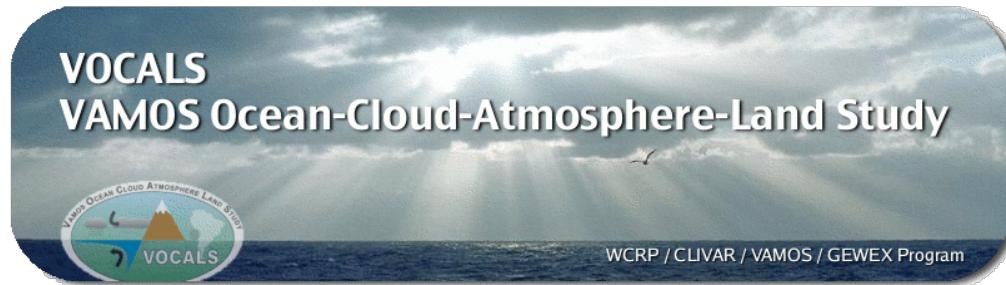


Simulating aerosols, radiative forcing, and impacts on marine stratocumulus during VOCALS REx

Scott Spak, Marcelo Mena-Carrasco, Pablo Saide,
Greg Carmichael



Motivations: Policy Relevance

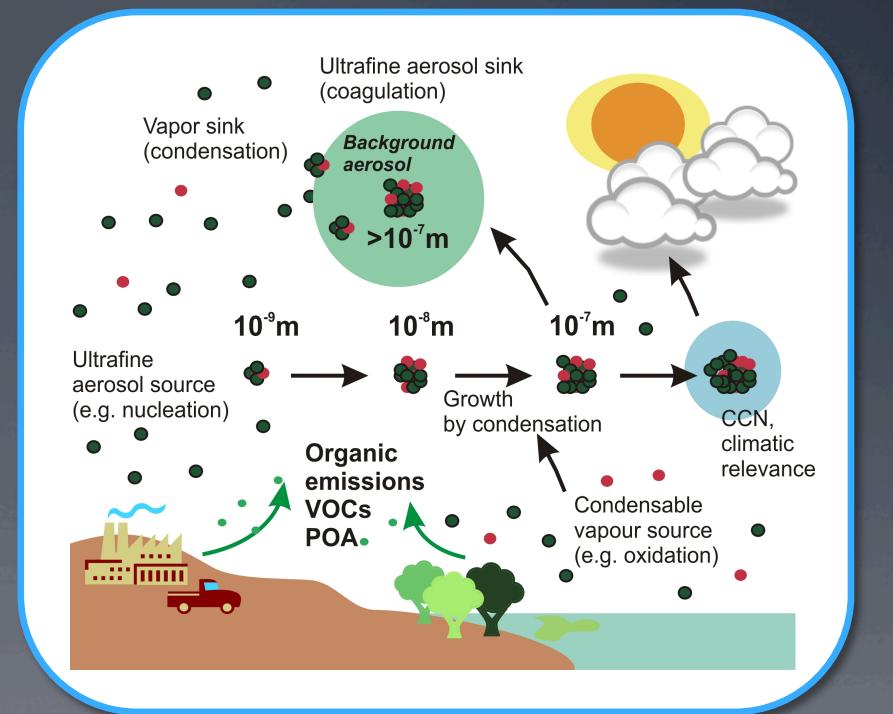
- How have anthropogenic emissions altered SEP Sc? Which sources matter most? How are they expected to change?
- Unintended regional/global climate consequences of Santiago's urban development?
- What about geo-engineering proposals?
- The SEP, beyond clouds and climate
 - traditional criteria air pollution questions for human health
 - direct radiative forcing and BC:S strategies
 - acid deposition to the southern oceans

Motivation → Approach

Simulate & evaluate basic properties of chemical transport during REx

then aerosol effects on cloud microphysics & climate

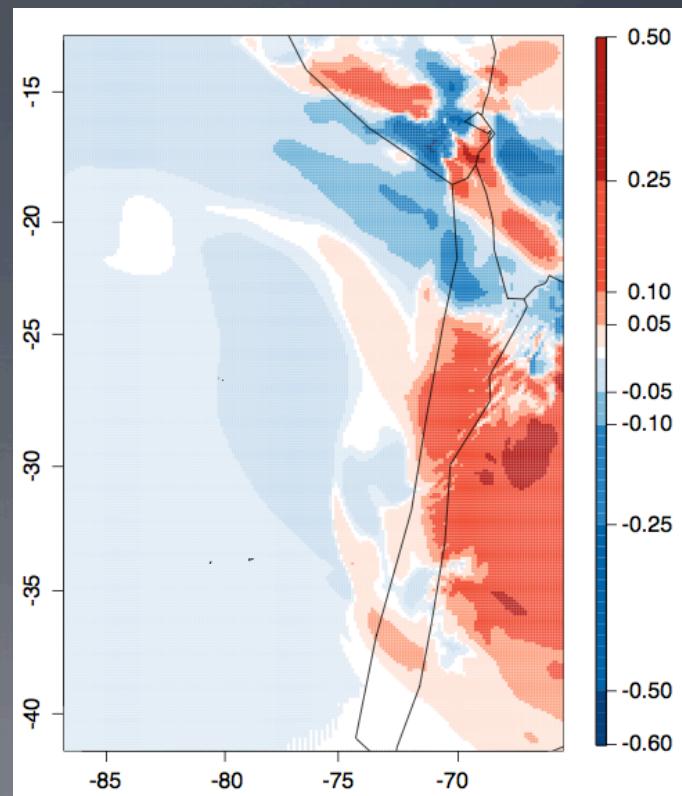
then response to changes in human activities



Building on ... tracer modeling

- Daily variability in SO_4 driven by synoptic condition: SEP Subtropical High location & strength
- DMS importance by omission: anthropogenic emissions can't account for observed offshore SO_4 & SO_2 concentrations
- Coastal and Altiplano emissions entrained from FT to clouds in WRF-STEM

Δ Cloud-level LPS SO_4 ($\mu\text{g}/\text{m}^3$)
RFo3-RFo6 & RF10 – other flights



Model Configuration

WRF-Chem v3.1.1 @ 12 km, 27 layers

- CBM-Z + MOSAIC (8 bins), Lin microphysics, Grell cumulus
- No DMS yet!
- MYNN v2.5 PBL
- Direct, 1st & 2nd indirect, semi-direct aerosol radiative forcing
- Restarted every 5 days with 1 day spin-up for continuity
- MOZART 4 boundary conditions
- **Emissions**

daily 1 km FINN fires (Wiedinmeyer *et al.*, GMDD, 2010)

+ MEGAN biogenics

+ VOCA anthropogenic emissions ON/OFF through 10/27

Campaign chemical transport evaluation

No DMS, too much OC (FINN + MOZART)

	Observed	Mean Bias	RMSE	FB*	FE*
O ₃ (ppb)	37.25	-3.53	10.47	-0.12	0.26
CO (ppb)	65.68	2.60	20.40	0.06	0.13
SO ₂ (ppt)	52.71	-32.00	152.04	-1.25	1.49
SO ₄ (μg/m ³)	0.43	-0.01	0.73	0.34	1.09
NO ₃ (μg/m ³)	0.011	0.002	0.035	NA	NA
NH ₄ (μg/m ³)	0.08	-0.01	0.11	0.35	1.31
OC (μg/m ³)	0.13	0.37	0.52	1.58	1.81

*Fractional bias = (2 x bias)/(observed + modeled)

Bolded values meet US EPA/community performance standards

Italicized values exceed targets

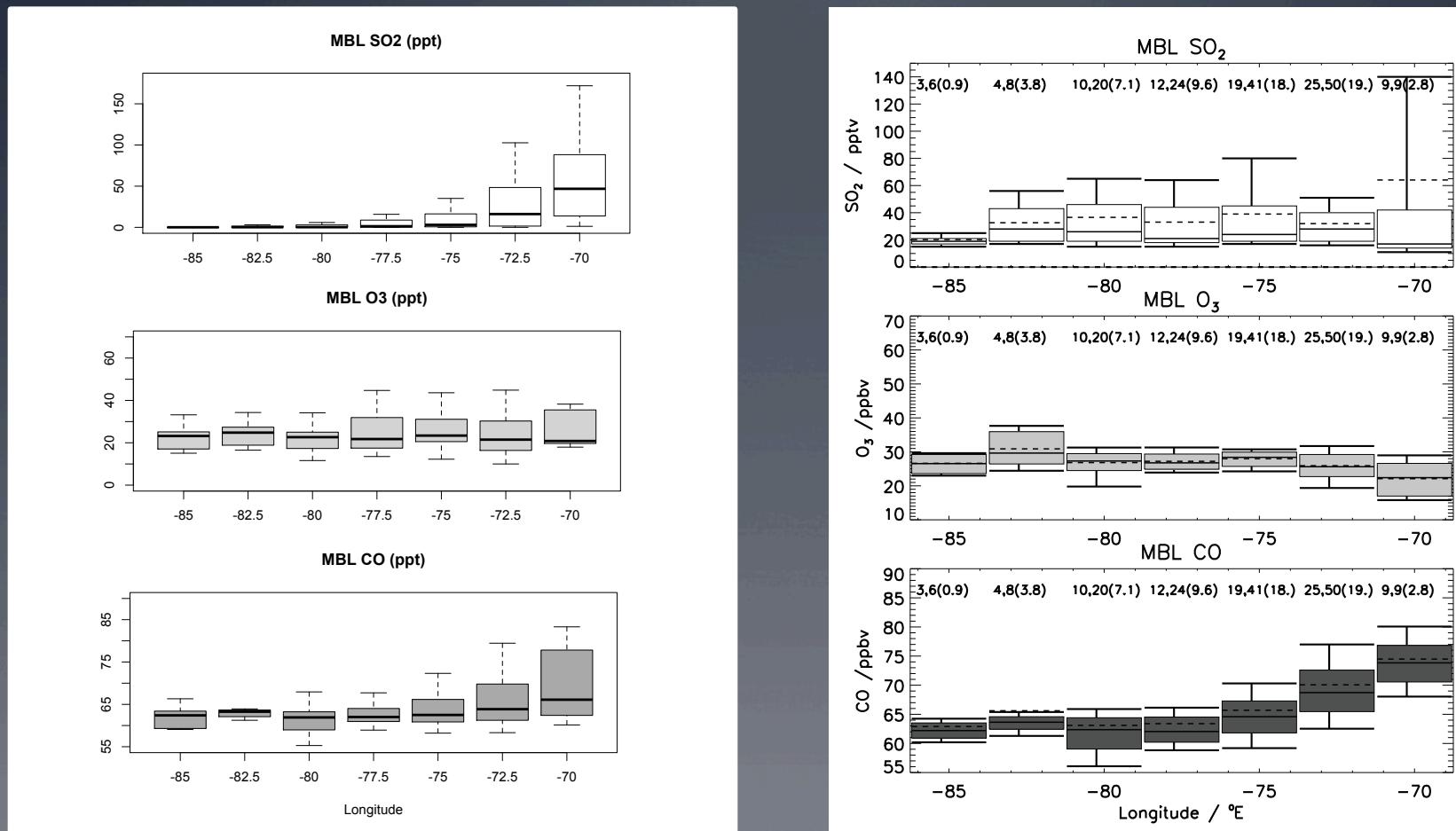
20° S chemical transport evaluation

all C-130 flights @ 1 minute average, processed per Allen *et al.* (ACPD)

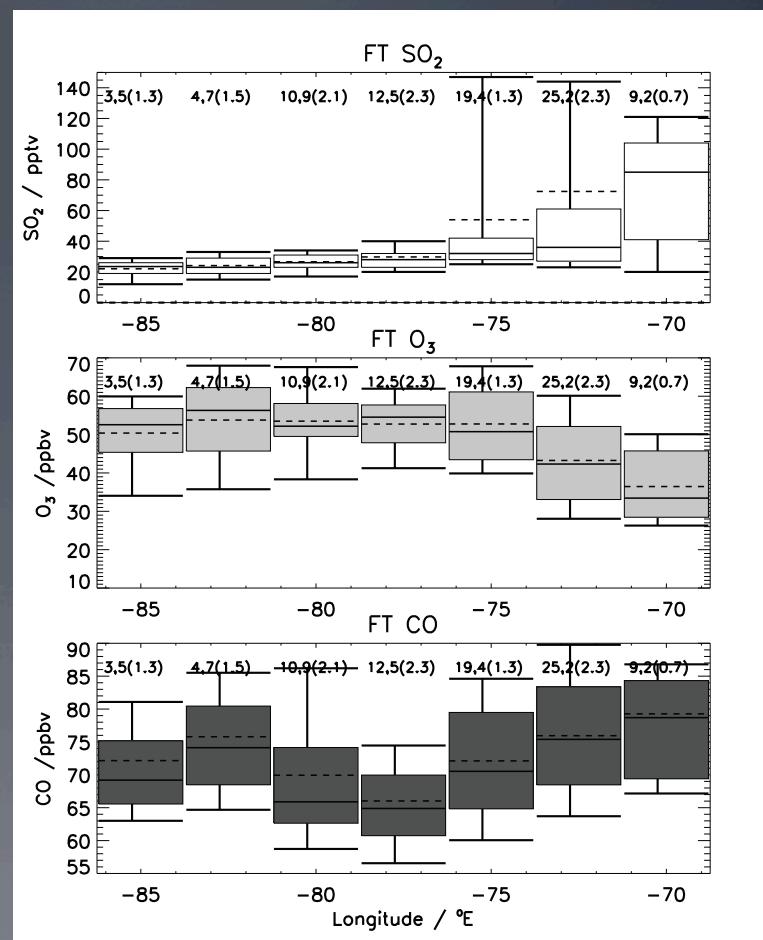
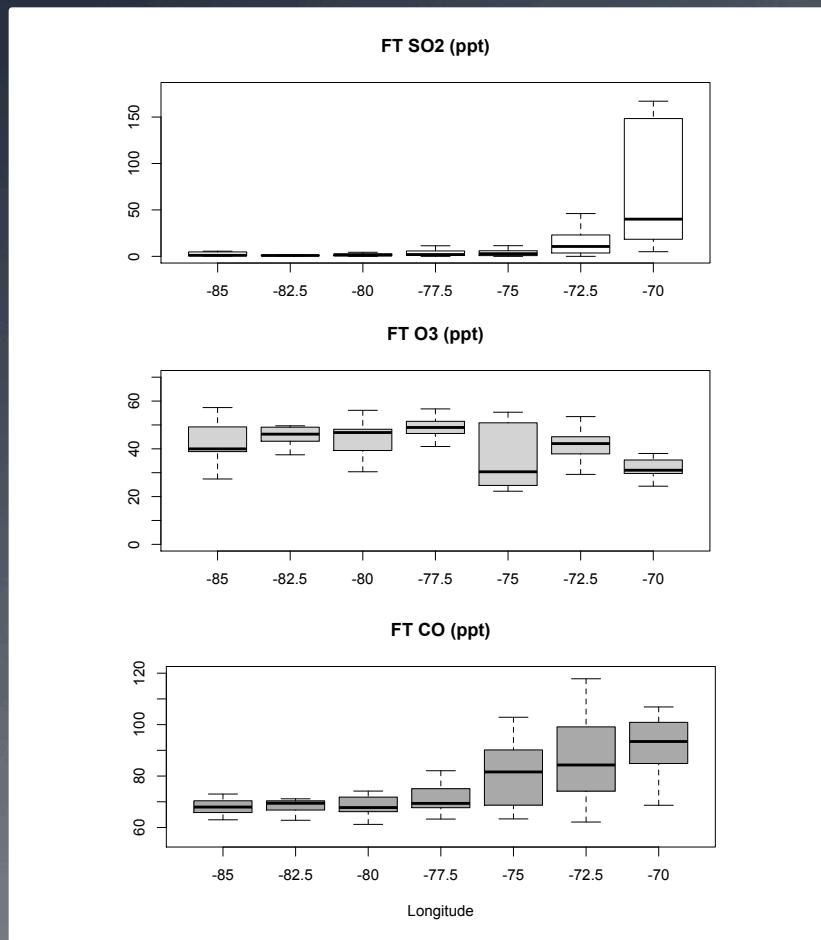
	Observed	Mean Bias	RMSE	FB*	FE*
O ₃ (ppb)	34.84	-5.71	10.83	-0.22	0.30
CO (ppb)	65.89	0.83	16.37	0.03	0.11
SO ₂ (ppt)	34.31	-16.06	61.75	-1.21	1.52
SO ₄ ($\mu\text{g}/\text{m}^3$)	0.47	-0.07	0.63	0.06	1.13
NO ₃ ($\mu\text{g}/\text{m}^3$)	0.011	0.001	0.021	NA	NA
NH ₄ ($\mu\text{g}/\text{m}^3$)	0.08	-0.02	0.10	-0.03	1.36
OC ($\mu\text{g}/\text{m}^3$)	0.12	0.33	0.47	1.44	1.81

Bolded values meet US EPA/community performance standards
Italicized values exceed targets

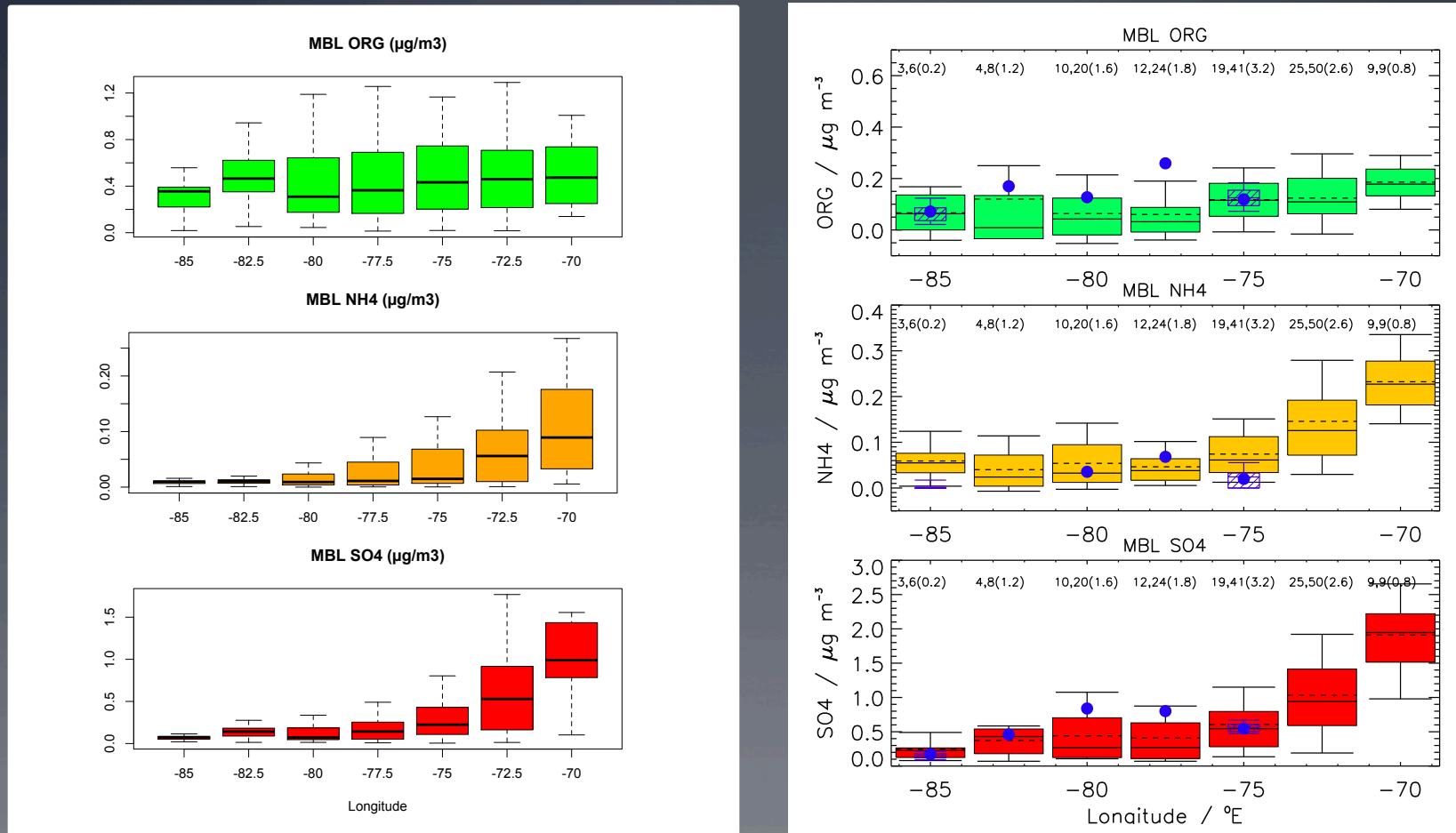
WRF-Chem MBL Gases @ 20°S vs. Allen *et al.* (2011)



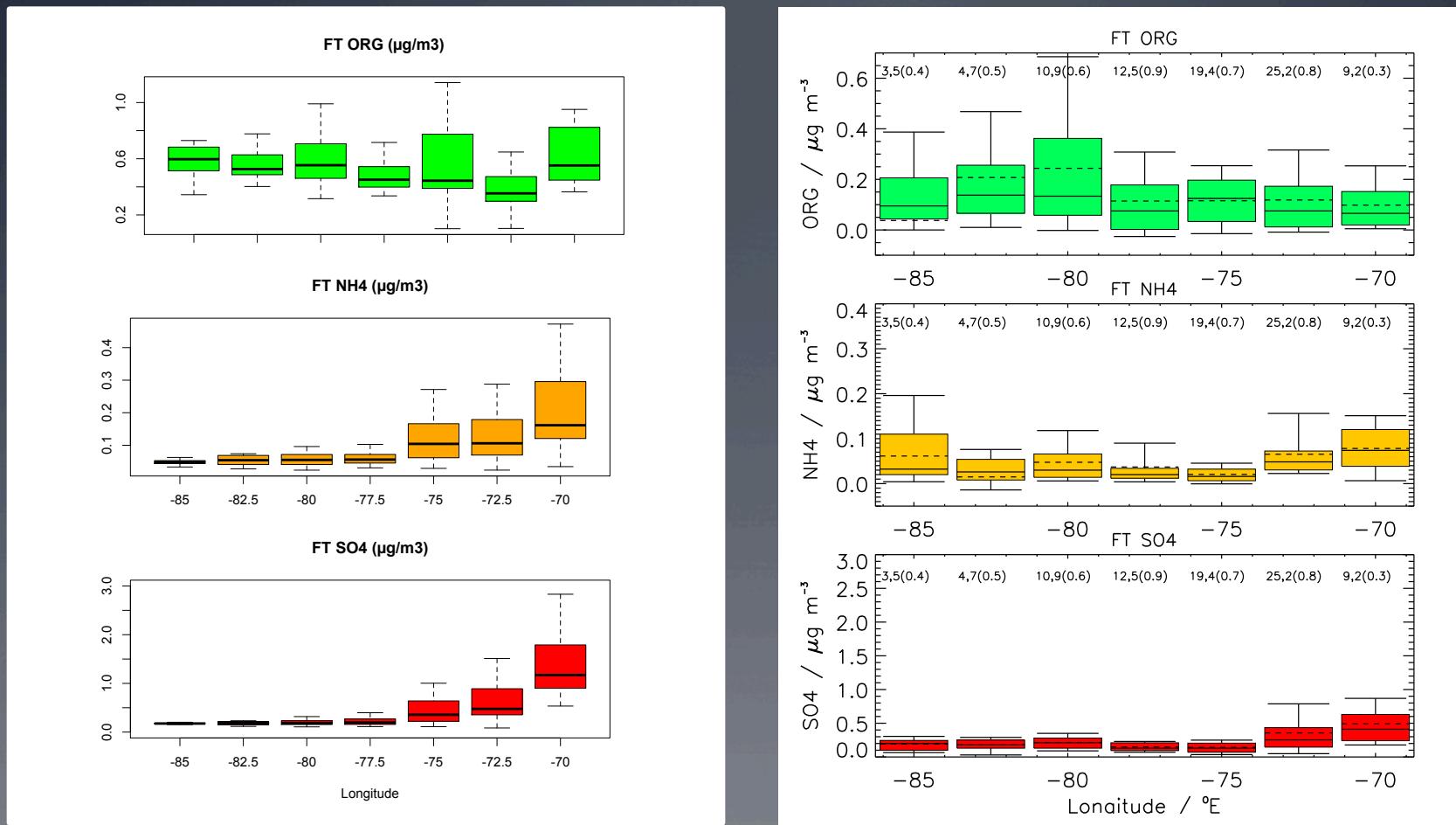
FT Gases: whiskers better than boxes



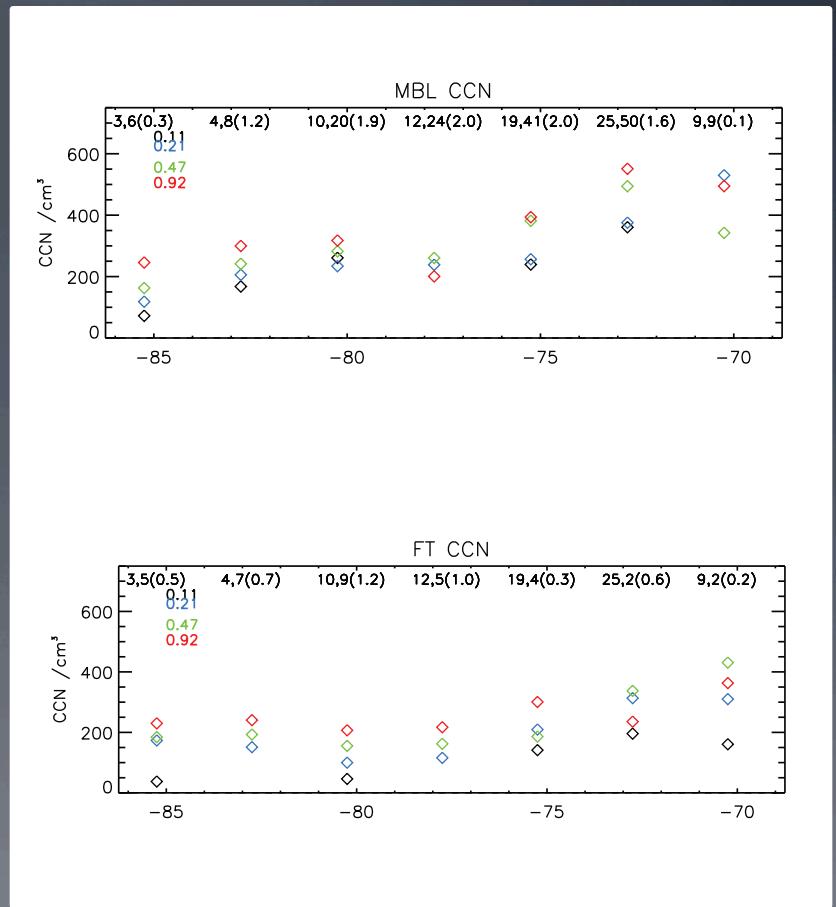
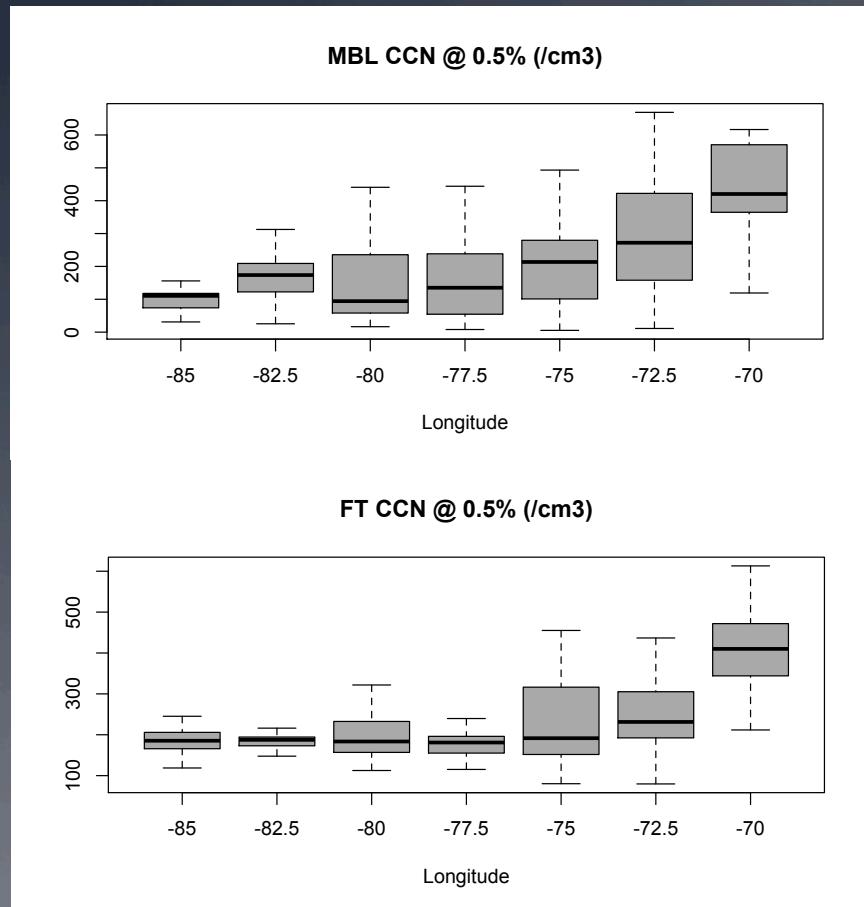
MBL aerosol composition



FT aerosol composition

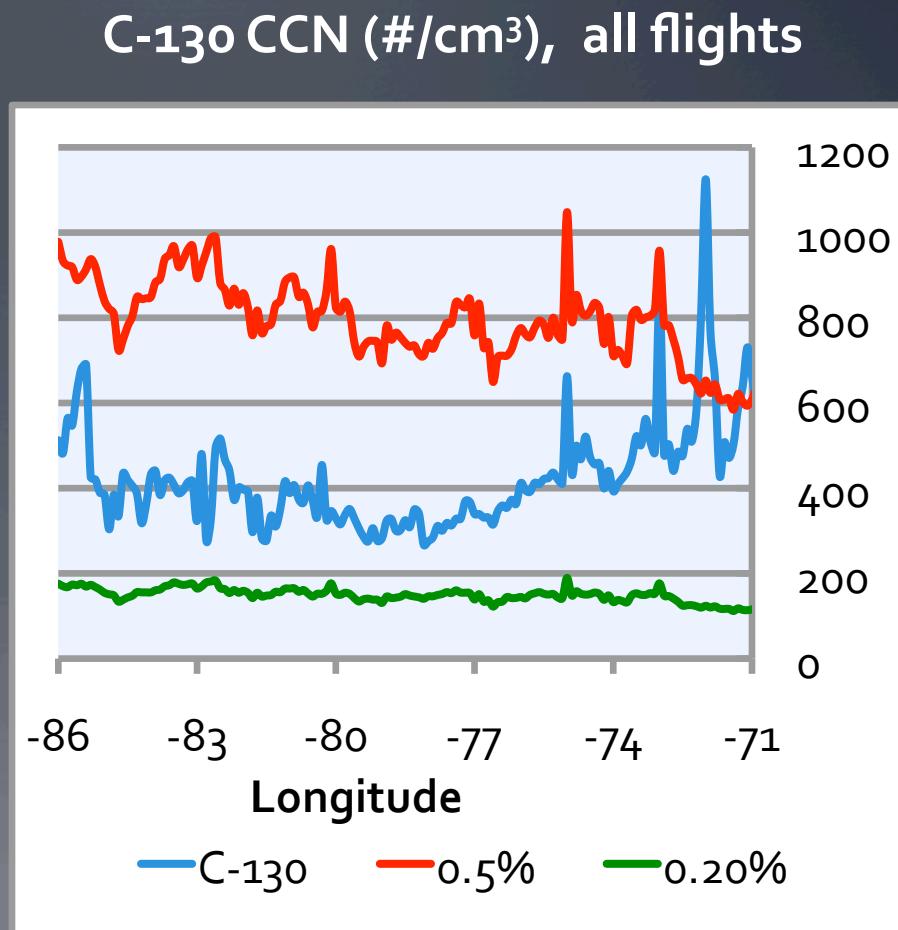


Cloud Condensation Nuclei



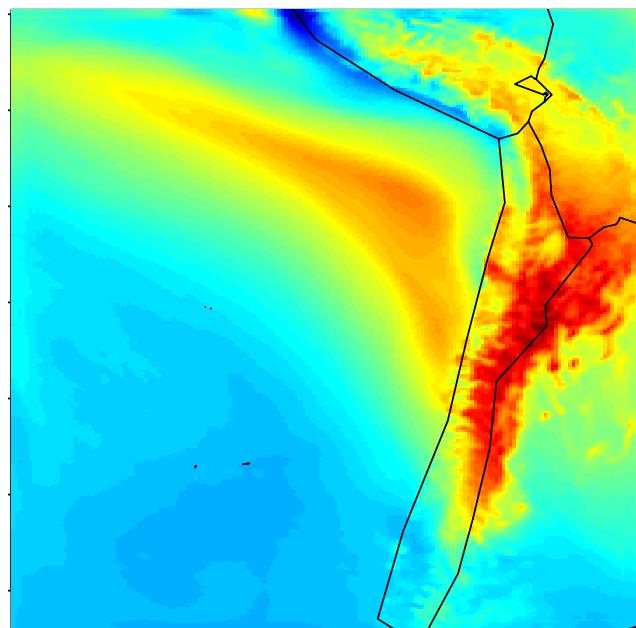
A word about CCN in WRF-Chem...

- WRF-Chem assumes 0.2% supersaturation for aerosol coupling
- CCN not well simulated by any WRF supersaturation level: observed @ 0.55 – 0.75% < modeled CCN @ 0.5%
- Variability along flight paths encouraging

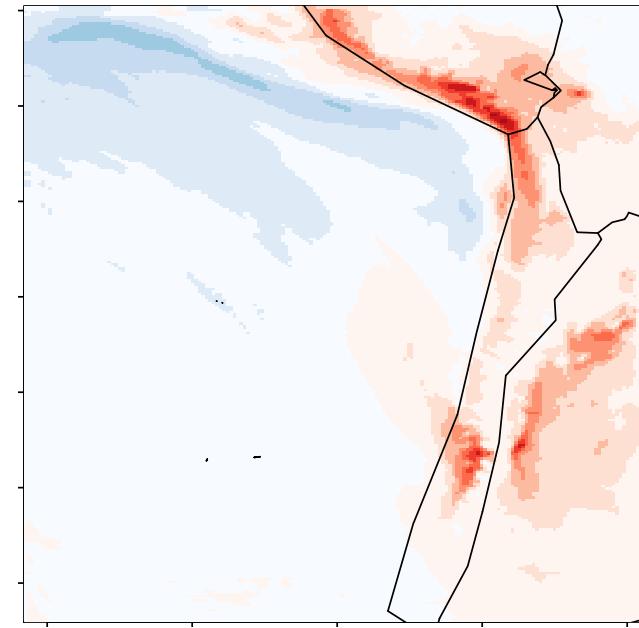


Average 10/15 – 10/27 ozone (ppb), cloud level

Base case

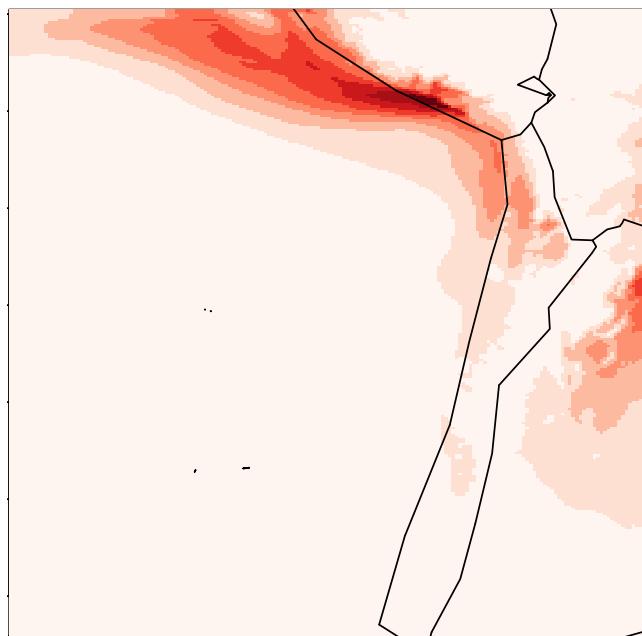


Anthropogenic impact

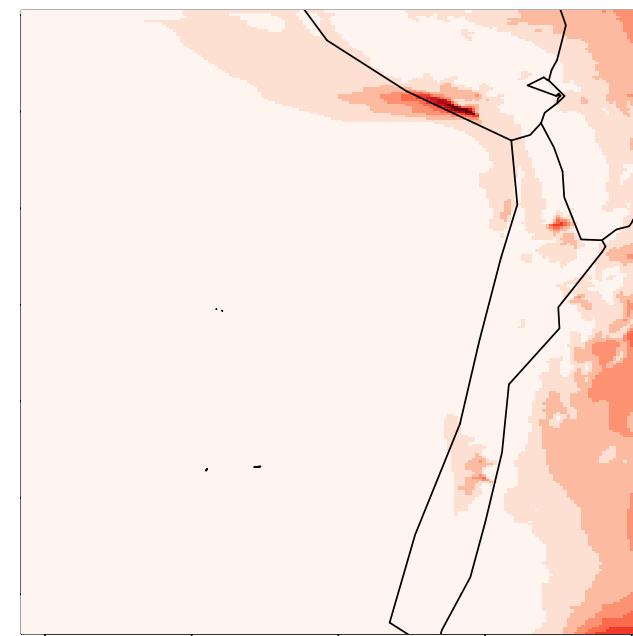


Anthropogenic impact: cloud-level aerosol properties

Sulfate ($\mu\text{g}/\text{m}^3$)



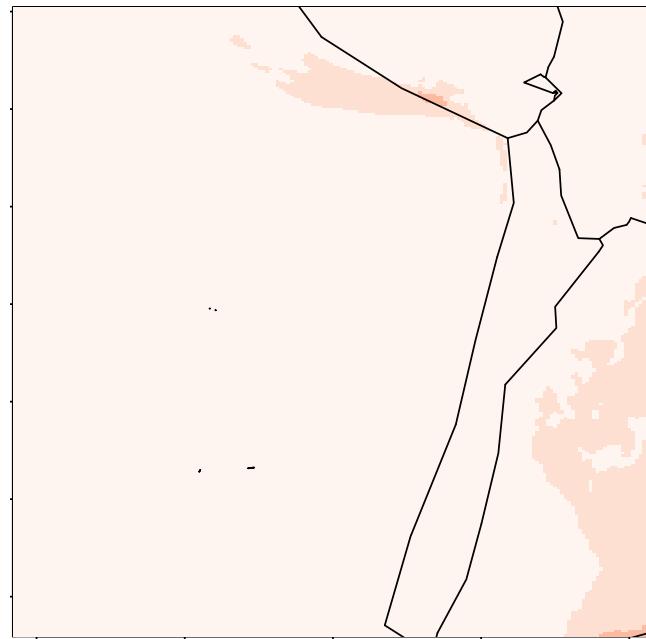
Number concentration (cm^{-3})



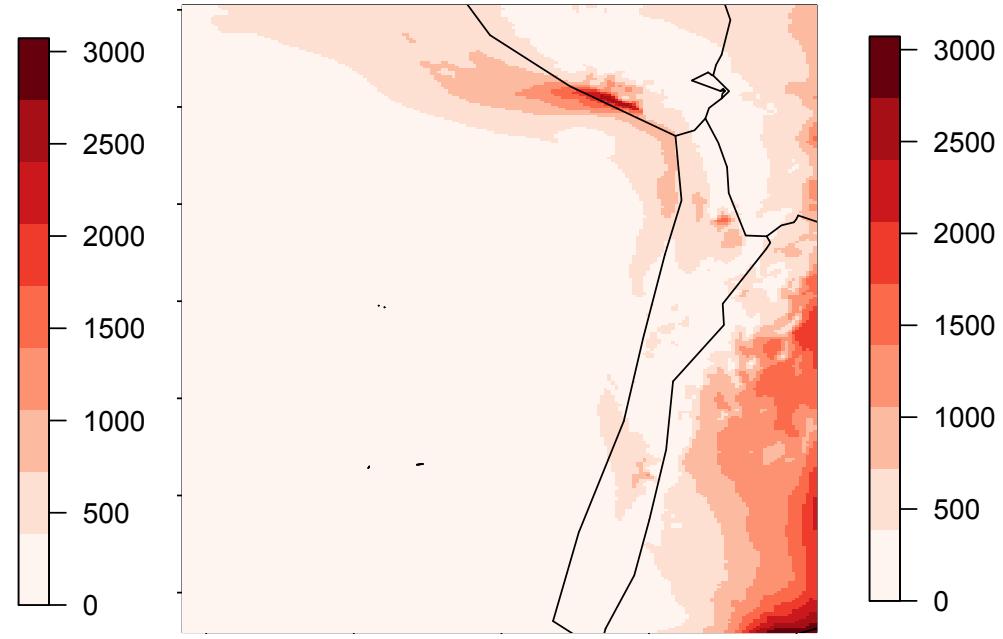
Number concentration enhancement by LPS >> central Chile

Anthropogenic impact: cloud-level CCN (cm^{-3})

0.2% supersaturation



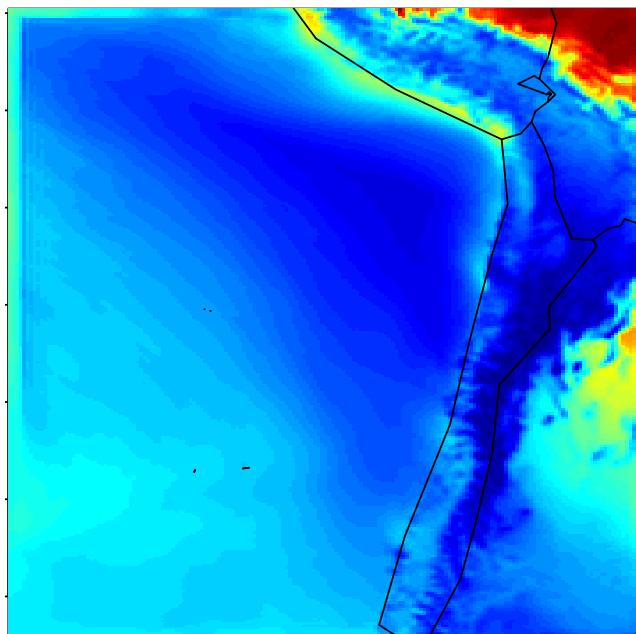
0.5% supersaturation



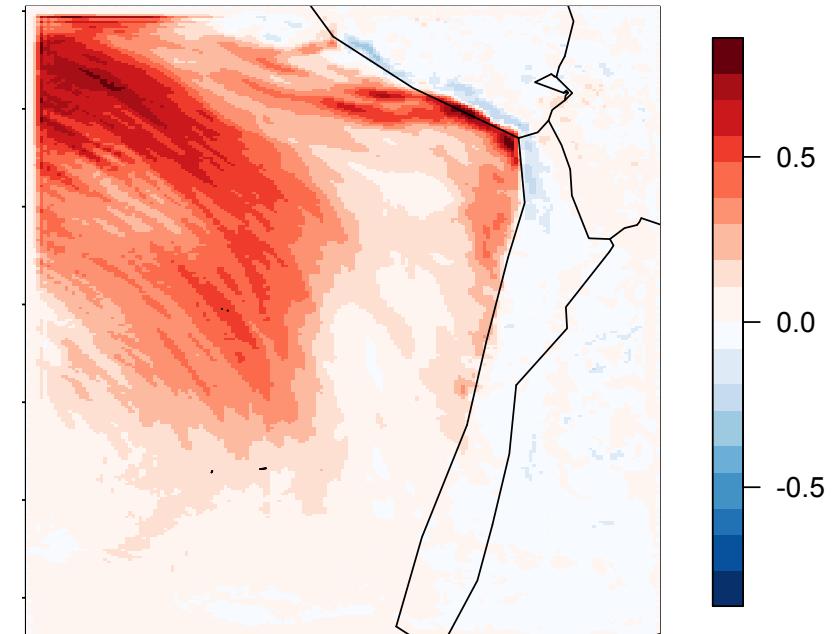
Order of magnitude underestimate in local coastal effect in WRF-Chem

Water vapor (g/kg), cloud level

Base case



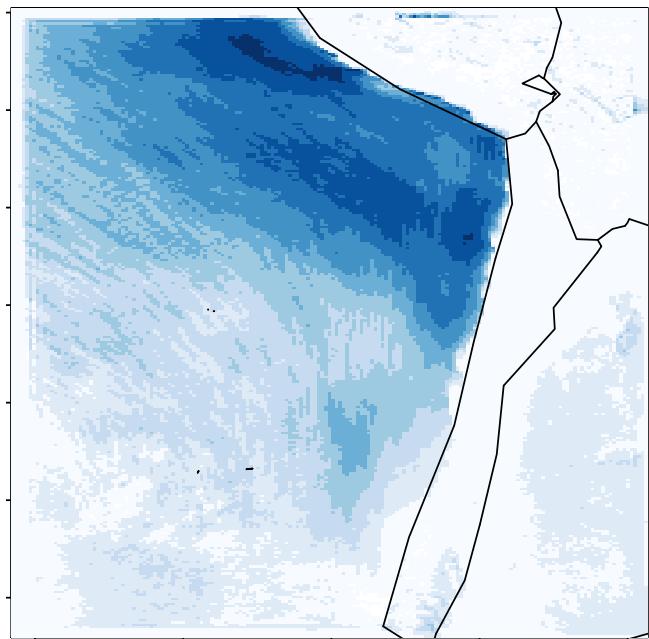
Anthropogenic impact



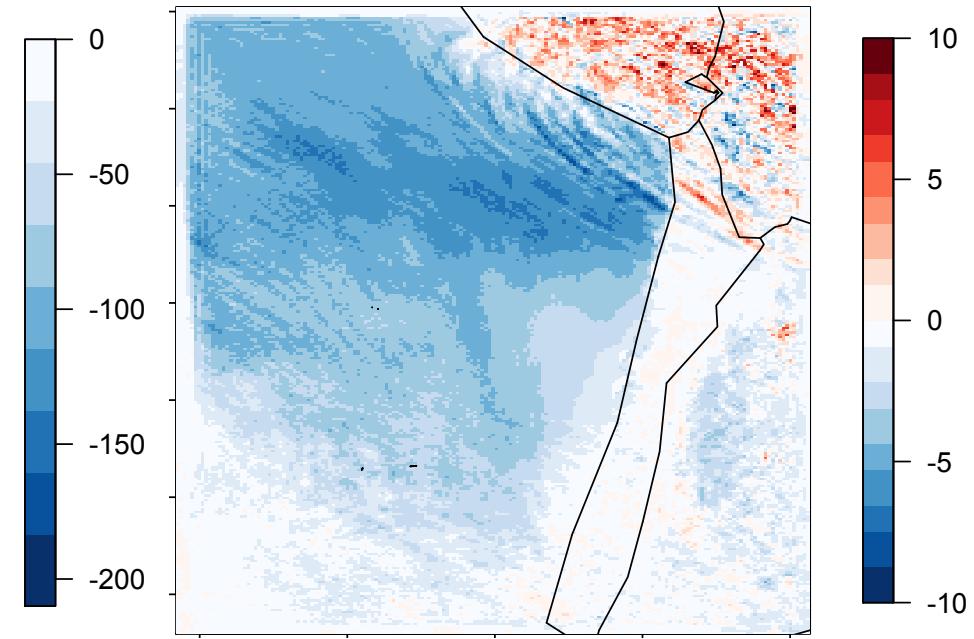
20° S changes due to LPS (coastal) and central Chile (offshore)

Radiative forcing (W/m^2)

Downwelling SW @ SFC

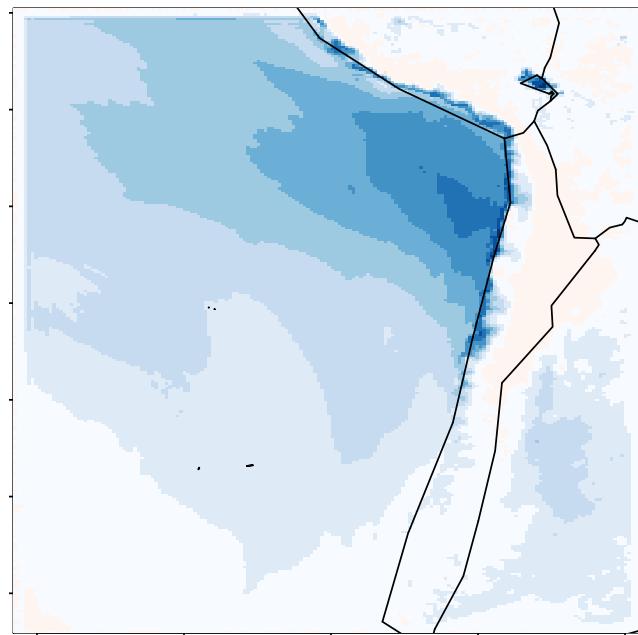


Upwelling LW @ TOA

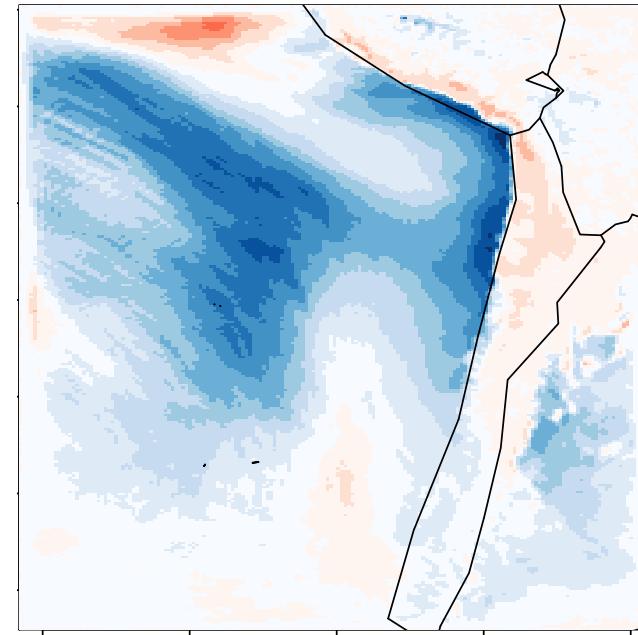


Anthropogenic impact on Temperature (K)

Surface



Cloud-level



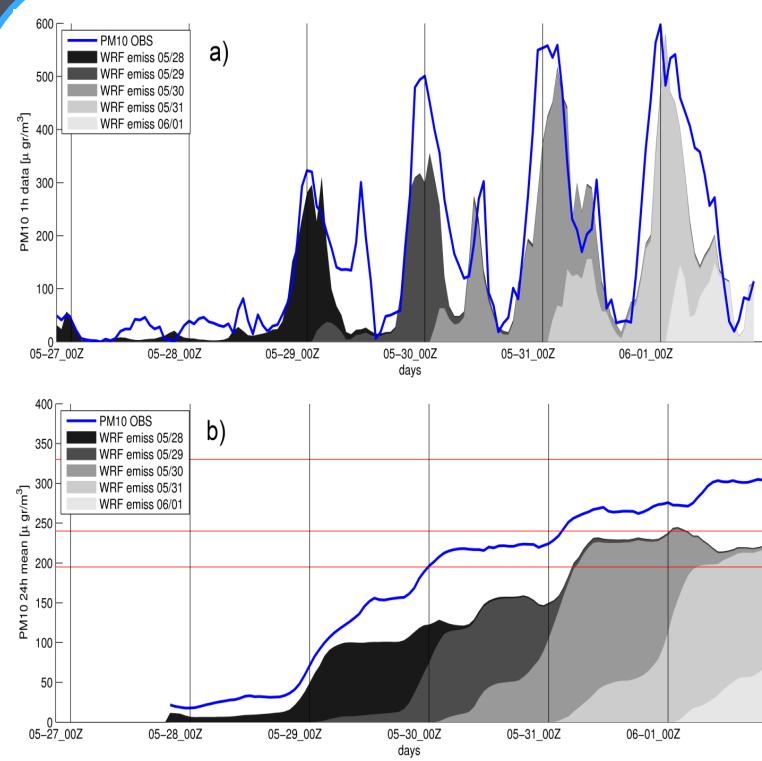
Coastal surface cooling by brighter clouds,
offshore cloud-level cooling by longer cloud lifetime

Conclusions

- Emissions & chemical transport: cautious but confident
 - ✓ Most species of interest simulated as well or better than regulatory modeling standards, prior airborne field campaigns
 - ✓ aerosols offshore @ 20S: central Chile + northern large point sources
 - ✓ natural emissions are key to improvement: DMS & biomass burning essential, sea salt algorithms and OC too high
- Modeling anthropogenic aerosol influence on clouds & climate: large but uncertain
 - ✓ Very strong anthropogenic indirect radiative forcing
 - ✓ *Average >0.5 °C surface & in-cloud cooling*
 - ✓ CCN diverges from observations, insensitive to anthropogenic emissions

Ongoing extensions

- Santiago daily operational WRF-Chem PM₁₀/PM_{2.5} forecast
- Adjoint WRF-Chem aerosol direct/indirect/semi-direct effects
- Effects of Santiago's development on regional climate and SEP Sc
 - land use/density/transportation
 - vehicle/energy technology



Next steps

- 1 km simulations on new UI supercomputer to resolve clouds & changes in Santiago's urban form
- Ocean-atmosphere-aerosol coupling WRF-Chem with ROMS
 - effects of aerosols on SEP SSTs, ENSO
 - aerosol vs. ocean contributions to Sc variability
- Adjoint applications
 - **emissions sectors & aerosol composition impacting 20° s clouds**
 - **SO₂ emissions inversion:** an initial test for 4DVAR
 - constraining box/column simulations with REx observations to **isolate process errors** in aerosol-microphysics interactions

Larger Questions

- How much skill must >LES models demonstrate to be useful?
 - primary: aerosols (concentrations, AOD, CCN, CDN, CN)
 - secondary: clouds (LWP, drizzle/rain rate, τ_{CLOUD} , brightness temperature)
 - tertiary: climate (SST, ocean/atmosphere energy balance, radiative forcing)
- Beyond “lots of CCN” geoengineering, other scenarios of interest?
- VOCALS “summary for policymakers” on atmosphere-ocean-cloud interactions
 - confidence in processes, answers to hypotheses
 - community metrics for model performance: criteria & target
 - specific process improvements needed
 - REx findings, long-term reanalyses, projections

Acknowledgements

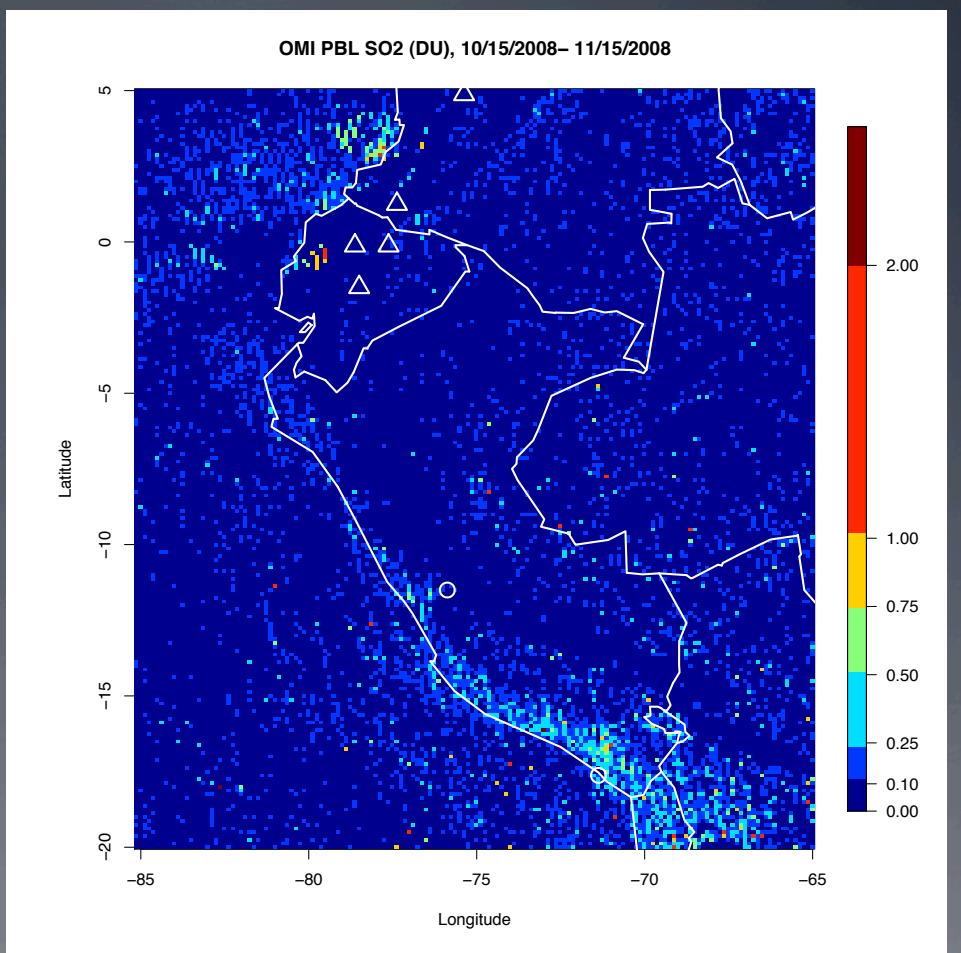
- VOCALS science and observational teams
- Data & Modeling
 - C. Wiedinmeyer, NCAR
 - L. Emmons, NCAR
 - J. Fast, PNNL

Funding



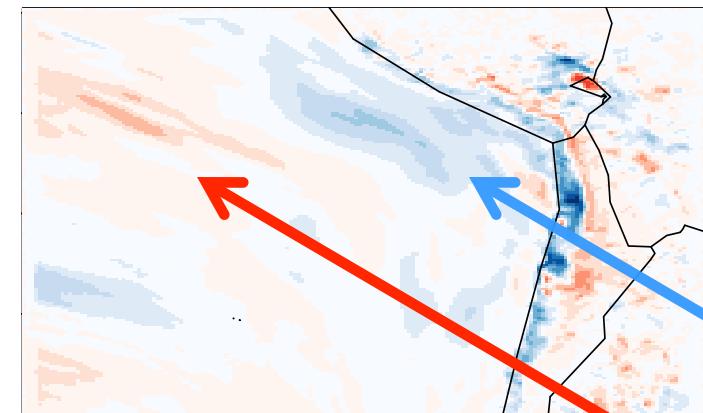
Building on... VOCA Emissions Inventory

- Anthropogenic
 - EDGAR 3.2 FastTrack 2000 + Bond *et al.* (2004) BC/OC @ LandScan
 - CONAMA/CODELCO 2008 for Chile
 - 1,400+ point sources
 - Municipal-level residential, industrial, mobile area sources
- Volcanoes & Peruvian smelters from OMI SO₂ during VOCALS REx inverted as in Cairn *et al.* (2007)

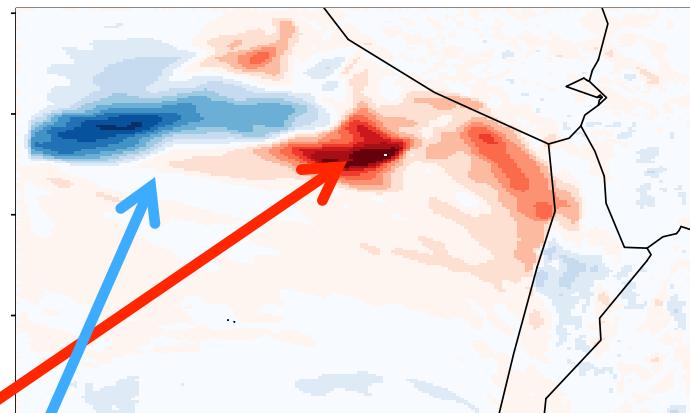


Episodic temperature (K): 10/15/08 only

Surface



Cloud top



Point source sulfate -> brighter coastal clouds, surface cooling

Less drizzle, inhibited convection -> offshore surface warming