Supporting materials:

Atmospheric mercury speciation dynamics at the high-altitude Pic du Midi Observatory, southern France

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Table S1. Summary of the atmospheric Hg species, ancillary data, and linear correlations during the 44 identified high PBM events (associated p-values: *, p<0.05, **, p<0.01, no star, p>0.05)

Events	Local Time	Max PBM ($pg\ m^{-3}$)	GEM ($ng\ m^{-3}$)	GOM ($pg\ m^{-3}$)	CO (ppb)	O_3 (ppb)	CPC (nbp/cm^3)	RH (%)	R^2 (GEM *vs* PBM)	R^2 (GOM *vs* PBM)	R^2 (CO *vs* PBM)	R^2 (O_3 *vs* PBM)	R^2 (RH *vs* PBM)
Middle and upper tropospheric intrusions high PBM events													
#1	12/18/2011 23:20	45	1.67	17	114	40.4		86	-0.88**	0.89**	-0.21	0.48*	-0.07
#2	12/24/2011 10:45	69	1.47	15	99	44.6		23	-0.81**	0.4	0.01	-0.1	0.36
#3	12/25/2011 7:25	52	1.73	26	103	42.7		47	-0.60*	0.37*	0.41*	0.15	0.12
#4	01/14/2012 12:35	52	1.67	105	88	42		15	-0.67*	-0.1	-0.26	-0.16	0
#5	01/18/2012 19:30	33	1.55	31	102	42.4		21	-0.65*	0.83**	0	0.4	-0.57*
#6	01/26/2012 6:10	44	1.72	22	114	42.2	441	19	-0.70**	0	0	-0.23	0
#7	02/03/2012 20:40	34	1.91	13	138	41.6	584	57	-0.45*	0.39*	-0.51*	-0.35	-0.63**
#8	02/05/2012 2:40	38	2.10	15	115	47.5	484	47	-0.66*	0.02	0	0.57*	-0.33
#9	02/07/2012 22:00	47	1.64	39	145	51.5	308	22	-0.77*	0.36	0.28	0.63**	-0.53*
#10	02/11/2012 2:40	49	1.62	29	135	44.1	307	19	-0.82**	0.07	-0.28	-0.61**	-0.46*
#11	02/12/2012 7:20	35	1.65	16	122	47.1	173	51	-0.54*	-0.03	-0.1	-0.05	-0.41*
#12	02/21/2012 7:00	85	1.47	25	128	50.2	150	6	-0.76**	0.62**	0.62**	0.39**	-0.81**
#13	02/22/2012 3:40	74	1.76	22	145	52.1	263	7	-073**	0.06	0.66**	0.17	-0.64**
#14	03/01/2012 10:20	40	1.82	53	121	60.8	907	24	-0.58*	0.84**	-0.14	0.96**	-0.73*
#15	03/05/2012 12:20	47	1.69	18	115		345	87	-0.54**	0.85**	-0.31*		-0.27
#16	03/09/2012 3:40	62	1.50	43	129		377	13	-0.70**	0.94**	-0.72**		-0.36
#17	03/10/2012 1:40	98	1.37	70	127		298	4	-0.76**	0.51**	0.03		-0.87**
#18	03/12/2012 14:20	42	1.43	69	113		337	20	-0.62*	0.14*	0.18		-0.16
#19	03/14/2012 0:55	40	1.38	131	111	50.3	1609	20	-0.48**	0.06	0.02	0.13	0.18
#20	03/23/2012 23:55	51	1.34	68	105	56.2	539	13	-0.89**	0.37*	0	-0.1	-0.02
#21	04/07/2012 1:25	38	1.82	26	126	59.5	1369	47	-0.40*	0.80**	0.75**	0.77**	-0.60**
#22	04/09/2012 1:25	41	1.86	42	110	60.9	391	91	-0.69*	0.97**	0.11	0.90**	-0.94**
#23	04/16/2012 13:25	39	1.57	38	121	55.4	600	59	-0.45*	0.81**	0.67*	0.48*	-0.72*
#24	04/17/2012 7:25	84	1.54	29	123	62.5	253	5	-0.84**	0.84**	0.34	0.58*	-0.83**
#25	04/22/2012 8:50	33	1.65	17	116	66.9	206	92	-0.80*	0.29	0.96**	-0.02	-0.46
#26	05/16/2012 5:05	69	1.25	153	90	98.5		6	-0.78**	0.62**	-0.30*	0.61**	-0.63**
#27	07/15/2012 10:00	34	1.55	92	99	55.4	397	32	-0.58*	0.88**	0.31	0.82**	-0.77**
#28	09/14/2012 2:10	59	1.40	21	116			6	-0.80*	0.26	0.56		-0.54
#29	09/20/2012 2:15	54	1.55	29	104	61.7		4	-0.57*	-0.36	0.34	0.57*	-0.48
#30	11/13/2012 3:15	55	1.56	58				4	-0.58**	0.54*			-0.74**
Anthropogenic impacted high PBM events													
#31	01/15/2012 19:15	48	1.98	19	144	45.7		46	0.70**	-0.60*	0.57*	0.39	-0.07
#32	01/27/2012 0:10	34	1.97	5	129	39.7	1915	94	0.12	-0.80**	0.95**	-0.14	0.2
#33	03/03/2012 3:35	54	2.14	9	147		3351	82	0.38*	0	0.75**		0.03
#34	03/15/2012 18:15	53	1.91	16	160	70	2587	57	-0.07	-0.04	0.38	0.51*	0
#35	06/09/2012 10:45	33	1.75	42	96	60.2	4638	60	0.78**	-0.82**	0.39	-0.21	0.68*
#36	08/28/2012 10:10	39	2.03	39	169	60.9	158	50	-0.03	0.27	0.42	0.45*	-0.52*
Mixed high PBM events													
#37	01/24/2012 19:30	64	2.00	13	102	44.4	238	10	0	0.18	0.1	0.38	-0.66**
#38	02/13/2012 5:20	41	1.84	27	125	47	306	23	0.33	0.86**	0	0.23	-0.92**
#39	02/18/2012 12:20	40	1.67	29	94	43.9	593	36	-0.2	0.2	-0.04	0	-0.38*
#40	02/27/2012 19:00	33	2.03	64	103	62.7	1135	4	0.37	0.72*	0.2	0.77*	-0.92**
#41	03/11/2012 14:20	42	1.49	60	126		419	14	-0.31	0.91**	0		-0.81**
#42	03/16/2012 22:15	35	1.85	101	111	55.7	2278	50	-0.17	0.74**	-0.46*	-0.76**	0
#43	07/10/2012 7:20	34	1.65	51	107	56.6	1058	70	-0.27	0.11	0.06	0.09	-0.35
#44	08/29/2012 10:50	45	1.55	73	114	50.2	122	57	-0.08	0	0.09	0.05	-0.08

Table S2. Pearson correlation coefficient r^2 between PBM/GOM ratio, and PBM concentration as well as Hysplit simulated air mass meteorological parameters and atmospheric pollutants during the 38 high PBM events (6 high PBM events related to anthropogenic pollution were not included) (Pearson correlation coefficient: *, $p<0.05$, **, $p<0.01$, no star, $p>0.05$).

Correlation r^2	PBM/GOM ratio	PBM concentration	Air pressure	Air Temp	RH	Solar radiation	CO	Ozone
PBM concentration	0.21*							
Air pressure	-0.07	-0.43**						
Air Temp	-0.18*	-0.39**	0.74**					
RH	0.03	-0.02	0.41**	0..09				
Solar radiation	-0.33**	-0.04	0	0.21**	0			
CO	0.04	0.02	-0.01	-0.11	0	-0.09		
Ozone	-0.11	0.02	-0.10	-0.02	-0.07	0.64**	-0.05	
Particle number concentration	-0.22*	-0.15*	0	0.06	-0.02	0.05	-0.03	0

Table S3. Summary of the atmospheric Hg species, ancillary data, and linear correlations during the 61 identified high PBM events

Events	Local Time	Max GOM (pg m^{-3})	GEM (ng m^{-3})	PBM (pg m^{-3})	CO (ppb)	O_3 (ppb)	CPC (nbp/cm^3)	RH (%)	R^2 (GEM *vs* GOM)	R^2 (PBM *vs* GOM)	R^2 (CO *vs* GOM)	R^2 (O_3 *vs* GOM)	R^2 (RH *vs* GOM)
Middle and upper troposphere intrusions high GOM events													
#1	11/30/2011 19:00	97	1.91	2	91	51.2		12	-0.40*	0	-0.23	0.70*	-0.38
#2	02/25/2012 0:20	95	1.62	9	94	50.7	321	33	-0.90*	0	-0.22	0.99**	-0.92*
#3	03/13/2012 12:55	136	1.50	24	102	52.1	499	13	-0.88*	0.27	-0.82*	0.74*	-0.28
#4	05/14/2012 2:25	150	1.72	10	97	72.1		5	-0.58*	0.12	0	0.63*	-0.56*
#5	05/14/2012 12:25	112	1.73	15	111	68.1		16	-0.65*	0.82**	-0.38	0.81*	-0.73*
#6	05/14/2012 23:05	167	1.42	12	95	66.2			-0.73**	0	-0.59*	0.76**	
#7	05/16/2012 5:05	153	1.25	69	90	98.5		6	-0.81**	0.61*	-0.50*	0.94**	-0.55*
#8	05/17/2012 1:05	134	1.64	3	93	56.8		22	-0.58*	-0.38	-0.80**	0.38*	-0.95**
#9	05/22/2012 22:25	91	1.54	5	104	53.2		95	-0.66*	0.10	0	0.83**	-0.62*
#10	06/04/2012 5:30	126	1.85	15	99	61.8	512	30	-0.51*	0.43	0	0.84**	-0.92**
#11	06/05/2012 6:10	187	1.68	12	102	68.1	237	20	-0.52*	0	-0.01	0.45*	-0.65*
#12	06/09/2012 4:45	189	1.40	18	94	61.1	343	45	-0.73**	0	-0.50*	0.38*	-0.52*
#13	06/25/2012 1:50	240	1.84	17	92	70.3	295	13	-0.47*	0	-0.14	0.80**	-0.74*
#14	07/12/2012 11:20	156	1.36	20	99	62.3	503	26	-0.69*	0	0	0.70*	-0.92**
#15	07/15/2012 10:00	92	1.55	34	99	55.4	397	32	-0.67**	0.89**	0.34	0.87**	-0.94**
#16	07/17/2012 6:40	223	1.28	7	91	50.9	144	46	-0.83**	-0.38	-0.19	0.70**	-0.12
#17	07/19/2012 3:20	104	1.44	16	100	59.3	1045	36	-0.72**	-0.10	-0.25	0.67*	-0.59*
#18	07/22/2012 4:00	227	1.52	12	116	60.9	99	11	-0.68**	0.08	0.36	0.83**	-0.78**
#19	07/23/2012 2:00	248	1.45	18	112	61.5	166	11	-0.69**	0	-0.05	0.42*	-0.38*
#20	08/01/2012 8:30	118	1.69	10	115	69.6	666	30	-0.61**	-0.20	-0.61**	0.50*	-0.87**
#21	09/15/2012 2:50	251	1.22	0	91	66.0		25	-0.26*	0	0.42	0.64*	-0.31
#22	09/16/2012 0:50	295	1.48	0	85	71.3		16	-0.52*	0	-0.59	0.62*	-0.90**
#23	10/16/2012 22:55	110	1.43	1	83	41.4		63	-0.82**	-0.32	-0.50*	0.59*	-0.62
#24	11/07/2012 20:35	109	1.31	23	95	70.3		11	-0.42*	0.46*	-0.03	0.89**	-0.28
Lower free troposphere over the North Atlantic Ocean high GOM events													
#25	12/22/2011 12:05	164	0.95	0	83	41.4		12	-0.40*	-0.24	0	-0.36*	-0.04
#26	01/14/2012 22:35	119	2.42	17	88	42.7		12	-0.54*	-0.08	-0.46	-0.66*	-0.21
#27	01/16/2012 5:15	100	1.55	28	79	41.1		38	-0.80**	0.33	-0.55*	-0.62**	0.22
#28	03/17/2012 2:15	140	1.65	25	113	54.4	1028	60	-0.77**	0	-0.45*	-0.53*	0.22
#29	06/07/2012 12:10	131	1.34	12	82	32.8	406	59	-0.87**	0	-0.76**	-0.55*	-0.53*
#30	06/26/2012 4:30	209	1.59	25	78	23.3	134	43	-0.44*	0.34*	-0.53*	-0.45*	-0.75**
#31	06/30/2012 5:05	151	1.45	11	96	60	760	51	-0.92**	-0.77**	-0.86**	-0.42*	0
#32	08/17/2012 8:35	98	1.55	1	94	46.2	562	51	-0.95**	-0.91**	-0.86**	-0.76**	-0.03
#33	10/03/2012 10:50	95	1.61	18	87	49.8		44	-0.36*	0.35*	-0.64*	-0.75*	-0.89**
Mixed high GOM events													
#34	12/23/2011 18:45	109	1.80	7	136	38.6		38	-0.82**	0	0.09	-0.06	0.71*
#35	01/13/2012 22:35	122	1.80	23	88	42.7		12	0	-0.02	0	-0.19	-0.15
#36	02/25/2012 18:20	91	1.63	3	88	41.4	216	78	0.06	0	-0.02	0.07	0.26
#37	03/13/2012 22:55	151	1.38	19	102	50	1404	15	-0.87*	0.11	-0.74*	0.26	-0.78*
#38	03/14/2012 12:55	115	1.53	23	114	51.5	1263	23	-0.88*	0	-0.02	0.04	-0.20
#39	03/16/2012 10:15	94	1.91	24	103	54.7	907	27	0	-0.03	-0.59*	-0.76**	-0.68**
#40	03/31/2012 0:55	109	1.65	9	101	58.5	329	33	-0.12	0.01	-0.15	0.22	-0.32
#41	04/01/2012 10:55	105	1.85	7	97	53.1	766	36	0.01	-0.03	-0.61*	0	-0.59*
#42	04/09/2012 7:25	145	1.85	11	110	61.5	226	32	-0.06	-0.06	0	0.15	-0.33
#43	04/09/2012 18:10	93	1.46	22	114	62.2	1325	31	-0.24	0.07	-0.55*	0.30	-0.52*
#44	04/17/2012 17:25	93	1.65	3	91	44.8	348	93	-0.50*	-0.29	-0.21	-0.02	0
#45	05/29/2012 6:25	116	2.12	6	106	58.5	480	38	0.20	-0.32	-0.59*	0.64**	-0.86**
#46	05/30/2012 8:25	133	2.15	7	104	58.5	398	29	0.12	-0.26	-0.76**	0.54*	-0.92**
#47	06/05/2012 10:10	211	1.53	10	83	38.3	1061	48	-0.50*	-0.08	-0.22	0	-0.08

#48	06/16/2012 5:55	91	1.50	9	99	56.3	1971	68	-0.58*	-0.58*	-0.03	0	-0.42*
#49	06/27/2012 4:30	178	1.45	6	89	57.8	475	22	-0.53*	-0.50*	-0.75**	0.23	-0.73**
#50	06/29/2012 3:05	100	1.42	23	94	64.1	1005	36	-0.87**	-0.10	-0.57*	0	-0.79*
#51	07/16/2012 2:40	143	1.89	22	105	57.3	118	17	0	0.62*	0.33	0.80**	-0.66**
#52	07/18/2012 0:40	185	1.89	1	96	49.8	296	19	0.27	-0.58*	0.06	0.09	-0.56*
#53	07/20/2012 23:20	105	1.48	7	102	43.4	436	62	-0.68**	-0.30	-0.08	-0.06	-0.10
#54	08/10/2012 21:15	119	1.59	4	104	64.2	621	28	-0.38*	-0.21	-0.22	0.07	-0.73*
#55	08/11/2012 7:15	105	1.46	24	96	48.0	224	30	-0.94*	0.88*	-0.19	-0.37	-0.74*
#56	08/12/2012 3:15	115	1.55	10	103	55.0	732	41	-0.65**	0	-0.75**	0.08	-0.53*
#57	08/16/2012 10:35	100	1.75	18	142	58.1	450	37	-0.02	0.02	0.33	0.34	-0.13
#58	08/29/2012 6:50	135	1.36	15	113	51.6	146	62	-0.69**	0	0	0.1	-0.39*
#59	09/08/2012 20:50	92	1.68	6	129	58.8		61	-0.58*	-0.48	0.04	-0.06	-0.39
#60	09/23/2012 13:20	153	1.34	7	84	54.3		40	-0.77**	0.17	-0.89**	-033	-0.96
#61	10/02/2012 4:50	189	1.48	0	80	49.5		45	-0.84**	-0.84**	-0.68*	-0.32	0.16

Figure S1 Annual time series of GEM, GOM and PBM concentrations at PDM Observatory

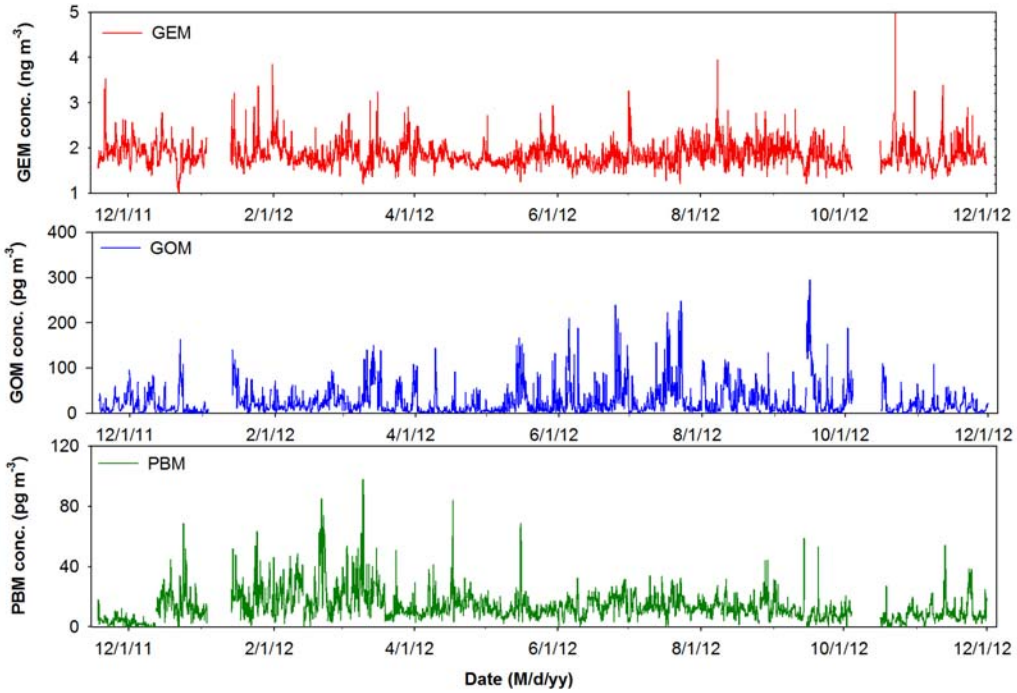

Figure S2. 240-h Hysplit air mass backward trajectories and air pressure for the 30 high PBM events that showed striking anti-correlations between PBM and GEM concentrations.

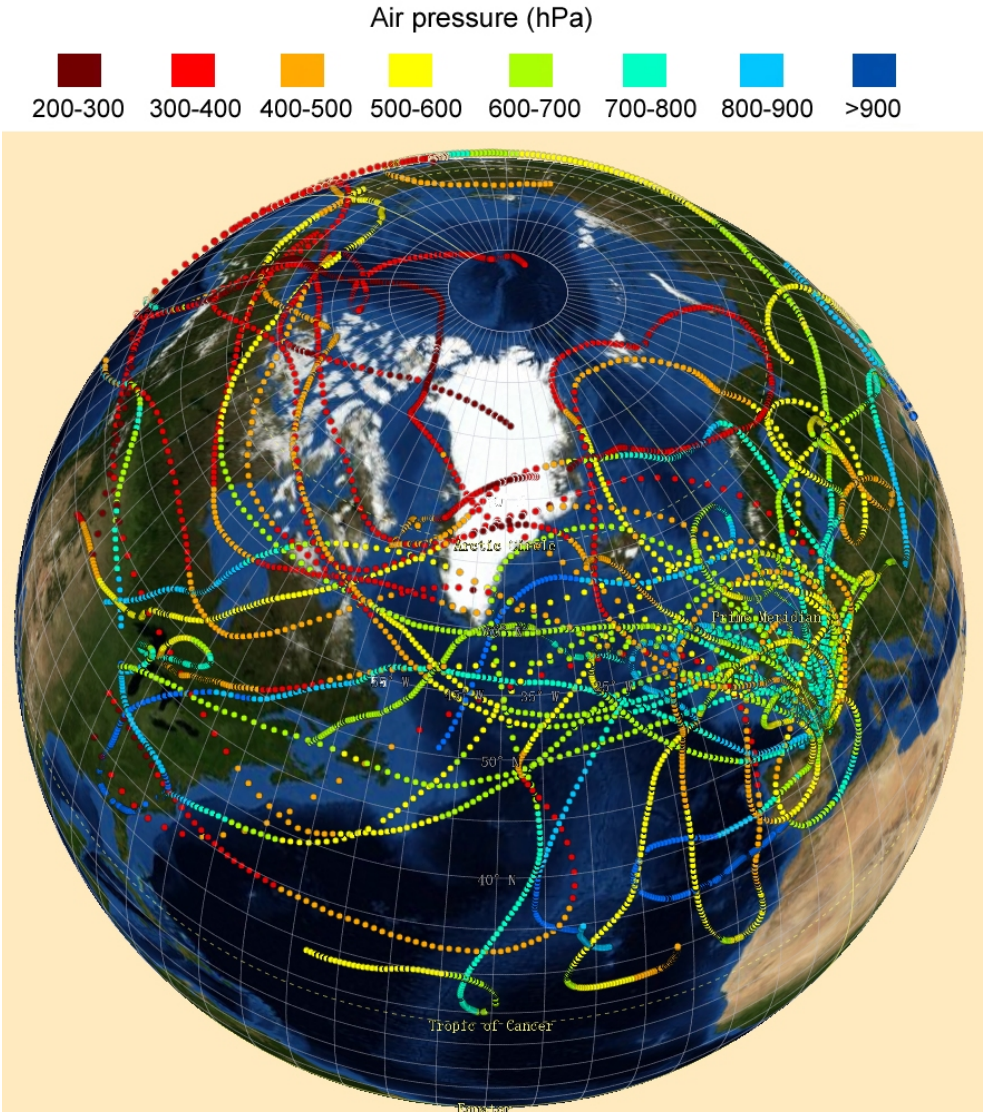

Figure S3. 240-h Hysplit air mass backward trajectory and air pressure for the 6 high PBM events related to anthropogenic pollution.

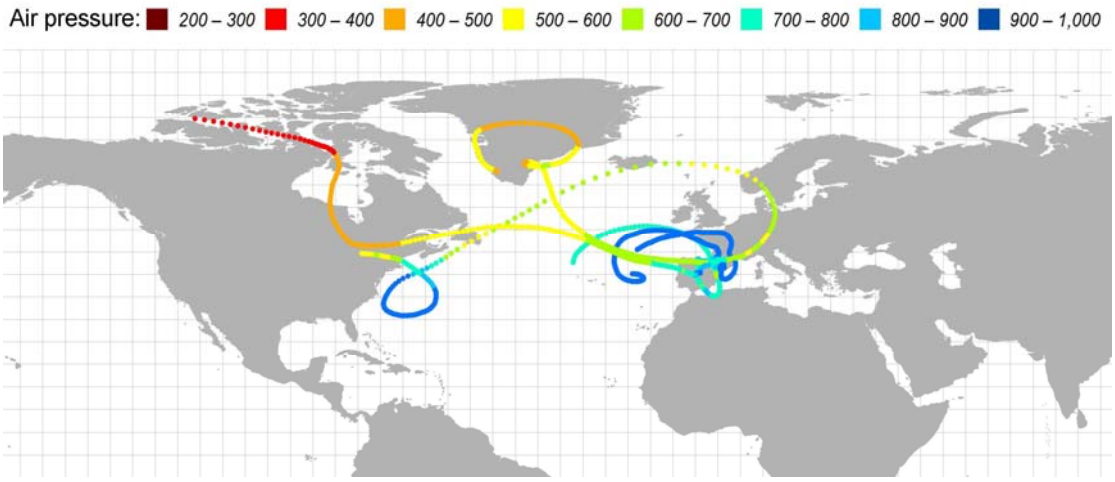

Figure S4. 240-h Hysplit air mass backward trajectory and air pressure for the 8 mixed high PBM events

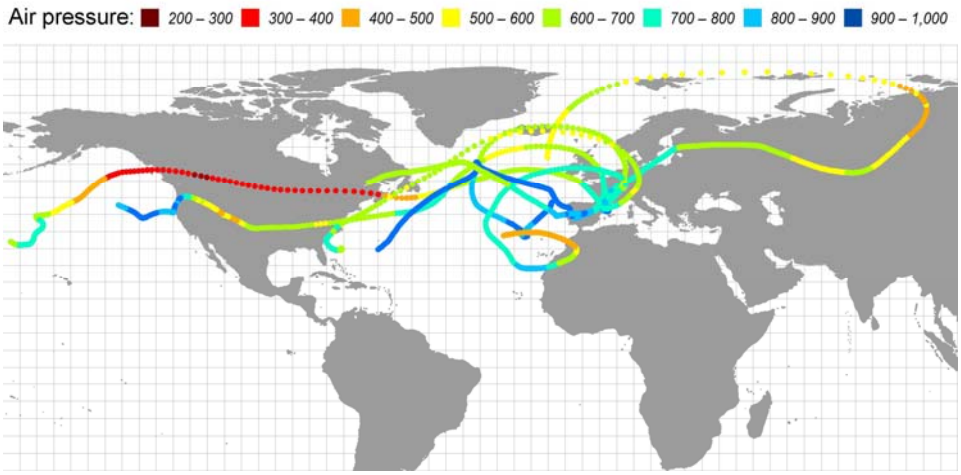

Figure S5. 168-h Hysplit air masses backward trajectory and air pressure for the 24 high GOM events with significant positive correlations between GOM and ozone concentrations.

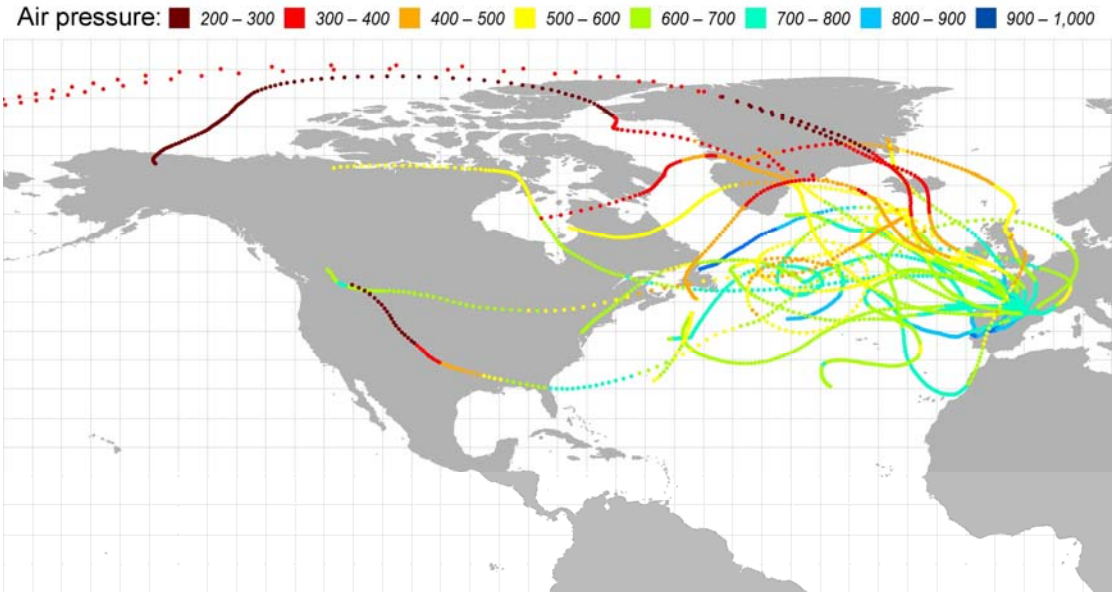

Figure S6. 168-h Hysplit air mass backward trajectory and air pressure for 9 high GOM events with significant anti-correlations between GOM and ozone concentrations.