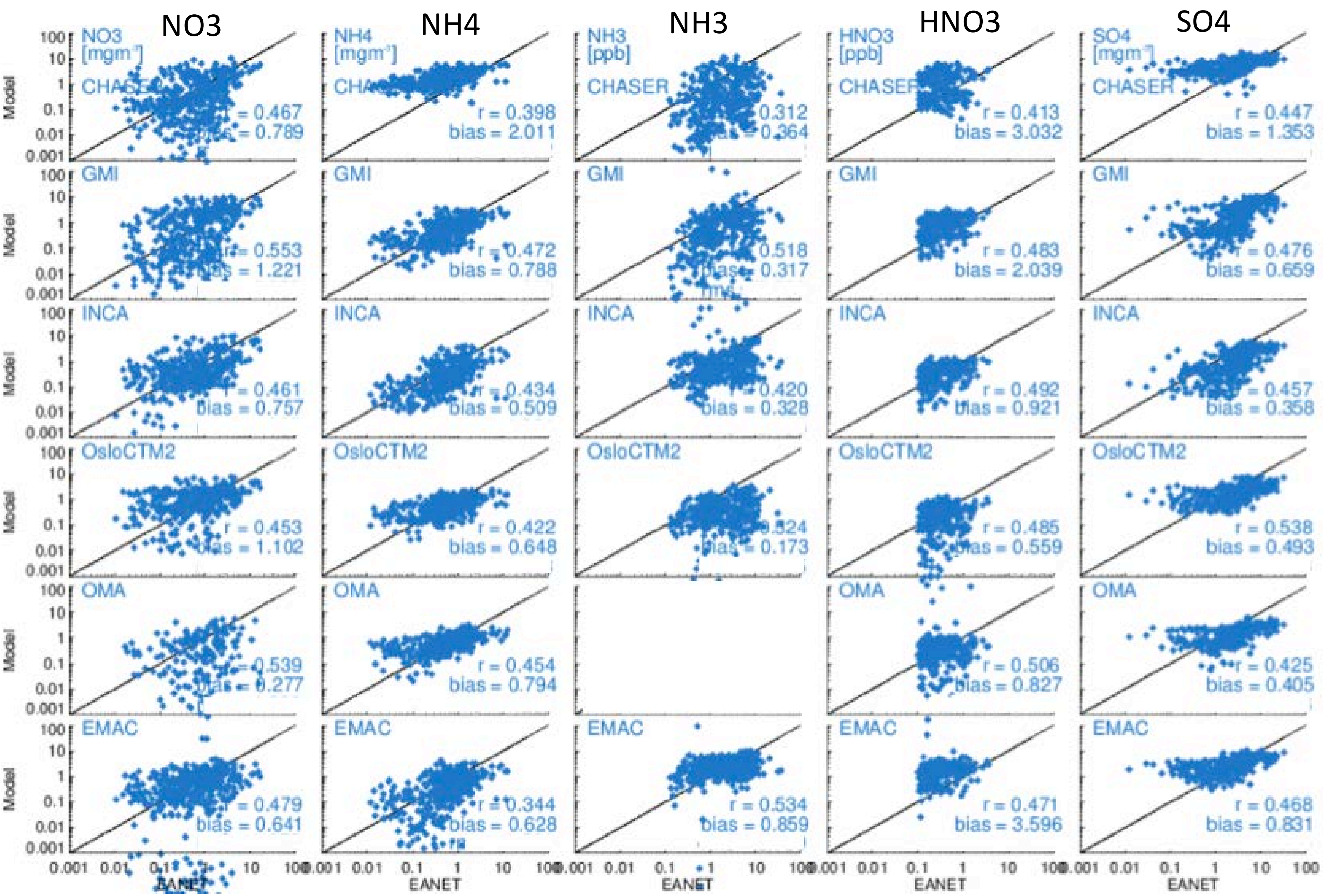
Current Status

model	modeler	Current status
CHASER	Kengo Sudo	Submitted
EMAC	Karydis Vlassis	Submitted
EMEP	Svetlana G. Tsyro	Submitted partially
GISS-MATRIX	Susanne Bauer Kostas Tsigaridis	Submitted partially
GISS-OMA	Susanne Bauer Kostas Tsigaridis	Submitted partially
GMI	Huisheng Bian	Submitted
INCA	Didier Haugluztaine	Submitted
OsloCTM2	Gunnar Myhre Ragnhild B. Skeie	Submitted but will update
OsloCTM3	Gunnar Myhre Ragnhild B. Skeie	In processing

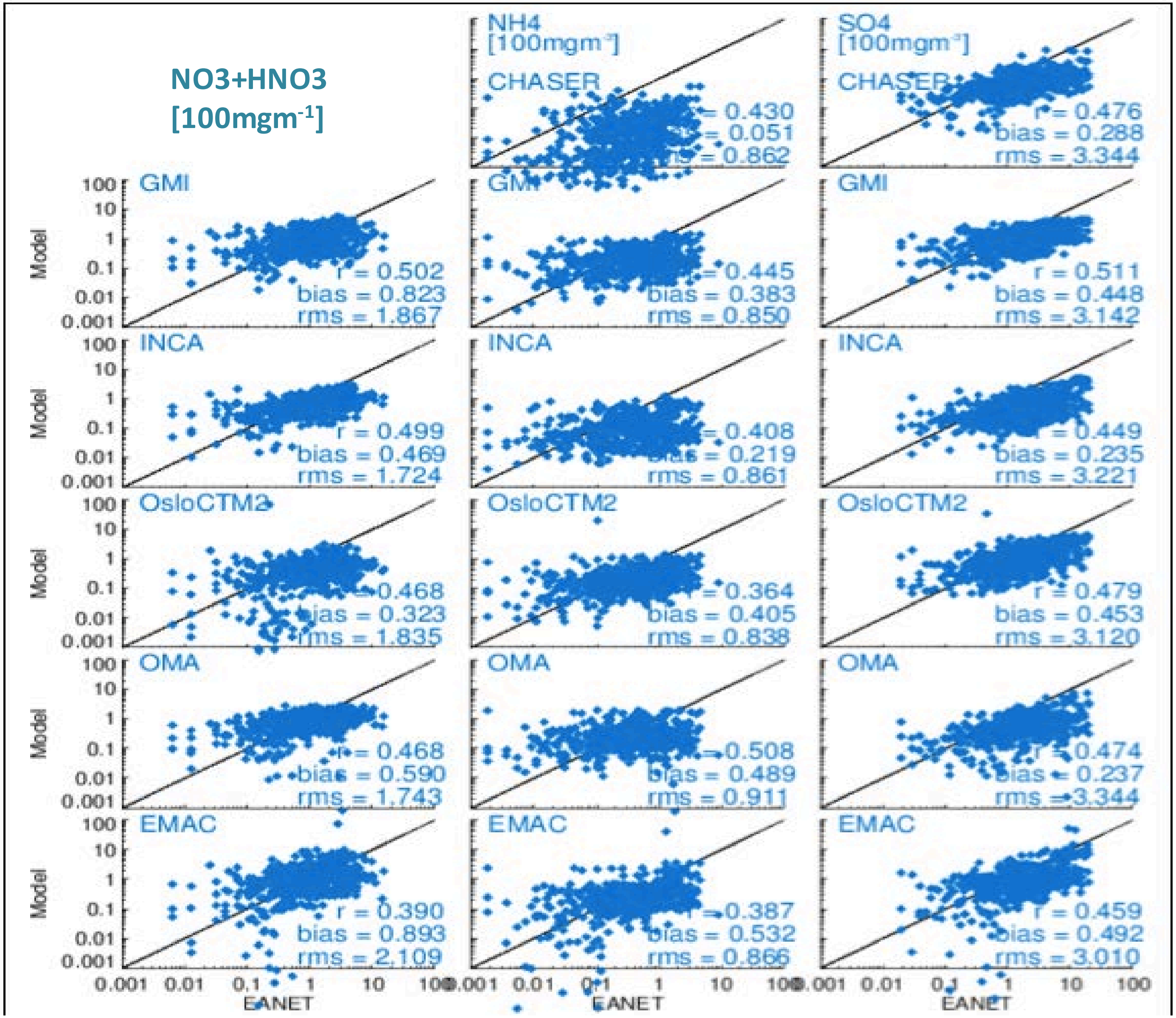
Surface observation sites



Surface mixing rations between EANET and the models over East Asia

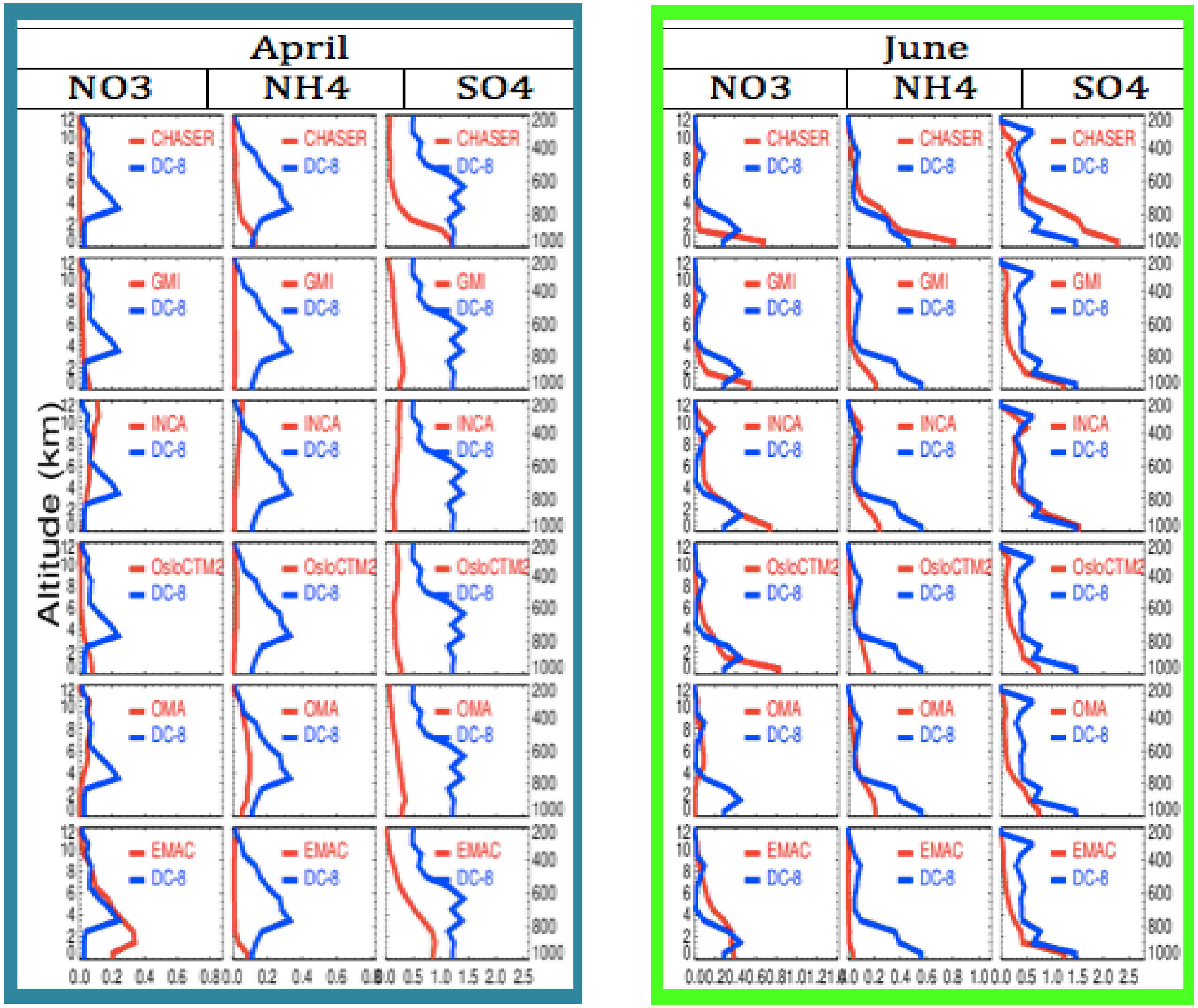


Wet deposition between EANET and the models over East Asia



Horizontal resolutions of the models range 2-3 degree

Tracer vertical profiles between ARCTAS and the models

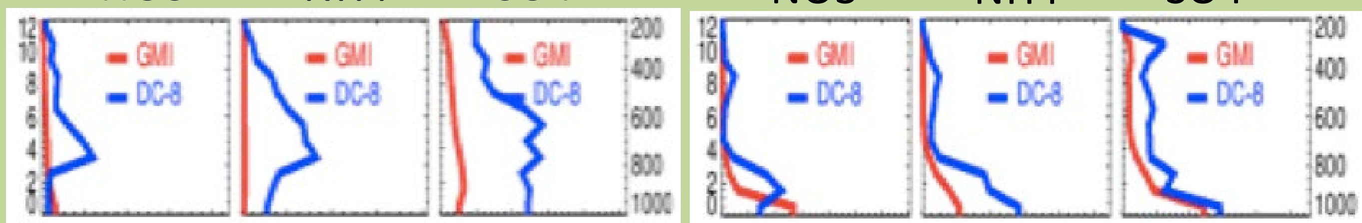


Tracer vertical profiles between ARCTAS and the models

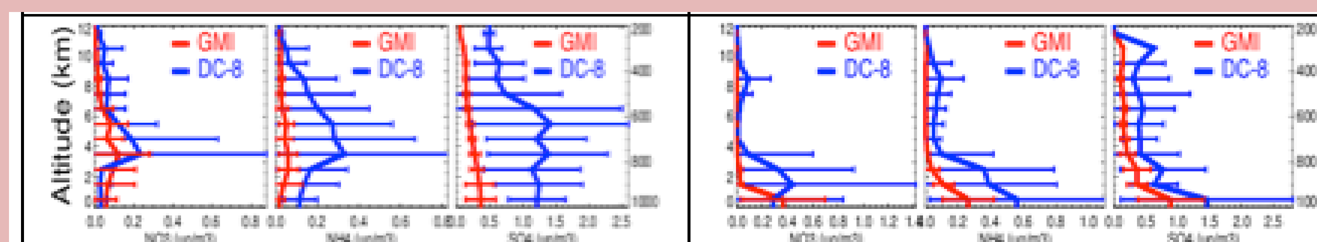
April | June

NO3 NH4 SO4 | NO3 NH4 SO4

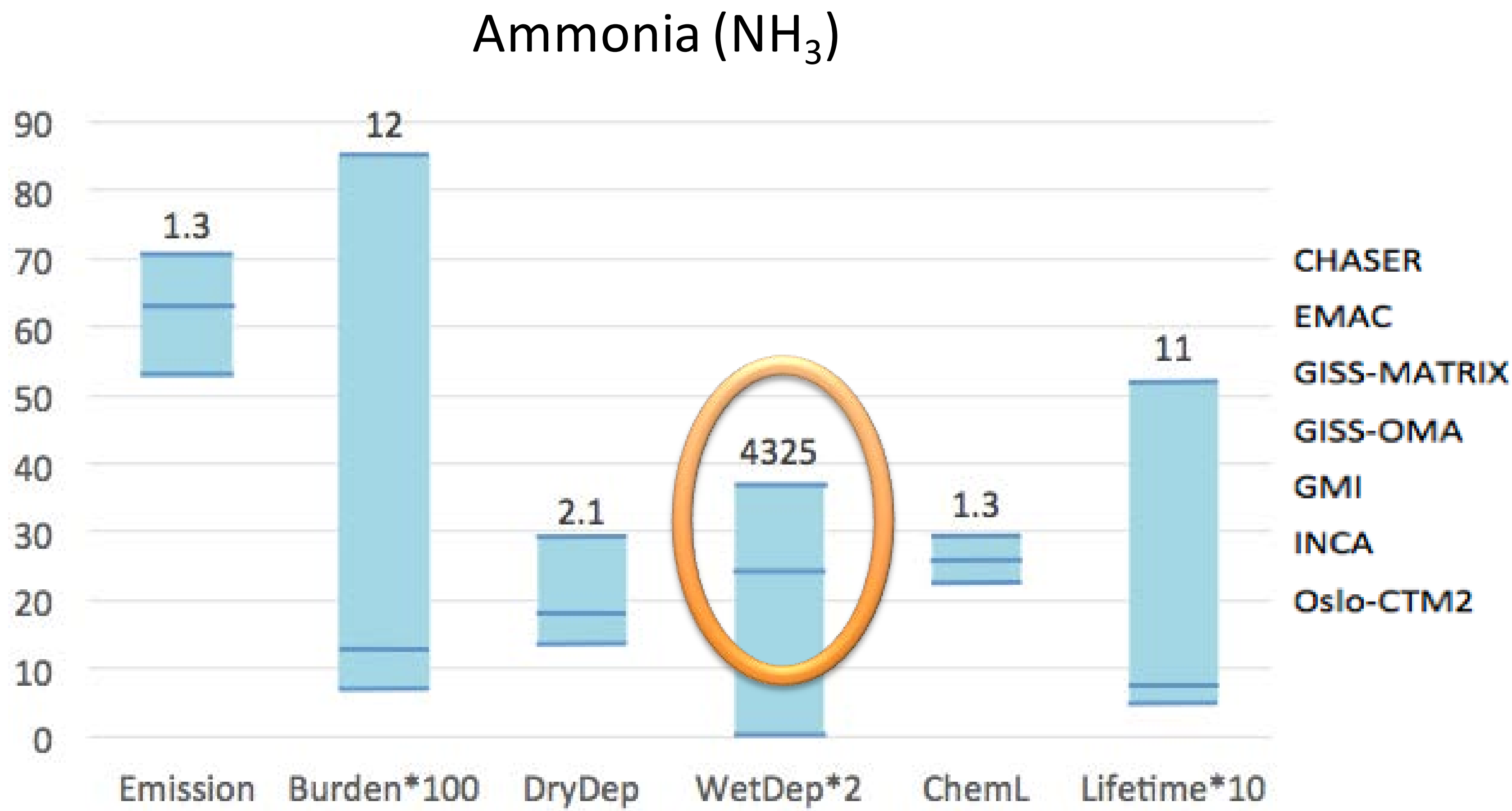
Monthly GMI



Daily GMI



Compare among models



Diversity in wet deposition is huge!

NH_3 Wet Deposition

Henry's Law constant H(T) of pure water

$$H(T) = H^{\ominus} * \exp\left(-\frac{\Delta_{sol}H}{R}\left(\frac{1}{T} - \frac{1}{T^{\ominus}}\right)\right)$$

Further correction by pH to give H(T,pH)

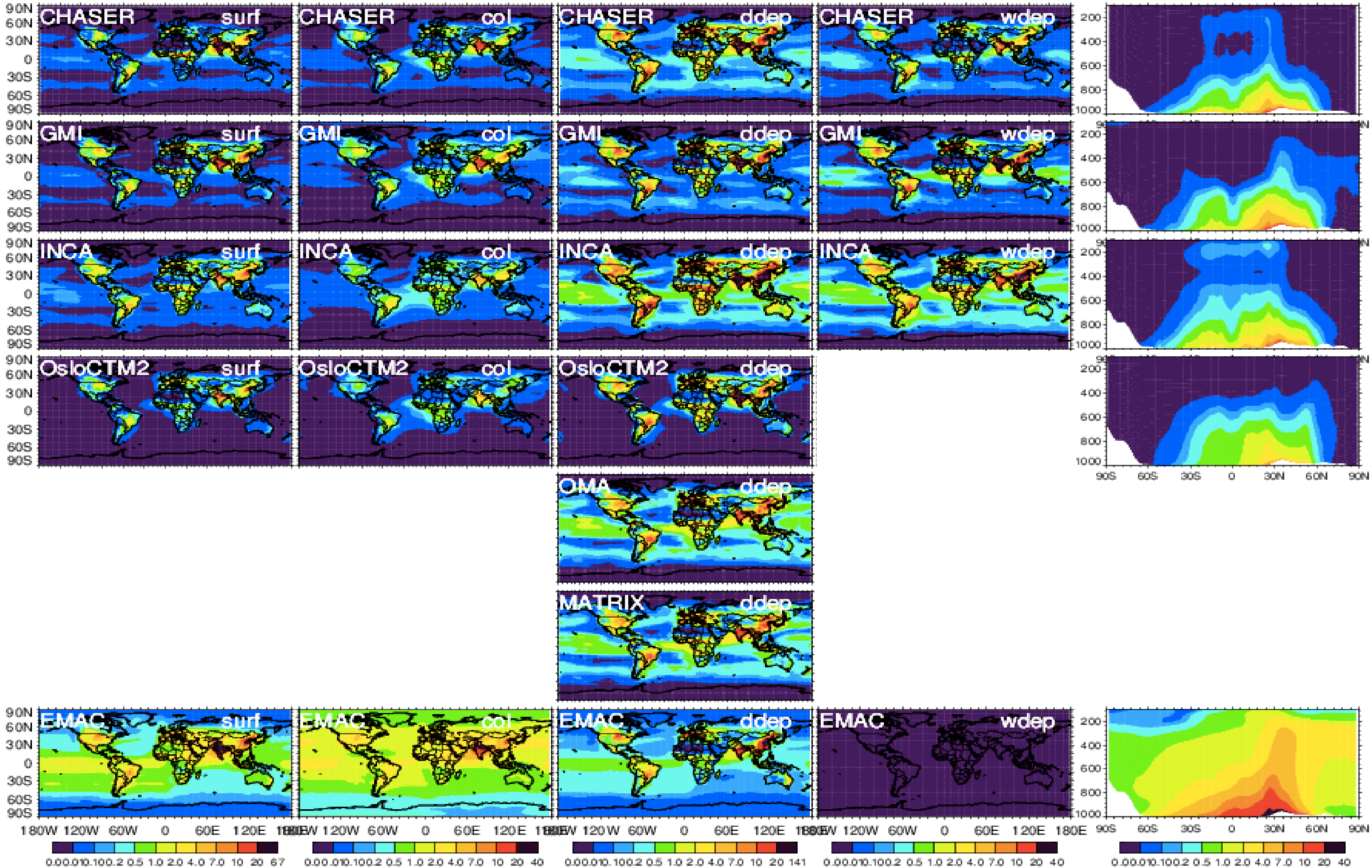
$$H(T, pH) = H^{\ominus} * f(pH) * \exp\left(-\frac{\Delta_{sol}H}{R}\left(\frac{1}{T} - \frac{1}{T^{\ominus}}\right)\right)$$

To give Effective Henry's law constant $H^{\ominus *} = H^{\ominus} * f(pH)$

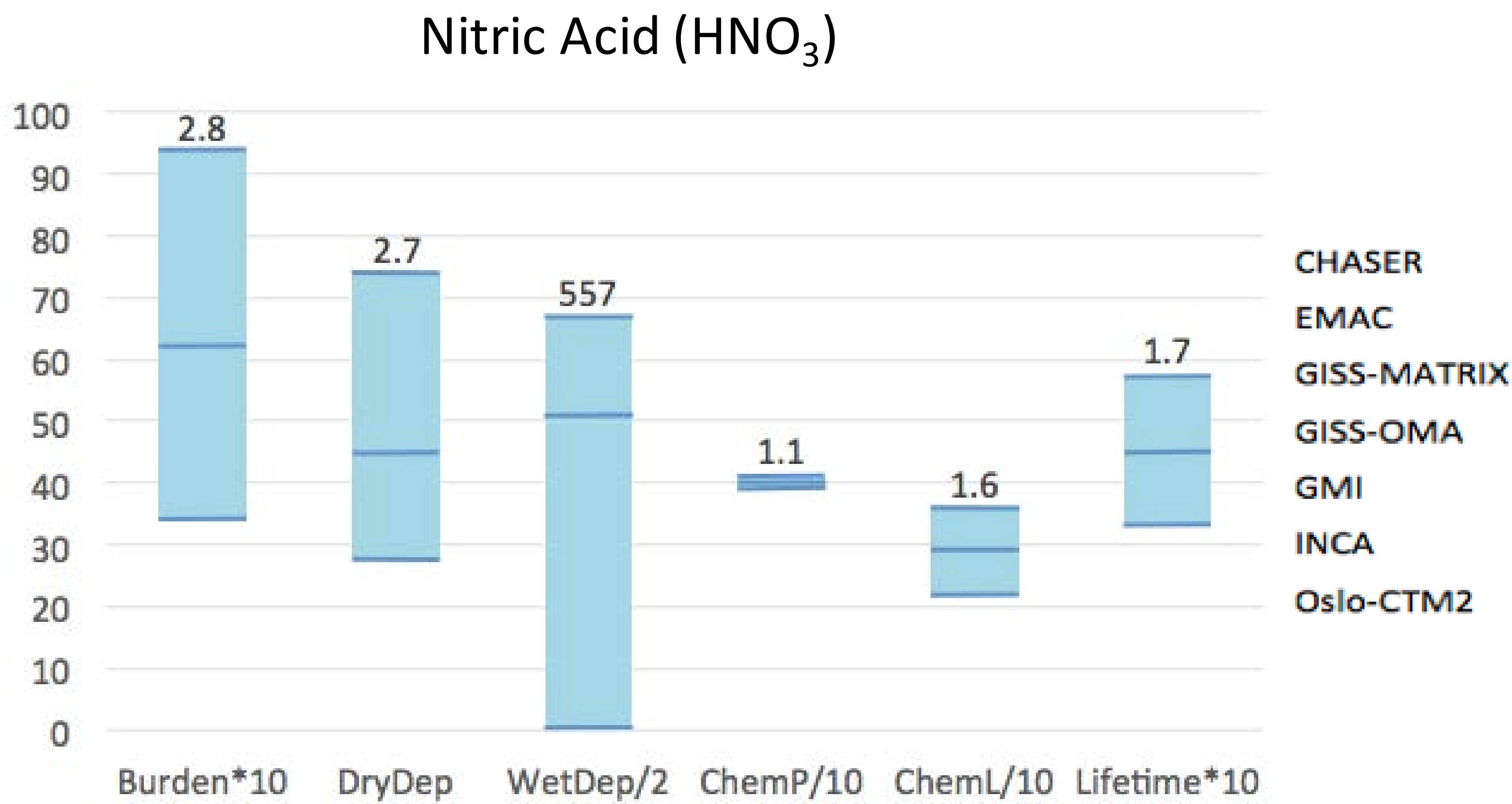
AeroCom Model	$H^{\ominus *}$ (M/atm)	f(pH) = 1.5*10^(9-pH)	$-\Delta_{sol}H/R$ (K)
CHASER	$3.0*10^5$	f(5.5)	3400
EMAC	$5.8*10^1$	1	4085
GISS-MATRIX	$1.0*10^1$	1	3416
GISS-OMA	$1.0*10^1$	1	3416
GMI	$1.05*10^6$	f(5.1)	4200
INCA*	$7.4*10^1$	f(pH)	3400
Oslo-CTM2	$3.3*10^6$	f(4.5)	

Compare among models

NH_3 distributions of surface, column, drydep, wetdep, and vertical zonal mean



Compare among models

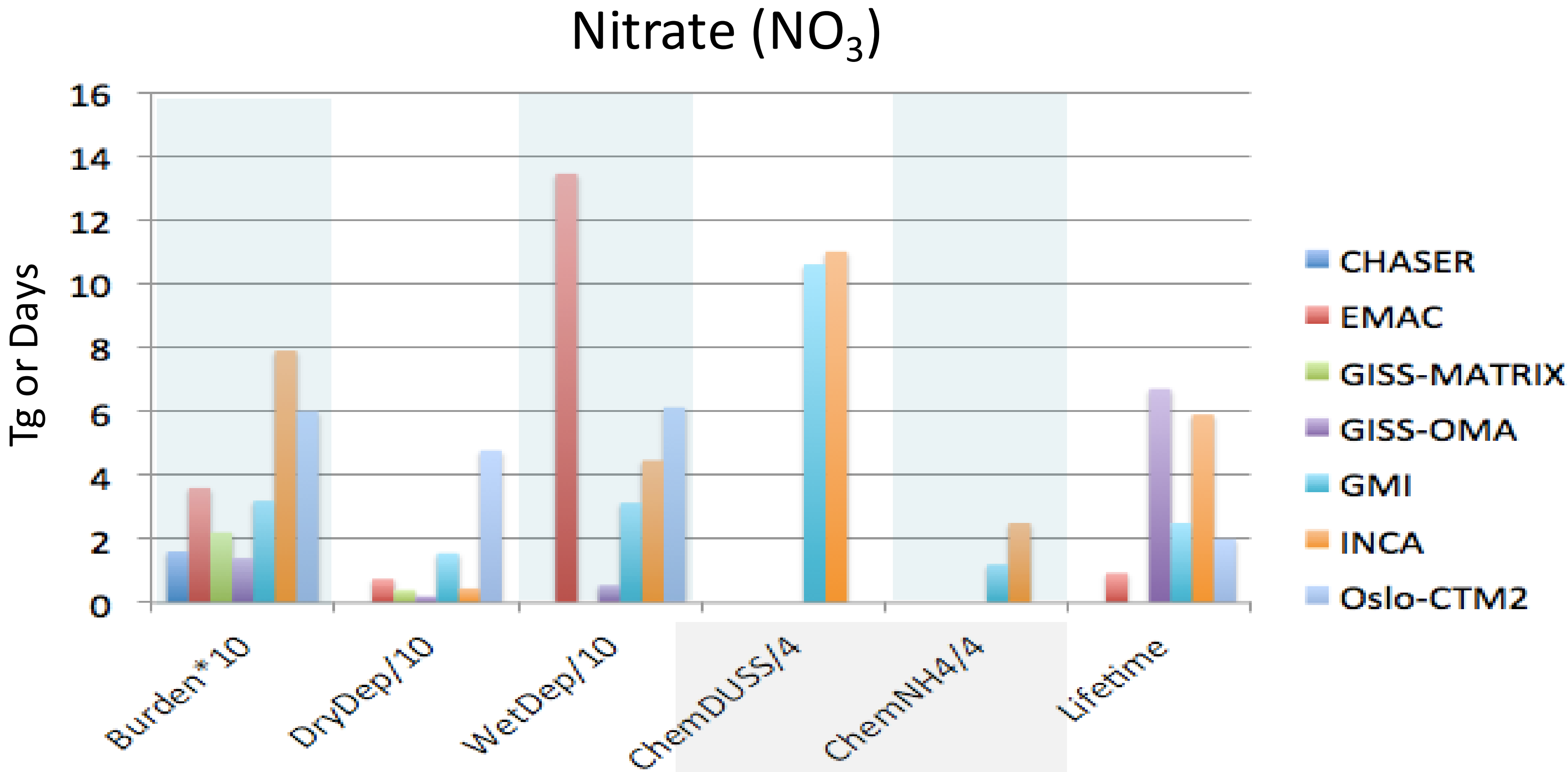


All models use online coupling NOx-HOx-O3-aerosol chemistry mechanism

There is almost 3 times difference in HNO3 global burden

Compare among models

> 80% of nitrate formation via ChemDUSS



Process	Mean	Range	Ratio (Max/Min)
Burden	0.37	0.14 – 0.79	5.6
DryDep	13.6	2.1 – 47.7	22.7
WetDep	55.4	5.5 – 134.4	24.4
ChemDUSS	43.2	42.3 – 44.1	1.04
ChemNH4	7.3	4.8 – 9.8	2.0
Lifetime	3.6	0.93 – 6.7	7.2

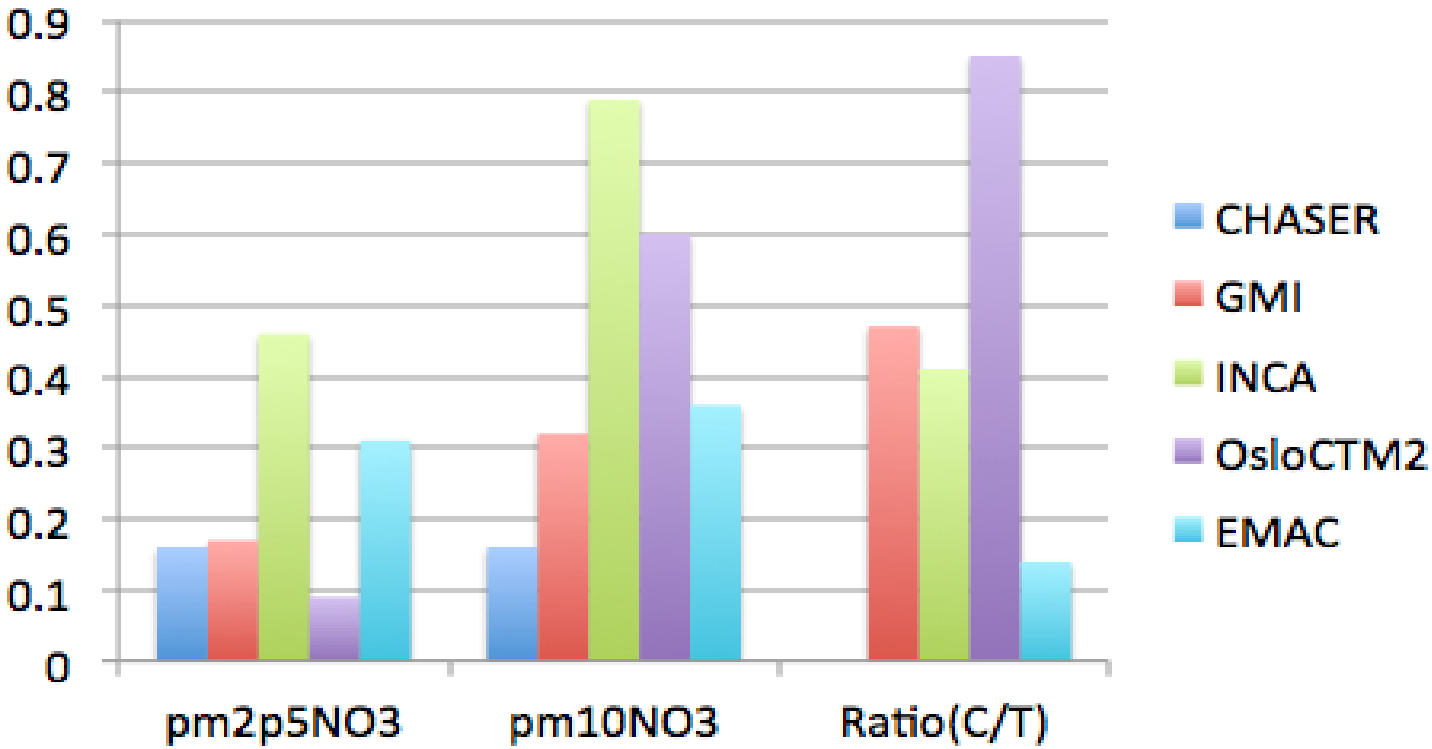
Chemical mechanism of nitrate formation

AeroCom Model	CHEM-EQM	CHEM Dust	CHEM Sea Salt	METHOD (CHEMDUSS)	HNO3 produced
CHASER	ISORROPIA-I	NO	NO	----	CHASER
EMAC	ISORROPIA-I I	YES	NO	ISORROPIA-I I	ECHAM5-MESSy2
GISS-MATRIX	ISORROPIA-I I	YES	NO	ISORROPIA-I I	GISS-MATRIX
GISS-OMA	ISORROPIA-I I	YES	NO	ISORROPIA-I I	GISS-OMA
GMI	RPMAIRS	YES	YES	First order loss rate	GMI
INCA*	INCA	YES	YES	First order loss rate	LMDzv4-INCA
Oslo-CTM2	EQSAM_v03d	NO	YES	EQSAM (run twice)	Oslo-CTM2

All models use online coupling NOx-HOx-O3-aerosol chemistry mechanism
Aerosols in all models in thermodynamically stable state except Oslo-CTM2 in metastable
All models are GCMs except GMI and Oslo-CTM2 that are CTMs

Compare among models

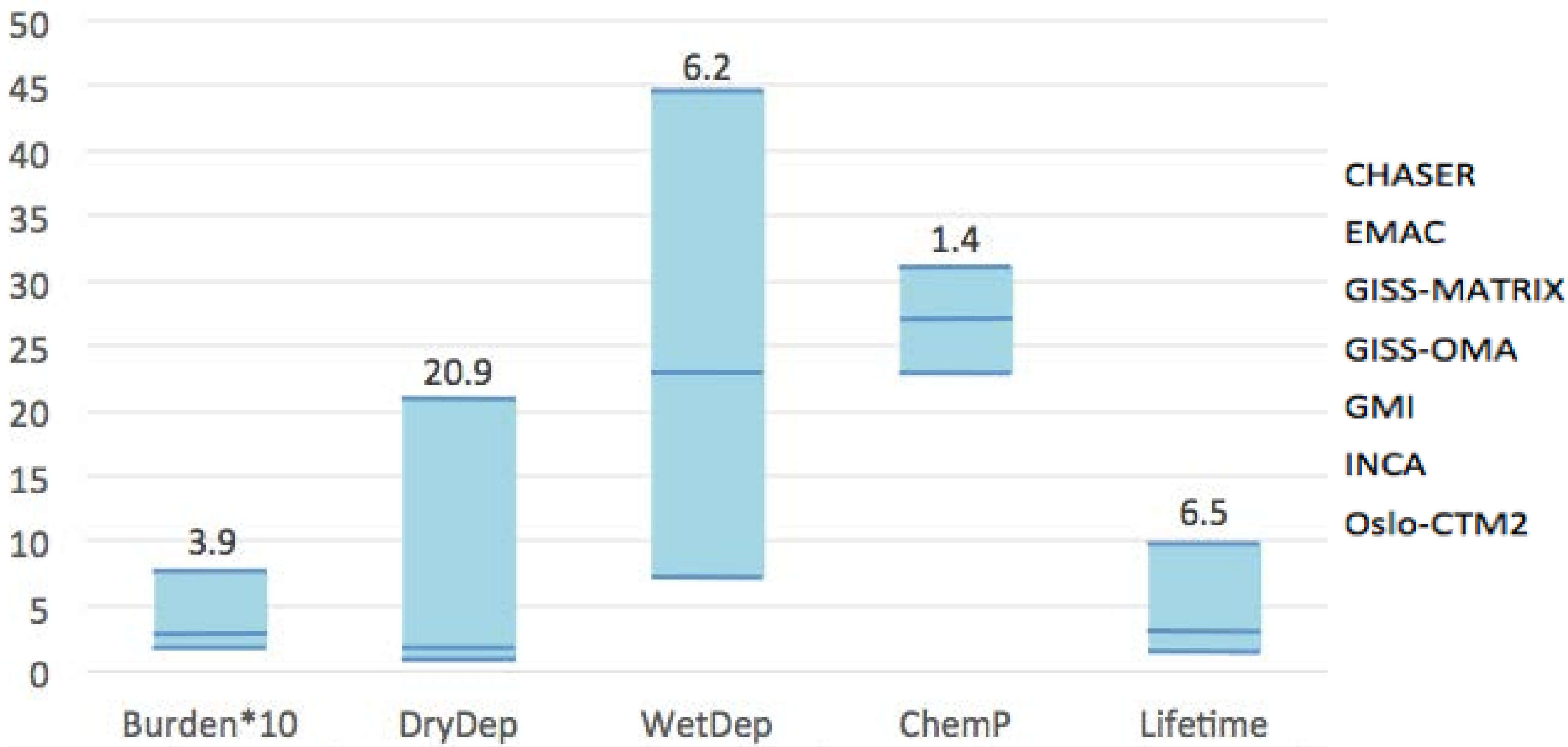
Global annual fine and coarse mode NO_3 and the coarse mode ratio



Coarse mode fraction ranges from 0 to 84%

Compare among models

Ammonium (NH_4)



Conclusion

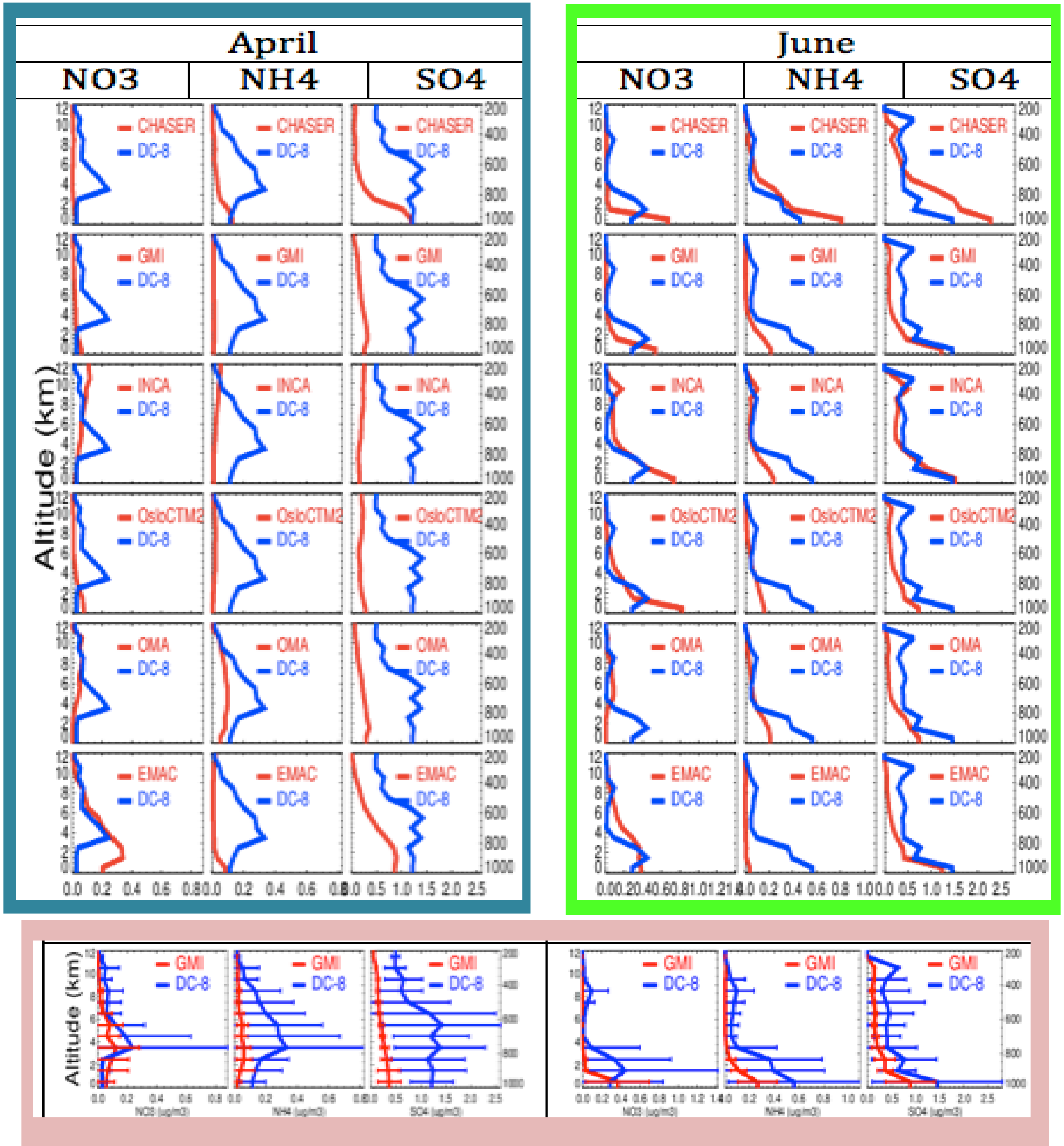
- The diversity of NO_3 simulation is larger than that of SO_4. The mean global burden of NO_3 is ~1/4 of that of SO_4 in 2008.
- The simulations of aerosol tracers (SO_4, NO_3, and NH_4) agree with observations better than gas precursors (NH_3 and HNO_3).
- NH_3 differs huge with and without applying pH correction for NH_3 wet removal.
- Improvement of wet and dry depositions is also needed for the simulation of HNO_3, NO_3, and NH_4.
- It is critical to correctly account for the NO_3 formation on dust and sea-salt.
- Size distribution varies dramatically among the models. A large fraction of NO_3 exists in a coarse mode according to measurements. Coarse mode fraction is typically larger than 60% over oceans.

Conclusion

Manuscript draft and figures/tables can be downloaded from
http://croc.gsfc.nasa.gov/gocart_4_aerocom/

Refer experiment information to
https://wiki.met.no/aerocom/phase3-experiments

Tracer vertical profiles between ARCTAS and the models



NH_3 Wet Deposition

Henry's Law constant:

$$H(T) = H^{\Theta} * \exp\left(-\frac{\Delta_{sol}H}{R}\left(\frac{1}{T} - \frac{1}{T^{\Theta}}\right)\right)$$

For NH_3, Henry's Law constant H^{Θ} at 298K is further corrected by pH value to get effective Henry's law constant $H^{\Theta*}$

AeroCom Model	$H^{\Theta*}$ (M/atm)	$-\Delta_{sol}H/R$ (K)	
CHASER	3.0e+5	3400	pH = 5.5
EMAC	5.8e+1	4085	
GISS-MATRIX	1.0e+1	3416	
GISS-OMA	1.0e+1	3416	
GMI	1.05e+6	4200	5.0
INCA*	7.4e+1	3400	
Oslo-CTM2	3.3e+6		4.5

* With an additional pH-correction

NH3 Wet Deposition

$$NH_3 + H_2O \Leftrightarrow NH_3 \cdot H_2O$$
$$NH_3 \cdot H_2O \Leftrightarrow NH_4^+ + OH^-$$

$$[NH_3^T] = [NH_3 \cdot H_2O] + [NH_4^+]$$
$$= p_{NH3} H^\Theta \left(1 + \frac{K_{al}[\mathrm{H}^+]}{K_w}\right)$$
$$\approx p_{NH3} \left(H^\Theta \frac{K_{al}[H^+]}{K_w}\right)$$

$K_{al} = [NH_4^+][OH^-] / [NH_3 \bullet H_2O] \approx 1.5 \times 10^{-5}$

$K_w = [H^+][OH^-]$

$= 1.0 \times 10^{-14}$ at 298 K in pure water

Therefore, effective Henry Law Constant is:

$$H^{\Theta^*} = H^\Theta \frac{K_{al}[H^+]}{K_w}$$
$$= 1.5 * 10^{+09} * 10^{-pH} * H^\Theta$$

$$\text{If pH} = 5, \text{then } H^{\Theta^*} = 1.5 * 10^{+04} * H^\Theta$$

pH ranging from 4.5 (Oslo-CTM2) to 5.5 (CHASER)