DIVERSITY OF VERTICAL DISTRIBUTION OF SULFUR SPECIES (SO_2+SULFATE) IN AEROCOM-II MODELS

Qian Tan[1,2], Mian Chin[3], Valentina Aquila[3], Michael Hoepfner[4], Andrew Rollins[5], Rushan Gao[5]

[1]BAERI, [2]NASA Ames, [3]NASA GSFC, [4]Karlsruhe Institute of Technology (Germany), [6]NOAA ESRL

ENVISAT/MIPAS, VIRGAS/POSIDON field campaign, multiple aircraft campaigns, and AeroCOM Project

Bay Area Environmental Research Institute

OUTLINE

- AEROCOM II Inter-comparison
  - ***Model-to-model difference in SO_2 and sulfate vertical distributions***
  - ***Comparison with available observations in UTLS***
  - ***Possible reasons for model-to-model differences***
    - **Sources and sinks budget**

Background & Motivation: observations show variations in the stratospheric aerosols in recent decades, while possible sources are not well understood.

Increase in background stratospheric aerosol observed with lidar at Mauna Loa Observatory and Boulder, Colorado

David Hofmann,[1,2] John Barnes,[1,3] Michael O'Neill,[1,2] Michael Trudeau,[1,2] and Ryan Neely[1,2]

Received 27 May 2009; revised 24 June 2009; accepted 6 July 2009; published 4 Augus

CALIPSO detection of an Asian tropopause aerosol

J.-P. Vernier,[1] L. W. Thomason,[1] and J. Kar[2]

Received 27 December 2010; revised 22 February 2011; accepted 28 February 2011; published

The Persistently Variable "Background" Stratospheric Aerosol Layer and Global Climate Change

S. Solomon,[1,2*] J. S. Daniel,[1] R. R. Neely III,[1,2,5,6] J.-P. Vernier,[3,4] E. G. Dutton,[5] L. W. Thomason[3]

Major influence of tropical volcanic eruptions on the stratospheric aerosol layer during the last decade

J.-P. Vernier,[1,2] L. W. Thomason,[1] J.-P. Pommereau,[2] A. Garnier,[2] A. Hauchecorne,[2] L. Blanot,[2,4] C. Trepte,[1]

Received 25 March 2011; revised 21 April 2011; accepted 30 April 2011;

Recent anthropogenic increases in SO_2 from Asia have minimal impact on stratospheric aerosol

R. R. Neely III,[1,2,3] O. B. Toon,[1,4] S. Solomon,[5] J.-P. Vernier,[6,7] C. Alvarez,[2,3] J. M. English,[8] K. H. Rosenlof,[2] M. J. Mills,[8] C. G. Bardeen,[8] J. S. Daniel,[2] and J. P. Thayer[9]

Received 19 December 2012; revised 11 February 2013; accepted 15 February 2013; published 13 March 2013.

Post-Pinatubo Evolution and Subsequent Trend of the Stratospheric Aerosol Layer Observed by Mid-Latitude Lidars in Both Hemispheres

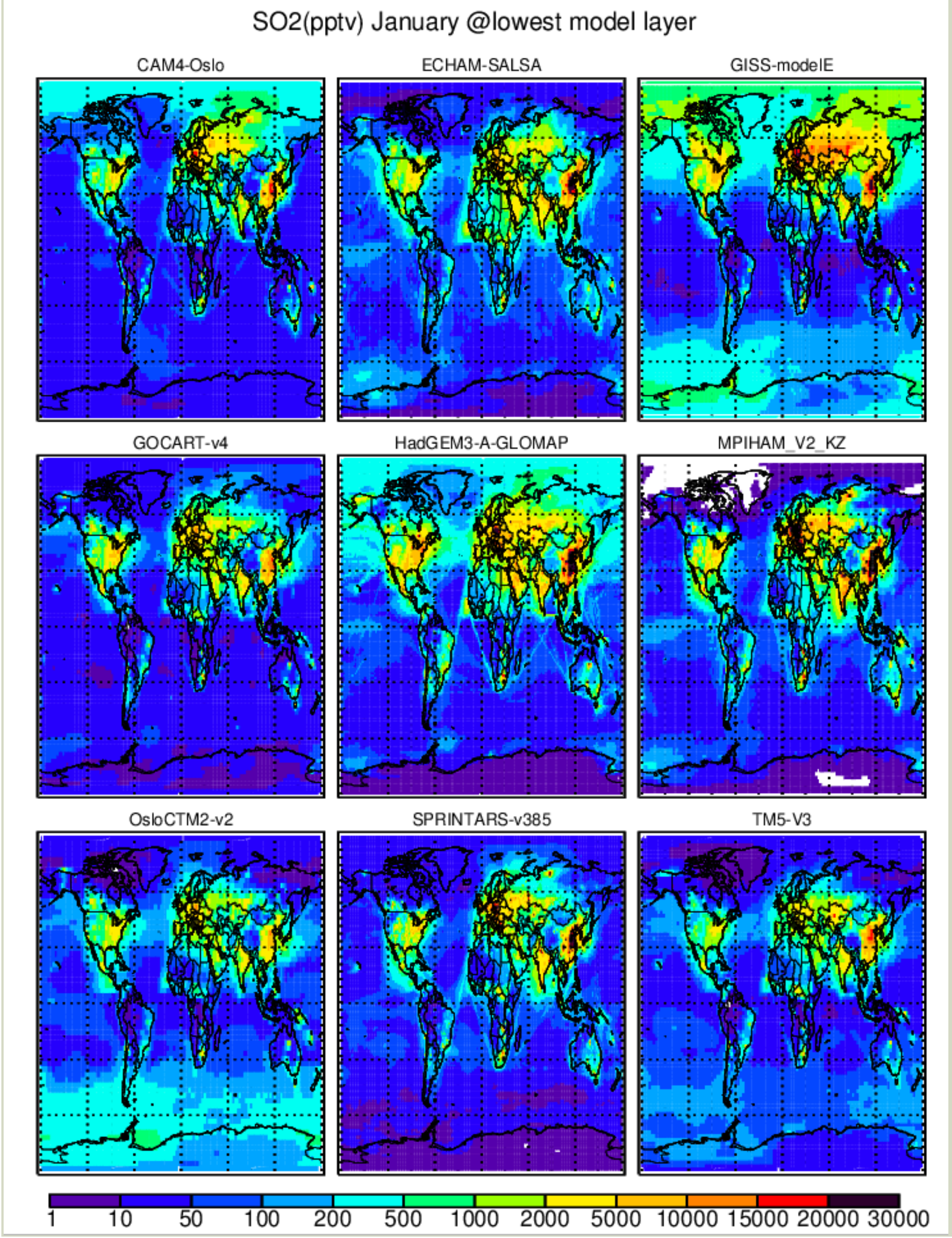
Tomohiro Nagai[1,3], Ben Liley[2], Tetsu Sakai[1], Takashi Shibata[3]
[1]*Meteorological Research Institute, Tsukuba,*
[2]*National Institute of Water and Atmospheric Research, La*
[3]*Graduate School of Environmental Studies, Nagoya Univer*
[4]*National Institute for Environmental Studies, Tsuk*

Increase in upper tropospheric and lower stratospheric aerosol levels and its potential connection with Asian pollution

J.-P. Vernier[1,2], T. D. Fairlie[2], M. Natarajan[2], F. G. Wienhold[3], J. Bian[4], B. G. Martinsson[5], S. Crumeyrolle[6], L. W. Thomason[2], and K. M. Bedka[2]

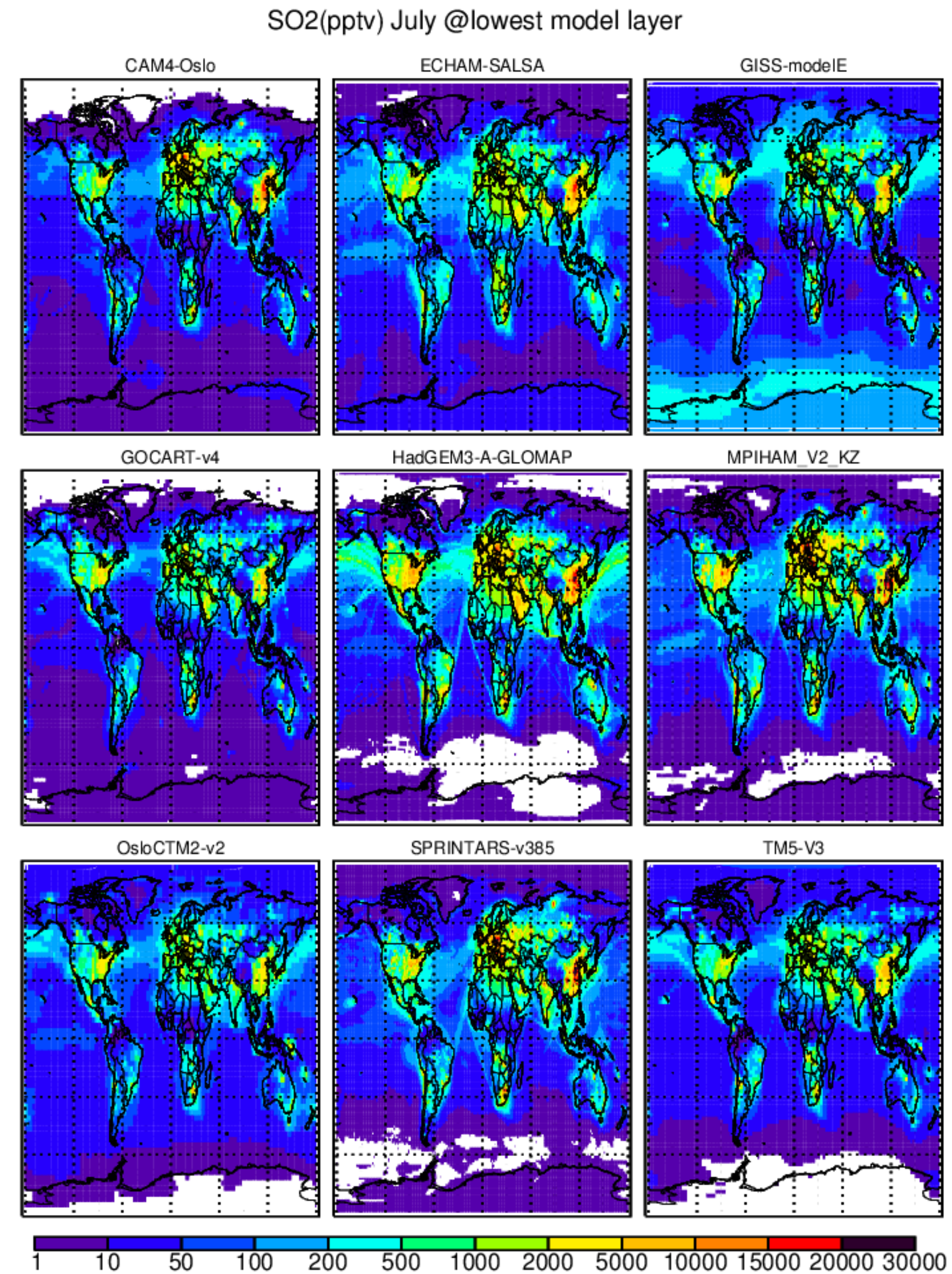
AEROCOM PHASE II MODELS

	Resolution	# Layers	Type	MET
CAM4-Oslo	2.5 x 1.89	26	CTM	GCM nudge to NCEP Reanalysis
GOCART-v4	2.5x2	30	CTM	GMAO-GOES-4
GOCART-v5	1.25x1	72	CTM	GMAO-GOES-5
OsloCTM2-v2	2.8125x2.81	60	CTM	ECMWF ERA-Interim
SPRINTARS-v385	1.125x1.121	56	CTM	Japanese GCM nudge to NCEP Reanalysis
TM5-V3	3x2	34	CTM	ECMWF ERA-Interim, or Forecast
ECHAM-SALSA	1.875x1.865	31	Online	GCM nudge to ECMWF
GISS-MATRIX	2.5x2	40	Online	GCM nudge to NCEP Reanalysis
GISS-modelE	2.5x2	40	Online	GCM nudge to NCEP Reanalysis
HadGEM2-ES	1.875x1.25	38	Online	GCM nudge to ERA Interim
HadGEM3-A-GLOMAP	1.875x1.25	38	Online	GCM nudge to ERA Interim
MPIHAM_V2_KZ	1.875x1.865	31	Online	GCM nudge to ECMWF
MERRA2	0.625x0.5	72	Online	MERRA reanalysis with aerosol assimilation



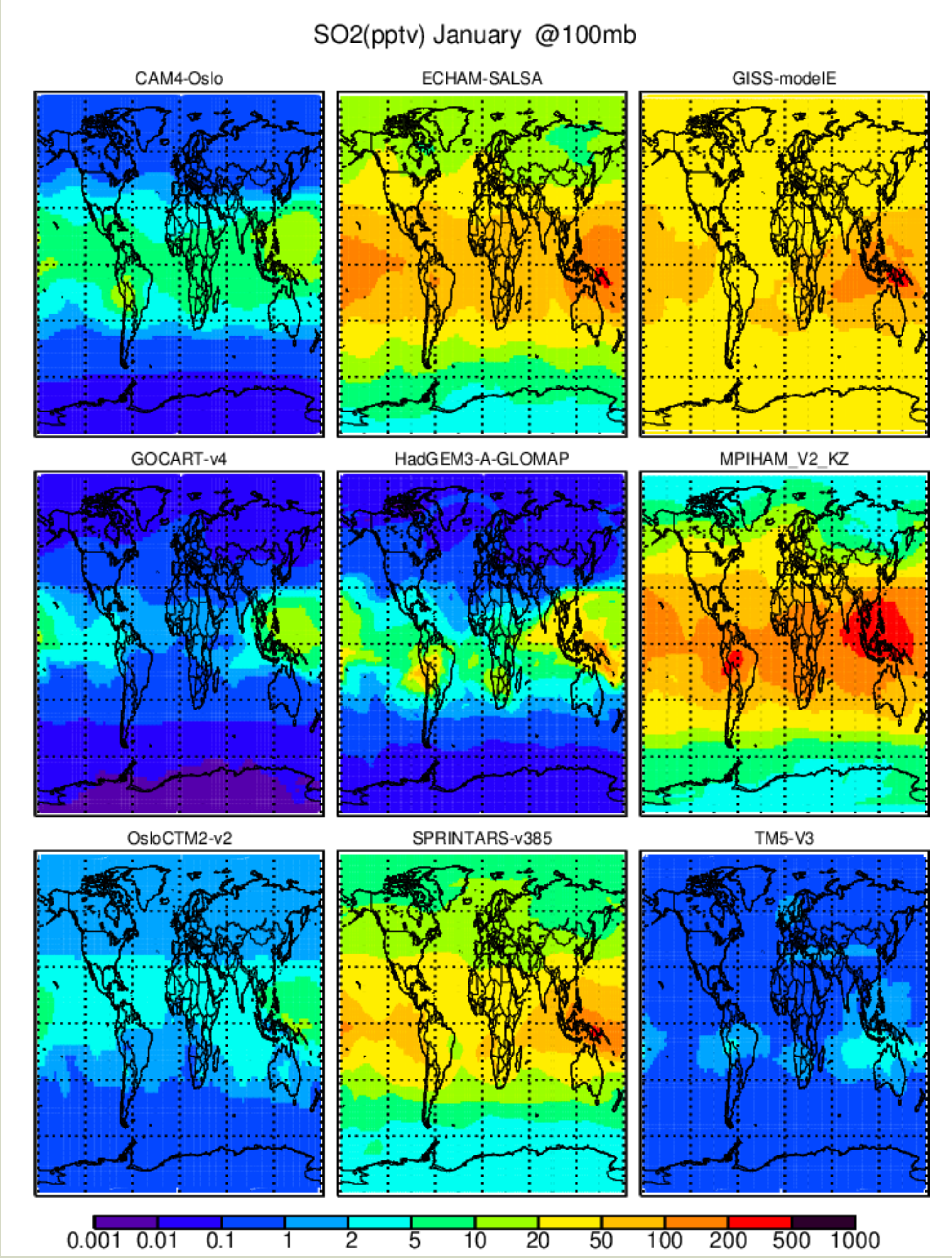
SO_2
January, 2006
@ lowest model layer

- Modeled SO_2 at the lowest layer follows emission patterns.
- In remote regions, the difference can be significant.



SO_2 July, 2006 @ lowest model layer

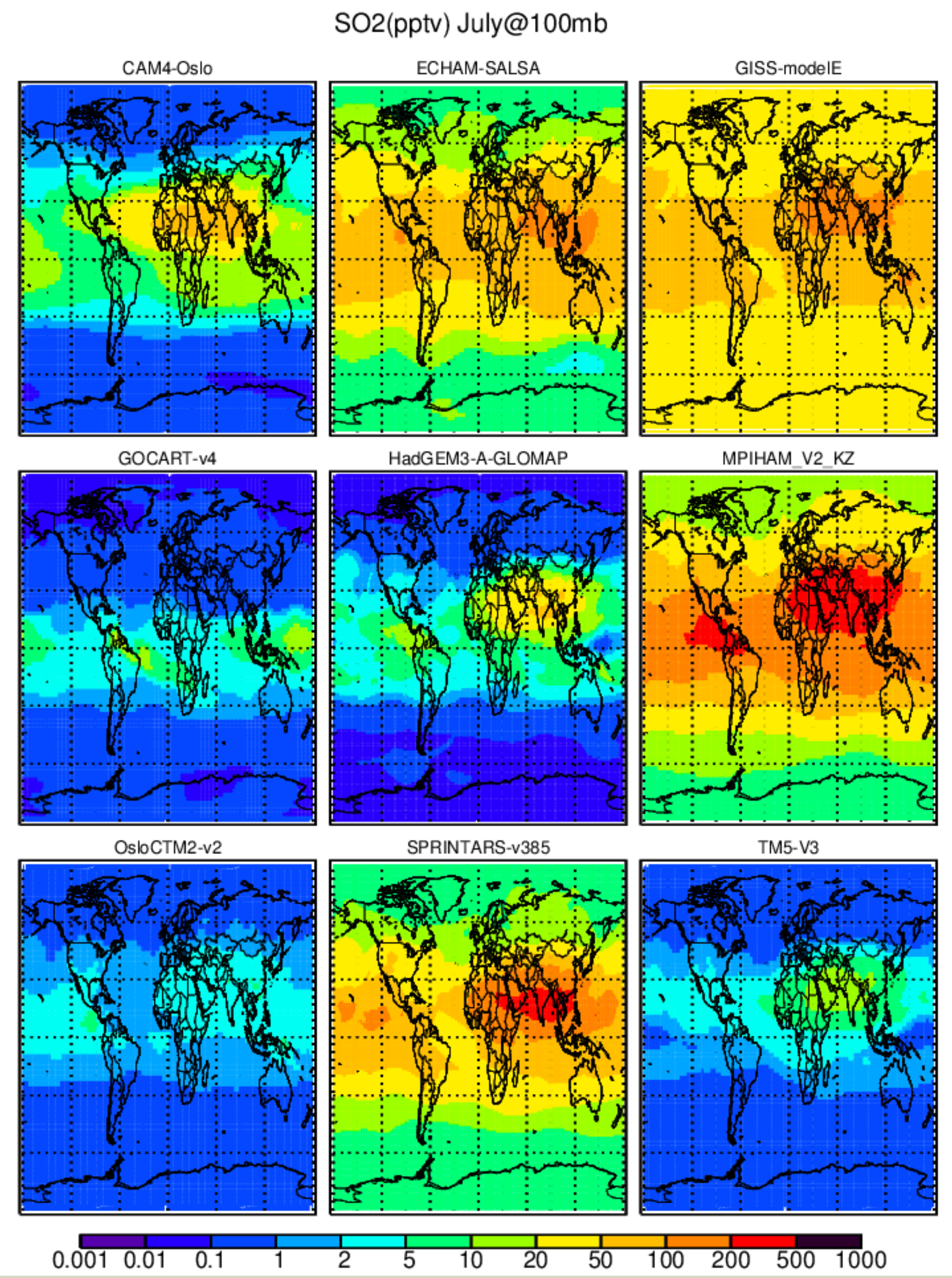
- In summer (July), modeled SO_2 at the lowest layer show less model-to-model difference than winter (January)

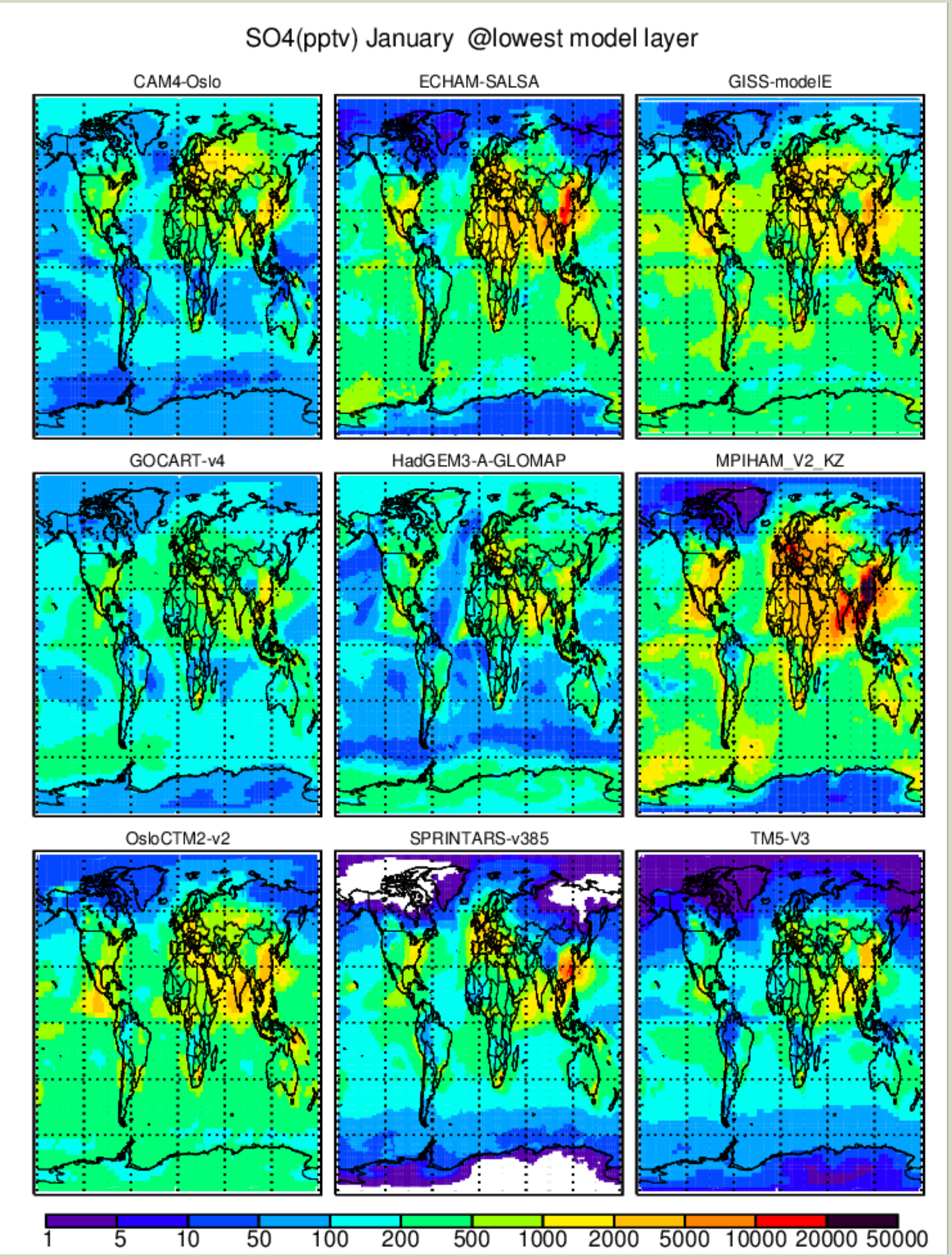


SO_2 January 2006 @ 100mb

- In UTLS, the model divergence (%) grows larger

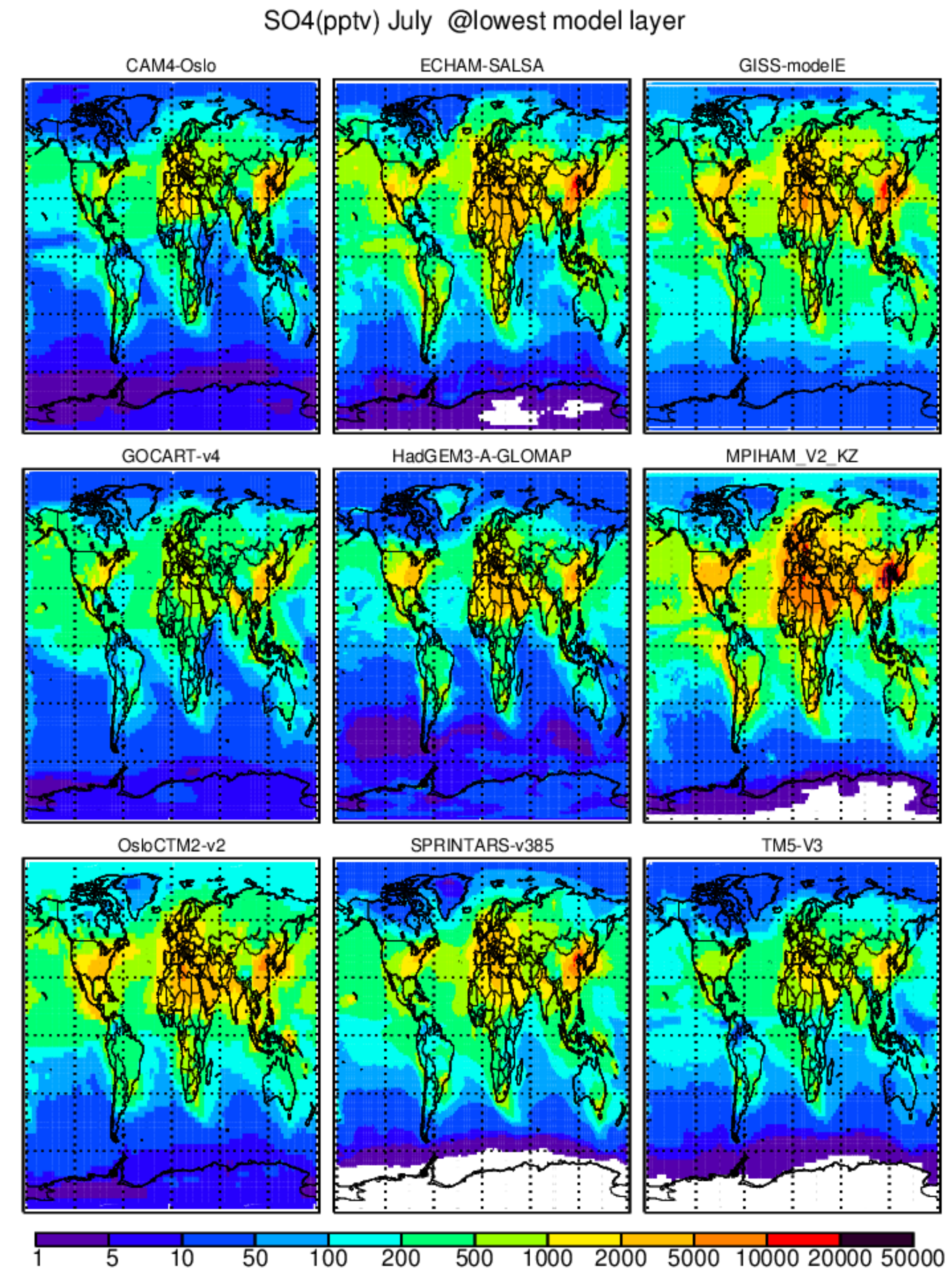
SO_2
July, 2006
@ 100mb





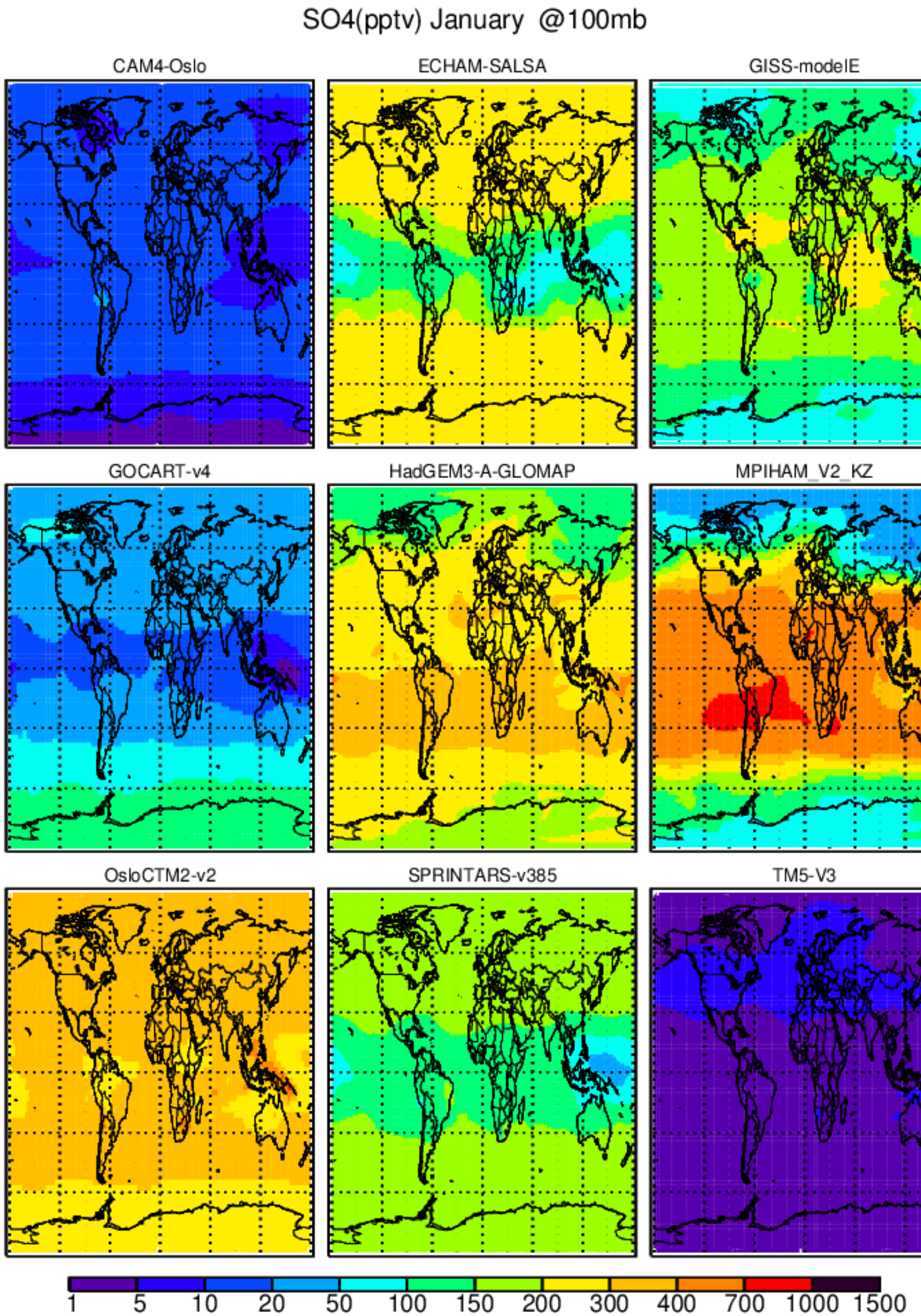
SO_4
January, 2006
@ lowest model layer

- Modeled sulfate at the lowest layer show larger model-to-model difference than SO_2



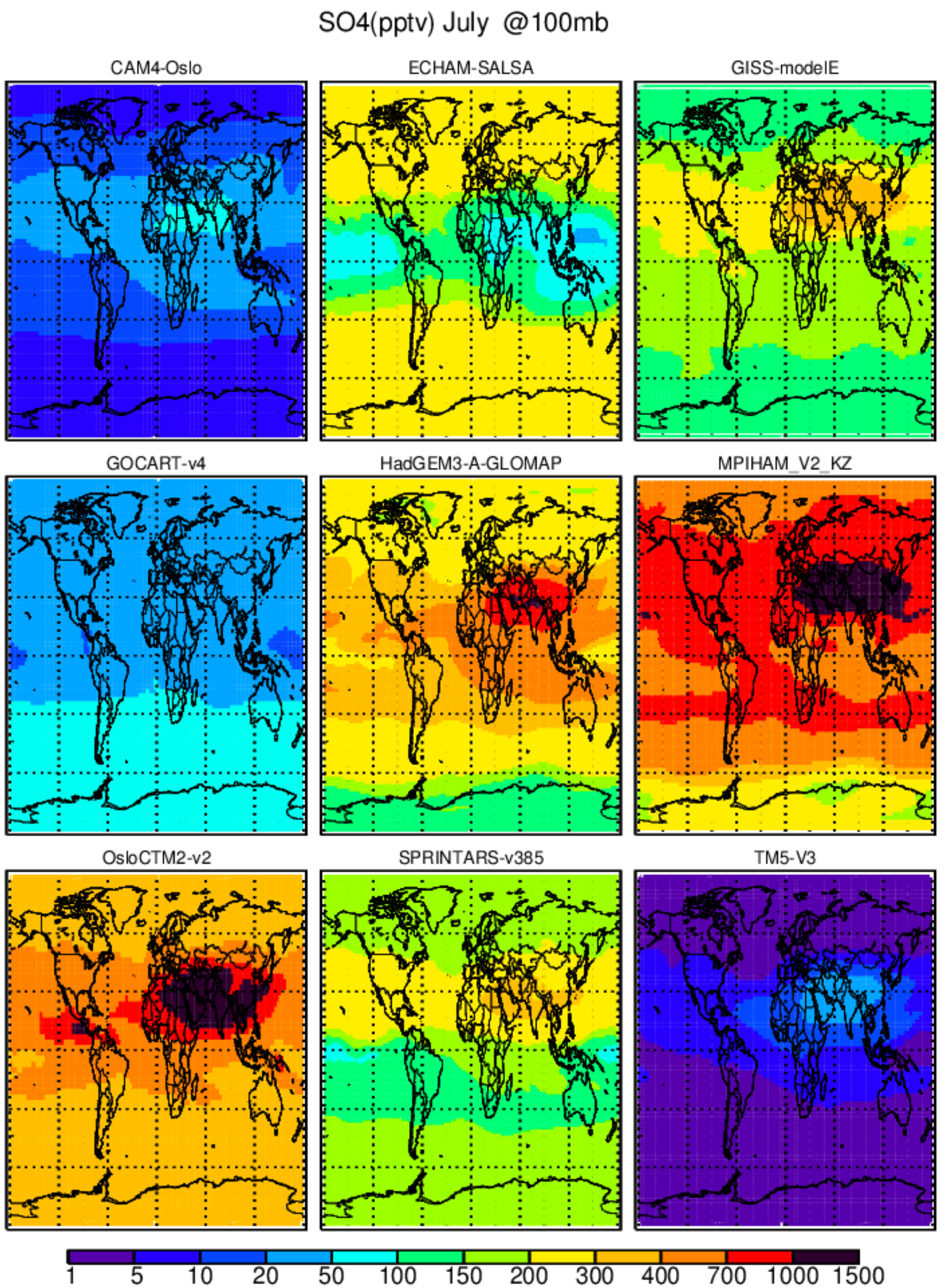
SO_4
July, 2006
@ lowest model layer

- In summer (July), modeled sulfate at the lowest layer show less model-to-model difference than winter (January)



SO_4 January, 2006 @ 100mb

- SO_4 shows large model divergence (%)
- The latitudinal gradient is also different
  - Tropics can be high/ same/ low than high latitude



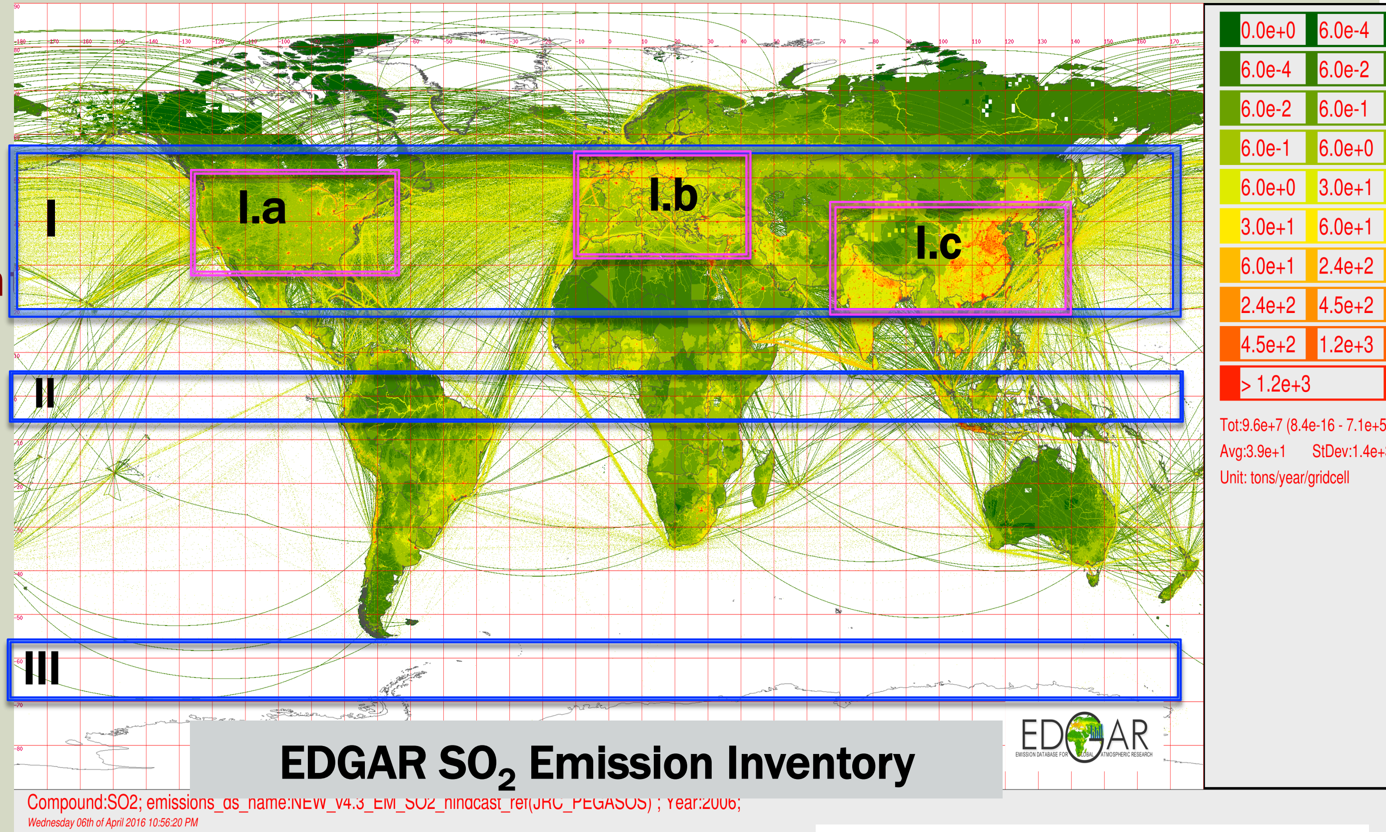
SO_4 July, 2006 @ 100mb

- SO_4 mixing ratio shows larger model divergence in July than January.

Model divergence can lead to different quantification of UTLS sulfur budget.

Checked variables:

- SO_2
  -- primary pollutant
- SO_4
  -- PM2.5/Climate
- SO_4:SO_2
  -- chemistry +wet scavenging
- SO_2+SO_4
  -- total S

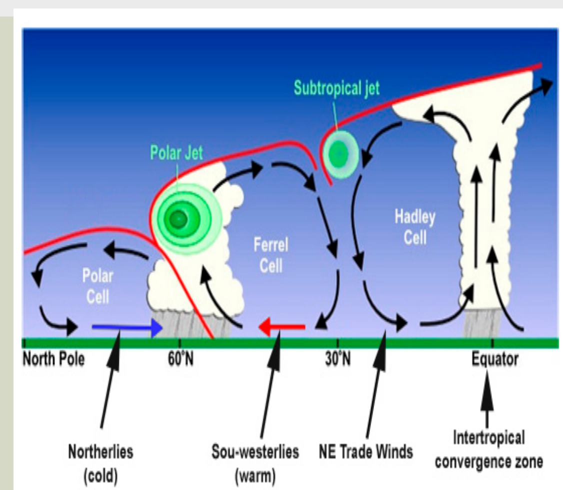


❖ Regions:

I. Emission hot spots

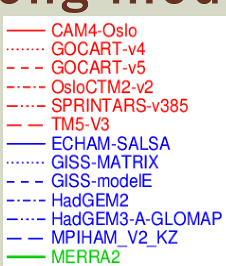
II. Tropics: major S-T exchange path

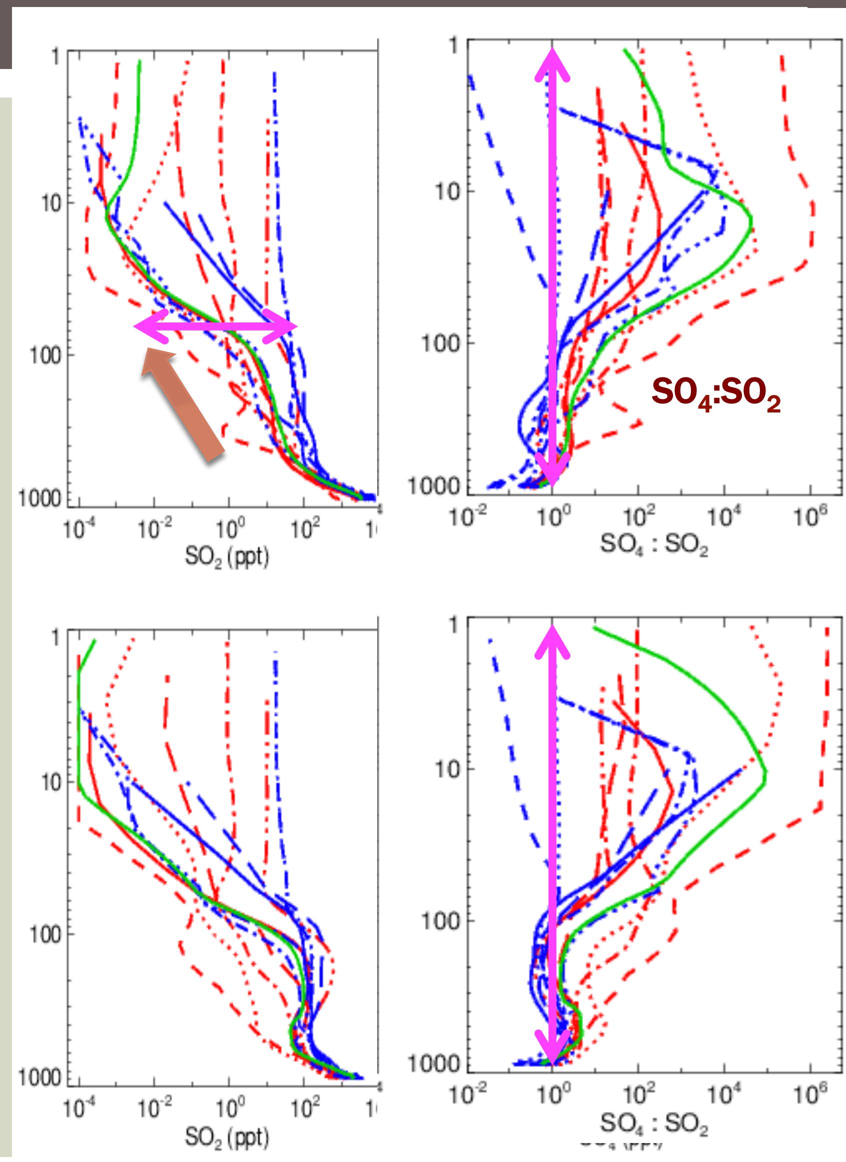
III. Southern ocean: remote region

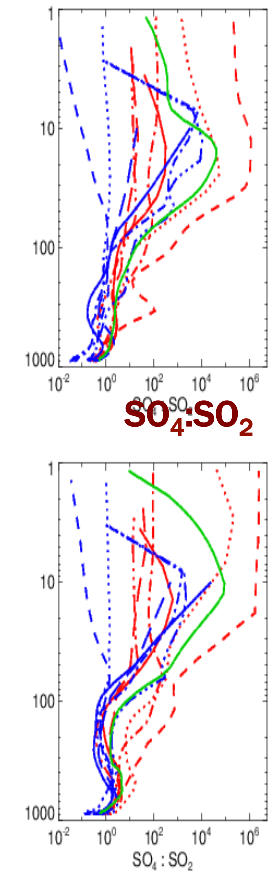


VERTICAL PROFILE OF SULFUR IN AEROCOM MODELS -- SOURCE REGIONS

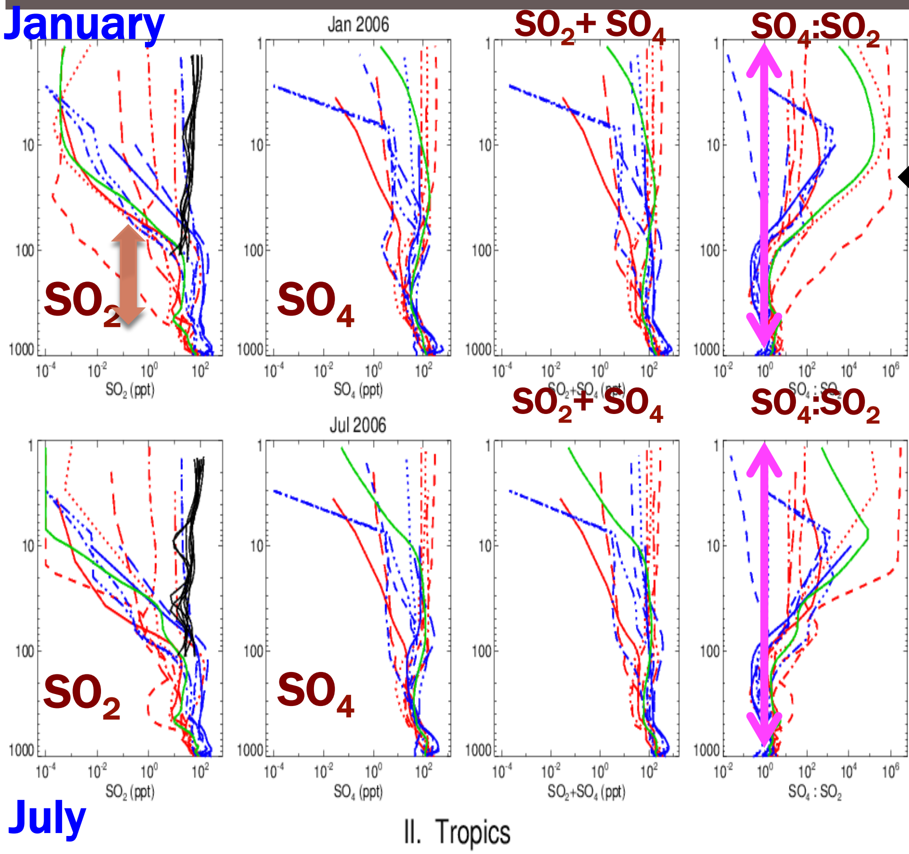
- Most models are consistent at the surface and diverge moving upward.
- SO_2 has larger vertical gradient than SO_4 near the source regions.
- Model-to model difference is bigger for SO_2 than SO_4
- $SO_4:SO_2$ is also different among models.



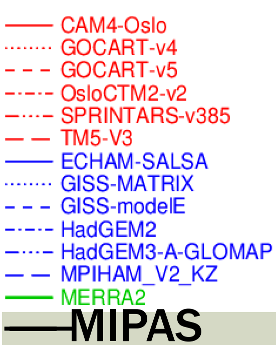




VERTICAL DISTRIBUTION OF SULFUR IN AEROCOM MODELS:
-- REMOTE REGIONS

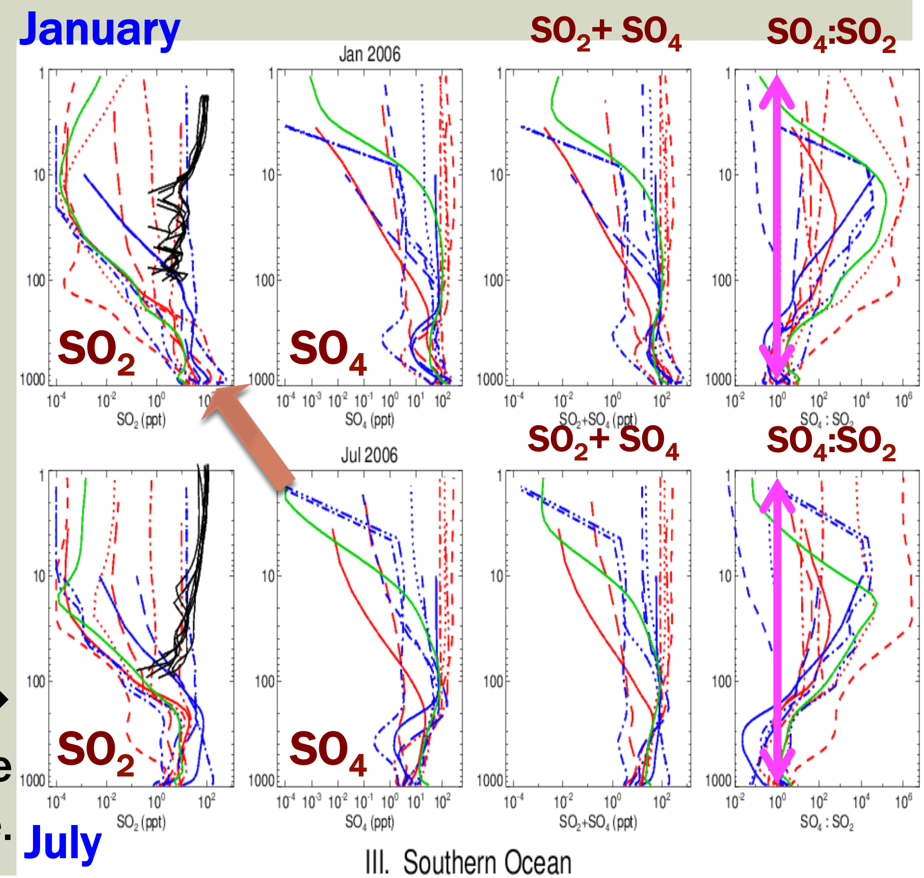


←**Tropics:** stronger vertical mixing.



Southern Ocean→

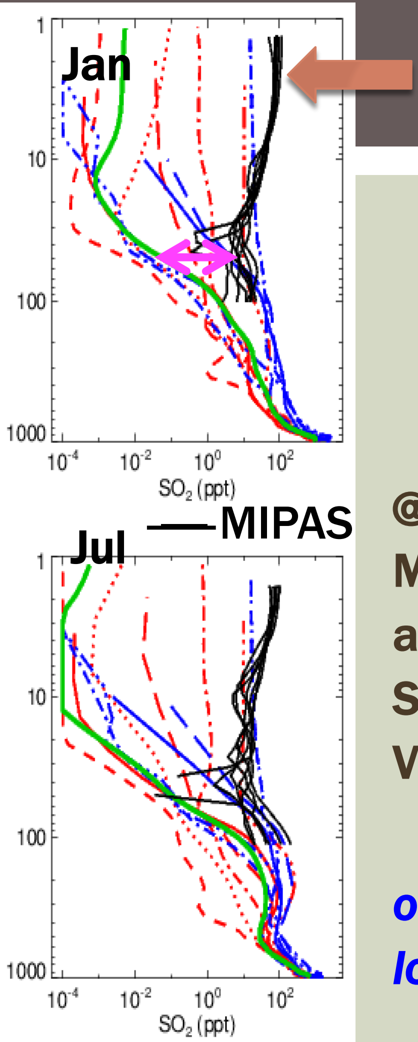
Model-to-model difference starts from surface.



COMPARISON WITH OBSERVATIONS

- Available observations
  - *In-situ aircraft measurements*
    - **Challenging requirements for instruments**
      - SO_2 and sulfate concentration change several magnitudes vertically
      - In UTLS, SO_2 mixing ratio is below detection limit of most current SO_2 instruments.
    - **Sparse spatial/temporal coverage.**
  - *Satellite measurement*
    - **Good spatial and temporal coverage**
    - **Confidence level in the accuracy of retrievals, esp. the vertical profile of chemical species might not be high.**
  - *No perfect aircraft/satellite measurement yet available to constrain modelled vertical profile.*

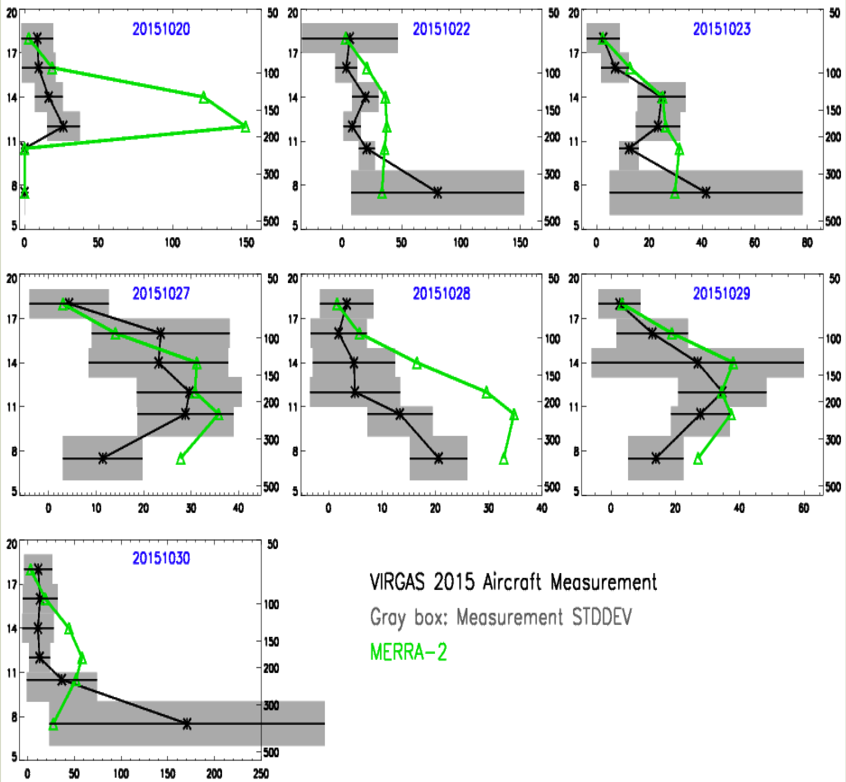
SO_2 IN UTLS: MODELS VS OBSERVATIONS



- MIPAS (satellite) SO_2 is ~ upper limit of models
  - *@ 30-45km: sulfate --evaporate-> H_2SO_4 --photolyzed-> SO_2 in high altitude is not included in models.*

@ lower stratosphere: MERRA-2 shows reasonable agreement with the in-situ SO_2 measurement made in VIRGAS 2015

-- MIPAS might overestimate the SO_2 in the lower stratosphere

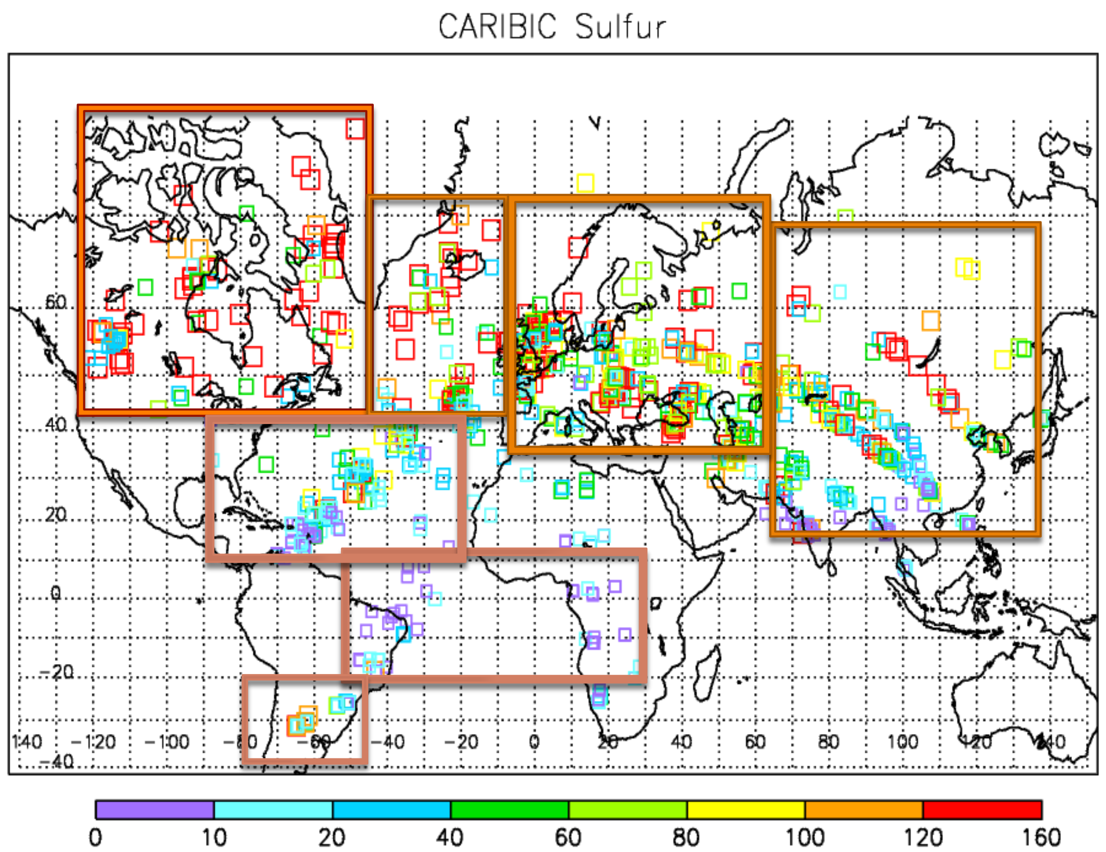


SO_2: MIPAS vs models.

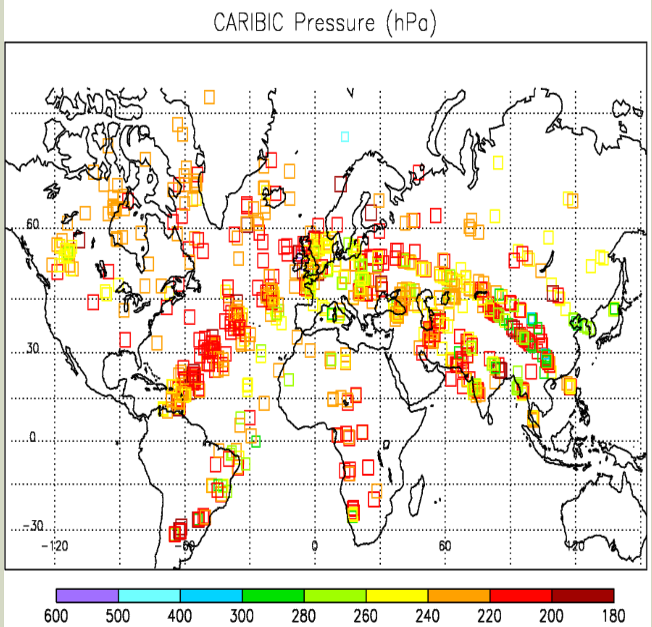
MIPAS: Höpfner et al. 2013, 2015

VIRGAS data from R. Gao & A. Rollins

OBSERVED SO_4 IN UPPER TROPOSPHERE



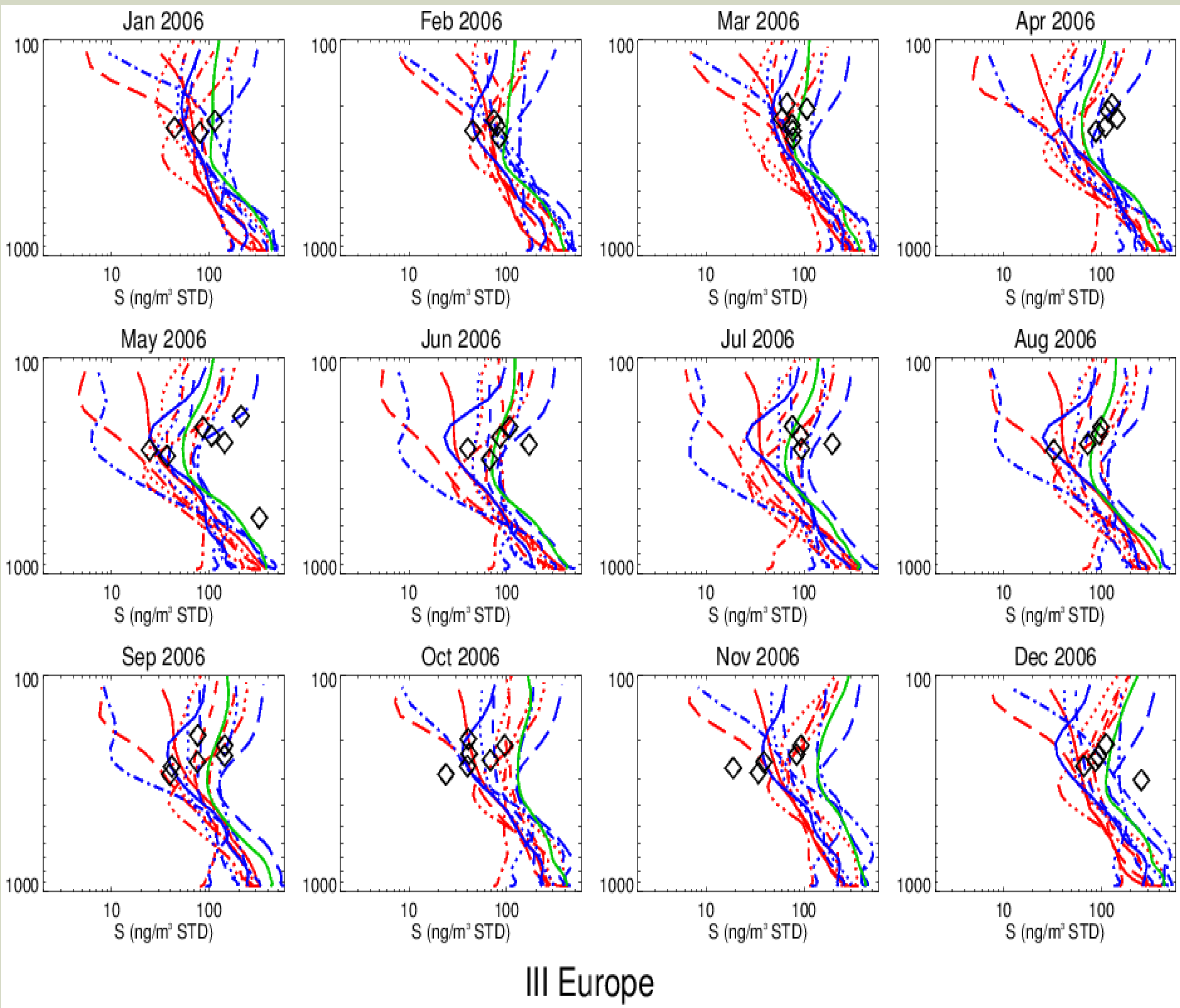
1, North America
2. Northern Atlantic
3. Europe
4. Asia
5. Mid-Atlantic
6. Tropics
7. South America



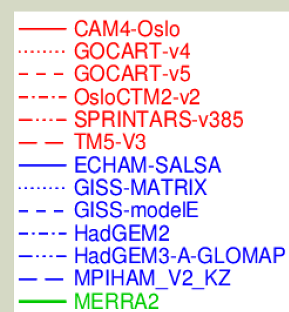
CARIBIC:

- **Air-borne measurement**
- **Cruise altitude (200-300hpa)**
- **2005-2013**
- **TR: 100min**
- **Uncertainty: 12%**

SO_4 IN UT: OBSERVATION VS MODELS -- SOURCE REGION

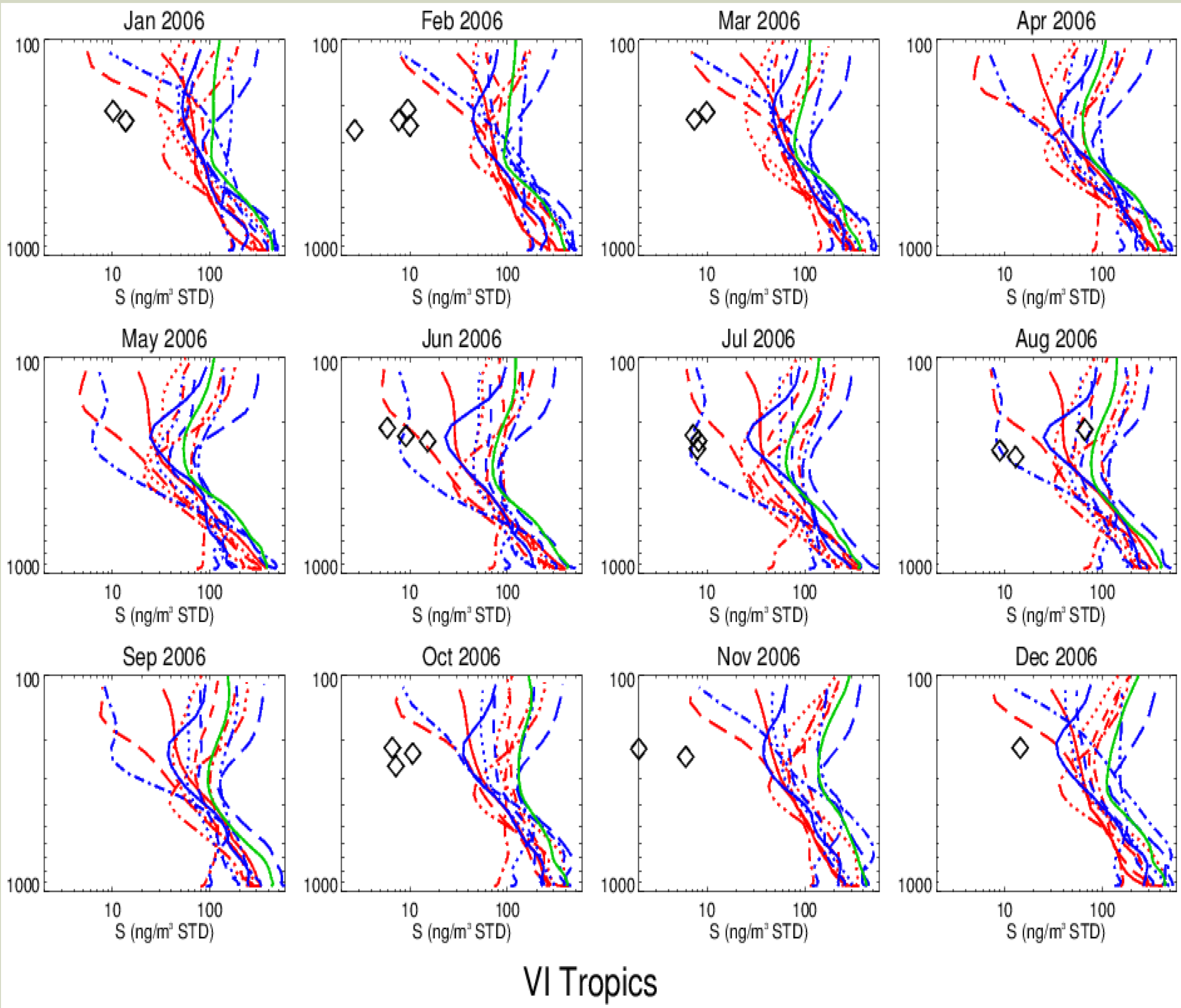


- CARIBIC measurements show large year-to-year variation of SO_4 aerosols
- Model-to-model variation is large.
- They overlap

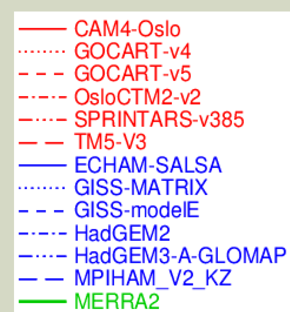


◆ CARIBIC measurements.

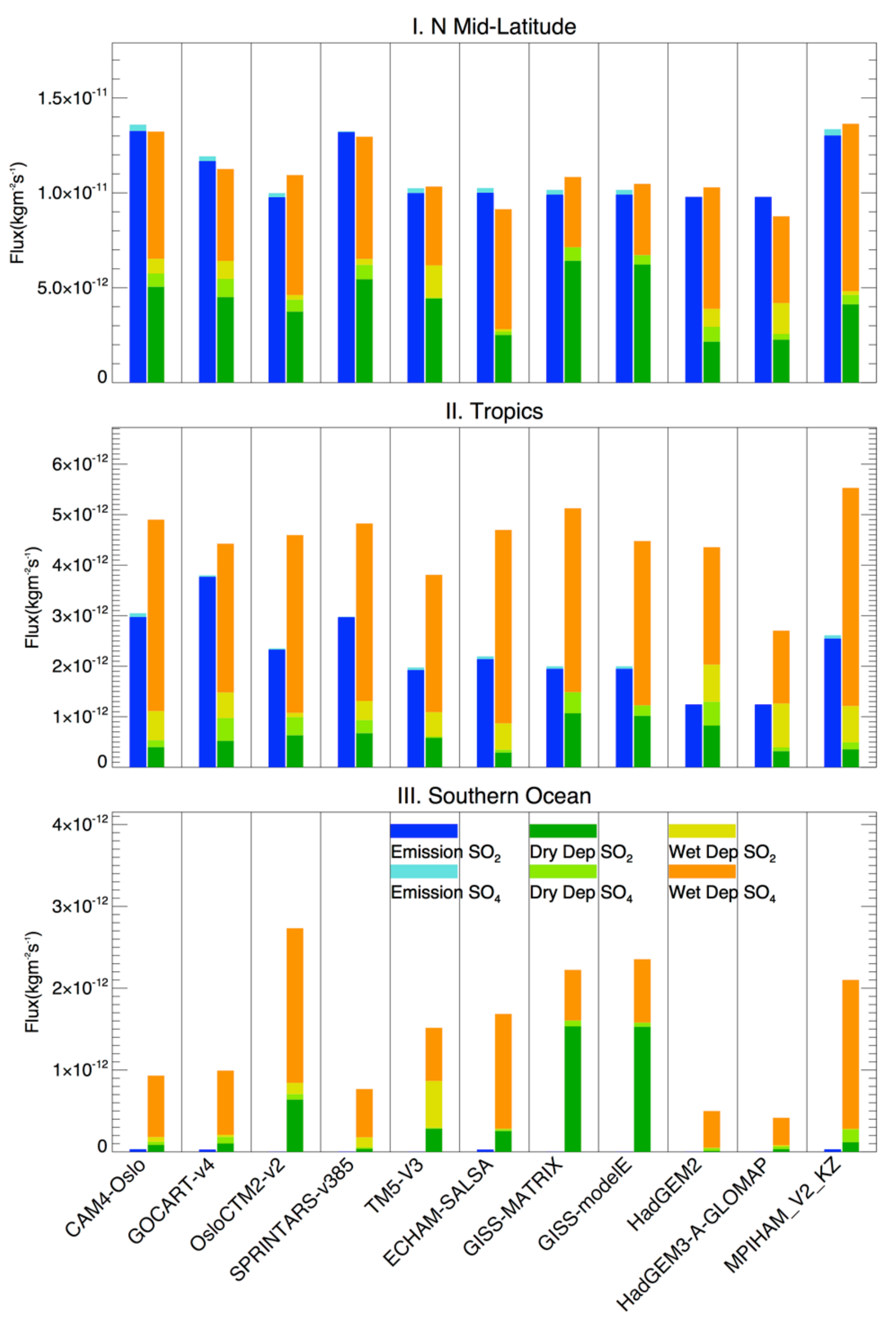
SO_4 IN UT: OBSERVATION VS MODELS --TROPICS



- CARIBIC measurements show less variation of SO_4 aerosols.
- Model-to-model variation is large.
- Models tend to **overestimate** SO_4



◆ **CARIBIC measurements.**

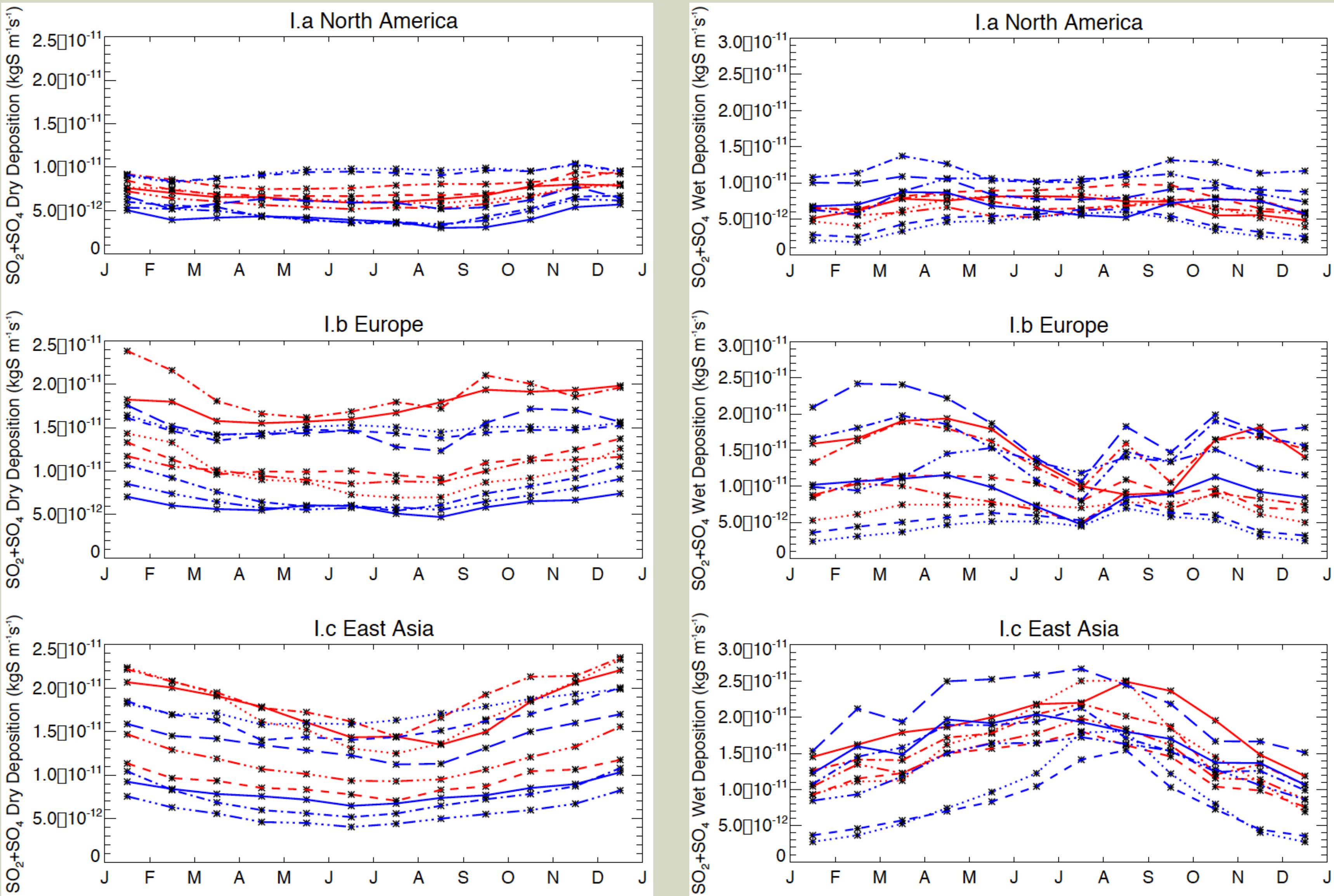


What do saved model diagnoses tell us about possible reasons for model divergence?

Budget of modeled processes

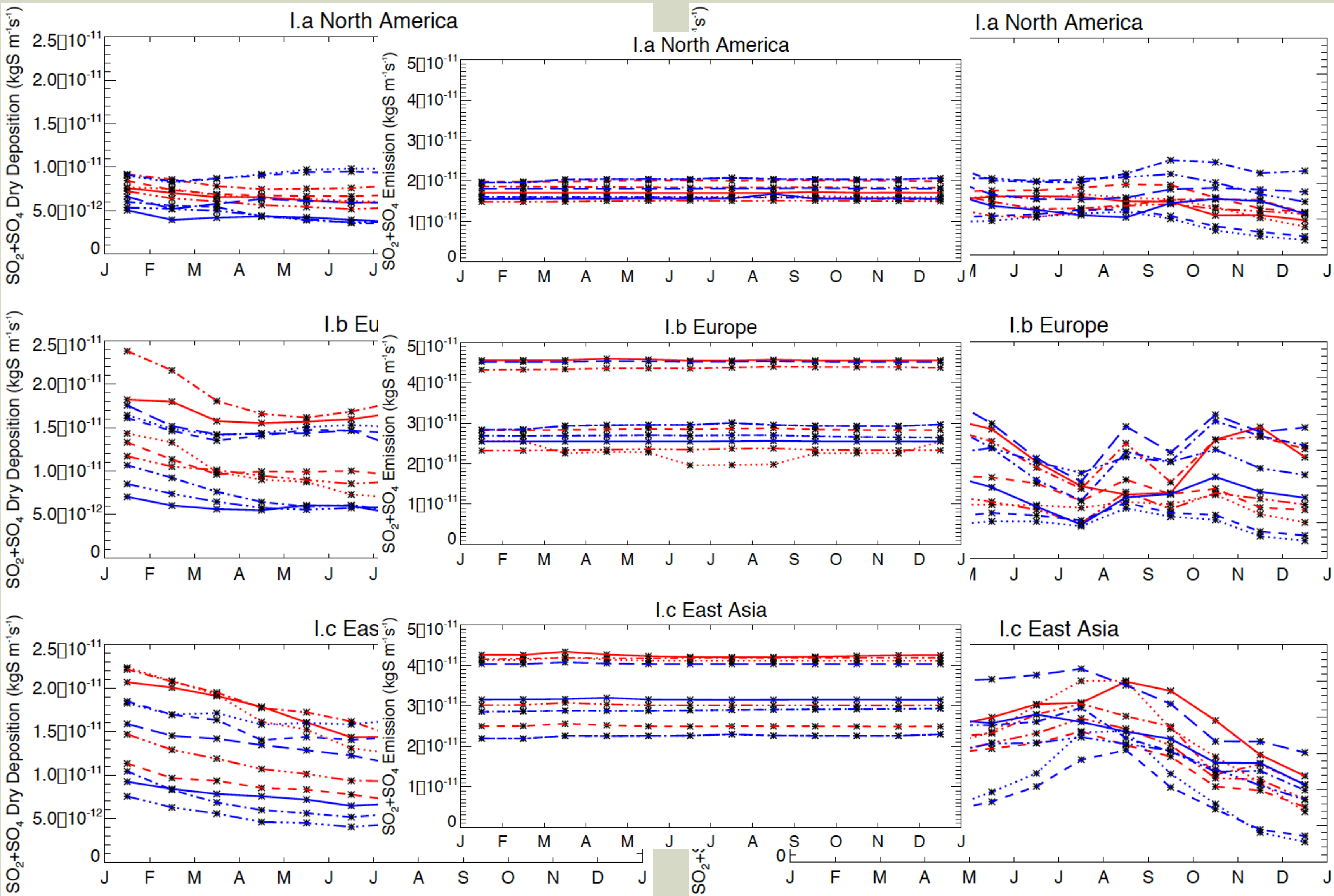
- Near the source regions, dry+wet deposition is about the same as emissions
- Models have different pathways (chemical/physical) to remove emissions.
- In remote regions, more variation in the calculated deposition fluxes.

Dry and Wet Deposition Near Major Source Regions (absolute magnitude)



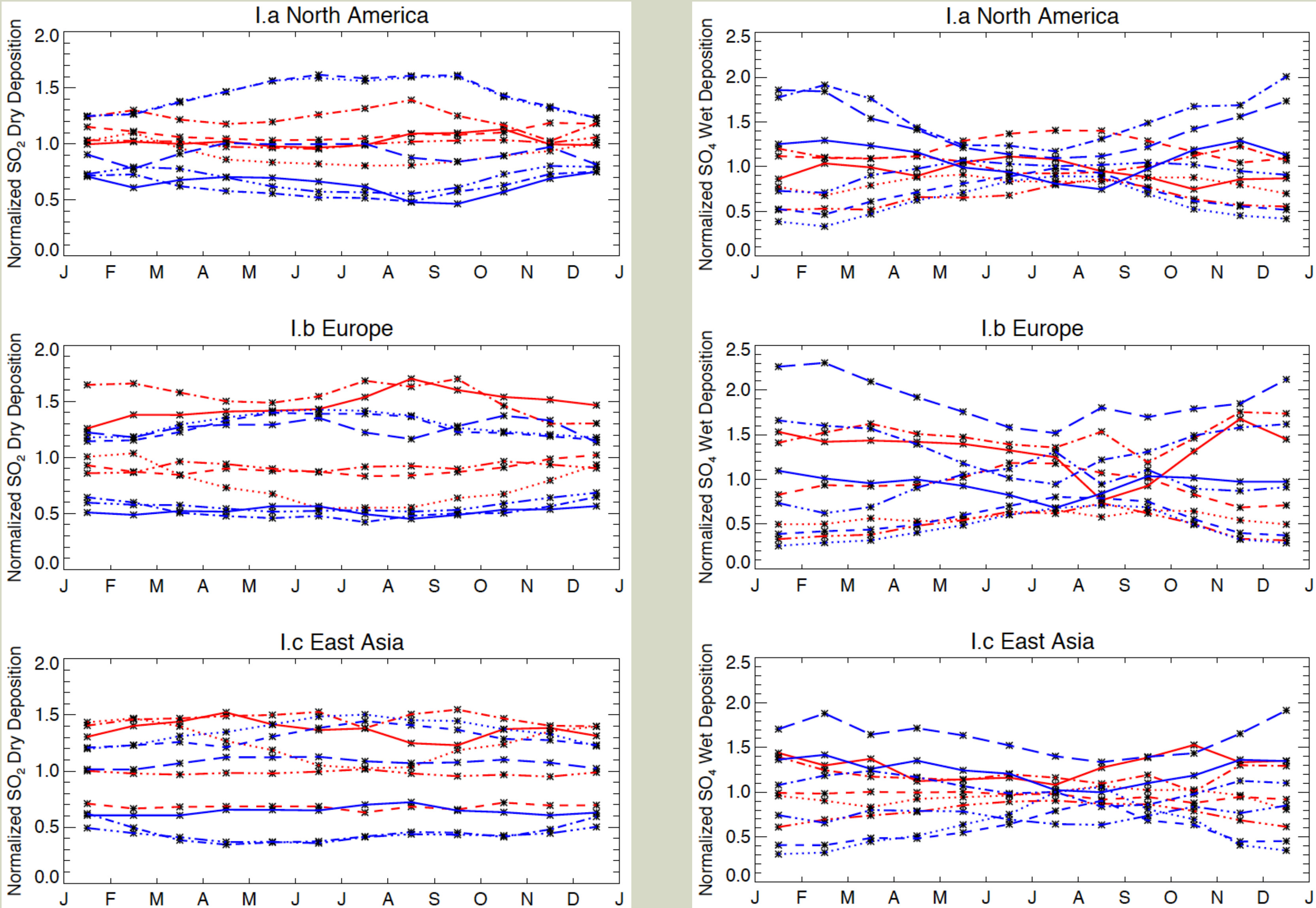
NA: emissions are better constrained, more surface concentration measurements are available

Dry and Wet Deposition Near Major Source Regions (absolute magnitude)



NA: emissions are better constrained, more surface concentration measurements are available

Dry and Wet Deposition Near Major Source Regions (Normalized)



Dry deposition shows 50%-150% variations, wet deposition: 30%-200%, winter more divergence.

CONCLUSION

- Vertical distribution of sulfur species in the AEROCOM-II models.
  - ***SO_2 shows larger variations than sulfate.***
  - ***Model divergence (% difference)***
    - **Grows vertically.**
    - **Bigger in regions with sparse/no observations.**
  - ***Models balance emissions with different pathways.***
    - **Dry deposition shows less variation than wet deposition.**
  - ***Correct surface concentration and column AOD might not be sufficient to guarantee models will give reliable quantification of UTLS aerosol source attribution.***

WISH LIST:
-- What might help to estimate the source attribution of UTLS aerosols?

- Synthetic tracer(s)
  - ***Distinguish contribution impacts from vertical transport, wet removal, and chemical processes***
- Vertical motion
  - ***Possible/feasible model diagnoses for vertical fluxes?***
    - **Large scale and convective transport**
    - **Limited aircraft measurements suggested high frequency waves (w ~0.5m/s) in upper troposphere might not captured by global models.**
- Flux between troposphere and stratosphere?
  - ***One more constrain, in addition to surface mixing ratio, surface fluxes and column AOD.***