Wintertime fine particle events in Wisconsin during the 2009 LADCO Winter Nitrate Study:

meteorological event characterization & meteorological + photochemical model evaluation

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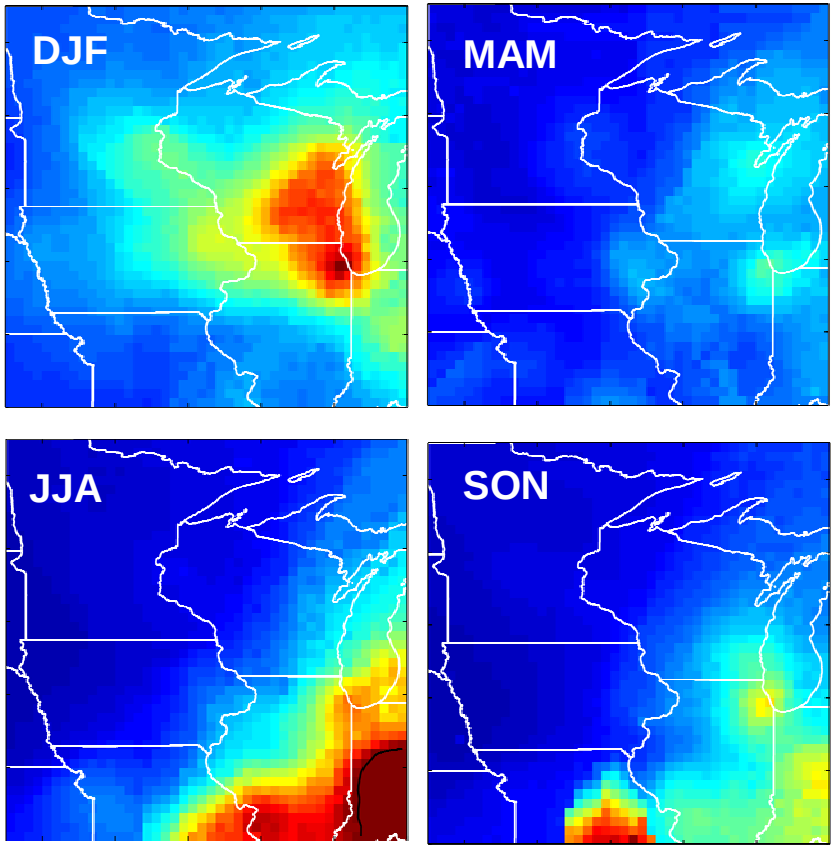
Motivations

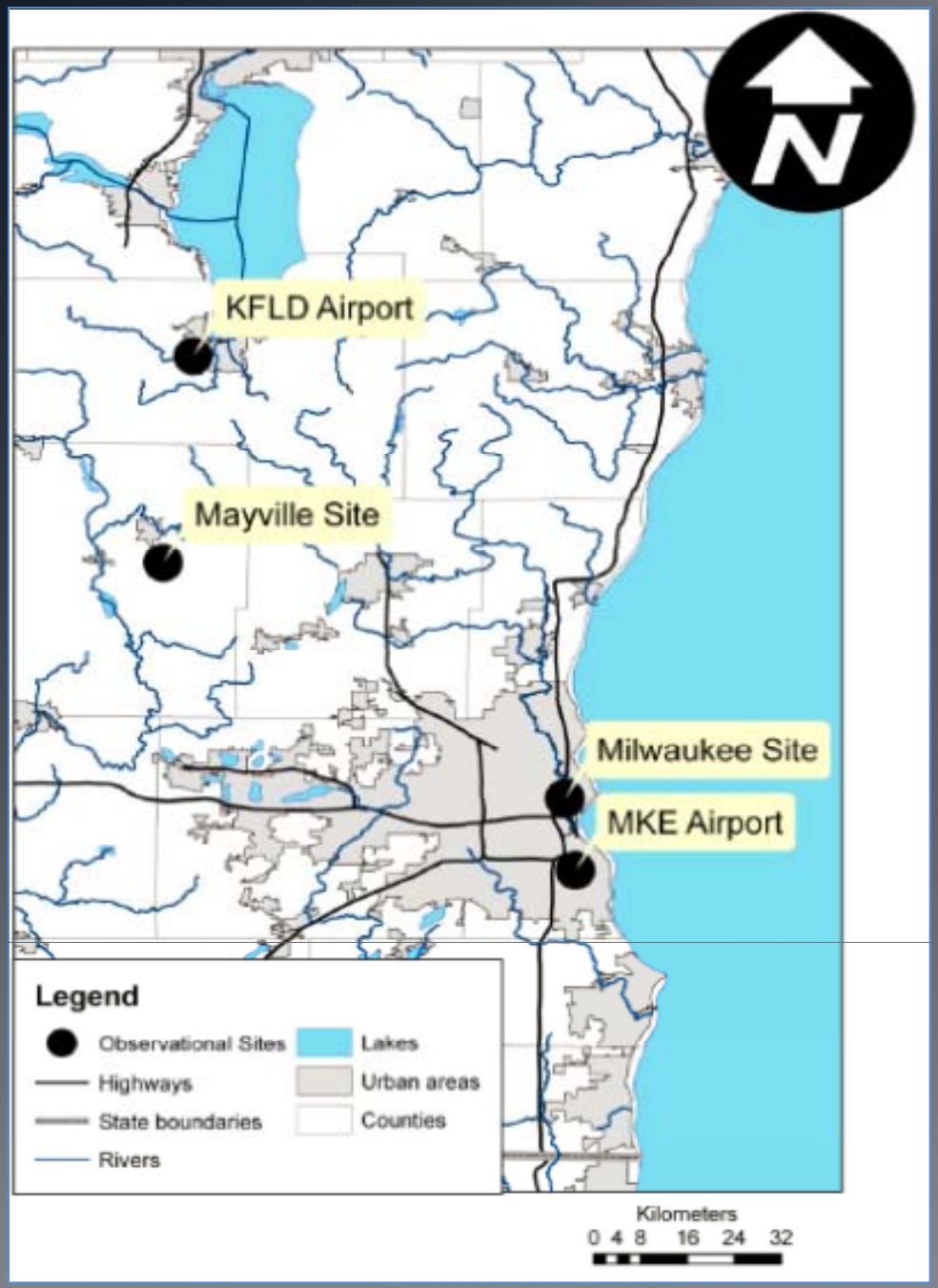
- Recurrent cold weather PM2.5 episodes greatly influence regional air quality.
- Similarities to California's Central Valley (Pun and Seigneur 1999; McMurry, Shepherd et al. 2004), Northwestern Europe and the Po Valley of Italy (Schaap, van Loon et al. 2004; Putaud, Van Dingenen et al. 2010).

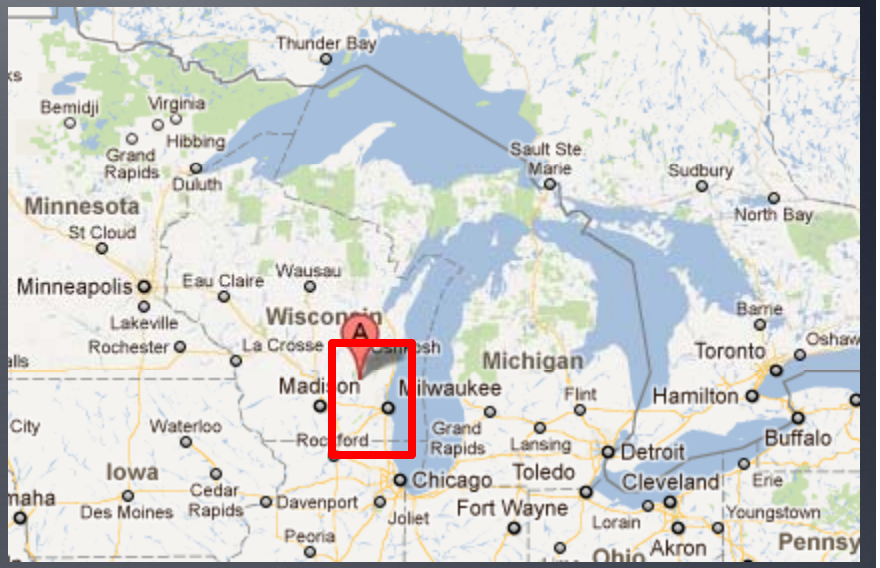
2009 AQS PM2.5



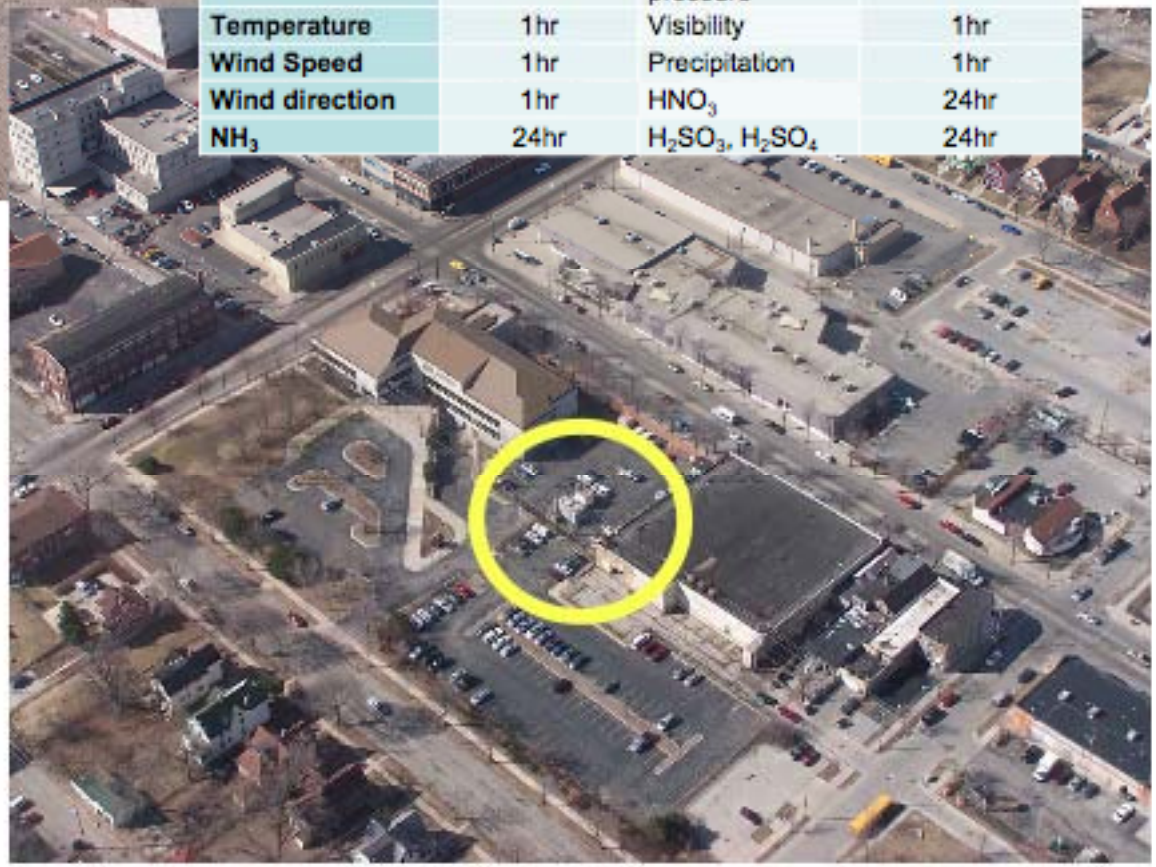
- Limited process studies to date
  - Basic conceptual model from LADCO (2009):

    Episodes tightly coupled with meteorological conditions
  - Ammonium nitrate comprises a large fraction of the PM2.5 during episodes (Chu 2004; McMurry *et al.* 2004; Blanchard & Tanenbaum 2008; Klatzman *et al.* 2009; LADCO 2009; Pitchford *et al.* 2009)


KFLD Airport
Mayville Site
Milwaukee Site
MKE Airport
Legend
Observational Sites
Highways
State boundaries
Rivers
Lakes
Urban areas
Counties
Kilometers
0 4 8 16 24 32


Thunder Bay
Bemidji
Virginia
Hibbing
Grand Rapids
Duluth
Sault Ste Marie
Sudbury
North Bay
Minnesota
St Cloud
Minneapolis
Eau Claire
Wausau
Lakeville
Rochester
La Crosse
Michigan
Madison
Milwaukee
Flint
Barrie
Toronto
Hamilton
Buffalo
Waterloo
Grand Rapids
Lansing
Detroit
Iowa
Cedar Rapids
Des Moines
Davenport
Chicago
Toledo
Cleveland
Erie
Joliet
Fort Wayne
Lorain
Youngstown
Peoria
Akron

Species	Averaging time (hr)	Species	Averaging time (hr)
$PM_{2.5}$	1hr	$PM_{2.5}$	24hr 1 in 3days
SO_4	1hr	NH_4	24hr 1 in 3days
NO_3	1hr	NO_3	24hr 1 in 3days
NH_4	1hr	SO_4	24hr 1 in 3days
HNO_3	1hr	OC	24hr 1 in 3days
NH_3	1hr	EC	24hr 1 in 3days
NO_y	1hr	O_3	1hr
NOx (note 6)	1hr	Relative Humidity	1hr
SO2 (note 7)	1hr	Surface pressure	1hr
Temperature	1hr	Visibility	1hr
Wind Speed	1hr	Precipitation	1hr
Wind direction	1hr	HNO_3	24hr
NH_3	24hr	H_2SO_3, H_2SO_4	24hr



Science Questions for the Study

Composition: Typical chemical composition during episodes and non-episodes?

Urban-rural contrast: Differences in $PM_{2.5}$ concentrations (frequency and severity), chemical composition, and source regions?

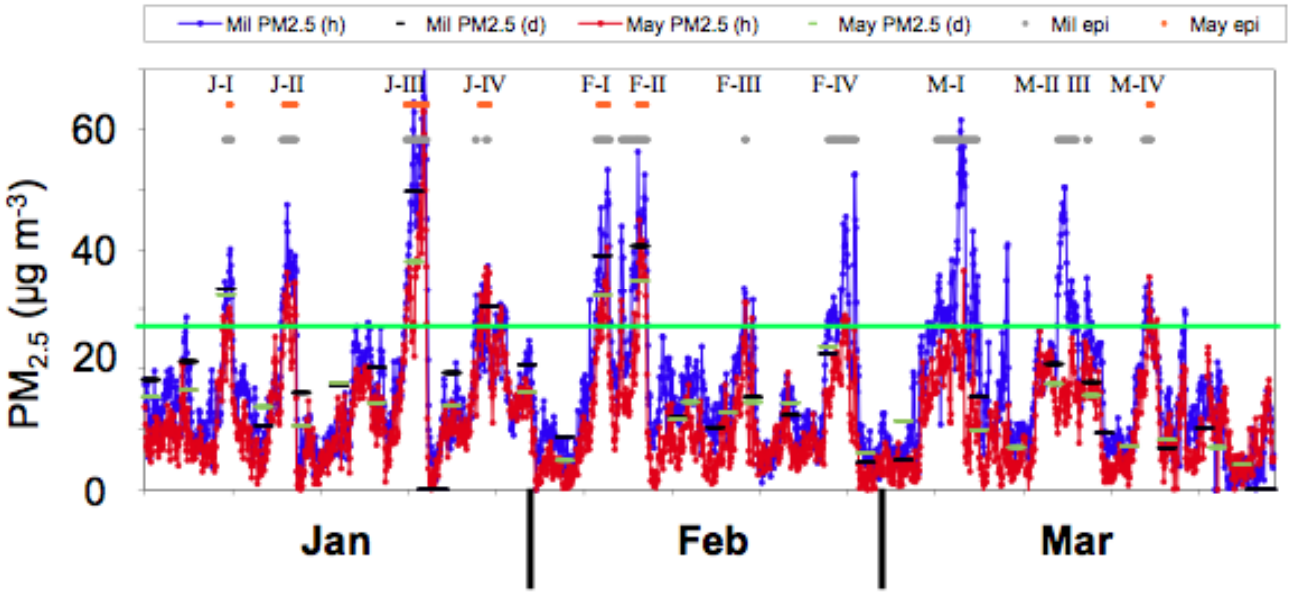
Meteorology: What meteorological conditions favor winter-time episodes? How can we best use this information to improve wintertime episode forecasting?

Nitrate formation chemistry: What do the data tell us the nitrate formation chemistry leading to events?

Sensitivity of episodes:

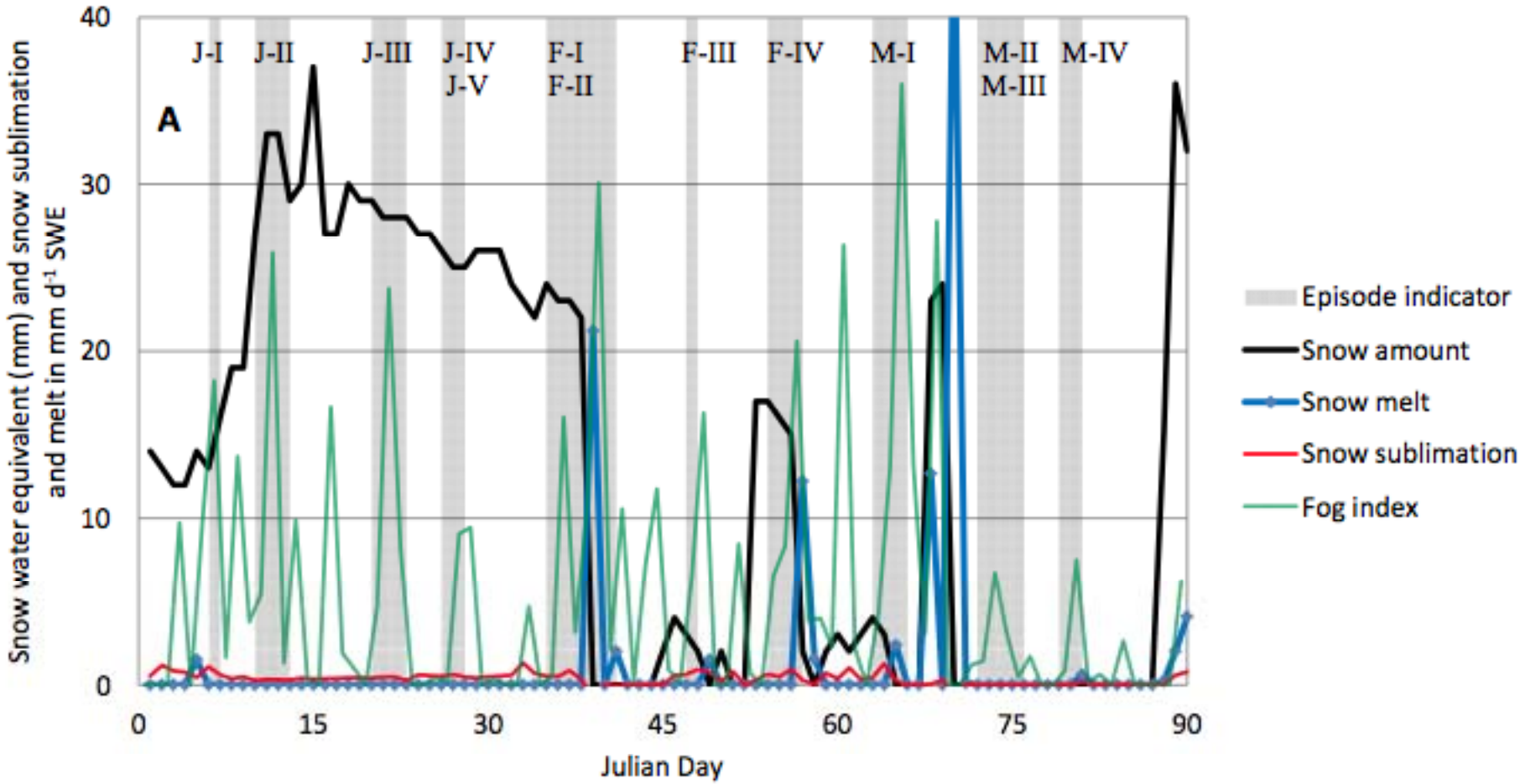
- How sensitive are concentrations to hypothetical changes in total nitrate, total ammonia, and total sulfate?
- What sources categories have leverage on episodes? Do local sources have influence?

3D Model skill: Can photochemical modeling accurately predict $PM_{2.5}$ concentrations during the observed winter-time episodes?



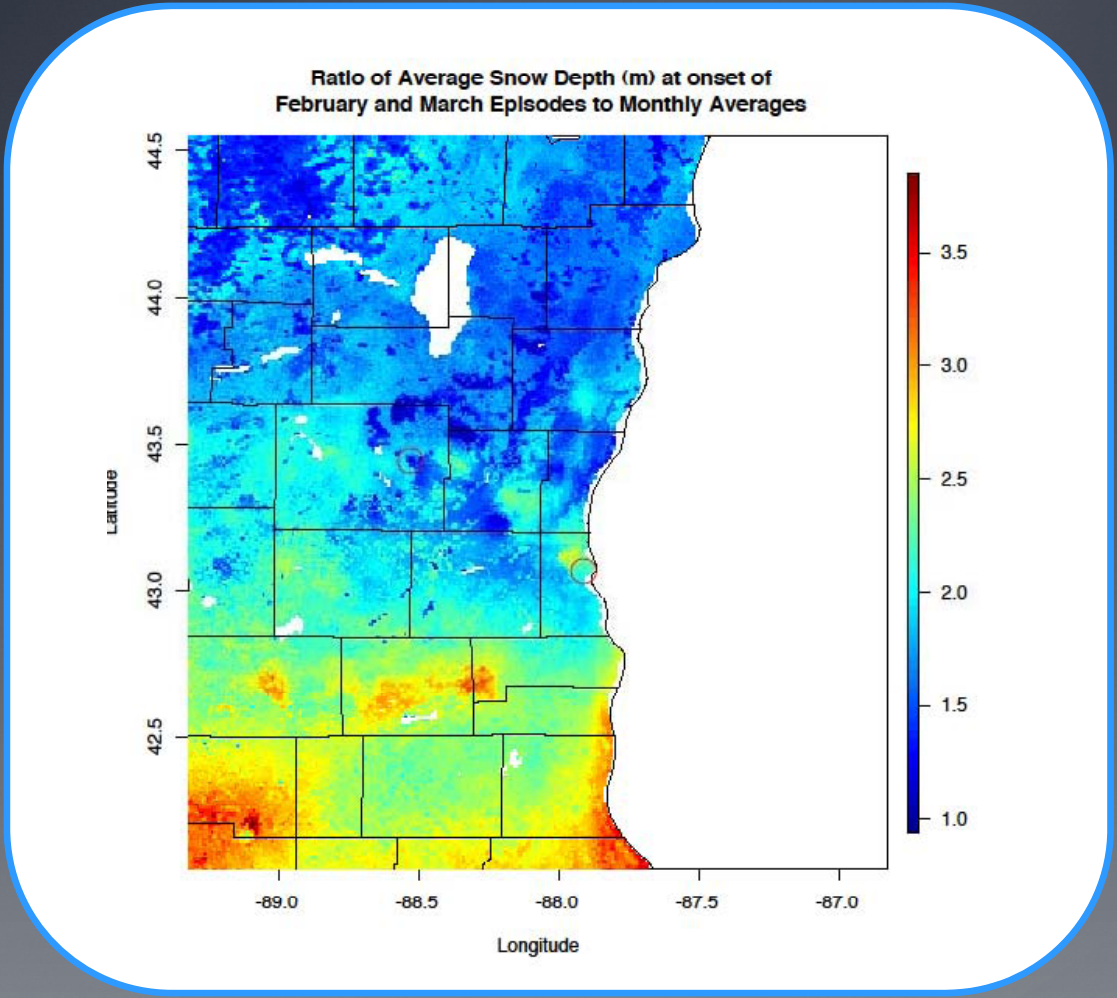
Parameter	Milwaukee (Urban)	Mayville (Rural)
$PM_{2.5}$ (µg m^{-3})	17.1	11.7
Total nitrate (µg m^{-3})	5.6	4.8
Total ammonia (µg m^{-3})	3.3	3.3
Gas ammonia (ppb)	2.3	2.4
Nitrate aerosol / total nitrate	78%	69%
NO_y (ppb)	27	6.3
Temperature (°C)	-3	-5
Ozone (ppb)	22	31
OC (µg m^{-3})	3.6	3.2
EC (µg m^{-3})	0.5	0.3
Gas Ratio (d'less)	1.5	1.7

Meteorology



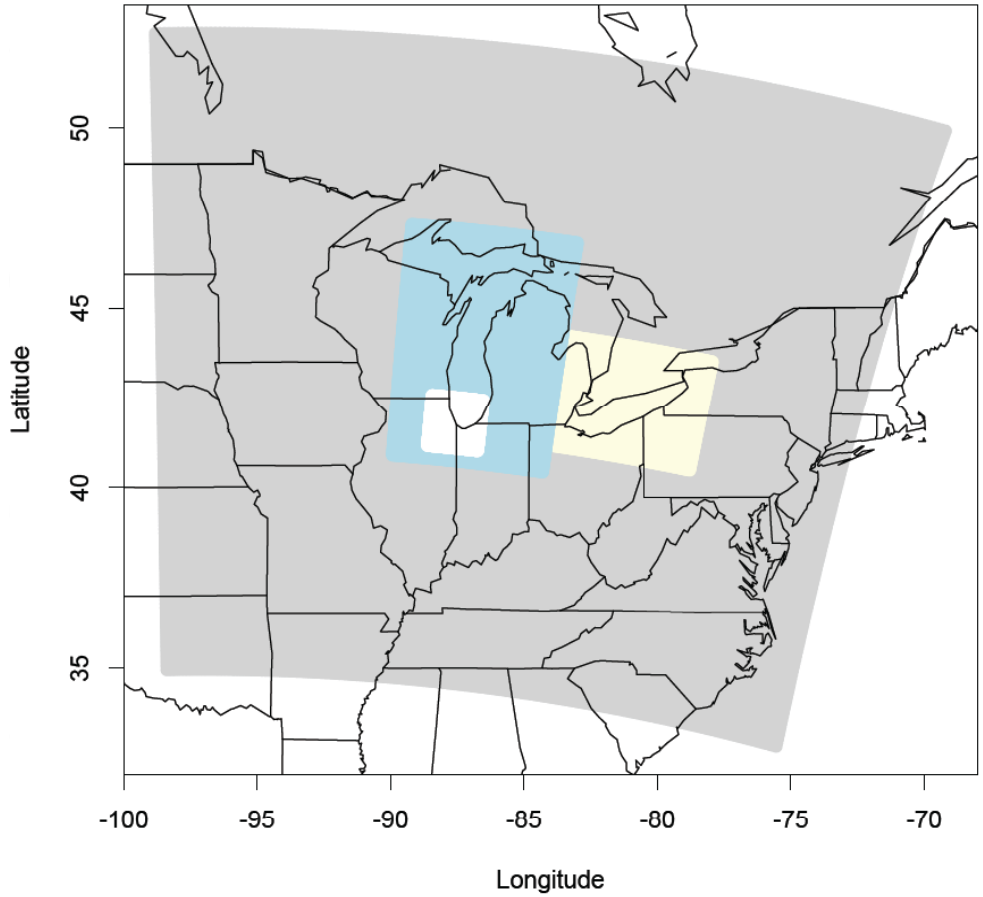
- Episodes began under similar synoptic conditions – arrival of a surface low pressure system.
- Episodes were marked by inversions with warm, moist air and low wind speeds.
- Snow cover and fog were both correlated with episode occurrence.
- Regional snow cover was present over southeastern Wisconsin and northern Illinois at the onset of late winter episodes and usually melted by episode end.

1 km SNODAS snow melt strongly correlated with PM2.5 episode intensity



Model Configuration

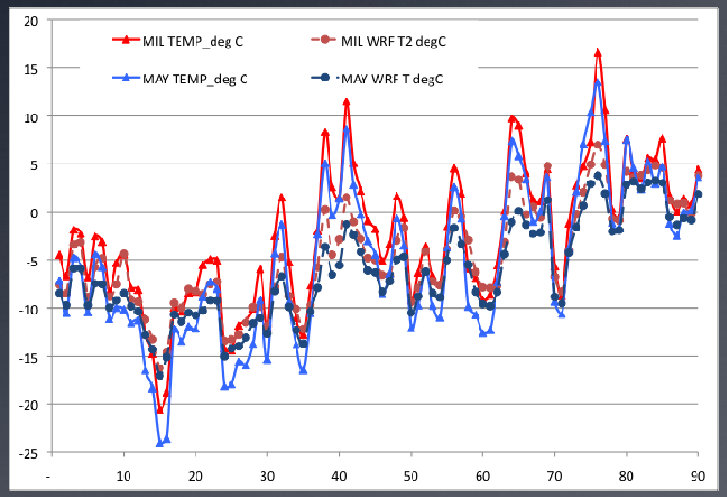
- Community Multiscale Air Quality Model (CMAQ) v4.7.1
  - CB05 gas phase / AERO5 aerosol module
  - ACM2 PBL closure
  - Mass-conserving advection
  - 14 vertical layers
- LADCO's 12 km regional modeling grid
  - Hourly boundary conditions from a 36 km simulation (with the same configuration) covering the continental United States.
- Meteorology
  - WRF 3.1.1 with the RPO configuration selected by Iowa DNR, SESARM, and LADCO
  - ACM2 PBL closure
  - Pleim-Xu land surface module
  - RRTM radiation
  - Morrison microphysics
  - Kain-Fritsch cumulus
  - Boundary meteorology from the North American Regional Reanalysis 3-hourly
  - Analysis nudging on NARR above the PBL (no direct observational nudging)
- Emissions
  - LADCO's 2008 emissions inventory used for 12km domain.
  - Day-specific biomass burning emissions from MODIS fire detection products.
  - NEI with day- specific biomass burning will be used for 36 km continental simulations.



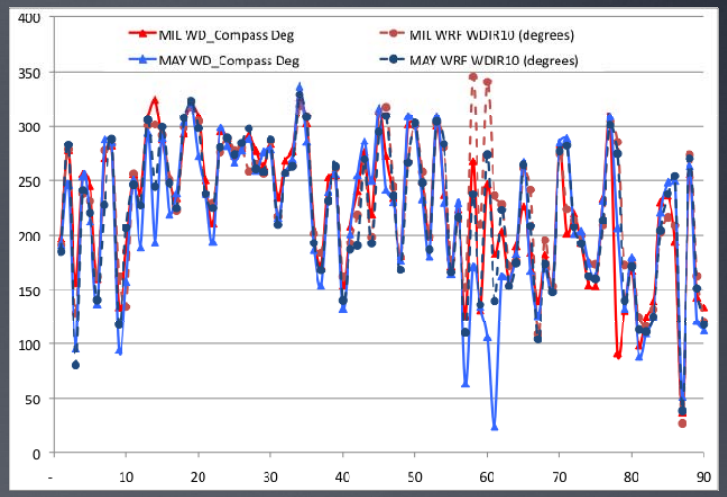
- Process Analysis
  - Chemical and process rates stored for all layers up to 550 m, with focus on NOy processing and N2O5 heterogeneous chemistry

Daily meteorology

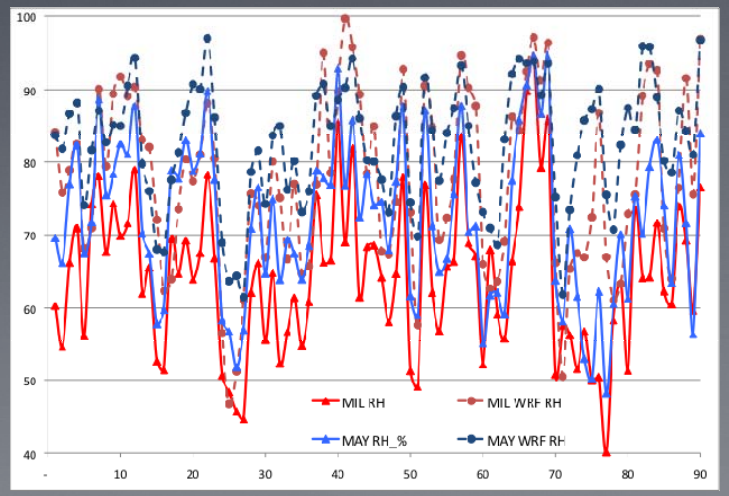
Temperature (ºC)



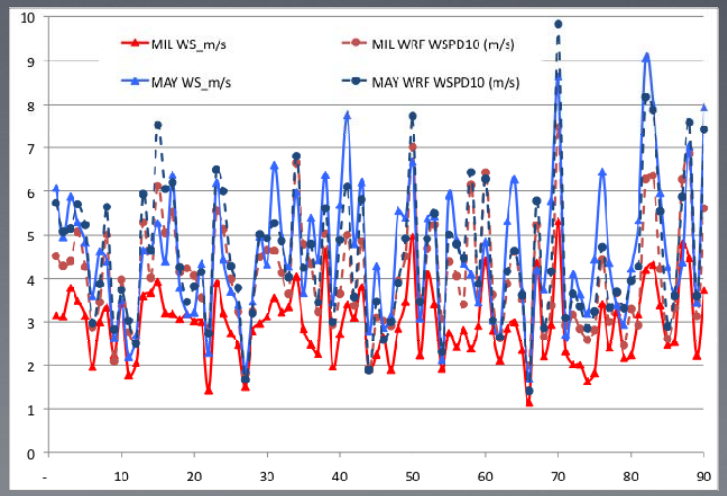
Wind Direction (degrees)



Relative Humidity (%)



Wind Speed (m/s)



Performance statistics by episode

Milwaukee

		J-I	J-II	J-III	J-IV	J-V	F-I	F-II	F-III	F-IV	M-I	M-II	M-III	M-IV	M-V
Temperature	Observed	-3.6	-4.0	-8.2	-4.8	-13.0	-11.7	-1.0	2.3	2.9	2.8	7.0	4.5	10.3	7.1
	WRF	-5.0	-5.2	-9.3	-7.5	-14.0	-12.1	-5.6	-3.5	-0.1	-0.3	2.0	0.9	0.0	4.2
	Bias	-1.4	-1.1	-1.1	-2.7	-1.0	-0.4	-4.6	-5.8	-3.1	-3.1	-5.0	-3.7	-10.3	-2.9
	RMSE	3.9	1.7	1.5	3.1	1.3	1.0	6.3	6.9	3.2	5.1	6.2	6.3	10.3	6.2
Wind Direction	Observed	229.8	284.9	247.2	234.7	293.2	318.6	178.8	207.1	179.2	179.5	180.8	163.3	192.7	178.9
	WRF	234.2	281.7	238.0	238.4	293.8	309.4	185.8	212.1	163.4	186.5	207.3	175.8	187.8	177.7
	Bias	4.40	-3.27	-9.23	3.74	0.56	-9.20	7.08	4.95	-15.80	6.96	26.55	12.46	-4.84	-1.22
	RMSE	71.8	7.5	44.0	34.7	46.6	10.9	20.4	23.3	16.1	65.3	106.7	27.8	16.6	107.5
Wind Speed	Observed	3.1	3.8	2.2	2.0	1.5	2.6	2.4	2.4	2.8	2.7	2.3	1.4	2.0	2.1
	WRF	4.2	4.5	2.6	3.3	1.8	2.9	3.3	3.4	3.3	4.2	3.4	2.5	2.8	2.8
	Bias	1.1	0.7	0.5	1.3	0.3	0.2	0.9	1.0	0.5	1.6	1.0	1.1	0.8	0.7
	RMSE	1.7	0.9	0.8	1.4	0.4	0.4	1.2	1.3	0.8	2.0	1.6	1.3	0.8	0.9
RH	Observed	62.6	73.1	78.8	77.0	48.7	67.2	76.0	76.1	74.5	70.3	79.2	58.9	66.1	57.9
	WRF	75.9	89.1	90.1	90.1	57.8	85.4	80.1	85.1	88.9	79.0	88.8	72.5	98.4	72.2
	Bias	13.3	16.0	11.3	13.1	9.1	18.2	4.2	9.0	14.4	8.7	9.6	13.6	32.2	14.3
	RMSE	20.3	16.8	11.9	14.8	12.7	18.6	13.6	15.8	16.1	15.8	16.4	21.0	32.3	22.5

PM2.5	Observed	13.1	33.6	36.1	50.3	28.9	31.4	37.6	35.4	30.3	31.3	33.8	35.5	31.6	29.4
	CMAQ	16.6	30.7	29.2	24.8	38.8	31.2	36.4	25.6	27.5	25.9	22.5	38.4	19.6	28.3
	Bias	3.5	-2.9	-6.8	-25.5	9.9	-0.2	-1.2	-9.8	-2.8	-6.2	-11.3	2.9	-12.0	-1.1
	RMSE	9.5	11.0	7.9	27.0	11.6	5.3	11.4	17.8	5.4	16.9	16.8	9.9	12.7	13.4
Nitrate	Observed	1.6	-	5.7	6.6	2.8	3.0	3.8	4.8	3.5	3.5	3.5	2.8	4.5	2.8
	CMAQ	1.3	1.3	2.7	4.1	0.9	0.8	3.0	2.0	2.9	2.6	2.5	1.7	3.2	2.3
	Bias	-0.2	-	-2.3	-2.4	-1.9	-2.2	-0.7	-2.8	-0.6	-0.9	0.2	-1.1	-1.4	-0.5
	RMSE	1.2	-	5.6	3.6	2.0	2.2	1.6	2.9	0.6	1.1	0.8	1.3	1.4	0.6
Ammonium	Observed	3.0	14.3	-	19.3	10.7	12.1	10.9	11.1	8.8	9.4	5.9	11.8	6.0	8.2
	CMAQ	3.5	11.2	7.3	6.0	10.4	8.9	9.6	5.8	6.0	5.6	4.1	14.3	4.6	6.8
	Bias	0.4	-4.2	-	-13.3	-0.3	-3.2	-1.5	-5.3	-2.8	-3.9	0.1	2.4	-1.4	-1.7
	RMSE	2.3	4.4	-	14.2	0.9	4.0	4.0	7.9	3.0	6.2	3.8	4.6	1.6	3.9
Sulfate	Observed	1.6	6.2	-	7.4	3.7	3.9	4.3	5.1	4.3	4.2	4.1	4.5	3.7	3.5
	CMAQ	1.5	3.7	3.1	3.2	3.4	2.9	3.7	2.5	2.9	2.7	2.0	4.8	2.6	3.2
	Bias	-0.1	-2.5	-	-4.2	-0.3	-1.0	-0.6	-2.6	-1.4	-1.6	-1.6	0.3	-1.1	-0.5
	RMSE	0.9	2.7	-	4.5	0.5	1.4	1.3	3.4	1.5	2.3	2.5	1.4	1.1	1.7
EC	Observed	0.3	0.7	0.4	4.0	-	0.6	0.8	0.5	-	0.5	0.5	0.8	0.6	-
	CMAQ	0.7	1.0	1.2	0.7	1.7	1.3	1.6	0.9	0.8	1.2	0.8	1.4	0.7	1.1
	Bias	0.3	0.3	0.6	-3.2	-	0.6	0.9	0.5	-	0.3	0.5	0.5	0.1	-
	RMSE	0.6	0.6	0.7	3.2	-	0.6	1.4	1.0	-	0.4	0.7	0.6	0.2	-
OC	Observed	3.3	3.4	3.1	-	-	4.7	5.9	3.6	-	7.8	4.1	5.1	6.3	-
	CMAQ	1.7	2.8	2.8	2.1	5.2	3.8	4.1	2.4	2.6	2.9	2.0	3.3	1.6	2.8
	Bias	-1.7	-0.6	-0.7	-	-	-0.9	-1.2	-1.0	-	-5.3	-1.6	-1.0	-4.7	-
	RMSE	2.5	1.6	0.7	-	-	1.0	2.0	2.1	-	5.4	2.3	1.8	4.8	-

Suggests a correlation matrix…

Focus on fog & snow cover

RH the strongest predictor of episode intensity &model skill

		Highest R^2	Explained by	
PM2.5	Observed	23%	RH Obs	✔
	CMAQ	29%	RH WRF	✔
	Bias	53%	RH WRF	✔
	RMSE	43%	WS Bias	
Nitrate	Observed	40%	RH Obs	✔
	CMAQ	36%	WD WRF	
	Bias	40%	T WRF	
	RMSE	35%	T WRF	
Ammonium	Observed	23%	WD RMSE	
	CMAQ	19%	WD RMSE	
	Bias	25%	RH Obs	✔
	RMSE	27%	WS Bias, RH Obs	✔
Sulfate	Observed	25%	RH obs	✔
	CMAQ	18%	WD RMSE	
	Bias	45%	RH Obs	✔
	RMSE	45%	RH Obs	✔
EC	Observed	9%	WS Bias	
	CMAQ	44%	RH WRF	✔
	Bias	13%	WS Bias	
	RMSE	18%	RH Obs	✔
OC	Observed	28%	WD WRF	
	CMAQ	46%	RH WRF	✔
	Bias	25% - 31%	5 variables	✔
	RMSE	40% - 45%	3 variables	✔

Multiple WRF settings to identify the role of snow

- RPO 2007/2008 SIP configuration with NARR analysis nudging
- Snow cover included as base case — too persistent
- Test without snow cover
- WI DNR WRF – identical physics & dynamics, but 5-day restarts from NCAR FNL analysis. Snow still too persistent.

Meteorology evaluation: Milwaukee

		No Episode		Local Episode		Total	
		Base	No Snow	Base	No Snow	Base	No Snow
Temperature (°C)	Observed	-3.6		1.4		-2.7	
	WRF	-1.8	-5.0	0.0	-2.4	-1.5	-4.6
	Bias	1.8	-1.4	-1.4	-3.8	1.3	-1.8
	RMSE	5.8	3.9	5.3	5.5	5.7	4.2
RH (%)	Observed	62.6		72.8		64.4	
	WRF	63.5	75.9	79.4	83.8	66.4	77.3
	Bias	1.0	12.6	6.6	12.4	2.0	12.9
	RMSE	16.3	20.3	17.6	16.8	16.6	19.7
P (hPa)	Observed	992.6		989.2		991.9	
	WRF	995.4	996.1	993.6	993.7	995.1	995.7
	Bias	2.9	3.6	4.4	4.6	3.1	3.7
	RMSE	3.5	3.9	4.7	4.9	3.8	4.1
Wind speed (m/s)	Observed	3.1		2.3		3.0	
	WRF	3.1	4.2	3.3	3.3	4.7	4.1
	Bias	1.0	1.1	1.0	1.0	1.8	1.1
	RMSE	2.6	1.7	1.5	1.4	2.5	1.6
Wind direction (°)	Observed	229.8		201.5		224.7	
	WRF	229.2	234.2	210.8	209.6	225.9	229.8
	Bias	-0.6	4.4	9.2	8.1	1.1	5.1
	RMSE	63.2	71.8	58.5	63.3	79.3	70.4

Snow cover effectively reduces bias during pollution events, but only slightly improves RMSE or R^2

Meteorology evaluation: Mayville

		No Episode		Local Episode		Total	
		Base	No Snow	Base	No Snow	Base	No Snow
Temperature (°C)	Observed	-5.2		-6.4		-5.3	
	WRF	-5.4	-6.1	-5.5	-8.4	-5.4	-6.3
	Bias	-0.1	-0.9	0.9	-2.0	-0.1	-1.0
	RMSE	6.6	4.3	4.7	4.1	6.4	4.3
RH (%)	Observed	71.4		83.8		72.2	
	WRF	85.5	81.9	92.1	90.2	85.9	82.4
	Bias	14.1	9.0	8.3	7.0	13.7	-1.0
	RMSE	20.3	4.3	12.7	4.1	16.9	4.3
P (hPa)	Observed	987.9		983.7		987.6	
	WRF	982.6	982.9	979.5	979.7	982.4	982.7
	Bias	-5.3	-5.0	-4.3	-4.0	-5.2	-4.9
	RMSE	5.7	5.3	4.5	4.3	5.7	5.3
Wind speed (m/s)	Observed	4.6		3.7		4.6	
	WRF	5.0	4.6	3.7	3.4	4.0	4.5
	Bias	0.3	-0.1	-0.1	-0.4	0.3	-0.1
	RMSE	2.0	1.5	1.2	1.2	1.0	1.5
Wind direction (°)	Observed	215.5		200.0		214.6	
	WRF	223.1	222.1	215.5	217.9	222.7	221.9
	Bias	7.6	6.6	15.6	17.9	8.1	7.3
	RMSE	72.0	71.1	52.3	52.9	71.0	70.1

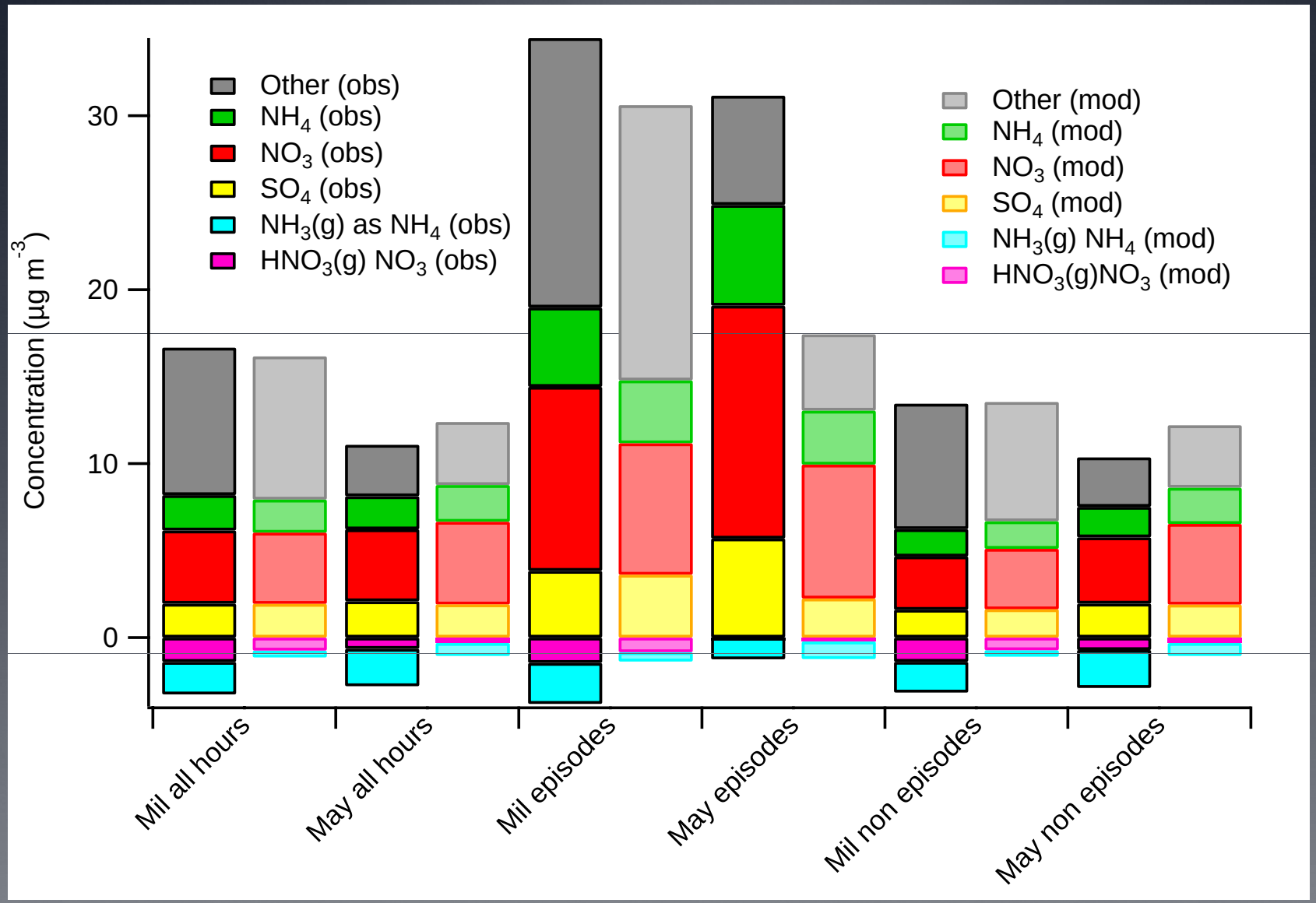
Snow cover improves performance over Mayville during episodes more than under normal conditions

Comparison at WNS Sites

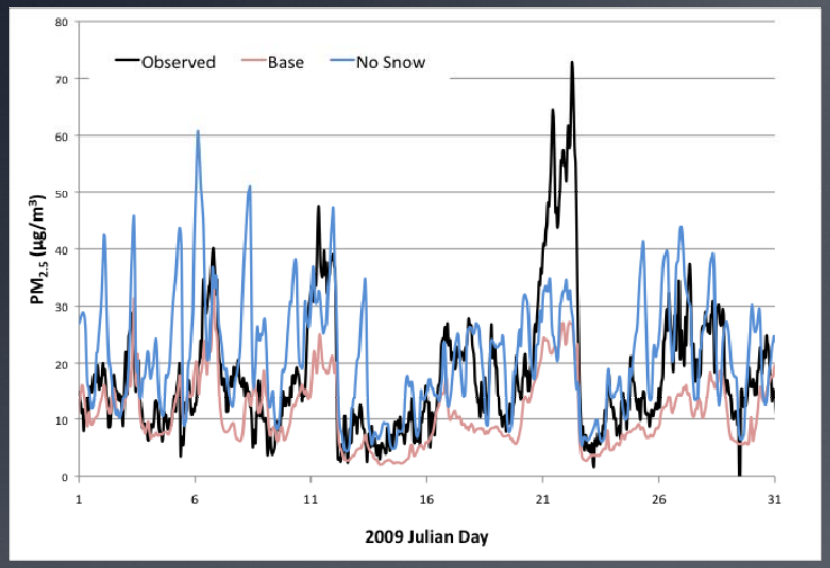
		Milwaukee				Mayville			
		No Epsode		Local Episode		No Epsode		Local Episode	
		Base	No Snow	Base	No Snow	Base	No Snow	Base	No Snow
PM2.5	Observed	13.06		35.75		10.29		33.27	
	CMAQ	13.04	18.71	29.15	31.18	11.03	10.76	19.14	18.84
	Bias	-0.05	5.67	-6.60	-4.64	1.61	0.48	-14.13	-14.44
SO4	Observed	1.61		4.12		1.99		5.62	
	CMAQ	1.61	1.27	3.49	2.49	1.82	1.23	2.68	2.44
	Bias	0.10	-0.30	-0.33	-1.38	-0.14	-0.73	-2.61	-2.89
NO3	Observed	3.01		11.22		3.85		14.90	
	CMAQ	3.27	4.08	7.10	7.75	4.59	4.29	7.78	7.68
	Bias	0.29	0.07	-3.70	-2.99	0.73	0.28	-7.50	-7.48
NH4	Observed	1.59		4.88		1.77		6.21	
	CMAQ	1.51	1.63	3.46	3.22	2.03	1.70	3.28	3.15
	Bias	-0.06	0.03	-1.19	-1.48	0.31	-0.08	-3.02	-3.10
EC	Observed	0.33		1.13		0.25		0.46	
	CMAQ	0.60	1.06	1.37	1.59	0.28	0.28	0.46	0.46
	Bias	0.23	0.69	0.07	0.44	0.02	0.05	0.03	0.06
OC	Observed	3.31		4.81		3.06		4.46	
	CMAQ	1.25	2.06	3.03	3.51	0.76	0.80	1.34	1.40
	Bias	-2.12	-1.27	-1.68	-0.79	-2.35	-2.16	-3.02	-2.92

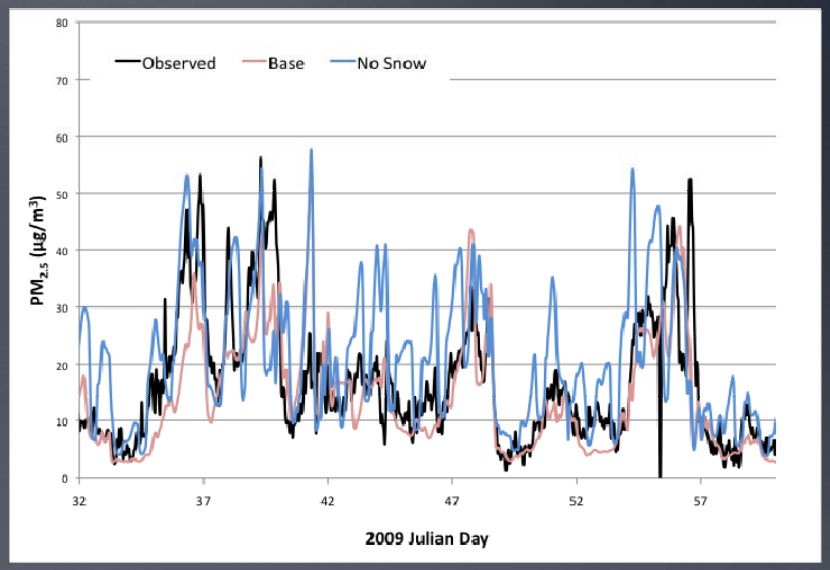
- Snow more important for simulating concentrations during Mayville episodes, Milwaukee during non-episodes
- Neither WRF simulation definitively better

Aerosol mass & composition

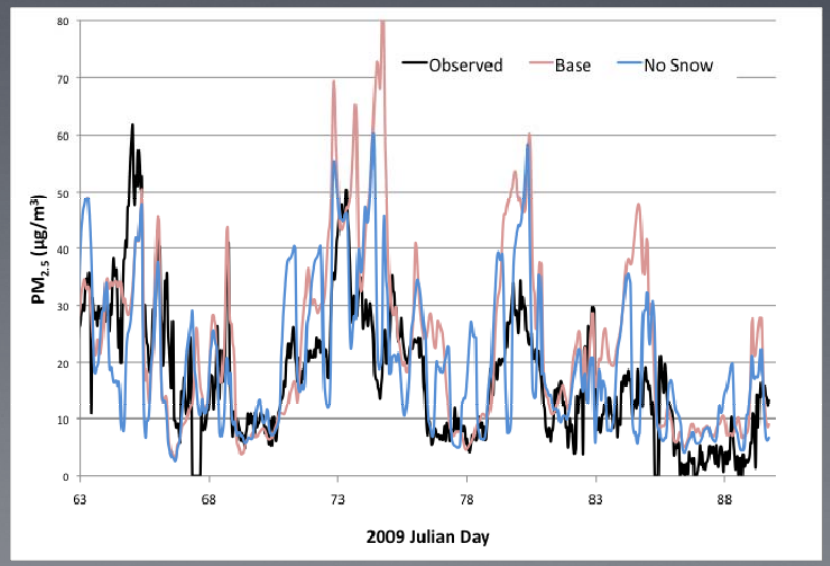


Milwaukee Hourly $PM_{2.5}$

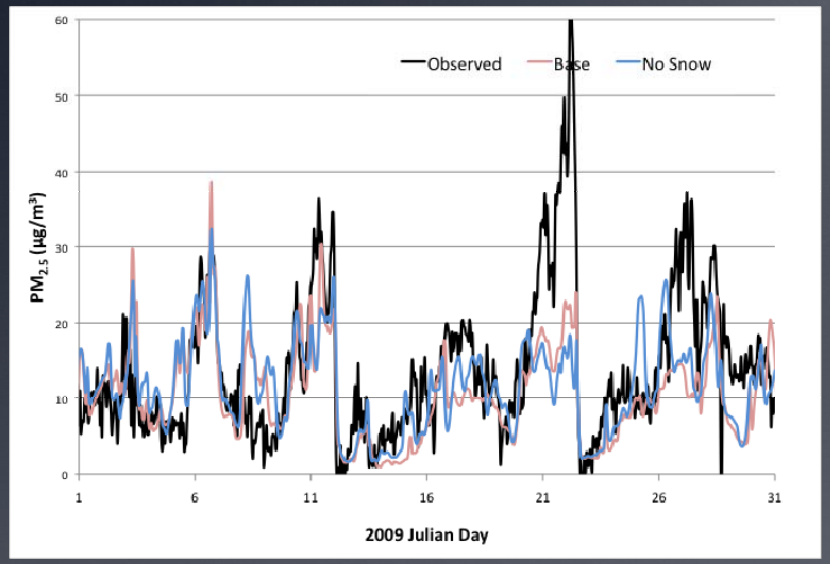


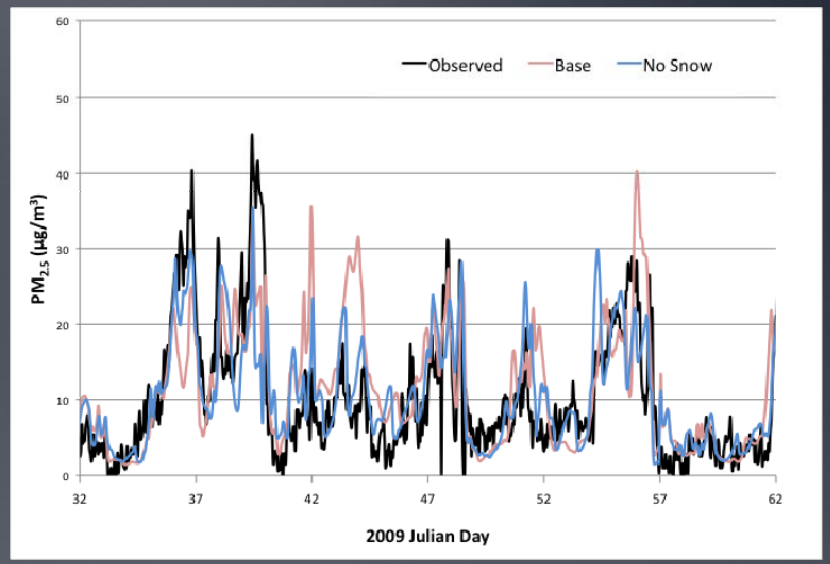


Snow cover degrades performance during cold periods in January, but then leads to stronger late season episodes



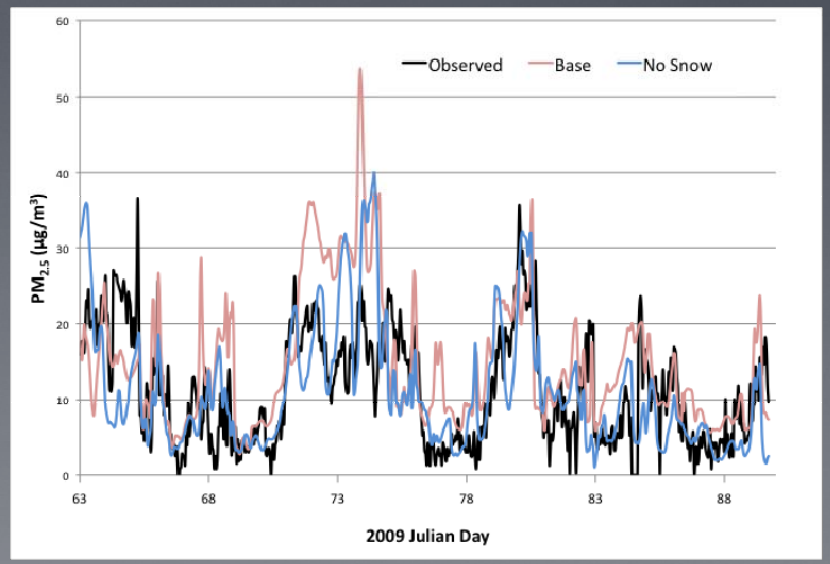
Mayville Hourly $PM_{2.5}$



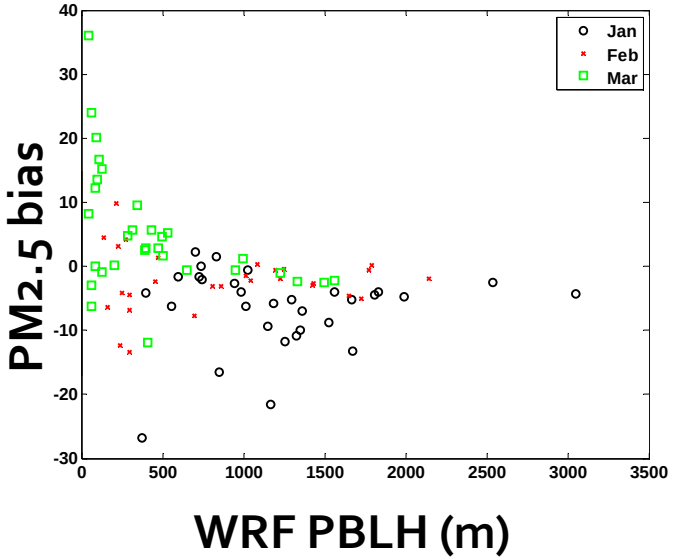


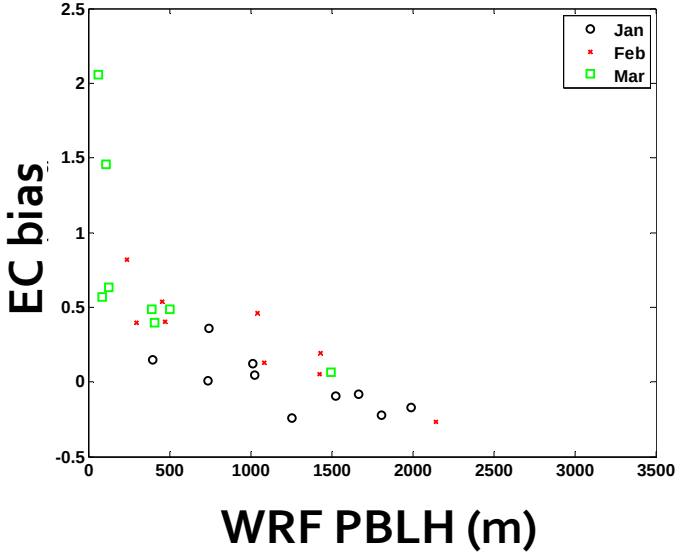
Nearly all episodes simulated, with few false alarms

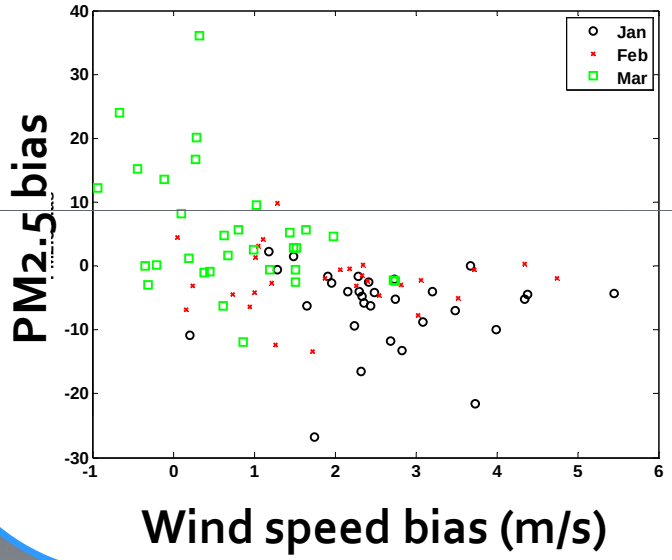
Observations often fall between the two simulations



PBL dynamics are crucial to model skill







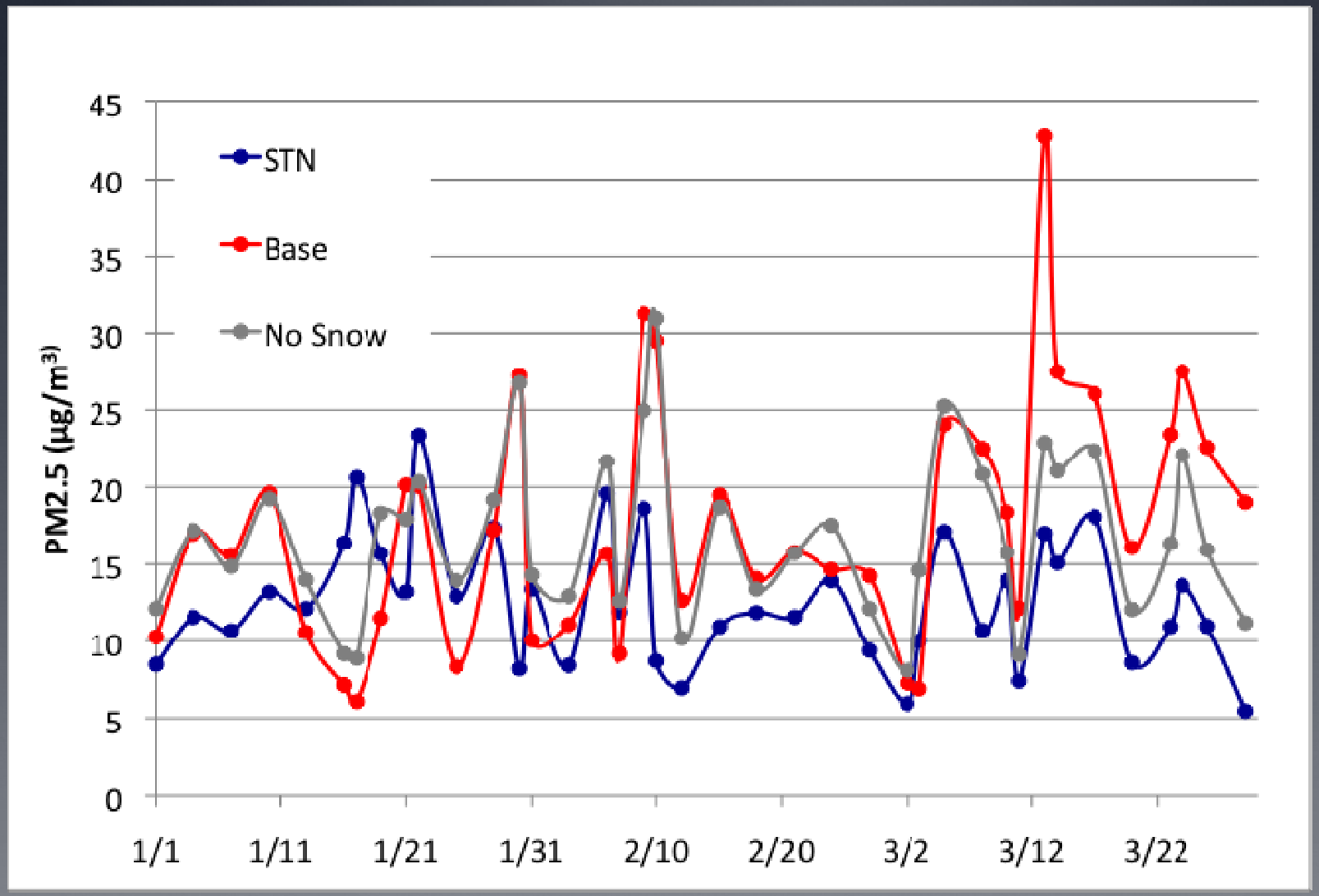
Shallow model mixed layer and low bias in wind speed drive overprediction in primary & secondary PM2.5 concentrations during wintertime fine particle events. RH bias also a factor.

Regional Average Concentrations

			CMAQ		Bias		Error	
Network	Species	Observed	Base	No Snow	Base	No Snow	Base	No Snow
AQS	Ozone	29.78	35.04	34.64	5.26	4.86	10.62	8.31
IMPROVE	PM2.5	6.56	8.60	8.23	1.64	1.27	3.69	2.70
	SO4	1.77	2.57	1.71	0.79	-0.07	1.13	0.60
STN	PM2.5	12.13	15.38	15.60	3.26	3.47	7.91	5.96
	NO3	3.00	3.02	3.76	0.03	0.76	2.19	1.58
	NH4	1.66	1.96	1.74	0.30	0.08	1.06	0.62
	SO4	2.35	3.58	2.24	1.23	-0.11	1.83	1.02

- WRF + CMAQ + LADCO inventory + 36 km BCs perform as expected for secondary inorganics and total $PM_{2.5}$
- Regional daily R^2 for No Snow >2x Base
- No obvious winner at individual sites

Midwest average $PM_{2.5}$ @ STN sites



Conclusions

1. Results suggest meteorology (PBL height & RH) the limiting factor for model skill in episode intensity, followed by emissions, with few indications of missing or erroneous processes of importance

2. From observations, snow melt & sublimation critical to improving RH & PBL height during episodes—but interactive snow effects not currently in common use for air quality modeling

3. Frequent snow cover updates at high resolution necessary as modeling moves toward finer and finer scales: SNODAS

Ongoing efforts

Additional refinement for

- RH: interactive snow effects in WRF
- daily SNODAS fields: snow cover, snow depth
- observational nudging at surface sites & tall towers
- higher resolution: vertical (75 layers) & horizontal (4 km, 1.33 km)

2009 WNS period as primary test case for Uiowa's Midwest forecasting and model evaluation

- Emissions, especially NH_4
- Chemical transport: CMAQ 5.0 & WRF-Chem

online photolysis, sectional aerosols, aerosol-cloud interactions

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