

Supporting Information:

Magnitude, Trends, and Impacts of Ambient Long-Term Ozone Exposure in the United States from 2000-2015

Karl M. Seltzer¹, Drew T. Shindell^{1,2}, Prasad Kasibhatla¹, Christopher S. Malley³

5 ¹Nicholas School of the Environment, Duke University, Durham, NC, USA

²Duke Global Health Initiative, Duke University, Durham, NC, USA

³Stockholm Environmental Institute, Department of Environment and Geography, University of York, York, UK

Correspondence to: Karl M. Seltzer (kms147@duke.edu); Drew T. Shindell (drew.shindell@duke.edu)

10

	Table S1: Evaluation of the ANN predictions using the original TOAR observations. Note: RMSE = Root-mean squared error. ...	2
	Table S2: Population-weighted exposure concentrations from 2000-2015. Note: DJF = December-February (all months within same calendar year); MAM = March-May; JJA = July-August; SON = September-November.	3
15	Table S3: Agriculture-weighted exposure concentrations from 2000-2015. Note: JAS = July-September; MAM = March-May; MJJ = May-July.	4
	Table S4: Meteorological adjusted and non-adjusted trends of all metrics (population-weighted and agriculture-weighted, respectively) from 2000-2015.	5
	Table S5: Estimated human-health and crop-loss impacts attributable to long-term O ₃ exposure from 2000-2015.	6

20

	Figure S1: TOAR observations (left) of the April-September average of the daily 1-hour maximum concentration (Jerrett et al. 2009 averaging metric) and predictions using the ANN (center). Concentrations are reported in ppb. Note: color bar has non-uniform intervals. Right: Scatter plot comparing TOAR observations (left) and ANN predictions (center), with each dot representing a monitoring locations. The mean bias (ppb) and root mean square error (ppb) are included in the scatter plot panel.	7
25	Figure S2: TOAR observations (left) of the annual average of the maximum daily 8-hour concentration (Turner et al. 2016 averaging metric) and predictions using the ANN (center). Concentrations are reported in ppb. Note: color bar has non-uniform intervals. Right: Scatter plot comparing TOAR observations (left) and ANN predictions (center), with each dot representing a monitoring locations. The mean bias (ppb) and root mean square error (ppb) are included in the scatter plot panel.	7
30	Figure S3: TOAR observations (left) of the July-September average of the M12 agriculture metric (8:00-20:00 local time) and predictions using the ANN (center). Concentrations are reported in ppb. Note: color bar has non-uniform intervals. Right: Scatter plot comparing TOAR observations (left) and ANN predictions (center), with each dot representing a monitoring locations. The mean bias (ppb) and root mean square error (ppb) are included in the scatter plot panel.	8
35	Figure S4: TOAR observations (left) of the July-September sum of the daily AOT40 agriculture metric (8:00-20:00 local time) and predictions using the ANN (center). Concentrations are reported in ppmh. Note: color bar has non-uniform intervals. Right: Scatter plot comparing TOAR observations (left) and ANN predictions (center), with each dot representing a monitoring locations. The mean bias (ppmh) and root mean square error (ppmh) are included in the scatter plot panel.	8
40	Figure S5: The p-values for the trends presented in Fig. 2 from the main text.	9
	Figure S6: The p-values for the trends presented in Fig. 4 from the main text.	9
	Figure S7: 2010 USA population (top left) and wheat (top right), maize (bottom left), and soybean (bottom right) production maps. Dots in top right panel represent monitoring locations with available data on January 1, 2010 and dots in bottom right panel represent monitoring locations with available data on July 1, 2010.	10
45	Figure S8: Comparison of the M12 and AOT40 concentration-response curves for soybeans and maize. To note: the concentrations are different for the M12 (ppb) and AOT40 (ppmh) metrics.	10

Performance Metric	Year	MDA1	MDA8	M12	AOT40
		April – Sept. Avg.	Annual Avg.	July – Sept. Avg.	July – Sept. Sum
Mean Bias ppb: MDA1, MDA8, M12 ppmh: AOT40	2000	0.11	0.47	0.57	-0.18
	2001	-0.60	-0.13	0.52	0.52
	2002	-0.60	-0.23	-0.89	-0.30
	2003	-1.26	-0.29	-0.69	-0.17
	2004	0.67	0.29	0.38	0.31
	2005	-0.61	-0.22	0.51	0.37
	2006	-1.09	-0.66	-0.50	0.02
	2007	-1.09	-0.43	-0.07	0.01
	2008	-0.98	-0.18	0.74	0.38
	2009	0.19	0.42	0.97	0.87
	2010	0.03	-0.36	0.86	0.73
	2011	-0.42	-0.11	0.17	0.64
	2012	-1.16	-0.31	-0.64	-0.16
	2013	-0.12	-0.04	0.29	0.39
	2014	-0.02	0.30	0.70	0.12
2015	-0.43	0.24	-0.07	0.24	
RMSE ppb: MDA1, MDA8, M12 ppmh: AOT40	2000	4.27	3.87	4.36	2.84
	2001	4.04	3.70	4.07	2.80
	2002	4.38	3.87	4.45	3.24
	2003	4.36	3.51	4.48	2.94
	2004	3.96	3.39	4.08	2.68
	2005	3.84	3.08	4.02	2.83
	2006	3.97	3.36	4.01	2.72
	2007	3.93	3.04	3.84	2.76
	2008	3.79	3.05	3.84	2.46
	2009	3.44	2.99	3.70	2.32
	2010	3.41	2.78	3.82	2.48
	2011	3.38	2.75	3.58	2.50
	2012	3.59	2.83	3.73	2.42
	2013	3.17	2.74	3.36	2.11
	2014	3.15	2.64	3.30	1.83
2015	3.07	2.31	3.06	1.92	
r ² [unitless]	2000	0.80	0.77	0.81	0.81
	2001	0.81	0.74	0.80	0.78
	2002	0.80	0.75	0.79	0.79
	2003	0.77	0.75	0.75	0.78
	2004	0.77	0.76	0.76	0.76
	2005	0.79	0.77	0.77	0.77
	2006	0.79	0.76	0.76	0.76
	2007	0.81	0.79	0.81	0.77
	2008	0.83	0.80	0.81	0.81
	2009	0.81	0.78	0.82	0.84
	2010	0.82	0.80	0.81	0.80
	2011	0.84	0.82	0.83	0.83
	2012	0.83	0.80	0.83	0.83
	2013	0.82	0.76	0.81	0.81
	2014	0.80	0.76	0.79	0.80
2015	0.81	0.79	0.77	0.77	

Table S1: Evaluation of the ANN predictions using the original TOAR observations. Note: RMSE = Root-mean squared error.

Year	MDA1	MDA8				
	[ppb]	[ppb]				
	April – Sept. Avg.	Annual Avg.	DJF Avg.	MAM Avg.	JJA Avg.	SON Avg.
2000	53.67	39.88	27.69	45.21	49.97	36.64
2001	54.65	40.10	27.10	44.58	51.54	37.20
2002	55.88	40.31	27.90	44.29	52.72	36.33
2003	52.34	39.12	28.09	43.34	48.35	36.70
2004	52.16	39.16	28.54	45.22	47.04	35.85
2005	54.73	40.43	27.79	45.05	50.37	38.51
2006	52.93	40.09	29.13	45.76	49.84	35.63
2007	53.03	40.41	29.57	46.15	48.18	37.73
2008	51.11	39.81	30.23	44.92	47.39	36.72
2009	48.84	38.68	30.76	44.68	44.73	34.53
2010	51.17	40.15	30.63	45.38	46.70	37.91
2011	51.21	40.46	31.11	44.65	48.50	37.59
2012	51.79	40.67	31.34	45.50	48.98	36.86
2013	48.39	39.10	31.29	44.47	43.95	36.70
2014	48.19	39.21	31.73	44.74	43.60	36.75
2015	48.34	39.14	31.59	43.68	43.90	37.36

Table S2: Population-weighted exposure concentrations from 2000-2015. Note: DJF = December-February (all months within same calendar year); MAM = March-May; JJA = July-August; SON = September-November.

Year	M12 [ppb]		AOT40 [ppmh]			
	Maize (JAS)	Soybean (JAS)	Maize (JAS)	Soybean (JAS)	Wheat (MAM)	Wheat (MJJ)
2000	43.20	44.20	9.61	10.48	7.40	10.84
2001	42.23	42.40	9.60	9.68	6.97	11.69
2002	44.17	44.26	11.19	11.46	6.98	12.05
2003	42.36	42.04	9.63	9.46	7.05	10.51
2004	40.16	39.89	7.25	6.91	7.23	9.26
2005	43.79	44.20	9.87	10.22	7.37	10.74
2006	40.88	41.03	8.02	8.20	7.71	12.07
2007	41.42	41.89	7.86	8.33	6.90	8.52
2008	39.17	38.89	6.16	6.00	6.66	8.84
2009	37.26	36.67	4.94	4.54	6.48	8.40
2010	39.47	39.66	6.24	6.40	6.78	7.48
2011	40.27	40.61	7.79	8.00	6.48	11.20
2012	43.25	43.02	9.87	9.70	7.17	11.83
2013	38.87	38.76	5.77	5.51	5.93	7.47
2014	36.83	36.51	3.84	3.50	6.95	6.62
2015	37.58	37.24	4.39	4.20	5.73	5.45

Table S3: Agriculture-weighted exposure concentrations from 2000-2015. Note: JAS = July-September; MAM = March-May; MJJ = May-July.

Trend Type	MDA1 [ppb/yr]			MDA8 [ppb/yr]		
	West	East	Total	West	East	Total
Met. Adjusted	-0.32 [-0.25, -0.39]	-0.46 [-0.39, -0.52]	-0.41 [-0.35, -0.47]	0.04 [0.07, 0.00]	-0.04 [-0.01, -0.07]	-0.02 [0.01, -0.04]
Non-Adjusted	-0.31 [-0.21, -0.41]	-0.49 [-0.28, -0.69]	-0.43 [-0.28, -0.57]	0.06 [0.11, 0.01]	-0.08 [0.02, -0.17]	-0.03 [0.04, -0.10]

Trend Type	MDA8-JJA [ppb/yr]			MDA8-DJF [ppb/yr]		
	West	East	Total	West	East	Total
Met. Adjusted	-0.29 [-0.23, -0.35]	-0.46 [-0.4, -0.53]	-0.40 [-0.35, -0.45]	0.33 [0.35, 0.30]	0.33 [0.35, 0.31]	0.33 [0.35, 0.31]
Non-Adjusted	-0.29 [-0.17, -0.41]	-0.54 [-0.28, -0.80]	-0.45 [-0.27, -0.64]	0.35 [0.42, 0.29]	0.31 [0.37, 0.26]	0.33 [0.37, 0.28]

Trend Type	MDA8-MAM [ppb/yr]			MDA8-SON [ppb/yr]		
	West	East	Total	West	East	Total
Met. Adjusted	0.06 [0.11, 0.02]	-0.01 [0.04, -0.05]	0.02 [0.05, -0.02]	0.04 [0.07, 0.02]	-0.03 [0.01, -0.07]	-0.01 [0.02, -0.04]
Non-Adjusted	0.07 [0.13, 0.02]	-0.05 [0.07, -0.17]	-0.01 [0.07, -0.09]	0.10 [0.19, 0.02]	-0.03 [0.12, -0.17]	0.02 [0.13, -0.08]

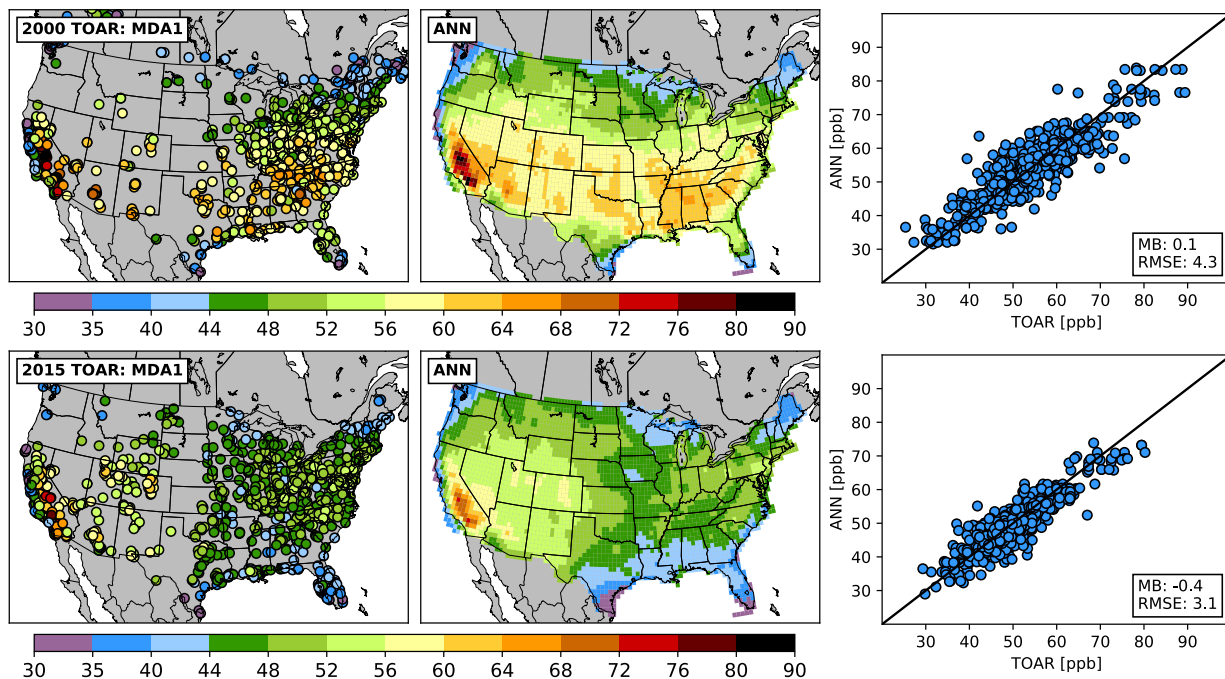
Trend Type	M12 Maize [ppb/yr]	M12 Soybean [ppb/yr]	AOT40 Maize [ppmh/yr]	AOT40 Soybean [ppmh/yr]	AOT40 Wheat MAM [ppmh/yr]	AOT40 Wheat MJJ [ppmh/yr]
Met. Adjusted	-0.22 [-0.19, -0.26]	-0.26 [-0.22, -0.30]	-0.27 [-0.22, -0.32]	-0.30 [-0.25, -0.36]	-0.04 [-0.02, -0.07]	-0.24 [-0.2, -0.28]
Non-Adjusted	-0.35 [-0.17, -0.54]	-0.39 [-0.19, -0.59]	-0.35 [-0.18, -0.51]	-0.39 [-0.21, -0.56]	-0.07 [-0.03, -0.11]	-0.29 [-0.12, -0.47]

Table S4: Meteorological adjusted and non-adjusted trends of all metrics (population-weighted and agriculture-weighted, respectively) from 2000-2015.

Year	Estimated Human-Health Impacts [# mortalities / 100,000 ppl / year]		Estimated Agriculture Impacts [% Relative Yield Lost]				
	J2009 [95% CI]	T2016 [95% CI]	M12 Maize	AOT40 Maize	M12 Soybeans	AOT40 Soybeans	AOT40 Wheat
2000	6.02 [2.04, 9.66]	10.81 [7.55, 13.79]	4.58	3.42	16.28	11.85	12.09
2001	6.34 [2.16, 10.18]	11.08 [7.74, 14.14]	4.26	3.42	14.94	10.95	11.39
2002	6.75 [2.30, 10.80]	11.33 [7.91, 14.46]	4.85	3.99	16.31	12.97	11.41
2003	5.73 [1.94, 9.22]	10.41 [7.25, 13.31]	4.28	3.43	14.66	10.70	11.53
2004	5.58 [1.89, 8.98]	10.27 [7.15, 13.13]	3.60	2.58	13.09	7.82	11.82
2005	6.40 [2.17, 10.27]	11.42 [7.96, 14.58]	4.70	3.51	16.26	11.57	12.05
2006	5.88 [1.99, 9.46]	11.11 [7.75, 14.18]	3.80	2.86	13.92	9.28	12.60
2007	5.90 [2.00, 9.49]	11.38 [7.94, 14.52]	4.02	2.80	14.57	9.42	11.28
2008	5.43 [1.84, 8.75]	11.08 [7.72, 14.15]	3.35	2.19	12.38	6.79	10.89
2009	4.77 [1.61, 7.70]	10.19 [7.09, 13.05]	2.84	1.76	10.78	5.13	10.60
2010	5.42 [1.84, 8.74]	11.32 [7.89, 14.46]	3.46	2.22	12.95	7.24	11.09
2011	5.58 [1.89, 8.99]	11.83 [8.26, 15.10]	3.68	2.78	13.63	9.06	10.59
2012	5.80 [1.96, 9.33]	12.11 [8.45, 15.46]	4.55	3.52	15.39	10.98	11.72
2013	4.84 [1.63, 7.82]	11.01 [7.66, 14.08]	3.24	2.05	12.28	6.23	9.71
2014	4.83 [1.63, 7.81]	11.22 [7.81, 14.35]	2.72	1.37	10.67	3.95	11.36
2015	4.95 [1.67, 8.00]	11.32 [7.88, 14.49]	2.87	1.57	11.18	4.76	9.38

Table S5: Estimated human-health and crop-loss impacts attributable to long-term O₃ exposure from 2000-2015.

60



65

Figure S1: TOAR observations (left) of the April-September average of the daily 1-hour maximum concentration (Jerrett et al. 2009 averaging metric) and predictions using the ANN (center). Concentrations are reported in ppb. Note: color bar has non-uniform intervals. Right: Scatter plot comparing TOAR observations (left) and ANN predictions (center), with each dot representing a monitoring locations. The mean bias (ppb) and root mean square error (ppb) are included in the scatter plot panel.

70

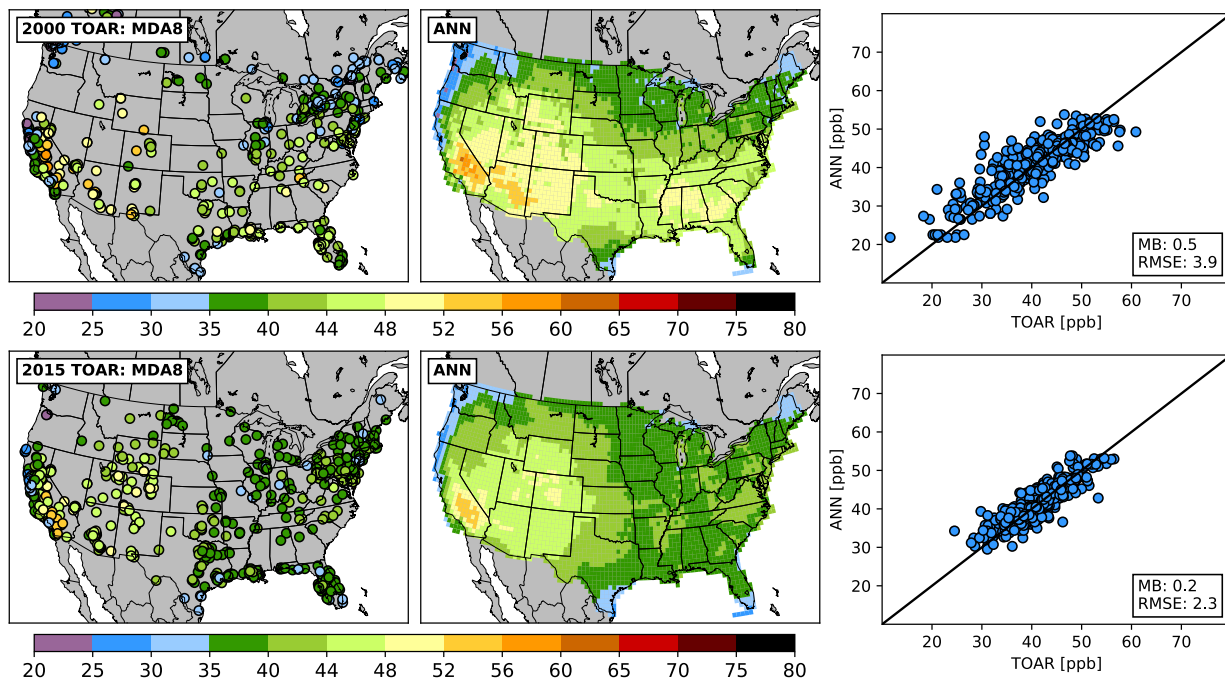
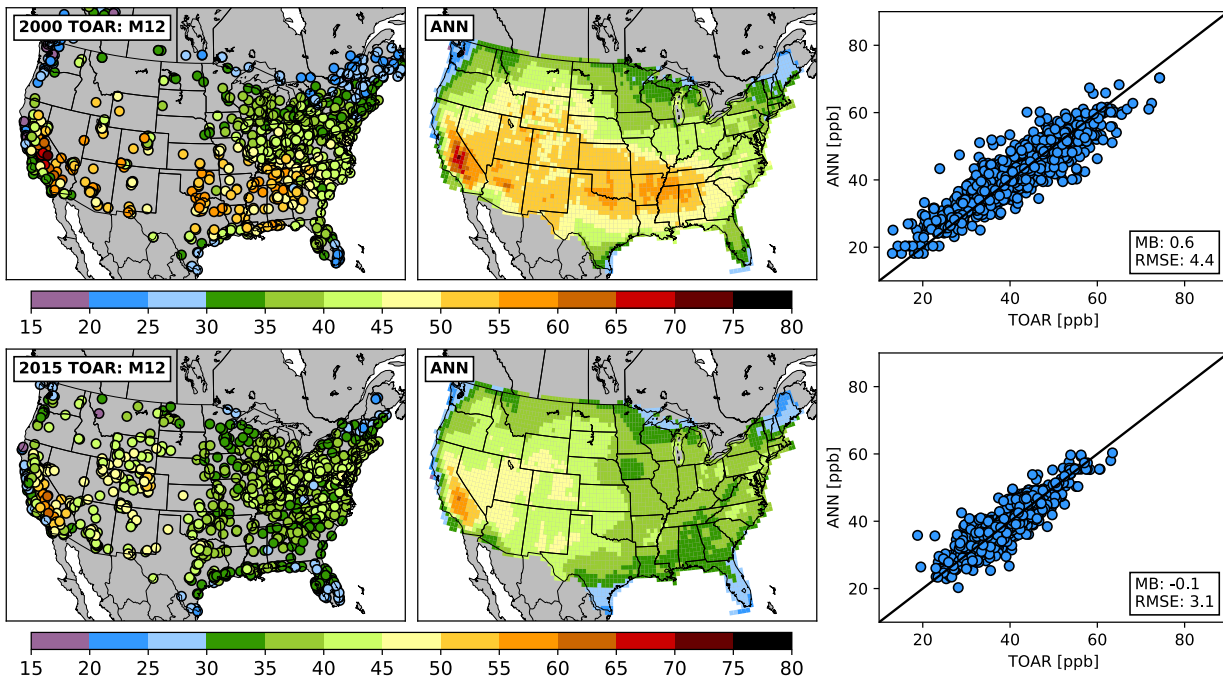
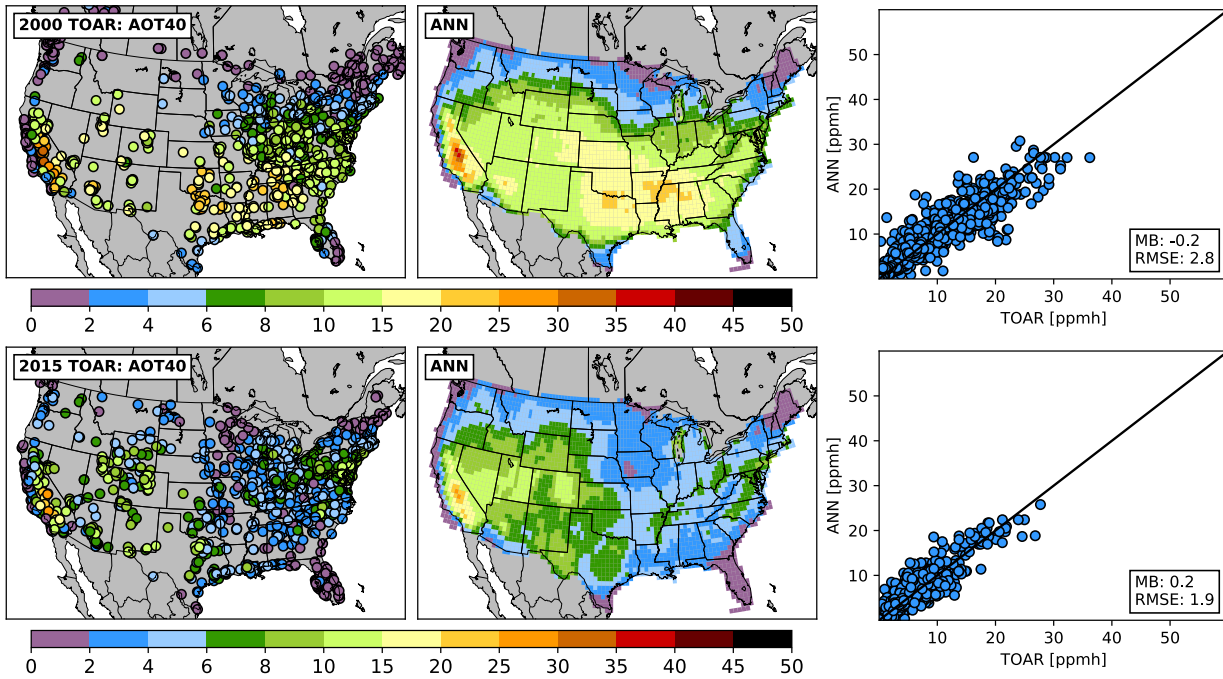


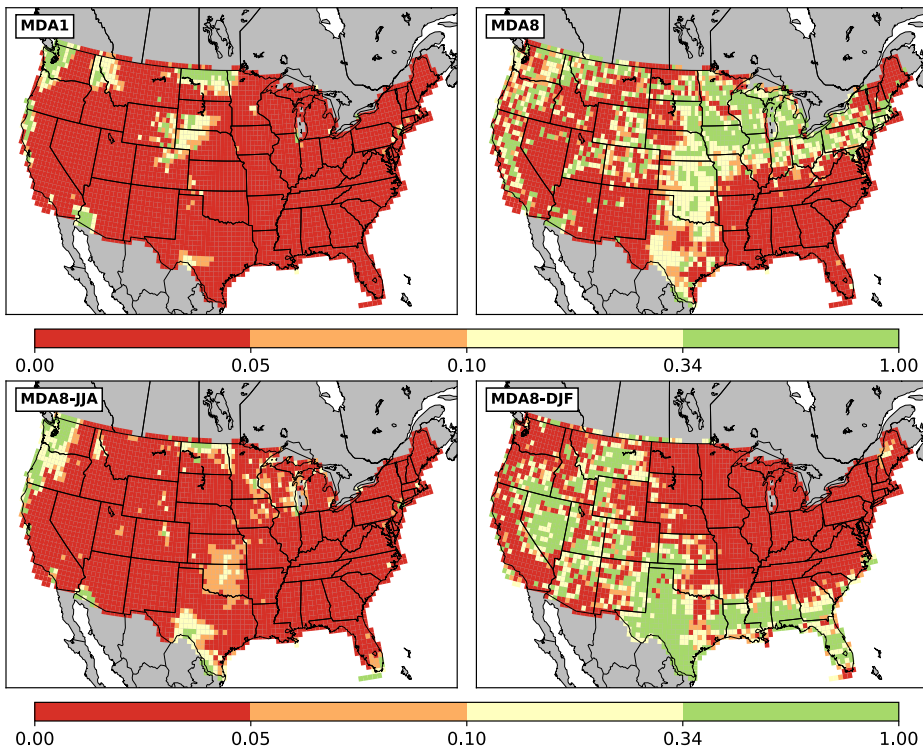
Figure S2: TOAR observations (left) of the annual average of the maximum daily 8-hour concentration (Turner et al. 2016 averaging metric) and predictions using the ANN (center). Concentrations are reported in ppb. Note: color bar has non-uniform intervals. Right: Scatter plot comparing TOAR observations (left) and ANN predictions (center), with each dot representing a monitoring locations. The mean bias (ppb) and root mean square error (ppb) are included in the scatter plot panel.



75 **Figure S3:** TOAR observations (left) of the July-September average of the M12 agriculture metric (8:00-20:00 local time) and predictions using the ANN (center). Concentrations are reported in ppb. Note: color bar has non-uniform intervals. Right: Scatter plot comparing TOAR observations (left) and ANN predictions (center), with each dot representing a monitoring locations. The mean bias (ppb) and root mean square error (ppb) are included in the scatter plot panel.



80 **Figure S4:** TOAR observations (left) of the July-September sum of the daily AOT40 agriculture metric (8:00-20:00 local time) and predictions using the ANN (center). Concentrations are reported in ppmh. Note: color bar has non-uniform intervals. Right: Scatter plot comparing TOAR observations (left) and ANN predictions (center), with each dot representing a monitoring locations. The mean bias (ppmh) and root mean square error (ppmh) are included in the scatter plot panel.



85 Figure S5: The p-values for the trends presented in Fig. 2 from the main text.

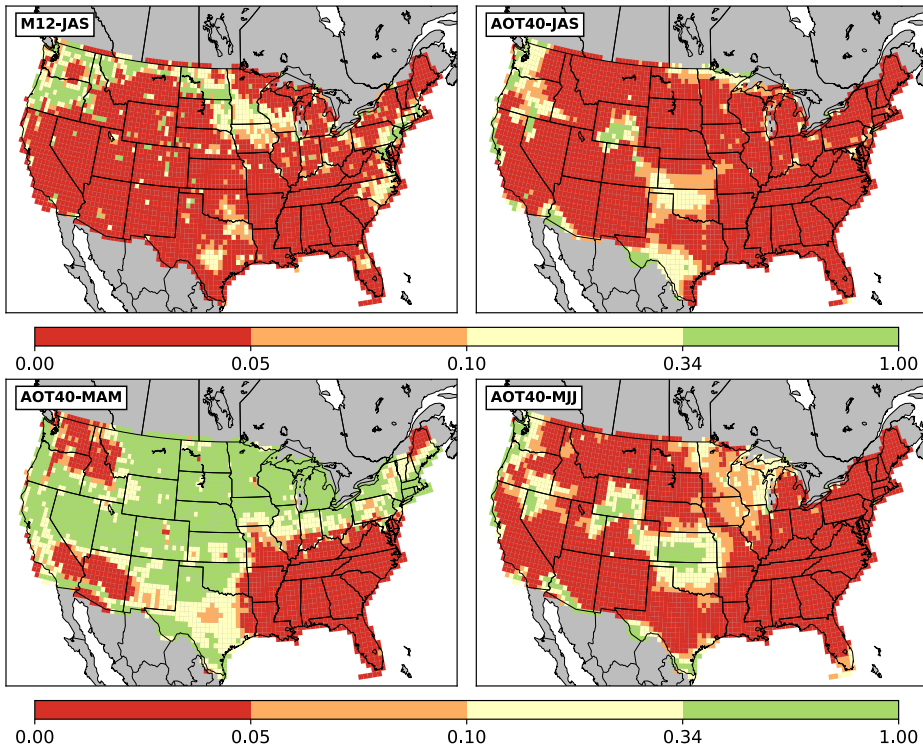
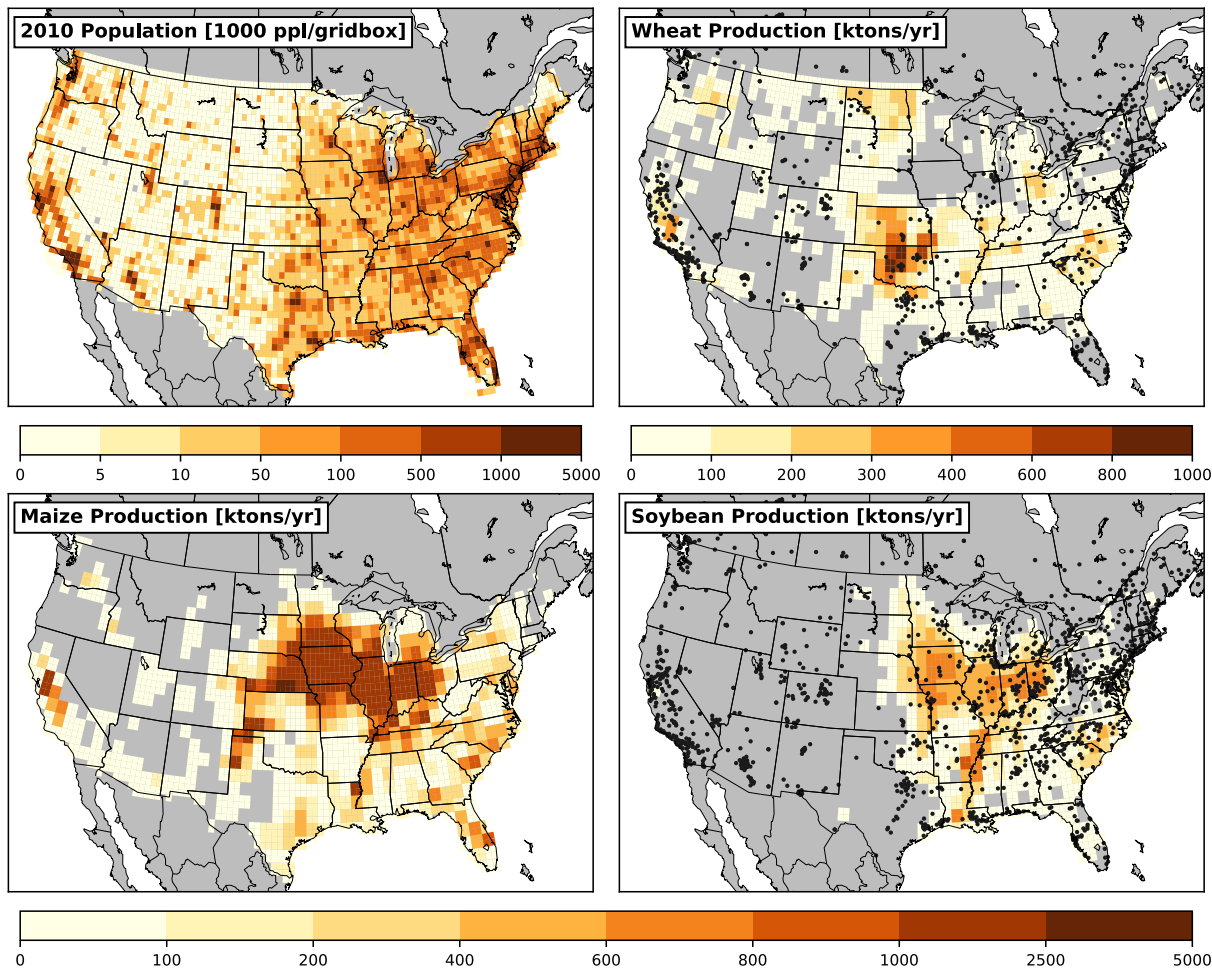
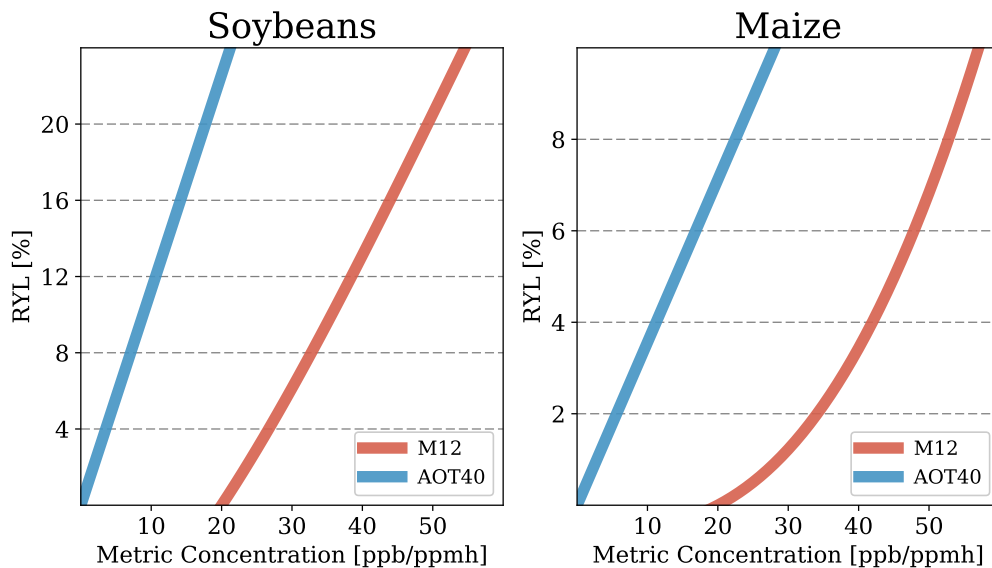


Figure S6: The p-values for the trends presented in Fig. 4 from the main text.



90 **Figure S7: 2010 USA population (top left) and wheat (top right), maize (bottom left), and soybean (bottom right) production maps. Dots in top right panel represent monitoring locations with available data on January 1, 2010 and dots in bottom right panel represent monitoring locations with available data on July 1, 2010.**



95 **Figure S8: Comparison of the M12 and AOT40 concentration-response curves for soybeans and maize. To note: the concentrations are different for the M12 (ppb) and AOT40 (ppmh) metrics.**