

Supporting Information to:

**Investigation of Mercury Emission Sources Using Hg Isotopic Compositions of
Atmospheric Mercury at the Cape Hedo Atmosphere and Aerosol Monitoring Station
(CHAAMS), Japan**

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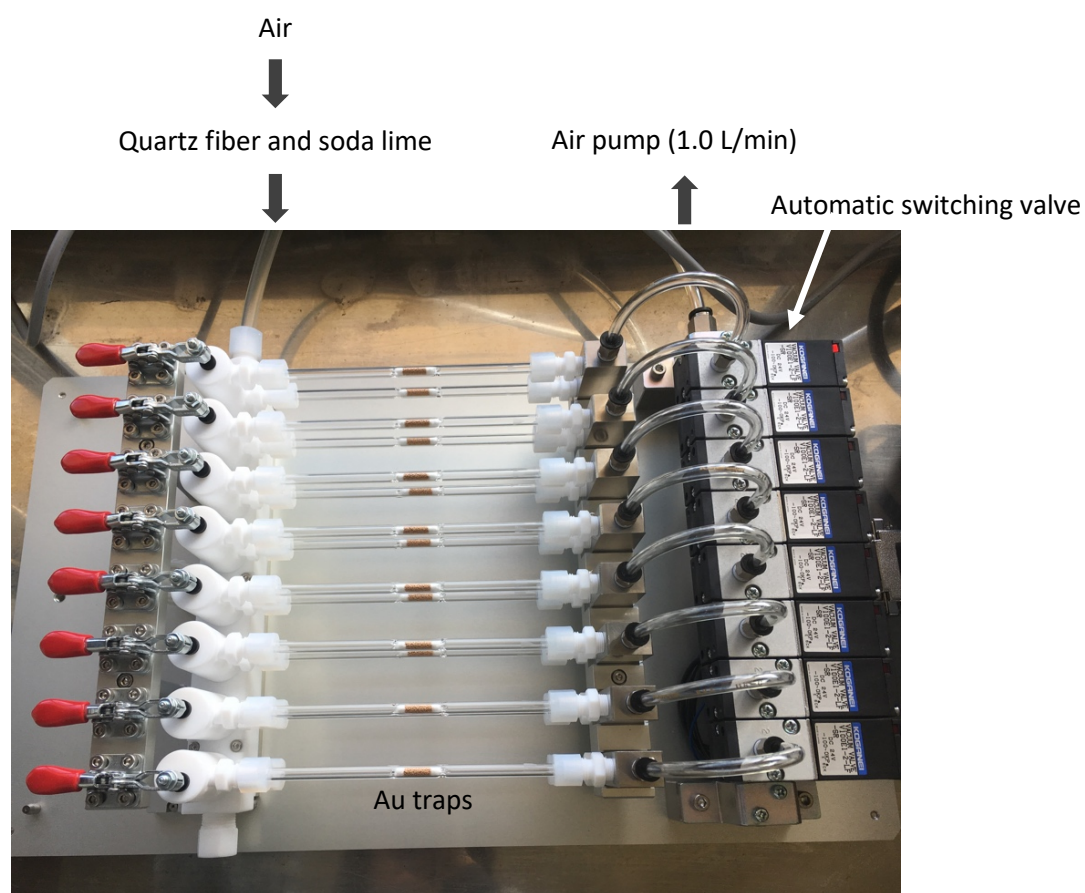


Figure S1: A GEM sampling system. For avoiding breakthrough of GEM across the trap, the air flow rate was adjusted to 1.0 L/min per trap. In order to measure Hg isotopes with the required accuracy, >10 ng of GEM was necessary. Two Au traps were set parallel, and eight rows were prepared for the long term monitoring. The automatic switching valve was set to switch every 72 hours.

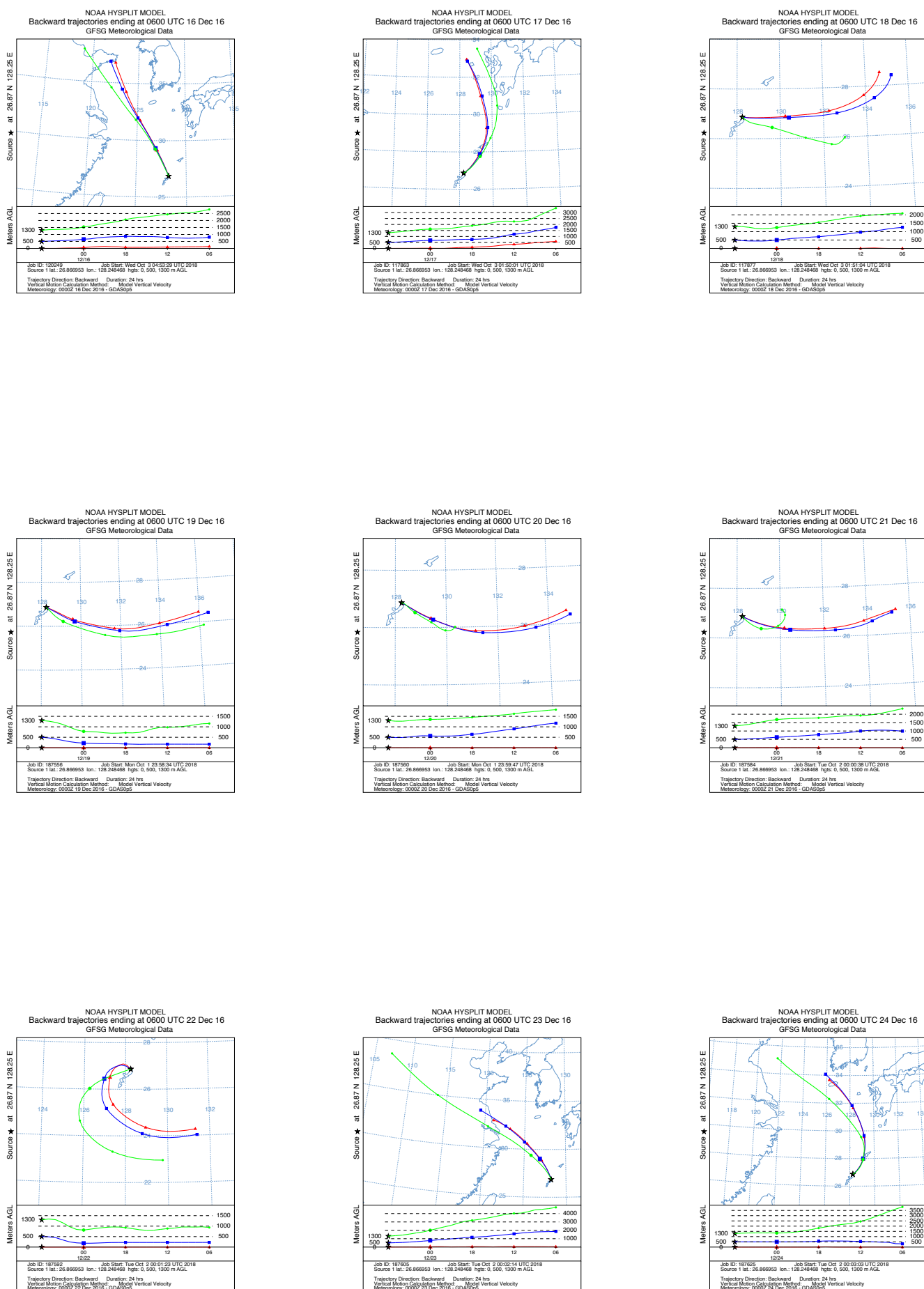


Figure SI 2. Air-mass back trajectories of 24-h duration

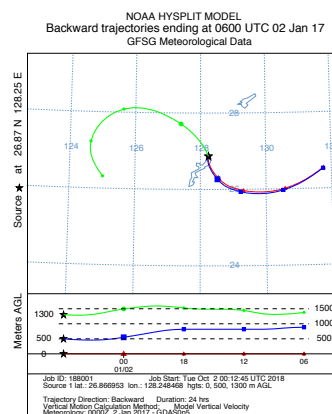
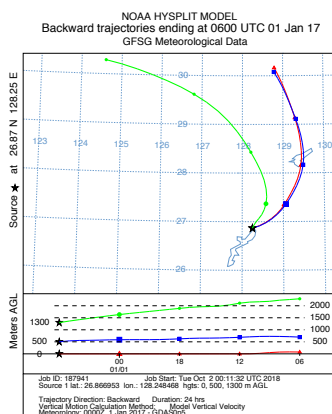
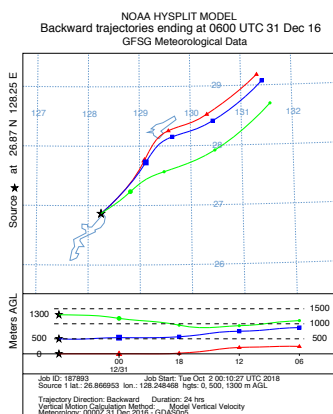
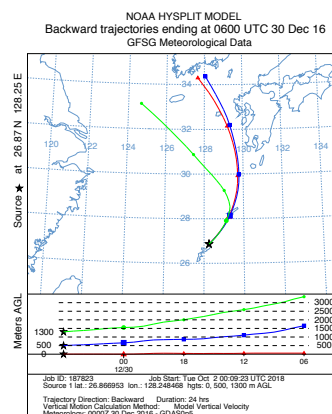
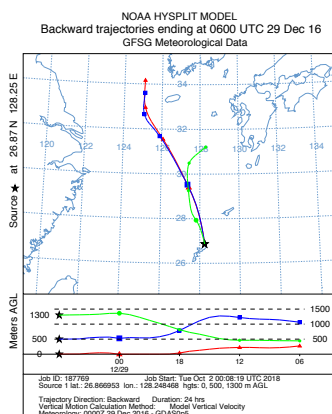
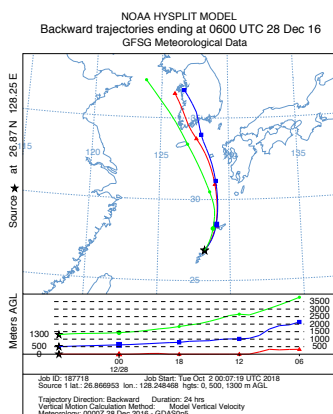
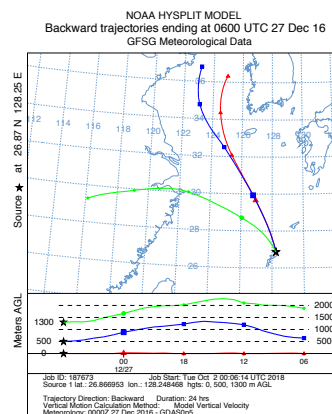
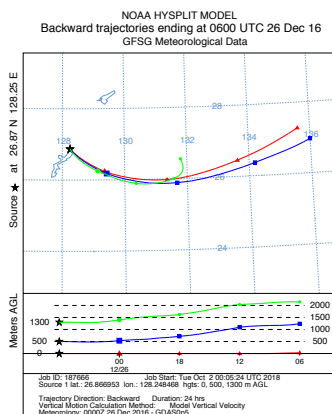
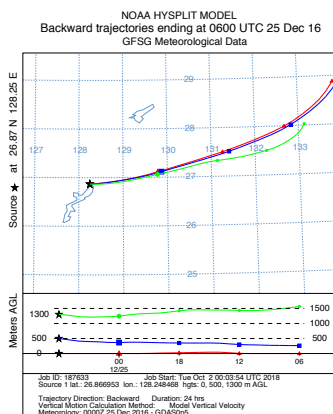


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

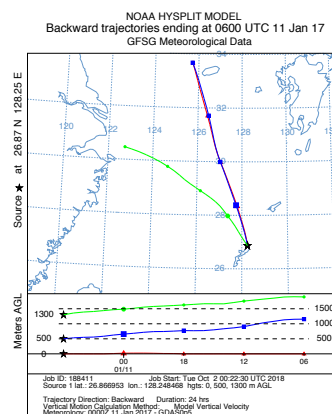
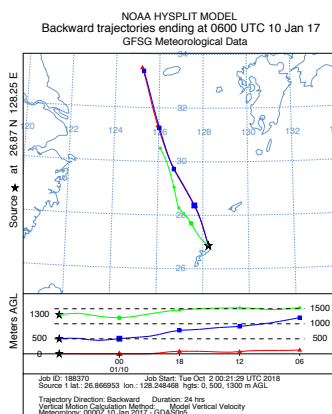
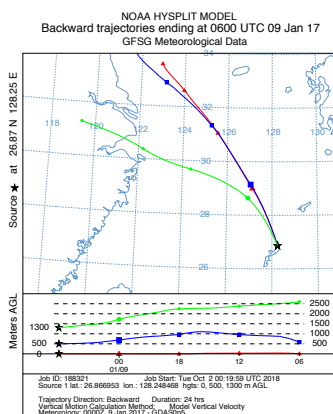
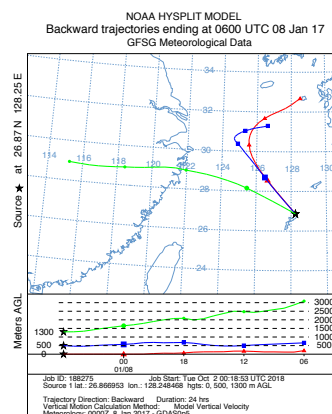
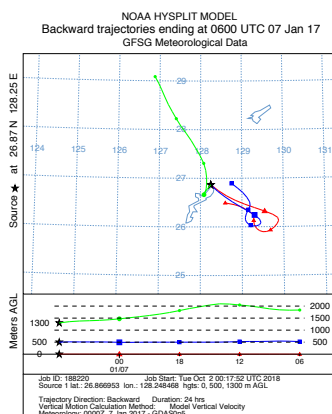
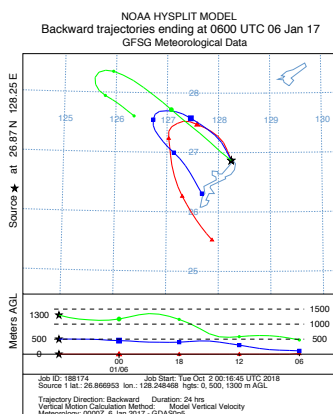
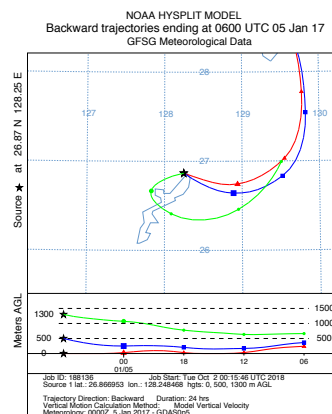
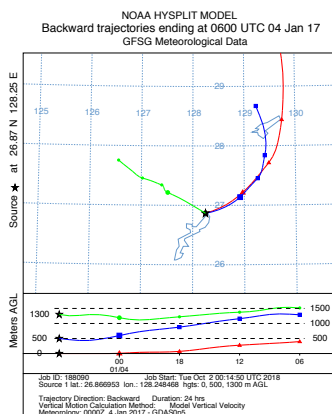
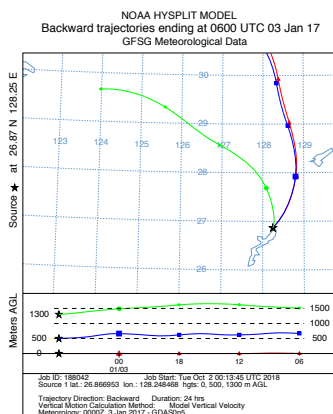


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

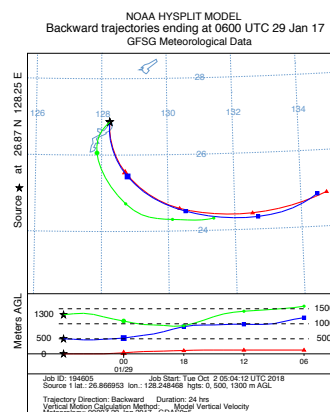
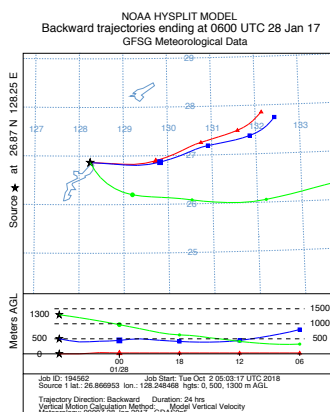
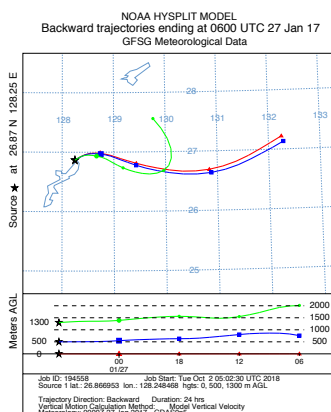
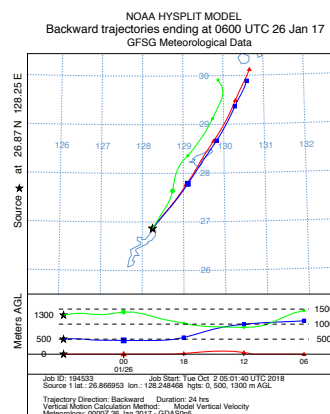
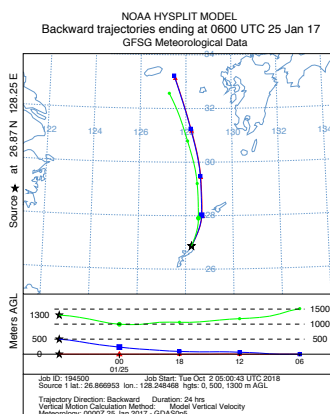
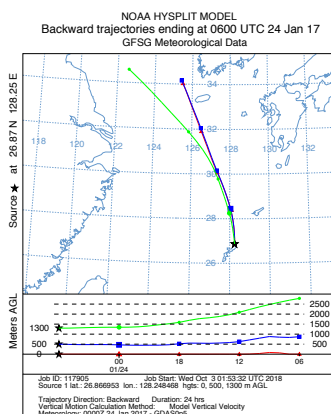
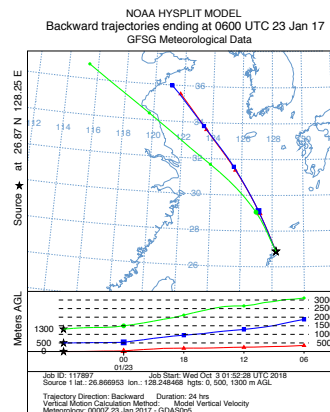
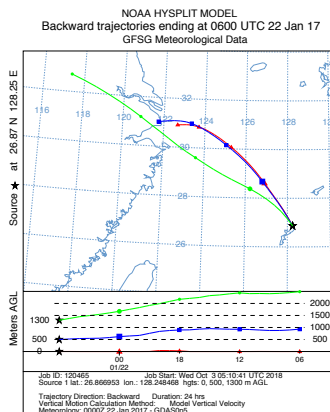
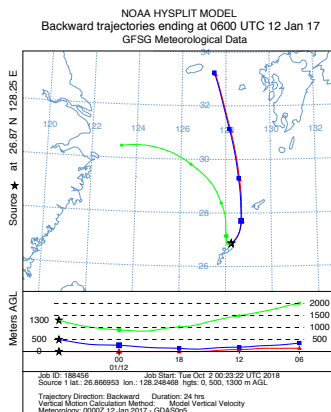


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

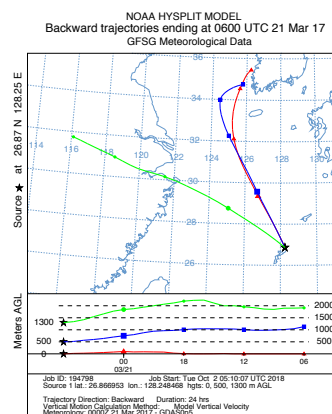
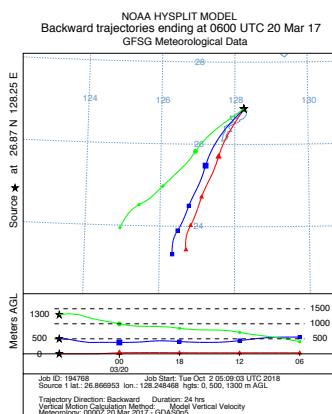
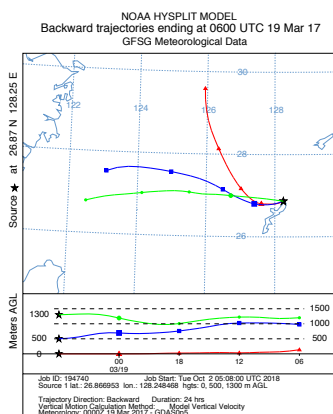
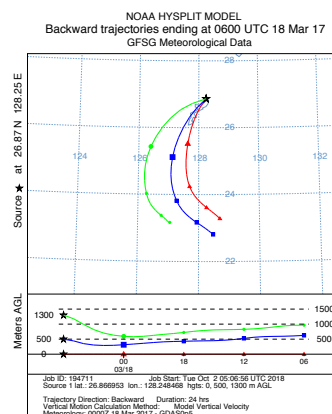
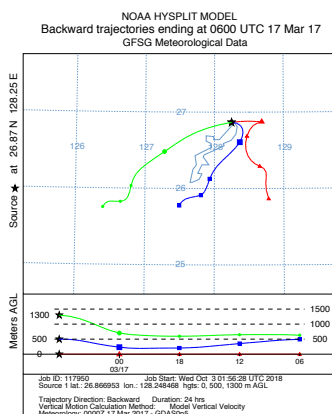
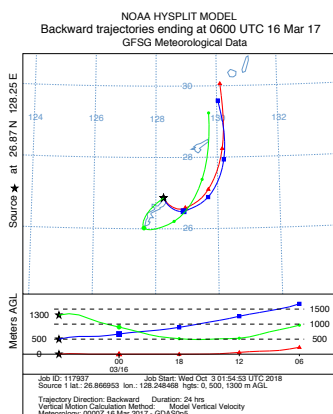
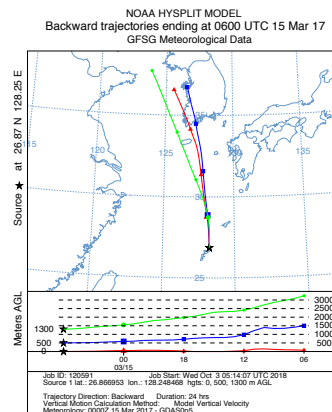
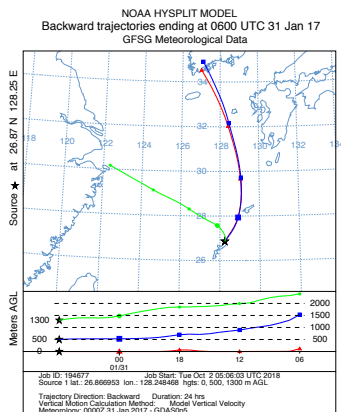
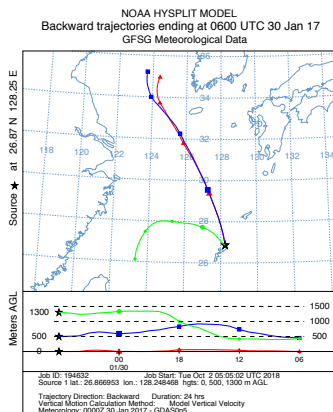


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

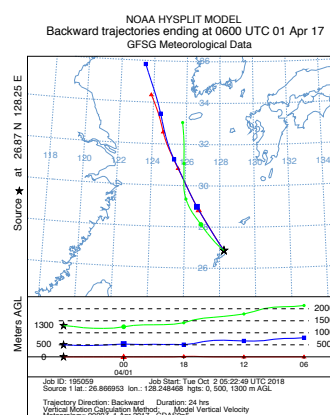
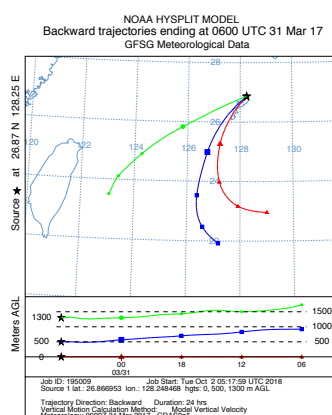
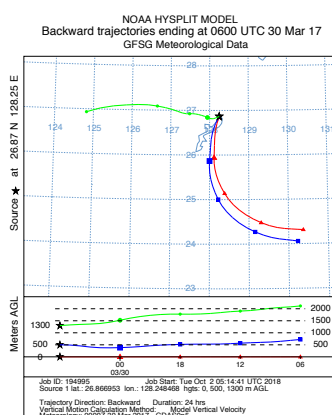
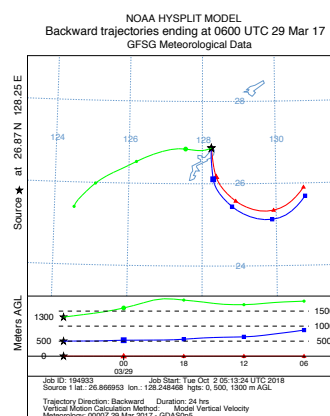
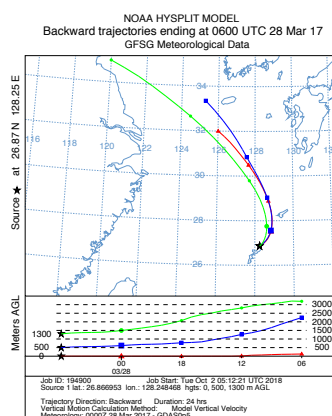
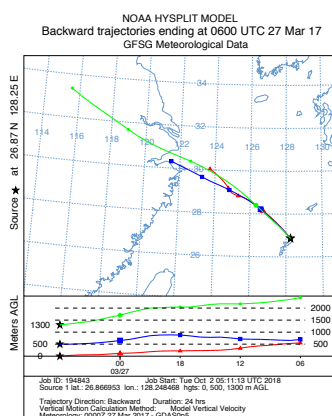
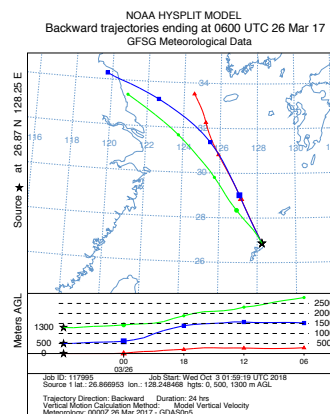
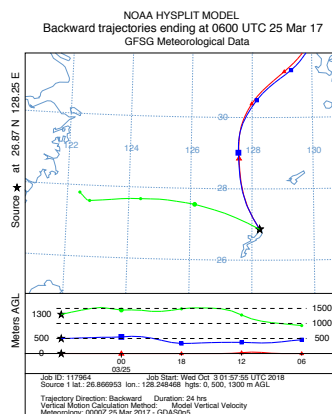
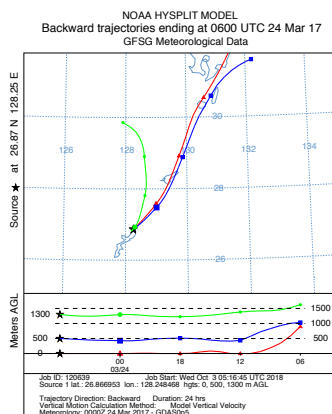


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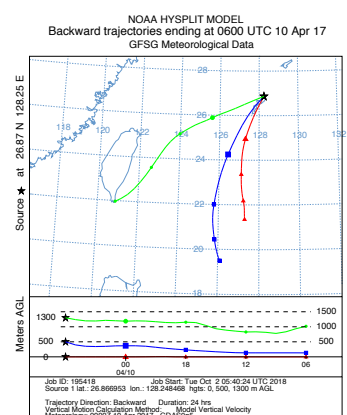
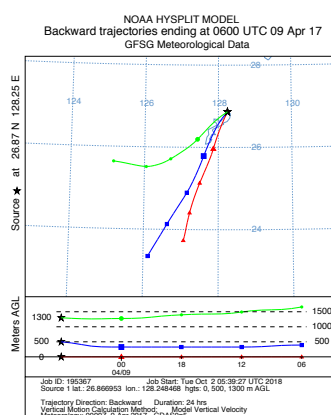
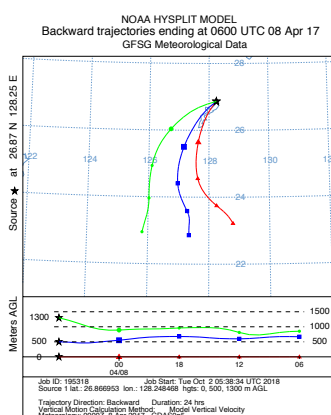
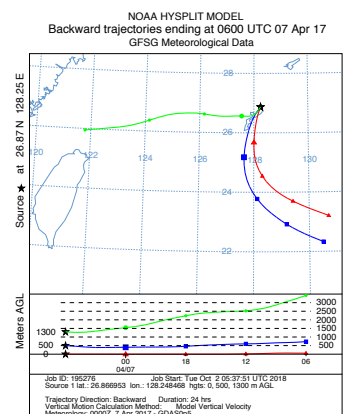
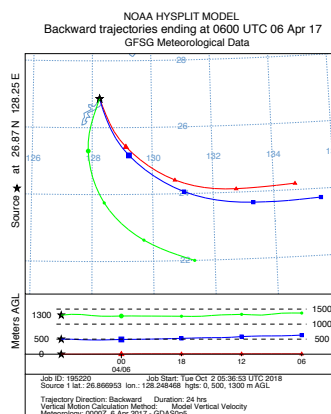
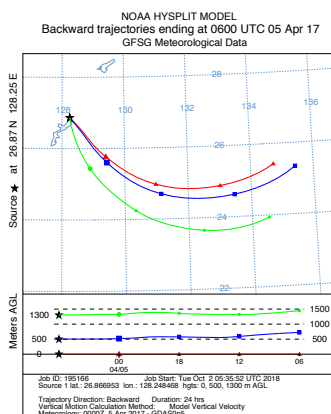
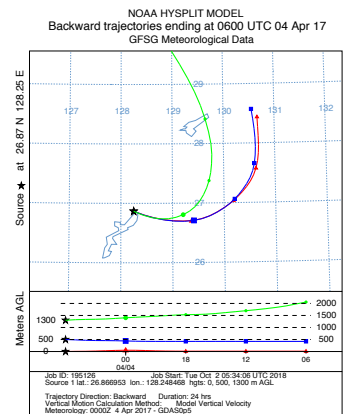
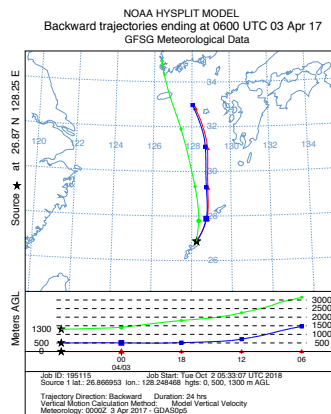
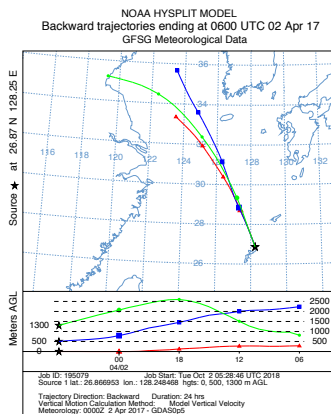


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

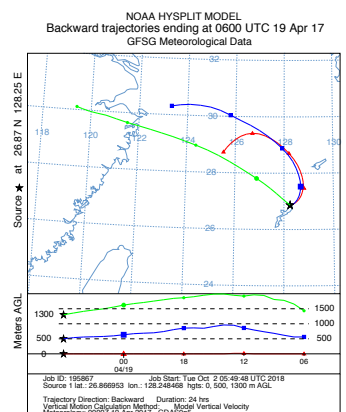
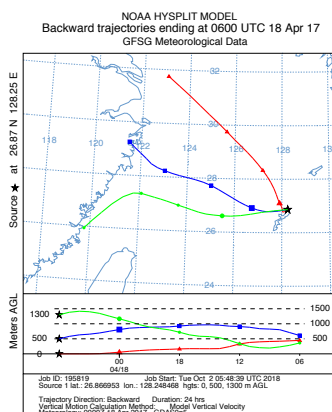
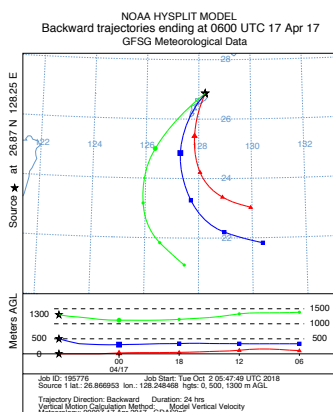
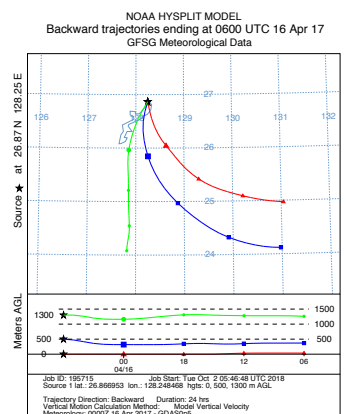
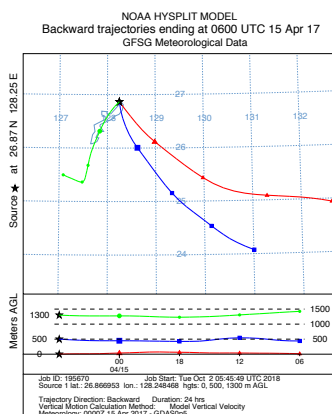
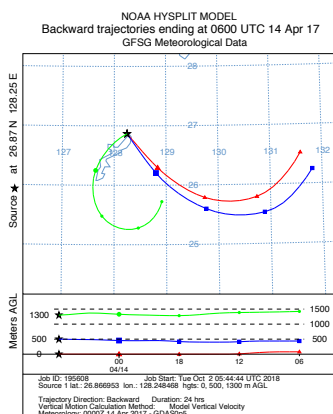
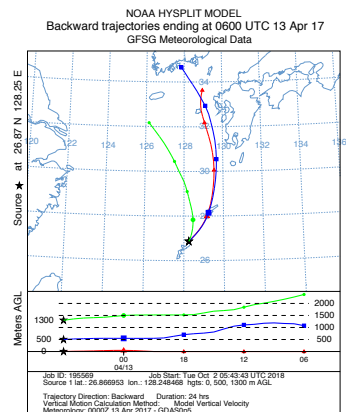
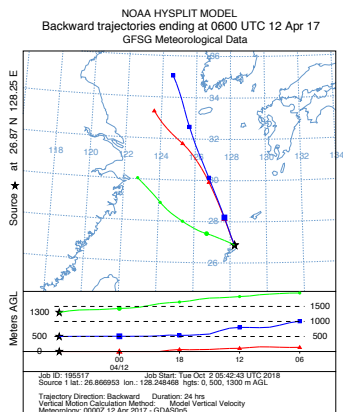
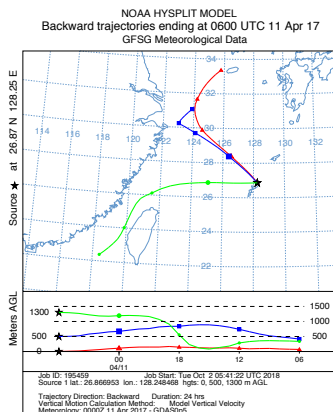


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

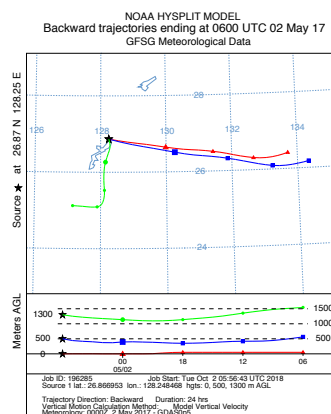
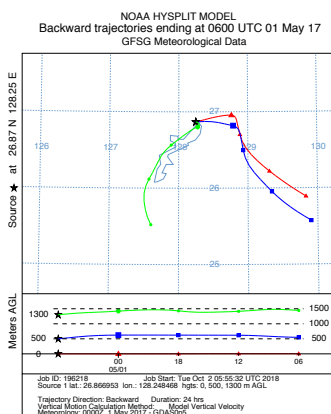
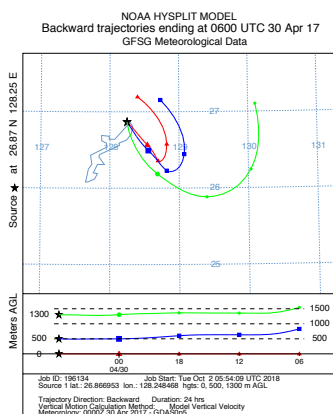
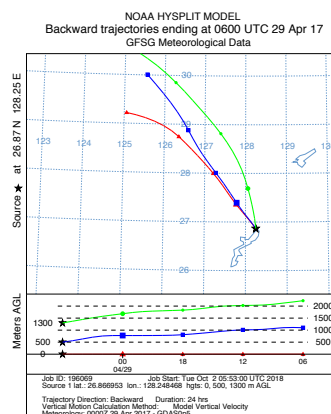
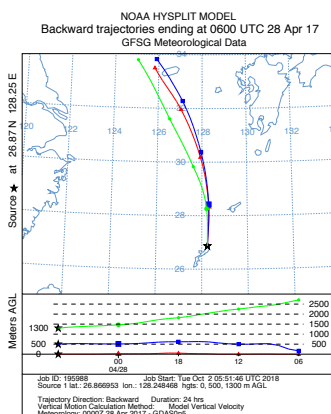
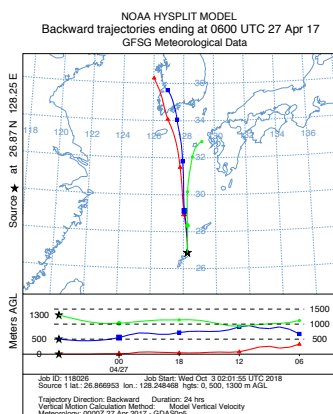
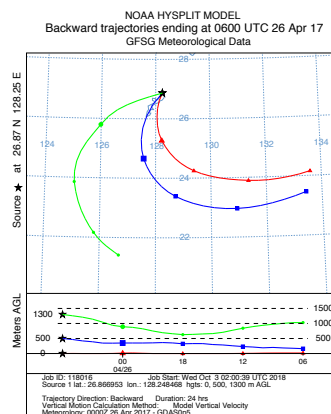
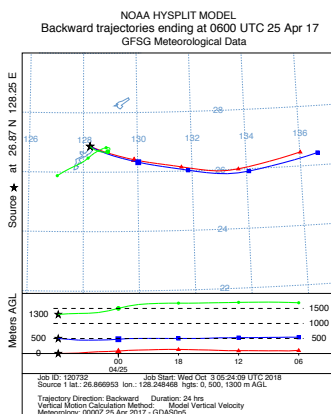
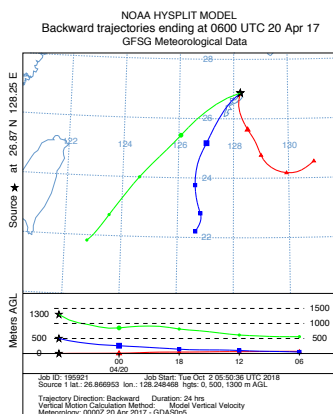


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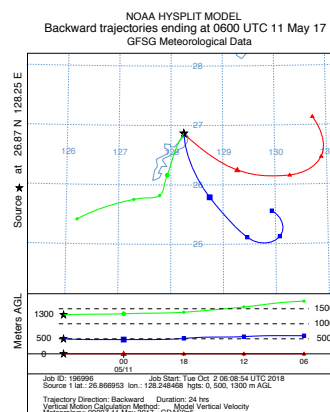
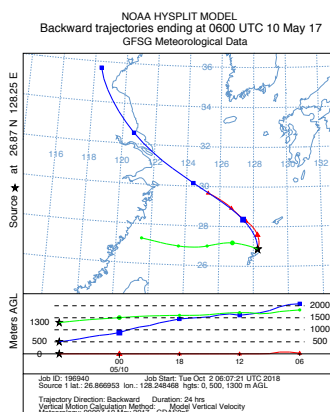
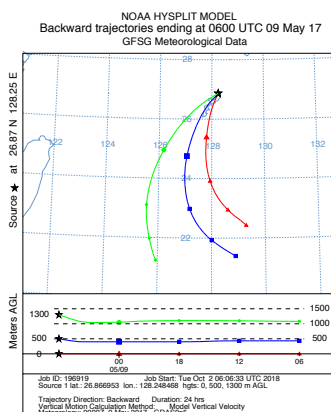
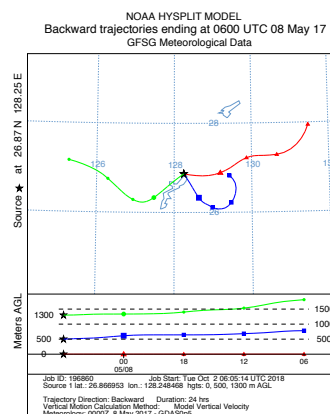
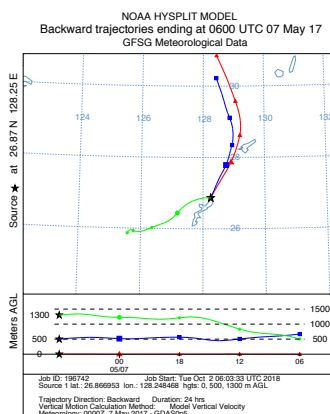
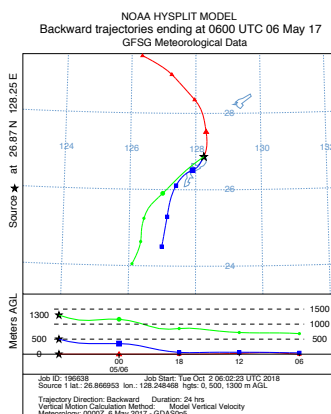
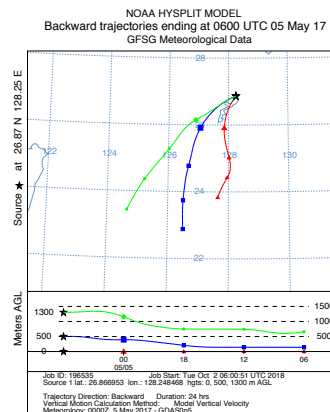
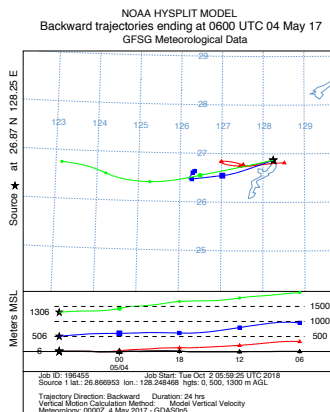
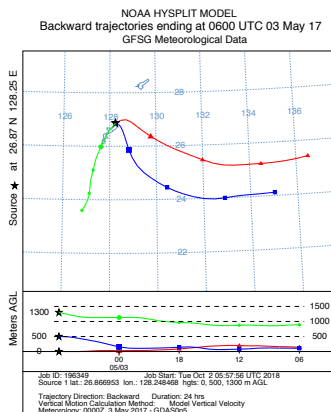


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

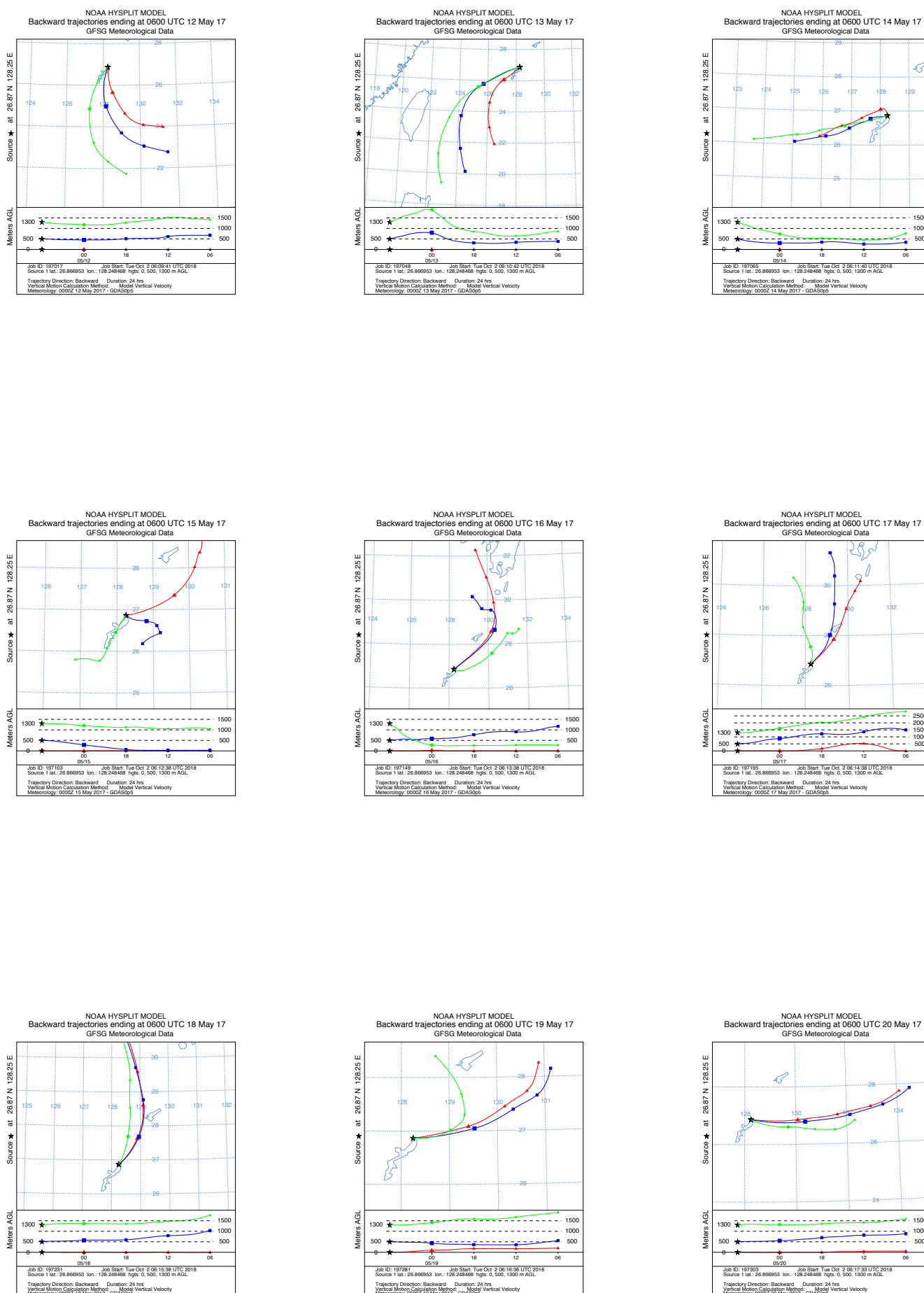


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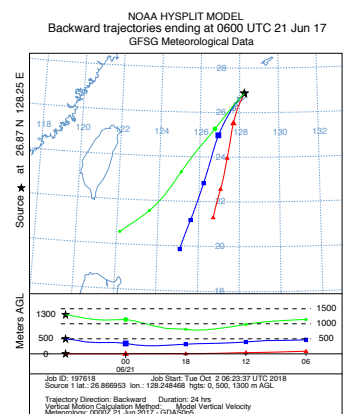
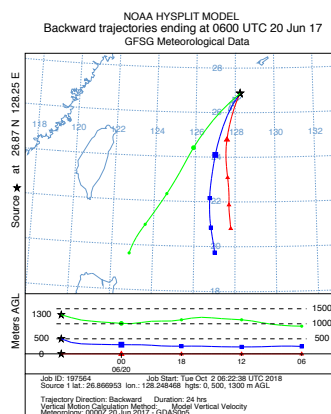
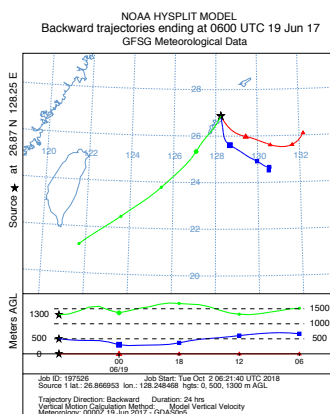
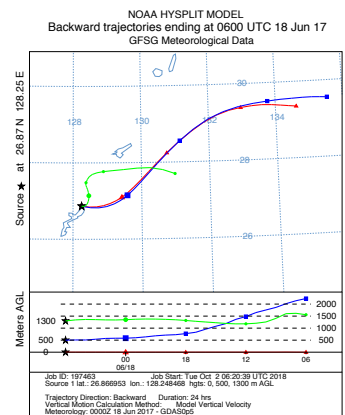
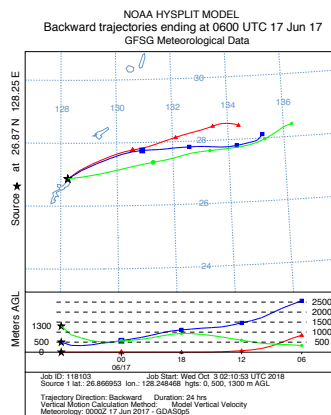
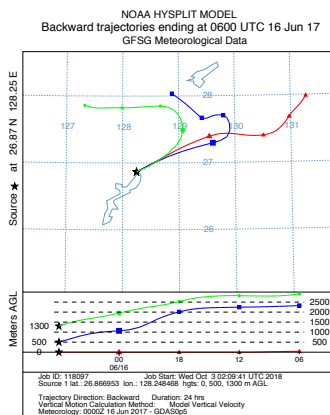
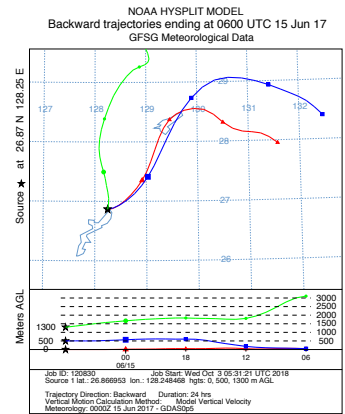
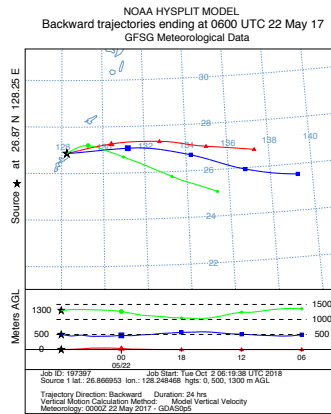
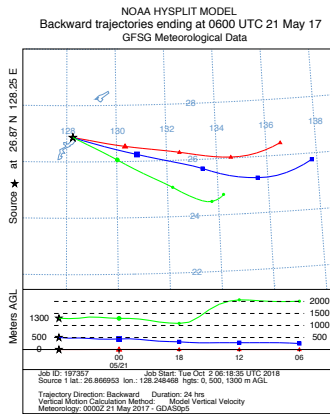


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

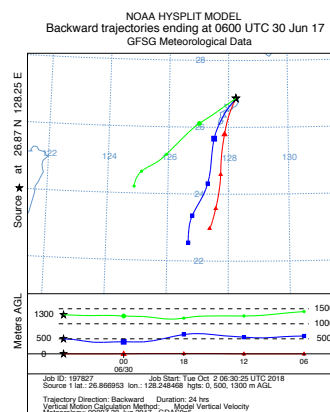
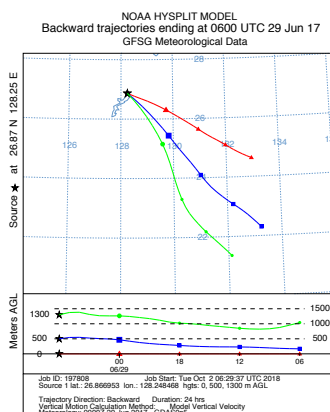
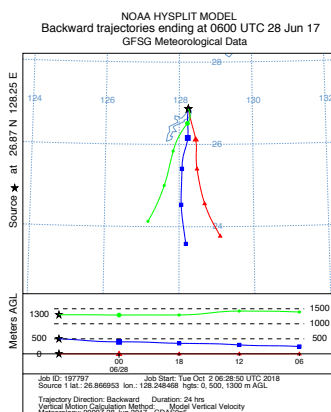
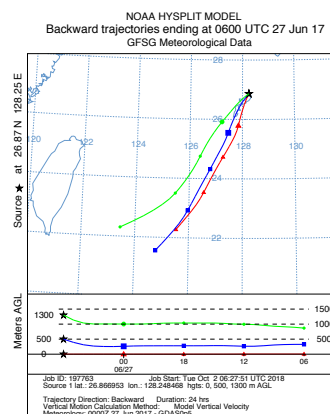
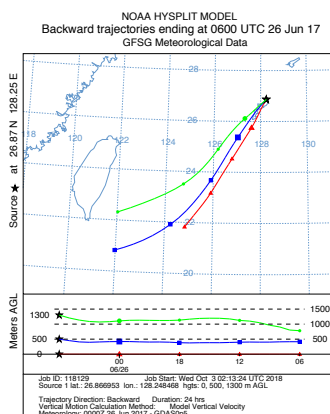
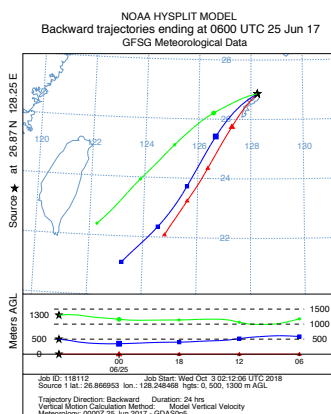
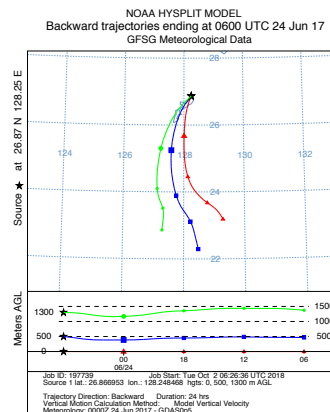
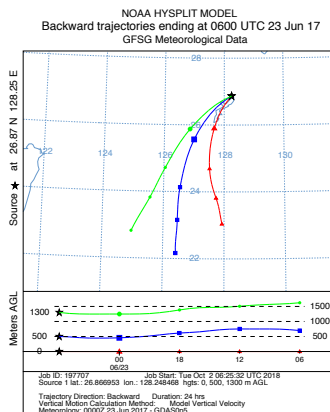
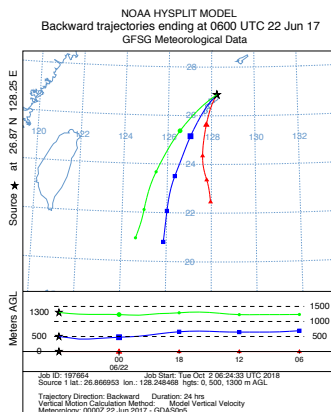


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

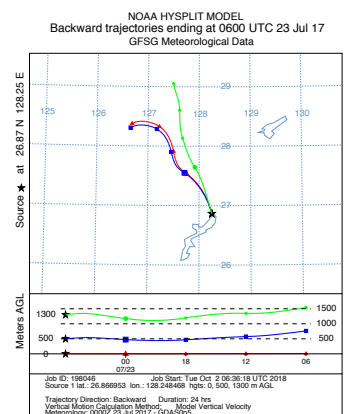
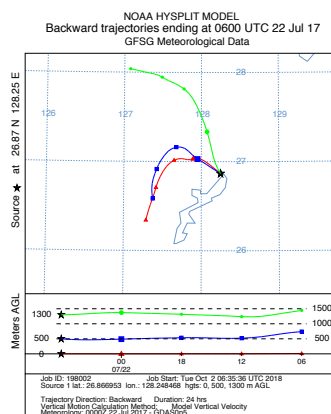
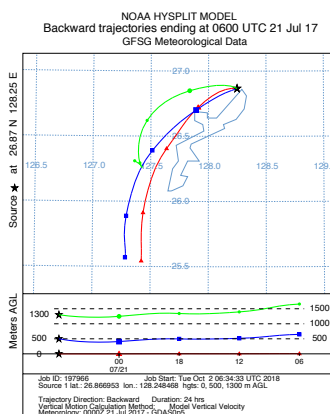
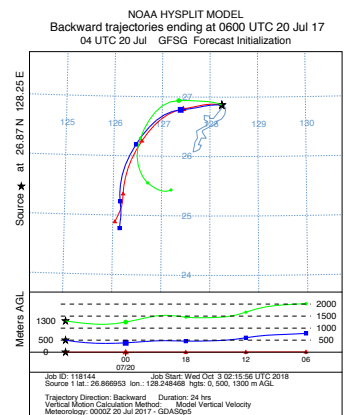
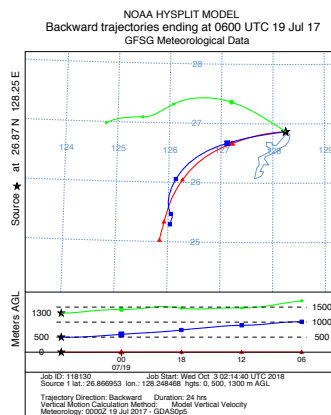
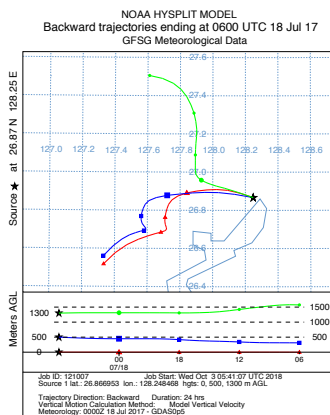
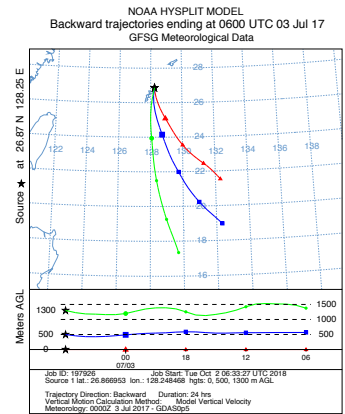
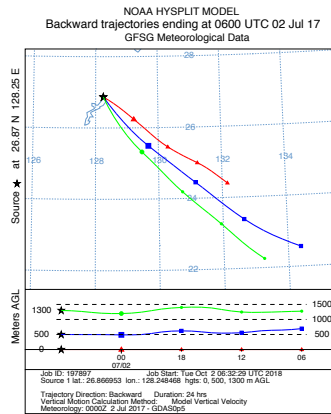
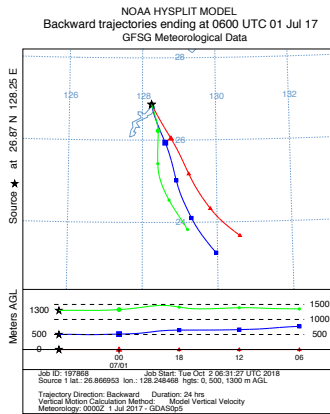


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

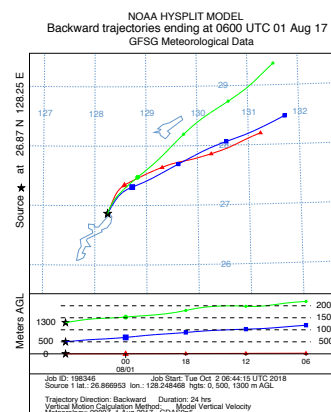
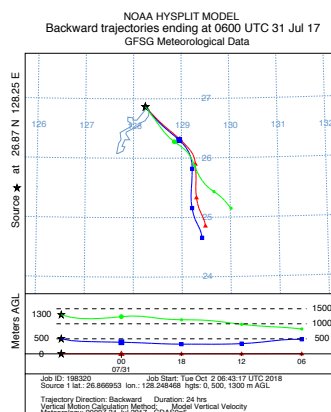
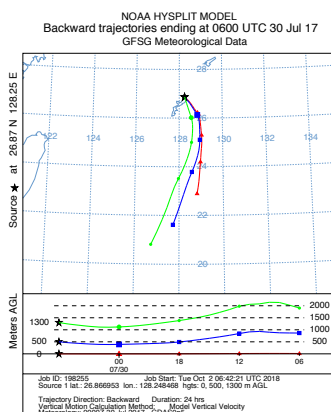
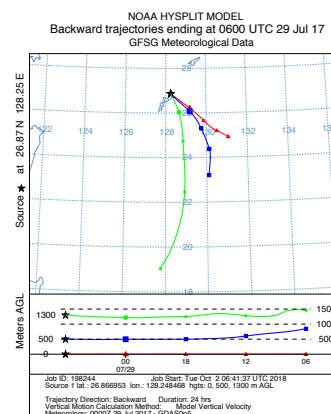
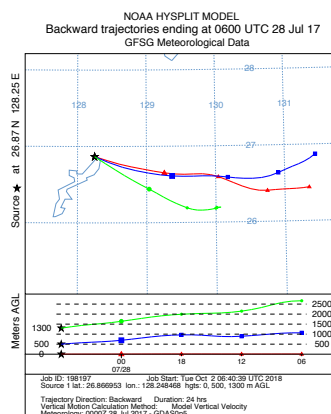
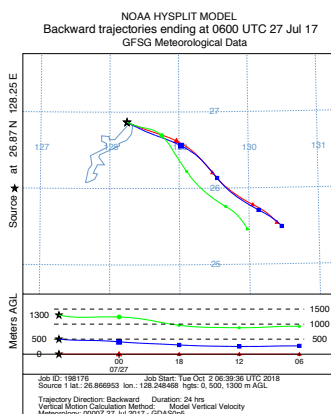
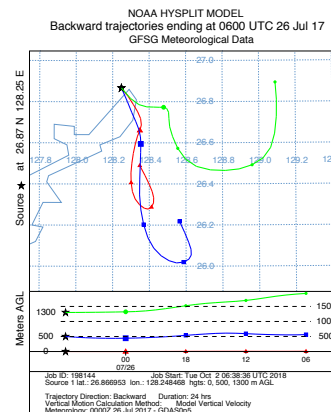
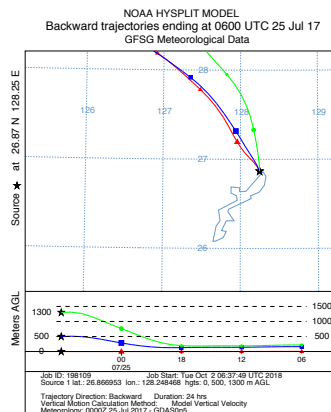
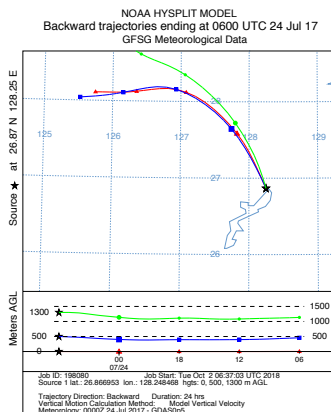


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

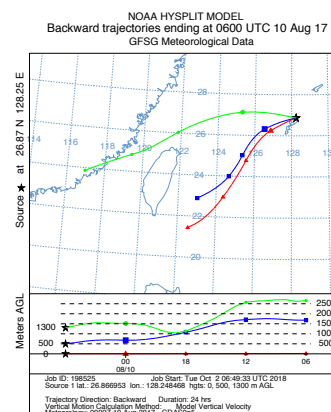
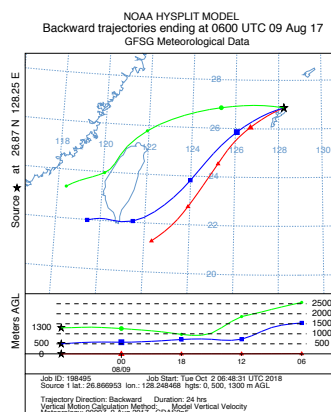
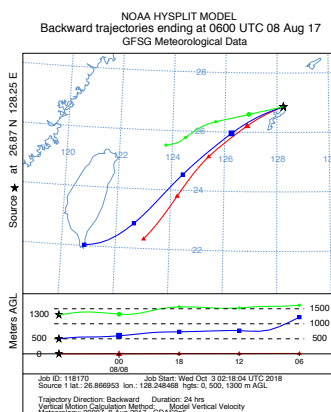
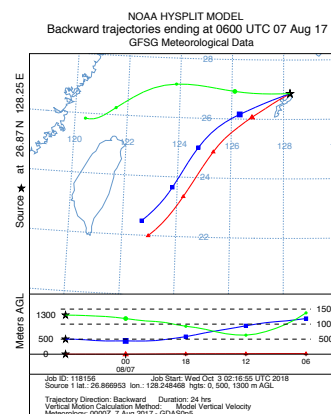
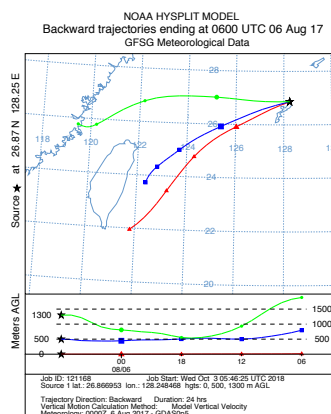
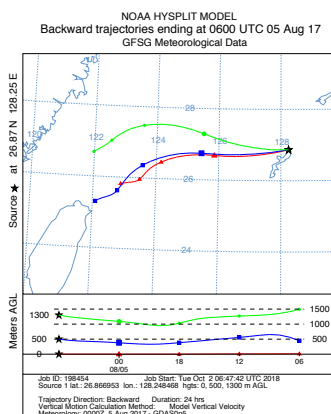
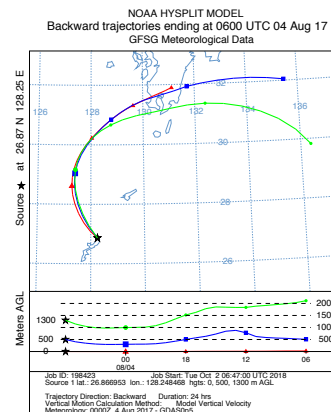
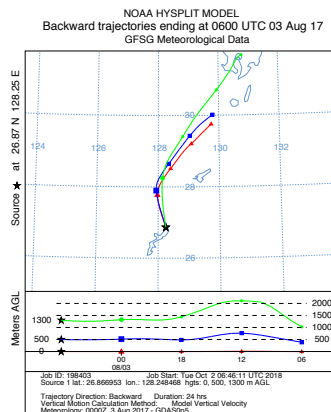
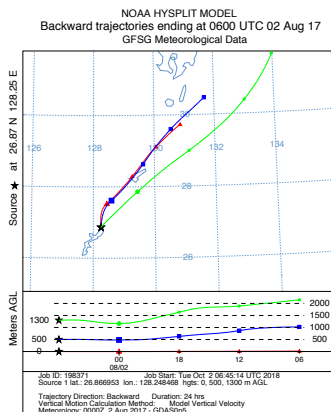


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

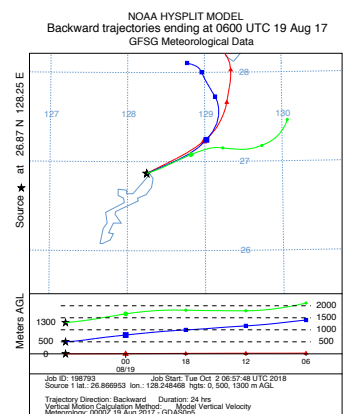
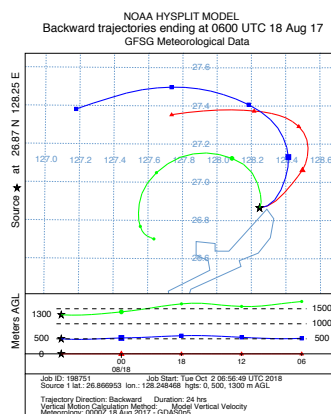
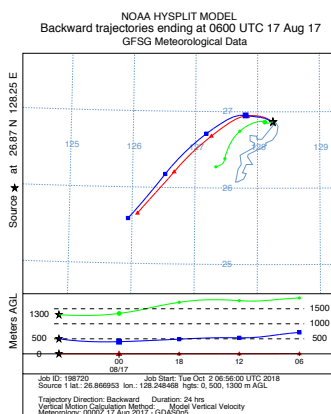
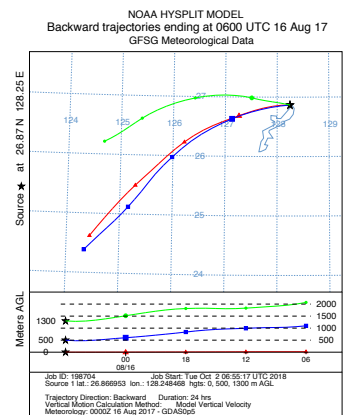
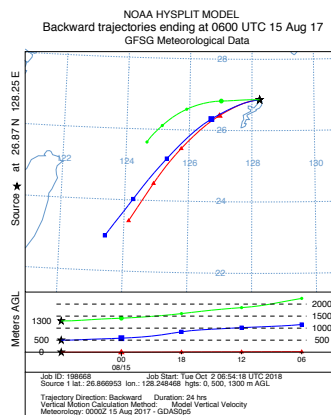
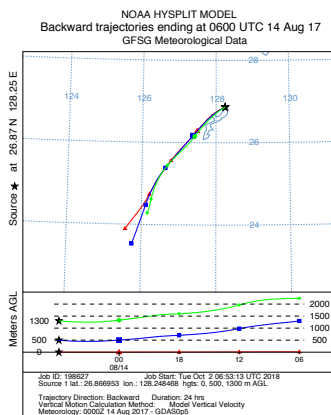
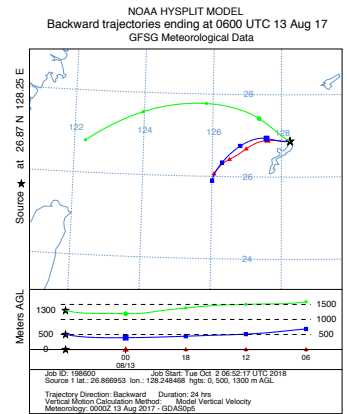
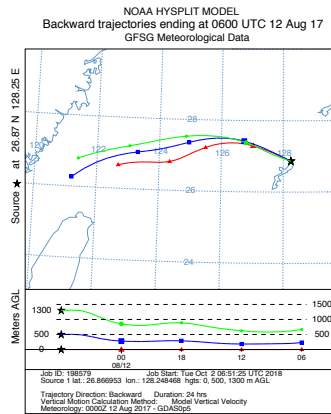
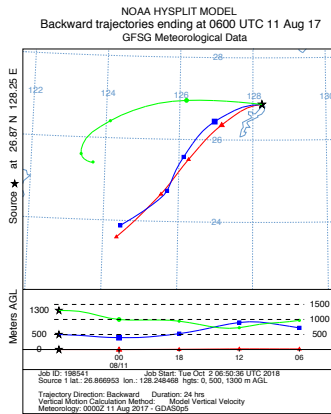


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

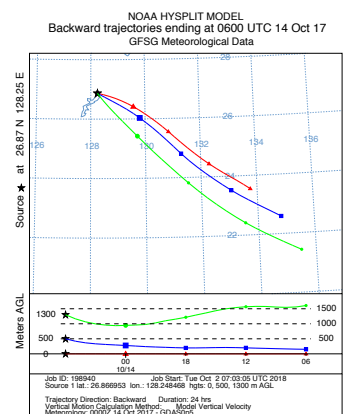
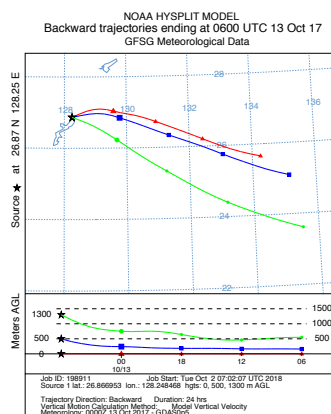
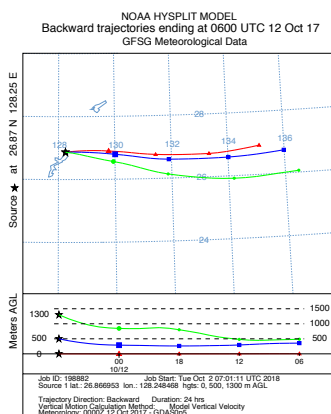
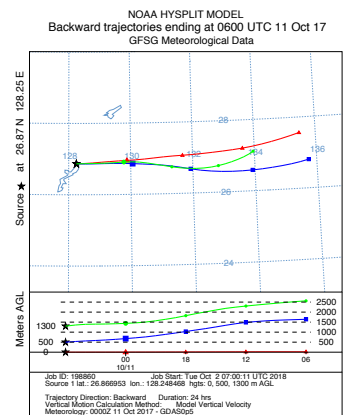
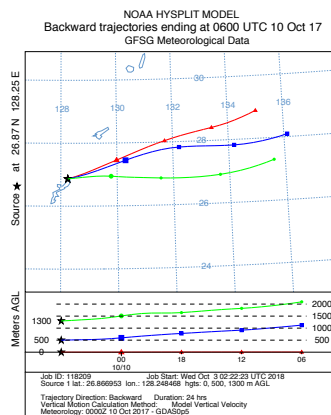
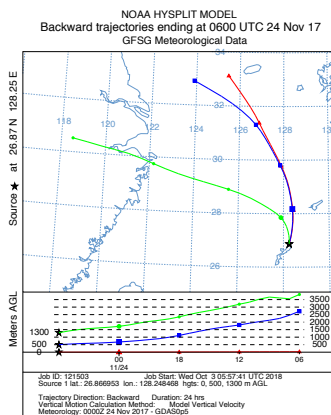
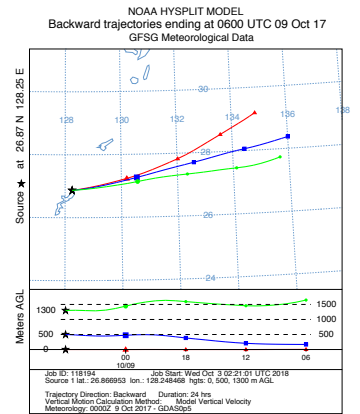
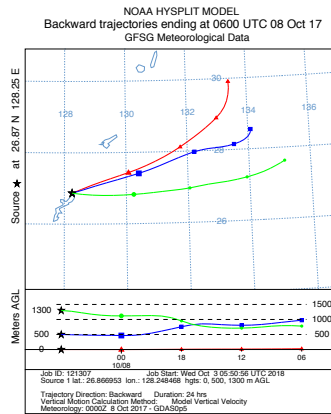
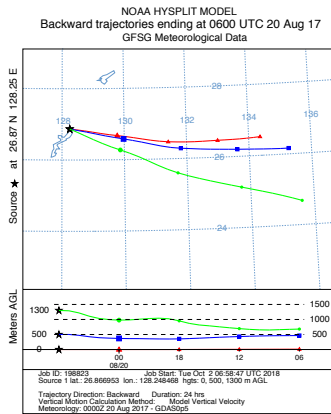


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

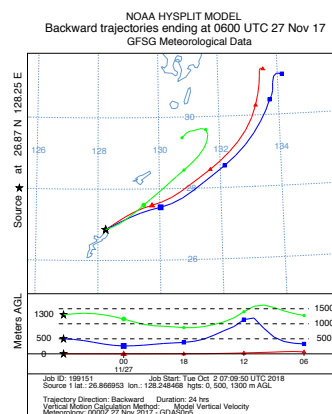
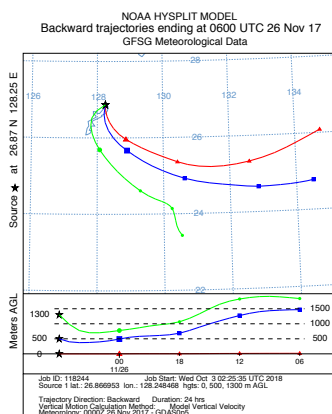
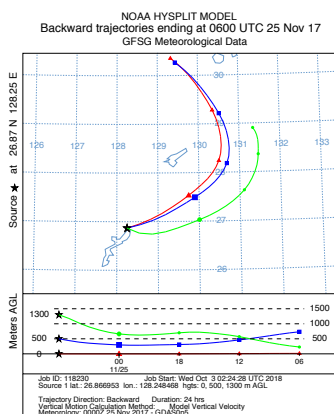
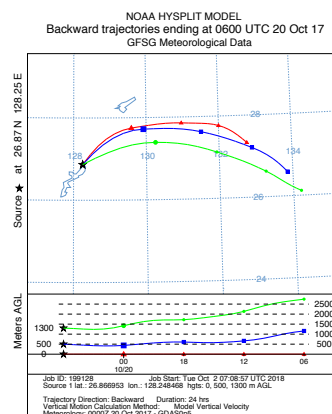
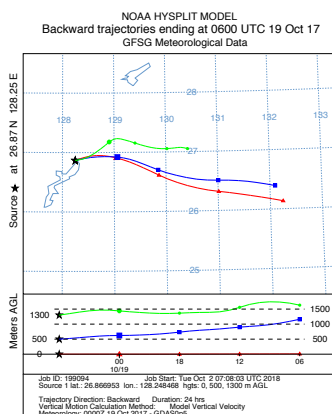
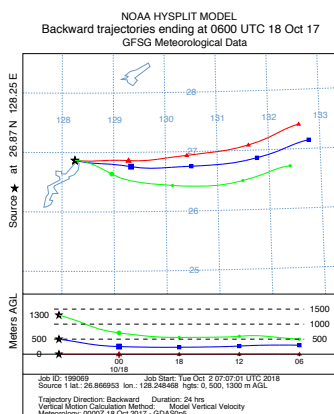
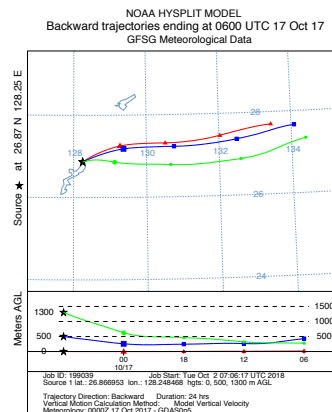
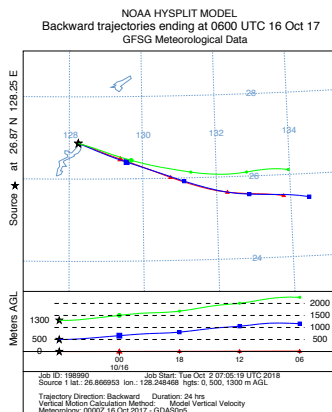
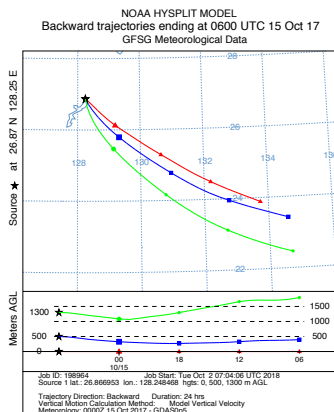


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

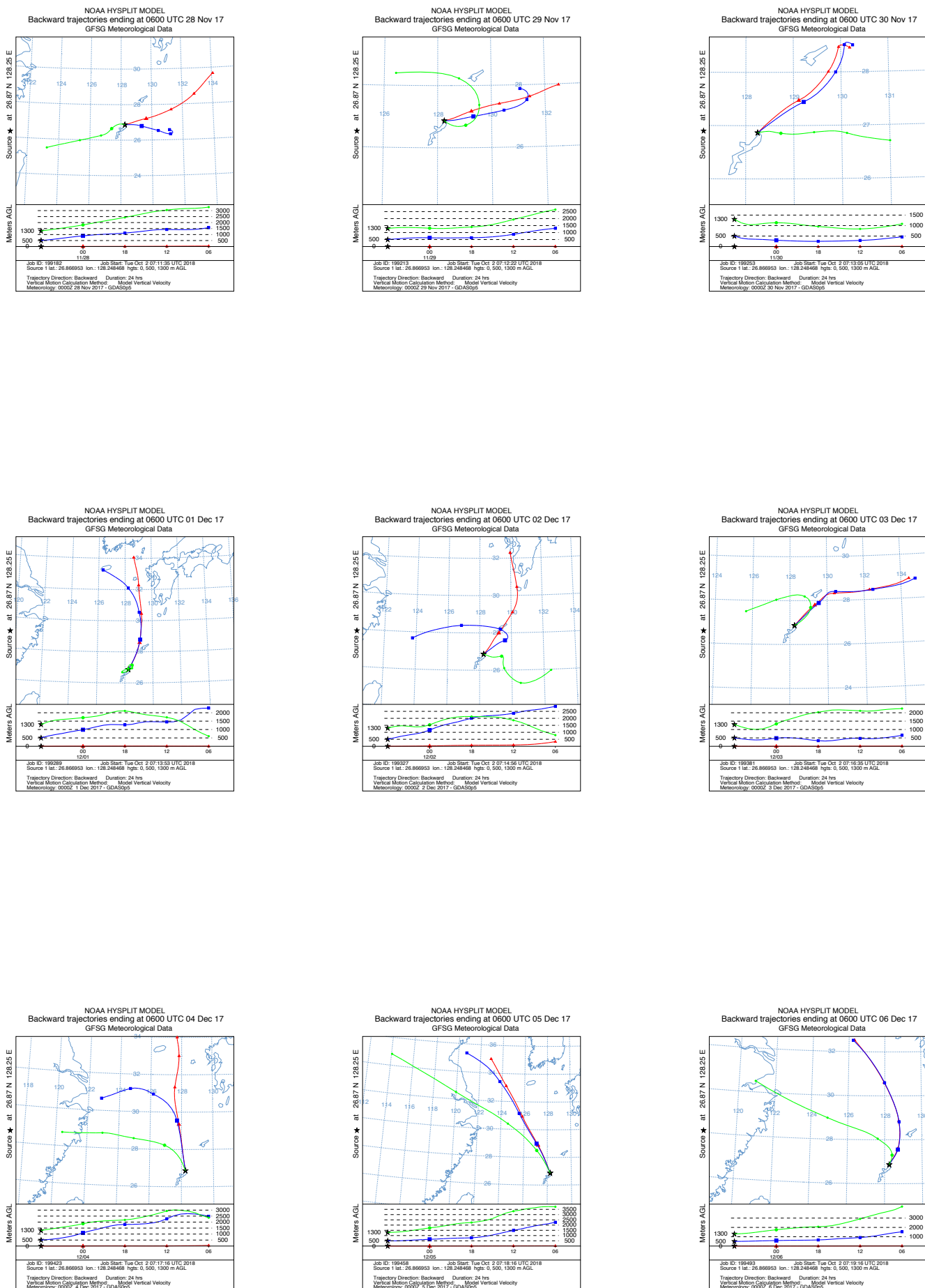


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

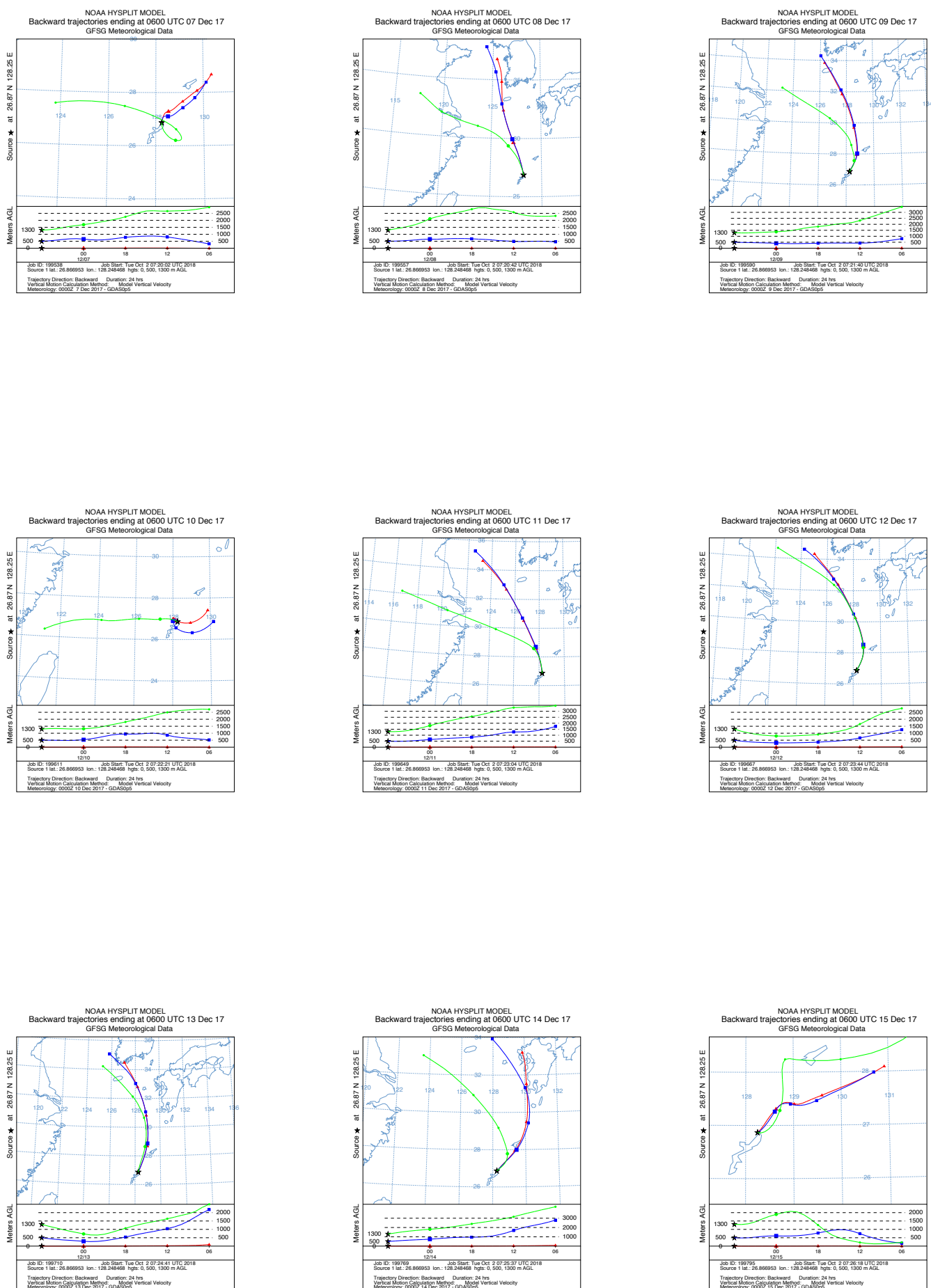


Figure SI 2. Air-mass back trajectories of 24-h duration (continue)

Table S1: Concentrations and isotopic compositions of GEM and wind directions during sampling periods at CHAAMS. Air-mass back trajectories of total run time of 24-hours were calculated for 3 days before sampling, and during sampling using the NOAA HYSPLIT model.

(N: North, S: South, W: West, E: East, NW: Northwest, NE: Northeast, SW: Southwest, SE: Southeast, MD*: Multiple directions)

■ Hg concentration and Hg isotopic compositions								■ Wind directions						
No.	Sampling Date		GEM	$\delta^{202}\text{Hg}$	$\Delta^{199}\text{Hg}$	$\Delta^{200}\text{Hg}$	$\Delta^{191}\text{Hg}$	Before Sampling			During Sampling			
	Start	End	ng/m ³	‰	‰	‰	‰	72-hour	48-hour	24-hour	Day 1	Day 2	Day 3	Day 4
1	2016/12/19	2016/12/22	1.3	-0.05	-0.24	-0.19	-0.23	N	N	E	E	E	E	SE
2	2016/12/22	2016/12/25	1.6	-0.07	-0.20	-0.07	-0.19	E	E	E	SW	NW	NC, SC	E
3	2016/12/25	2016/12/28	1.4	-0.22	-0.29	0.02	-0.18	SW	NW	NC, SC	E	E	NE	N
4	2016/12/28	2016/12/31	1.0	-0.14	-0.25	-0.15	-0.25	E	E	NE	N	NW	N	NE
5	2016/12/31	2017/1/3	1.3	-0.34	-0.18	0.05	-0.15	N	NW	N	NE	NW, N	E, W	N, NW
6	2017/1/3	2017/1/6	1.7	-0.26	-0.16	-0.05	-0.15	NE	NW, N	MD	N, NW	NE, NW	NE	MD
7	2017/1/6	2017/1/9	1.7	-0.19	-0.19	-0.10	-0.19	N, NW	NE, NW	NE	MD	MD	MD	NW
8	2017/1/9	2017/1/12	1.0	-0.09	-0.20	-0.09	-0.12	MD	MD	MD	NW	N	N, NW	N, NW
9	2017/1/25	2017/1/28	1.0	0.16	-0.32	-0.13	-0.22	NW	NW	N, NW	N	NE, NW	E	E
10	2017/1/28	2017/1/31	0.9	0.41	-0.21	0.01	-0.17	NW	N	N, NW	N, NW	SE	NW, W	N, NW
11	2017/3/18	2017/3/21	1.7	0.79	-0.13	-0.04	-0.12	N	NE	S, SW	S	W, NW	SW	NW
12	2017/3/27	2017/3/30	1.4	-0.09	-0.18	-0.09	-0.16	NE, N	N, NW	NW	NW	NW	SE, SW	SE, W
13	2017/3/30	2017/4/2	1.4	-0.41	-0.07	0.04	-0.17	NW	NW	SE, SW	SE, W	S, SW	NW	NW
14	2017/4/2	2017/4/5	1.1	-0.39	-0.14	-0.01	-0.17	SE, W	S, SW	NW	NW	N	NE	SE
15	2017/4/5	2017/4/8	1.3	-0.03	-0.14	-0.07	-0.15	NW	N	NE	SE	SE	SE, W	S
16	2017/4/8	2017/4/11	1.5	-0.30	-0.12	0.00	-0.10	SE	SE	SE, W	S	SW	SW	NW, W
17	2017/4/11	2017/4/14	1.7	-0.15	-0.26	-0.07	-0.07	S	SW	SW	NW, W	NW	N	E, SE
18	2017/4/14	2017/4/17	1.7	-0.32	-0.24	-0.14	-0.12	NW, W	NW	N	E, SE	SE, S	SE, SW	S
19	2017/4/17	2017/4/20	2.1	-0.28	-0.16	-0.03	-0.07	E, SE	SE, S	SE, SW	S	NW, W	NW	S
20	2017/4/28	2017/5/1	0.9	-0.25	-0.22	-0.07	-0.14	E	SE, S	N	N	NW	MD	SE, SW
21	2017/5/1	2017/5/4	1.0	-0.20	-0.15	-0.07	-0.11	N	NW	MD	SE, SW	E, S	SE, SW	E

22	2017/5/4	2017/5/7	1.4	-0.06	-0.19	-0.06	-0.13	SE, SW	E, S	SE, SW	E	S, SW	SW, NW	N, E
23	2017/5/7	2017/5/10	1.6	-0.42	-0.18	-0.01	-0.11	E	S, SW	SW, NW	N, E	E, W	S	NW, W
24	2017/5/10	2017/5/13	1.7	-0.15	-0.15	-0.02	-0.13	N, E	E, W	S	NW, W	MD	S	SW
25	2017/5/13	2017/5/16	1.4	-0.22	-0.22	-0.01	-0.13	NW, W	MD	S	SW	W	MD	NE
26	2017/5/16	2017/5/19	1.3	-0.40	-0.23	-0.11	-0.18	SW	W	MD	NE	NE, N	N	NE
27	2017/5/19	2017/5/22	1.4	-0.51	-0.20	-0.07	-0.11	NE	NE, N	N	NE	E	E	E
28	2017/6/18	2017/6/21	1.2	-0.36	-0.16	-0.05	-0.16	NE, N	MD	E	NE	SE, SW	S, SW	S, SW
29	2017/6/21	2017/6/24	1.3	-0.22	-0.23	-0.09	-0.14	NE	SE, SW	S, SW	S, SW	S, SW	S	S
30	2017/6/27	2017/6/30	0.9	-0.12	-0.19	-0.03	-0.21	SW	SW	SW	SW	S	SE	S, SW
31	2017/6/30	2017/7/3	1.4	0.01	-0.29	-0.10	-0.24	SW	S	SE	S, SW	S, SE	SE	S
32	2017/7/21	2017/7/24	2.3	-2.15	-0.05	0.04	-0.05	NW, W	SW, S	SW	SW	NW, SW	NW	NW
33	2017/7/24	2017/7/27	2.6	-1.77	-0.08	-0.02	-0.13	SW	NW, SW	NW	NW	NW	MD	SE
34	2017/7/27	2017/7/30	1.6	-1.76	-0.08	-0.02	0.03	NW	NW	MD	SE	E	SE, SW	S
35	2017/7/30	2017/8/2	2.4	-1.47	-0.05	0.01	-0.04	SE	E	SE, SW	S	SE	NE	NE
36	2017/8/2	2017/8/5	2.1	-1.46	-0.09	0.01	-0.10	S	SE	NE	NE	N	NE	W
37	2017/8/9	2017/8/12	2.2	-1.70	-0.04	0.02	-0.05	SW, W	SW, W	SW	SW	SW, W	SW, W	W
38	2017/8/12	2017/8/15	2.2	-1.73	0.00	0.00	0.03	SW	SW, W	SW, W	W	SW, W	SW	SW
39	2017/8/15	2017/8/18	2.1	-2.01	-0.09	0.00	-0.14	W	SW, W	SW	SW	SW, W	SW	MD
40	2017/8/18	2017/8/20	2.3	-2.01	-0.08	-0.05	-0.14	SW	SW, W	SW	MD	NE	E, SE	E
41	2017/10/11	2017/10/14	2.1	-1.26	-0.09	-0.07	-0.03	NE, E	E	E	E	E	SE	SE
42	2017/10/14	2017/10/17	2.0	-1.14	-0.09	-0.06	-0.08	E	E	SE	SE	SE	E	E
43	2017/10/17	2017/10/20	1.6	-1.38	-0.14	-0.04	-0.10	SE	SE	E	E	E	E	E
44	2017/11/27	2017/11/30	1.7	-0.73	-0.16	-0.04	-0.11	NW	NE	SE	NE	MD	MD	NE, E
45	2017/11/30	2017/12/3	1.7	-0.53	-0.13	-0.07	-0.15	NE	MD	MD	NE, E	N	MD	MD
46	2017/12/3	2017/12/6	1.3	-0.62	-0.11	-0.03	-0.07	NE, E	N	MD	MD	NW, N	NW	NW
47	2017/12/6	2017/12/9	1.5	-0.38	-0.21	-0.04	-0.18	MD	NW, N	NW	NW	MD	NW	NW

48	2017/12/9	2017/12/12	1.4	-0.56	-0.19	-0.07	-0.13	NW	MD	NW	NW	MD	NW	NW
49	2017/12/12	2017/12/15	1.5	-0.36	-0.12	-0.01	-0.12	NW	MD	NW	NW	N	N, NW	NE

*When it is difficult to determine the direction of the trajectory, the result is written as MD.

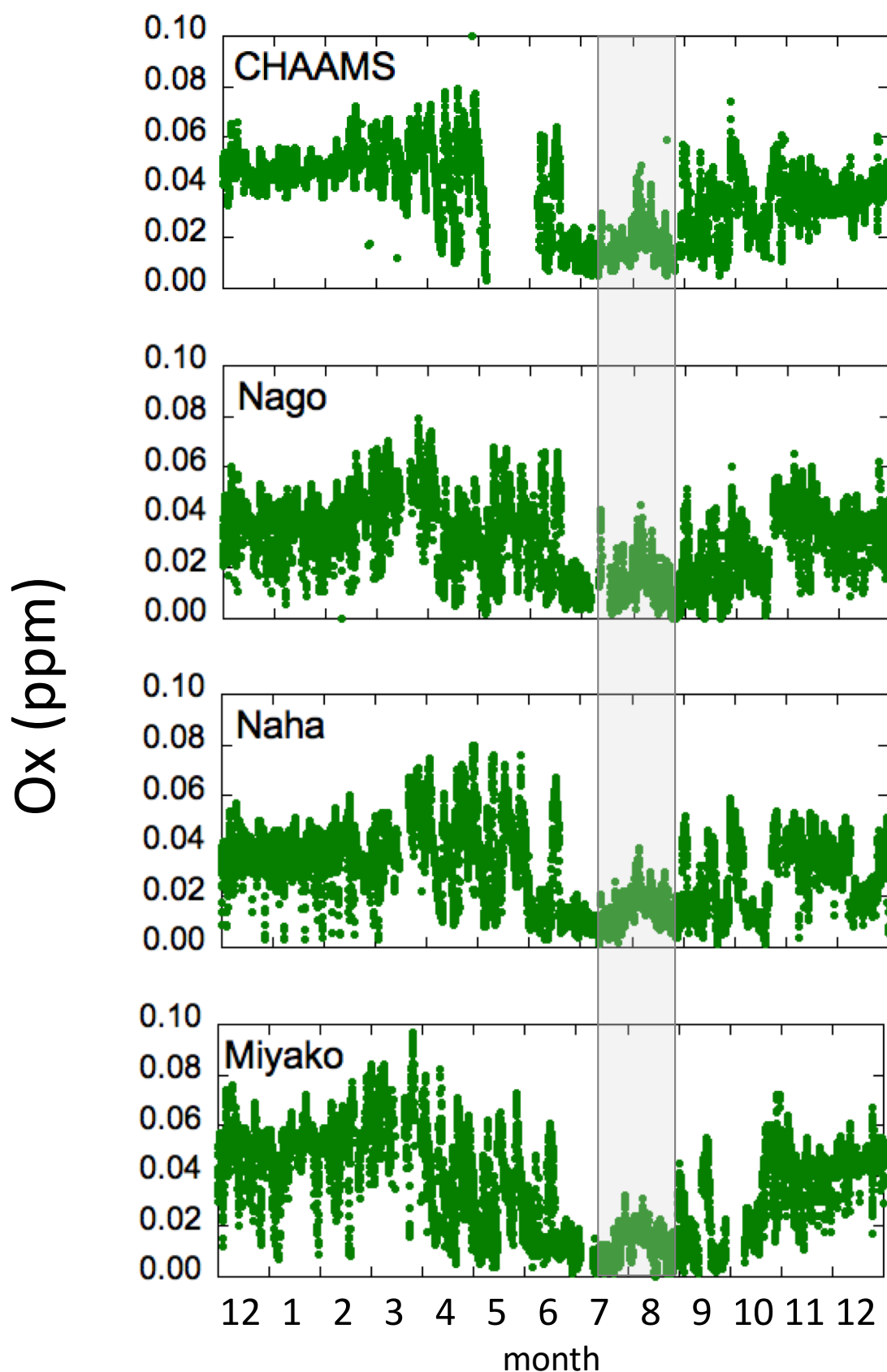


Figure S3. Ox concentrations at the monitoring stations (*preliminary results from <http://soramame.taiki.go.jp>). From late July to August (gray bars), the relatively small peaks indicate low $\delta^{202}\text{Hg}$ values in the GEM samples.