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# Evaluating cloud processes in particulate matter forecasting

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# Outline

- Challenges in modelling and evaluating cloud processing of gases and aerosols
- Experience from model evaluation for ICARTT field campaign period, focusing on cloud processes
  - *ICARTT-CTC (Chemical transformation and transport by cloud) aircraft study*
  - *Regional scale (15-km res.): surface (network data) vs. aloft (aircraft measurement); sensitivity study*
  - *Hi-resolution (2.5-km res.) case study (cloud processing of urban/ industrial plumes)*
- Lessons learnt and future work

# Challenges in modelling cloud processes

- Clouds play a major role in the transformation/production of PM
- Relatively well known aqueous inorganic reactions (e.g., sulphur chemistry)
- Poorly known aqueous-phase reactions involving organics and their impact on secondary organic aerosol formation
- Bulk vs. size-resolved cloud droplets for aqueous-phase chemistry calculation
- (Meteorological) model skill in predicting cloud dynamics and microphysics
- Scale dependency on process representation
- Feedback to cloud microphysics and dynamics

# Challenges in evaluating cloud processes

- Very difficult to conduct direct evaluation; most evaluations are indirect (inferred)
- Lack of appropriate data
- Spatial and temporal disparity between observations (e.g., aircraft) and model predictions
- Evaluation methodology

# ICARTT-CTC

- Total of 23 research flights between July 19 and August 18, 2004; all but 4 were over southern Great Lakes area; stratocumulus (Sc) and towering cumulus (Tcu); measurements from 5 Tcu and 4 Sc flights are used for this model evaluation study.

- Measurements on-board:

Trace gases (CO, O3, NOx, **SO2**, HNO3, HCHO, H2O2);

***For particles 0.01 – 20 µm:***

TSI **SMPS** and TSI **APS** (inboard);

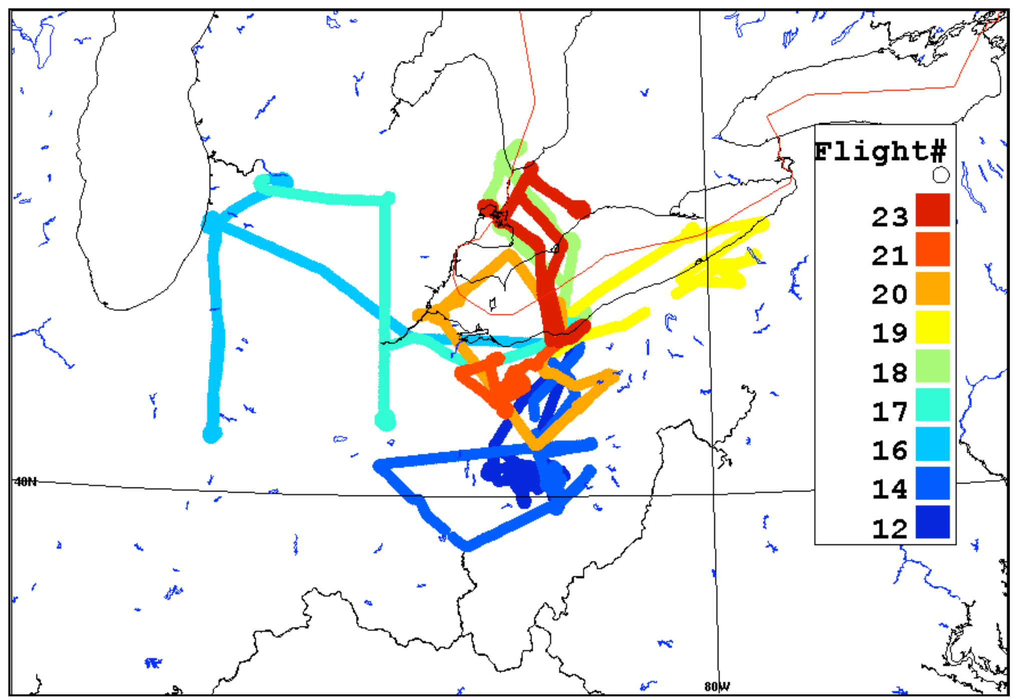
PMS PCASP and PMS FSSP300 (outboard);

***For cloud droplets:***

PMS FSSP100 and FSSP300;

***For chemical composition (aerosol and residual):***

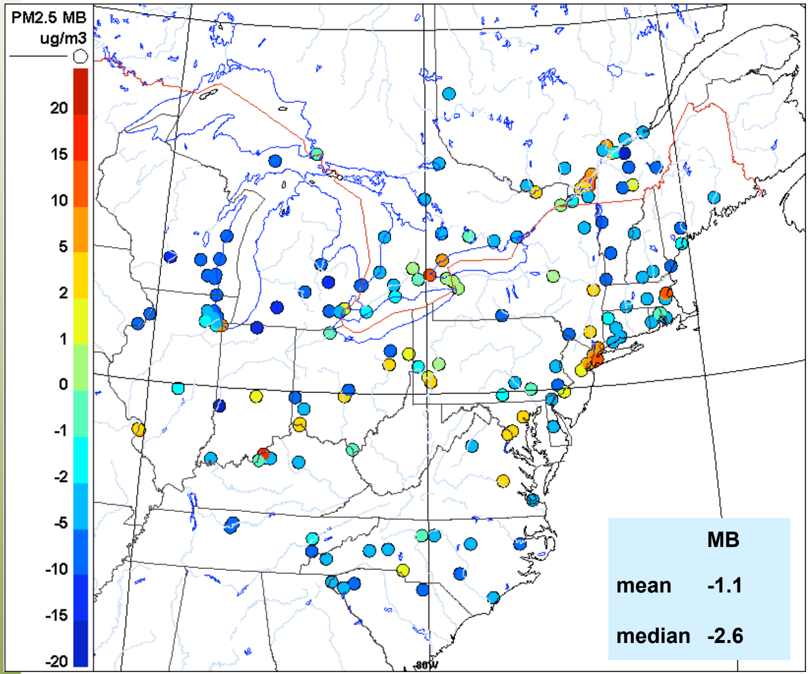
**Q-AMS**, **PILS**, and **cloud water samples**.

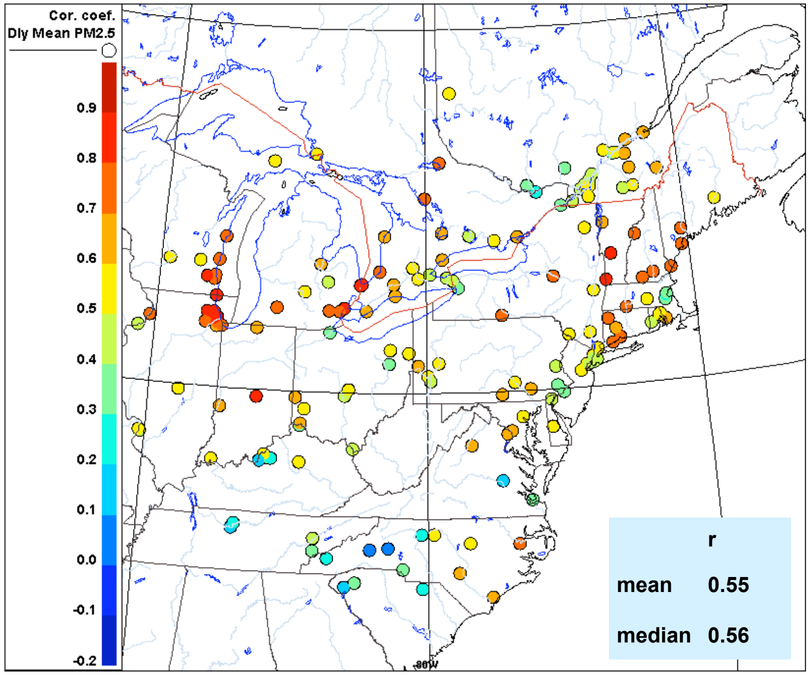


# Evaluation at regional scale

- Evaluation period: July 14 – August 18, 2004
- AURAMS runs at 42- and **15-km** resolutions, July 7 – August 19, 2004
- Evaluation statistics against surface networks: AIRNOW $PM_{2.5}$ (TEOM, hourly observations); IMPROVE speciated $PM_{2.5}$ (filter, 24-hour every 3 days).
- Evaluation aloft against aircraft measurements: vertical profiles for each flight; model sampling along flight tracks (grids containing flight path, no interpolation; sampling from hourly model output for entire flight period ~ 3 to 4 hours)
- Sensitivity to in-cloud oxidation

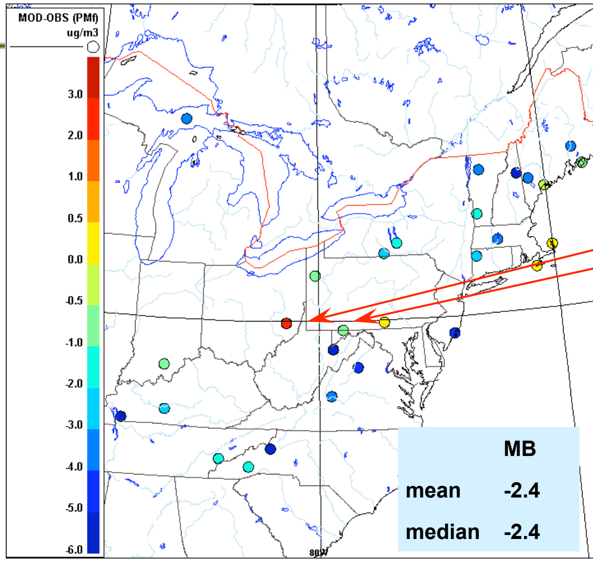
# Comparison against AIRNOW Daily mean $PM_{2.5}$ (MB and r)

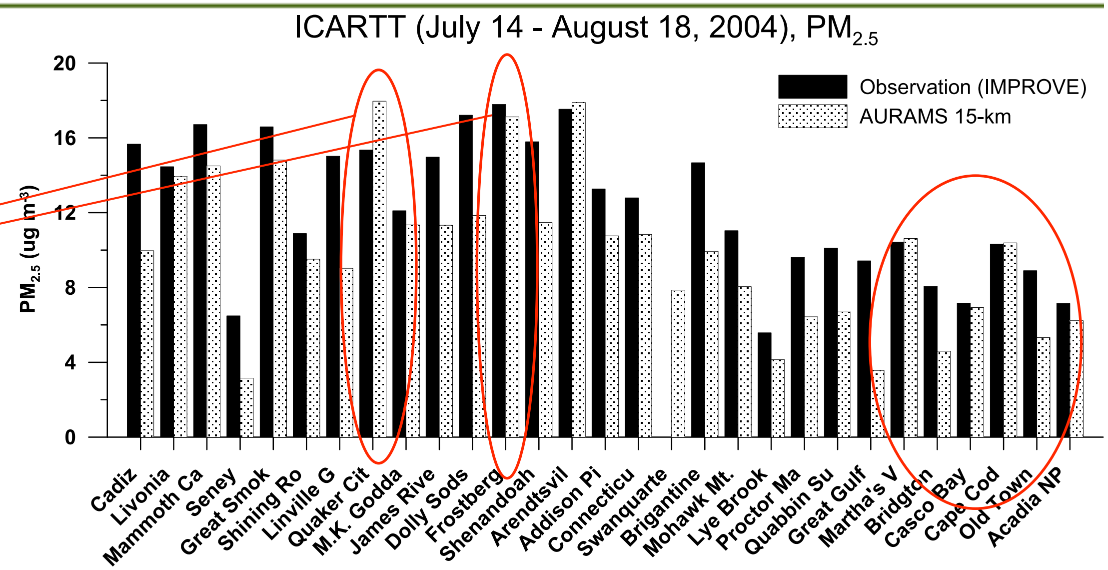


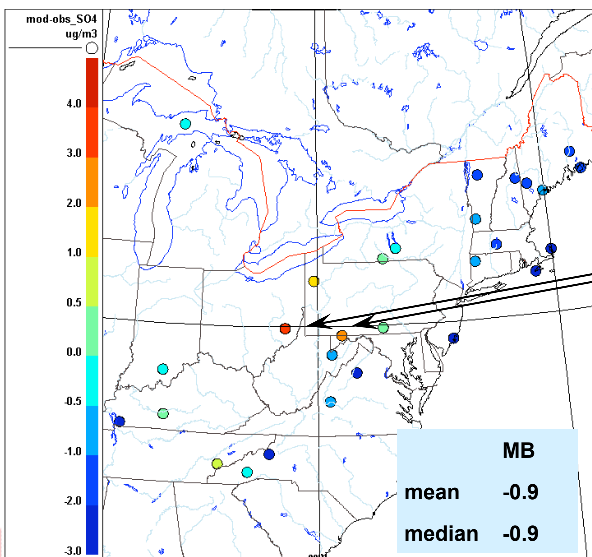



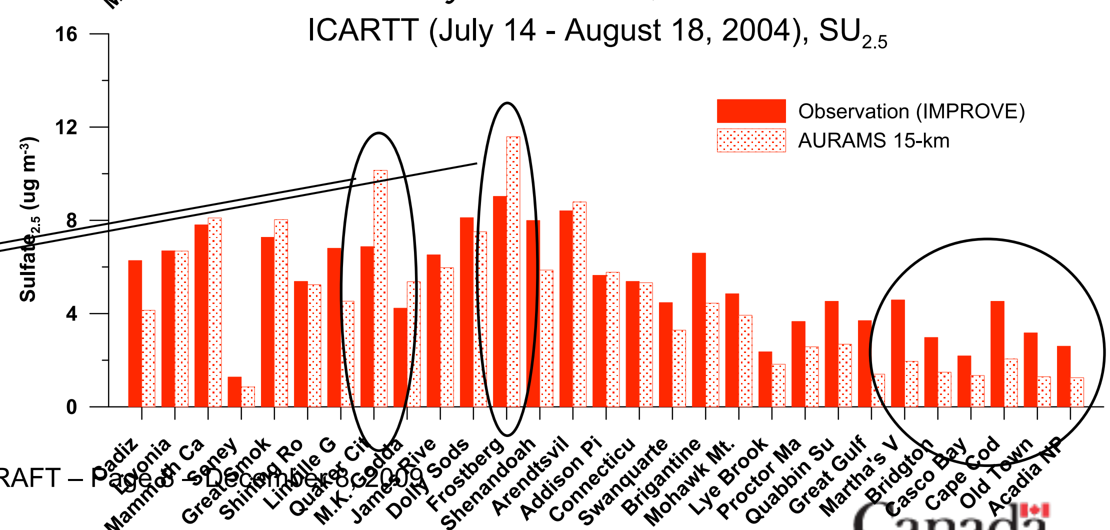

- The distribution of MB is more skewed towards negative: model under predicted PM2.5 at most of the sites except for some of the urban sites.
- The distribution of r is more normal (mean ≈ median), with lower correlation at more rural sites.

# Comparison against IMPROVE speciated $PM_{2.5}$ ($PM_{2.5}$ and $SU_{2.5}$; 24-hr sampling, 1 in 3 days)



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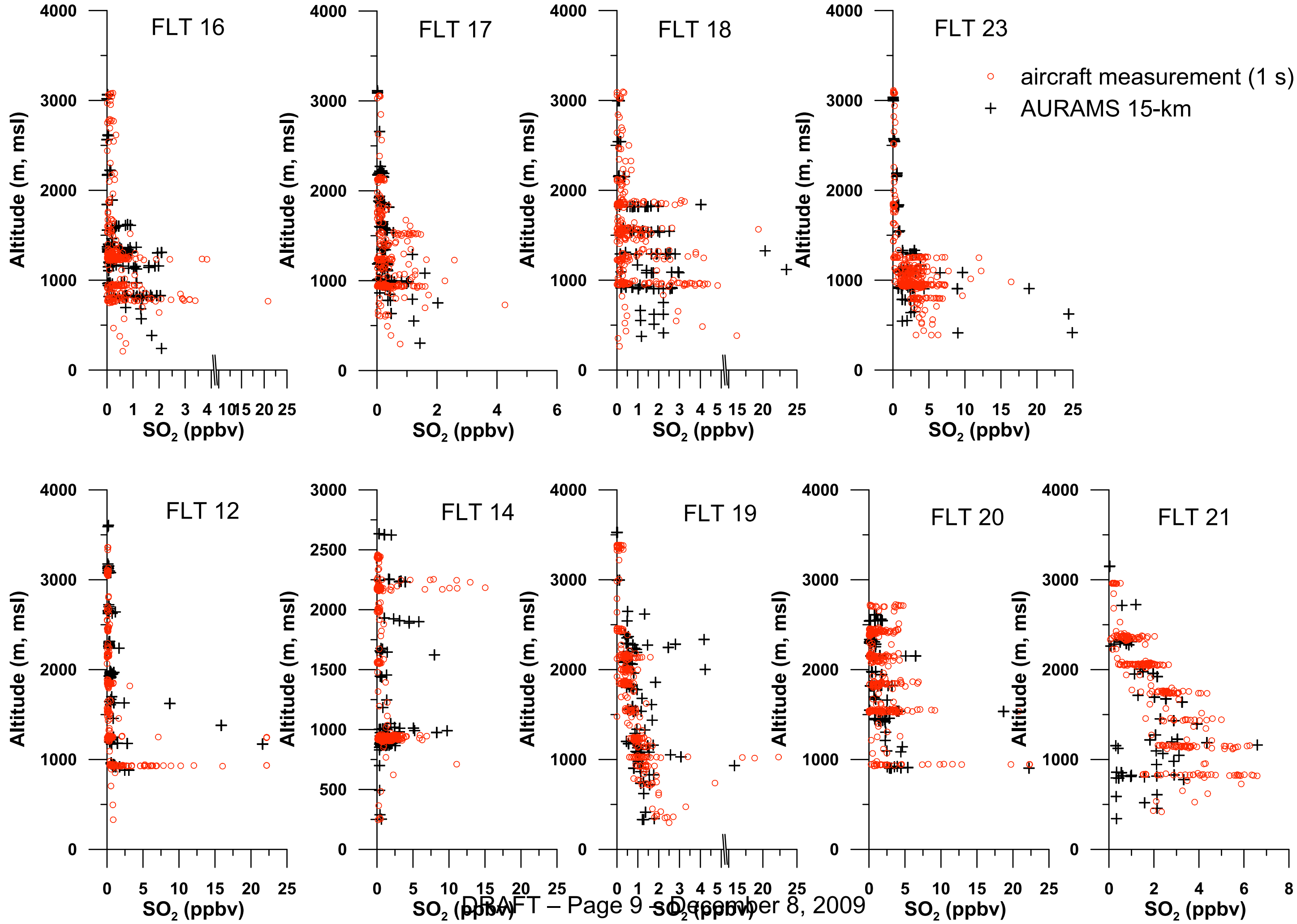
# Comparison against aircraft measurement - $SO_2$



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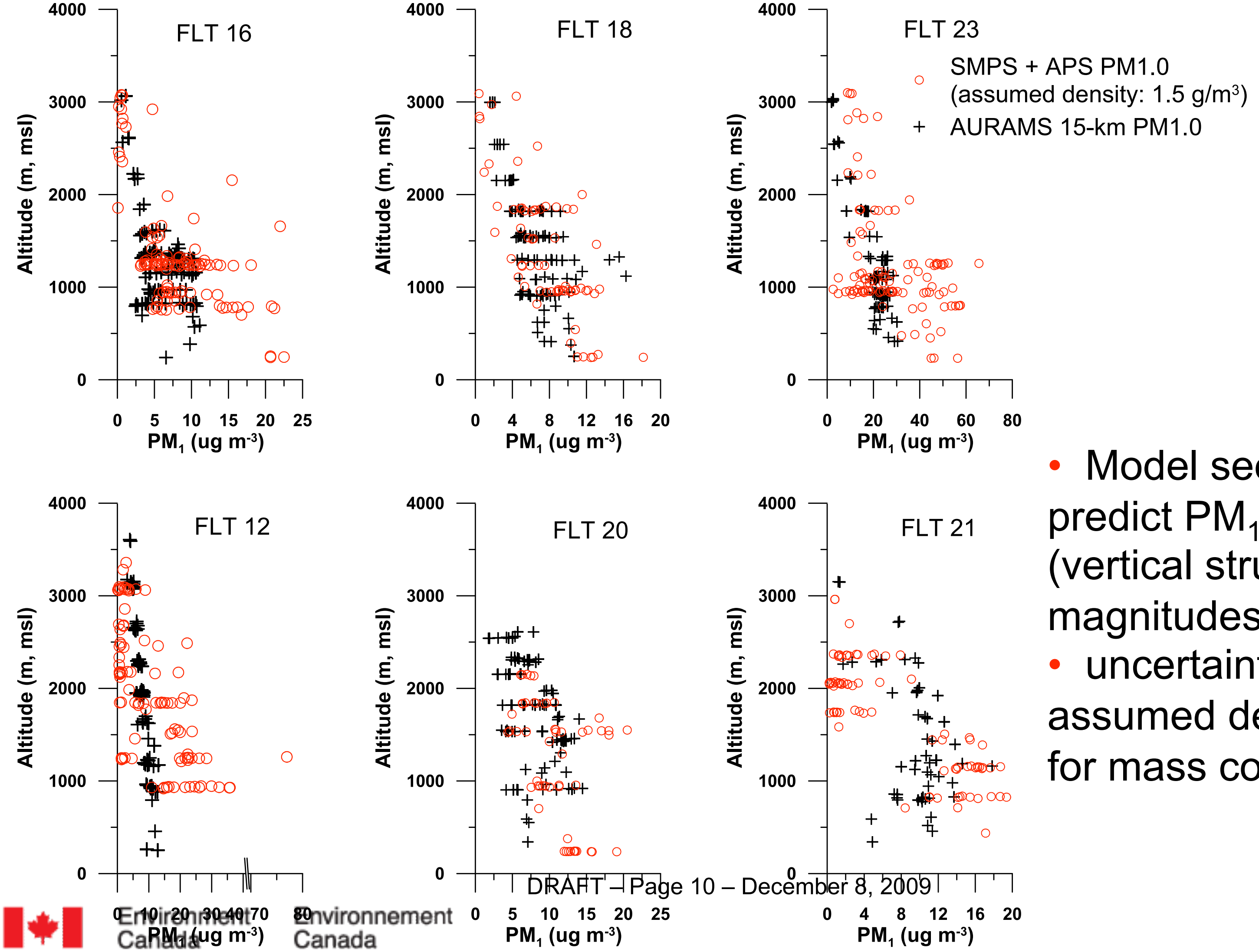
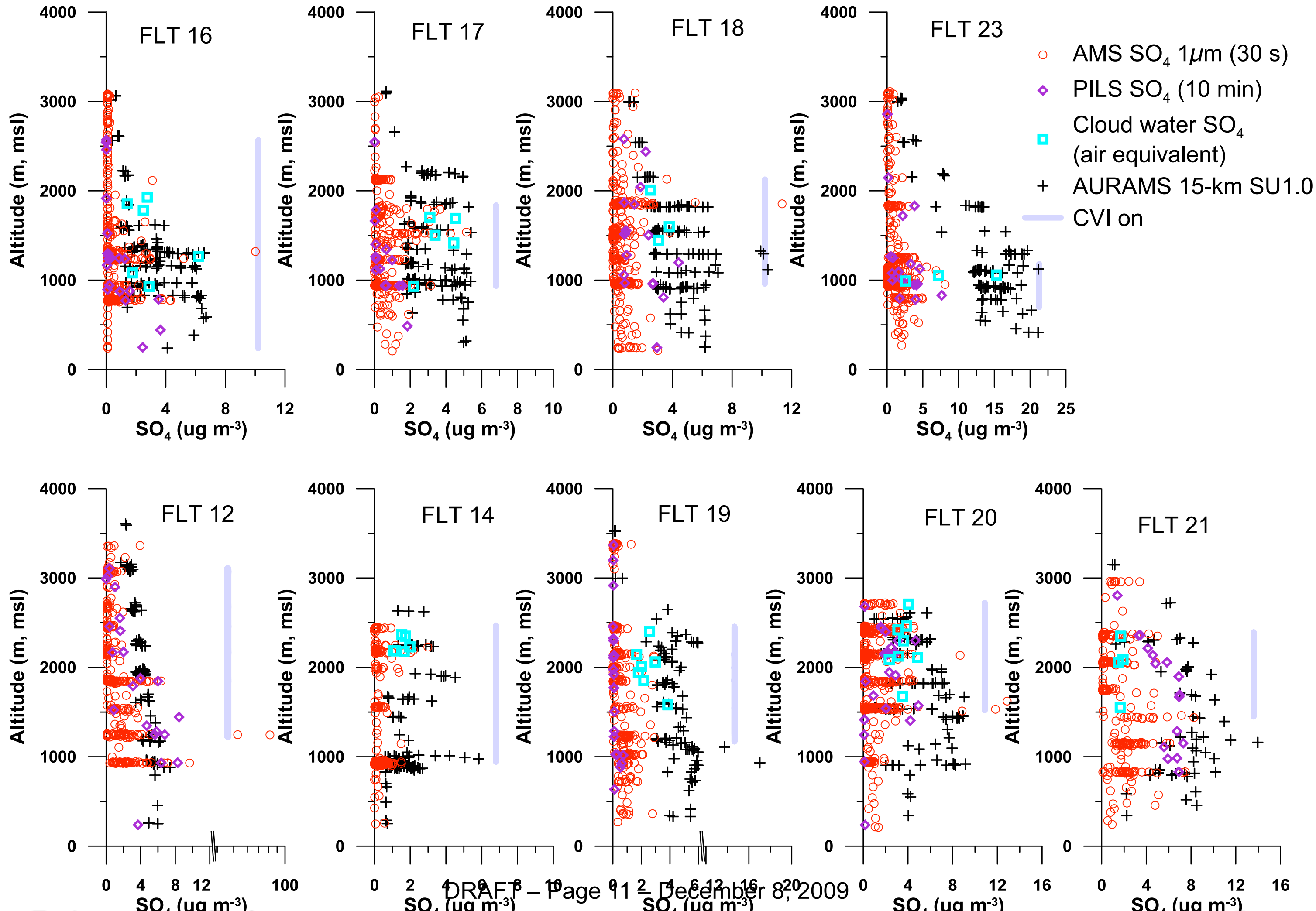
# Comparison against aircraft measurement (APS, SMPS) – $PM_{1.0}$




- Model seems to predict $PM_{1.0}$ well (vertical structure and magnitudes),
- uncertainty in assumed density used for mass conversion

# Comparison against aircraft measurement (AMS, PILS, and cloud water samples) – sulfate



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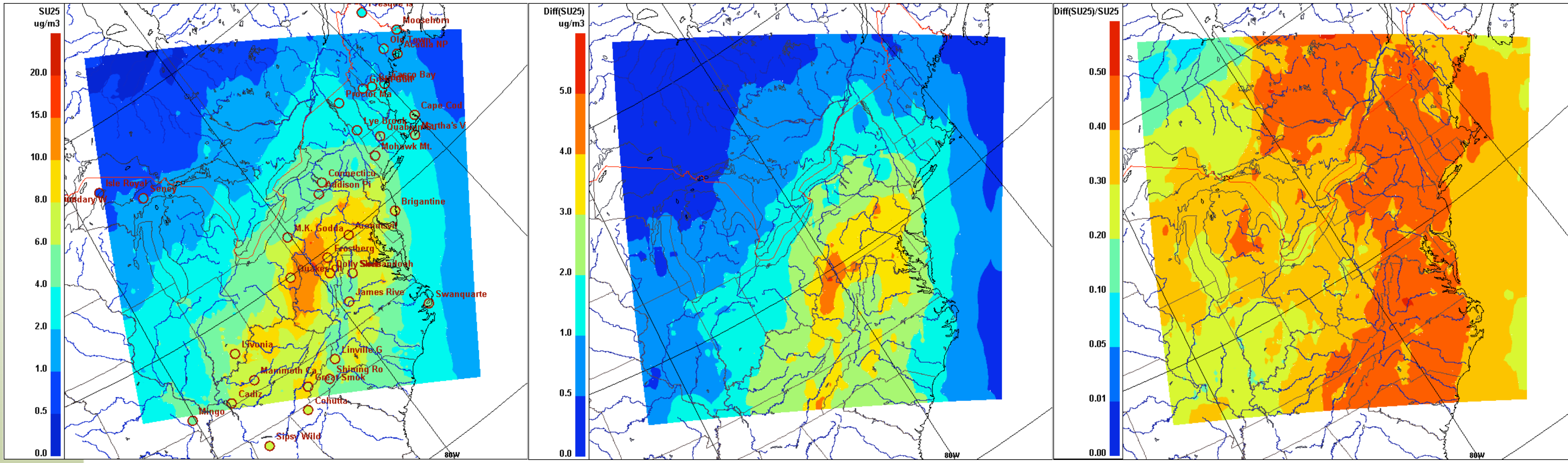
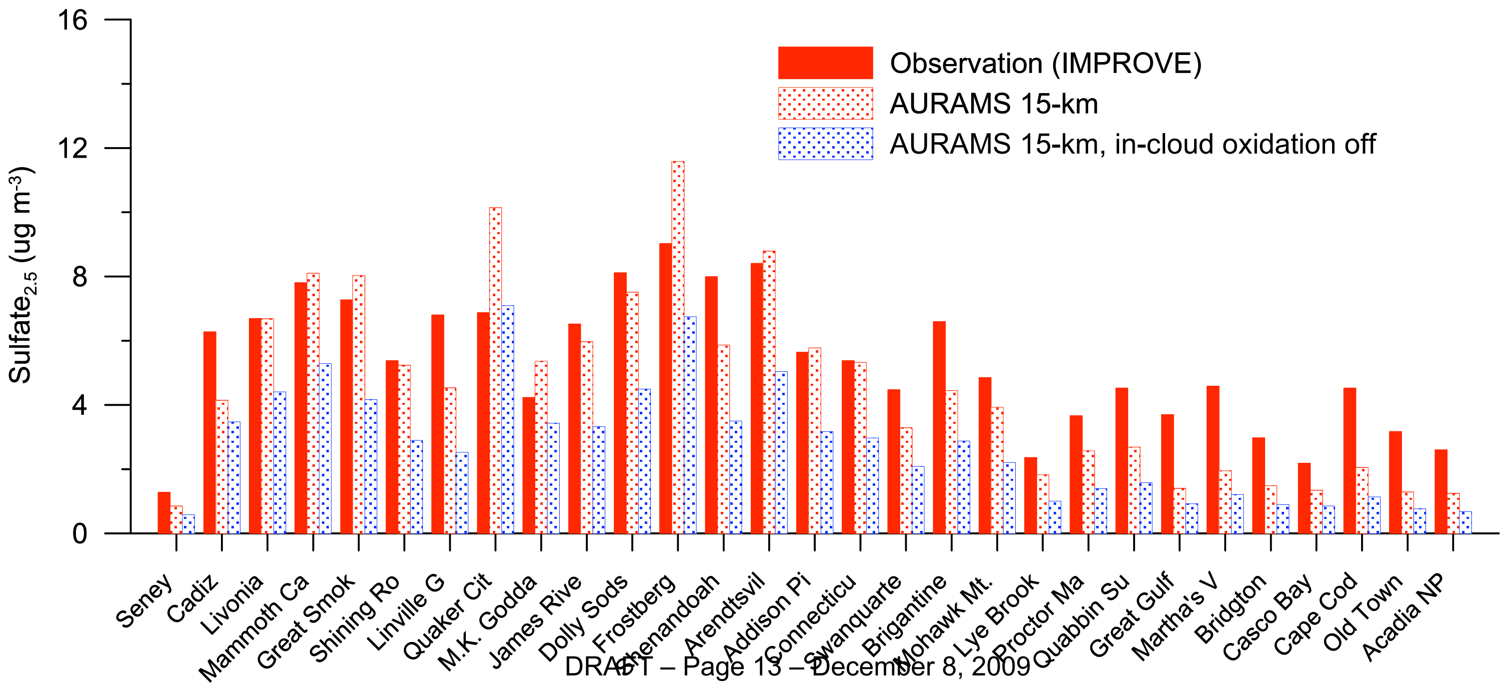
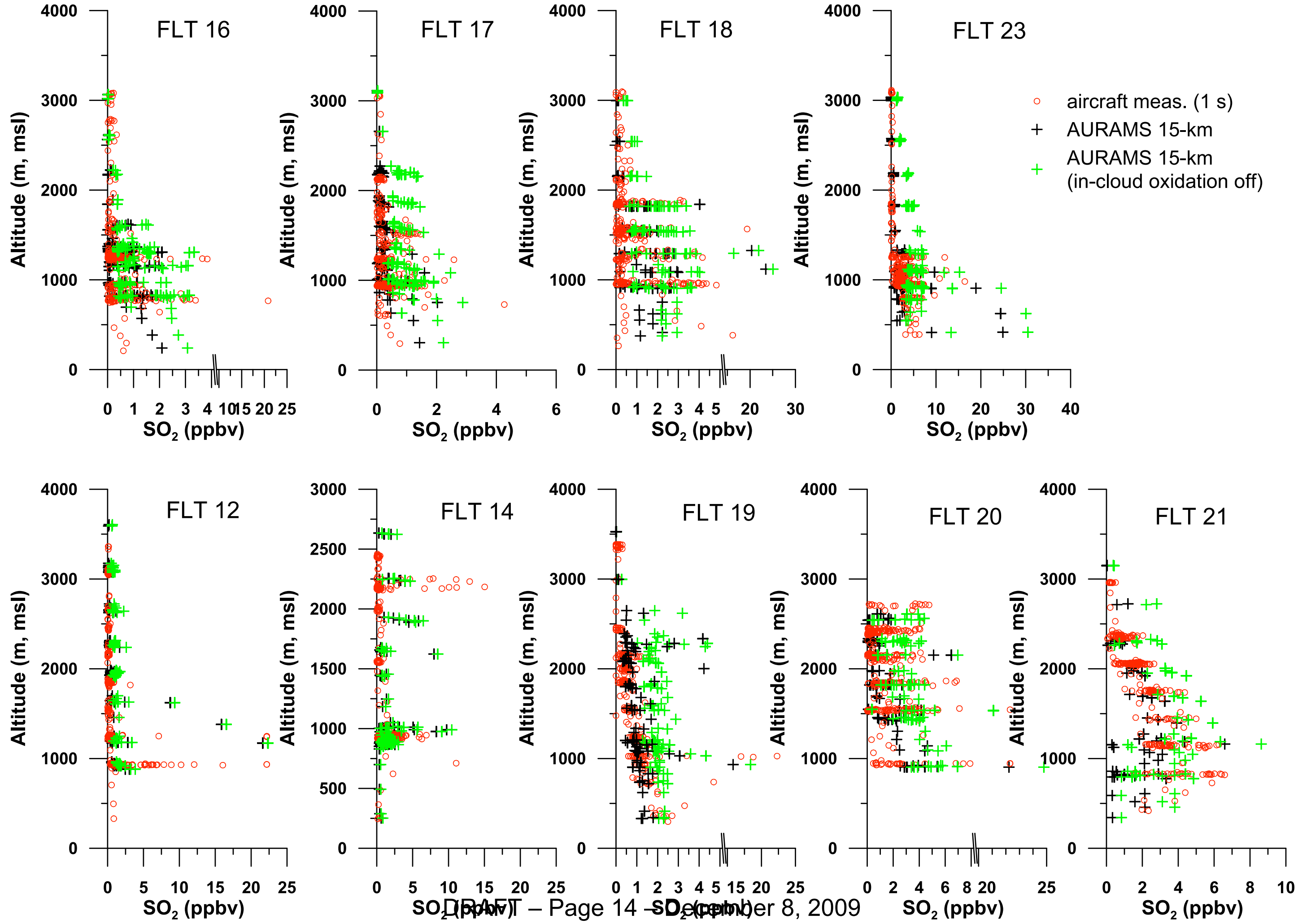
# Sensitivity to in-cloud oxidation at ground level

SU25, 2004/0714 – 2004/0818 SU25(basecase)-SU25(0cldoxi) Delta SU25 / SU25(basecase)

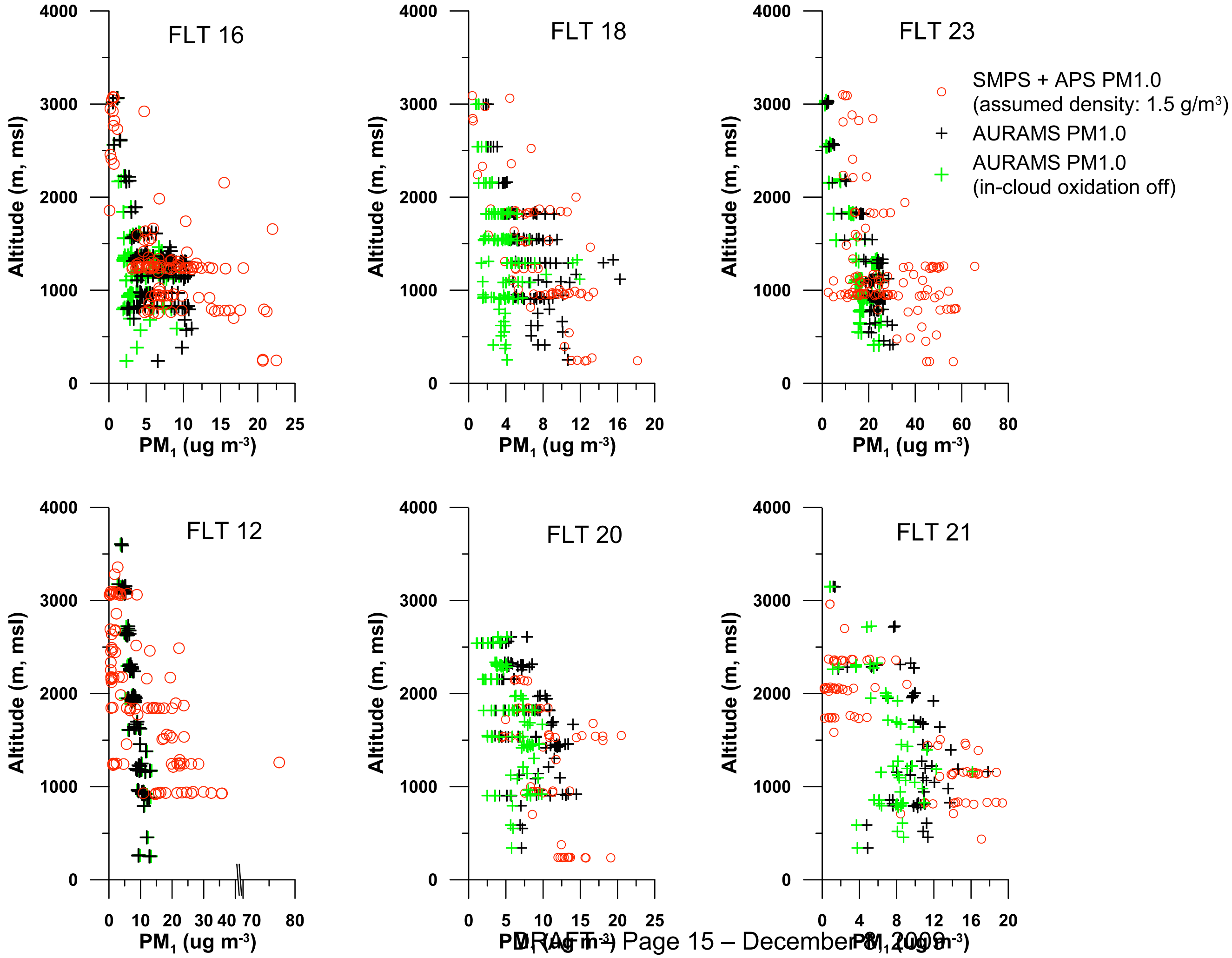


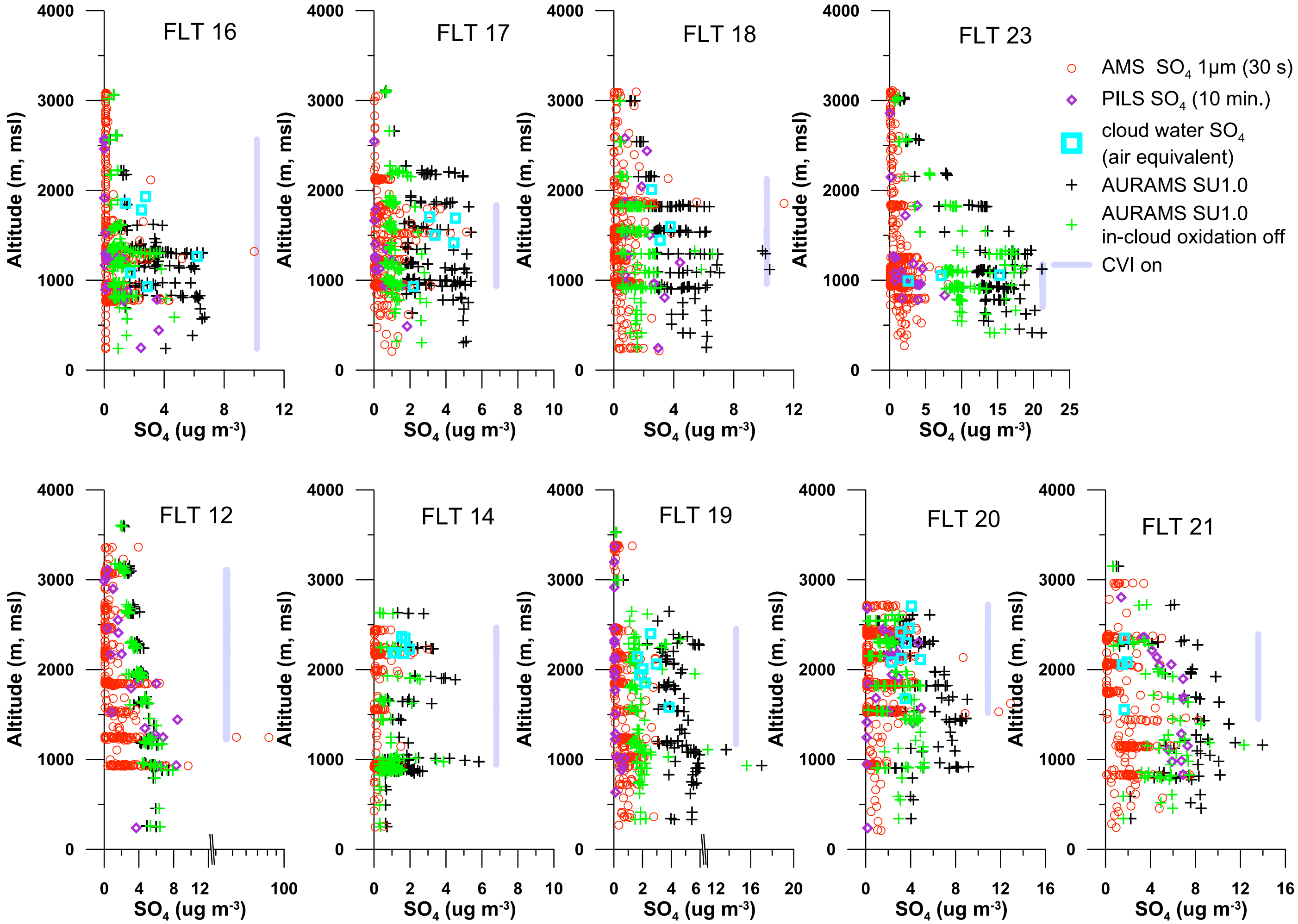
# Sensitivity to in-cloud oxidation aloft – $SO_2$



# Sensitivity to in-cloud oxidation aloft – PM1.0



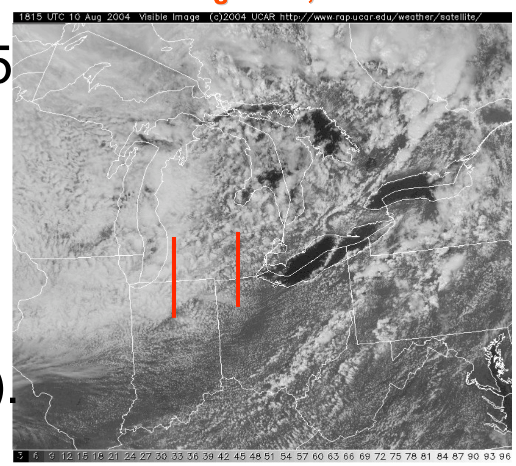
# Sensitivity to in-cloud oxidation aloft – sulfate

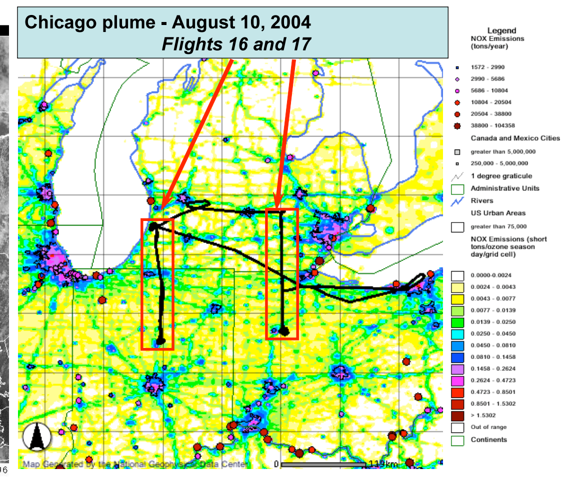


# Hi-res case study

- Flight 16 and 17 (August 10, 2004), cloud (Sc) processing in plumes downwind of Chicago area (under westerly flow).
- Aircraft sampling along two north-south lines: (FLT 16) 200 km east of Chicago (~86W) and (FLT 17) 200 km further east (~84W); below and in cloud.
- AURAMS simulation at 2.5 km resolution (cascading 42- to 15- to 2.5-km)
- Model output at every 2 minutes; sampling along flight track (given flight location at 2-min intervals).

1815 Z August 10, 2004



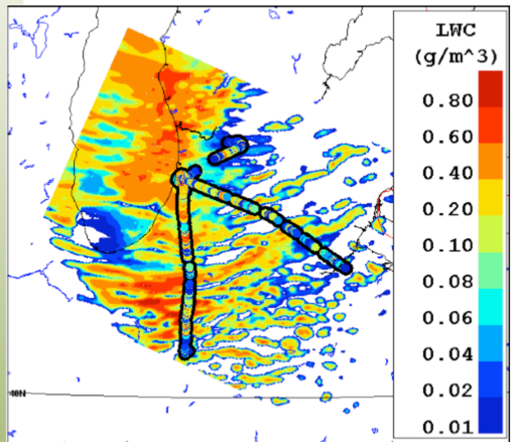

➢ Also used as a case study for the 7th WMO cloud modelling workshop, (chemistry) case 5;

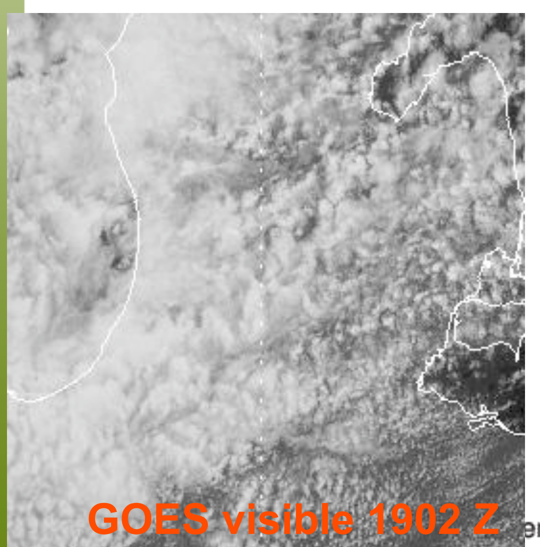
➢ Participating models in the case 5 study: AURAMS (EC, W. Gong et al.), MesoNH (LA/CNRS, M. Leriche et al.), and WRF-CHEM (NOAA/ESRL & CIRES, S.-W. Kim et al.)

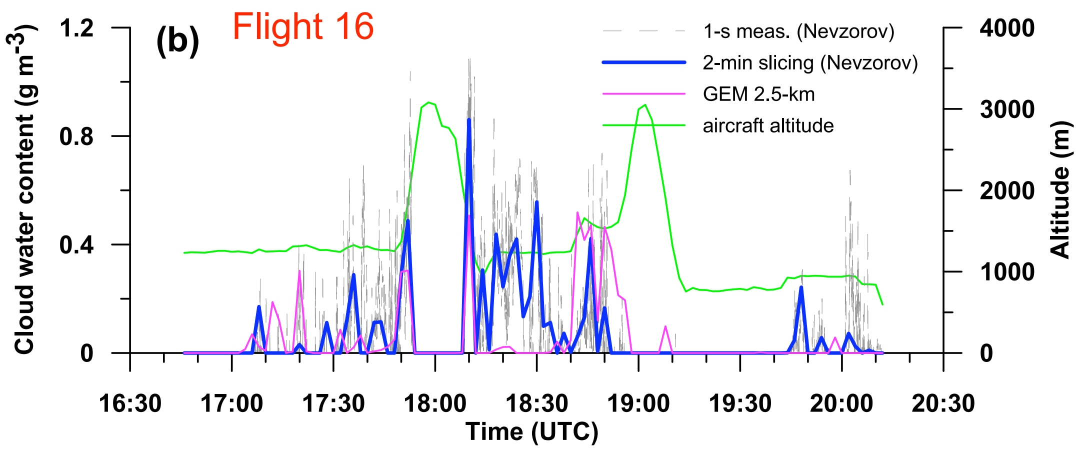
*Gong et al. (2009), Air Pollution Modeling and Its Application XX, edits. S.T. Rao and D. Steyn, Springer.*

# Liquid water content (LWC)

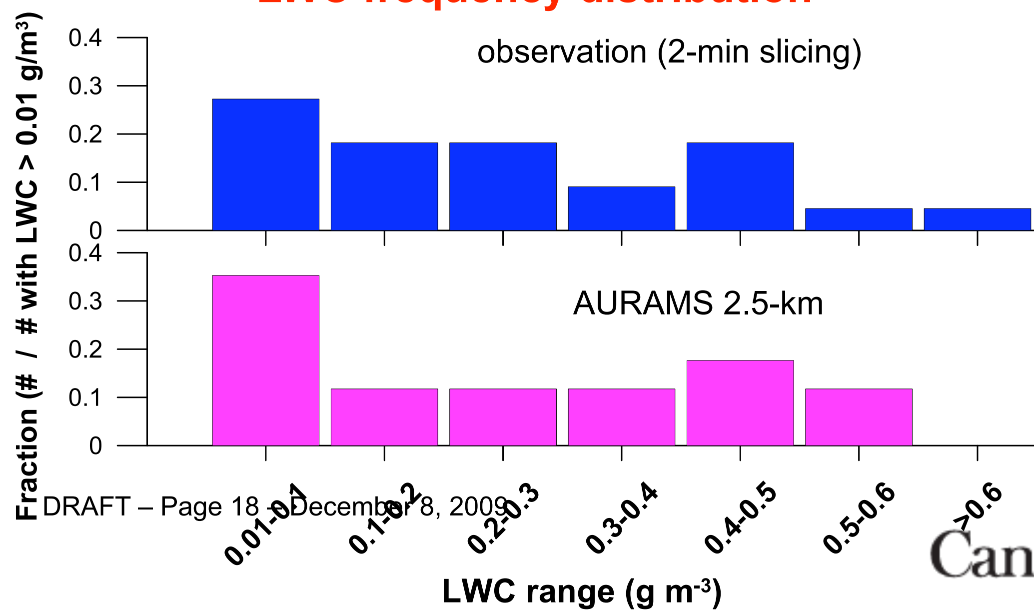
AURAMS (GEM) 2.5-km, 1235 m, 19 Z



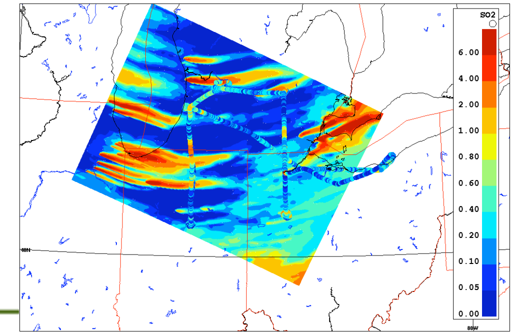



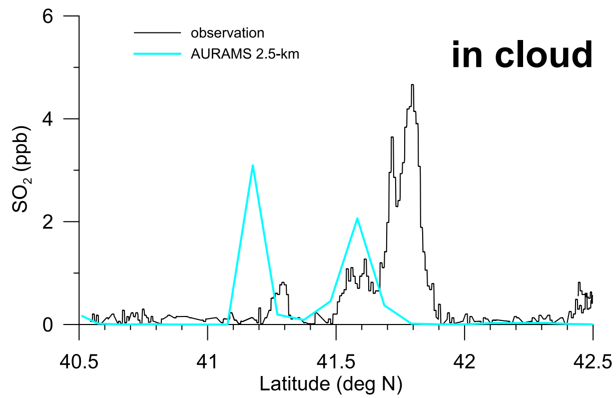

LWC frequency distribution

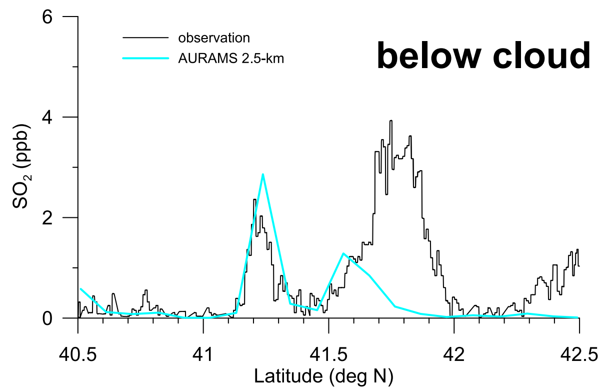


# Gaseous species – $SO_2$

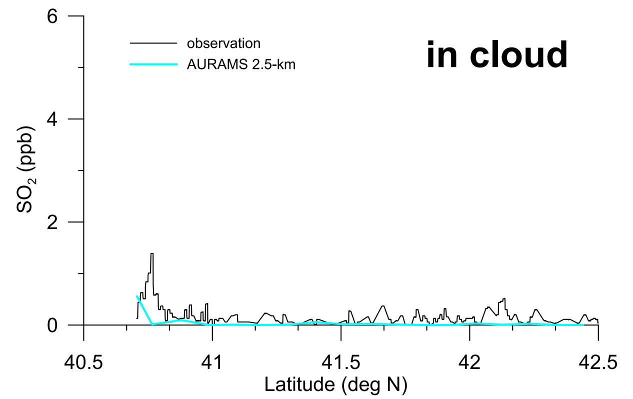


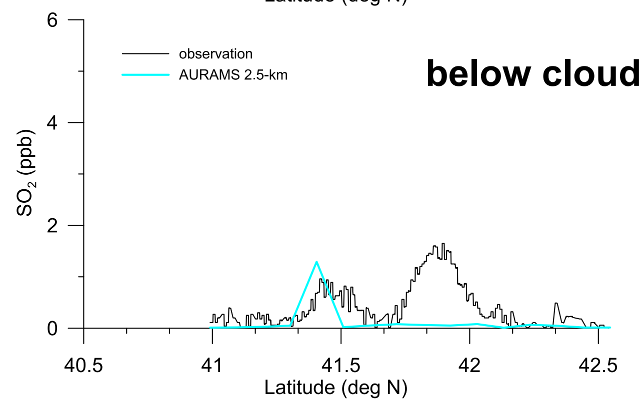
## FLT 16 (86 W)



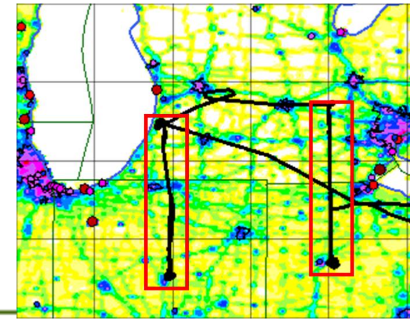

## FLT 17 (84 W)



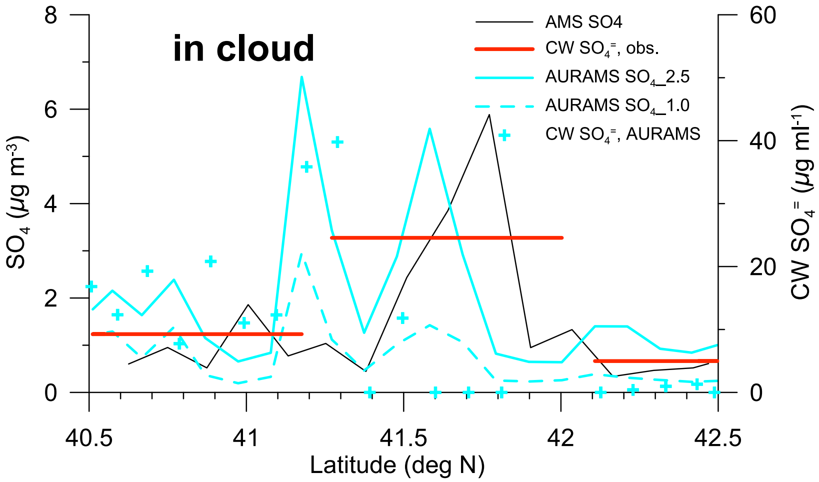




**Both observation and model show depletion of SO2 at cloud level downwind (FLT 17) – an indication for in-cloud oxidation?**

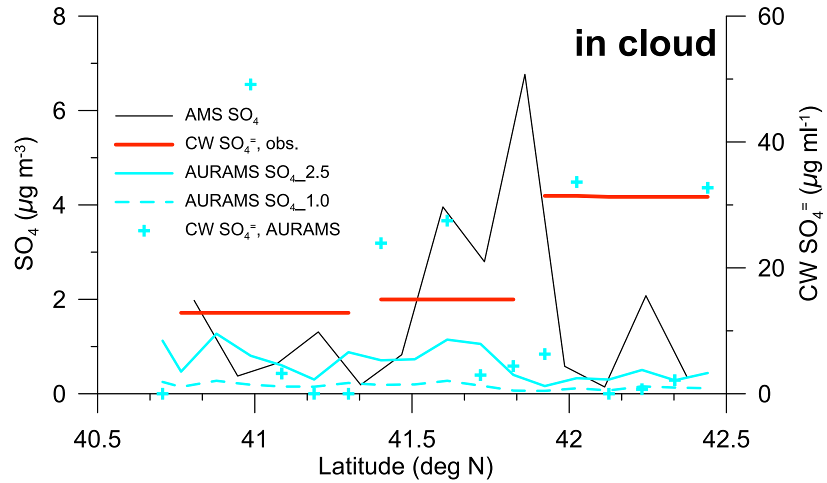
# Particle $SO_4$ and CW $SO_4^=$

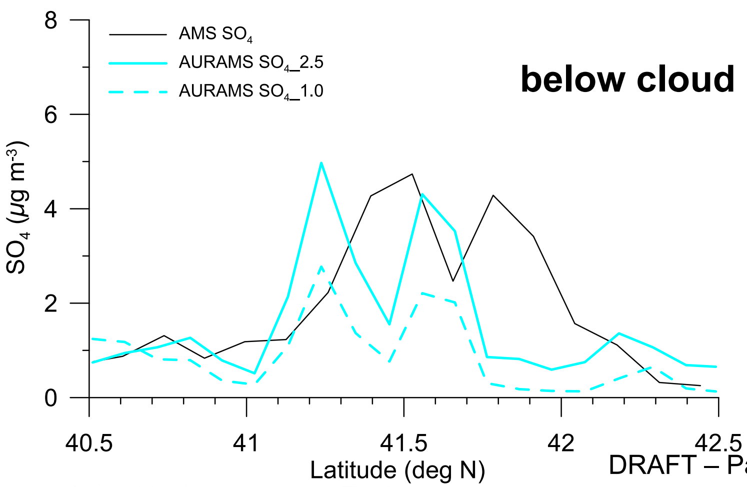


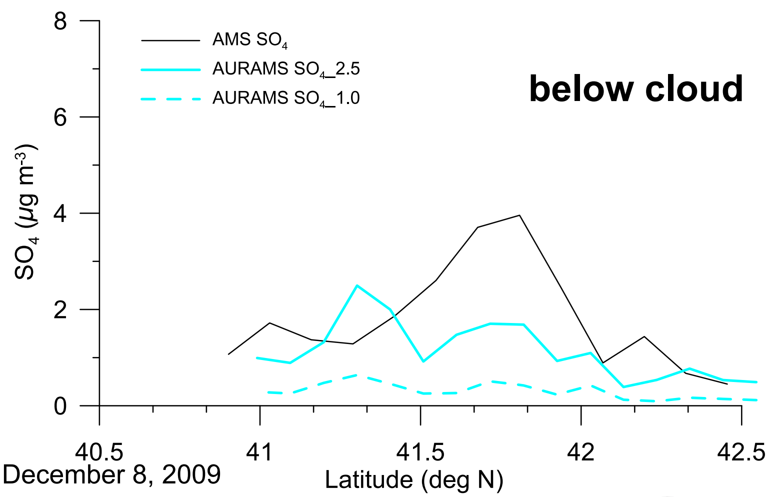
## FLT 16 (86 W)



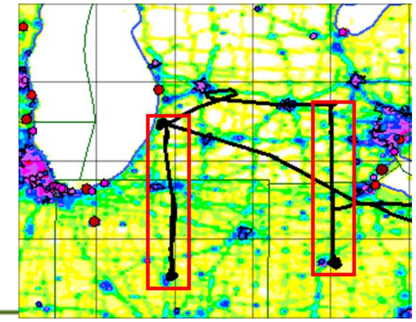

## FLT 17 (84 W)

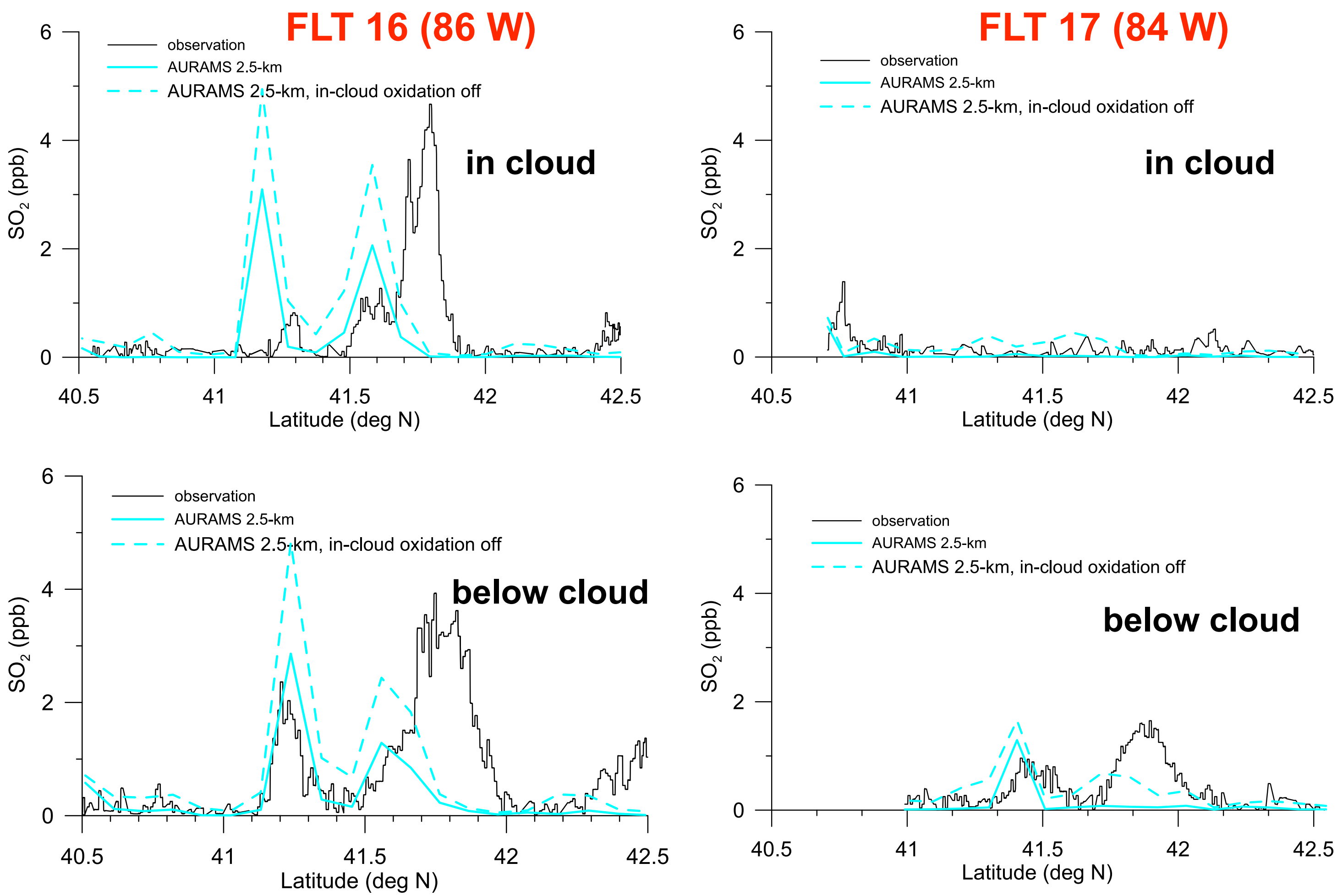


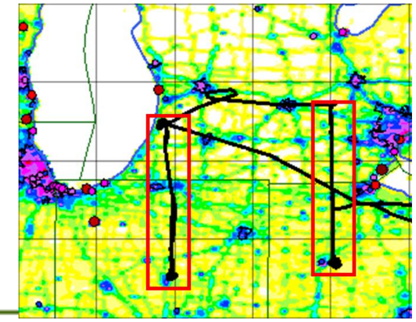



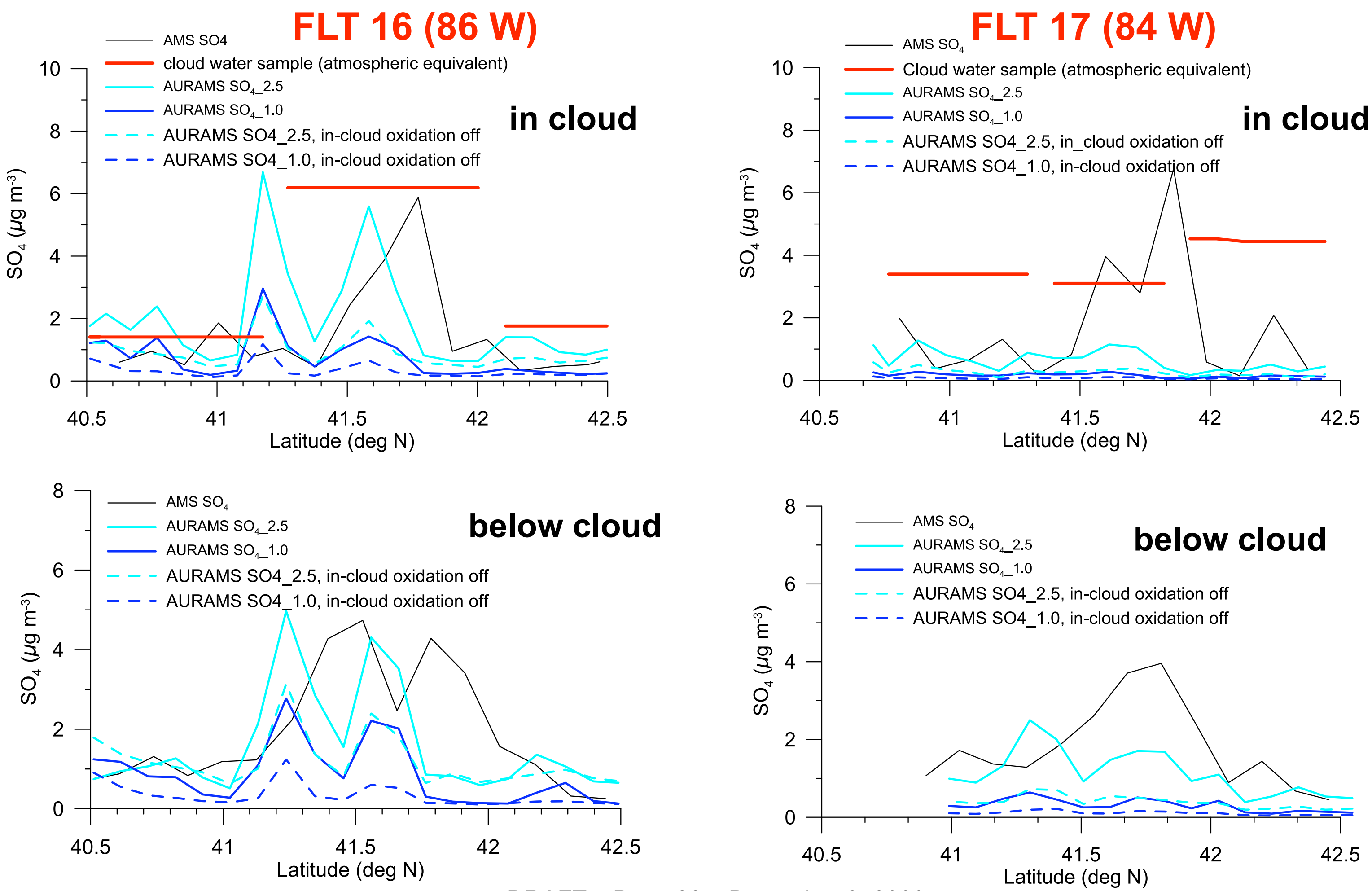
# Sensitivity on in-cloud oxidation – $SO_2$





# Sensitivity on in-cloud oxidation – $SO_4$





# Lessons learnt and future work

- The modelling of cloud processing of gas and aerosols is complicated because it is controlled by several factors, e.g., cloud and precursors – both having high spatial and temporal variability; model uncertainties are high.
- The model is shown to have some skill in modelling the cloud process, more successful in some cases than others (e.g., capturing plumes at hi-resolution); there is observational evidence for significant aqueous-phase production.
- More quantitative (definitive) evaluation is still challenging: availability of (and uncertainty in) observational data; evaluation methodology (spatial & temporal disparity/mismatch between model and observation).
- Existing evaluation of modelled cloud properties for the ICARTT study (Zhang et al., 2007) showed that the meteorological model (GEM) over predicted cloud liquid water content in general for this study period, which ***may*** lead to model over-prediction of aqueous-phase sulfate production.
- There is a predictability issue; the challenge is to quantify (if possible) the uncertainties in model prediction.

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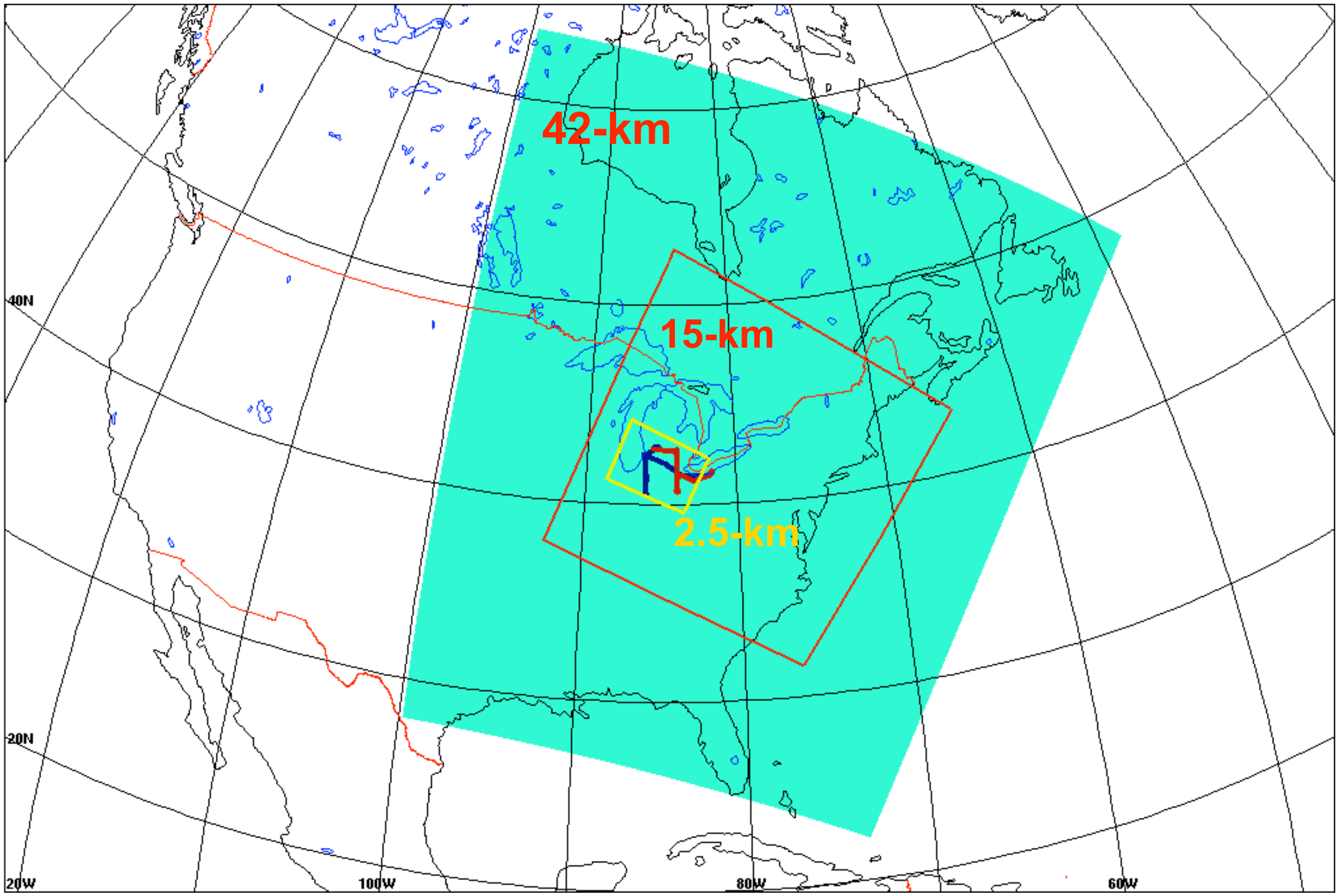
# Thank you!

# AURAMS simulation domains for case 5

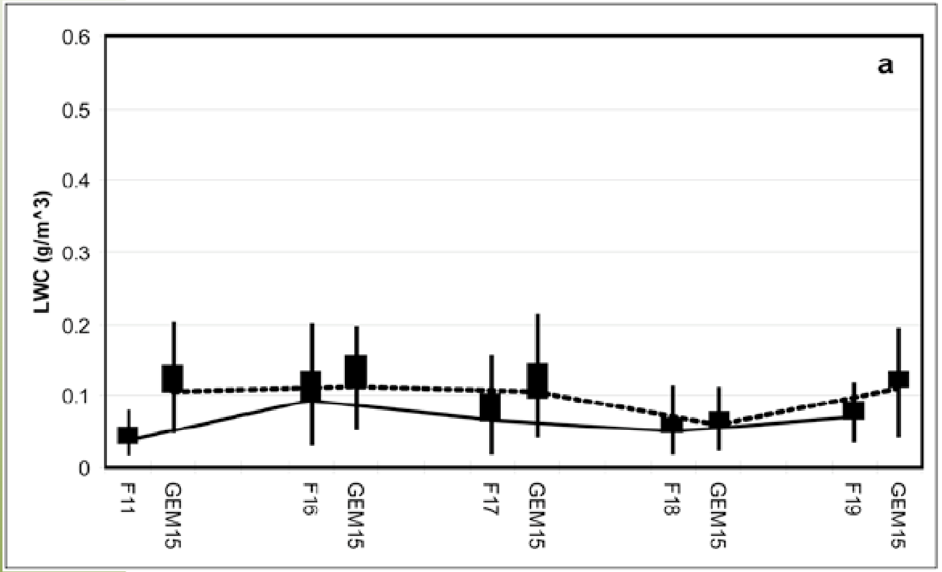


*Courtesy of Junhua Zhang*

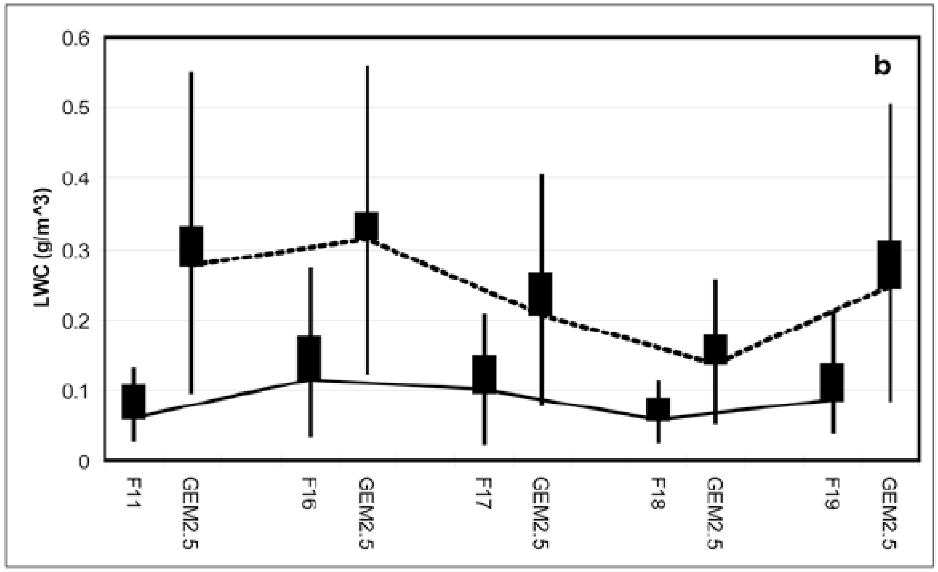
# Model Grid Scale LWC Comparison, 5 Sc Flights

**Aircraft: solid line GEM: dashed line**

## GEM15




## GEM2.5

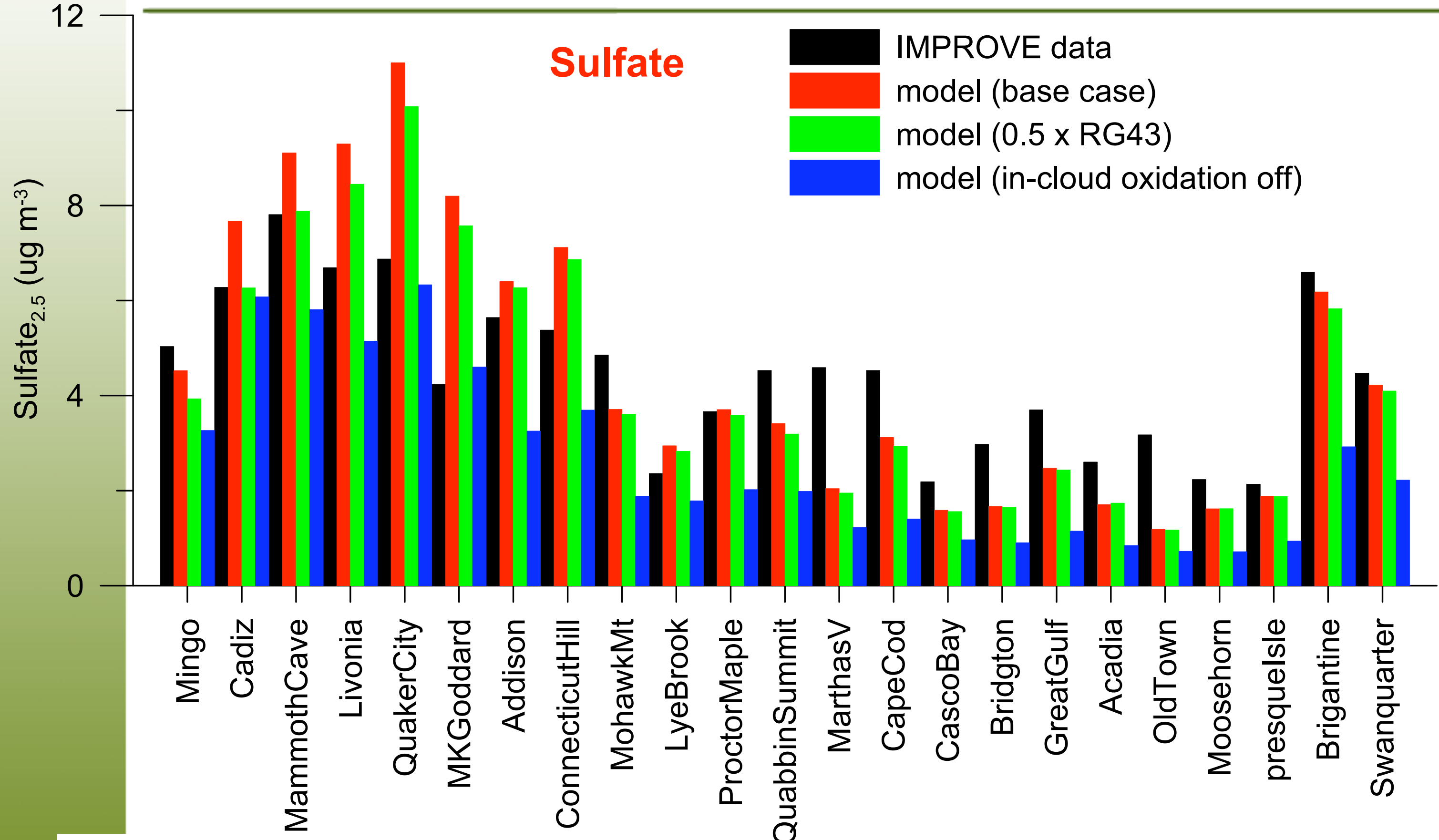



| | Aircraft_15 | GEM15 |
|---|---|---|
| Mean | 0.09 | 0.13 |

| | Aircraft_2.5 | GEM2.5 |
|---|---|---|
| Mean | 0.13 | 0.29 |

# In-cloud oxidation vs. clear-air oxidation

Sites are arranged on the axis to go from west (left) to east (right)




Modelled sulfate is more sensitive to in-cloud than clear-air oxidation.

Clear-air oxidation has greater impact closer to sources than farther downwind.

- Potential over-prediction of in-cloud production due to over-prediction of cloud water by GEM (Zhang et al., 2007 JGR)
- Possible error in emission from power plants.