Electronic Supplementary Material (ESI) for Environmental Science: Atmospheres.

SUPPORTING INFORMATION:

Anthropogenic Secondary Organic Aerosol and Ozone Production from Asphalt-Related Emissions

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Table S1: Nationwide usage and application, in-use, and total emission factors averages and 95% ranges for the Monte Carlo simulations.

Statistic	Usage [Gg]					
	Asphalt Cement	Hot-Mix	Warm-Mix	Cutback	Emulsified	Roofing
Average	15416	12235	3182	153	1960	2911
2.5%	14218	11271	2855	137	1785	2602
97.5%	16619	13211	3517	169	2136	3225
Uncertainty Applied	±25% uncertainty in usage at the state-level aggregations from the Asphalt Institute survey. ±25% uncertainty in hot- and warm-mix splits at the state-level.					
Statistic	Application Emission Factor [g kg^{-1}]					
	Asphalt Cement	Hot-Mix	Warm-Mix	Cutback	Emulsified	Roofing
Average	--	3.92	2.11	406.68	97.58	0.20
2.5%	--	3.18	1.47	305.06	72.98	0.14
97.5%	--	4.38	2.41	509.20	121.98	0.22
Uncertainty Applied	--	Skewed, non-negative with a threshold max value equivalent to a 6-hr timescale of emission and approximate mean of a 5-hr timescale of emission		±25% uncertainty in NEI emission factors		Skewed, non-negative with a threshold max value equivalent to a 6-hr timescale of emission and approximate mean of a 5-hr timescale of emission.
Statistic	In-Use Emission Factor [g kg^{-1}]					
	Asphalt Cement	Hot-Mix	Warm-Mix	Cutback	Emulsified	Roofing
Average	--	1.00	1.00	1.01	1.00	18.30
2.5%	--	0.13	0.14	0.15	0.15	9.61
97.5%	--	2.42	2.42	2.42	2.39	31.56
Uncertainty Applied	--	Lognormally distributed with a mean value of ~1 g/kg				Lognormally distributed with a mean value of ~17.8 g/kg
Statistic	Total Emission Factor [g kg^{-1}]					
	Asphalt Cement	Hot-Mix	Warm-Mix	Cutback	Emulsified	Roofing
Average	--	4.92	3.11	407.69	98.58	18.50
2.5%	--	3.32	1.61	305.20	73.12	9.75
97.5%	--	6.80	4.83	511.61	124.36	31.78

Note: All emission factor distributions are illustrated in Fig. S1.

Table S2: Statewide, population weighted, summertime-averge SOA and MDA8 O_3 enhancements from asphalt-related emissions.

State	SOA [μg m^{-3}]					MDA8 O_3 [ppb]				
	SON	DJF	MAM	JJA	Annual	SON	DJF	MAM	JJA	Annual
AL	0.02	0.02	0.02	0.02	0.02	0.01	0.01	0.00	0.00	0.01
AZ	0.04	0.03	0.03	0.03	0.03	0.04	0.03	0.03	0.03	0.03
AR	0.02	0.02	0.02	0.02	0.02	0.01	0.02	0.00	0.00	0.01
CA	0.08	0.05	0.05	0.07	0.06	0.10	0.05	0.07	0.12	0.08
CO	0.02	0.02	0.02	0.02	0.02	0.02	0.01	0.02	0.03	0.02
CT	0.02	0.02	0.02	0.05	0.03	0.02	0.02	0.02	0.03	0.02
DE	0.03	0.02	0.03	0.04	0.03	0.02	0.02	0.02	0.01	0.02
DC	0.04	0.03	0.04	0.06	0.04	0.03	0.02	0.03	0.03	0.03
FL	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01	0.01
GA	0.03	0.02	0.02	0.03	0.02	0.01	0.02	0.01	0.00	0.01
ID	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.00	0.01
IL	0.04	0.03	0.04	0.06	0.04	0.03	0.02	0.04	0.04	0.03
IN	0.03	0.03	0.03	0.05	0.04	0.02	0.03	0.02	0.01	0.02
IA	0.02	0.03	0.03	0.03	0.03	0.01	0.02	0.02	0.00	0.01
KS	0.02	0.02	0.03	0.03	0.02	0.01	0.01	0.01	0.00	0.01
KY	0.03	0.03	0.03	0.04	0.03	0.01	0.02	0.01	0.00	0.01
LA	0.02	0.02	0.02	0.02	0.02	0.01	0.01	0.01	0.01	0.01
ME	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.00	0.00	0.01
MD	0.03	0.03	0.03	0.05	0.03	0.02	0.02	0.02	0.01	0.02
MA	0.02	0.02	0.02	0.03	0.02	0.01	0.01	0.01	0.01	0.01
MI	0.03	0.03	0.03	0.05	0.04	0.02	0.02	0.02	0.02	0.02
MN	0.02	0.03	0.04	0.03	0.03	0.01	0.02	0.03	0.02	0.02
MS	0.02	0.02	0.02	0.02	0.02	0.01	0.01	0.00	0.00	0.00
MO	0.03	0.02	0.03	0.03	0.03	0.01	0.02	0.01	0.00	0.01
MT	0.01	0.00	0.01	0.01	0.01	0.00	0.00	0.00	0.00	0.00
NE	0.02	0.02	0.03	0.03	0.02	0.01	0.01	0.01	0.01	0.01
NV	0.03	0.01	0.02	0.04	0.02	0.02	0.01	0.02	0.03	0.02
NH	0.01	0.02	0.01	0.02	0.02	0.01	0.01	0.01	0.00	0.01
NJ	0.03	0.03	0.03	0.06	0.04	0.03	0.02	0.03	0.04	0.03
NM	0.02	0.02	0.01	0.02	0.02	0.02	0.01	0.01	0.01	0.01
NY	0.03	0.03	0.03	0.06	0.04	0.03	0.02	0.03	0.07	0.04
NC	0.03	0.02	0.02	0.03	0.02	0.01	0.02	0.01	0.00	0.01
ND	0.01	0.01	0.01	0.01	0.01	0.00	0.01	0.01	0.00	0.00
OH	0.03	0.03	0.04	0.05	0.04	0.02	0.03	0.02	0.01	0.02
OK	0.02	0.02	0.03	0.03	0.03	0.01	0.02	0.01	0.01	0.01
OR	0.02	0.00	0.01	0.02	0.01	0.02	0.00	0.01	0.02	0.01
PA	0.03	0.03	0.03	0.04	0.03	0.02	0.02	0.02	0.01	0.02
RI	0.02	0.02	0.02	0.04	0.02	0.01	0.01	0.02	0.01	0.01
SC	0.02	0.02	0.02	0.02	0.02	0.01	0.02	0.00	0.00	0.01
SD	0.01	0.01	0.02	0.02	0.01	0.00	0.01	0.01	0.00	0.00
TN	0.03	0.02	0.03	0.03	0.03	0.01	0.02	0.01	0.00	0.01
TX	0.02	0.02	0.03	0.02	0.02	0.02	0.02	0.01	0.01	0.02
UT	0.02	0.01	0.01	0.02	0.01	0.01	0.01	0.01	0.01	0.01
VT	0.01	0.02	0.01	0.02	0.01	0.01	0.02	0.00	0.00	0.01
VA	0.02	0.02	0.02	0.03	0.03	0.01	0.02	0.01	0.00	0.01
WA	0.01	0.00	0.01	0.02	0.01	0.01	0.00	0.02	0.03	0.02
WV	0.02	0.02	0.02	0.03	0.02	0.01	0.02	0.01	0.00	0.01
WI	0.03	0.03	0.03	0.05	0.04	0.02	0.02	0.02	0.02	0.02
WY	0.01	0.01	0.01	0.01	0.01	0.00	0.00	0.00	0.00	0.00

Table S3: Summary of data in Fig. S3.

Region	OC NMB %				$PM_{2.5}$ NMB %				MDA8 O_3 NMB %			
NE	0.48	0.97	0.42	-0.19	0.11	0.21	0.07	-0.26	0.16	0.02	-0.14	0.04
SE	0.33	0.36	0.15	-0.16	0.03	0.11	-0.06	-0.28	0.11	-0.05	-0.12	0.11
MW	0.40	1.29	0.53	-0.32	0.16	0.24	0.21	-0.23	0.17	0.10	-0.13	0.09
PL	0.22	0.79	0.26	-0.46	0.14	0.17	0.08	-0.41	0.05	-0.03	-0.15	-0.02
SW	-0.09	0.04	0.20	-0.32	-0.23	-0.21	0.03	-0.32	-0.11	-0.11	-0.23	-0.13
NW	-0.11	0.85	0.41	-0.46	0.03	-0.08	0.27	-0.45	-0.03	-0.06	-0.18	-0.08
Season	SON	DJF	MAM	JJA	SON	DJF	MAM	JJA	SON	DJF	MAM	JJA

Region	OC n Observations				$PM_{2.5}$ n Observations				MDA8 O_3 n Observations			
NE	1314	1317	1324	1359	17592	18354	18395	18375	14632	11251	18082	18615
SE	844	818	853	912	15941	15927	16635	16718	13701	7810	18891	18477
MW	887	905	904	932	14614	15194	15265	15183	14059	7691	18189	18361
PL	2050	1964	2014	2030	23288	21974	22894	23591	25904	20983	28423	28668
SW	1344	1320	1347	1368	16397	16422	16786	17052	19788	17597	20660	21512
NW	641	596	620	644	11735	11390	11687	12099	1114	664	1397	2400
Season	SON	DJF	MAM	JJA	SON	DJF	MAM	JJA	SON	DJF	MAM	JJA

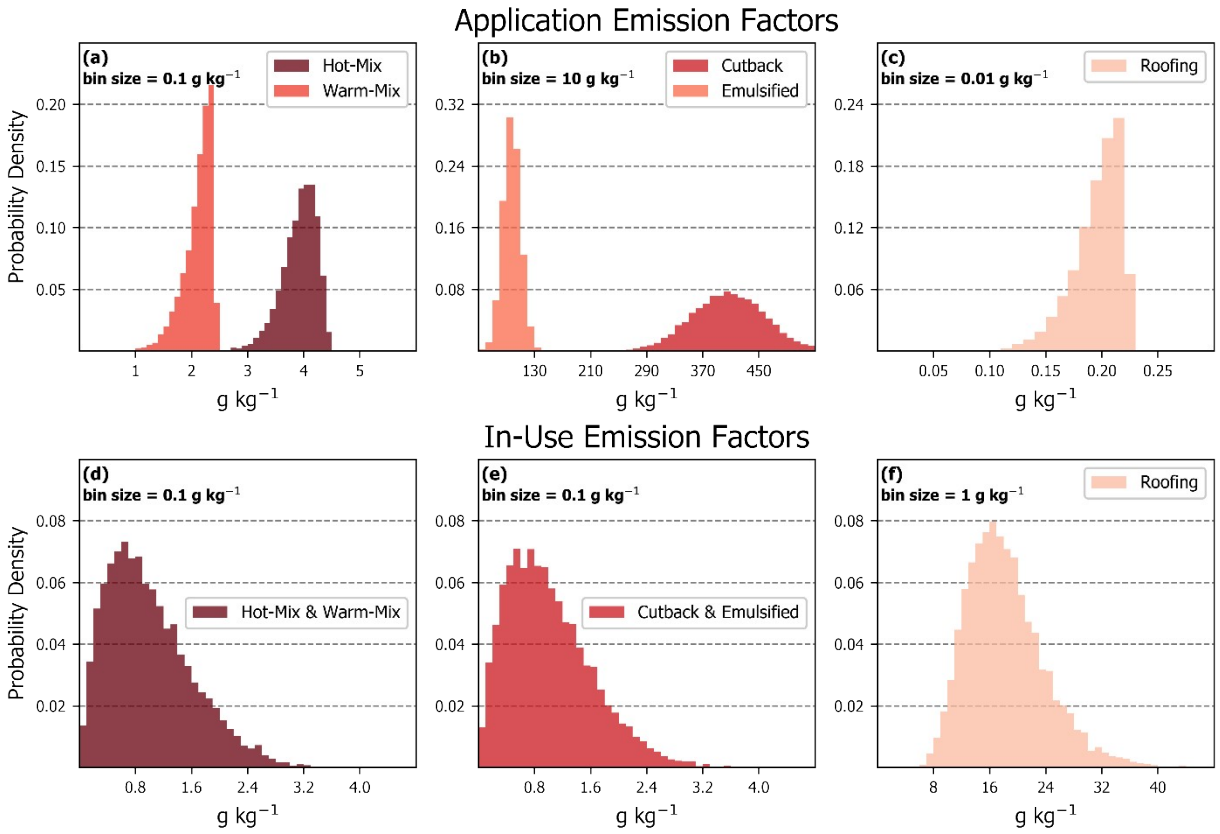

Figure S1. Application emission factors, in g kg^{-1} of liquid asphalt, for (a) hot- and warm-mix paving asphalt, (b) cutback and emulsified paving asphalt, and (c) roofing asphalt from the Monte Carlo analysis. In-use emission factors, in g kg^{-1} of liquid asphalt, for (d) hot- and warm-mix paving asphalt, (e) cutback and emulsified paving asphalt, and (f) roofing asphalt from the Monte Carlo analysis.

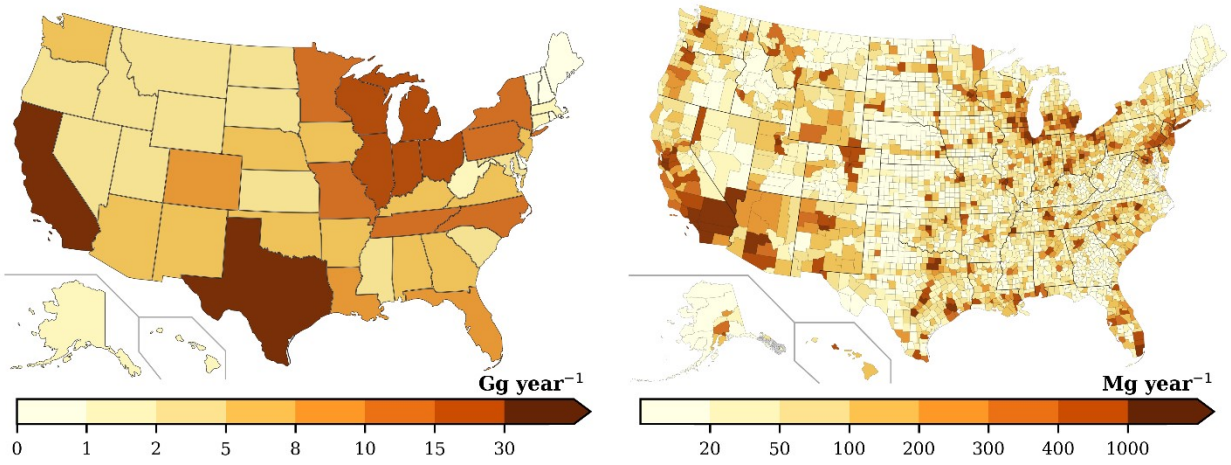

Figure S2. (Left) State-level and (Right) county-level of asphalt-related emissions.

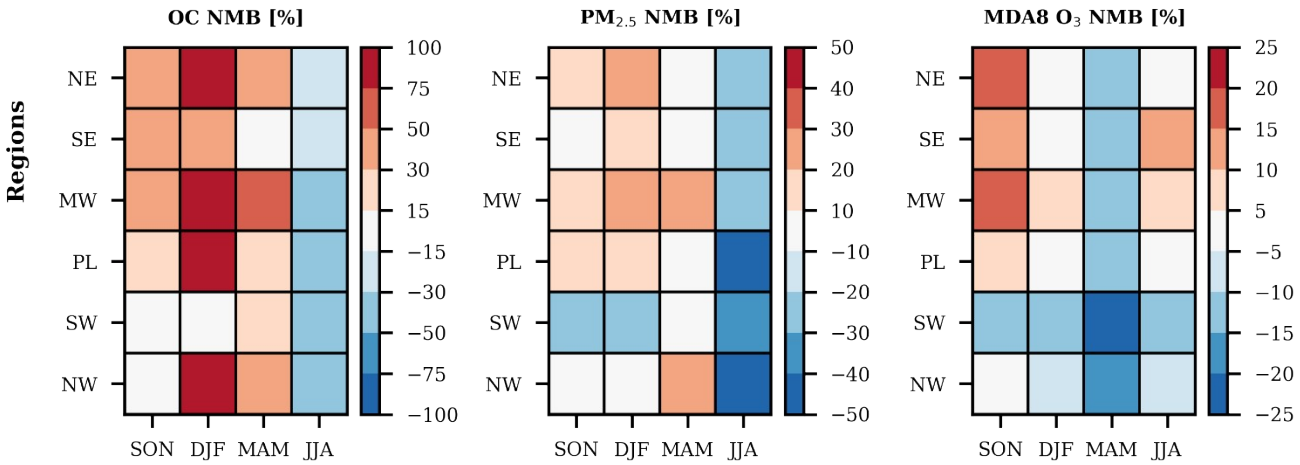

Figure S3. Modeling performance metrics for OC, $PM_{2.5}$, and maximum daily 8-hour O_3, disaggregated by season and region. Note: NE – U.S. EPA Regions 1, 2, 3; SE – U.S. EPA Region 4; MW – U.S. EPA Region 5; PL – U.S. EPA Regions 6, 7, 8; SW – U.S. EPA Region 9; NW – U.S. EPA Region 10; SON – fall months; DJF – winter months; MAM – spring months; JJA – summer months; NMB – normalized mean bias. A numerical summary of this data can be found in Table S3.

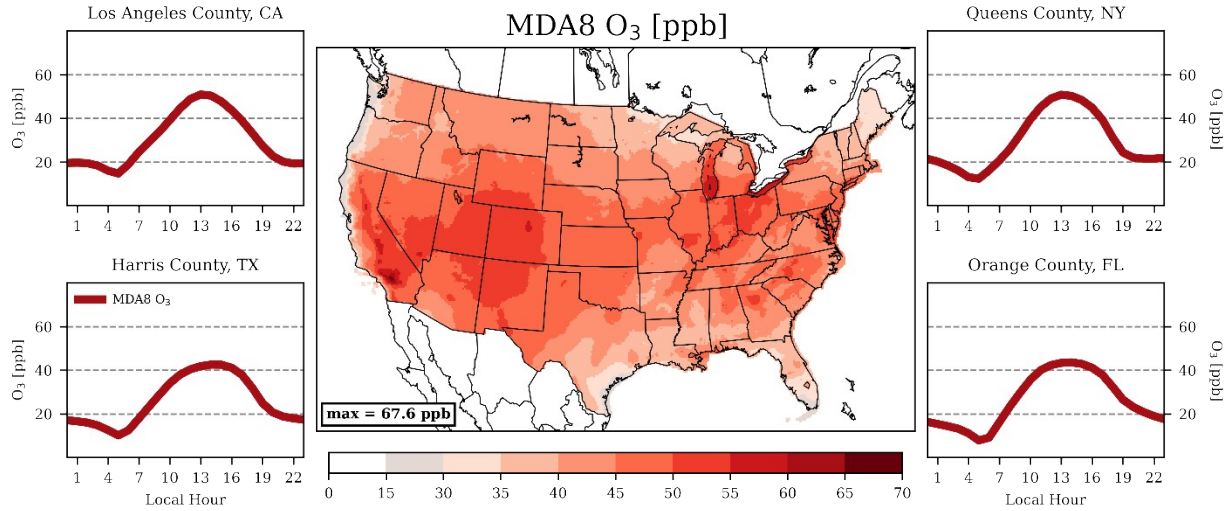

Figure S4. Summertime-averge MDA8 O_3 from the simulation that includes the new asphalt-related emissions (center). Diurnal, population-weighted O_3 (side panels). Note that Houston and Orlando are located in Harris County, TX and Orange County, Florida, respectively.

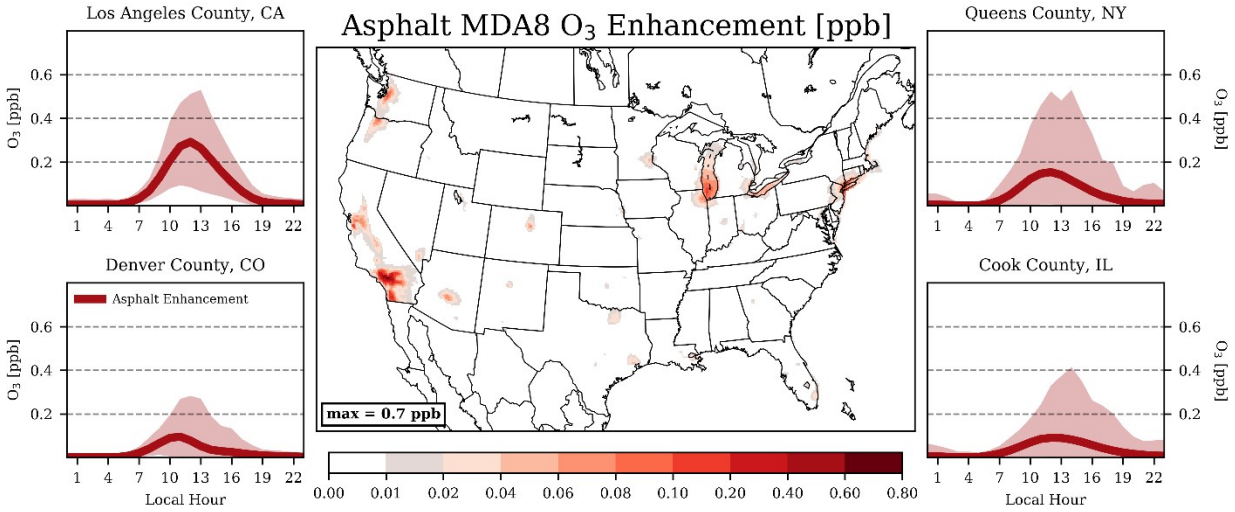

Figure S5. Summertime-averge MDA8 O_3 enhancements from asphalt-related emissions (center). Diurnal, population weighted O_3 enhancements from asphalt-related emissions for select counties (side panels) where the lines indicate the average asphalt-related enhancements for each hour of the day and shading represents the asphalt-related enhancements for 95% of all days.