Strong influence of deposition and vertical mixing on secondary organic aerosol concentrations in CMAQ and CAMx

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Introduction

➤ **Background:** (1) Secondary organic aerosol (SOA) affects global climate and ambient air quality. (2) Regional chemistry transport models (RCTM) are widely used to estimate SOA concentrations implemented with specific SOA modeling approach.

➤ **Gap:** (1) The two most commonly applied regulatory Air Quality Models (CMAQ: Community Multiscale Air Quality; CAMx: Comprehensive Air quality Model with extensions) predict very different organic aerosol (OA) concentrations with identical emissions, meteorological inputs and aerosol modeling approach (1.5-dimensional Volatility Basis Set[Koo et al., 2014]). (2) These existed differences suggest that transformation and fate pathways could be the underlying reasons.

➤ **Goal:** (1) Identifies processes that strongly affect the simulation of organic aerosols and quantifies sensitivity to process treatments

Methodology

➤ **Model simulation:** (1) Case study development.
➤ **Case study selection and demonstration**
➤ **Case study characterization:** (1) Concentration time series. (2) Vertical profiles.
➤ **Process Diagnostic:** (1) Mass analysis. (2) Deposition investigation.

Table 1. CMAQ and CAMx model configurations

Model option	CMAQ	CAMx
Model version	Version 5.0.1	Version 5.41
Horizontal resolution	36/12 km	36/12 km
Vertical layers	NZ = 14	NZ = 14
Horizontal advection	PPM	PPM
Vertical advection	PPM	Implicit
Horizontal diffusion	Spatially varying	Spatially varying
Vertical diffusion	Eddy diffusion with updated ACM2 option (Using PBL directly from WRFv3.4)	Eddy diffusion (Using Kz calculated from WRFv3.4 PBL)
Meteorology	WRFv3.4	WRFv3.4
Emissions	CSAPR and MEGANv2.1	CSAPR and MEGANv2.1
Dry deposition	Pleim and Ran (2011)	Zhang (2003)
Oxidant chemistry mechanism	CB05	CB05
Aerosol scheme	1.5D-VBS	1.5D-VBS

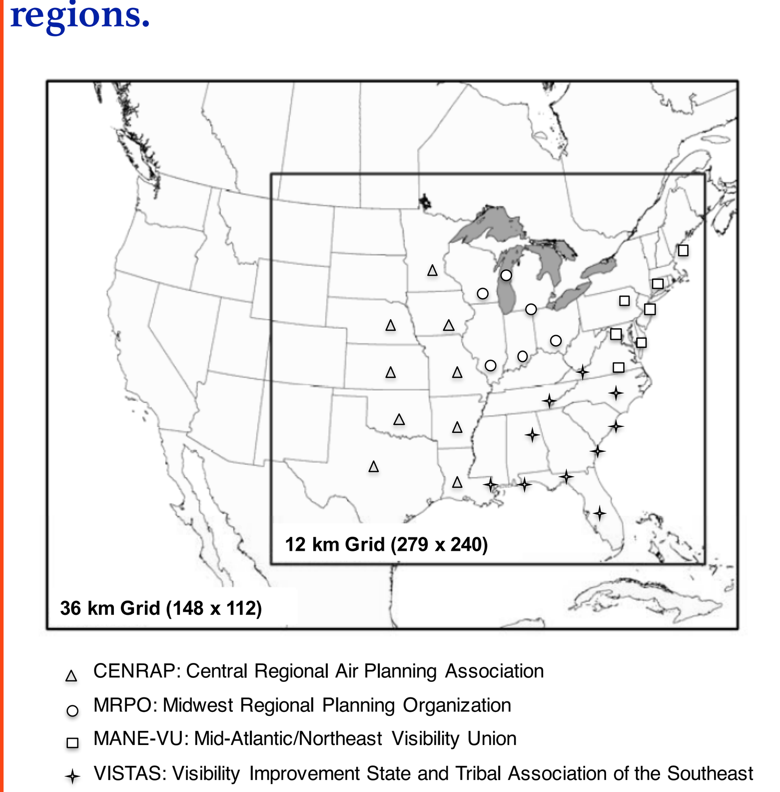


Figure 1. The 36 and 12km simulation domains. Four sets of symbols represent U.S. regional Planning Organization (RPO) regions.

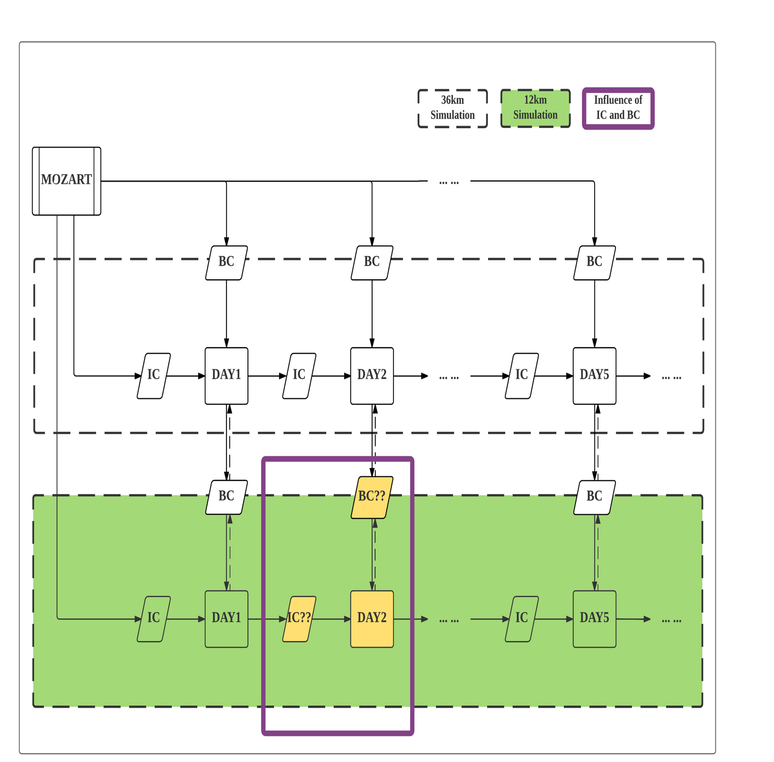
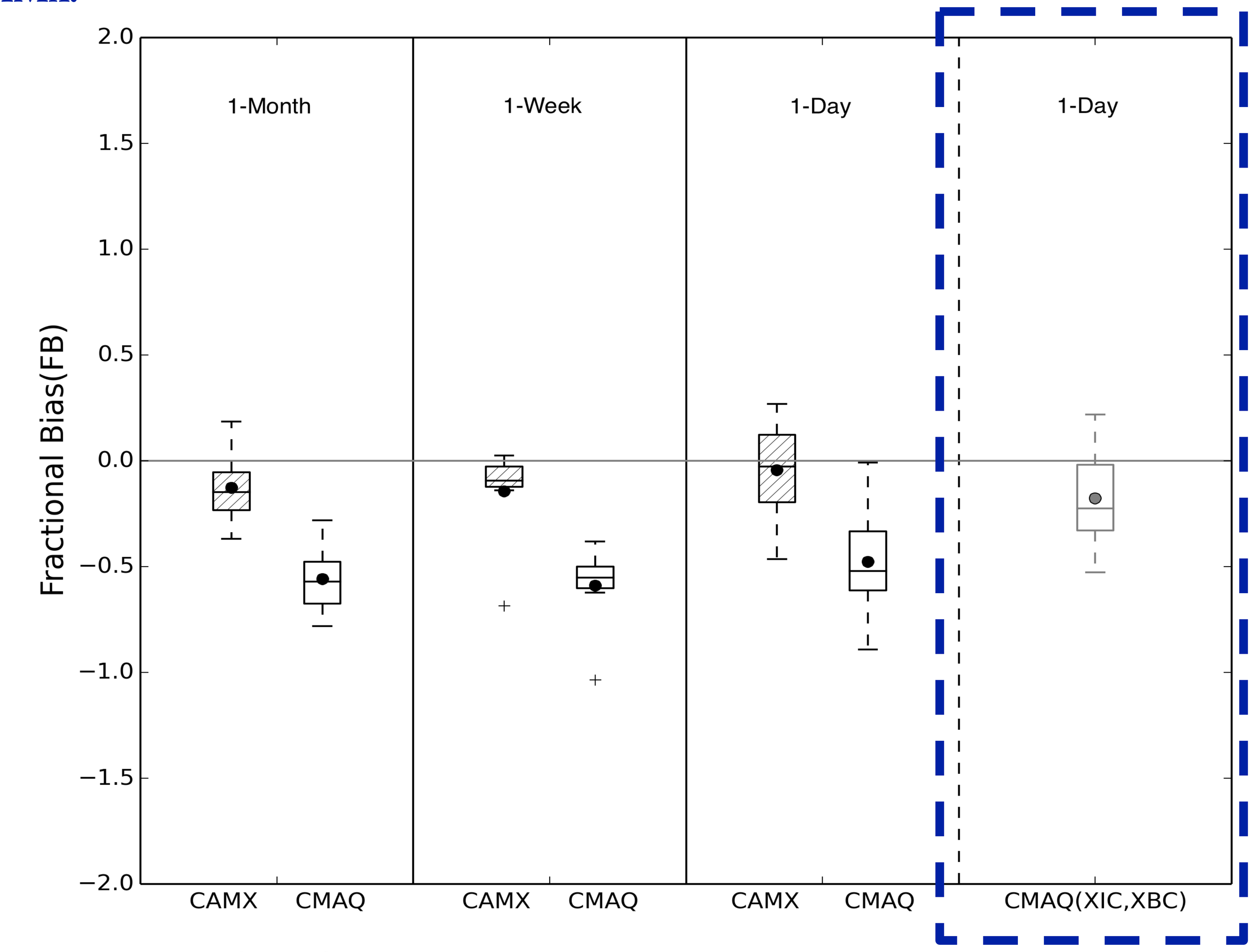


Figure 2. Scheme of CAMx and CMAQ simulation. IC/BC represents initial and boundary condition

Results

➤ **Case study selection and demonstration**

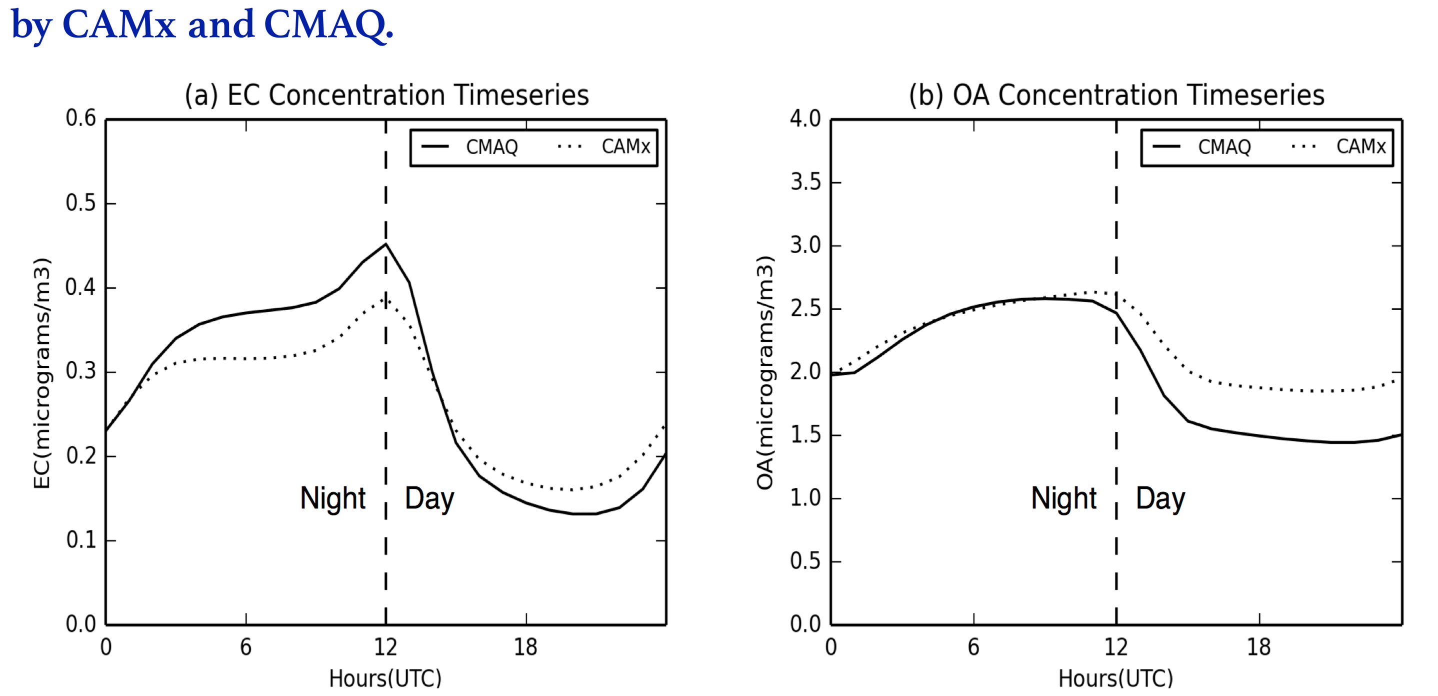
Figure 3. Fractional biases of OC modeled by CAMx and CMAQ at all monitoring sites including IMPROVE and STN for the summer episodes. 1-Month represents results of the 1-month simulation (August). 1-Week represents results of 1-week simulation (Aug. 1st to 7th) including two days of observations (Aug. 2nd and 5th). 1-Day represents results of the 1-day simulation (Aug. 2nd). CMAQ (XIC, XBC) represents the modified CMAQ simulation using conversions of IC and BC from CAMx.



➤ **Case study characterization**

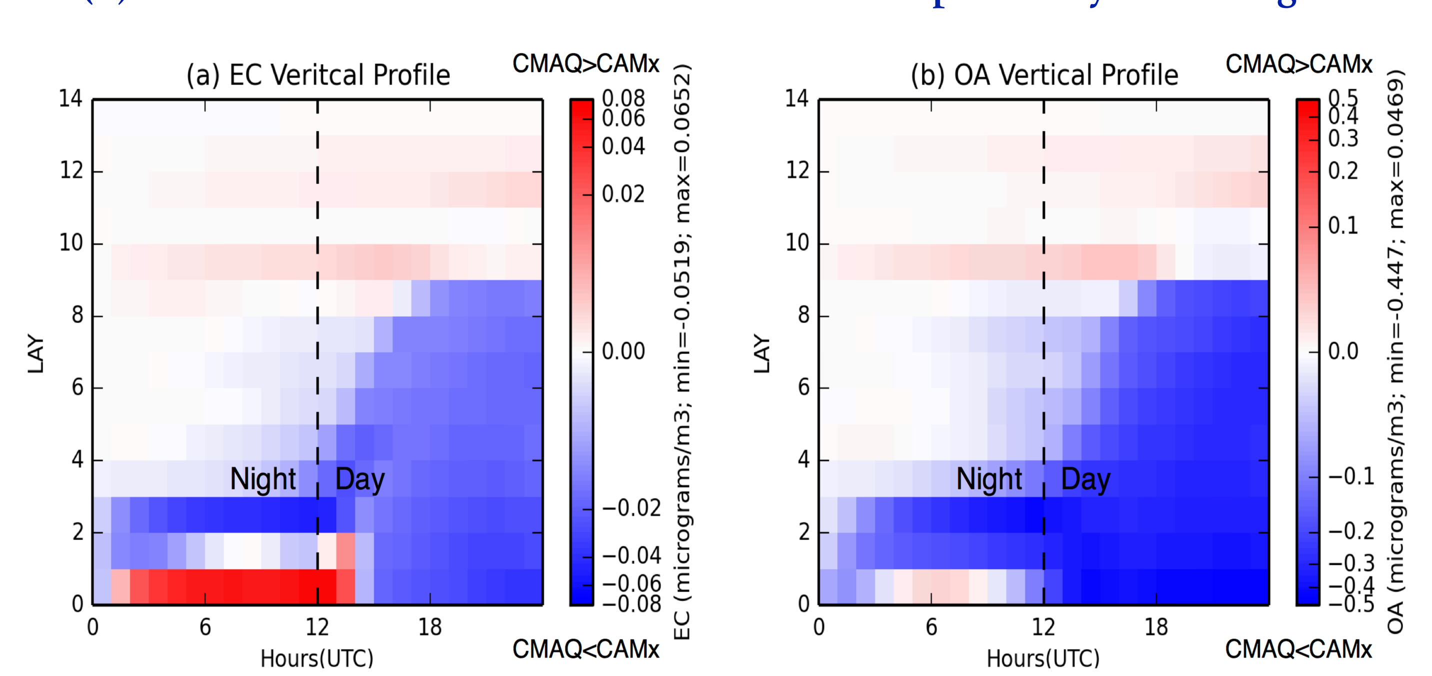
1. Concentration time series

Figure 4. Characterization of case study (Aug. 2nd) shown with time series of (a) EC and (b) OA surface layer concentrations at all domains on Aug. 2nd modeled by CAMx and CMAQ.



2. Vertical profiles

Figure 5. Differences in grid distributions between CAMx and CMAQ of (a) EC and (b) OA concentrations at all domains and atmospheric layers on Aug. 2nd.



➤ **Process diagnostic**

3. Mass analysis

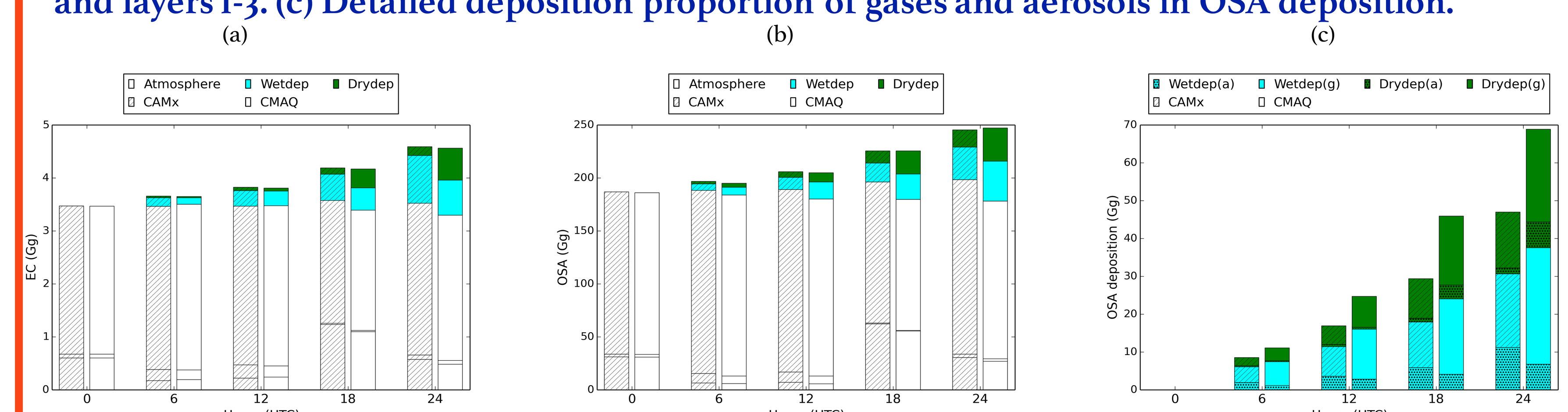
$$M_{i,t+\Delta t} = M_{i,t0} + F_{in,i,\Delta t} - F_{out,i,\Delta t} + E_{i,\Delta t} + P_{i,\Delta t} - L_{i,\Delta t} - D_{i,\Delta t} - W_{i,\Delta t}$$

M_i (mass of species); F (transition flux of species); E (source emission); P (chemical production of species); L (chemical loss of species); D (dry deposition of species); W (wet deposition of a species).

Table 2. Process quantifications of EC and OSA by using process analysis in CAMx and CMAQ
(OSA = SVOC (g) + SOA (a), Including all semi-volatile gases involved in SOA formation in both the gas (g) and aerosol (a) phases)

		CAMx					CMAQ				
	Hours	0h	6h	12h	18h	24h	0h	6h	12h	18h	24h
EC	Emissions	0.00	0.21	0.40	0.80	1.23	0.00	0.19	0.36	0.74	1.15
	Atmmass	3.46	3.45	3.45	3.56	3.51	3.47	3.51	3.48	3.40	3.30
	Wetdep	0.00	-0.16	-0.29	-0.49	-0.89	0.00	-0.12	-0.27	-0.41	-0.65
	Drydep	0.00	-0.03	-0.06	-0.12	-0.17	0.00	-0.02	-0.06	-0.36	-0.61
	Transport	0.00	-0.02	-0.05	-0.09	-0.13	0.00	0.00	-0.02	-0.05	-0.07
	Chemistry	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
OSA	Emissions	0.00	0.49	0.93	1.85	2.87	0.00	0.38	0.71	1.51	2.41
	Atmmass	185.72	187.18	187.86	195.05	197.47	185.44	183.29	179.64	179.10	177.68
	Wetdep	0.00	-6.00	-11.26	-17.67	-30.27	0.00	-7.34	-15.67	-23.89	-37.45
	Drydep	0.00	-2.42	-5.42	-11.36	-16.23	0.00	-3.66	-8.65	-21.95	-31.36
	Transport	0.00	-1.02	0.48	-1.23	-5.20	0.00	-0.95	0.63	-0.23	-2.61
	Chemistry	0.00	10.41	17.41	37.74	60.58	0.00	9.24	16.76	38.15	61.57

Figure 6. Distribution of (a) EC and (b) OSA fates (atmosphere: white; dry deposited: green; wet deposited: cyan). Within the atmospheric compartment, two solid lines denote masses in layer 1 and layers 1-3. (c) Detailed deposition proportion of gases and aerosols in OSA deposition.

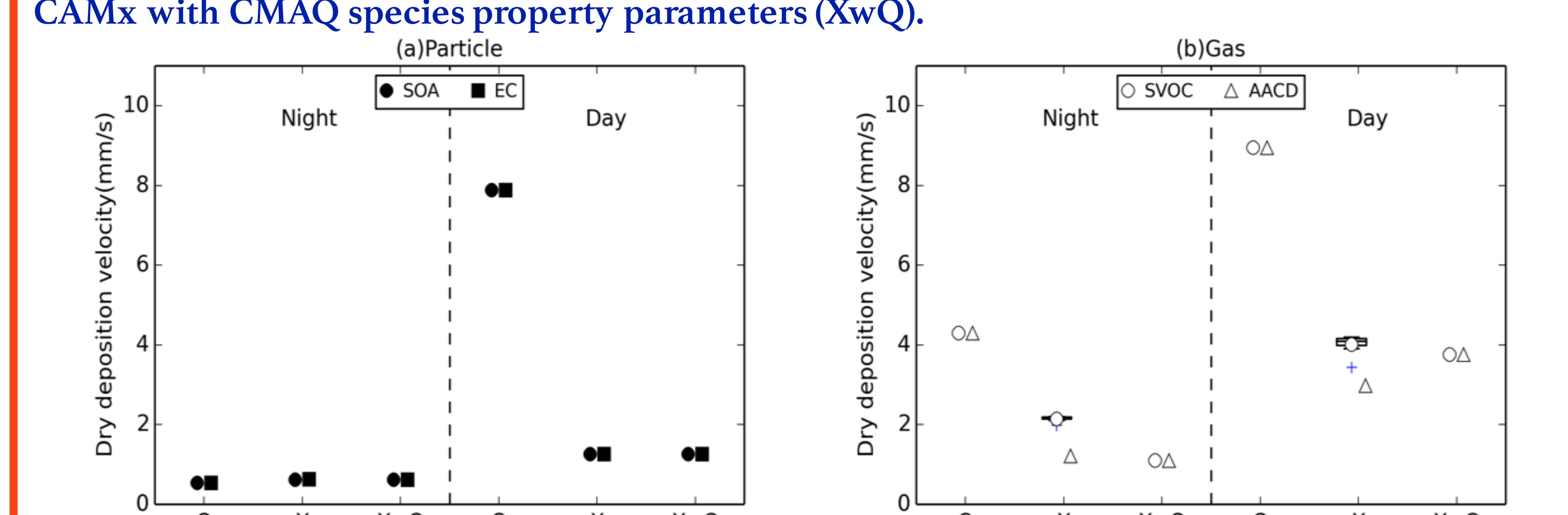


4. Deposition investigation

$$F_{D,t} = C_t \times V_{D,t} \times \Delta t$$

F (deposition flux); C (species concentration); V (species deposition velocity);

Figure 7. Mean dry deposition velocities for (a) aerosols (SOA) and (b) SVOC and acetic acid (AACD) gases at nighttime (0-12 UTC) and daytime (12-24 UTC) for CMAQ (Q), CAMx (X) and CAMx with CMAQ species property parameters (XwQ).



Conclusions

(1) CMAQ had higher gas and particle deposition velocities than CAMx, leading to more SVOCs and SOA deposited. (2) Despite immediate differences between the models, bias was not previously attributed to deposition because of compensating effects of vertical mixing. CMAQ retained more mass in the surface layer during the nighttime; this difference is likely due to distinct methods that CAMx and CMAQ calculate vertical diffusivities. (3) Overall, deposition, particularly dry, is uncertain and ill-constrained in regional models due to lack of deposition flux measurements for model evaluation.

Future work

Implement CAMx deposition methodology [Zhang et al., 2001, 2003] in CMAQ as an option to test whether it improves the CMAQ deposition process.

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