Supplemental information for:
A Direct sensitivity approach to predict hourly ozone resulting from compliance with the National Ambient Air Quality Standard

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DDM Background

DDM calculates spatially and temporally varying sensitivity coefficients which are computed as the partial derivative of the atmospheric diffusion equation with respect to the input of interest, Equation 1.

$$\boldsymbol{S_{ij}(t)} = \widetilde{\boldsymbol{P}}_{\boldsymbol{j}} \frac{\partial \boldsymbol{C_i(t)}}{\partial \boldsymbol{p_j}} = \widetilde{\boldsymbol{P}}_{\boldsymbol{j}} \frac{\partial \boldsymbol{C_i(t)}}{\partial(\boldsymbol{\epsilon_j} \widetilde{\boldsymbol{P}}_{\boldsymbol{j}})} = \frac{\partial \boldsymbol{C_i(t)}}{\partial \boldsymbol{\epsilon_j}} \qquad \text{Equation S-1}$$

S_{ij}, the normalized sensitivity coefficient (ppb / fractional change in p_j), represents the change in concentration of species i (C_i) for and incremental change in input parameter j (p_j). $\widetilde{P}_j$ (x,t) is the normalized input and ε_j is a scaling variable[1]. In general terms, the sensitivity coefficient tells us how a model output (ozone concentration) will change if a model input (emissions of NO_x or VOC) is perturbed. This first order sensitivity coefficient, $S_{ij}(t)$ is quite accurate for small perturbations, but can only be used to approximate a linear response. Second (and third) order derivatives can be taken to give higher order sensitivity coefficients, S^2_{ij} etc[2]. The sensitivities can be expressed such that a new concentration is estimating using modeled coefficients with the first three terms of the Taylor series expansion, Equation 2.

$$\boldsymbol{C}(+\Delta\boldsymbol{\epsilon}) = \boldsymbol{C}(\boldsymbol{0}) + \Delta\boldsymbol{\epsilon}\boldsymbol{S}(\boldsymbol{0}) + \frac{\Delta\boldsymbol{\epsilon}^{\boldsymbol{2}}}{\boldsymbol{2}}\boldsymbol{S}^{\boldsymbol{2}}(\boldsymbol{0}) + \ \ldots + \frac{\Delta\boldsymbol{\epsilon}^{\boldsymbol{n}}}{\boldsymbol{n}!}\boldsymbol{S}^{\boldsymbol{n}}(\boldsymbol{0}) + \boldsymbol{R}_{\boldsymbol{n+1}} \qquad \text{Equation S-2}$$

Here $\Delta\epsilon$ represents the relative change in emissions (for instance $\Delta\epsilon$ = -0.2 would be equivalent to reducing emissions by 20%), C(0) is the concentration under baseline conditions (no perturbation in emissions) and R_{n+1} is a remainder term.

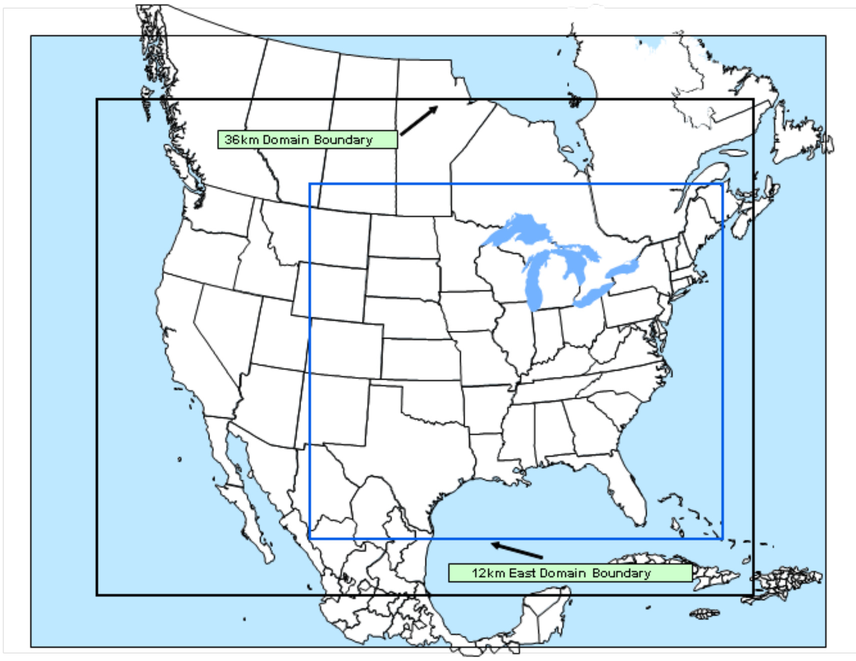
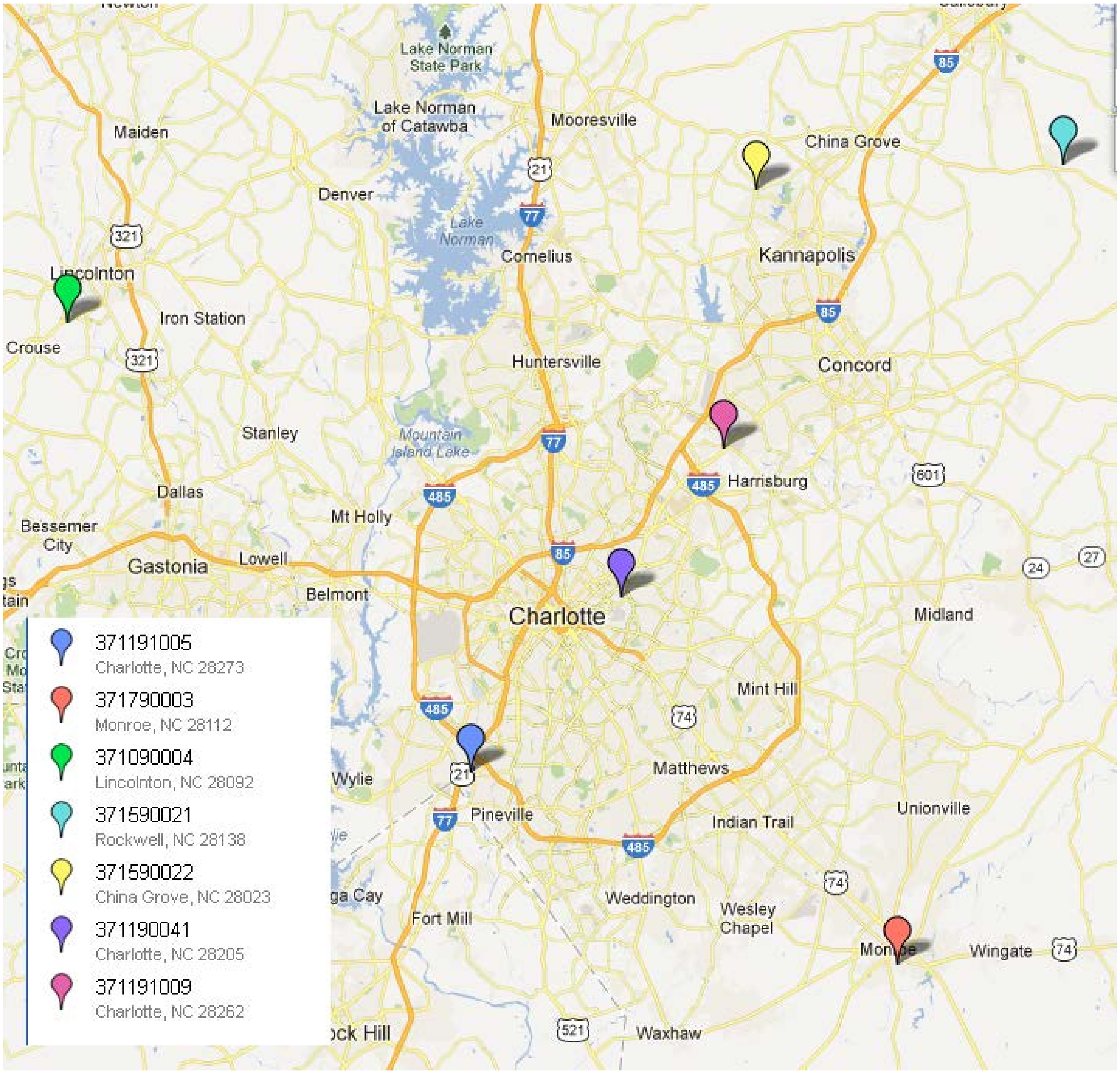


Figure S1. Map of the 36km continental US and 12km Eastern US modeling domains used in this work.



Figures S2. Map of Charlotte area monitoring sites evaluated in this work.

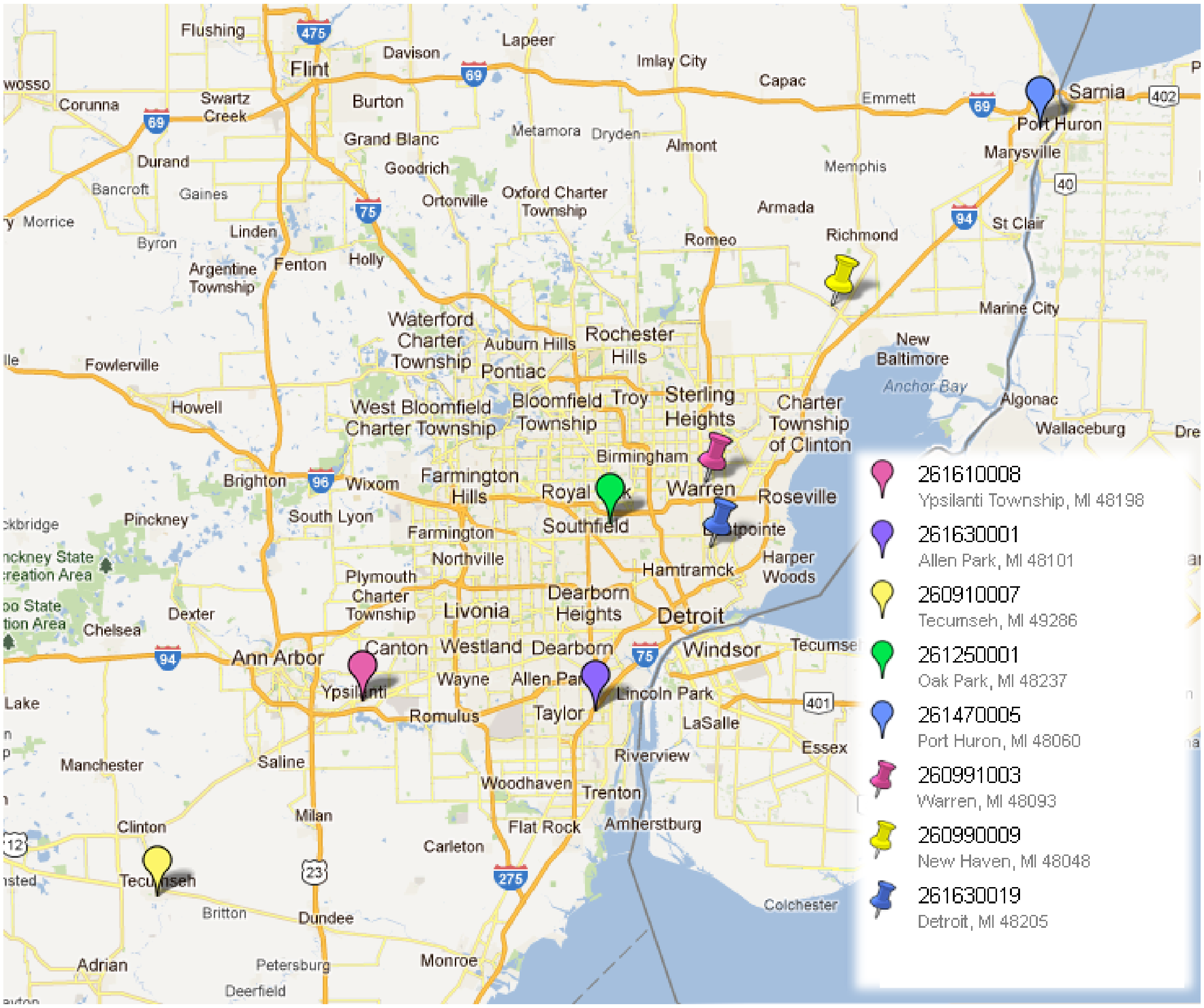


Figure S3. Map of Detroit area monitoring sites evaluated in this work.

Table S1. Summary of model performance statistics of modeled ozone compared to measured ozone at Charlotte area monitoring sites for July and August 2005.

Metric	Hourly ozone		8-hr daily maximum ozone	
	All values	➢ 60 ppb	All values	➢ 60 ppb
Number	10018	1482	422	167
Mean Obs (ppb)	32.8	73.2	56.1	72.9
Mean Mod (ppb)	46.0	71.2	60.9	73.1
Mean Bias (ppb)	13.2	-2.0	4.8	0.2
Mean Error (ppb)	15.8	8.7	7.9	6.5
Normalized mean bias (%)	40.3	-2.8	8.6	0.3
Normalized mean error (%)	48.3	11.9	14.1	9.0

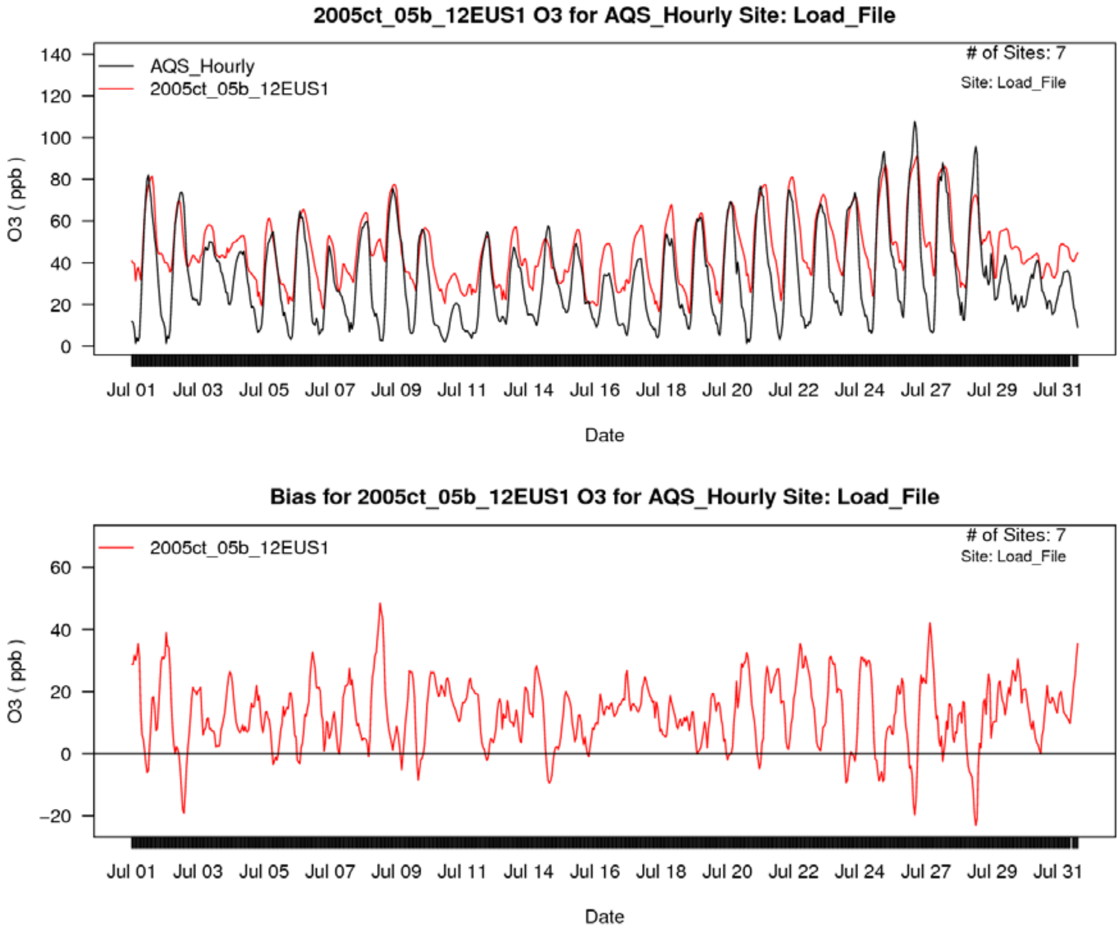


Figure S4. Time series of measured and modeled average hourly ozone concentrations at seven Charlotte area ozone monitors for July 2005.

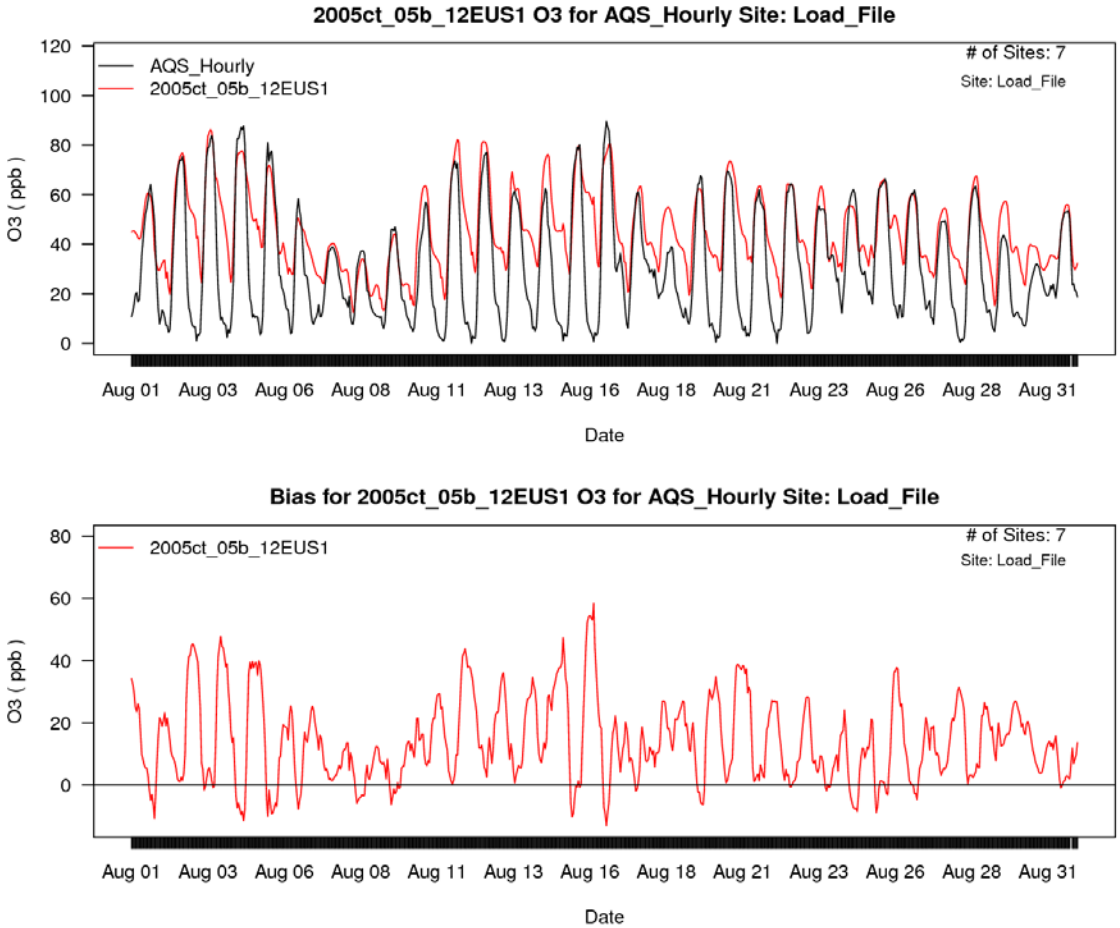


Figure S5. Time series of measured and modeled average hourly ozone concentrations at seven Charlotte area ozone monitors for August 2005.

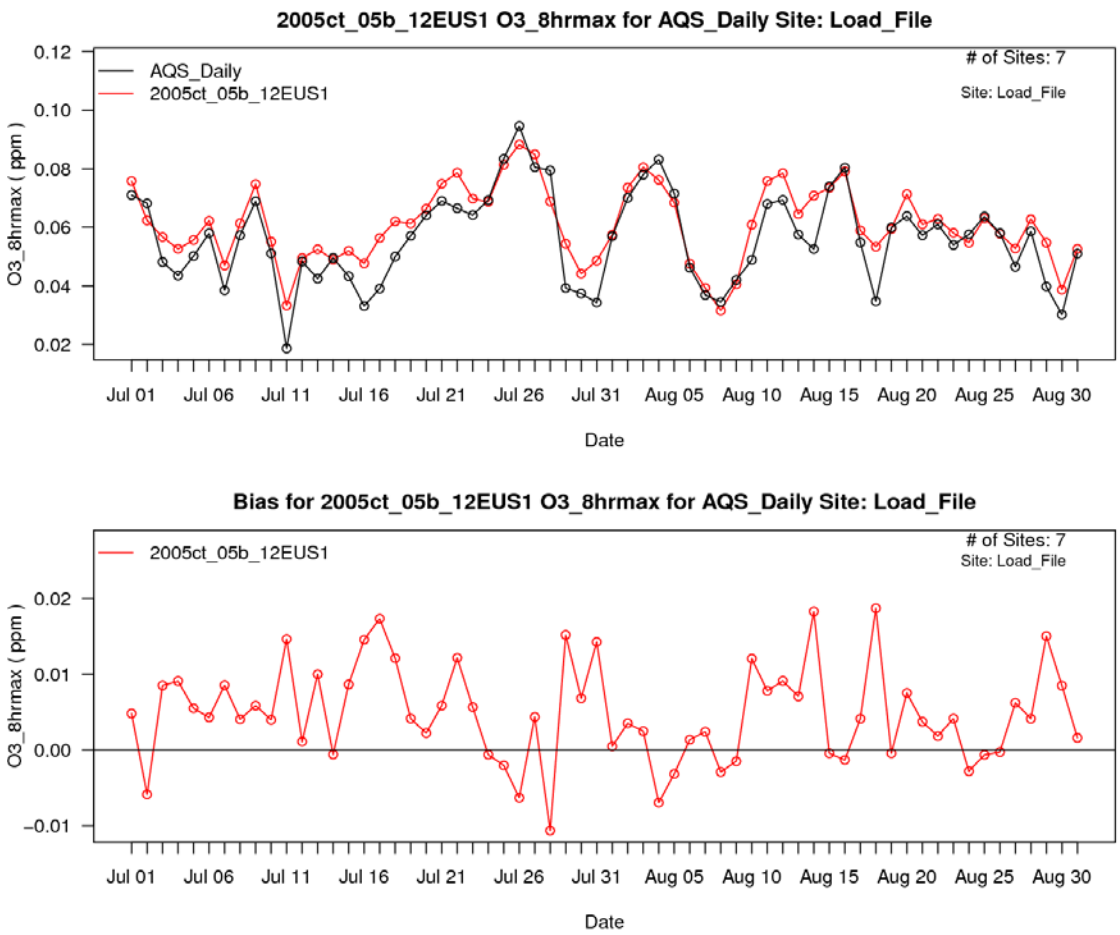


Figure S6. Time series of measured and modeled average 8-hr daily maximum ozone concentrations at seven Charlotte area ozone monitors for July and August 2005.

Table S2. Summary of model performance statistics of modeled ozone compared to measured ozone at Detroit area monitoring sites for July and August 2005.

Metric	Hourly ozone		8-hr daily maximum ozone	
	All values	➢ 60 ppb	All values	➢ 60 ppb
Number	11,329	1362	295	88
Mean Obs (ppb)	33.6	71	53.7	71.7
Mean Mod (ppb)	37.9	63.2	55.9	67.7
Mean Bias (ppb)	4.4	-7.8	2.2	-4.1
Mean Error (ppb)	11.2	11.7	7.8	7.7
Normalized mean bias (%)	13.0	-10.9	4.1	-5.7
Normalized mean error (%)	33.4	16.6	14.5	10.7

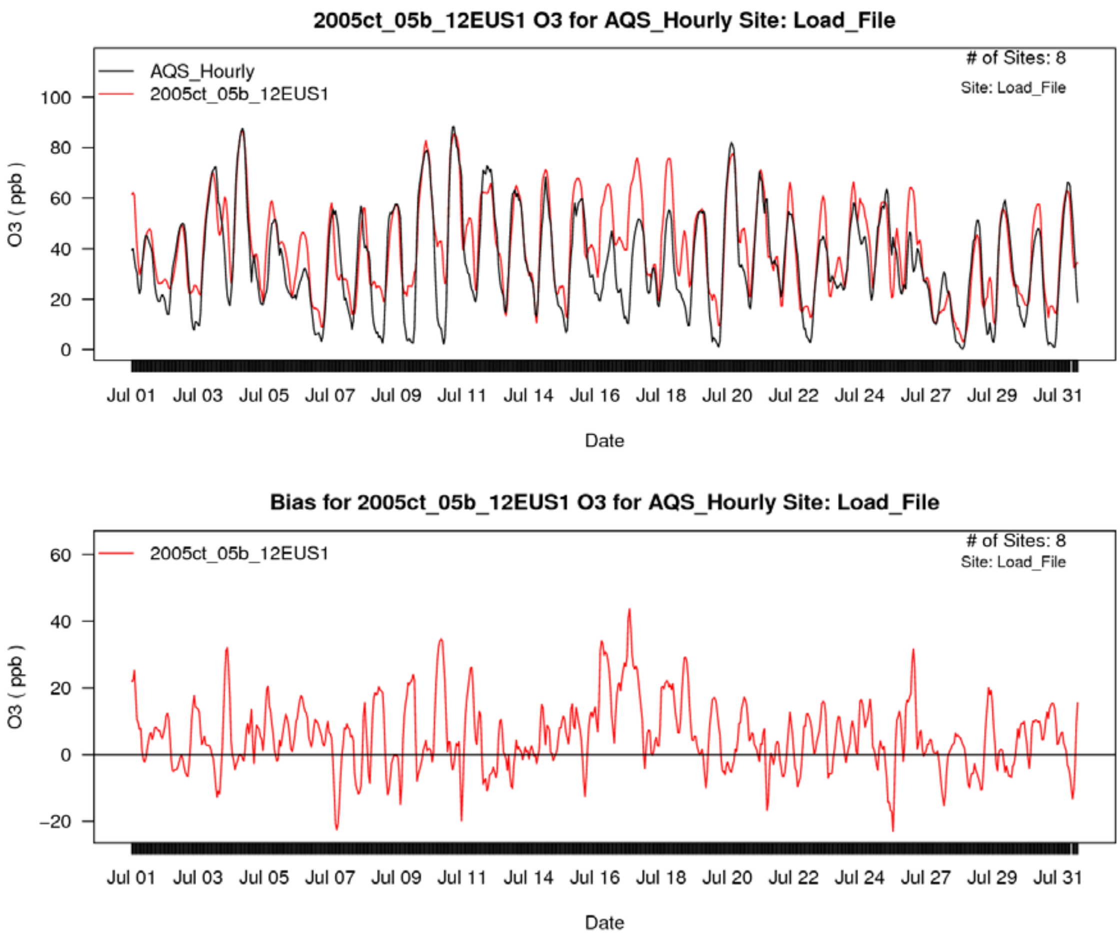


Figure S7. Time series of measured and modeled average hourly ozone concentrations at eight Detroit area ozone monitors for July 2005.

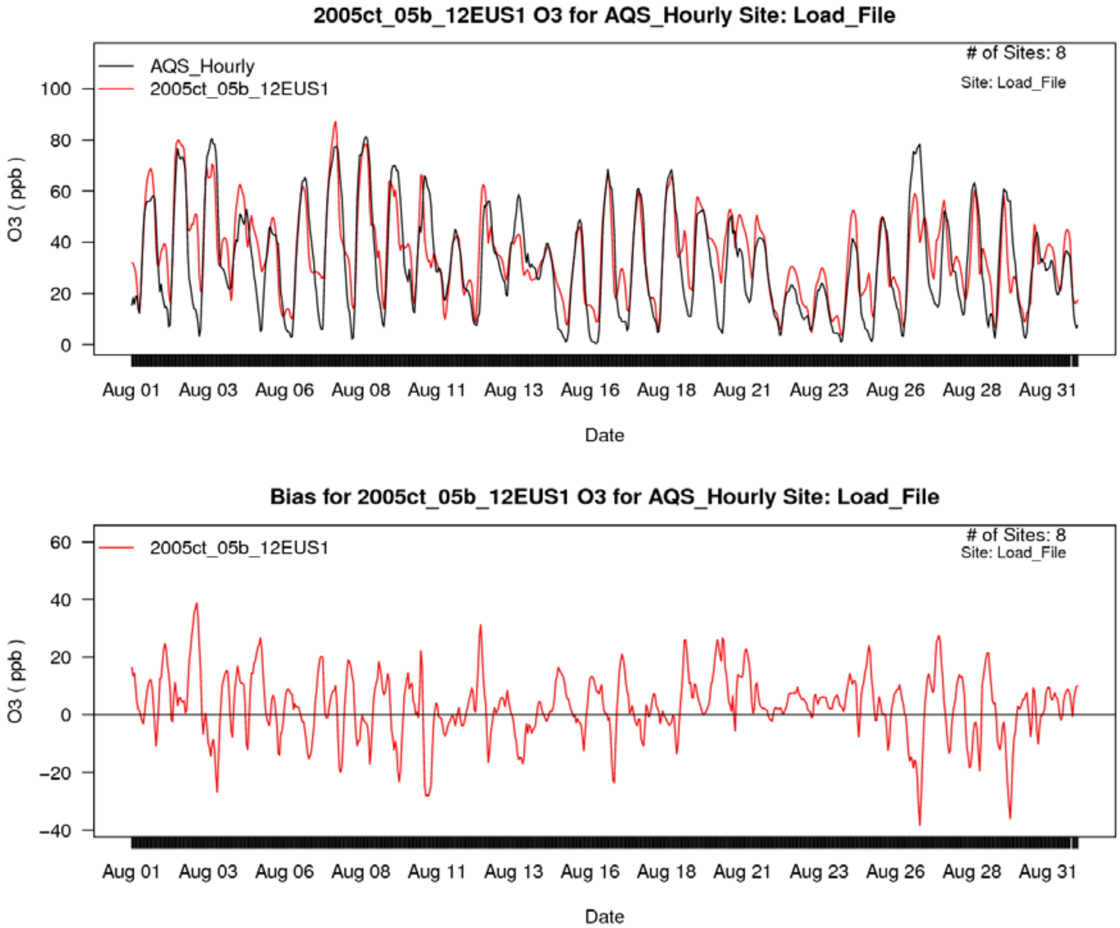


Figure S8. Time series of measured and modeled average hourly ozone concentrations at eight Detroit area ozone monitors for August 2005.

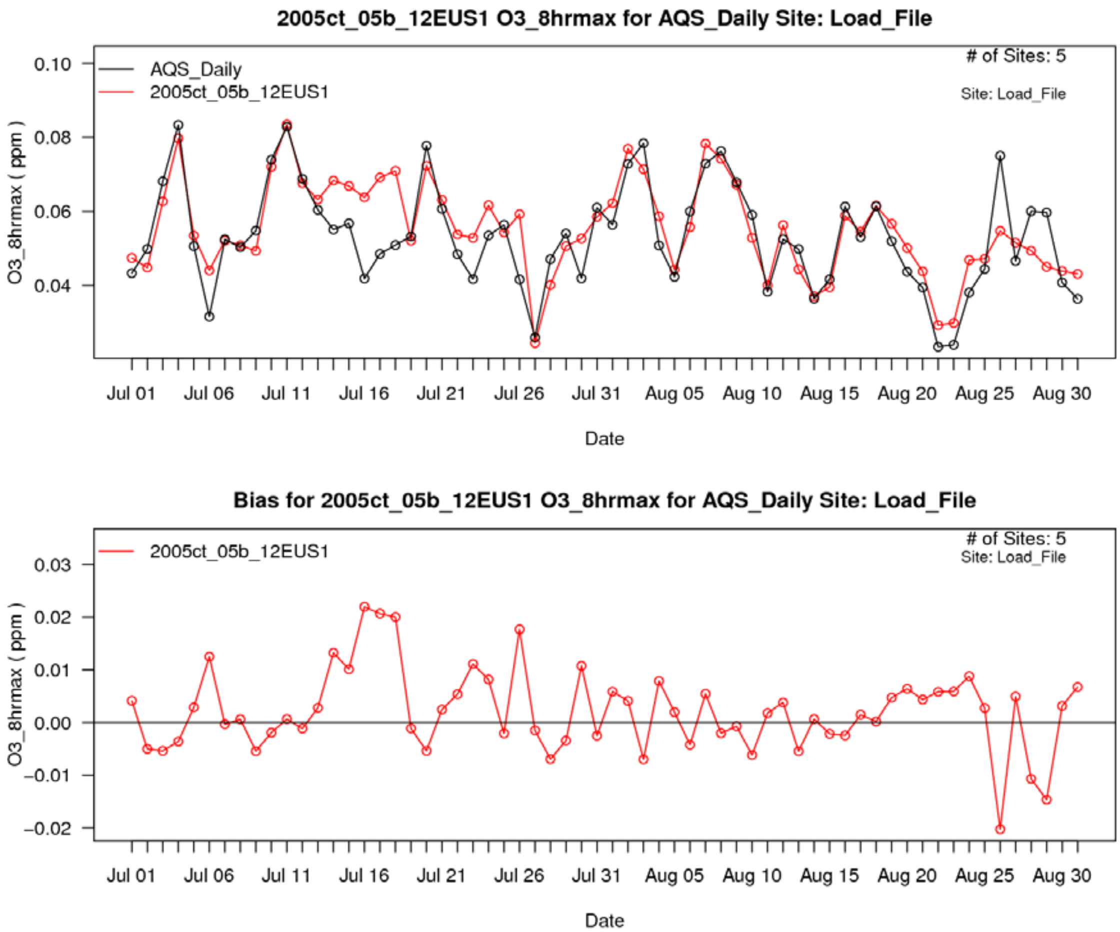


Figure S9. Time series of measured and modeled average 8-hr daily maximum ozone concentrations at eight Detroit area ozone monitors for July and August 2005.

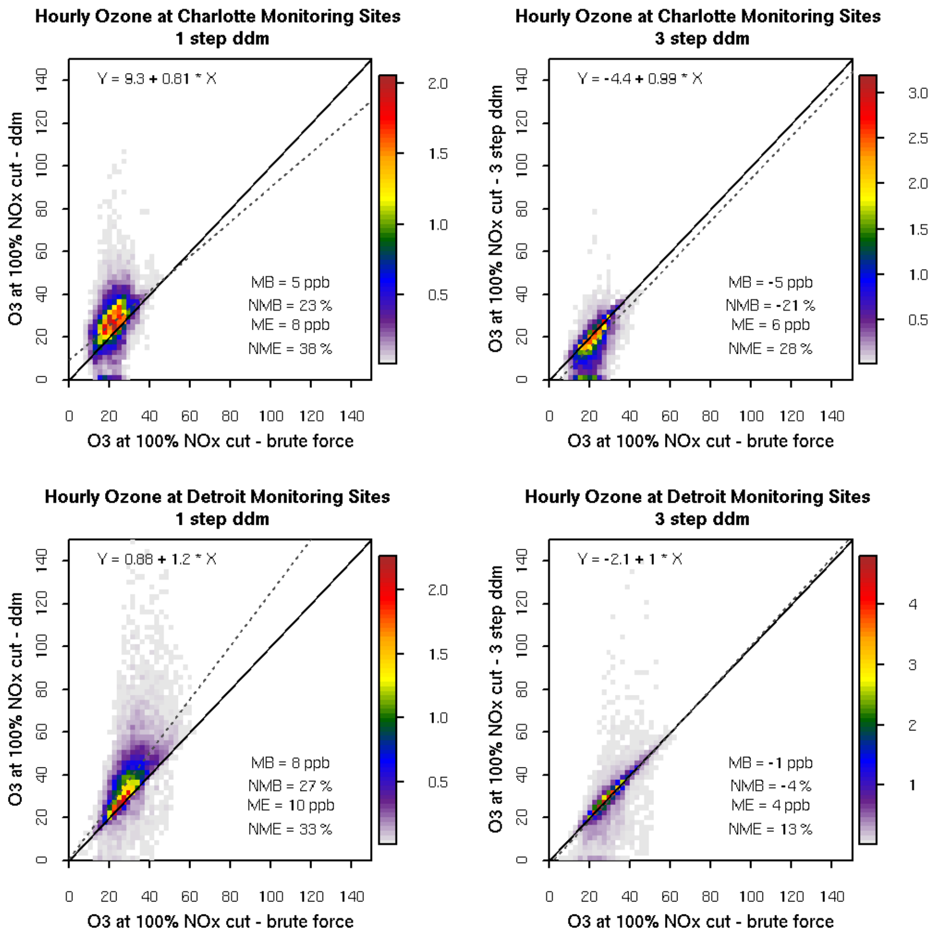


Figure S10. Density scatter plots comparing ozone predictions using DDM sensitivities to 100% NOx cuts to model predictions from runs with brute force emissions cuts at Charlotte (top) and Detroit (bottom) sites. Colors indicate the percentage of points that fall at each spot on the plot. One step DDM adjustment results are shown in left-hand plots and three step DDM adjustment results are shown in right-hand plots.

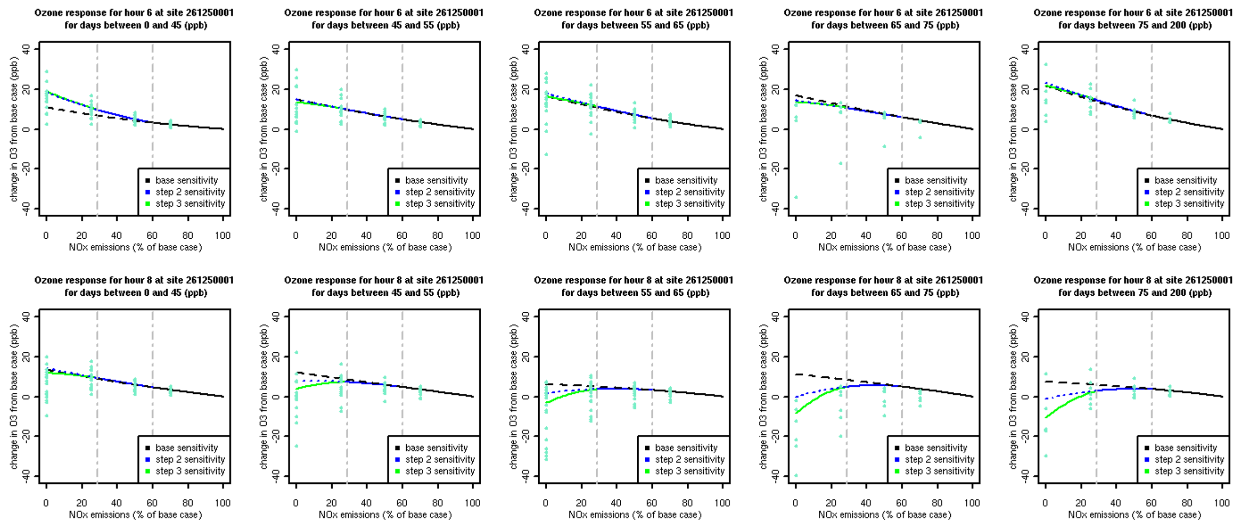


Figure S11. Depiction of 3-step DDM adjustment approach. Each panel shows the change in ozone over the entire range of NO_x reductions. The solid line shows the path followed for the 3-step DDM adjustment, while the dotted lines show changes in ozone that would be predicted if the based or step 2 sensitivities were used down to 0 NO_x emissions. Turquoise dots show change in ozone from model runs employing brute force NO_x emissions cuts on the days that are included in each bin for each monitoring site. Top panels show all five bins for Detroit site 25125001 at 6am. Bottom panels show all five bins for Detroit site 25125001 at 8am.

References:

1. Yang, Y. J.; Wilkinson, J. G.; Russell, A. G., Fast, direct sensitivity analysis of multidimensional photochemical models. *Environ. Sci. Technol.* **1997,** *31* (10), 2859-2868.
2. Hakami, A.; Odman, M. T.; Russell, A. G., High-order, direct sensitivity analysis of multidimensional air quality models. *Environ. Sci. Technol.* **2003,** *37* (11), 2442-2452.