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Atmospheric Chemistry and Physics Discussions

Supplement of

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

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Table S1: 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.

All Obs

		GEM (ng m^{-3})		RM (pg m^{-3})		O_3 (ppbv)		WV (g kg^{-1})	
Location	Season	mean	σ	mean	σ	mean	σ	mean	σ
LABS	Winter	1.73	0.27	24	9	36.2	7.0	5.9	2.8
LABS	Spring	2.20	0.26	27	18	52.9	5.3	8.5	2.7
LABS	Summer	1.43	0.13	15	20	22.2	5.0	14.2	1.6
LABS	Fall	1.70	0.15	11	5	29.2	6.8	13.2	3.5
MBO	Spring	1.60	0.17	32	46	48.7	8.1	3.6	1.1
MBO	Summer	1.44	0.10	44	45	42.4	8.3	5.0	1.5
DRI	Winter	1.73	0.57	14	16	--	--	3.1	1.0
DRI	Spring	1.60	0.26	25	22	--	--	3.5	1.0
DRI	Summer	1.36	0.35	68	42	49.2	9.6	4.4	1.3
DRI	Fall	1.66	0.38	39	26	--	--	3.6	1.0
NV02	Summer	1.79	1.12	32	16	40.1	6.2	4.5	2.0
SPL	Spring	1.59	0.19	27	22	44.9	7.4	3.9	0.8
SPL	Summer	1.59	0.21	34	22	36.7	8.0	4.6	1.1
All-Sites	All Seasons	1.65	0.32	30	24	40.3	7.2	6.0	1.6

WV Filtered Obs

Location	Season	mean	σ	mean	σ	mean	σ	mean	σ
LABS	Winter	1.71	0.29	25	9	37.7	6.0	5.0	2.4
LABS	Spring	2.21	0.28	31	18	52.4	5.1	7.6	2.2
LABS	Summer	1.42	0.12	15	20	22.0	5.1	14.1	1.5
LABS	Fall	1.70	0.15	11	5	29.2	6.8	13.2	3.5
MBO	Spring	1.62	0.16	33	47	50.7	7.2	3.1	0.8
MBO	Summer	1.40	0.11	64	49	45.8	7.6	3.7	0.7
DRI	Winter	1.79	0.55	17	17	--	--	2.7	0.6
DRI	Spring	1.60	0.25	27	24	--	--	3.1	0.6
DRI	Summer	1.28	0.29	76	45	50.6	9.1	3.7	0.8
DRI	Fall	1.66	0.37	44	27	--	--	3.2	0.6
NV02	Summer	1.55	0.71	34	17	40.6	5.7	3.7	1.2
SPL	Spring	1.59	0.21	28	21	46.2	6.7	3.7	0.7
SPL	Summer	1.57	0.23	42	20	38.2	6.8	4.1	0.8
All-Sites	All Seasons	1.62	0.29	34	25	41.3	6.6	5.5	1.3

Std Geos-Chem Model

		GEM (ng m^{-3})		RM (pg m^{-3})		O_3 (ppbv)		WV (g kg^{-1})	
Location	Season	mean	σ	mean	σ	mean	σ	mean	σ
LABS	Winter	1.77	0.18	142	59	52.6	7.1	3.7	2.6
LABS	Spring	2.10	0.27	87	31	52.9	5.2	6.5	2.7
LABS	Summer	1.61	0.06	21	17	32.2	10.0	9.9	2.0
LABS	Fall	1.89	0.15	36	24	46.8	6.7	10.3	1.2
MBO	Spring	1.84	0.14	111	55	51.2	8.9	1.6	0.9
MBO	Summer	1.67	0.09	135	58	50.9	11.6	2.3	1.2
DRI	Winter	1.87	0.13	57	35	--	--	1.6	0.8
DRI	Spring	1.91	0.11	69	36	--	--	2.1	0.8
DRI	Summer	1.70	0.08	72	29	52.3	7.4	3.0	1.1
DRI	Fall	1.74	0.08	63	28	--	--	2.3	0.9
NV02	Summer	1.76	0.07	62	21	48.1	7.2	3.0	1.1
SPL	Spring	1.79	0.14	116	41	57.9	4.7	1.4	0.3
SPL	Summer	1.71	0.09	119	43	60.4	4.8	1.9	0.7
All-Sites	All Seasons	1.80	0.12	84	37	50.5	7.4	3.8	1.3

Table S2: 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.

			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 S2 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

12 Table S2 cont.

			RM:GEM		RM:O_3		RM:WV	
Site	Season	Parameter	Obs	Model	Obs	Model	Obs	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

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Table S2 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 S2 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 S1: Maps of regions around study sites (green stars) in Taiwan and North America.

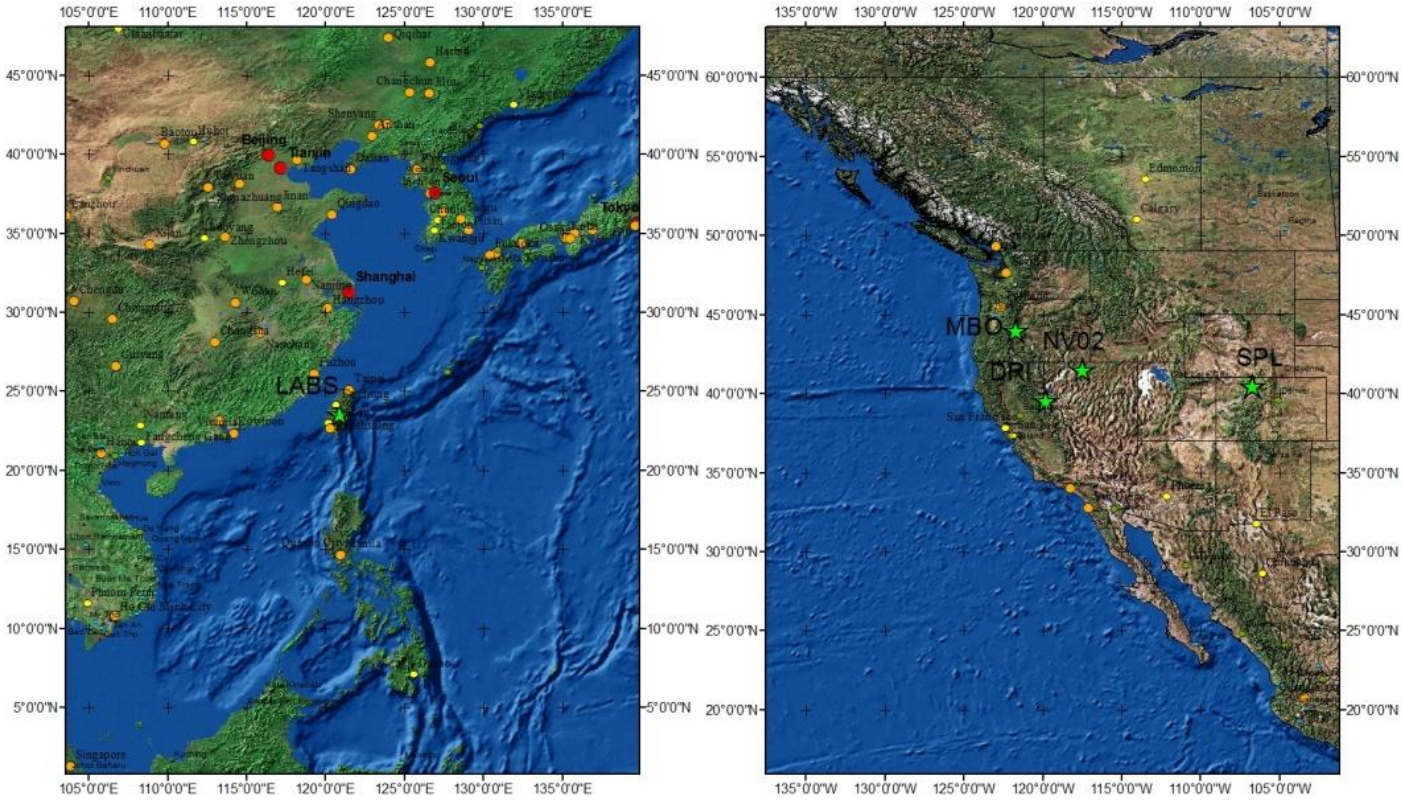


Figure S2: Upper air forecast maps for June 21, 2007 (A) and June 24, 2007 (B) for the contiguous U.S., which revealed a large scale subsidence event that was consistent with the source of the RM having been conversion of GEM in the FT. On June 21 at 300 mbar a ridge was off shore of the Pacific Northwest 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

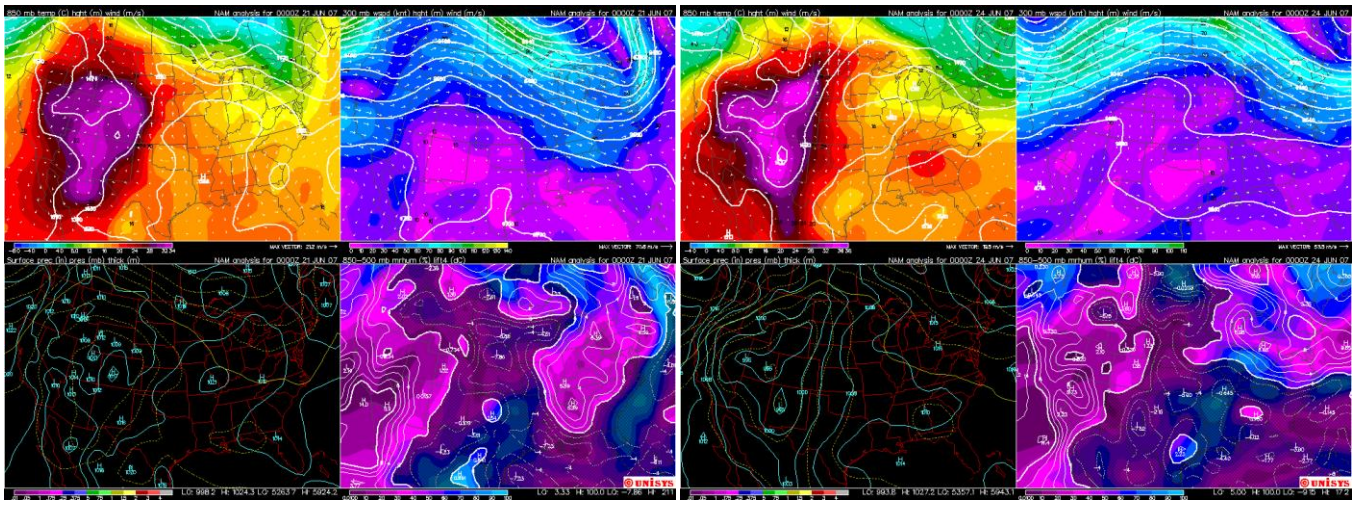
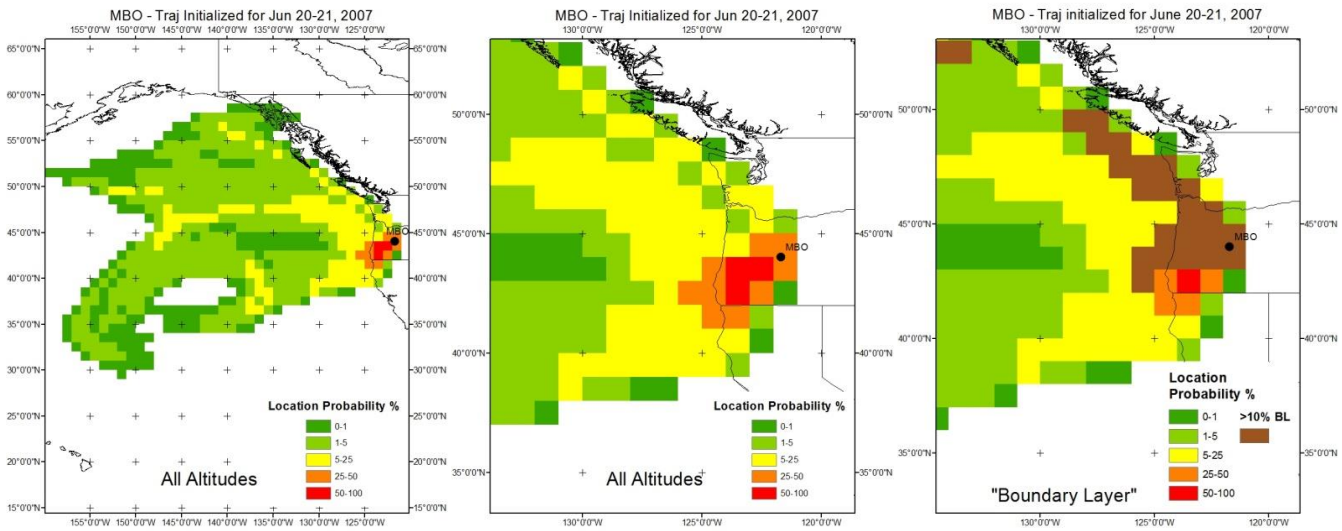
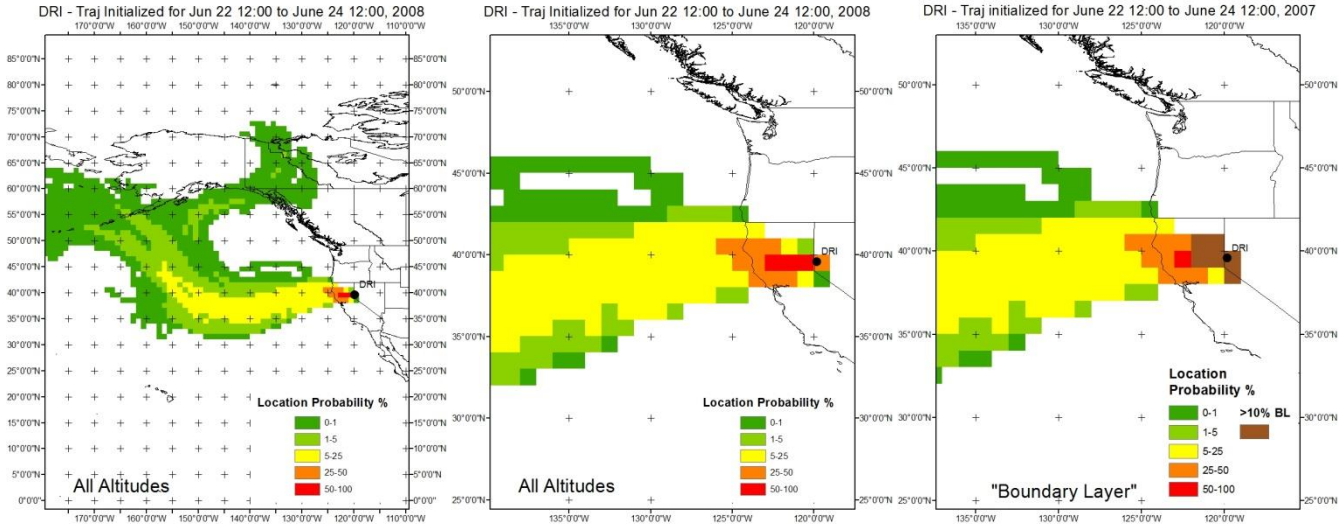
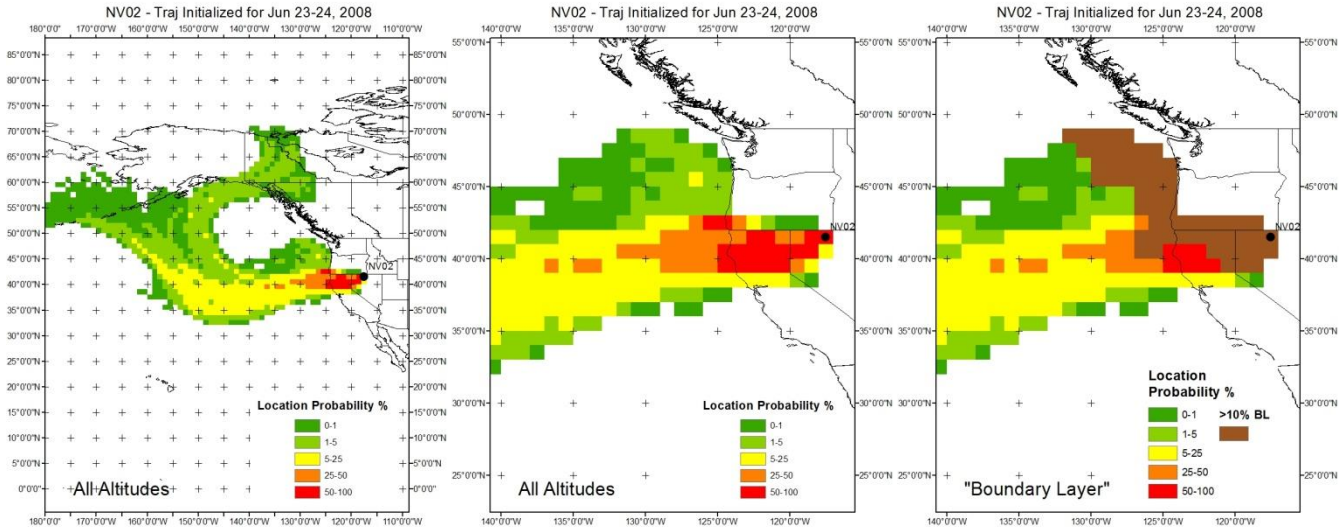


Figure S3: Gridded frequency distributions (GFDs) of 120-hr HYSPLIT back trajectories with input meteorology from the National Centers for Environmental Prediction (NCEP) reanalysis data, for RGM events observed in sequence at MBO, DRI, and NV02 (times UTC). Gridded frequency distributions were generated 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 S3


MBO - Traj Initialized for Jun 20-21, 2007
MBO - Traj Initialized for Jun 20-21, 2007
MBO - Traj initialized for June 20-21, 2007
MBO
Location Probability %
0-1
1-5
5-25
25-50
50-100
>10% BL
All Altitudes
"Boundary Layer"


DRI - Traj Initialized for Jun 22 12:00 to June 24 12:00, 2008
DRI - Traj Initialized for Jun 22 12:00 to June 24 12:00, 2008
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
All Altitudes
"Boundary Layer"


NV02 - Traj Initialized for Jun 23-24, 2008
NV02 - Traj Initialized for Jun 23-24, 2008
NV02 - Traj Initialized for Jun 23-24, 2008
NV02
Location Probability %
0-1
1-5
5-25
25-50
50-100
>10% BL
All Altitudes
"Boundary Layer"