Revisiting the organic aerosol in southeast US

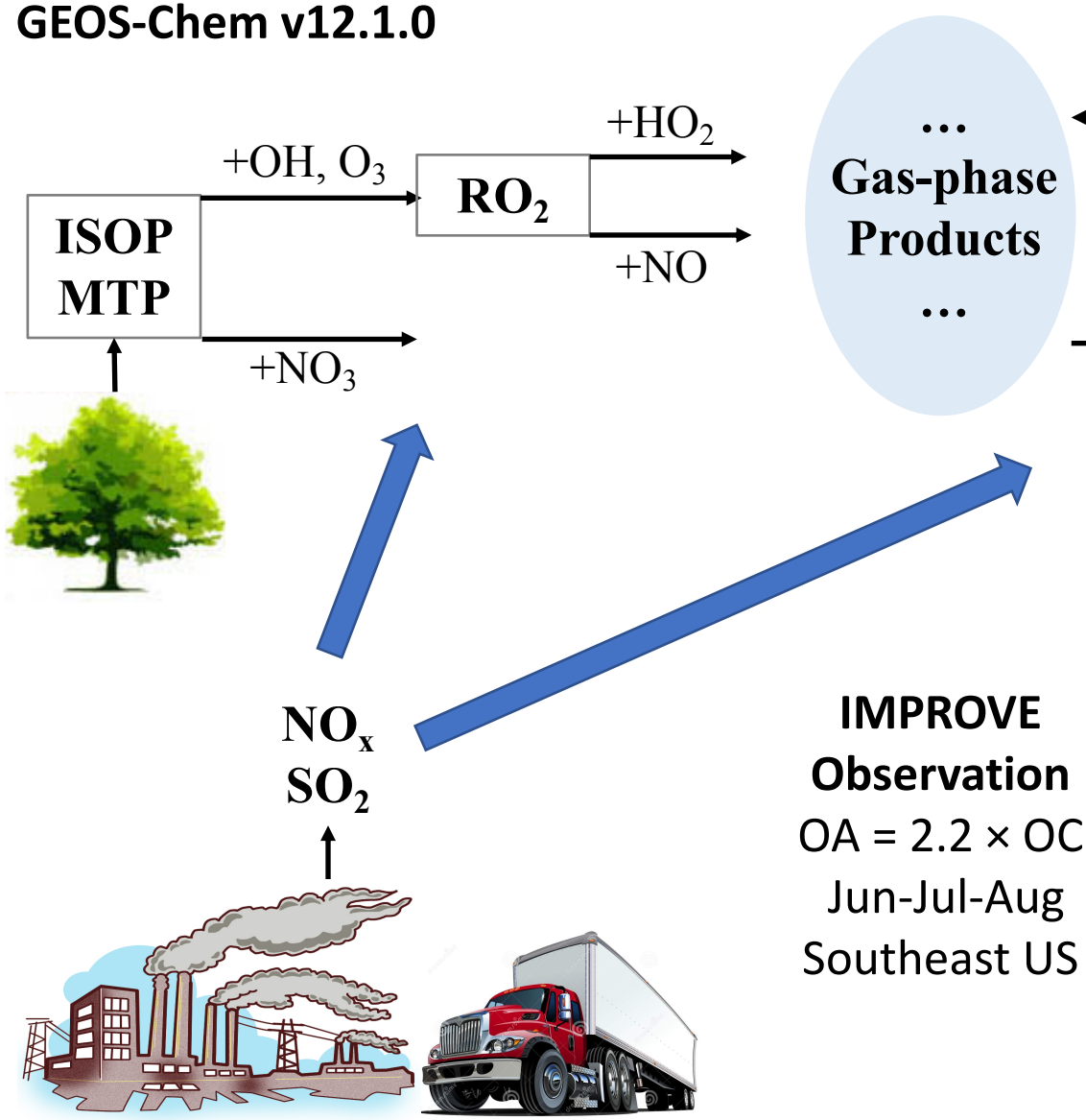
Yiqi Zheng, Jingqiu Mao
University of Alaska Fairbanks

May 9th, 2019
The 9th International GEOS-Chem Meeting
Harvard University, Cambridge, MA

Coauthors: Joel Thornton (U. Washington), Nga Lee Ng (Georgia Tech), Erin McDuffie (Dalhousie U.), Jose Jimenez (CU Boulder), Eloise Marais (U. Leicester)
Funding support: NOAA

Anthropogenic influence on BSOA formation

GEOS-Chem v12.1.0



Volatility Basis Set (VBS) *[Pye et al. 2010]*

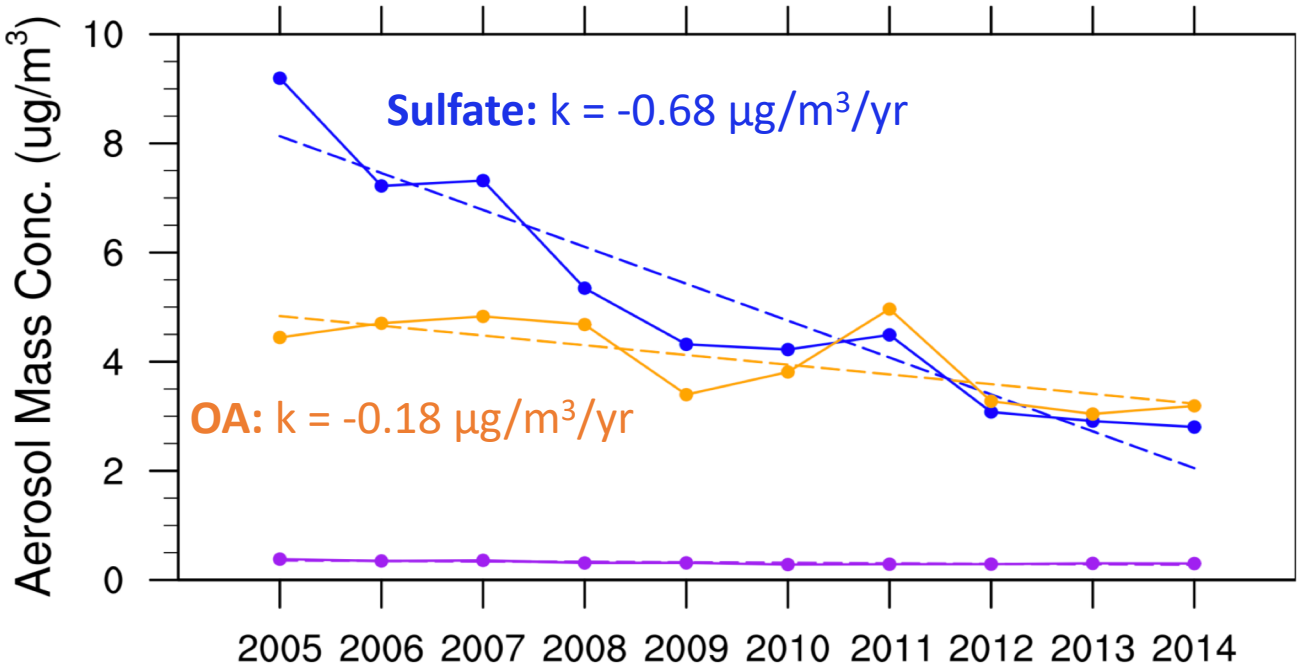
6 ISOP-SOA species *[Marais et al. 2016]*

IMPROVE Observation

OA = 2.2 × OC

Jun-Jul-Aug

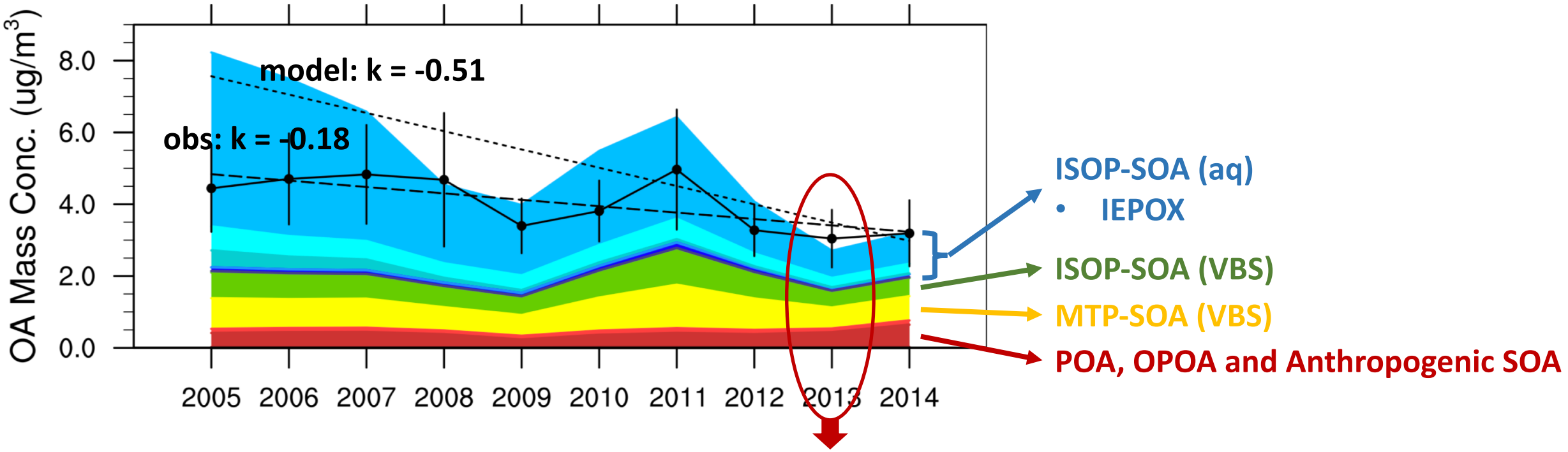
Southeast US



Model overestimates the 10-yr OA trend and the ISOP contribution

GEOS-Chem v12.1.0, 2°×2.5°, SEUS, JJA-average

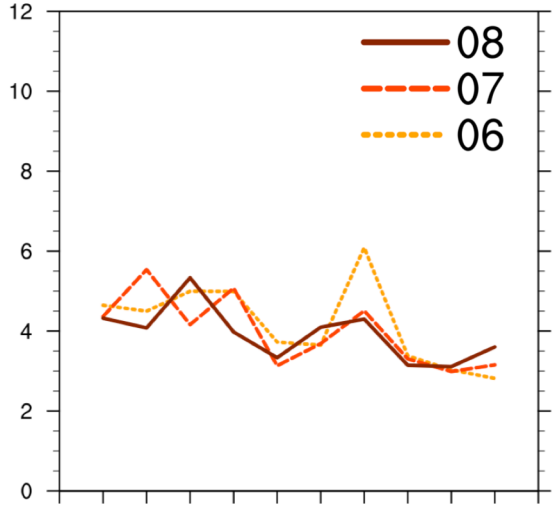
Units of trend (k): μg/m^3/year



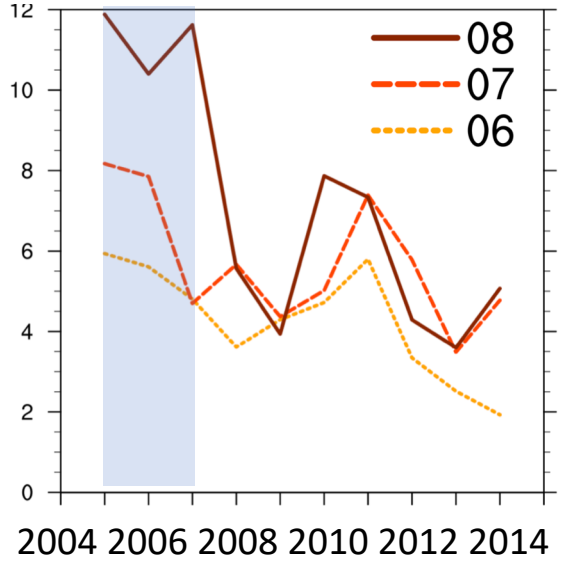
Fraction in total OA (%)	GC v12.1.0	Zhang et al., 2018	Xu et al., 2018
ISOP-SOA	58	13	21-36
MTP-SOA	22	42	> 19-34

Large month-to-month variation in 2005-2007

IMPROVE OA [μg/m³]



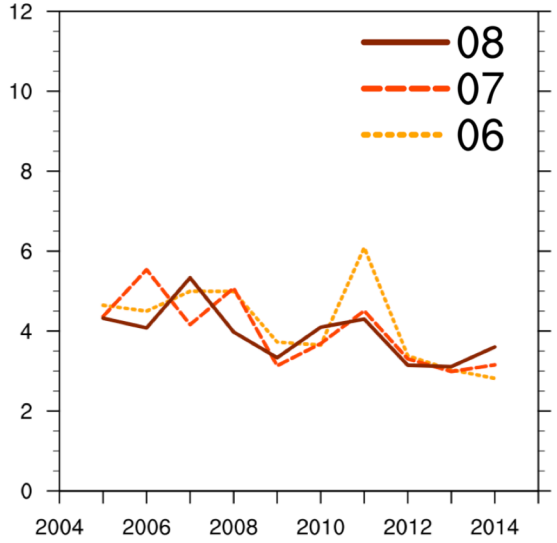
Model OA (default) [μg/m³]



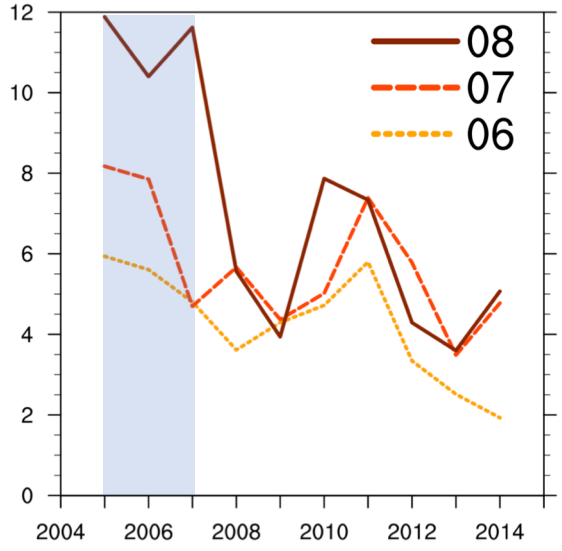
Large J-J-A variation in 2005-2007 dominated by IEPOX-SOA

Large month-to-month variation due to aerosol acidity [H^+]

IMPROVE OA [µg/m³]

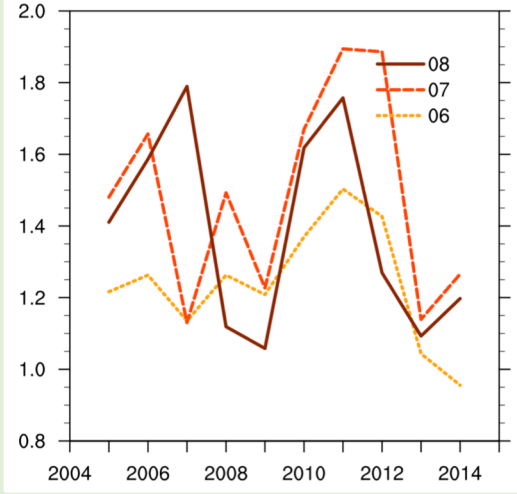


Model OA (default) [µg/m³]

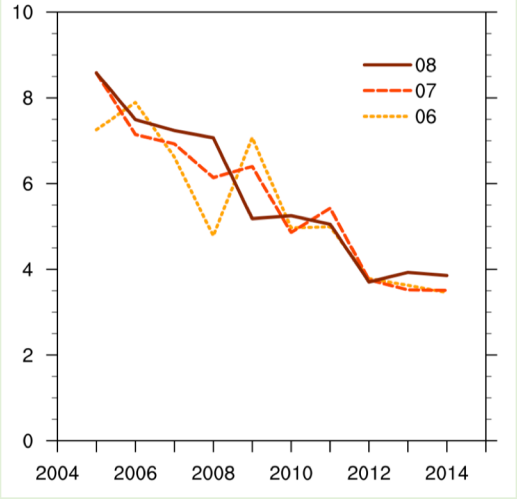


Model

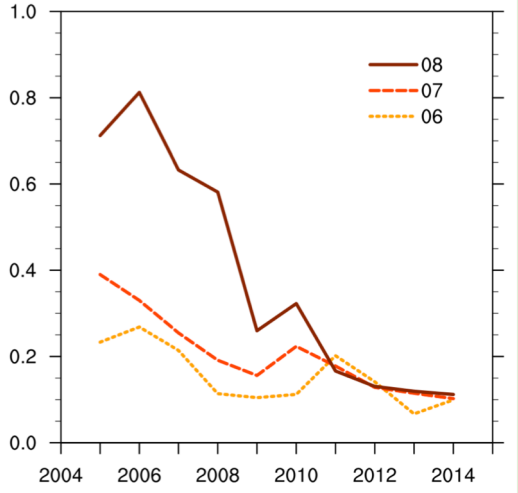
$ISOP_{emis}$ [mg/m²/hr]



Model SO_4 [µg/m³]

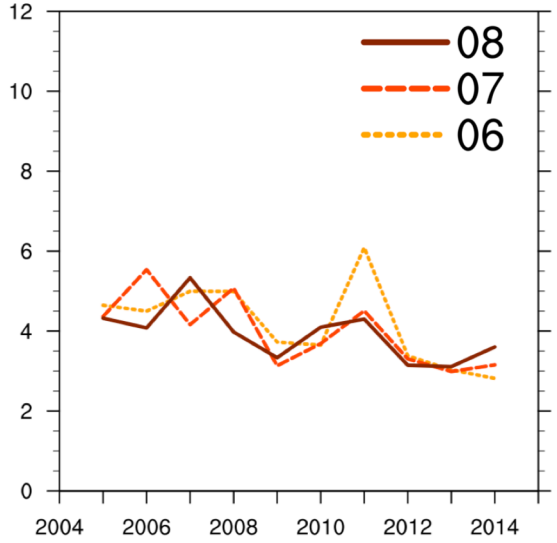


Model H^+ [mol/L]

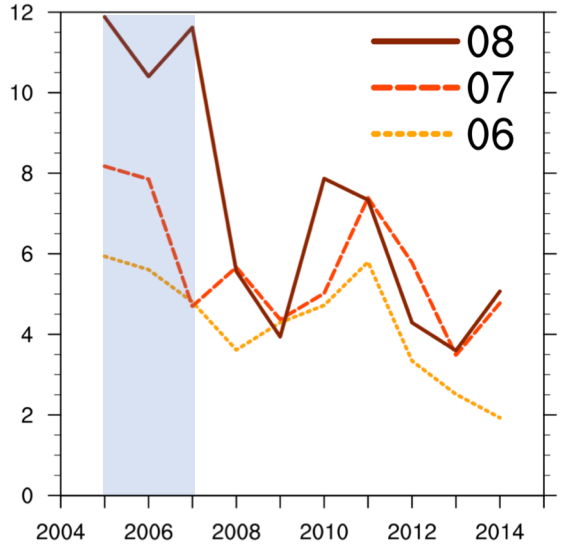


High [H^+] in Aug 2005-2007 due to insufficient NH_3

IMPROVE OA [µg/m³]

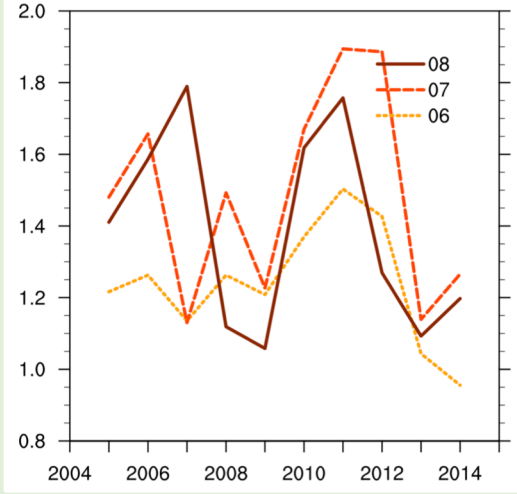


Model OA (default) [µg/m³]

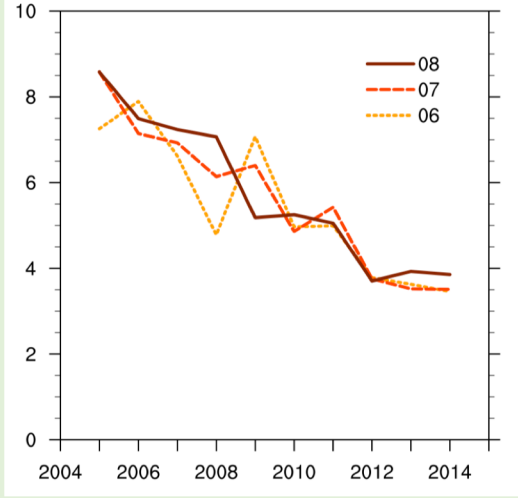


Model

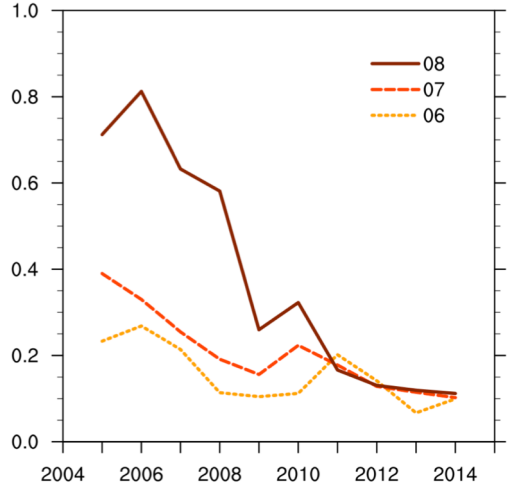
$ISOP_{emis}$ [mg/m²/hr]



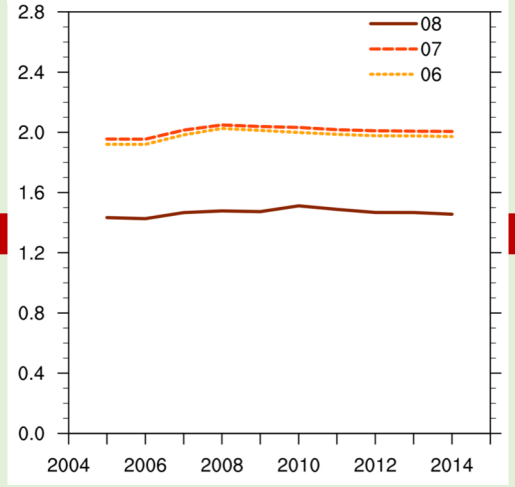
Model SO_4 [µg/m³]



Model H^+ [mol/L]

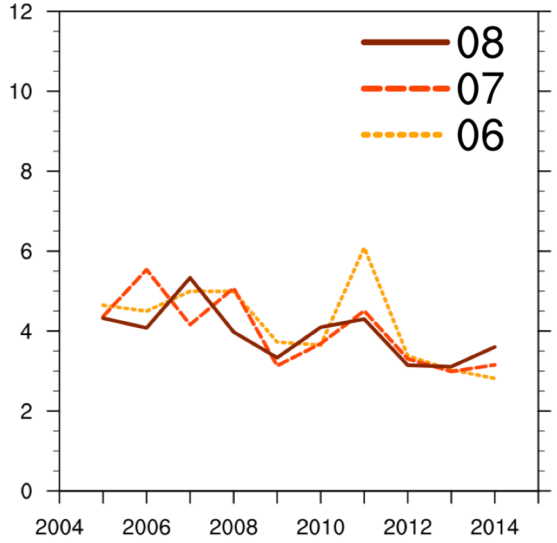


NH_{3_emis} [x10^8 kg]

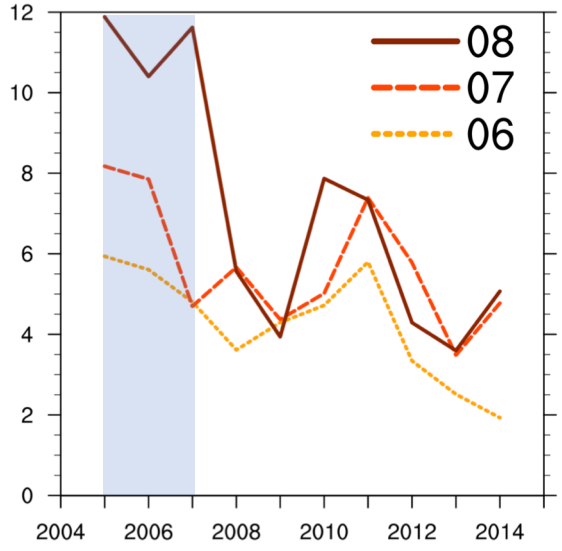


High $[H^+]$ in Aug 2005-2007 due to insufficient NH_3

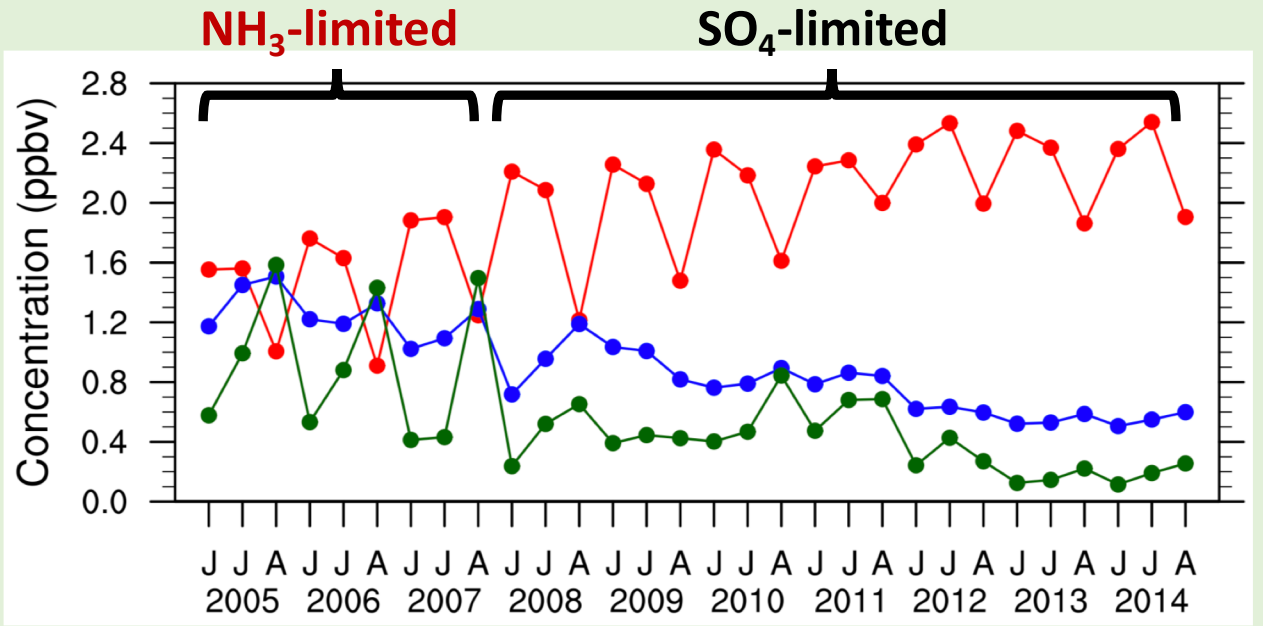
IMPROVE OA [μg/m³]



Model OA (default) [μg/m³]



Model



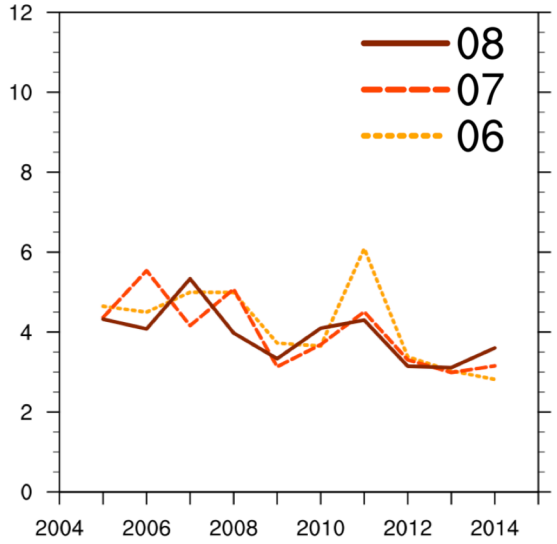
IEPOX-SOA vs NH_3

2005-2007 r^2 = 0.87 (-)

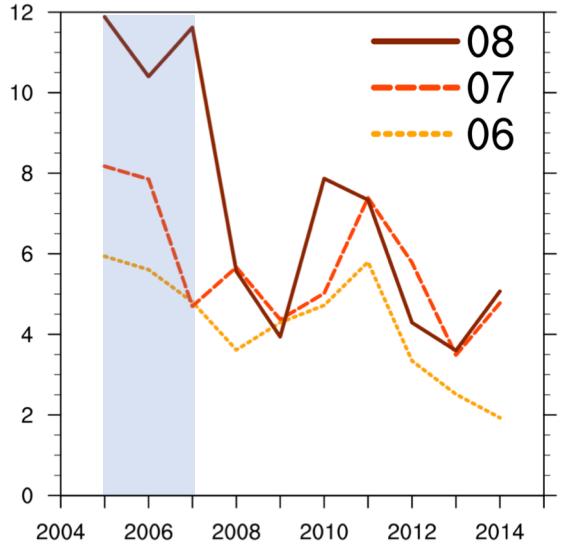
2008-2014 r^2 = 0.26 (-)

Large month-to-month variation in 2005-2007

IMPROVE OA [μg/m³]



Model OA (default) [μg/m³]



High aerosol acidity [H^+]
(in an NH_3-limited regime)

↓

High IEPOX-SOA

←

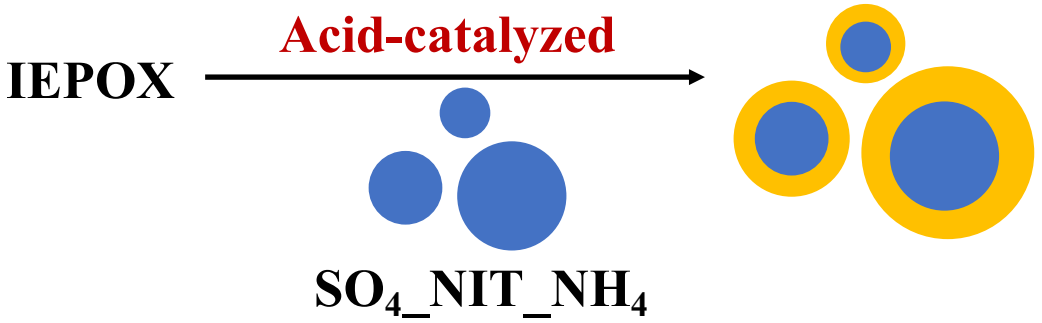
Why the model-obs. difference?

- IEPOX-SOA too sensitive to [H^+]
- [H^+] too high
- Overestimate IEPOX-SOA

Model Development

- Coating effect for IEPOX uptake
- Fixed [H^+]
- Update MTP-chemistry (future work)

Coating: reducing γ_{IEPOX} and γ_{IEPOX} sensitivity to $[H^+]$



Model – Default

γ_{IEPOX} = Function (H^+, …)

Rate = Function (γ_{IEPOX} , $A_{sulfate}$)

Model – Coating

Fitted based on *[Gaston et al., 2014].*

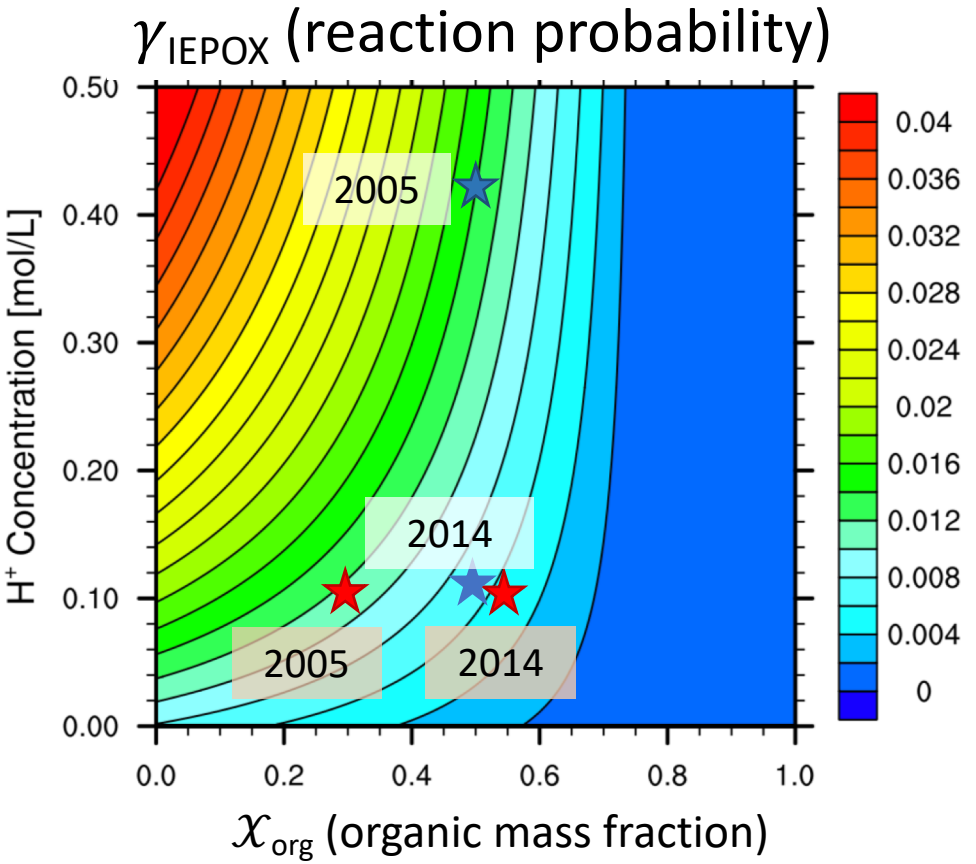
γ_{IEPOX} = Function (H^+, …)

γ_{new} = γ_{IEPOX} * (1 – 1.3 * χ_{org})

Rate = Function (γ_{new} , $A_{sulfate+org}$)

★ Modeled $[H^+]$ and χ_{org}

★ Observed χ_{org} and assuming $[H^+]$=0.1 mol/L (2013 SOAS campaign data)



Model development: coating effect

Model – Default

γ_{IEPOX} = Function (H^+, …)

Rate = Function (γ_{IEPOX} , $A_{sulfate}$)

Model – Coating

γ_{IEPOX} = Function (H^+, …)

γ_{new} = γ_{IEPOX} * (1 – 1.3 * χ_{org})

Rate = Function (γ_{new} , $A_{sulfate+org}$)

Model – Coating_fixedH

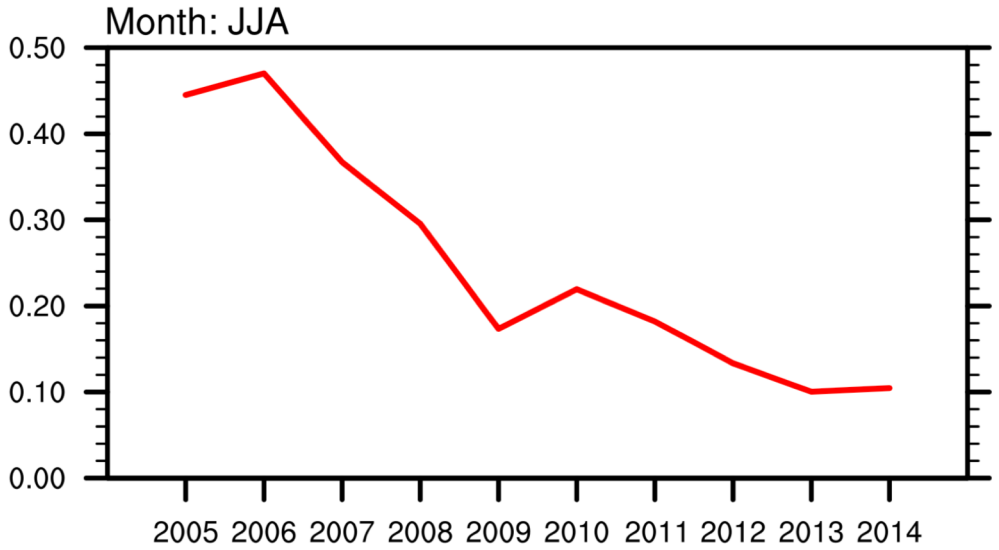
SOAS data

γ_{IEPOX} = Function (H^+=0.1 mol/L, …)

γ_{new} = γ_{IEPOX} * (1 – 1.3 * χ_{org})

Rate = Function (γ_{new} , $A_{sulfate+org}$)

Model H^+ [mol/L]



Trend of acidity

Current model: decrease in acidity

[Weber et al. 2016]: remain unchanged

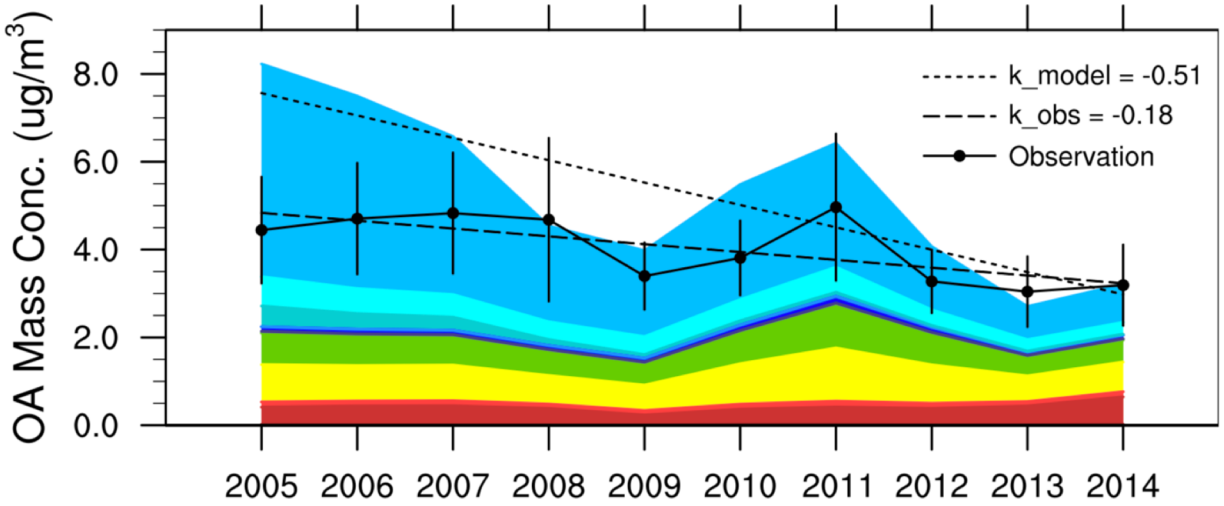
[Silvern et al. 2017]: increase in acidity

Adding coating effect improves model performance

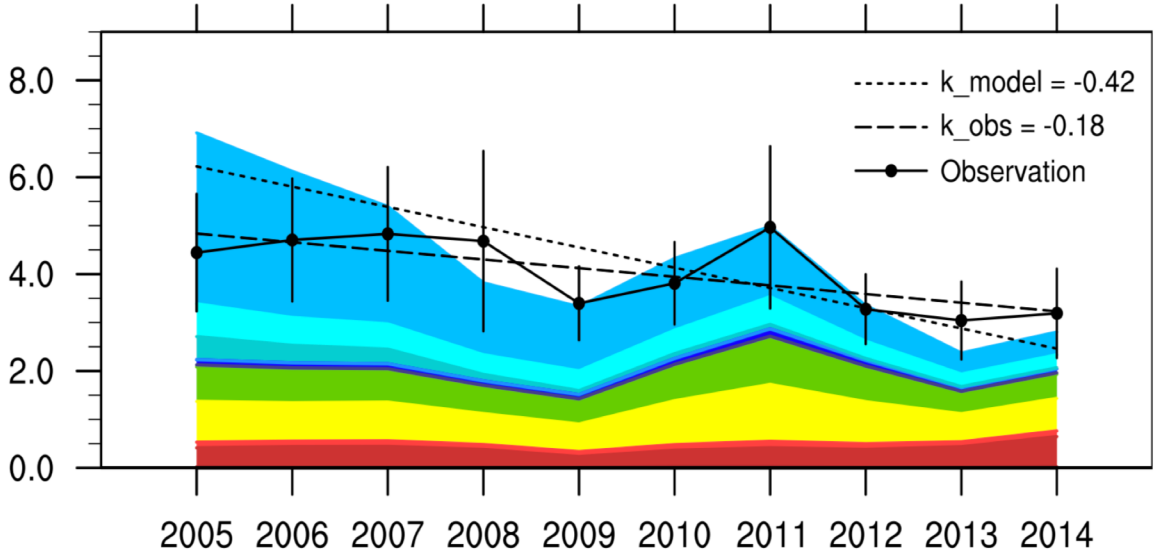
Month: JJA

Observation:
k = -0.18
Units: µg/m³/year

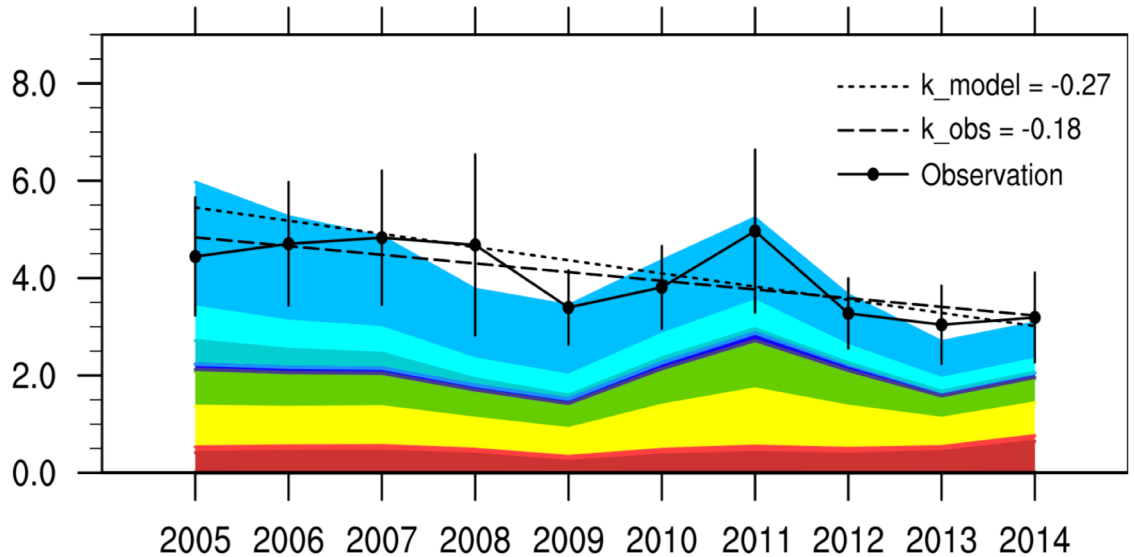
Default: k = -0.51



Coating: k = -0.42

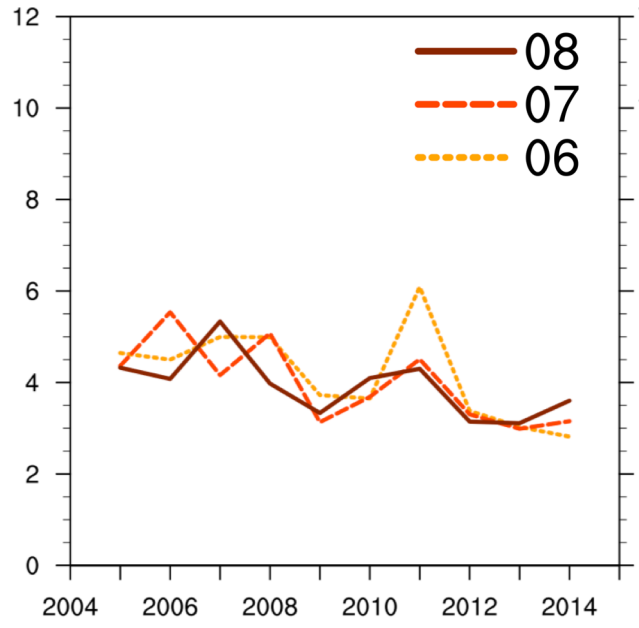


Coating_fixedH: k = -0.27

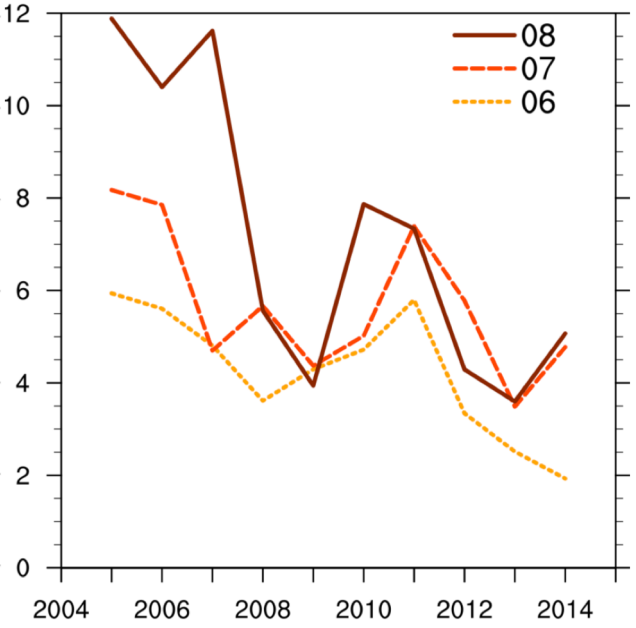


Adding coating effect improves model performance

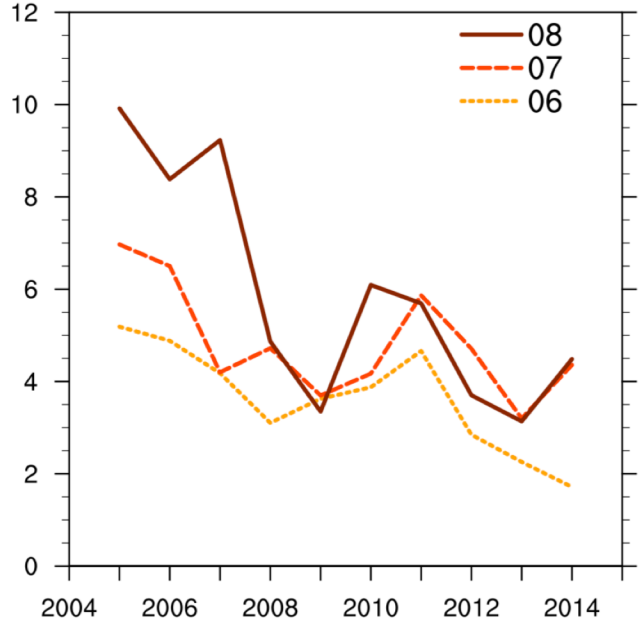
Observed OA



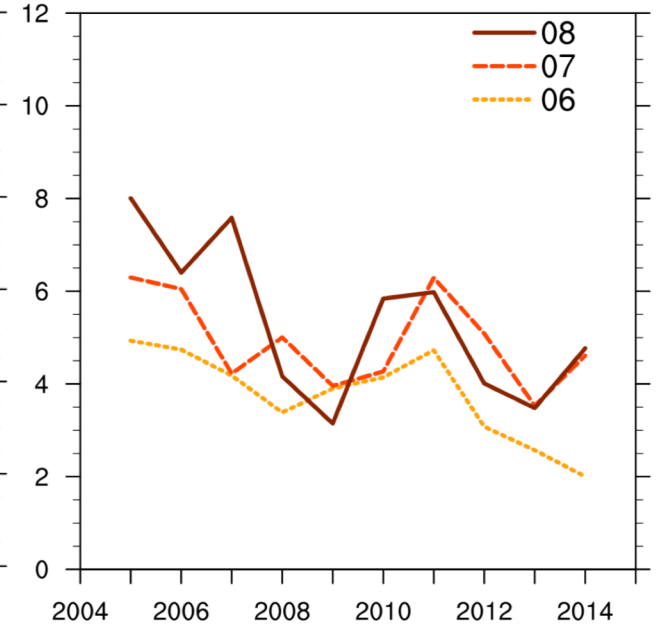
Model OA (default)



Model OA (coating)

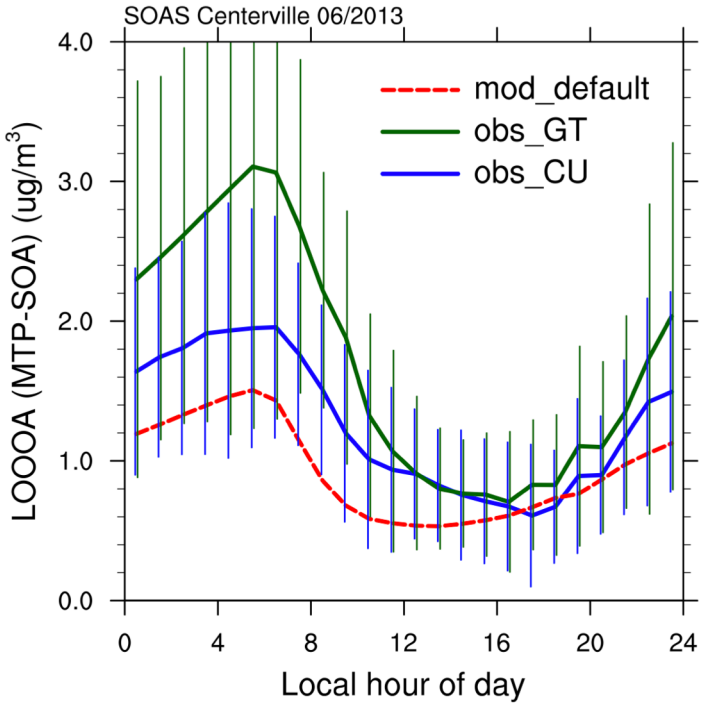


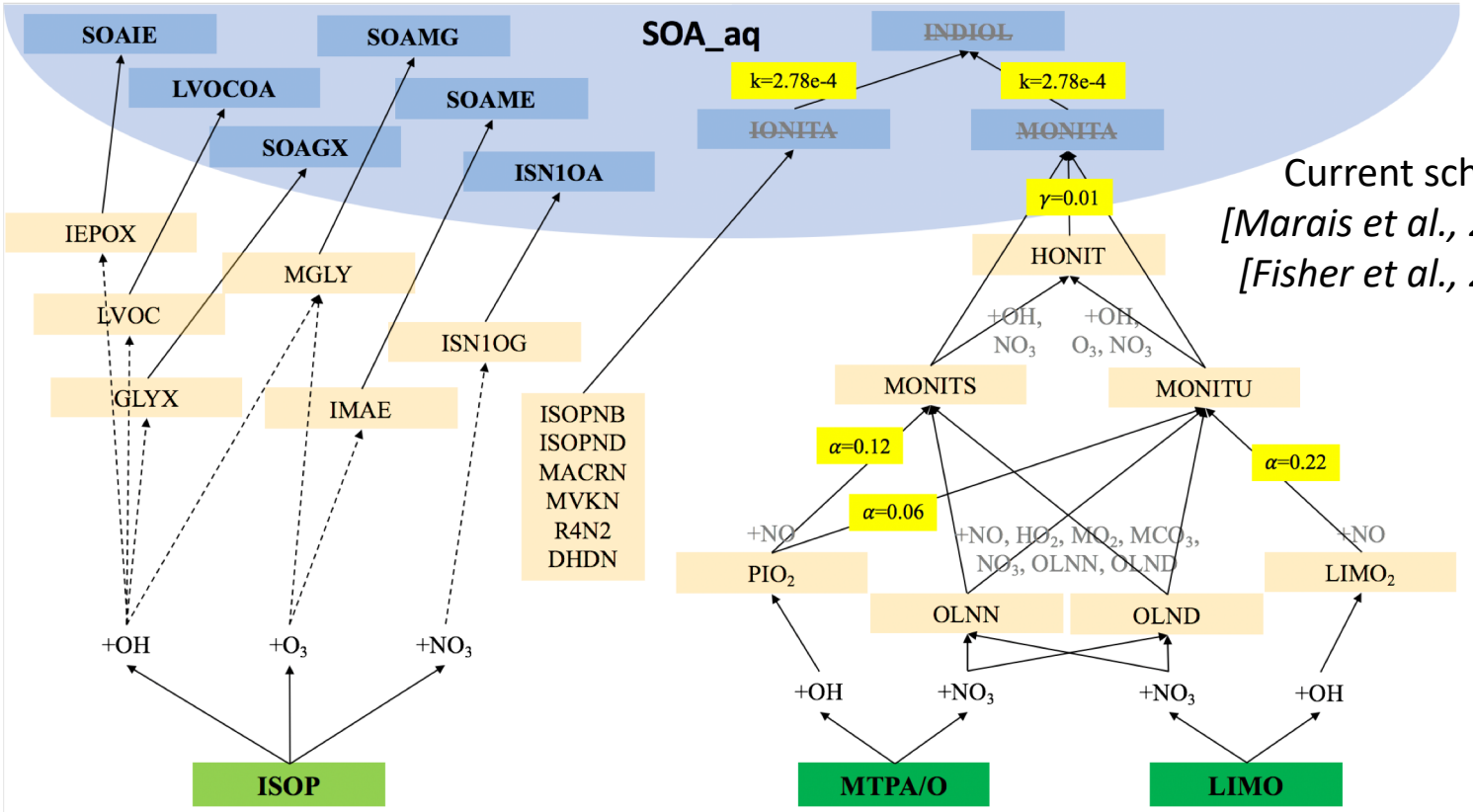
Model OA (coating_fixedH)



Summary & Future work

- Current model overestimates the 10-year OA trend (high IEPOX-SOA and acidity in Aug 2005-2007).
- Coating effect for IEPOX uptake & fixed [H^+] improves model performance.
- Underestimation of MTP-SOA?





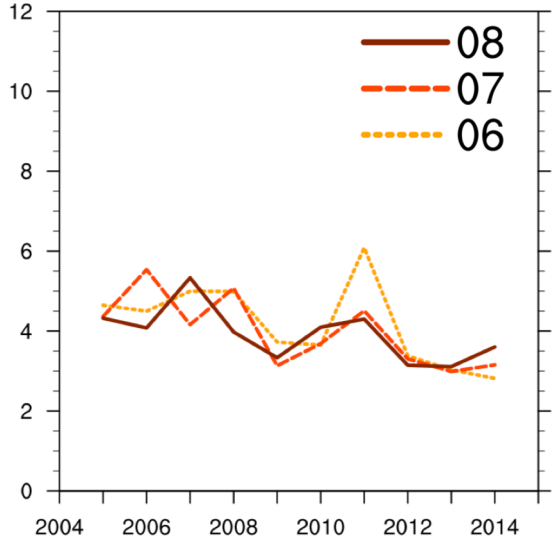
Current scheme:
[Marais et al., 2016]
[Fisher et al., 2016]

Issues to resolve: Loss of mass; lack of observational constraint; double counting with VBS.
MTP-chemistry will be updated in collaboration with Prof. Sally Ng's group.

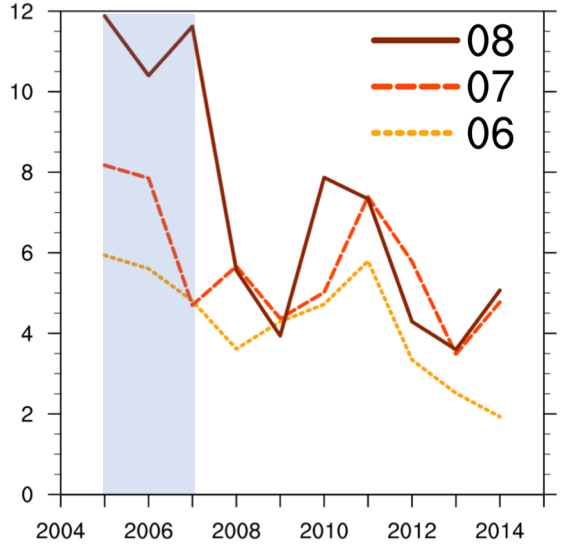
backup

Large month-to-month variation dominated by IEPOX-SOA

IMPROVE OA [μg/m³]

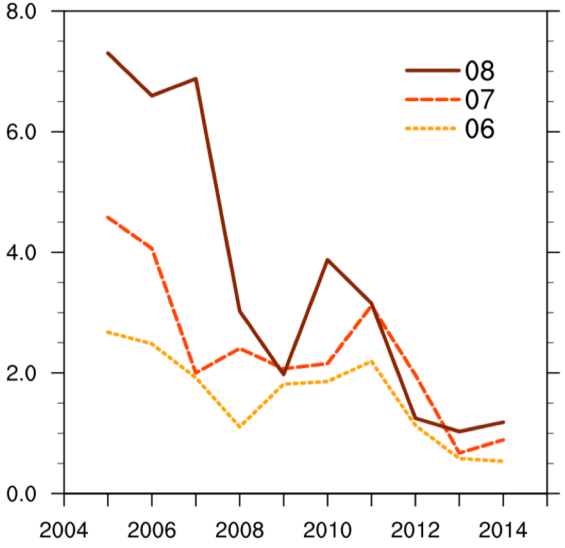


Model OA (default) [μg/m³]

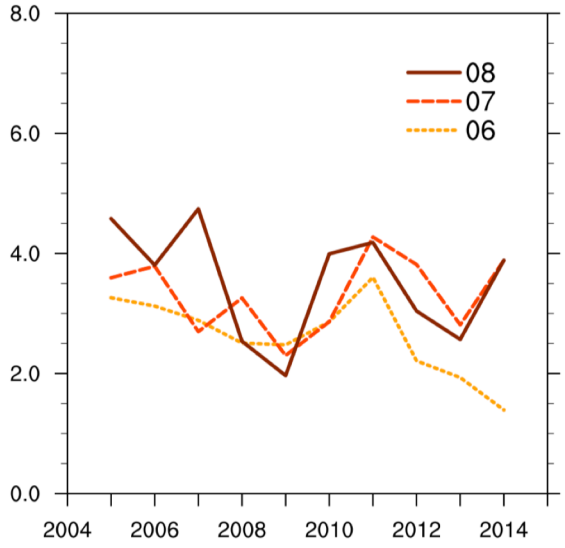


Dominated by IEPOX-SOA

IEPOX-SOA [μg/m³]

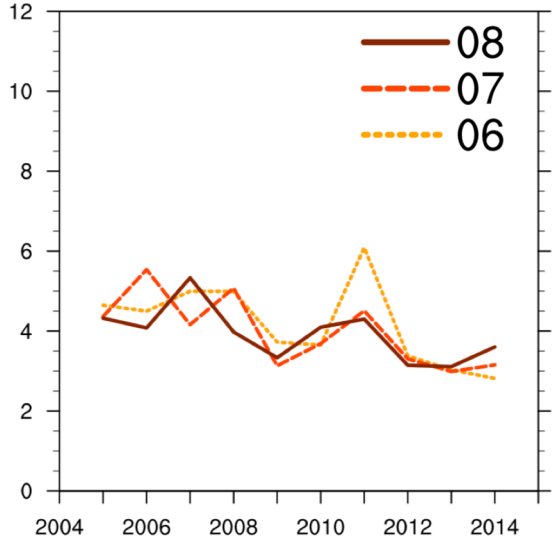


OA – IEPOX-SOA [μg/m³]

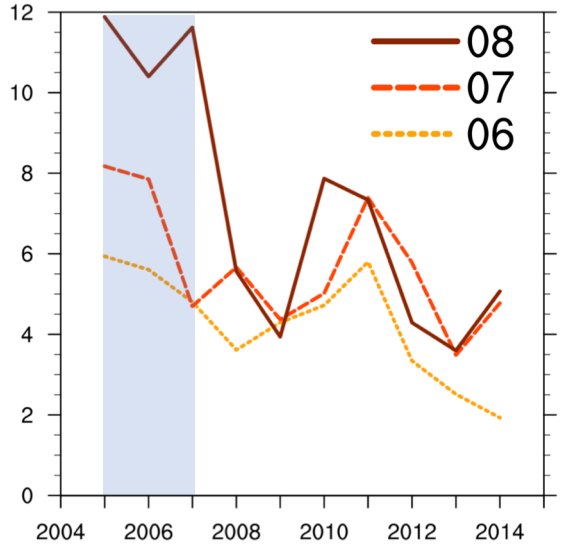


Observed OA show no big difference between Jun-Jul-Aug

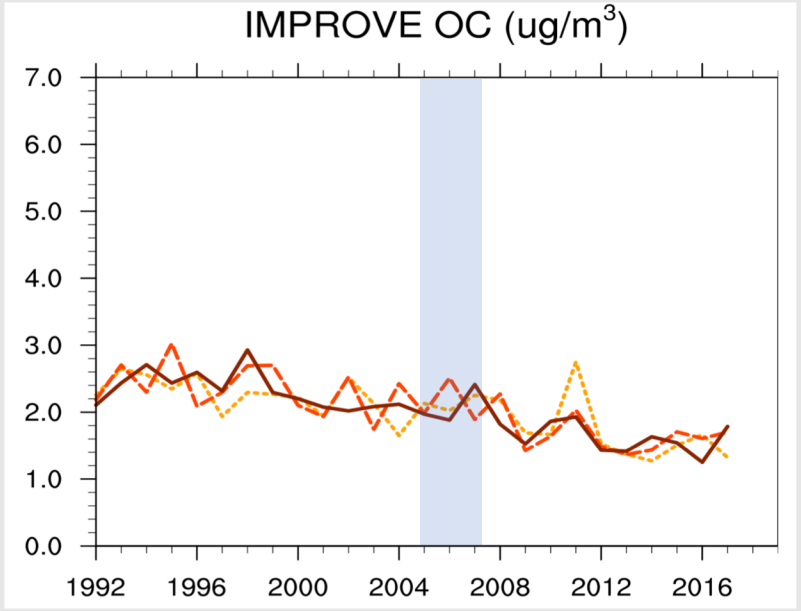
IMPROVE OA [μg/m³]

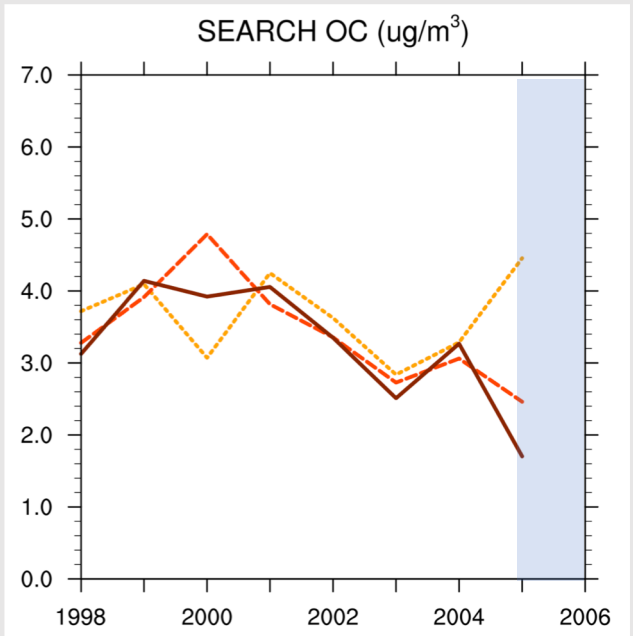


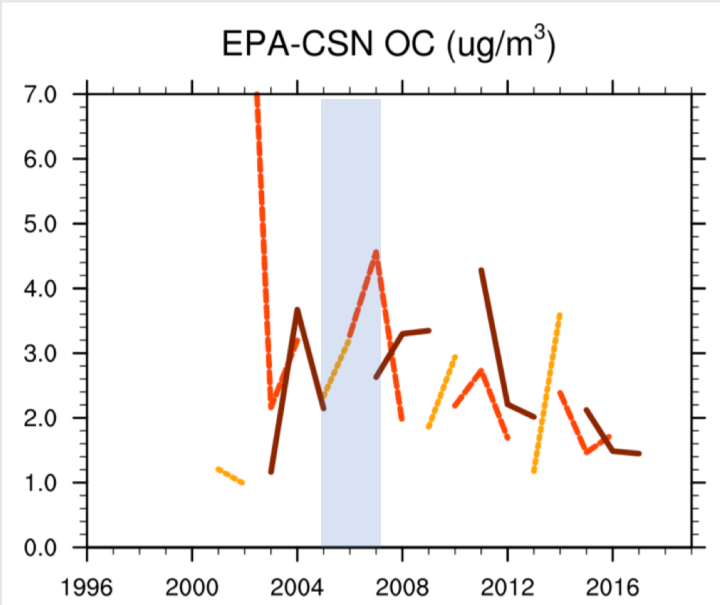
Model OA (default) [μg/m³]

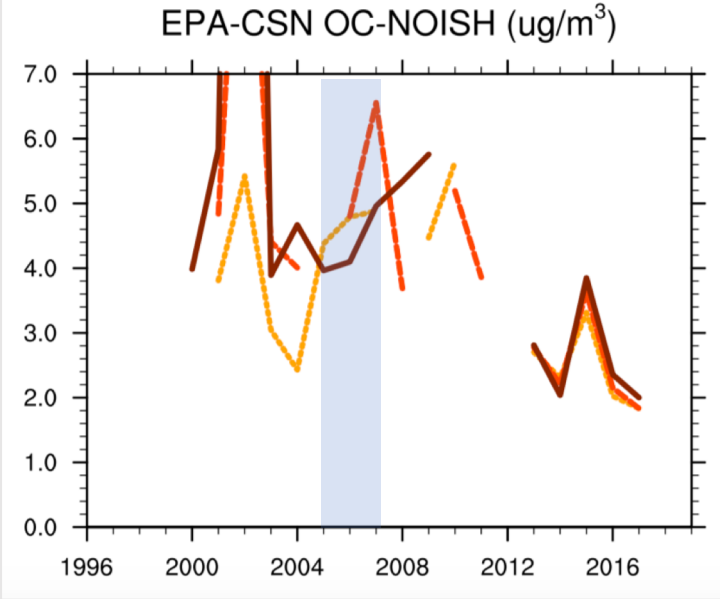


Observation

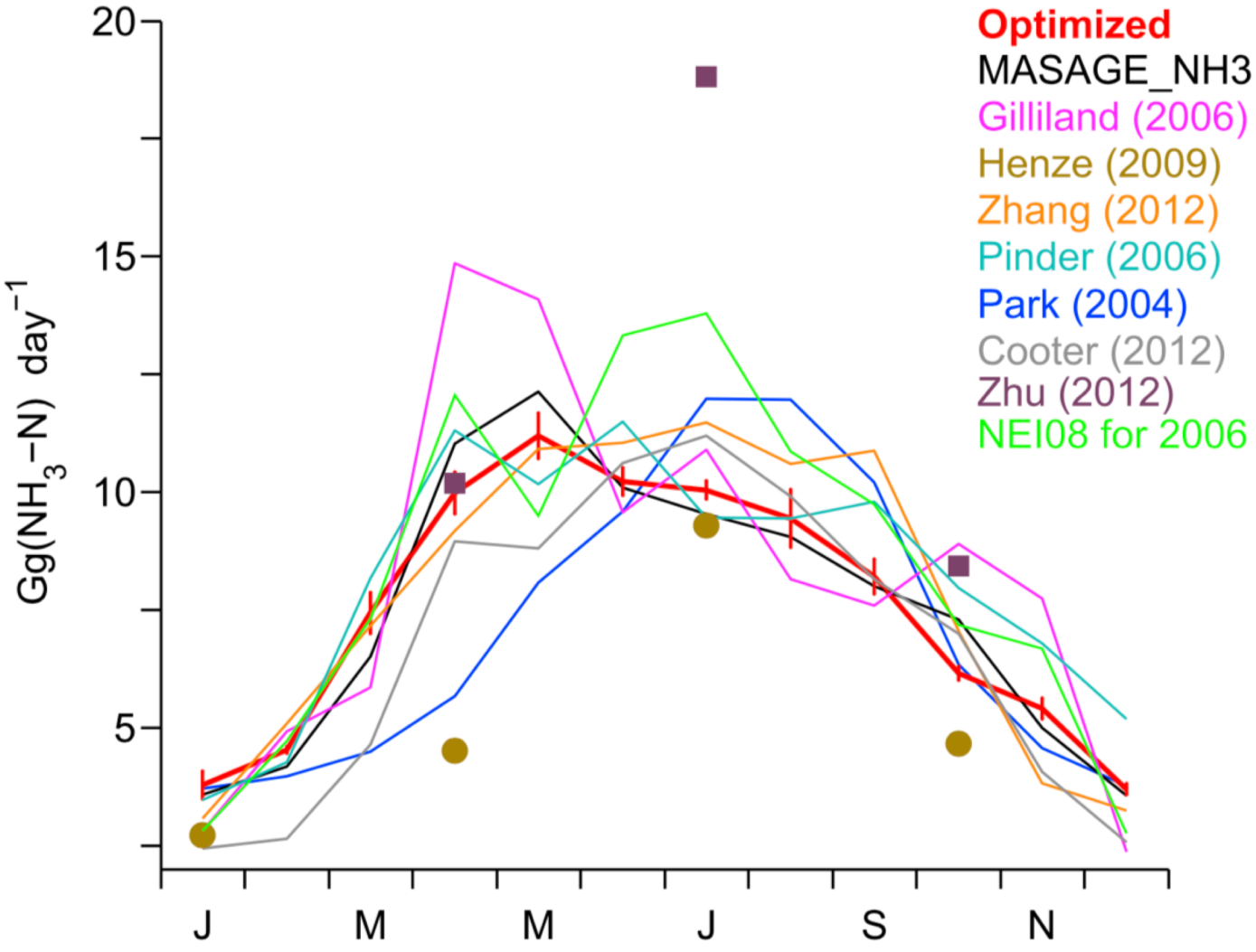








NH_3 emission

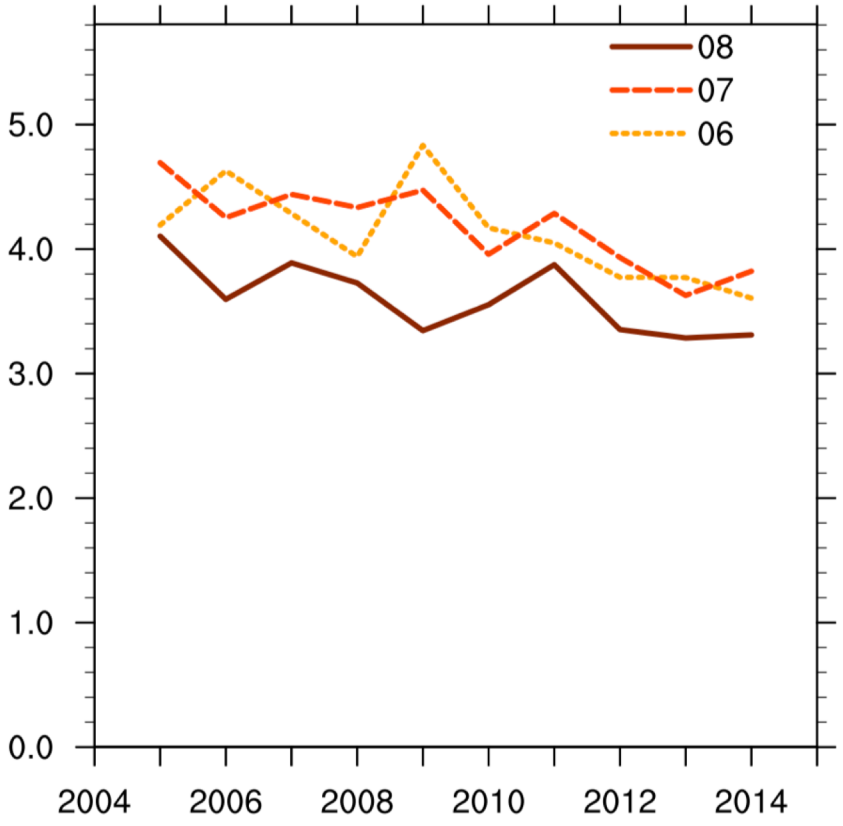


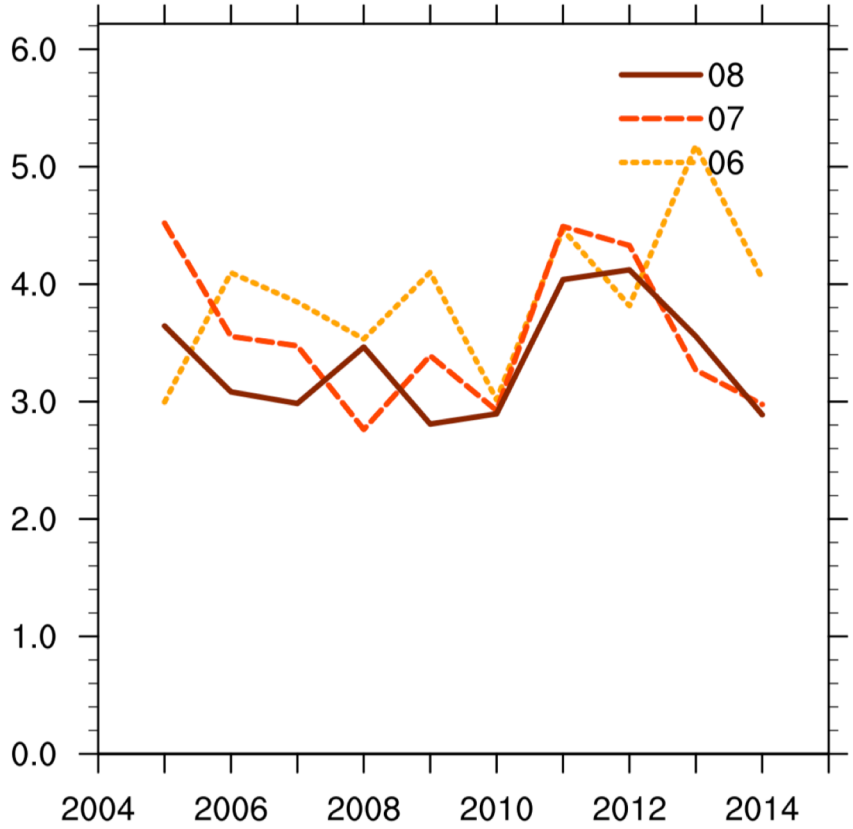
[Paulot et al., 2014]

Modeling vs. Observation NH_x

Modeling

$NH_x = NH_3 + NH_4$ (ppbv)



Observation (NADP)

NH_x WetDep Flux ($x10^{-5}$ kg/m^2)