

Table S1. Average RMS error (ppb) in predicting a subset of the observing sites for range of interpolation parameters. The numbers in bold denote the final set of parameters used for the interpolation and their RMS error.

$\beta =$	US				EU			
	2.0	2.5	3.0	3.5	2.0	2.5	3.0	3.5
D=10 km, L=500 km	6.53	6.42	6.44	6.50	6.56	6.52	6.61	6.72
D=10 km, L=250 km	7.12	7.08	7.12	7.17	6.70	6.72	6.81	6.92
D=25 km, L=500 km	6.65	6.43	6.38	6.39	6.96	6.78	6.74	6.78
D=25 km, L=250 km	7.20	7.09	7.06	7.08	7.02	6.93	6.93	6.97
D=50 km, L=500 km	7.17	6.85	6.68	6.60	7.39	7.15	7.03	6.98
D=50 km, L=250 km	7.58	7.41	7.31	7.26	7.36	7.25	7.18	7.15

Table S2. Domain mean number of air quality extreme events (AQX) and the surface MDA8 O₃ concentration (ppb) corresponding to the 84th, 50th, and 16th percentiles for each month of the year and day of the week for the 2005-2006 grid-cell interpolated observations (-O) and the UCI CTM (-M) in the US and EU. The 84th and 16th percentile values are given relative to the 50th percentile.

		Unit	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
US-O	AQX	#	0.0	0.3	0.9	11.1	14.0	37.6	30.0	15.2	9.9	0.9	0.0	0.0
	O ₃ 84%	ppb	+7.2	+6.9	+7.0	+8.7	+9.8	+12.7	+11.5	+10.3	+12.3	+10.9	+8.2	+7.4
	O ₃ 50%	ppb	30.5	36.2	43.4	49.5	49.8	51.8	51.5	48.9	43.6	35.3	31.3	28.1
	O ₃ 16%	ppb	-8.5	-7.9	-6.7	-8.6	-11.3	-14.8	-15.1	-13.3	-12.9	-9.9	-7.9	-8.1
US-M	AQX	#	0.4	0.4	1.6	4.8	12.7	25.2	25.4	33.8	12.3	2.9	0.4	0.1
	O ₃ 84%	ppb	+8.5	+10.6	+12.4	+14.4	+19.7	+24.6	+23.5	+24.5	+23.2	+19.4	+14.9	+8.9
	O ₃ 50%	ppb	48.6	53.9	62.4	70.9	74.4	77.5	79.3	79.2	73.2	60.2	51.7	45.3
	O ₃ 16%	ppb	-9.9	-8.4	-10.5	-12.5	-15.6	-21.1	-23.8	-23.0	-18.8	-13.1	-9.4	-10.6
EU-O	AQX	#	0.0	0.5	1.7	17.4	28.9	19.9	46.0	4.0	1.3	0.1	0.1	0.0
	O ₃ 84%	ppb	+7.0	+6.4	+6.8	+7.6	+11.1	+14.0	+17.2	+12.0	+11.7	+7.4	+7.6	+7.8
	O ₃ 50%	ppb	27.4	33.8	41.3	45.1	44.7	43.6	43.8	37.2	35.2	28.5	25.3	24.1
	O ₃ 16%	ppb	-7.7	-7.1	-6.3	-6.2	-6.7	-9.0	-12.0	-9.3	-7.4	-6.7	-7.9	-8.2
EU-M	AQX	#	0.0	0.0	0.6	2.9	24.9	35.7	32.5	16.4	6.9	0.2	0.0	0.0
	O ₃ 84%	ppb	+6.9	+9.3	+10.2	+11.3	+12.9	+14.9	+17.3	+14.9	+15.5	+14.3	+6.1	+5.9
	O ₃ 50%	ppb	44.3	51.6	59.3	67.9	73.9	76.1	73.5	69.6	65.5	53.5	46.4	42.6
	O ₃ 16%	ppb	-8.6	-8.1	-7.5	-9.4	-13.1	-17.7	-19.8	-18.3	-14.6	-6.5	-7.0	-9.5
			Sun	Mon	Tue	Wed	Thu	Fri	Sat					
US-O	AQX	#	8.7	7.5	8.8	9.1	10.8	13.3	11.8					
	O ₃ 84%	ppb	+14.2	+14.1	+14.2	+15.1	+14.8	+14.9	+14.5					
	O ₃ 50%	ppb	40.9	40.2	40.4	40.3	40.5	40.6	41.0					
	O ₃ 16%	ppb	-12.1	-12.4	-12.8	-12.5	-12.6	-12.6	-12.5					
US-M	AQX	#	11.5	10.3	8.8	9.0	9.7	10.1	10.5					
	O ₃ 84%	ppb	+24.7	+24.2	+24.3	+24.5	+24.3	+23.9	+24.5					
	O ₃ 50%	ppb	62.0	62.5	62.5	62.4	62.6	62.6	62.2					
	O ₃ 16%	ppb	-15.8	-16.2	-16.2	-16.0	-16.4	-16.3	-16.0					
EU-O	AQX	#	8.5	8.2	9.0	9.1	11.3	12.3	11.6					
	O ₃ 84%	ppb	+11.7	+11.9	+12.0	+12.8	+13.2	+13.3	+12.8					
	O ₃ 50%	ppb	36.6	35.4	35.4	35.4	35.5	35.5	36.1					
	O ₃ 16%	ppb	-10.5	-10.9	-11.4	-11.5	-11.3	-11.4	-10.9					
EU-M	AQX	#	9.1	10.5	10.0	10.0	10.6	10.3	9.5					
	O ₃ 84%	ppb	+20.0	+20.7	+20.8	+20.9	+20.9	+20.7	+20.1					
	O ₃ 50%	ppb	58.3	58.2	58.2	58.3	58.2	58.2	58.3					
	O ₃ 16%	ppb	-14.5	-13.8	-13.4	-13.7	-13.7	-13.7	-14.3					

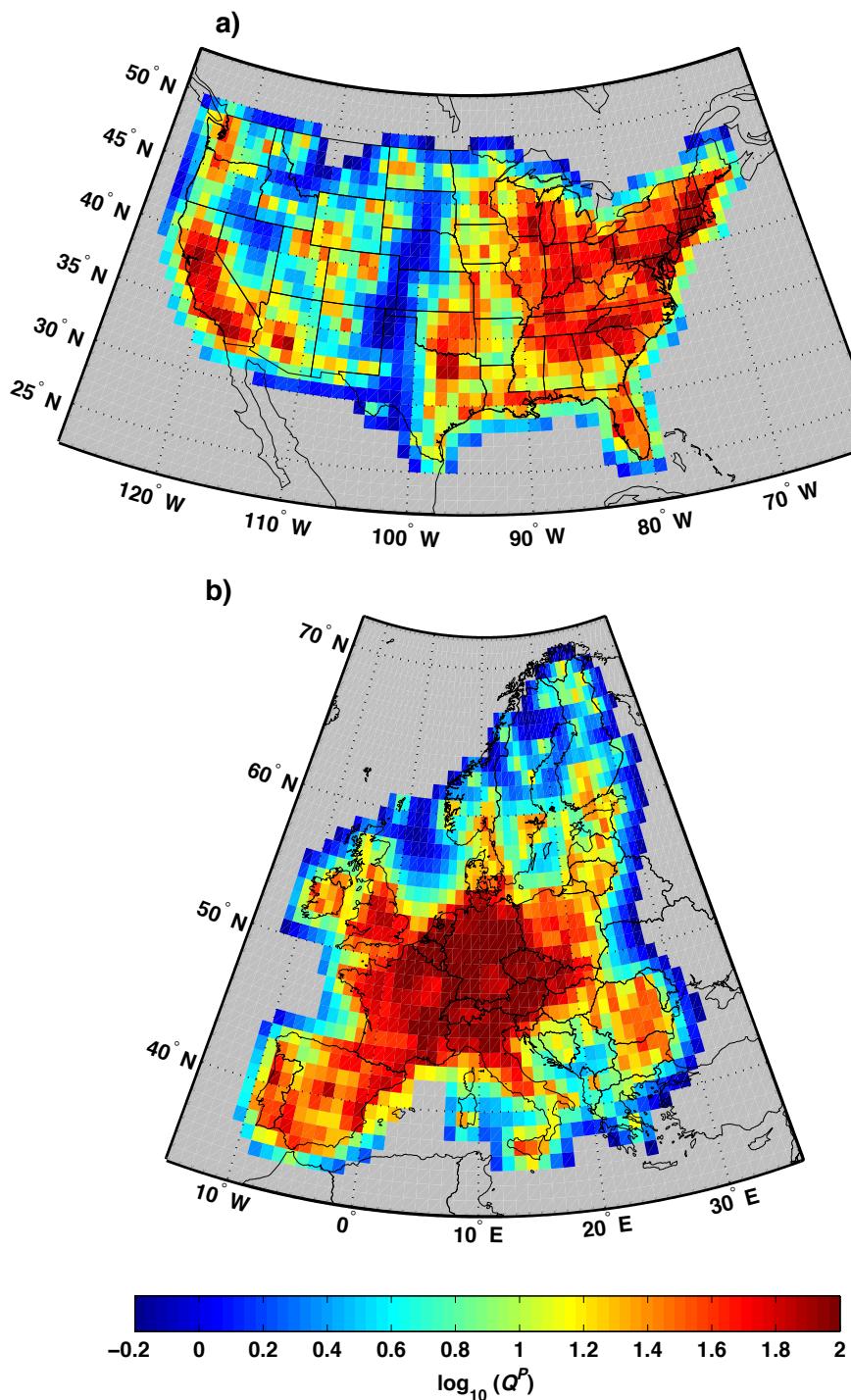


Figure S1. Logarithm of the quality of prediction (Q^P) in the (a) US and (b) EU.

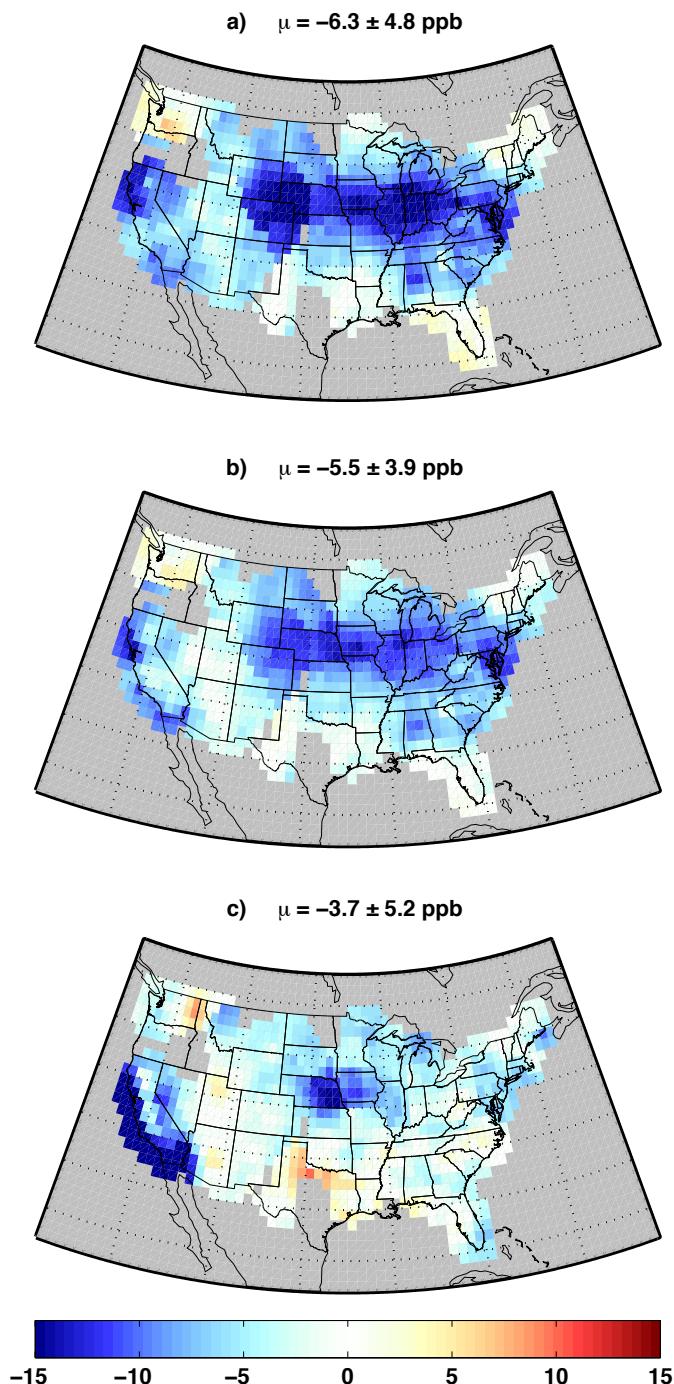


Figure S2. Bias of the gridded MDA8 O₃ concentration (ppb) created using only AQS stations vs. using only CASTNET stations for years 2000-2009 (bias = AQS minus CASTNET). Biases calculated using independent sampling and are shown for the (a) 25th, (b) 50th, and (c) 95th percentiles. This mask includes only grid cells with a Q^P greater than 0.10 for both data sets. The area-weighted mean bias and 1σ for each percentile is given with the graph. All mean biases are negative.

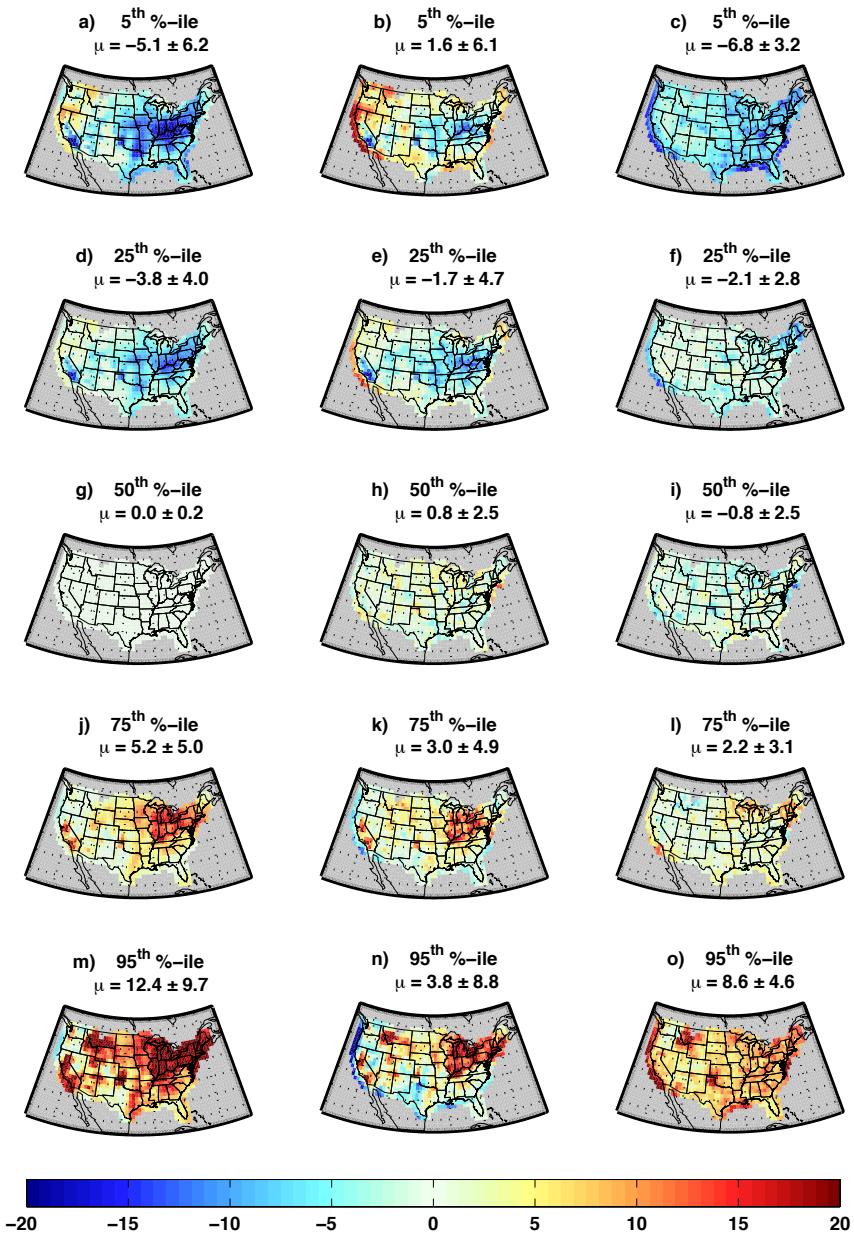


Figure S3. Model mean bias (MB = CTM minus observed) of surface MDA8 O₃ (ppb) in the US. Left column (**a, d, g, j, m**) shows MB calculated using independent sampling, middle column (**b, e, h, k, n**) concurrent sampling, and right column (**c, f, i, l, o**) their difference (independent minus concurrent). First row (**a, b, c**) shows results for the 5th percentile, second row (**d, e, f**) 25th percentile, third row (**g, h, i**) 50th percentile, fourth row (**j, k, l**) 75th percentile, and fifth row (**m, n, o**) 95th percentile. The median of each dataset was subtracted prior to calculating the percentiles and the resulting MB. Percentiles values are calculated as the mean of the value corresponding to the percentile value $\pm 2.5\%$.

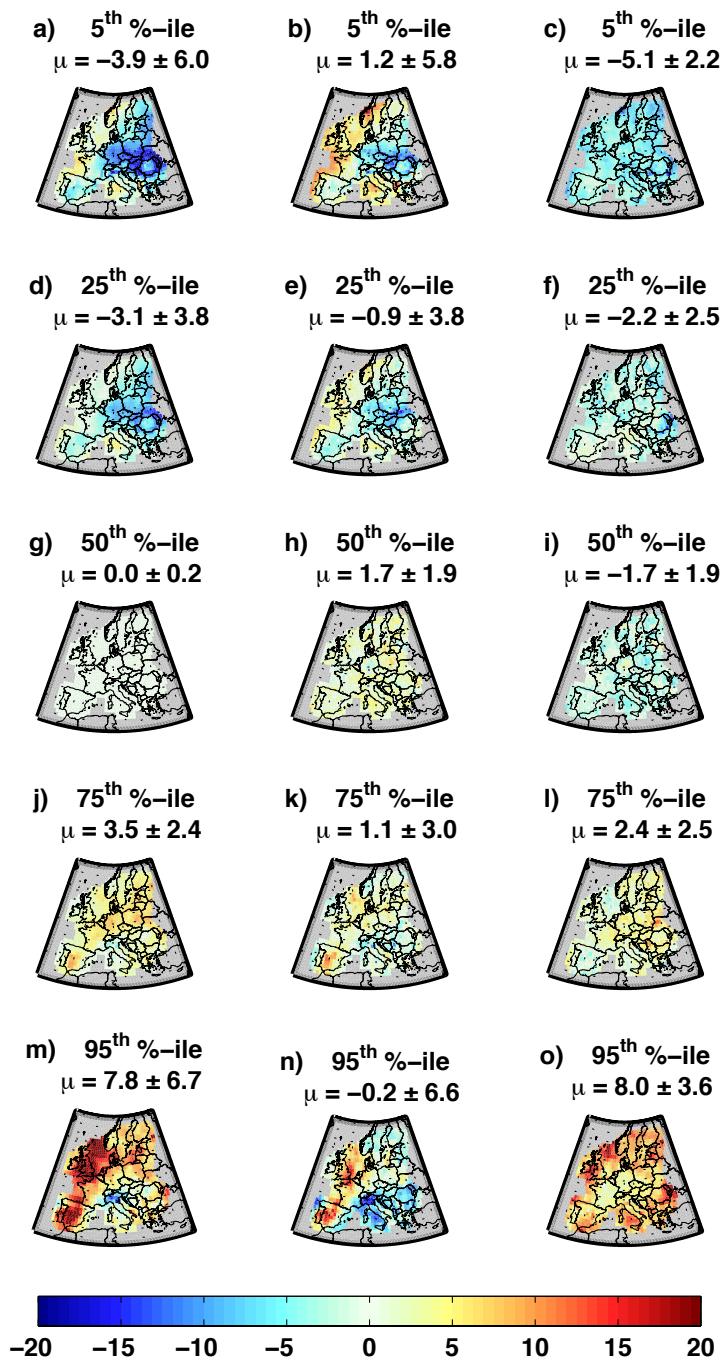


Figure S4. Same as Fig. S3, but for EU.

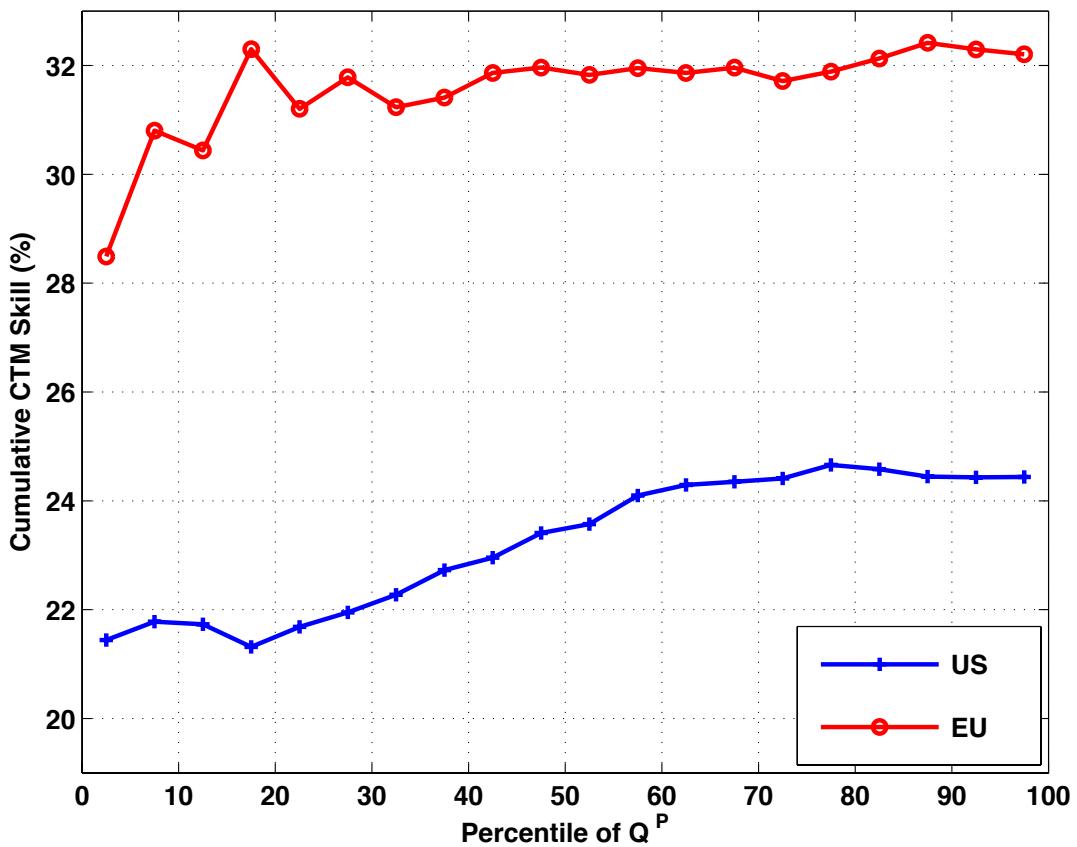


Figure S5. Cumulative average CTM skill (%) for each 5th percentile of Q^P for the US and EU.

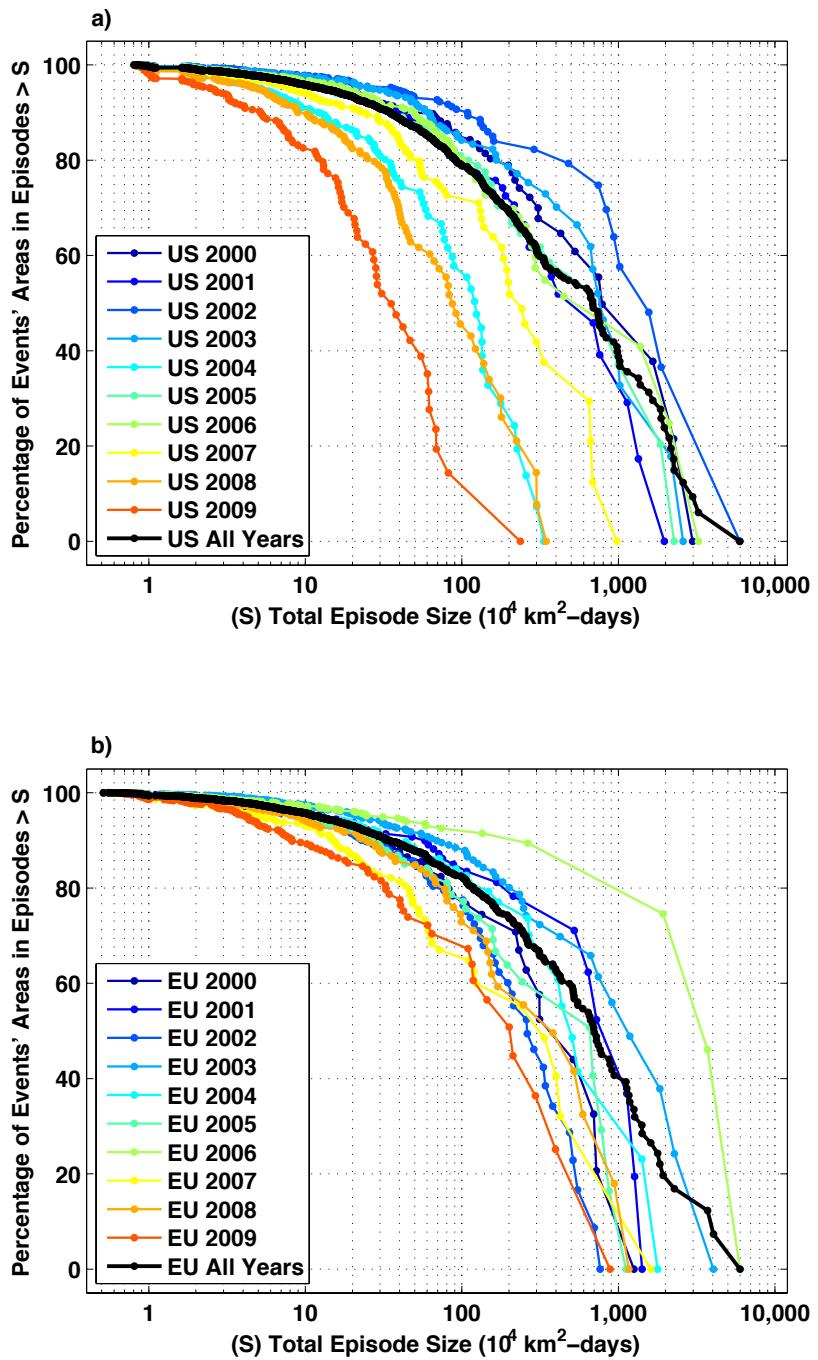


Figure S6. Complementary cumulative distribution function of the percentage of the total areal extent of all individual AQX events (defined as 100 worst days in decade) as a function of episode size ($10^4 \text{ km}^2\text{-days}$) they are clustered into for the (a) US and (b) EU in the 10-yr observations for each year from 2000 to 2009 and the full decade.

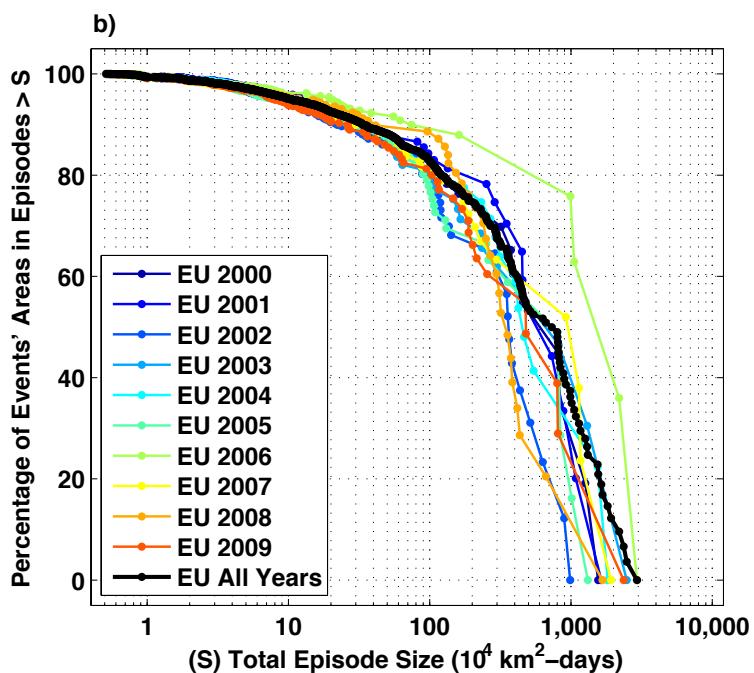
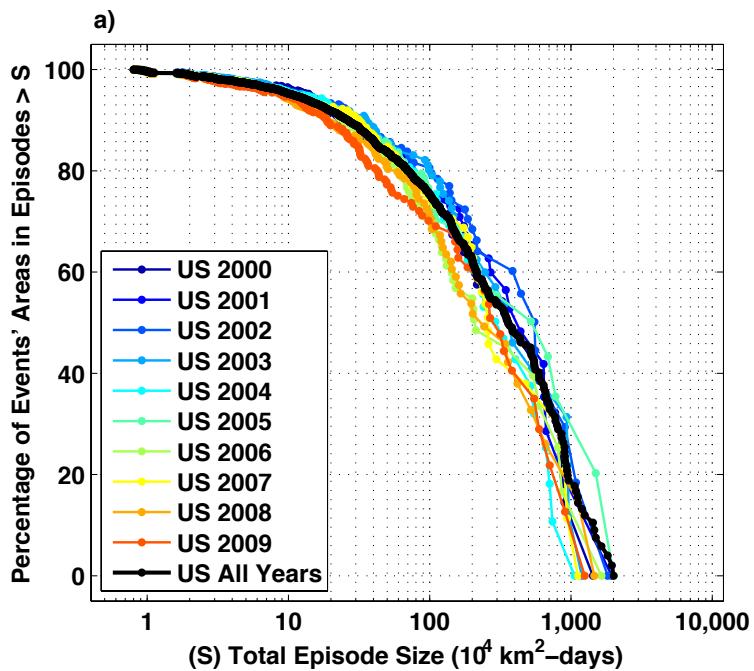


Figure S7. Same as Fig. S6 except that AQX events are normalized so that each grid cell has 10 AQX events per year.

