Community Multiscale Air Quality (CMAQ) Model Supports North American Airborne Mercury Experiment (NAAMEX) : Model Forecast and Post-data Analysis in Model Evaluation

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Li Pan[1,2*], Jerry Lin[3], Dan Jaffe[4], Seth Lyman[4], and Pruek Pongprueksa[3]

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ARL Air Resources Laboratory — Conducting research and development in the fields of air quality, atmospheric dispersion, climate, and boundary layer

INTRODUCTION

▪ **North American Airborne Mercury Experiment (NAAMEX):** EPA Clean Air Mercury Rules require mercury emission control from coal power generating plants. However, the limitation of our understanding on mercury leads to the scientific uncertainties of mercury. Therefore, high quality mercury measurements are needed to reduce these uncertainties. In the year of 2010 from October 25 to November 18, mercury species including GEM, HG2 and TAM and other species including CO, CO2, O3, NOX, HOX, aerosol and halogens were measured on aboard in the total of ten flights of NCAR C-130 aircraft. This project is led by the professor Dan Jaffe from U of Washington and funding supports are from NSF, EPA, NOAA and EPRI. For more details on this project, please refer to http://www.atmos.washington.edu/jaffegroup/modules/NAAMEX. In the support of this field experiment, Community Multiscale Air Quality (CMAQ) Mercury Model at the version of 4.6 was used to perform the forecasting simulation during the whole period of flight measurement. After this campaign, the post-data analysis was conducted by comparing model results with observation measurements in order to evaluate the CMAQ model performance in the prediction of mercury transport transformation and deposition in the atmosphere.

FORECAST

- Domains
  - Polar Stereographic (187 x 187 x 22; 108 x 108km) ref_lat=90.0, ref_lon=-90.0, truelat1=45.0; std_lon=-90.0;
  - Lambert Conformal (175 x 115 x 22; 36 x 36km) ref_lat=40.0, ref_lon=-97.0, truelat1=33.0; truelat2=45.0; std_lon=-97.0;
- Meteorology
  - WRF version 3.2;
  - NCEP FNL from GDAS and NOAA GFS;
  - MCIP version 3.6 to get CMAQ ready meteorology input files;
- Emissions
  - Polar domain: IPCC emissions re-gridded;
  - Lambert domain EPA NEI 2005 processed by SMOKE;
- IC/BC
  - Polar domain: GRAHM (Global/Regional Atmospheric Heavy Metals);
  - Lambert domain: Extracted from outputs of Polar domain;
- CMAQ
  - CMAQ version 4.6 with cb05hg_ae4_aq mechanism;
- Outputs
  - http://aqm.lamar.edu

Table 1. The summary of flight status on NAMMEX

Flights	Date	Time	ASL	Path	Modeled Domain
#01	10/21	19:00–24:00	1813–5378	South Dakota → Wyoming → Colorado;	US_175x115x22 / Lambert conformal
#02	10/25	13:17–17:32	381–6523	Colorado → Kansas → Missouri → Illinois → Michigan;	US_175x115x22 / Lambert conformal
#03	10/26	12:10–16:14	3934–7062	Crossing Atlantic Ocean from US to Europe;	NH_187x187x22 / polar stereographic
#04	10/27	12:02–13:37	372–6643	European flight in France;	NH_187x187x22 / polar stereographic
#05	11/03	13:05–15:05	619–6033	European flight in France;	NH_187x187x22 / polar stereographic
#06	11/03	16:45–18:22	490–7008	European flight in France;	NH_187x187x22 / polar stereographic
#07	11/04	12:07:30–18:27	3409–6211	Crossing Atlantic Ocean from Europe to US;	NH_187x187x22 / polar stereographic
#08	11/05	16:32:30–22:22	632–6963	Maine → New York → Michigan → Iowa → Nebraska → Colorado	US_175x115x22 / Lambert conformal
#09	11/16	15:27:30–23:52	408–6919	Colorado → New Mexico → Arizona → New Mexico	US_175x115x22 / Lambert conformal
#10	11/18	15:05:00–25:00	25–7258	Colorado → Utah → Wyoming → California → Colorado	US_175x115x22 / Lambert conformal

Table 2. The summary of measured and modeled Ozone (ppbv), TAM, GEM and HG2 (pg/std_m3) concentrations in NAAMEX flights.

Flight	Species	Min		Max		Mean		STD		#Num
		OBS	MOD	OBS	MOD	OBS	MOD	OBS	MOD	
#01	O3	27.8	44.1	53.8	61.4	43.3	53.6	6.6	5.8	114
	TAM	1305.9	1337.8	1822.1	1402.5	1542.0	1371.0	77.0	14.9	105
	GEM	1334.6	1272.7	1748.0	1352.5	1506.0	1309.0	79.4	22.1	105
	HG2	1.0	8.0	184.4	106.9	59.6	60.9	47.3	29.7	79
#02	O3	27.0	36.0	82.8	60.0	40.6	53.9	13.5	5.3	71
	TAM	1448.0	1336.0	2403.0	1429.0	1720.0	1377.0	219.0	24.3	76
	GEM	1135.0	1286.0	2121.0	1394.0	1680.0	1324.0	224.0	34.1	76
	HG2	0.4	22.4	342.0	89.9	89.6	55.1	89.5	21.1	50
#03	O3	44.2	31.0	78.2	37.1	57.5	34.3	7.7	1.6	91
	TAM	1411.8	1322.0	2197.9	1387.0	1770.0	1346.0	143.0	18.6	67
	GEM	**908.5**	1251.0	1957.6	1296.0	1629.0	1275.0	186.0	10.8	67
	HG2	18.9	40.6	503.3	92.7	157.0	70.7	91.5	14.8	61
#04	O3	67.0	34.1	104.0	39.1	86.9	35.0	102.0	1.2	30
	TAM	1312.3	1340.4	2227.8	1440.0	1615.0	1362.0	205.0	24.8	34
	GEM	1320.9	1254.8	1925.3	1418.6	1577.0	1288.0	154.0	42.2	34
	HG2	9.8	65.7	385.9	119.8	87.3	80.7	105.0	13.4	22
#05	O3	58.2	26.2	77.7	34.7	67.9	31.0	7.2	2.1	32
	TAM	1050.6	1343.1	2108.7	1515.7	1482.0	1364.0	277.0	33.2	29
	GEM	**930.3**	1224.2	1835.8	1437.5	1333.0	1259.0	248.0	41.5	29
	HG2	30.5	78.2	302.1	129.9	154.0	105.0	77.5	17.0	28
#06	O3	62.6	24.5	87.0	41.5	71.3	33.0	5.6	6.6	36
	TAM	1029.5	1309.7	1596.4	1369.9	1269.0	1353.0	147.0	33.3	33
	GEM	**939.7**	1210.5	1503.4	1318.5	1186.0	1276.0	133.0	26.7	33
	HG2	13.3	39.7	240.2	112.6	108.0	74.5	70.3	21.9	26
#07	O3	65.6	36.1	135.3	42.2	88.6	38.9	14.3	1.6	132
	TAM	972.9	1356.0	1577.0	1401.7	1290.0	1378.0	133.0	12.4	105
	GEM	**872.5**	1264.9	1308.0	1366.2	1131.0	1318.0	102.0	29.3	105
	HG2	9.2	13.7	464.5	123.1	168.0	58.3	88.2	37.6	100
#08	O3	57.9	54.4	296.8	64.4	92.9	61.3	41.2	2.7	90
	TAM	914.6	1272.2	1835.4	1455.6	1323.0	1356.0	179.0	29.0	111
	GEM	**348.6**	1270.2	1927.5	1337.3	1201.0	1299.0	236.0	11.9	111
	HG2	1.5	2.2	589.6	118.4	151.0	59.6	118.0	22.5	94
#09	O3	34.5	41.2	149.0	65.0	77.4	55.4	19.3	7.8	166
	TAM	842.7	1343.7	2284.4	1548.1	1134.0	1371.0	155.0	28.7	104
	GEM	NA	NA	NA	NA	NA	NA	NA	NA	NA
	HG2	NA	NA	NA	NA	NA	NA	NA	NA	NA
#10	O3	41.4	27.3	98.8	64.1	73.0	54.5	15.3	9.5	192
	TAM	892.9	1354.1	1927.1	1716.0	1080.0	1384.0	185.0	64.4	188
	GEM	**641.7**	1260.7	1806.0	1631.1	897.0	1304.0	175.0	65.3	188
	HG2	5.6	58.5	439.1	111.2	188.0	79.6	82.5	11.9	184

Table 3 the summary of linear correlations between observation and model simulation for Ozone, TAM, GEM and HG2 in NAAMEX flights

Flight	Species	Slope	Intercept	R^2	#Num
#01	O3	-0.211	62.8	0.056	114
	TAM	-0.067	1474.8	0.119	105
	GEM	0.167	1057	0.360	105
	HG2	0.181	50.2	0.084	79
#02	O3	0.221	44.94	0.310	71
	TAM	0.077	1245.7	0.479	76
	GEM	0.124	1116.4	0.660	76
	HG2	0.070	48.7	0.094	50
#03	O3	-0.030	35.8	0.017	91
	TAM	0.067	1226.8	0.272	67
	GEM	0.006	1265.4	0.010	67
	HG2	-0.032	75.9	0.040	61
#04	O3	-0.020	36.7	0.026	30
	TAM	0.087	1221.1	0.530	34
	GEM	0.204	967.3	0.560	34
	HG2	0.057	75.7	0.200	22
#05	O3	0.140	21.6	0.227	32
	TAM	0.090	1230.5	0.570	29
	GEM	0.083	1149.3	0.240	29
	HG2	-0.077	117.4	0.123	28
#06	O3	0.520	-4.1	0.197	36
	TAM	-0.160	1558.5	0.515	33
	GEM	-0.102	1397.6	0.259	33
	HG2	-0.080	83.3	0.067	26
#07	O3	0.020	37.2	0.033	132
	TAM	-0.044	1435.7	0.227	105
	GEM	0.190	1102.0	0.441	105
	HG2	0.090	43.1	0.044	100
#08	O3	-0.007	62.0	0.012	90
	TAM	0.013	1339.8	0.061	111
	GEM	0.007	1291.0	0.020	111
	HG2	-0.002	58.9	0.000	94
#09	O3	0.320	31.0	0.615	166
	TAM	0.124	1231.0	0.448	104
	GEM	NA	NA	NA	NA
	HG2	NA	NA	NA	NA
#10	O3	0.270	34.7	0.190	192
	TAM	0.310	1052.0	0.780	188
	GEM	0.295	1039.6	0.630	188
	HG2	-0.005	80.7	0.001	184

Table 4. Species Correlations among GEM, HG2, TAM and Ozone in the measurement or in the model simulation for the NAAMEX flight #01

Specie vs.. Specie	Slope	Intercept	R^2	#Num
HG2				
Obs_HG2 vs.. Mod_HG2	0.18	50.2	0.084	79
Obs_GEM vs.. Obs_HG2	-0.29	497.2	0.219	79
Obs_O3 vs.. Obs_HG2	0.39	41.8	0.003	78
Mod_GEM vs.. Mod_HG2	-1.24	1680.0	0.778	121
Mod_O3 vs.. Mod_HG2	4.38	-170.4	0.813	121
GEM				
Obs_GEM vs.. Mod_GEM	0.167	1057.0	0.360	105
Obs_GEM vs.. Obs_CO	89.893	-92828.6	0.392	68
Obs_GEM vs.. Obs_O3	0.035	-9.9	0.188	100
Mod_GEM vs.. Mod_CO	0.639	-753.9	0.336	121
Mod_GEM vs.. Mod_O3	-0.211	329.1	0.534	121
TAM				
Obs_TAM vs.. Mod_TAM	-0.067	1474.8	0.119	105
Obs_GEM vs.. Mod_GEM	0.167	1057.0	0.360	105
Obs_TAM vs.. Obs_GEM	0.715	404.2	0.480	105
Obs_TAM vs.. Obs_HG2	0.120	-128.9	0.030	79
Mod_TAM vs.. Mod_GEM	-0.480	1966.1	0.113	121
Mod_TAM vs.. Mod_HG2	1.480	-1966.1	0.548	121

Model Evaluation

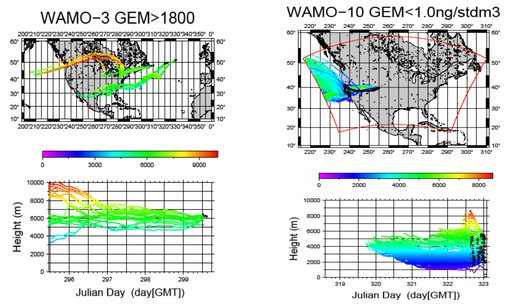
•**HG2**
- Observation shows HG2 is highly related to GEM but it is not related to ozone;
- Model shows HG2 is highly related to GEM and ozone;
- Is the reaction of ozone with GEM still important to HG2 in reality as the prediction in model?

•**GEM**
- Observation shows that GEM is positively correlated to CO and ozone;
- Model shows that GEM is positively correlated to CO but it is negatively correlated to ozone;
- For GEM, a good emission + a bad chemical mechanism => a good model prediction of GEM only if it is in a emission dominating flight;

•**TAM**
- Observation shows that TAM variation is dominated by GEM alternation;
- Model shows that HG2 alternation is more important than GEM alternation in TAM variation;
- TAM is badly predicted in the model even there is a good prediction of GEM in CMAQ ;

•**Overview**
- Airplane flew around South Dakota, Wyoming and Colorado;
- These regions are considered to be clean in CMAQ, which means GEM concentrations are very closed to model background concentration;
- GEM variation is the joint effects of mercury transport and chemistry as the chemistry is crucial;
- The GEM prediction in CMAQ shows that its concentrations alone the flight path only vary from 1272.7 to 1352.5 pg/stdm^3;
- As a result, TAM variation predicted in CMAQ is decided by HG2 variation;
- Meanwhile CMAQ predicts HG2 is mainly from GEM oxidation reaction with Ozone;
- As in the simulation of CMAQ, GEM is slowly removed from atmosphere through the pathway of converting to HG2 in a clean atmosphere condition;
- On the other hand, observation shows that GEM concentration has a big variation that is from 1334.6 to 1748.0 pg/stdm^3 and observed GEM is highly related to observed CO, which may indicate the emission influences in the areas that flight passed are strong;
- That may be the reason that ozone is positively correlated GEM in the observation;
- TAM variation is dominated by GEM alternation;
- HG2 correlated to GEM having a negative slope, but it doesn't correlated to ozone;
- Observation is showing that mercury emission contributes significantly to flight mercury measurements, with a slight influence from mercury gas phase chemistry in the air;

Fig. 1 Backward Trajectory Analysis: high GEM concentrations at high altitudes (left panel); low GEM concentrations at low altitudes (right panel);



What does it mean if GEM background concentration is lower than we understand (See Table 2) ?
1. Do we overestimate mercury emission especially for GEM natural sources ?
2. Do we ignore some important mercury sinking pathways ?
3. Do we underestimate GEM gas phase oxidation reactions in some circumstances ?

Table 5. the linear correlations between CO vs.. GEM and CO vs.. HG2 in NAAMEX flight measurements

Flight	Species	Slope	Intercept	Rsq.	#Num
#01	CO vs. GEM	0.004	1318.5	0.392	68
	CO vs. HG2	-3.780	62.8	0.000	58
#02	CO vs. GEM	0.036	147.7	0.290	50
	CO vs. HG2	-0.006	307.7	0.027	31
#03	CO vs. GEM	0.017	803.9	0.141	56
	CO vs. HG2	-0.013	800.9	0.361	51
#04	CO vs. GEM	0.013	998.0	0.084	25
	CO vs. HG2	-0.005	282.4	0.014	18
#05	CO vs. GEM	0.039	-196.9	0.636	18
	CO vs. HG2	0.003	17.3	0.035	18
#06	CO vs. GEM	0.036	-15.1	0.215	26
	CO vs. HG2	-0.008	384.0	0.046	21
#07	CO vs. GEM	0.016	383.0	0.566	83
	CO vs. HG2	0.000	154.9	0.000	79
#08	CO vs. GEM	0.030	-143.4	0.736	84
	CO vs. HG2	-0.009	548.0	0.205	73
#09	CO vs. GEM	NA	NA	NA	NA
	CO vs. HG2	NA	NA	NA	NA
#10	CO vs. GEM	0.005	454.6	0.334	151
	CO vs. HG2	0.001	71.3	0.095	147

Summary and Acknowledge

- CMAQ is capable of predicting atmospheric mercury in term of situations below;
  - CMAQ is able to predict GEM emission that is related to CO emission;
  - CMAQ is able to predict GEM oxidation reaction in the atmosphere;
  - CMAQ is able to predict GEM variation in the source regions;
  - CMAQ is able to predict TAM component;
  - CMAQ is able to predict TAM variation in the source regions;
- It is real a challenge to predict atmospheric HG2 in CMAQ;
  - CMAQ is unable to predict HG2 production reaction in the atmosphere;
  - CMAQ is unable to predict HG2 variations in all situations;
  - CMAQ is unable to predict ozone concentrations at high altitudes;
- Our assumed mercury background concentration is great than the flight measurement;
  - CMAQ is unable to predict GEM variations in the remote regions;
  - CMAQ is unable to predict TAM variations in the remote regions;

[1]NOAA/OAR/ARL, 1315 East West Hwy, Room 3316, Silver Spring, MD 20910; *li.pan @noaa.gov
[2]The Department of Mechanical Engineering, Lamar University, Beaumont, TX 77710;
[3] The Department of Civil Engineering, Lamar University, Beaumont, TX 77710;
[4] Atmospheric and Environmental Chemistry Department, University of Washington-Bothell, Bothell, WA 98011 ;