

S1. Definition of statistical measures

For quantitative comparison between the simulations we used statistical measures including correlation coefficient (R), root mean square error (RMSE), mean average error (MAE), mean bias (MB), normalized mean bias (NMB), and index of agreement (IOA). Definitions of these metrics can be found below:

$$R = \frac{(\overline{C_o} - \overline{C_p})(C_p - \overline{C_p})}{\sigma_{C_p}\sigma_{C_o}} \quad (1)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^n (C_p - C_o)^2}{n}} \quad (2)$$

$$MAE = \frac{1}{n} \sum_{i=1}^n |C_p - C_o| \quad (3)$$

$$MB = \frac{1}{n} \sum_{i=1}^n (C_p - C_o) \quad (4)$$

$$NMB = \frac{(\overline{C_p} - \overline{C_o})}{\overline{C_o}} \times 100\% \quad (5)$$

$$IOA = 1 - \frac{\sum_{i=1}^n (C_p - C_o)^2}{\sum_{i=1}^n (|C_p - \overline{C_o}| + |C_o - \overline{C_o}|)^2} \quad (6)$$

Where C_o is the observation value, C_p is the model value, σ is the standard deviation, and \overline{C} is the mean value

Table SM 1. Summary of model performance in capturing temperature at BAO 10m and 300m during Aug 1-15, 2014

T (C)- 10m	PBL			Met IC and BC		Initialization		Horizontal resolution	
	PBL1	PBL2	PBL3	Met5	Met6	Init4	Init5	Hor5	Hor5-12km
Mean Model	22.40	20.95	21.20	24.06	23.44	21.59	24.06	24.06	24.08
Mean Obs	21.67	21.67	21.67	21.67	21.67	21.67	21.67	21.67	21.67
R	0.89	0.89	0.89	0.86	0.89	0.71	0.86	0.86	0.88
RMSE	2.05	2.03	2.01	3.25	2.63	2.99	3.25	3.25	3.18
MAE	1.56	1.62	1.59	2.60	2.05	2.30	2.60	2.60	2.53
MB	0.74	-0.72	-0.46	2.40	1.77	-0.08	2.40	2.40	2.41
NMB	3.4%	-3.3%	-2.1%	11.1%	8.2%	-0.4%	11.1%	11.1%	11.1%
IAO	0.94	0.94	0.94	0.85	0.90	0.83	0.85	0.85	0.86
T (C) - 300m	PBL1	PBL2	PBL3	Met5	Met6	Init4	Init5	Hor5	Hor5-12km
Mean Model	21.91	20.95	21.30	23.58	22.89	20.31	23.58	23.58	23.52
Mean Obs	21.68	21.68	21.68	21.68	21.68	21.68	21.68	21.68	21.68
R	0.76	0.75	0.72	0.74	0.78	0.57	0.74	0.74	0.75
RMSE	2.16	2.14	2.10	2.79	2.27	3.09	2.79	2.79	2.80
MAE	1.69	1.73	1.68	2.24	1.76	2.45	2.24	2.24	2.21
MB	0.23	-0.73	-0.38	1.90	1.22	-1.37	1.90	1.90	1.85
NMB	1.1%	-3.4%	-1.8%	8.8%	5.6%	-6.3%	8.8%	8.8%	8.5%
IAO	0.87	0.85	0.84	0.78	0.85	0.72	0.78	0.78	0.79

Table SM 2. Summary of model performance in capturing relative humidity (RH) at BAO 10m and 300m during Aug 1-15, 2014

RH (%)	PBL			Met IC and BC		Initialization		Horizontal resolution	
	PBL1	PBL2	PBL3	Met5	Met6	Init4	Init5	Hor5	Hor5-12km
RH (%) -10m									
Mean Model	46.85	57.59	55.78	32.65	39.87	59.36	32.65	32.65	32.89
Mean Obs	46.47	46.47	46.47	46.47	46.47	46.47	46.47	46.47	46.47
R	0.78	0.69	0.73	0.63	0.64	0.53	0.63	0.63	0.71
RMSE	10.89	16.90	15.13	19.13	14.95	22.33	19.13	19.13	18.15
MAE	8.45	14.38	12.86	15.01	11.31	18.10	15.01	15.01	14.43
MB	0.38	11.12	9.31	-13.81	-6.60	12.90	-13.51	-13.51	-13.58
NMB	0.8%	23.9%	20.0%	-29.7%	-14.2%	27.7%	-29.7%	-29.7%	-29.2%
IAO	0.88	0.74	0.78	0.65	0.75	0.65	0.65	0.65	0.69
RH (%) -300m									
Mean Model	43.63	51.45	48.25	31.27	38.55	59.06	31.27	31.27	31.94
Mean Obs	38.70	38.70	38.70	38.70	38.70	38.70	38.70	38.70	38.70
R	0.64	0.59	0.48	0.53	0.52	0.41	0.53	0.53	0.57
RMSE	13.06	17.92	15.25	12.66	11.14	28.39	12.66	12.66	12.11
MAE	9.92	14.78	12.77	9.73	8.60	23.19	9.73	9.73	9.29
MB	4.93	12.75	9.55	-7.43	-0.15	20.36	-7.43	-7.43	-6.76
NMB	12.7%	32.9%	24.7%	-19.2%	-0.4%	52.6%	-19.2%	-19.2%	-17.5%
IAO	0.75	0.61	0.60	0.67	0.71	0.43	0.67	0.67	0.70

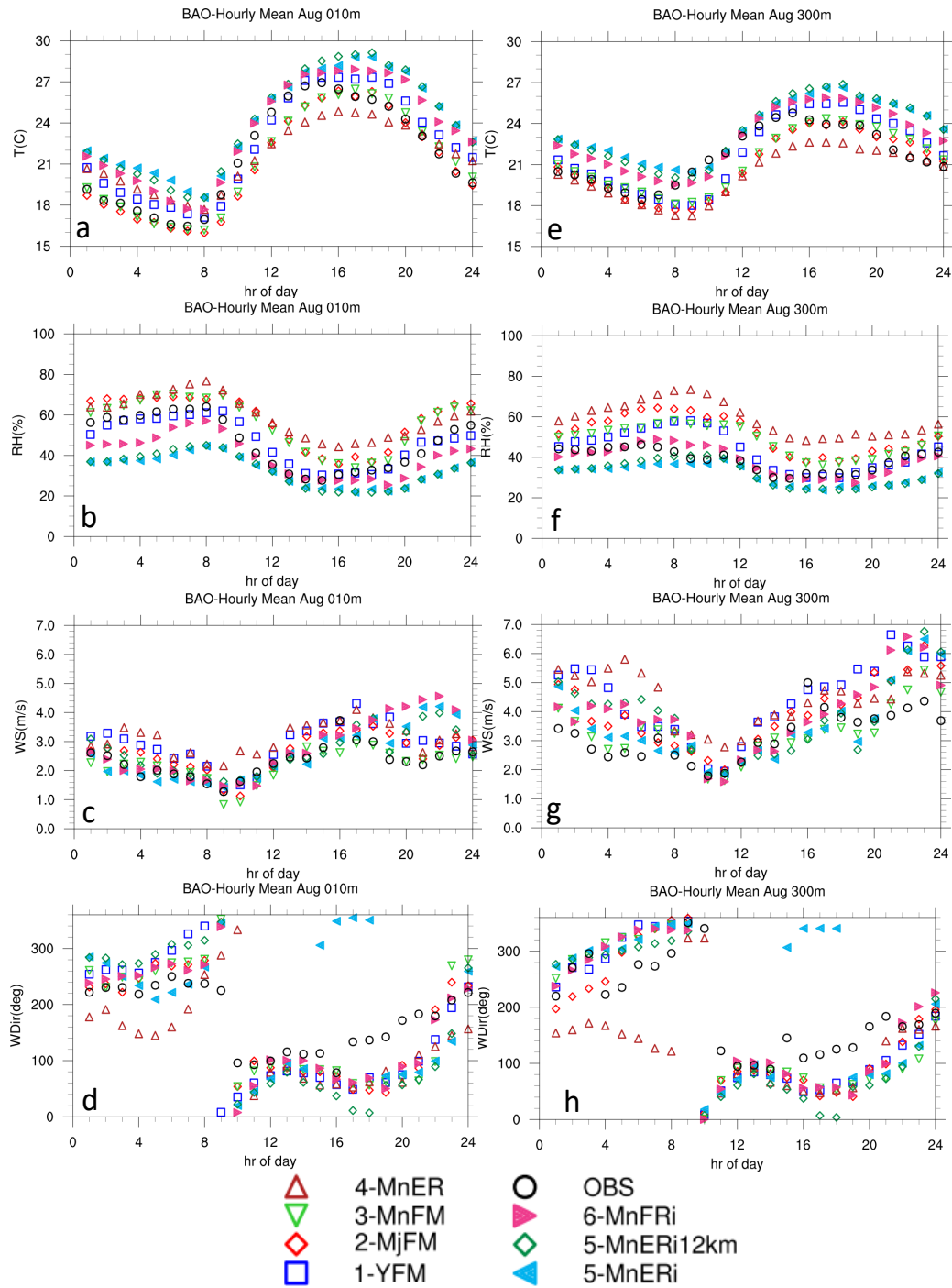


Figure SM 1. Average diurnal cycle of temperature (a, e), relative humidity (b, f), wind speed (c, g) and wind direction (d, h) for all tests and observation at BAO 10m and 300m. Averages are calculated for Aug 1 to 15, 2014.

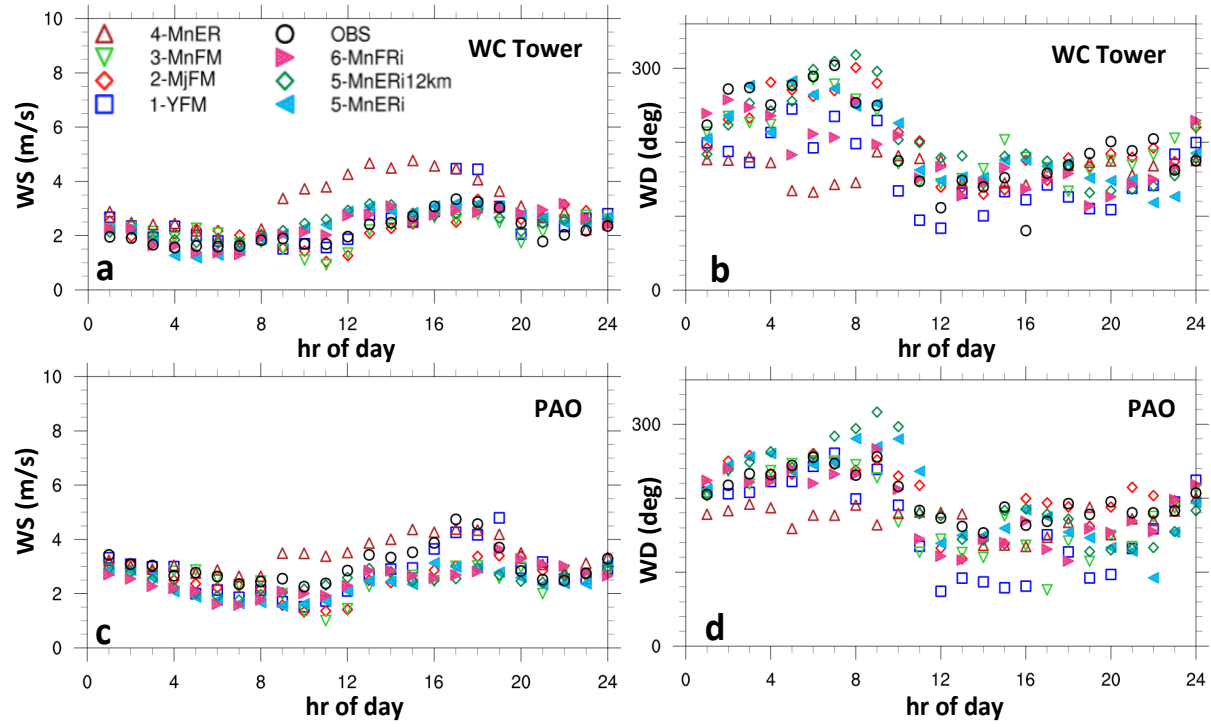


Figure SM 2. Average diurnal cycle of wind speed (WS) and direction (WD) at WC Tower and PAO sites. Averages are calculated for August 1 to 11, 2014.

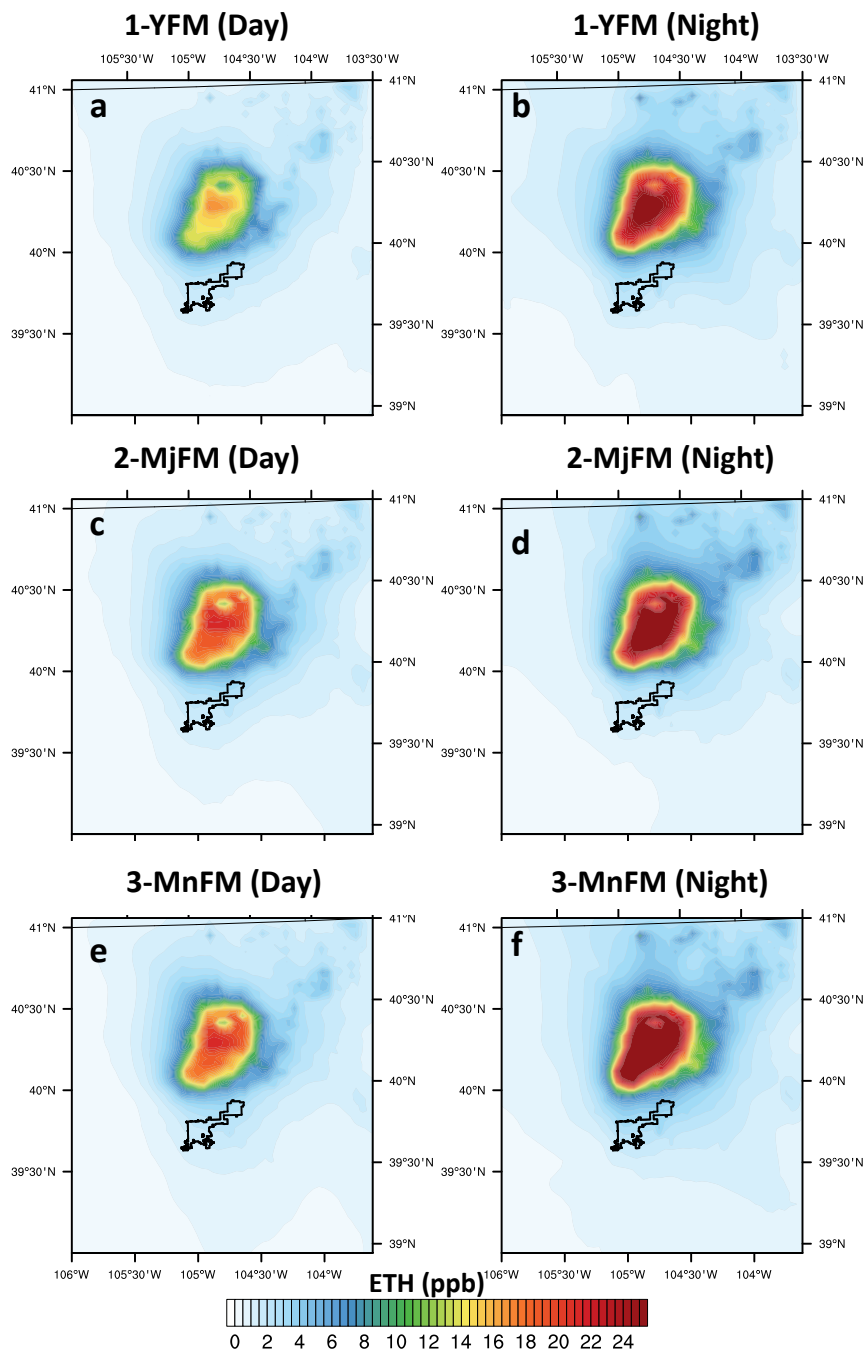


Figure SM 3. Surface ethane in sim 1 (1-YFM), sim 2 (2-MjFM), sim 3 (3-MnFm) averaged from August 1 to 15, 2014

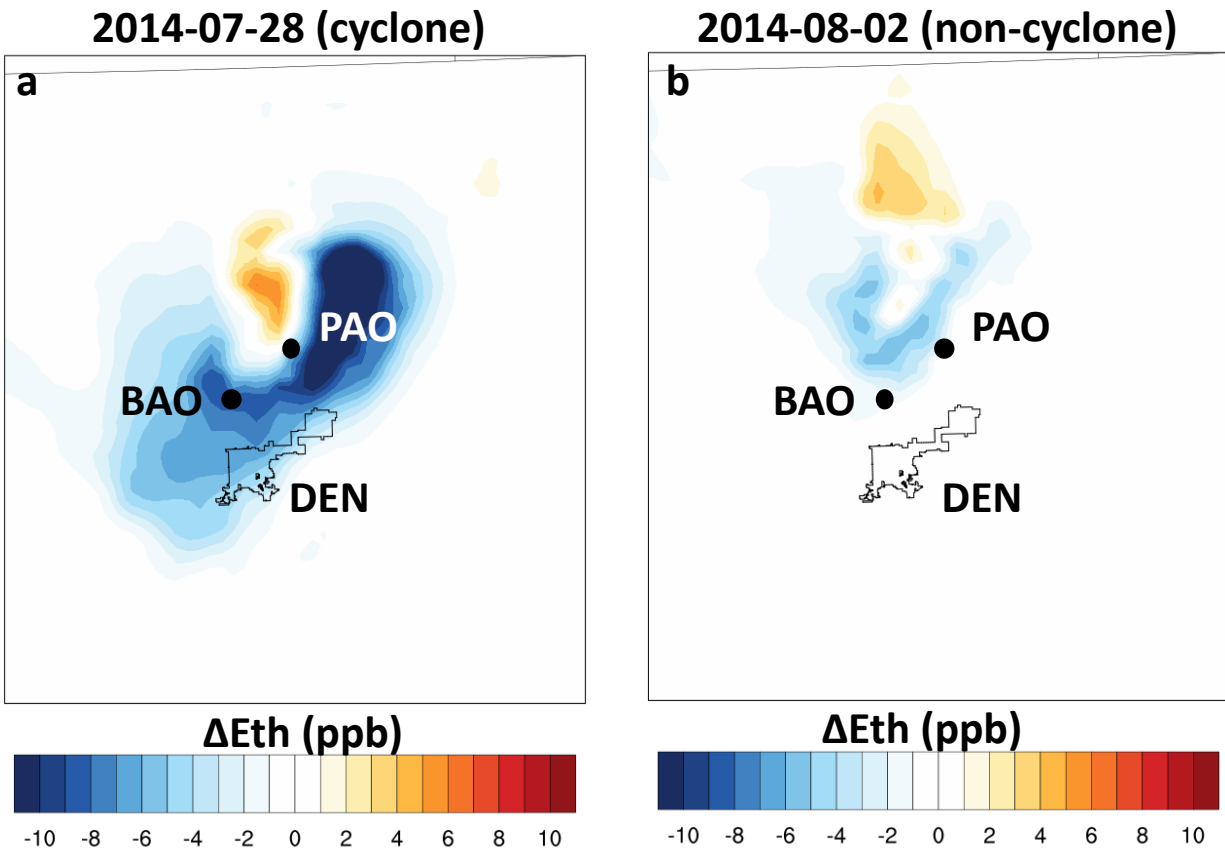


Figure SM 4. Difference in surface ethane concentration (ΔEth) between Met6 (6-MnFRi) and Met5 (5-MnERi) averaged during a) July 28, 2014 (Denver Cyclone) and b) August 2, 2014 (non-cyclone). The locations of BAO and PAO sites and Denver county are marked on the map.

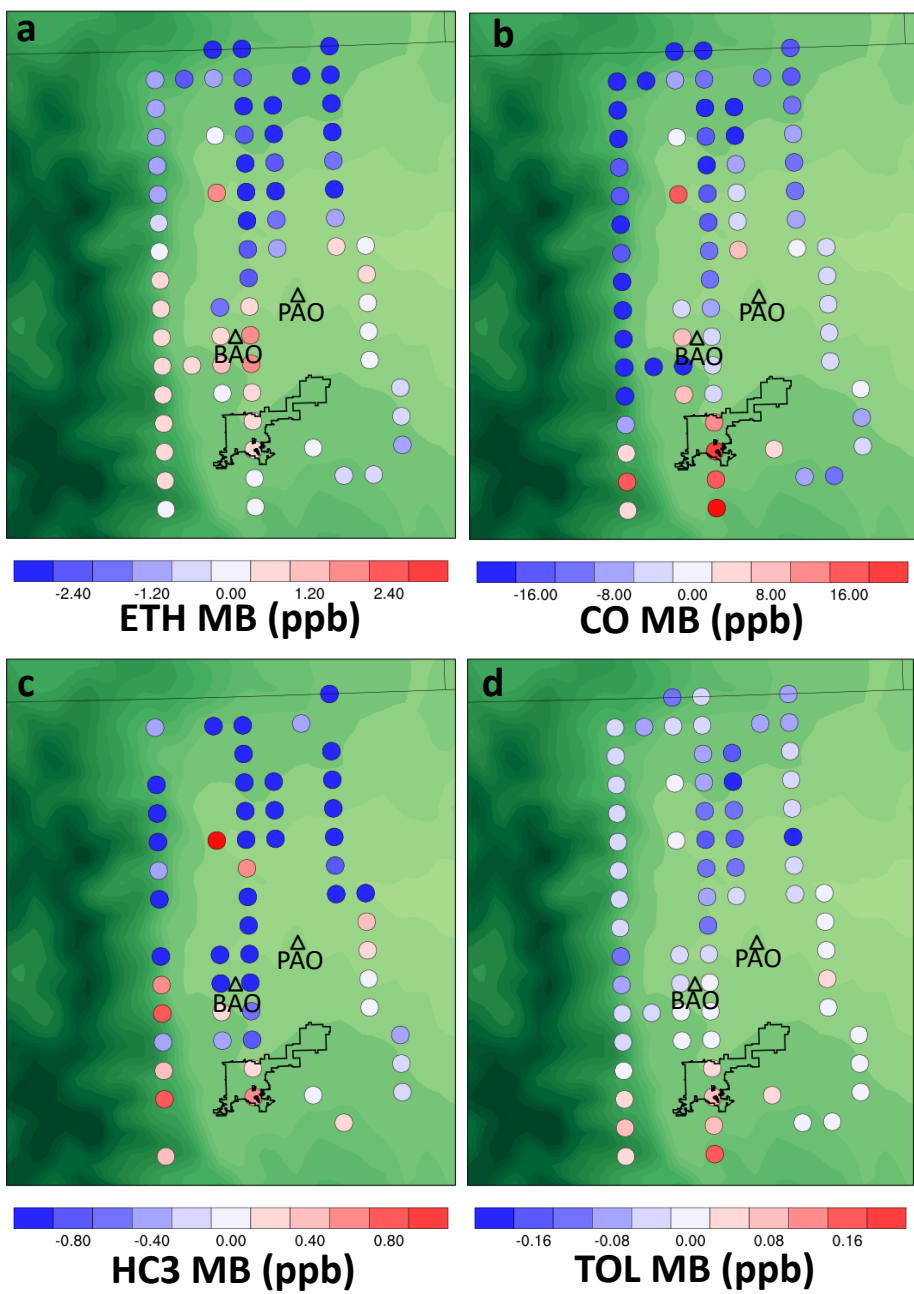


Figure SM 5. Mean biases of ethane (a), CO (b), HC3 (c), and TOL (d) in Em7 along the C130 PM flights limited to measurements below 1500m agl and grids with more than 4 measurement points. Outline of the Denver county and location of BAO and PAO sites are marked on the underlying terrain map.

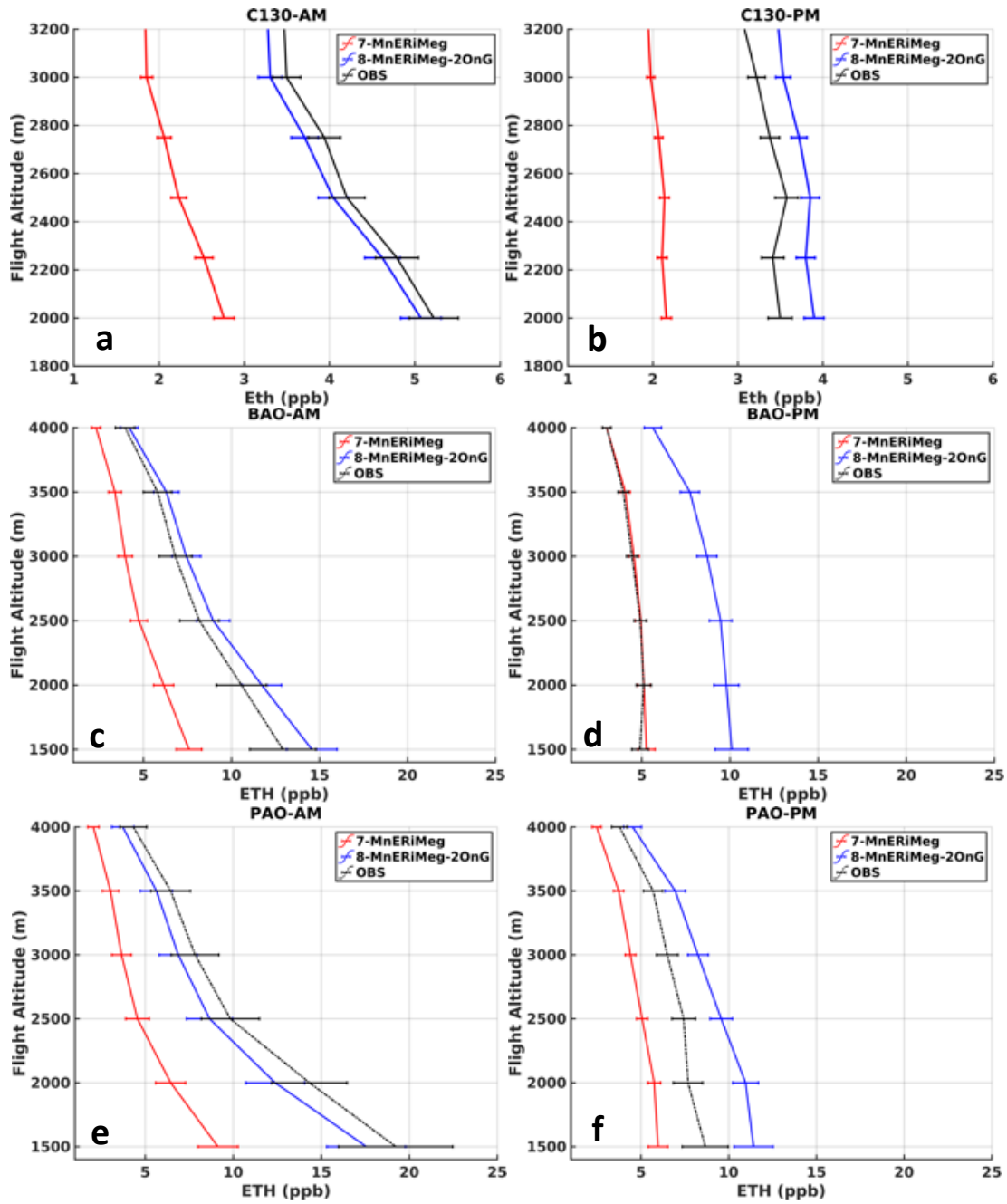


Figure SM 6. Sensitivity of ethane to oil and NG emission during C130-AM (a), C130-PM (b), P3-PAO AM (d), P3-PAO PM (c), P3-BAO AM (e), P3-BAO PM (f) averaged for August flights.