Supplement of Earth Syst. Sci. Data, 15, 5261–5279, 2023
https://doi.org/10.5194/essd-15-5261-2023-supplement

Supplement of

Spatiotemporally resolved emissions and concentrations of styrene, benzene, toluene, ethylbenzene, and xylenes (SBTEX) in the US Gulf region

Chi-Tsan Wang et al.

Correspondence to: Bok H. Baek (bhbaek@gmail.com)

Supplementary document:

S1. Emission Data

We applied the USEPA's 2011 National Emission Inventory (NEI) Emissions Modeling Platform (EMP) version 6 (USEPA, 2021b), based on the official 2011 NEI with the SMOKE model system to generate the year 2012 gridded hourly emissions for the CAMx modeling. The following emission source types were processed: 1) area, 2) point, 3) mobile, and 4) biogenic source types. The area emission sources included the following emission sectors: fugitive dust (afdust), commercial marine vessels (cmv), non-point source oil and gas industry (np_oilgas), rail, agriculture (ag), agriculture fire (agfire), non-point (nonpt), and nonroad. The stationary point sources included stationary point electric generating units (ptegu), point source fire or wildfire (ptfire), point source from non-electric generating unit point source (ptnonipm, which include most industrial processes but exclude the "egu" and "oilgas" industry), and point source oil and gas (pt_oilgas) sectors. The details of these emission sectors can be found in the technical supporting document (TSD) from the 2011 NEI EMP (USEPA, 2021b). The SBTEX emission from original NEI are shown in Table S1.

The official USEPA's NEI reported the county-level annual or monthly total emissions of criteria air pollutants (CAPs), including carbon monoxide (CO), nitrogen dioxide (NOx), sulfur dioxide (SO2), particulate matter (PM), and VOC. The SMOKE model system processed these county total emissions inventories and generated the spatially gridded and chemically speciated hourly emission data of model species for the CTM. In this study, the reduced chemical mechanism, Carbon Bond 06 revision 4 (CB6r4) (Yarwood and Jung, 2010), is applied to simulate the chemistry process. The CB6 mechanism uses 22 surrogate VOC species to represent all VOC-related compounds in the atmosphere (Ramboll, 2020b), such as aldehyde (ALDX), formaldehyde (FORM), acetone (ACET), methane (CH4), methanol (MEOH), benzene (BENZ), and others. Thus, the CTM employs these surrogate model species and reduced chemical mechanisms to simulate the radical cycle reactions of HO_x (OH and HO_2) and NOx (NO and NO_2), the ozone formation process, and other secondary air pollutants like $PM_{2.5}$ (Steyn and Rao, 2010).

According to the USEPA SPECIATE database documents (Simon et al., 2010; USEPA, 2021a) and the chemical speciation tool (Ramboll, 2020b), the CB6 model species like TOL and XYL can be derived from the explicit SBTEX chemical species; however, they are not limited to SBTEX. In fact, hundreds of real VOC species similar in chemical structure to toluene and xylene are included in the surrogate species, TOL and XYL (Ramboll, 2020b). Thus, the model species can not represent the explicit xylenes and toluene chemical compounds while they share the names of these species in the CTM model.

S2. Air Quality Modeling

The Comprehensive Air Quality Model with Extensions version 7.0, CAMx7.0 (RAMBOLL 2020) was implemented in this study to predict the SBTEX concentrations in the atmosphere. The base model simulation period is from April 20^{th} to September 30^{th}, 2012 (April 20^{th} to April 30^{th} are spin-up dates). The evaluated meteorological data from WRF version 3.8 over the U.S. continental region are provided by the USEPA Support Center for Regulatory Atmospheric Modeling (SCRAM) (USEPA, 2022). They are converted to the CAMx-ready format using the WRFCAMx version 4.8.1 program developed by the CAMx development team (RAMBOLL, 2020a). The photodissociation coefficients are calculated by the Tropospheric Ultraviolet-Visible (TUV) Radiation Model (Madronich, 1987) with Ozone Monitoring Instrument (OMI) daily data (NASA, 2021). The USEPA daily hemisphere CMAQ model results are used to calculate the boundary condition and initial condition (Hogrefe et al., 2021). The chemical mechanism is Carbon Bond 06 revision 4 (CB6r4) (Ramboll, 2020b). Figure 1 in manuscript shows that the main model domain is 12 km × 12 km (blue rectangle), with flexi-nesting to 4 km × 4 km (red rectangle).

To accurately simulate the decay of individual SBTEX, the base CAMx modeling results are immediately processed through the reactive tracer (RTRAC) probing tool step in CAMx. The base model predicted hourly oxidant concentrations, such as ozone (O_3), hydroxyl radical (OH), and nitrate radical (NO_3); those hourly oxidant concentrations and dry/wet depositions were used to estimate the decay of SBTEX concentrations. Thus, it is critical to confirm that the CAMx model can reasonably represent the oxidants concentrations in the base modeling run. The CAMx predictions are statistically evaluated with the 87 ozone monitoring sites data in the 4 km × 4 km model domain. The base modeling ozone evaluation results over the simulation period are shown in Table S2. The evaluation indicators followed the USEPA's model evaluation guidance (USEPA, 2023, 2006). It should be noted that the purpose of the base model is not for the ozone State Implementation Plan (SIP) or ozone control, but to simulate the SBTEX concentrations using the RTRAC feature in the CAMx modeling system. Therefore, the ozone performance may not be as good as the ozone SIP modeling application over the nonattainment region, which is relatively smaller than our 4 km × 4 km domain. The base modeling is fairly performed well over the Gulf region states (Correlation Coefficient (R) ≥ 0.55). The ozone performances in Texas and Louisiana are close to the observed USEPA AQS stations (Texas: R = 0.79, NMB = 1%; Louisiana: R = 0.77, NMB = 11%).

Additionally, because our base modeling shares the same simulation period as the Texas Commission on Environmental Quality (TCEQ) 2012 Ozone SIP modeling application (TCEQ, 2016), we verified our modeling results with TCEQ's simulated OH radical-related model species, including ozone, NO_2, and formaldehyde over the Dallas and Houston region. The detailed comparisons are shown in the supplementary documents (Fig. S1) and indicate that both modeling applications share a similar, good modeling performance. For Ozone, R is 0.75 in Dallas and 0.84 in Houston; for NO_2, R is 0.58 in Dallas and 0.76 in Houston; and for Formaldehyde, R is 0.74 in Dallas and 0.78 in Houston. While the model domains, meteorological input data, gridded hourly emission patterns, and CAMx model versions are different between these two applications, our results support our conducting the Reactive Tracer probing process with the base model.

S3. Missing HAPs emission from np_oilgas and ptnonipm

S3.1 The Imputation HAPs Emission in Non-Point Oil and Gas Industry (np_oilgas)

Fig. S2 and Table S5 shows all adjusted emission of "np_oilgas" (state total: 5803 t yr^{-1}; model domain: 2912 t yr^{-1}) are from Texas. Within this sector, increases in STEX can be attributed to a specific SCC code. Table S6 shows details of emission behaviors emission in "np_oilgas" for the imputation emission. The significant toluene increases are associated with tank condensate (729 t yr^{-1}), crude oil loading (589 t yr^{-1}), well dehydrator (581 t yr^{-1}), oil well (115 t yr^{-1}) and gas well (61 t yr^{-1}) pneumatic devices in oil and gas exploration and production process. The xylenes increases are from oil and gas production processes such as oil well artificial lift (such as beam pump) (2,062 t yr^{-1}), produced water at oil well (426 t yr^{-1}), hydraulic fracturing engines (91 t yr^{-1}), and all processes at oil well (73 t yr^{-1}). The ethylbenzene imputations are from crude petroleum oil well pneumatic devices (162 t yr^{-1}), storage tanks (109 t yr^{-1}), gas well dehydrators (53 t yr^{-1}), on-shore oil well pneumatic pumps (34 t yr^{-1}) and produced water at the well (28 t yr^{-1}). The imputations of styrene in the "np_oilgas" sector are relatively small (1.9 t yr^{-1}), and most of them (69%) are from hydraulic fracturing engines. Other styrene are from engine combustion (0.47 t yr^{-1}, 24%) and artificial lift (0.13 t yr^{-1}, 0.4%).

Figure S2 shows the STEX-adjusted emission spatial distributions. In Fig. S2a, the largest toluene emission increase (19 t yr^{-1}) occurred near the industry in Beaumont, which has many industrial storage tanks. The largest xylenes increase (Fig. S2b), 35 t yr^{-1}, near Fort Worth city; the most significant ethylbenzene increase (Fig. S2c), 4 t yr^{-1}, is near Longview; the largest styrene increase (Fig. S2d) 0.02 t yr^{-1}, is located near Karnes, McMullen, Dewitt, and Live Oak County. Those STEX emissions happened in counties with many oil and gas industry storage or oil wells, reflecting the emission process patterns.

Figure S3a shows the temporal profile of “np_oilgas” adjusted emission by hourly total over the modeling domain from May 1st to Sep 30th. The oil and gas industry adjust emission reflects the flat industrial emission pattern caused by oil well or VOC evaporation processes, and the daytime hour (L.T. 9:00 to L.T. 16:00) has a higher (~ 15%) emission rate than night-time, and during the day there was an increase of 10 tons hr^{-1} compared to 65 tons hr^{-1} at night. This pattern is caused by styrene emission from fracturing engines and other engine combustion for the oil and gas well operation. Further, xylene contributes about 50% of STEX, and toluene contributes about 40% of STEX. That increase shows that the adjusted HAPs emission inventory in “np_oilgas” sector that relates to the oil well, storage tank, and the fracturing engine process is now being captured in adjusted inventory.

S3.2 The Imputation HAPs Emission in Point Source Non-Electricity Generation Unit (ptnonipm)

Fig. S4 and Table S5 shows all adjusted STEX emission of the industrial point source emission sector: “ptnonipm” (six states total, including F.L., TX, LA, MS, GA, and AL: 2315 t yr^{-1}; model domain total: 1,483 t yr^{-1}). Adjusted STEX emissions of “ptnonipm” have nearly half of that increase from styrene (741 t yr^{-1}) in Louisiana in the model domain.

Table S7 shows for "ptnonipm" that most of the styrene increases are from manufacturing processes of synthetic rubber dryers (374 t yr^{-1}), manufacturing fiberglass (134 t yr^{-1}), and polystyrene (130 t yr^{-1}). For toluene, increased emissions were from manufacturing pesticides (105 t yr^{-1}), paint evaporation (48 t yr^{-1}), landfill fugitive emission (23 t yr^{-1}), and chemical manufacturing and evaporation. The xylenes imputations were from operating wastewater (33 t yr^{-1}), water-base paint (29 t yr^{-1}), solvent-based paint (22 t yr^{-1}), coating (19 t yr^{-1}), and sealing (18 t yr^{-1}) in chemical evaporation processes. The ethylbenzene imputations were from the plastics production from the extruder (9 t yr^{-1}), operating water-base paint (7 t yr^{-1}), fugitive emission in chemical manufacturing (5 t yr^{-1}), utilizing solvent-base paint (5 t yr^{-1}), and wastewater treatment (4 t yr^{-1}).

Figure S4 shows the spatial distribution of STEX imputed emission for "ptnonipm." In this figure, the toluene (Fig S4a) shows the most considerable emission difference (103 tons yr^{-1}) happened near Baton Rouge, Louisiana. This adjusted emission in Baton Rouge is caused by producing pesticides. The most considerable xylenes difference (Fig. S4b), 21 t yr^{-1}, happened near Montgomery, Alabama. This xylenes adjusted emission is caused by the building material industry (painting and sealing process) in the southwest of Montgomery. The largest ethylbenzene difference (Fig. S4c), 5 t yr^{-1}, is at Birmingham, Alabama. This emission is near the steel industry. The largest styrene difference (Fig. S4d), 364 t yr^{-1}, is near Lake Charles in Louisiana. This emission is caused by the polymer industry that produces synthetic rubber or other polymers.

Figure S3b shows a temporal profile of “ptnonipm” adjusted emission by hourly total over the modeling domain from May 1st to Sep 30th. The “ptnonipm” change emission reflects a relatively flat industrial emission pattern caused by polymer manufacturing or VOC evaporation. The daytime hour (L.T. 9:00 to L.T. 16:00) has a higher (~ 15%) emission rate than night-time, and during the day there was an increase of 3 tons hr^{-1} compared to 19 tons hr^{-1} at night. This pattern is caused by xylene emission from operating wastewater and paint evaporation. Further, styrene contributes more than 50% of STEX, and toluene contributes about 25% of adjusted STEX. That increase shows that the adjusted HAPs emission inventory in the “ptnonipm” sector related to pesticide manufacturing, paint evaporation, building material industry, and steel industry, are now being captured in adjusted inventory for “ptnonipm”.

Other missing STEX emissions were found from the "pt_oilgas" and "ptegu" sectors, but they were insignificant with a total emission of STEX of 185 t yr^{-1}. (Figure S5, S6).

Tables:

Table S1. The Annual emission rates (metric tons yr^{-1}) of styrene, benzene, toluene, ethylbenzene, and xylene (SBTEX) for the National Emission Inventory (Base)

Emission Sectors[#]	Scenario	BENZENE tons yr^{-1}	TOLUENE tons yr^{-1}	XYLENES tons yr^{-1}	ETHYLBENZENE tons yr^{-1}	STYRENE tons yr^{-1}	Total tons yr^{-1}
agriculture fire (agfire)	Base	1,128	745	0	0	0	1,873
commercial marine vehicle (cmv)	Base	103	16	24	10	11	164
non-point source (nonpt)	Base	3,070	16,932	5,156	1,188	777	27,123
non-road vehicle (nonroad)	Base	4,752	13,506	14,265	2,682	171	35,376
on-road vehicle (onroad)	Base	10,495	**43,657**	**27,271**	**7,472**	309	**89,204**
fire emission (ptfire)	Base	**46,052**	10,909	4,355	0	0	61,316
rail road (rail)	Base	10	14	20	8	9	61
residential wood combustion (rwc)	Base	395	92	26	0	0	513
non-point oil gas industry (np_oilgas)	Base	5,421	1,596	3,097	228	1	10,343
electricity power plants unit (ptegu)	Base	277	129	58	34	7	505
point source emission other than electricity generation unit (ptnonipm)	Base	7,305	2,232	2,351	594	**2,170**	14,652
point source emission of oil and gas industry (pt_oilgas)	Base	510	252	169	29	1	961
Total	Base	79,518	90,080	56,792	12,245	3,456	242,091

Table S2. The base CAMx model evaluation for ozone over modeling states: MB (mean bias), ME (mean error), RMSE (root mean square error), FB (fractional bias), FE (fractional error), NMB (normalized mean bias), NME (normalized mean error), and R (correlation coefficient).

State (sites counts)	MB	ME	RMSE	FB	FE	NMB	NME	R
Alabama (4)	7.6	12.43	16.09	28.09	52.33	27.03	44.19	0.67
Florida (4)	7.27	11.37	15.07	24.44	35.93	22.47	35.14	0.55
Louisiana (24)	2.93	9.99	12.85	4.26	53.34	10.92	37.27	0.77
Mississippi (6)	8.92	11.97	15.15	30.99	39.52	27.87	37.4	0.67
Texas (49)	0.3	8.85	12.08	-3.05	45.98	1.02	30.4	0.79

Table S3. The physical parameters used in CAMx for reactive tracer modeling.

No.	Model species name	Henry's law constants (M/atm)	T fact K	Molecular weight	Rscale
1	**TOLUENE**	1.60E-01	4000	92.14	1
2	**XYLENES**	1.57E-01	5633	106.1	1
3	**ETHYLBENZ**	1.20E-01	5100	106	1
4	**STYRENE**	2.90E-01	4800	104.1	1
5	**BENZENE**	1.80E-01	4000	78	1

Table S4. The chemical reaction parameters used in CAMx for reactive tracer modeling.

Toxicants	Oxidants	A ($ppm^{-1}min^{-1}$)	Ea (K)	B	T_{ref} (K)
BENZENE	OH	3.39E+03	1.90E+02	0	300
TOLUENE	OH	2.66E+03	-3.40E+02	0	300
XYLENES	OH	2.41E+04	0.00E+00	0	300
XYLENES	NO_3	5.27E-01	0.00E+00	0	300
ETHYLBENZ	OH	1.03E+04	0.00E+00	0	300
ETHYLBENZ	NO_3	1.77E-01	0.00E+00	0	300
STYRENE	OH	8.56E+04	0.00E+00	0	300
STYRENE	O_3	2.51E-02	0.00E+00	0	300
STYRENE	NO_3	2.21E+03	0.00E+00	0	300

Table S5. The state total annual emissions (tons yr^{-1},) for the 2012 National Emission Inventory (Base), and the emission scenario adjusted in this study (Adj).

		Alabama		Florida		Georgia		Louisiana		Mississippi		Texas		Total by species		Sector Total	
Sector	Species	Base	Adj	Base	Adj	Base	Adj	Base	Adj	Base	Adj	Base	Adj	Base	Adj	Base	Adj
electricity power plants unit (ptegu)	toluene	24	+2.2 (9%)	51	+0.35 (0.7%)	13	+0.34 (3%)	17	+1.2 (7%)	10	+0.007 (0.06%)	70	**+3.6 (5%)**	185	+7.7 (4%)	535	+13 (2%)
	xylenes	10	+1.1 (11%)	26	+0.16 (0.6%)	5.2	+0.01 (0.2%)	11	+0.48 (4%)	6.4	+0.002 (0.03%)	225	**+1.7 (1%)**	283	+3.5 (1%)		
	styrene	4.7	+0.001 (0%)	5.8	+0.0003 (0.01%)	0.34	+0.001 (0.4%)	0.54	+0.02 (3%)	0.32	0	5.8	**+0.03 (0.5%)**	17	+0.05 (0.3%)		
	ethylbenzene	5.7	+0.55 (10%)	14	+0.08 (0.53%)	3.7	+0.01 (0.3%)	4.6	+0.29 (6%)	2.8	+0.001 (0.03%)	18	**+0.9 (5%)**	49	+1.8 (4%)		
point source emission of oil and gas industry (pt_oilgas)	toluene	13	+6.4 (48%)	2.5	+0.69 (28%)	4.2	+0.09 (2%)	27	+43 (160%)	17	+1.7 (10%)	183	**+45 (24%)**	246	+96 (39%)	437	+172 (39%)
	xylenes	11	+1.9 (17%)	1	+0.87 (86%)	1.2	+0.09 (7%)	14	**+30 (220%)**	9.1	+1.4 (15%)	107	+29 (27%)	143	+63 (44%)		
	styrene	1.1	+0.01 (1%)	0.04	+0.007 (18%)	0.2	0	0.5	**+0.21 (44%)**	0.33	+0.05 (14%)	5.2	+0.009 (0.2%)	7.4	+0.28 (4%)		
	ethylbenzene	3.2	+0.55 (18%)	0.40	+0.07 (17%)	0.4	+0.05 (12%)	4.7	**+5.7 (123%)**	3.6	+0.49 (14%)	29	+4.7 (16%)	41	+12 (28%)		
point source emission other than electricity generation unit (ptnonipm)	toluene	304	+51 (17%)	350	+126 (36%)	425	+167 (39%)	609	**+194 (32%)**	253	+48 (19%)	1,039	+88 (8%)	2,979	+674 (23%)	9,951	+2,315 (23%)
	xylenes	705	+84 (12%)	252	+79 (31%)	70	**+146 (208%)**	444	+47 (11%)	344	+39 (11%)	1,098	+115 (10%)	2,914	+510 (17%)		
	styrene	433	+24 (5%)	723	+48 (7%)	341	+122 (36%)	236	**+420 (178%)**	251	+35 (14%)	1,298	+362 (28%)	3,282	+1,012 (31%)		
	ethylbenzene	85	+12 (14%)	57	+19 (34%)	115	+26 (23%)	125	+14 (11%)	79	+15 (18%)	315	**+34 (10%)**	776	+119 (15%)		
non-point oil gas industry (np_oilgas)	toluene	133	0	12	0	0	0	1041	0	118	0	19	**+2,371 (12,605%)**	1,322	+2,371 (179%)	4,253	**+5,803 (136%)**
	xylenes	209	0	5.9	0	0	0	2441	0	46	0	28	**+2,938 (10,339%)**	2,731	+2,938 (108%)		
	styrene	0.08	0	0.0003	0	0	0	0.33	0	0.03	0	0.76	**+1.9 (251%)**	1.2	+1.9 (160%)		
	ethylbenzene	15	0	2	0	0	0	166	0	13	0	4.0	**+492 (12,423%)**	200	+492 (247%)		
State Total		1,957	+183 (9%)	1,501	+275 (18%)	979	461 (47%)	**5,141**	**+756 (15%)**	1,153	+141 (12%)	**4,445**	**+6,487 (146%)**	15,176	+8,303 (55%)	15,176	+8,303 (55%)

Table S6. The “np_oilgas” imputation STEX emissions by SCC code

Species	SCC	t yr^{-1}	Contribution	SCC description
toluene	2310021010	729	30.8%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Gas Production;Storage Tanks: Condensate
	2310011201	589	24.8%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Oil Production;Tank Truck/Railcar Loading: Crude Oil
	2310021400	581	24.5%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Gas Production;Gas Well Dehydrators
	2310010300	115	4.8%	Industrial Processes;Oil and Gas Exploration and Production;Crude Petroleum;Oil Well Pneumatic Devices
	2310021300	61	2.6%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Gas Production;Gas Well Pneumatic Devices
xylenes	2310000330	2,062	70.2%	Industrial Processes;Oil and Gas Exploration and Production;All Processes;Artificial Lift
	2310000550	426	14.5%	Industrial Processes;Oil and Gas Exploration and Production;All Processes;Produced Water
	2310000660	91	3.1%	Industrial Processes;Oil and Gas Exploration and Production;All Processes;Hydraulic Fracturing Engines
	2310010300	73	2.5%	Industrial Processes;Oil and Gas Exploration and Production;Crude Petroleum;Oil Well Pneumatic Devices
	2310011000	62	2.1%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Oil Production;Total: All Processes
styrene	2310000660	1	69.0%	Industrial Processes;Oil and Gas Exploration and Production;All Processes;Hydraulic Fracturing Engines
	2310021302	0.47	24.0%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Gas Production;Natural Gas Fired 4Cycle Rich Burn Compressor Engines 50 To 499 HP
	2310000330	0.13	6.6%	Industrial Processes;Oil and Gas Exploration and Production;All Processes;Artificial Lift
	2310021202	0.01	0.4%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Gas Production;Natural Gas Fired 4Cycle Lean Burn Compressor Engines 50 To 499 HP
ethylbenzene	2310010300	162	33.0%	Industrial Processes;Oil and Gas Exploration and Production;Crude Petroleum;Oil Well Pneumatic Devices
	2310021010	109	22.1%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Gas Production;Storage Tanks: Condensate
	2310021400	53	10.8%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Gas Production;Gas Well Dehydrators
	2310111401	34	7.0%	Industrial Processes;Oil and Gas Exploration and Production;On-Shore Oil Exploration;Oil Well Pneumatic Pumps
	2310000550	28	5.8%	Industrial Processes;Oil and Gas Exploration and Production;All Processes;Produced Water

Table S7. The “ptnonipm” imputation STEX emissions by SCC code

Species	SCC	ton yr^{-1}	Contribution	SCC description
toluene	30102601	105	15.1%	Industrial Processes;Chemical Manufacturing;Pesticides;Other Not Classified
	50100701	48	7.0%	Chemical Evaporation;Surface Coating Operations;Surface Coating Application - General;Paint: Solvent-base
	30588801	23	3.3%	Waste Disposal;Solid Waste Disposal - Government;Landfill Dump;Fugitive Emissions
	50100402	21	3.0%	Industrial Processes;Petroleum Industry;Fugitive Emissions;Specify in Comments Field
	33000199	20	2.9%	Industrial Processes;Miscellaneous Manufacturing Industries;Miscellaneous Industrial Processes;Other Not Classified
xylenes	30700121	33	6.2%	Industrial Processes;Pulp and Paper and Wood Products;Sulfate (Kraft) Pulping;Wastewater: General
	40200201	29	5.5%	Petroleum and Solvent Evaporation;Surface Coating Operations;Surface Coating Application - General;Paint: Water-base
	40200101	22	4.2%	Chemical Evaporation;Surface Coating Operations;Surface Coating Application - General;Paint: Solvent-base
	40202501	19	3.6%	Chemical Evaporation;Surface Coating Operations;Miscellaneous Metal Parts;Coating Operation
	40201726	18	3.4%	Chemical Evaporation;Surface Coating Operations;Metal Can Coating;End Sealing Compound
styrene	30102609	374	36.4%	Industrial Processes;Chemical Manufacturing;Synthetic Rubber (Manufacturing Only);Dryers
	30501215	134	13.0%	Industrial Processes;Mineral Products;Fiberglass Manufacturing;Curing Oven (Textile-type Fiber)
	30800901	130	12.6%	Industrial Processes;Rubber and Miscellaneous Plastics Products;Plastic Miscellaneous Products;Polystyrene: General
	30800730	65	6.3%	Industrial Processes;Rubber and Miscellaneous Plastics Products;Fiberglass Resin Products;Mechanical Resin Application: (non-vapor-suppressed)
	30101809	45	4.4%	Industrial Processes;Chemical Manufacturing;Plastics Production;Extruder
ethylbenzene	30101809	9	7.3%	Industrial Processes;Chemical Manufacturing;Plastics Production;Extruder
	40200201	7	6.0%	Chemical Evaporation;Surface Coating Operations;Surface Coating Application - General;Paint: Water-base
	30188801	5	4.1%	Industrial Processes;Chemical Manufacturing;Fugitive Emissions;General
	40200101	5	3.8%	Chemical Evaporation;Surface Coating Operations;Surface Coating Application - General;Paint: Solvent-base
	30600508	4	3.2%	Industrial Processes;Petroleum Industry;Wastewater Treatment;Oil/Water Separator

Table S8. The average concentration comparison between observational sites (Obs) and adjustment case model result (Adj)

					benzene (ppb)		toluene (ppb)		xylenes (ppb)		ethylbenzene (ppb)		styrene (ppb)		SBTEX (ppb)	
Group	Site ID	FIPs	Lat	Lon	Obs	Adj	Obs	Adj	Obs	Adj	Obs	Adj	Obs	Adj	Obs	Adj
Airport	481210034	48121	33.21	-97.19	0.131	0.088	0.149	0.207	0.080	0.105	0.023	0.021	0.003	0.001	0.385	0.422
Airport	481830001	48183	32.37	-94.71	0.500	0.152	0.272	0.116	0.158	0.073	0.037	0.016	0.022	0.009	0.989	0.366
Airport	483390078	48339	30.35	-95.42	0.194	0.132	0.277	0.264	0.135	0.118	0.043	0.032	0.022	0.064	0.671	0.610
Airport	484393009	48439	32.98	-97.06	0.129	0.128	0.142	0.350	0.075	0.153	0.027	0.038	0.007	0.003	0.380	0.671
Industry	220050004	22005	30.22	-90.96	0.355	0.395	0.289	0.321	0.165	0.101	0.049	0.033	0.032	0.031	0.890	0.881
Industry	220330009	22033	30.46	-91.17	0.272	0.488	0.388	0.545	0.240	0.211	0.063	0.060	0.034	0.092	0.998	0.910
Industry	480391003	48039	29.01	-95.39	0.161	0.122	0.172	0.099	0.087	0.046	0.027	0.012	0.006	0.007	0.452	0.285
Industry	481670005	48167	29.38	-94.93	0.421	0.182	0.291	0.108	0.277	0.075	0.089	0.017	0.013	0.007	1.091	0.389
Industry	482010024	48201	29.9	-95.32	0.270	0.248	0.373	0.648	0.314	0.255	0.091	0.067	0.027	0.048	1.075	1.266
Industry	482010036	48201	29.77	-95.1	0.940	1.049	0.741	0.477	0.575	0.309	0.155	0.115	0.126	0.081	2.536	2.031
Industry	482010057	48201	29.73	-95.23	1.248	0.820	0.739	0.762	0.462	0.343	0.155	0.101	0.055	0.142	2.661	2.168
Industry	482010058	48201	29.77	-95.03	0.360	0.980	0.345	0.408	0.266	0.294	0.056	0.097	0.020	0.059	1.047	1.838
Industry	482010061	48201	29.61	-95.01	0.461	0.260	0.323	0.182	0.164	0.090	0.067	0.024	0.055	0.013	1.070	0.570
Industry	482010069	48201	29.7	-95.26	0.204	0.488	0.415	0.622	0.278	0.278	0.076	0.077	0.114	0.103	1.087	1.568
Industry	482010307	48201	29.71	-95.25	0.334	0.531	0.469	0.698	0.412	0.310	0.124	0.086	0.099	0.116	1.438	1.742
Industry	482010617	48201	29.82	-94.99	0.194	0.551	0.299	0.245	0.120	0.184	0.027	0.052	0.006	0.028	0.646	1.060
Industry	482010803	48201	29.76	-95.17	0.284	0.811	0.882	0.514	0.304	0.260	0.062	0.089	0.052	0.070	1.583	1.744
Industry	482011015	48201	29.76	-95.08	1.064	0.912	1.266	0.393	0.457	0.263	0.108	0.099	0.096	0.065	2.991	1.667
Industry	482011035	48201	29.73	-95.25	0.252	0.579	0.498	0.666	0.338	0.299	0.080	0.086	0.059	0.100	1.228	1.730
Industry	482011039	48201	29.67	-95.12	0.222	0.430	0.335	0.384	0.201	0.173	0.042	0.047	0.015	0.047	0.815	1.034
Industry	482011049	48201	29.71	-95.22	0.420	0.671	0.569	0.760	0.341	0.341	0.108	0.095	0.023	0.151	1.462	2.019
Industry	482015504	48201	29.7	-95.11	0.383	0.533	0.638	0.411	0.348	0.185	0.093	0.054	0.043	0.058	1.503	1.240
Industry	482016000	48201	29.68	-95.25	0.232	0.370	0.529	0.553	0.397	0.244	0.088	0.064	0.033	0.089	1.280	1.319
Industry	482450009	48245	30.03	-94.07	0.207	0.437	0.228	0.257	0.177	0.120	0.029	0.037	0.002	0.010	0.642	0.861
Industry	482450011	48245	29.89	-93.99	0.558	0.333	0.240	0.105	0.227	0.050	0.052	0.017	0.007	0.014	1.084	0.452
Industry	482450014	48245	29.96	-93.89	0.448	0.246	1.045	0.089	0.213	0.040	0.065	0.013	0.010	0.052	1.781	0.441
Industry	482450017	48245	29.98	-93.95	0.335	0.373	0.214	0.170	0.175	0.081	0.056	0.032	0.090	0.078	0.870	0.734
Industry	482450018	48245	29.94	-94	0.312	0.406	0.208	0.181	0.158	0.085	0.053	0.034	0.012	0.042	0.742	0.748
Industry	482450019	48245	29.89	-93.97	0.393	0.483	0.206	0.142	0.152	0.060	0.054	0.021	0.012	0.020	0.817	0.726
Industry	482451035	48245	29.97	-94.01	0.208	0.373	0.291	0.169	0.209	0.080	0.044	0.031	0.010	0.041	0.761	0.692
Industry	482451050	48245	30.06	-94.09	0.315	0.548	0.276	0.343	0.250	0.156	0.074	0.046	0.014	0.010	0.929	1.103
Rural	220330013	22033	30.7	-91.05	0.168	0.135	0.124	0.158	0.065	0.065	0.025	0.019	0.026	0.007	0.409	0.385
Rural	220470009	22047	30.22	-91.31	0.184	0.175	0.200	0.135	0.119	0.045	0.033	0.014	0.020	0.012	0.556	0.382
Rural	481211007	48121	33.04	-97.13	0.082	0.088	0.199	0.250	0.081	0.114	0.012	0.026	0.001	0.002	0.375	0.480
Rural	481390016	48139	32.48	-97.02	0.102	0.074	0.102	0.125	0.056	0.064	0.020	0.014	0.003	0.002	0.282	0.279
Rural	481391044	48139	32.17	-96.87	0.110	0.043	0.095	0.064	0.056	0.027	0.020	0.008	0.008	0.001	0.290	0.142
Rural	481671034	48167	29.25	-94.86	0.121	0.070	0.098	0.047	0.035	0.026	0.018	0.006	0.000	0.001	0.273	0.151
Rural	482010029	48201	30.03	-95.67	0.217	0.104	0.223	0.320	0.130	0.129	0.041	0.034	0.011	0.004	0.622	0.590
Rural	482030002	48203	32.66	-94.16	0.279	0.100	0.197	0.084	0.102	0.077	0.025	0.013	0.037	0.002	0.640	0.276
Rural	482311006	48231	33.15	-96.11	0.123	0.042	0.122	0.077	0.062	0.032	0.023	0.009	0.004	0.001	0.333	0.160
Rural	482511008	48251	32.46	-97.16	0.113	0.061	0.109	0.109	0.049	0.069	0.023	0.012	0.005	0.001	0.298	0.252
Rural	482570005	48257	32.56	-96.31	0.110	0.042	0.077	0.081	0.038	0.034	0.016	0.009	0.000	0.001	0.242	0.167
Urban	481130069	48113	32.82	-96.86	0.081	0.168	0.239	0.521	0.332	0.217	0.054	0.058	0.003	0.005	0.708	0.969
Urban	482010055	48201	29.69	-95.49	0.178	0.200	0.213	0.706	0.157	0.275	0.048	0.072	0.011	0.006	0.606	1.260
Urban	484391018	48439	32.72	-97.1	0.144	0.119	0.288	0.364	0.128	0.191	0.028	0.041	0.033	0.005	0.620	0.721
Urban	484391062	48439	32.65	-97.2	0.087	0.107	0.225	0.343	0.086	0.194	0.022	0.035	0.014	0.005	0.434	0.685

Table S9

(a)

	Correlation Coefficient (R)					
Month	xylene	toluene	styrene	ethylbenzene	BENZ	SBTEX
1	0.7	0.59	0.73	0.71	0.52	0.73
2	0.71	0.63	0.53	0.67	0.51	0.72
3	0.68	0.71	0.69	0.66	0.47	0.81
4	0.58	0.48	0.61	0.6	0.61	0.69
5	0.76	0.62	0.54	0.83	0.68	0.84
6	0.73	0.51	0.57	0.71	0.67	0.75
7	0.84	0.62	0.6	0.8	0.68	0.79
8	0.66	0.57	0.63	0.7	0.49	0.7
9	0.8	0.65	0.59	0.8	0.56	0.81
10	0.74	0.71	0.65	0.82	0.6	0.81
11	0.74	0.53	0.72	0.8	0.79	0.77
12	0.67	0.4	0.56	0.66	0.53	0.72

(b)

	Normalized Mean Bias (%)					
Month	xylene	toluene	styrene	ethylbenzene	BENZ	SBTEX
1	-29.78	-3.85	135.07	-25.12	59.29	10.94
2	-19.24	1.43	164.16	-12.22	19.87	5.87
3	-33.41	-13.15	163.16	-26.86	21.59	-3.99
4	-44.24	-20.97	130.03	-38.46	0.93	-18.64
5	-23.7	-0.43	93.02	-15.97	45.77	10.71
6	-24.86	-4.01	56.83	-18.28	45.24	8.1
7	-20.45	5.53	105.34	-7.84	55.7	12.98
8	-33.62	-5.26	59.36	-23.58	36.7	1.21
9	-24.62	-4.12	37.66	-14.55	32.96	3.99
10	-34.47	-16.8	53.57	-20.92	48.08	-0.53
11	-31.66	-22.23	64.58	-14.05	28.85	-7.69
12	-31.73	-9.02	63.45	-3.04	27.15	1.95

Figures:

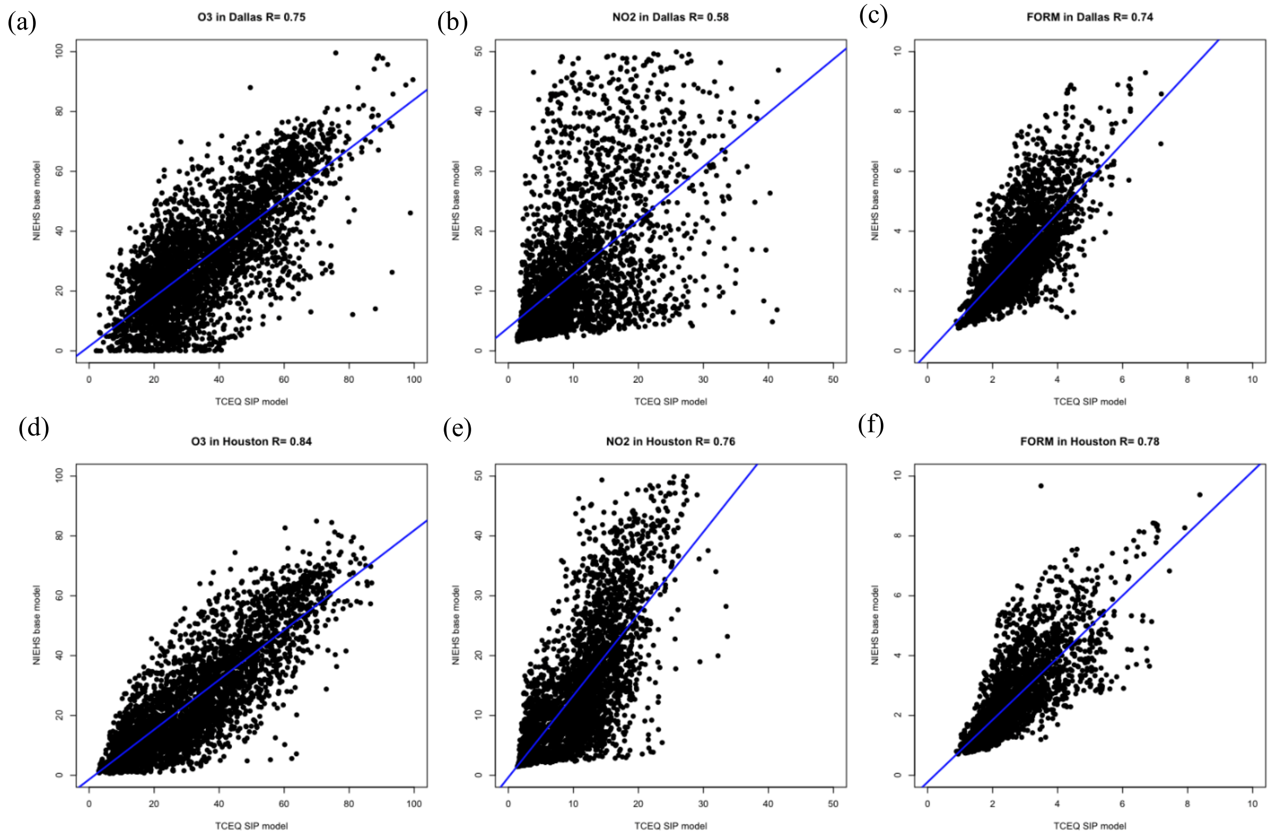

Figure S1. The model result comparison between TCEQ Ozone SIP application and our NIEHS base model result: a) ozone, b) NO_2, c) formaldehyde in Dallas, Texas; d) ozone, e) NO_2, and f) formaldehyde in Houston, Texas.

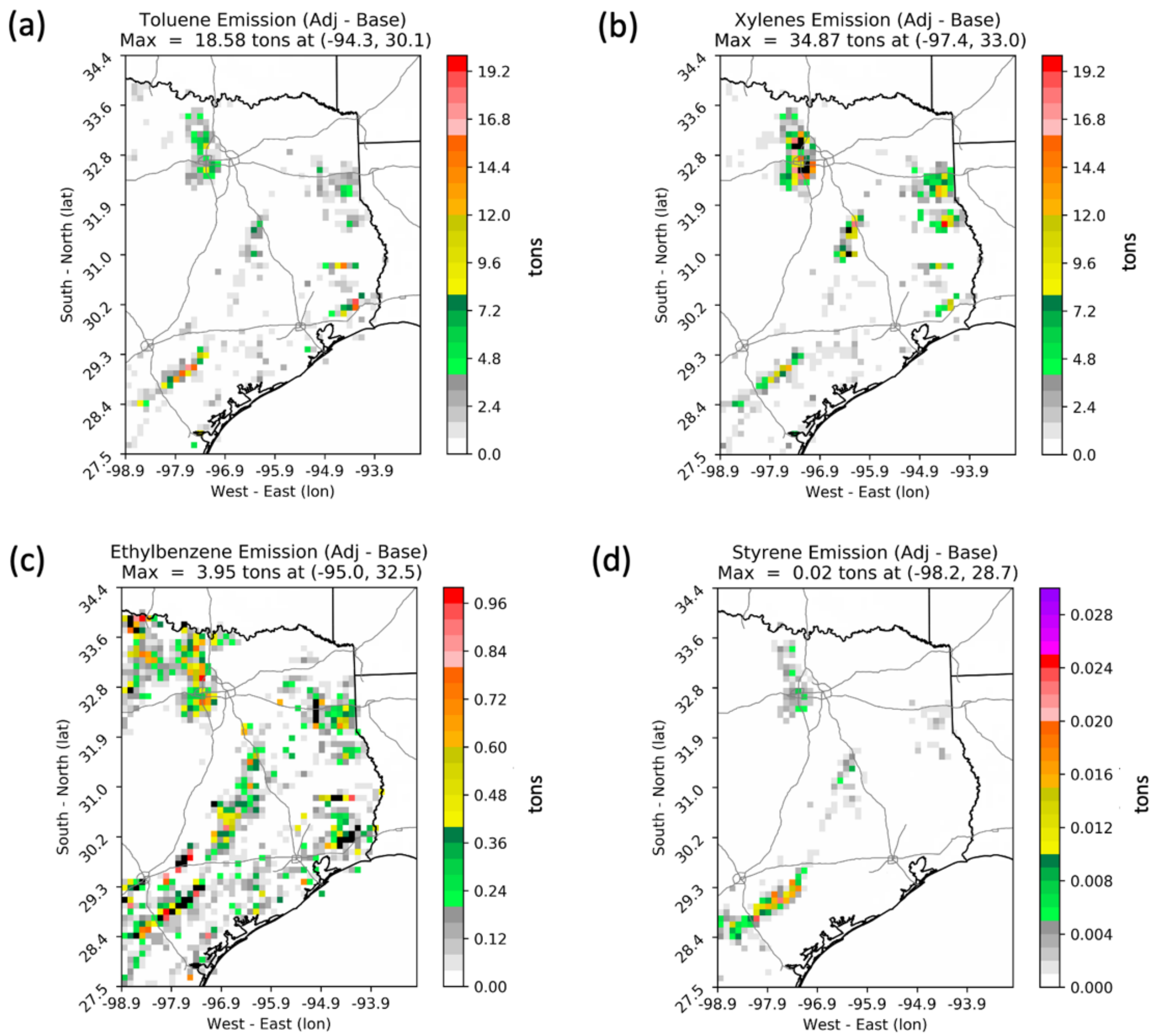

Figure S2. For the non-point source oil and gas industry emission sector (np_oilgas) the increase in annual emission rates for the 2012 Emission Inventory (Base), and the emission scenario adjusted in this study (Adj) of (a) toluene, (b) xylenes, (c) ethylbenzene, and (d) styrene

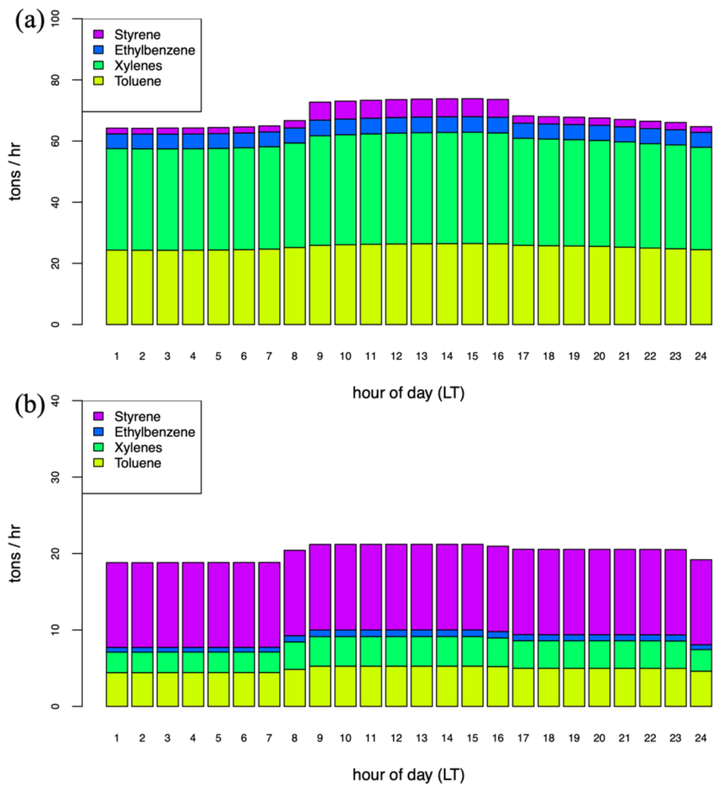

Figure S3. (a) For the "np_oilgas" imputation emission (Adj - Base) from May 1st to September 30th over the entire 12×12 km model domain. The bars are the hourly total of styrene, toluene, ethylbenzene, and xylenes (STEX) (tons hr^{-1}), and **(b)** is for the "ptnonipm" imputation emission.

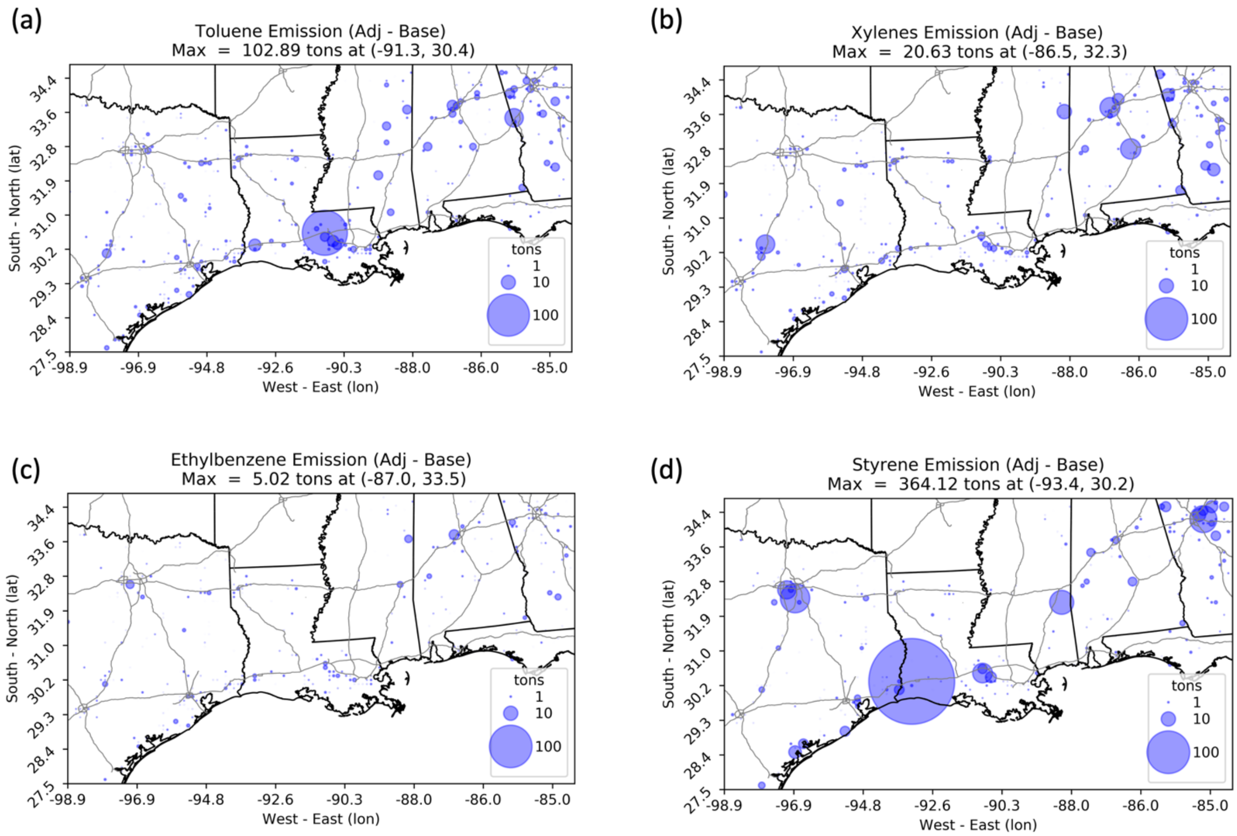

Figure S4. For the industry point source sector without electricity power plant emissions (ptnonipm) the increase in annual emission rates for the 2012 Emission Inventory (Base), and the emission scenario adjusted in this study (Adj) for (a) toluene, (b) xylenes, (c) ethylbenzene, and (d) styrene,

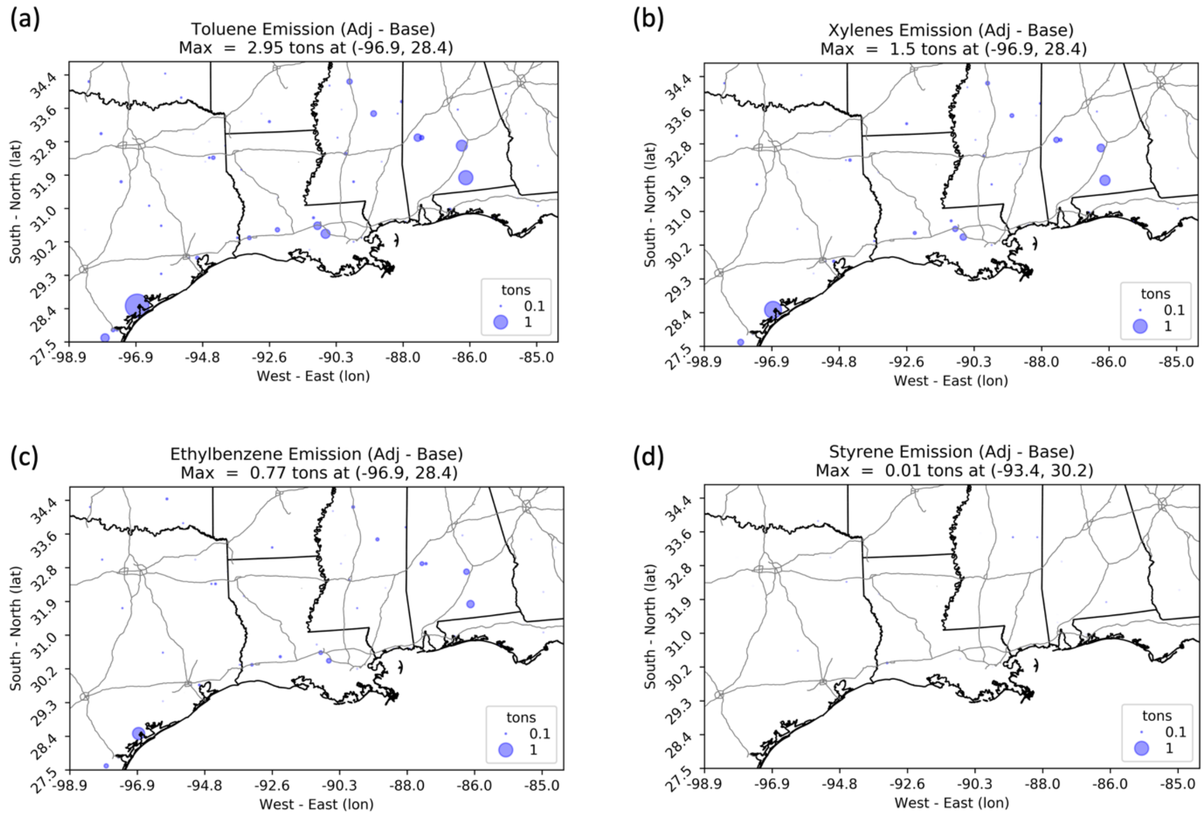

Figure S5 Delta emission (Adj-Base) of (a) toluene, (b) xylenes, (c) ethylbenzene, and (d) styrene, in electricity generation power plant emission sector (ptegu)

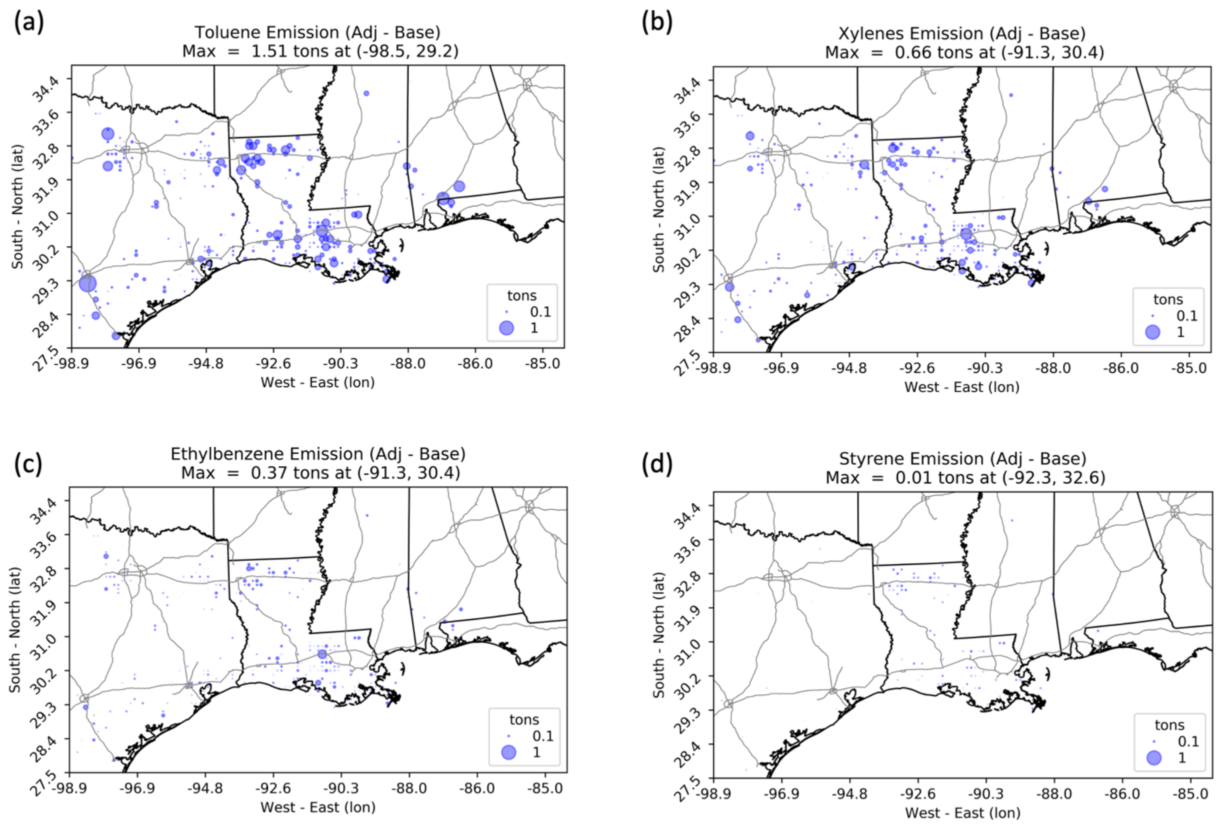

Figure S6 Delta emission (Adj-Base) of (a) toluene, (b) xylenes, (c) ethylbenzene, and (d) styrene, in point source oil and gas industry emission sector (pt_oilgas)

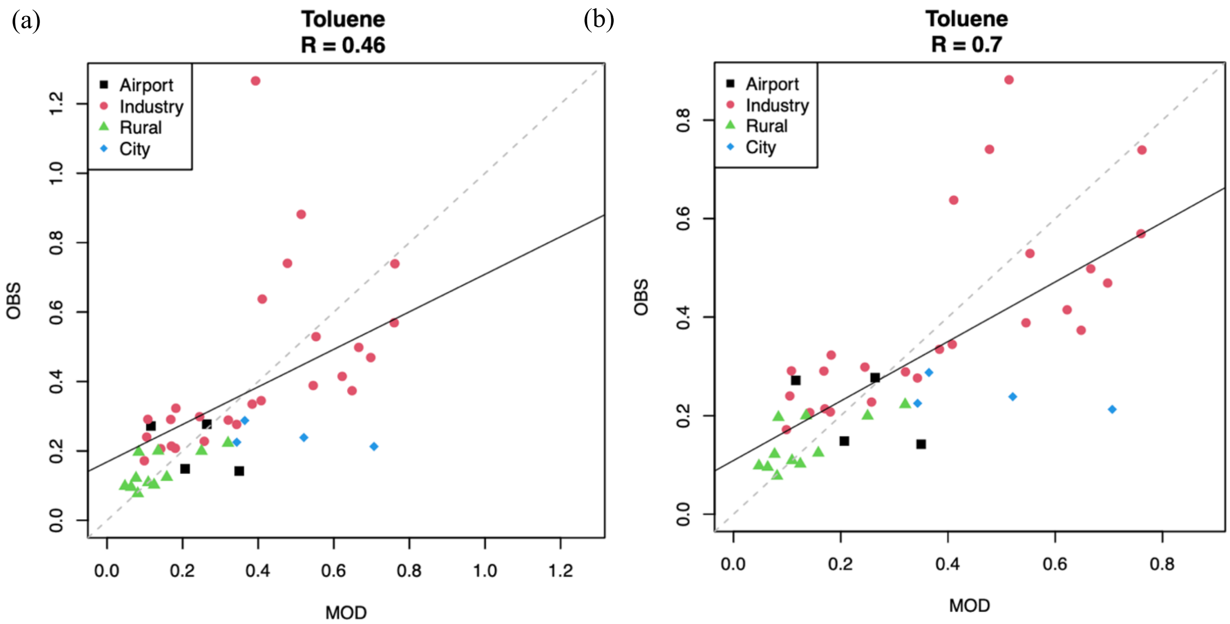

Figure S7 The average toluene concentration (ppb) comparison between model and observational data. (a) is original, and (b) is after removing two sites (Site ID: 482011015 and 482450014) with a significant underestimate.

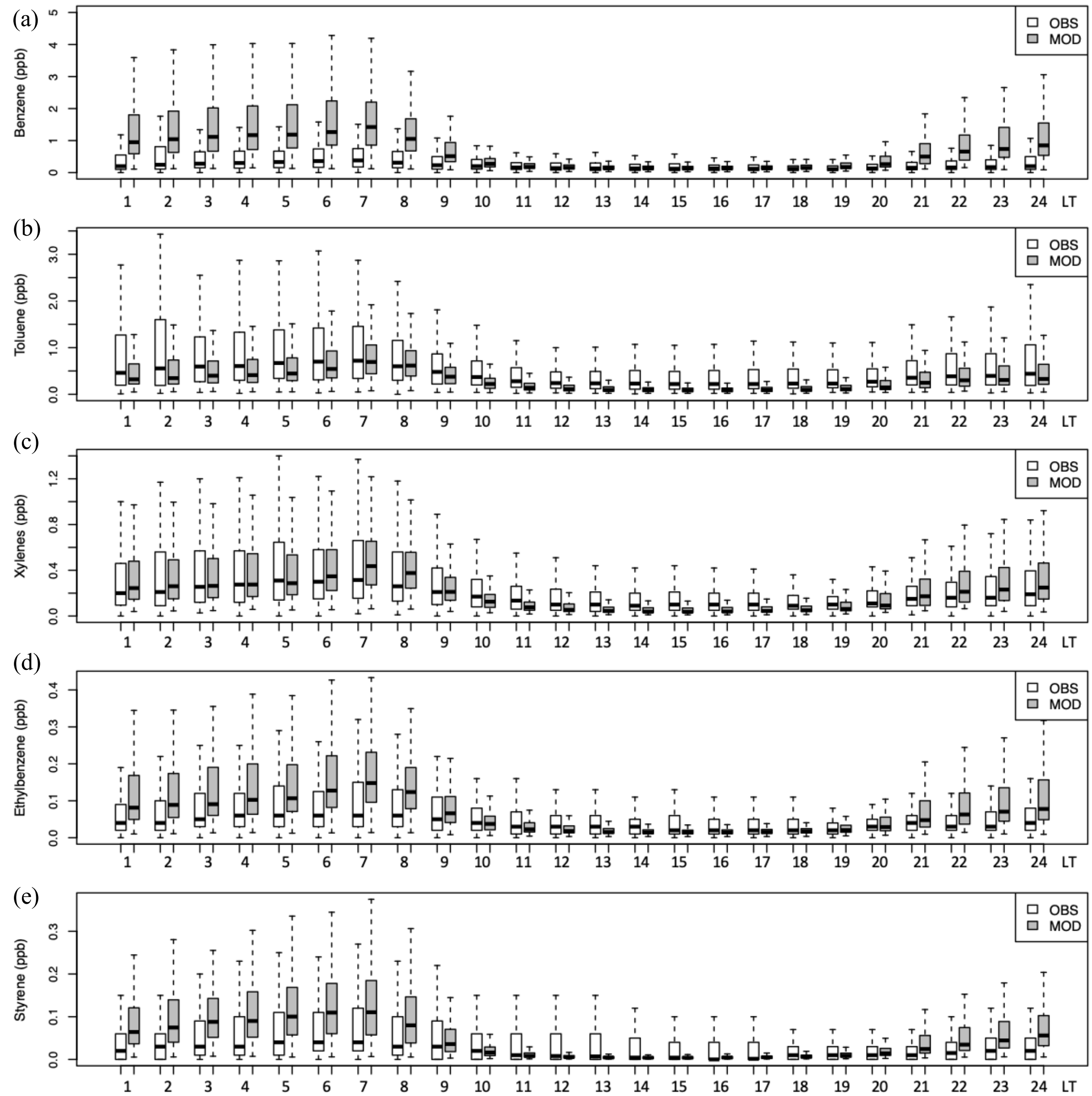

Figure S8. Diurnal pattern comparison of SBTEX in Houston (Site ID: 482010803, 482011015, 482010617) during the model simulation period (May 1st, 2012 to Sep 30th, 2012) for (a) benzene; (b) toluene; (c) xylenes; (d) ethylbenzene, and (e) styrene.

Reference:

Safety flare for burning combustible gas - has tangential inlet for non-flammable gas between housing and stack.

Hogrefe, C., Gilliam, R., Mathur, R., Henderson, B., Sarwar, G., Appel, K. W., Pouliot, G., Willison, J., Miller2, R., Vukovich1, J., Eyth, A., Talgo, K., Allen, C., and Foley, K.: CMAQv5.3.2 ozone simulations over the Northern Hemisphere: model performance and sensitivity to model configuration Office of Research and Development, 2021.

NASA: OZONE & AIR QUALITY: https://ozoneaq.gsfc.nasa.gov, last 2021.

RAMBOLL: CAMx7.00 User's Guide: https://camx-wp.azurewebsites.net/Files/CAMxUsersGuide_v7.00.pdf, last 2020a.

Ramboll: Speciation Tool User's Guide: https://www.cmascenter.org/speciation_tool/documentation/5.0/Ramboll_sptool_users_guide_V5.pdf, last 2020b.

Simon, H., Beck, L., Bhave, P. V., Divita, F., Hsu, Y., Luecken, D., Mobley, J. D., Pouliot, G. A., Reff, A., Sarwar, G., and Strum, M.: The development and uses of EPA's SPECIATE database, Atmospheric Pollution Research, 1, 196-206, 10.5094/apr.2010.026, 2010.

Steyn, D. G. and Rao, S. T.: Air Pollution Modeling and Its Application XX, Regional and intercontinental modelling, 2010.

TCEQ: TCEQ 2012 Modeling Platform Technical Support Document, https://wayback.archive-it.org/414/20210529054550/https://www.tceq.texas.gov/assets/public/implementation/air/am/modeling/gn/doc/TCEQ_2012_Modeling_Platform_TSD.pdf, 2016.

USEPA: Guidance on the Use of models and other analyses for demonstrating attainment of air quality goals for ozone, PM2.5 and Regional Haze, https://permanent.fdlp.gov/LPS74329/draft_final-pm-O3-RH.pdf, 2006.

USEPA: SPECIATE: https://www.epa.gov/air-emissions-modeling/speciate, last access: Aug 31, 2023, 2021a.

USEPA: National Emissions Inventory (NEI) 2011 Version 6 Air Emissions Modeling Platforrm (EMP): https://www.epa.gov/air-emissions-modeling/emissions-modeling-platforms, last access: JAN, 27th, 2022, 2021b.

USEPA: EPA's report on the environment: https://cfpub.epa.gov/roe/, last access: Aug. 31, 2023, 2023.

Yarwood, G. and Jung, J.: UPDATES TO THE CARBON BOND MECHANISM FOR VERSION 6 (CB6), 2010.