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Use of a global model to understand speciated atmospheric mercury observations at five high-elevation sites

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Table S-1: Mean daily concentrations and standard deviations for GEM, RM, O_3, and WV by site and by season for all observations, water-vapor screened observation (WV < 75th percentile by season), and modeled data from the standard run. Winter, spring, summer, fall, and are defined as December-February, March-May, June-August, September-November. N refers to the number of daily means used in the calculation for the mean and standard deviation for all parameters.

All Observations

			GEM		RM		O_3		WV	
Location	Season	N	mean	σ	mean	σ	mean	σ	mean	σ
LABS	Winter	31	1.73	0.27	24	9	36.2	7	5.9	2.8
LABS	Spring	29	2.2	0.26	27	18	52.9	5.3	8.5	2.7
LABS	Summer	29	1.43	0.13	15	20	22.2	5	14.2	1.6
LABS	Fall	14	1.7	0.15	11	5	29.2	6.8	13.2	3.5
MBO	Spring	183	1.6	0.17	32	46	48.7	8.1	3.6	1.1
MBO	Summer	80	1.44	0.1	44	45	42.4	8.3	5	1.5
DRI	Winter	214	1.73	0.57	14	16	--	--	3.1	1
DRI	Spring	189	1.6	0.26	25	22	--	--	3.5	1
DRI	Summer	159	1.36	0.35	68	42	49.2	9.6	4.4	1.3
DRI	Fall	143	1.66	0.38	39	26	--	--	3.6	1
NV02	Summer	64	1.79	1.12	32	16	40.1	6.2	4.5	2
SPL	Spring	32	1.59	0.19	27	22	44.9	7.4	3.9	0.8
SPL	Summer	25	1.59	0.21	34	22	36.7	8	4.6	1.1
All-Sites	All Seasons	1206	1.65	0.32	30	24	40.3	7.2	6	1.6

WV Filtered Observations

			GEM		RM		O_3		WV	
Location	Season	N	mean	σ	mean	σ	mean	σ	mean	σ
LABS	Winter	25	1.71	0.29	25	9	37.7	6	5	2.4
LABS	Spring	23	2.21	0.28	31	18	52.4	5.1	7.6	2.2
LABS	Summer	23	1.42	0.12	15	20	22	5.1	14.1	1.5
LABS	Fall	11	1.7	0.15	11	5	29.2	6.8	13.2	3.5
MBO	Spring	146	1.62	0.16	33	47	50.7	7.2	3.1	0.8
MBO	Summer	64	1.4	0.11	64	49	45.8	7.6	3.7	0.7
DRI	Winter	171	1.79	0.55	17	17	--	--	2.7	0.6
DRI	Spring	151	1.6	0.25	27	24	--	--	3.1	0.6
DRI	Summer	127	1.28	0.29	76	45	50.6	9.1	3.7	0.8
DRI	Fall	114	1.66	0.37	44	27	--	--	3.2	0.6
NV02	Summer	51	1.55	0.71	34	17	40.6	5.7	3.7	1.2
SPL	Spring	26	1.59	0.21	28	21	46.2	6.7	3.7	0.7
SPL	Summer	20	1.57	0.23	42	20	38.2	6.8	4.1	0.8
All-Sites	All Seasons	908	1.62	0.29	34	25	41.3	6.6	5.5	1.3

Table SI-1 Continued:

GEOS Chem Standard Model

			GEM		RM		O_3		WV	
Location	Season	N	mean	σ	mean	σ	mean	σ	mean	σ
LABS	Winter	31	1.77	0.18	142	59	52.6	7.1	3.7	2.6
LABS	Spring	29	2.1	0.27	87	31	52.9	5.2	6.5	2.7
LABS	Summer	24	1.61	0.06	21	17	32.2	10	9.9	2
LABS	Fall	11	1.89	0.15	36	24	46.8	6.7	10.3	1.2
MBO	Spring	188	1.84	0.14	111	55	51.2	8.9	1.6	0.9
MBO	Summer	84	1.67	0.09	135	58	50.9	11.6	2.3	1.2
DRI	Winter	204	1.87	0.13	57	35	--	--	1.6	0.8
DRI	Spring	166	1.91	0.11	69	36	--	--	2.1	0.8
DRI	Summer	177	1.7	0.08	72	29	52.3	7.4	3	1.1
DRI	Fall	154	1.74	0.08	63	28	--	--	2.3	0.9
NV02	Summer	77	1.76	0.07	62	21	48.1	7.2	3	1.1
SPL	Spring	32	1.79	0.14	116	41	57.9	4.7	1.4	0.3
SPL	Summer	29	1.71	0.09	119	43	60.4	4.8	1.9	0.7
All-Sites	All Seasons	1206	1.8	0.12	84	37	50.5	7.4	3.8	1.3

Table S-2: Interspecies correlation statistics by site and season. N is the number of daily means used in the calculations and $P < 0.05$ indicates a significant correlation according to ANOVA analysis. Observations were filtered according to the WV cutoff described in the text. Units for slopes are: RM:GEM = pg ng^{-1}, RM: O_3 = pg m^{-3} $ppbv^{-1}$, RM:WV = pg m^{-3} g^{-1} kg

			RM:GEM		RM:O_3		RM:WV	
Site	Season	Parameter	Obs	Model	Obs	Model	Obs	Model
LABS	Winter	N	25	31	22	31	25	31
		R	-0.27	-0.68	-0.05	0.93	-0.37	-0.77
		Slope	-8.7	-219	0.12	8	-1.5	-17
		Intercept	40	529	21	-260	33	207
		P < 0.05?	no	yes	no	yes	no	yes
LABS	Spring	N	23	29	23	29	23	29
		R	-0.45	0.03	-0.06	0.76	-0.87	-0.72
		Slope	-29	3.6	-0.21	4.5	-7.1	-8
		Intercept	95	79	43	-149	86	140
		P < 0.05?	yes	no	no	yes	yes	yes
LABS	Summer	N	23	24	23	24	23	24
		R	-0.64	0.57	0.02	0.62	-0.85	-0.5
		Slope	-107	146	0.09	1	-11	-4.2
		Intercept	167	-215	13	-12	176	63
		P < 0.05?	yes	yes	no	yes	yes	yes
LABS	Fall	N	11	11	11	11	11	11
		R	-0.44	-0.02	-0.36	0.37	0.05	-0.77
		Slope	-16	-3.7	-0.29	1.3	0.07	-15
		Intercept	39	42	20	-27	10	192
		P < 0.05?	no	no	no	no	no	yes

Table S-2 cont.

			RM:GEM		$RM:O_3$		RM:WV	
Site	Season	Parameter	Obs	Model	Obs	Model	Obs	Model
MBO	Spring	N	146	188	146	188	146	187
		R	-0.44	-0.53	0.14	0.82	0.17	-0.52
		Slope	-129	-205	0.88	5.1	10	-31
		Intercept	242	487	-12	-148	1.4	160
		P < 0.05?	yes	yes	no	yes	yes	yes
MBO	Summer	N	64	84	64	84	64	84
		R	-0.7	-0.42	0.43	0.85	-0.44	-0.65
		Slope	-316	-270	2.5	4.2	-19	-30
		Intercept	501	587	-60	-79	134	204
		P < 0.05?	yes	yes	yes	yes	yes	yes

Table S-2 cont.

Site	Season	Parameter	RM:GEM Obs	RM:GEM Model	RM:O_3 Obs	RM:O_3 Model	RM:WV Obs	RM:WV Model
DRI	Winter	N	171	244	--	--	171	153
		R	0.44	-0.57	--	--	-0.44	-0.5
		Slope	14	-207	--	--	-11	-15
		Intercept	-7	424	--	--	47	100
		P < 0.05?	yes	yes	--	--	yes	yes
DRI	Spring	N	151	276	--	--	151	184
		R	0.14	-0.64	--	--	-0.05	-0.29
		Slope	13	-205	--	--	-1.5	-13
		Intercept	5	46	--	--	31	99
		P < 0.05?	no	yes	--	--	no	yes
DRI	Summer	N	127	276	48	63	127	184
		R	-0.05	-0.44	0	0.27	-0.45	-0.59
		Slope	-8.4	-167	0.01	1.3	-27	-15
		Intercept	86	357	111	11.6	174	122
		P < 0.05?	no	yes	no	yes	yes	yes
DRI	Fall	N	114	244	--	--	115	153
		R	0.13	-0.57	--	--	0.04	-0.5
		Slope	9	-207	--	--	2	-15
		Intercept	29	424	--	--	38	100
		P < 0.05?	no	yes	--	--	no	yes

Table S-2 Cont.

			RM:GEM		$RM:O_3$		RM:WV	
Site	Season	Parameter	Obs	Model	Obs	Model	Obs	Model
NV02	Spring	N	51	77	51	77	51	77
		R	-0.5	-0.34	0.38	0.35	-0.67	-0.64
		Slope	-28	-110	1.2	1.1	-9.8	-13
		Intercept	75	256	-13	11	70	100
		P < 0.05?	yes	yes	yes	yes	yes	yes

Table S-2 cont.

			RM:GEM		$RM:O_3$		RM:WV	
Site	Season	Parameter	Obs	Model	Obs	Model	Obs	Model
SPL	Spring	N	26	33	26	33	26	33
		R	-0.59	-0.83	-0.14	0.28	-0.33	-0.27
		Slope	-60	-237	-0.44	2.5	-10	-32
		Intercept	124	541	48	-30	66	162
		P < 0.05?	yes	yes	no	no	no	no
SPL	Summer	N	20	31	20	31	20	31
		R	-0.56	-0.76	0.46	0.44	-0.33	-0.68
		Slope	-88	-362	1.4	4	-8	-43
		Intercept	175	737	-10	-121	74	201
		P < 0.05?	yes	yes	yes	yes	no	yes

Figure S-1: Maps of regions around study sites (green stars) in Taiwan and North America.

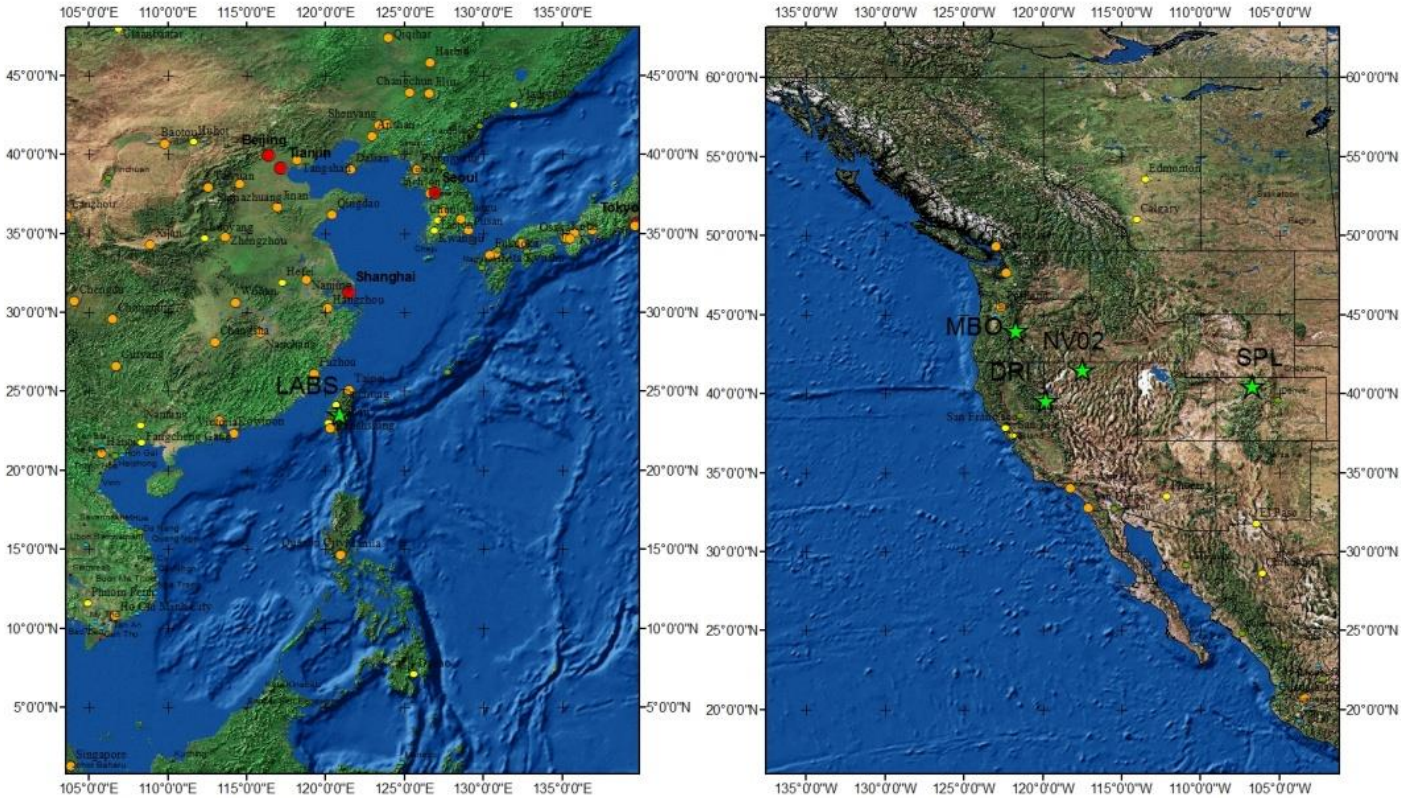
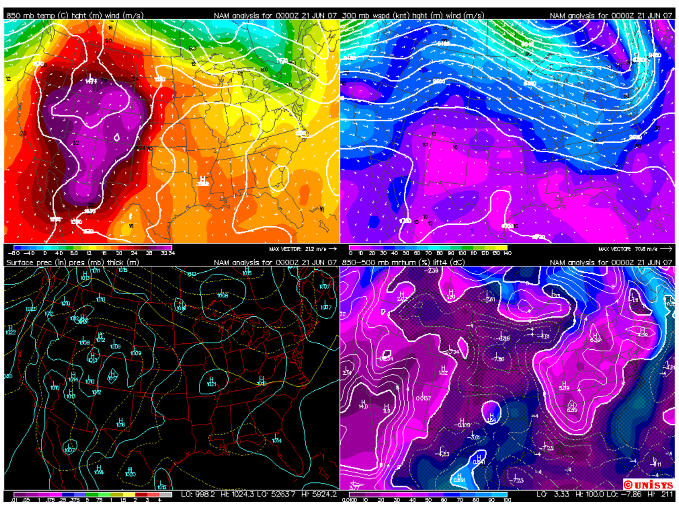


Figure S-2: Upper air forecast maps for June 21, 2007 (A) and June 24, 2007 (B) for the contiguous U.S., which revealed a large scale subsidence event that was consistent with the source of the RM having been conversion of GEM in the FT. On June 21 at 300 mbar a ridge was off shore of the Pacific Northwest and subsidence characterized by low RH between 1000 and 500 mbar extended from southern California northward through southern Washington, thus impacting MBO (Figure SI-2A). By June 24, the ridge at 300 mbar and the center of subsidence as indicated by RH fields had moved eastward to impact the sites in Nevada (Figure SI-2B). For each set of 4-panels in A and B, the two upper left panels show 850 mbar temperature, height, and wind vector fields, the two upper right panels show the 300 mbar wind fields, the two lower left panels show sea level pressure, and two lower right panels show 1000-500 mbar integrated relative humidity and lifted index fields. Images from http://weather.unisys.com.

A



B

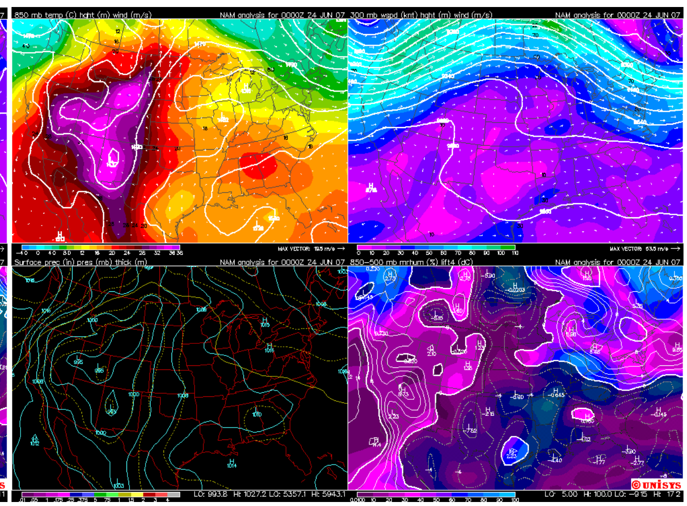
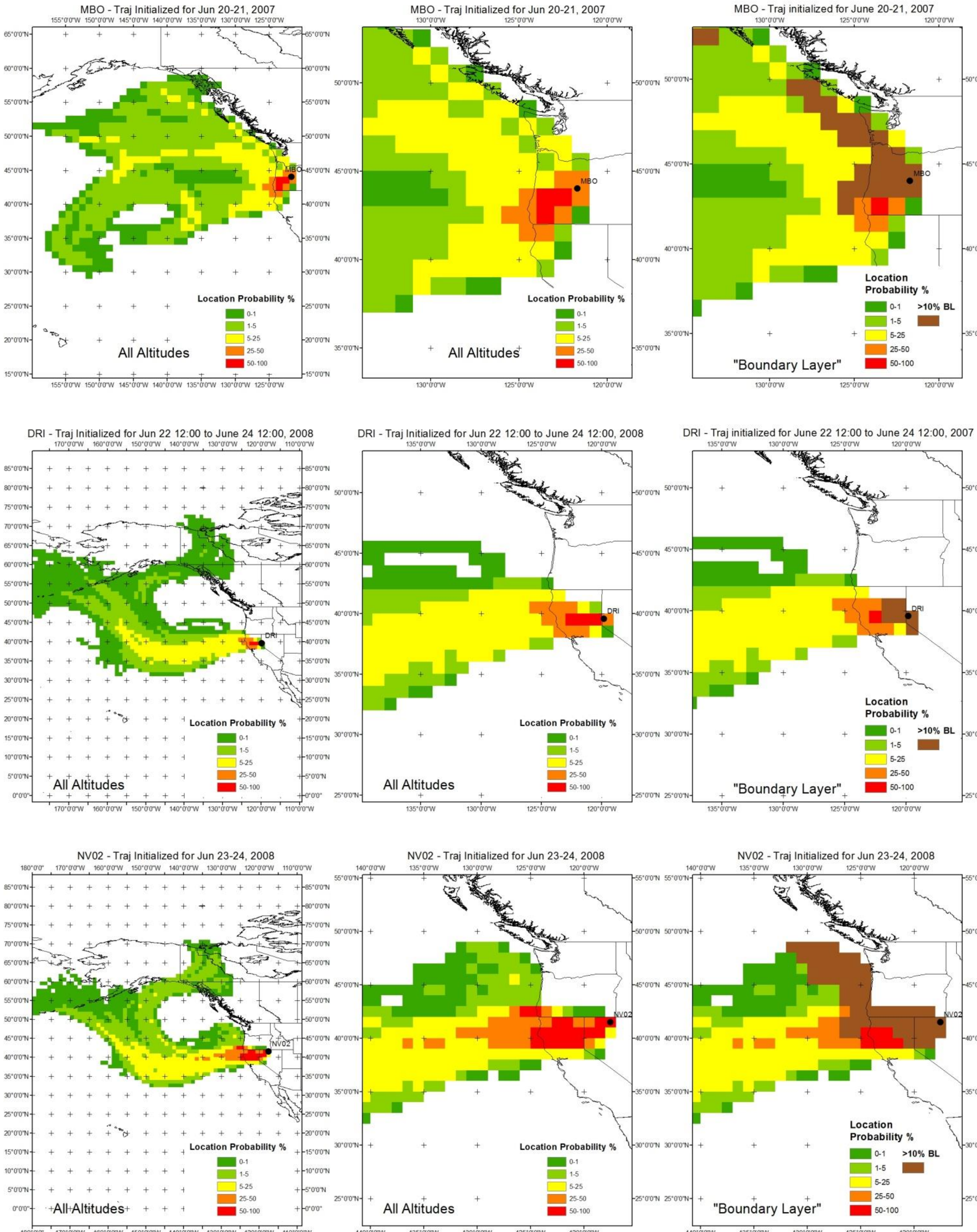


Figure S-3: Gridded frequency distributions (GFDs) of 120-hr HYSPLIT back trajectories with input meteorology from the National Centers for Environmental Prediction (NCEP) reanalysis data, for RGM events observed in sequence at MBO, DRI, and NV02 (times UTC). Gridded frequency distributions were generated by averaging the number of trajectory points in a 1^{o} x 1^{o} grid cell over the domain of interest. Location probability represents the fraction of trajectory points in a given cell relative to the number of trajectory points in the most populated cell (Gustin et al., 2012). For each trajectory point, the modeled altitude was compared against that of the planetary boundary layer. The colors show relative horizontal position probabilities of the trajectory points initialized by the times given above each plot and 9 starting locations in a $0.5x0.5^{o}$ grid with altitudes of 500, 1000, 1500, and 2000 m agl. Left panels show entire distribution of trajectory points over all altitudes 5-days backward in time, center panels shows close-up views of the event, and right panels depict those grid cells in brown that have > 10% of trajectory points below the modeled boundary layer (termed "Boundary Layer"). At each site these plots show general transport from west to east. The GFDs suggest that two air streams merged upwind of each site: a branch in the BL from the north, and a branch in the FT from more southerly directions. At each site the trajectories are consistent with the forecast maps in showing recently (< 1 day) subsided air, which is indicated by one or more grid cells in the GFDs with the maximum location probability (colored red) still remaining after the grid cells were identified with > 10% of points below the modeled boundary layer (colored brown).

Figure S-3



MBO - Traj Initialized for Jun 20-21, 2007
MBO
Location Probability %
0-1
1-5
5-25
25-50
50-100
All Altitudes
MBO - Traj Initialized for Jun 20-21, 2007
MBO
Location Probability %
0-1
1-5
5-25
25-50
50-100
All Altitudes
MBO - Traj initialized for June 20-21, 2007
MBO
Location Probability %
0-1
1-5
5-25
25-50
50-100
>10% BL
"Boundary Layer"
DRI - Traj Initialized for Jun 22 12:00 to June 24 12:00, 2008
DRI
Location Probability %
0-1
1-5
5-25
25-50
50-100
All Altitudes
DRI - Traj Initialized for Jun 22 12:00 to June 24 12:00, 2008
DRI
Location Probability %
0-1
1-5
5-25
25-50
50-100
All Altitudes
DRI - Traj initialized for June 22 12:00 to June 24 12:00, 2007
DRI
Location Probability %
0-1
1-5
5-25
25-50
50-100
>10% BL
"Boundary Layer"
NV02 - Traj Initialized for Jun 23-24, 2008
NV02
Location Probability %
0-1
1-5
5-25
25-50
50-100
All Altitudes
NV02 - Traj Initialized for Jun 23-24, 2008
NV02
Location Probability %
0-1
1-5
5-25
25-50
50-100
All Altitudes
NV02 - Traj Initialized for Jun 23-24, 2008
NV02
Location Probability %
0-1
1-5
5-25
25-50
50-100
>10% BL
"Boundary Layer"