Supplementary Material

ERA5 versus ERA-Interim

In comparing the ERA5 reanalysis with ERA-Interim, a few features stand out. First, we note that for many parameters, the general spatial patterns over the southeast Atlantic are consistent between the two products. Figure S1 shows generally similar spatial patterns, yet upon closer examination the differences at a given point in time are large (Figure S1). This results in generally better agreement between ERA5 and the observations, as compared with ERA-Interim (Figure S2).

For water vapor specifically, ERA-Interim still generally captures the presence of an elevated water vapor signal in the altitudes above the boundary layer, but often this high-humidity air was displaced to a lower altitude than what was observed (e.g., Figure S2).

It is not clear which changes from ERA-Interim to ERA5 may be responsible for this improvement, and previous work has shown the differences between the two are not due to any one factor, i.e. not solely due to ERA5's improved spatiotemporal resolution (Hoffmann et al., 2019). Nonetheless, it is reassuring that this newest ECMWF product agrees so well with the aircraft observations, and this gives us confidence that the ERA5 meteorology may be consistent with real-world meteorology over the continental source region as well.

Supplementary Figures

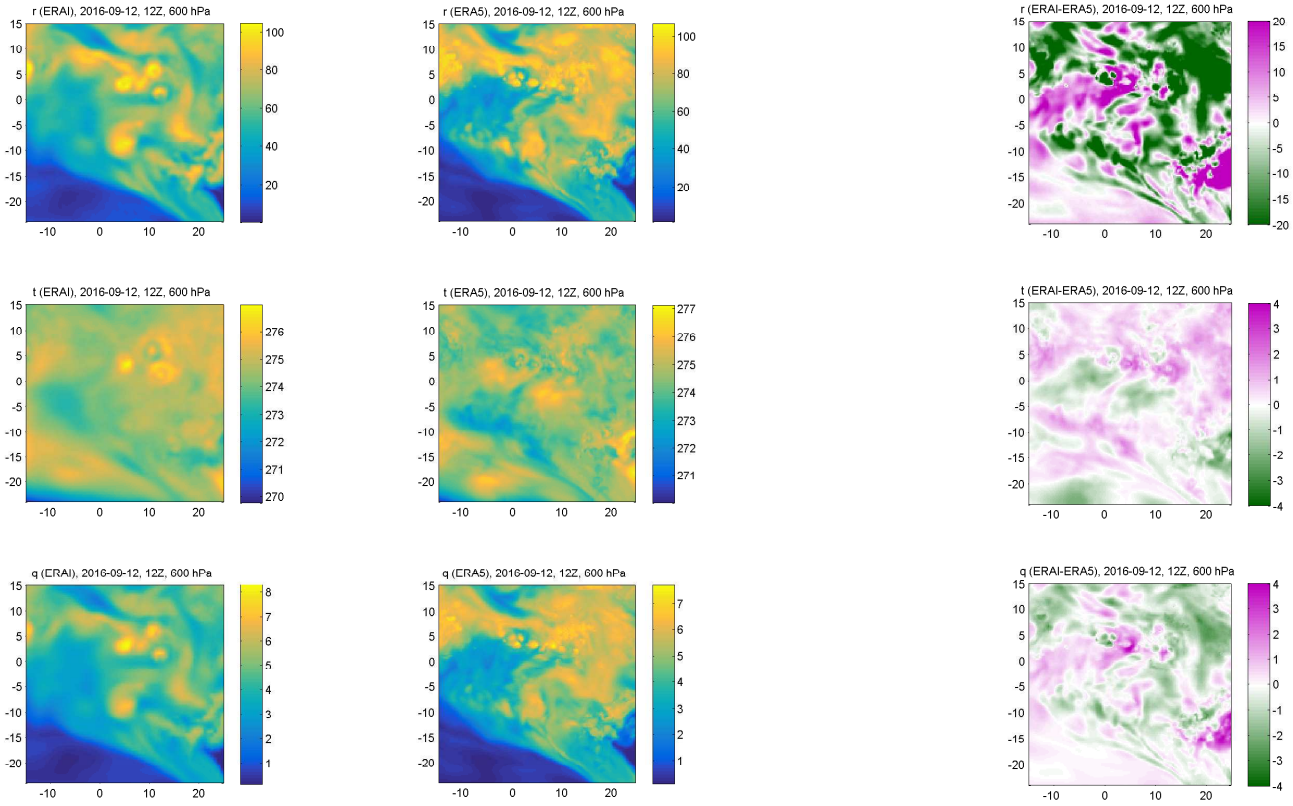


Figure S1. Maps showing an example of relative humidity (r, top), temperature (t, middle), and specific humidity (q, bottom) fields at 600hPa (approximately 4.4km) for 12 September 2016, 12Z for ERA-Interim (left), ERA5 (middle), and the difference between the two (right). While ERA5 shows much higher resolution, both products show areas of high humidity around (10°S, 5°E), and two areas of high temperature appear near the equator in both products, though significantly offset from one another by several degrees of longitude.

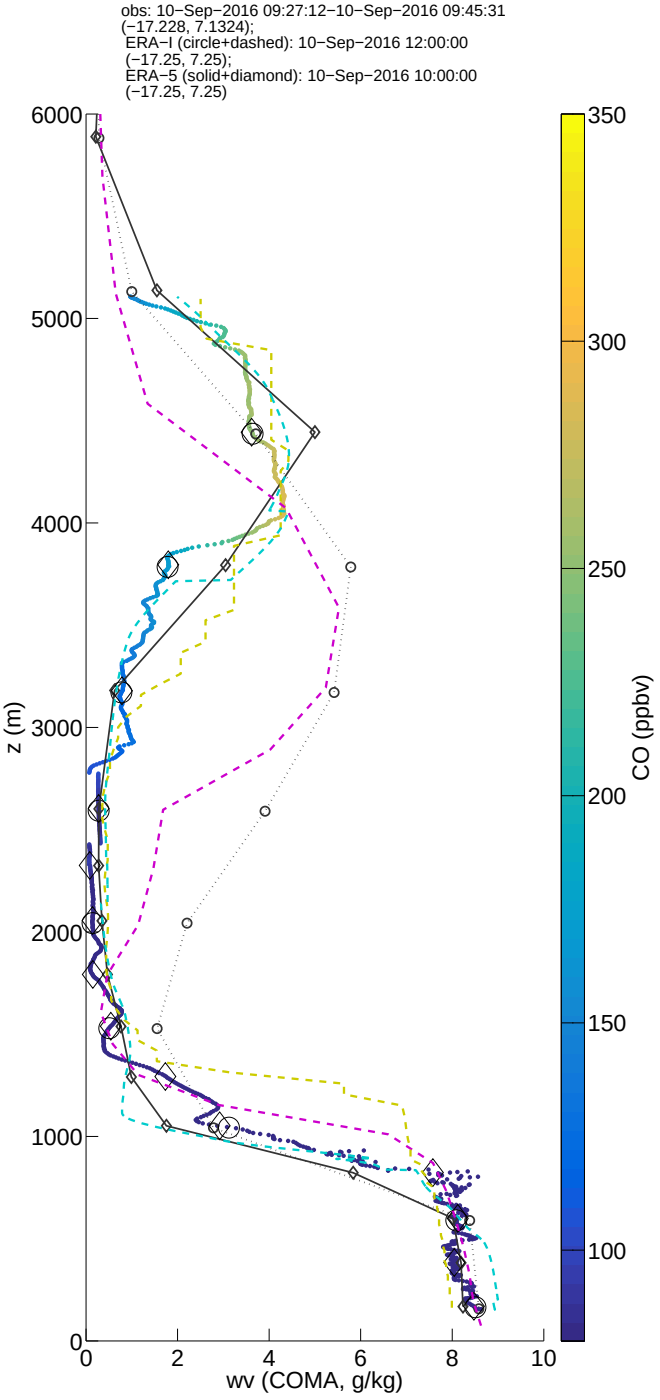


Figure S2. Profile from 10 Sep 2016 showing a common difference between ERA-Interim (dotted black line), ERA5 (solid black line), MERRA-2 (dashed purple line), WRF-CAM5 (dashed aqua line), and WRF-Chem (dashed gold line) as compared with the observed values (colored according to CO): namely, that the location of the plume magnitude was often lower in ERA-Interim than either ERA5 or the ORACLES observations. Larger hollow shapes indicate the 50-m observed averages around a given reanalysis height used in Figure 6.

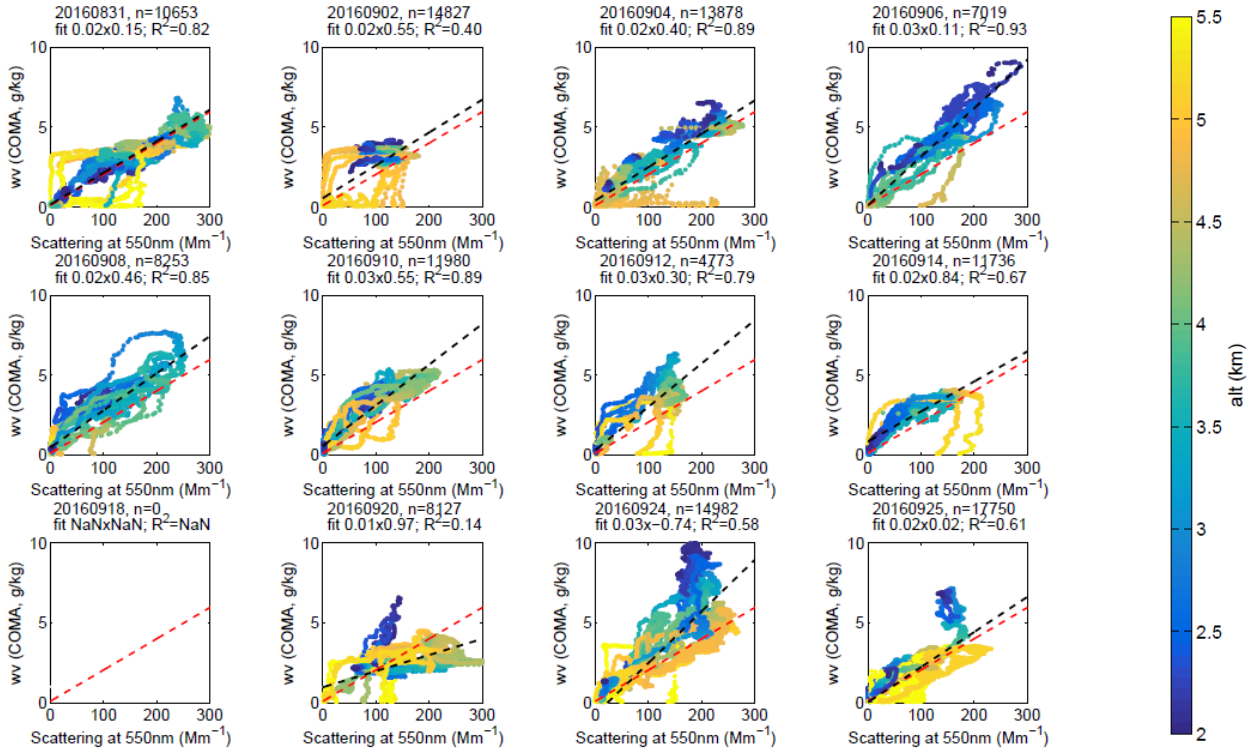
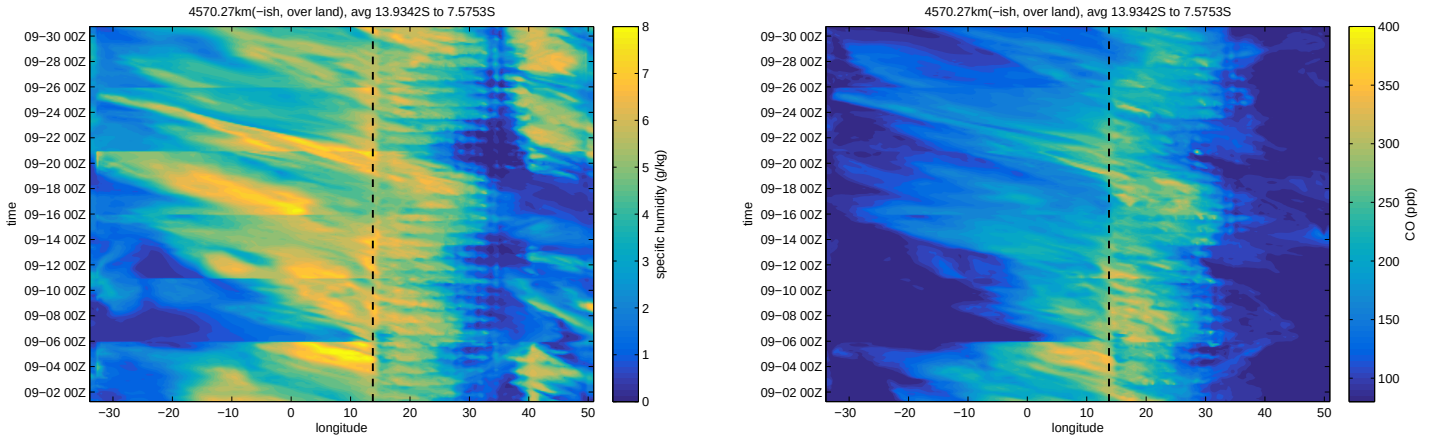


Figure S3. Similar to Figure 3 but here showing scattering at 550nm at actual temperature and pressure (measured by a 3-wavelength TSI Nephelometer) versus water vapor from COMA. The deviations from the coherent line at high altitudes (yellow points) are likely due to a known issue with the TSI Nephelometer in the 2016 deployment, where due to a clogged filter, dead volume within the instrument expanded into the sample volume and caused a delay in the scattering signal response when ascending and flying from polluted into pristine conditions, or when descending and flying from pristine into polluted air (i.e., exiting or entering the top of plume layer). The results are similar for total aerosol extinction (defined as aerosol scattering plus absorption).



(a) (b)

Figure S4. As in Figure 9, but showing water vapor and CO both from WRF-CAM5. The effects of the reinitialization (i.e., the 3-daily discontinuity) is evident.

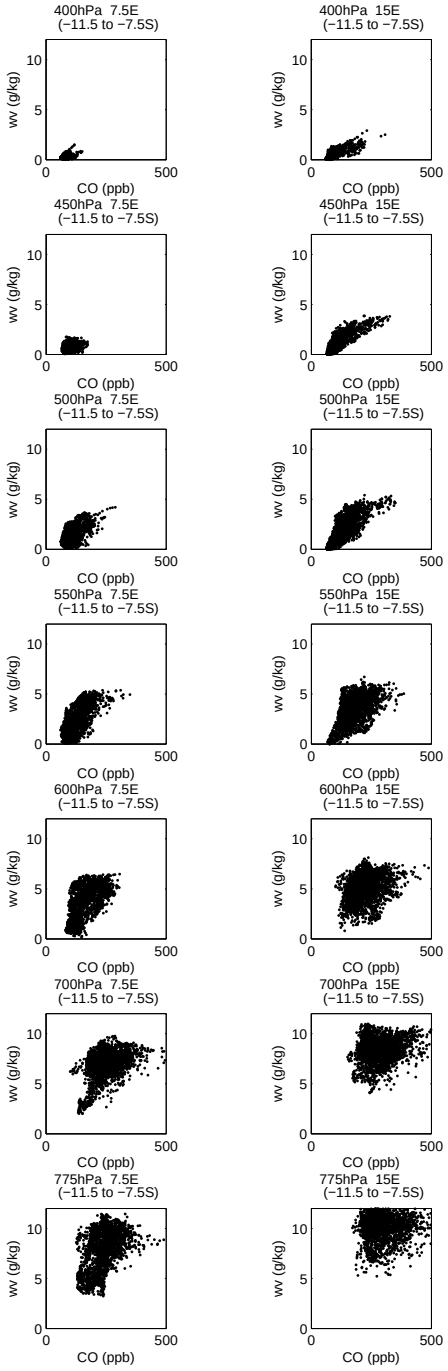


Figure S5. As in Figure 13, for selected altitude levels for the entire month of September (MERRA-2, every 3 hours), showing the dependence of the linear relationship on over-continent mixing between boundary-layer-influenced air and the dry, clean higher troposphere.

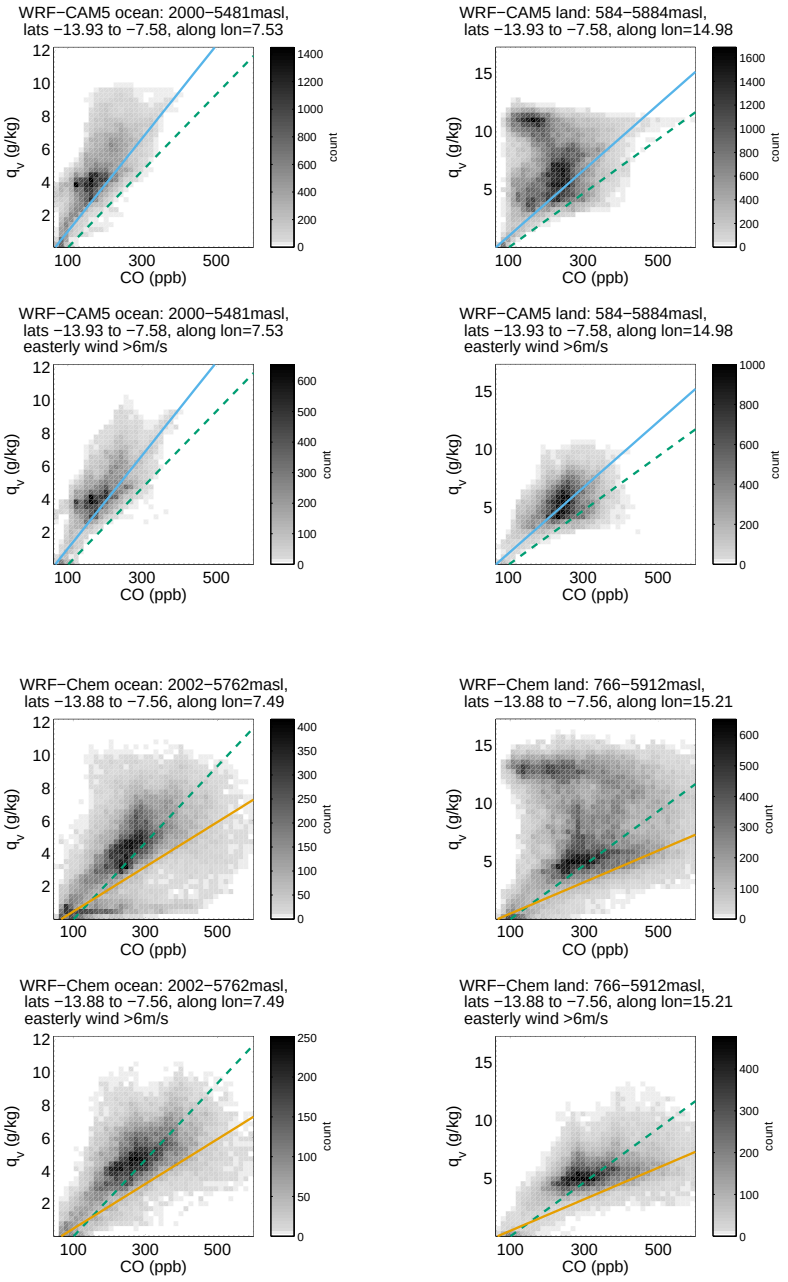


Figure S6. As in Figure 13 but for the two WRF models.

Table S1. Location and time of all aircraft profiles.

date	time		latitude (°)		longitude (°)		altitude (m)	
	start	end	start	end	start	end	start	end
Zone 1								
31-Aug-2016	11:27:11	11:33:00	-13.1267	-12.768	2.9636	2.6187	3830	5444
31-Aug-2016	11:33:01	11:55:00	-12.767	-13.0011	2.6177	2.841	5446	66
04-Sep-2016	11:09:06	11:21:44	-12.5693	-11.8787	2.4281	1.7692	188	4181
08-Sep-2016	10:55:37	11:11:30	-9.9071	-10.8162	0.0131	0.82224	5812	1095
08-Sep-2016	11:16:22	11:22:19	-11.07	-11.3561	1.0804	1.372	1181	73
08-Sep-2016	11:56:10	12:08:25	-13.1286	-13.8894	3.1628	3.9044	2151	5181
12-Sep-2016	10:59:51	11:21:39	-10.7495	-9.7424	0.72939	-0.3378	6467	399
12-Sep-2016	12:21:35	12:42:01	-12.6686	-13.7885	2.7179	3.8058	581	5811
12-Sep-2016	12:42:34	12:47:18	-13.8234	-14.1341	3.8398	4.1444	5822	4520
25-Sep-2016	10:41:33	11:05:27	-14.0069	-12.625	4.0053	2.6242	6447	73
25-Sep-2016	11:50:10	12:01:17	-10.4037	-9.8443	0.40059	-0.15597	1375	4533
25-Sep-2016	13:15:18	13:34:44	-12.2769	-13.4552	2.2755	3.4497	5799	82
Zone 2								
06-Sep-2016	11:31:46	11:39:59	-11.6386	-12.1721	9.0479	9.2325	165	2608
06-Sep-2016	11:50:48	11:57:28	-13.0133	-13.518	9.5255	9.702	2586	4661
18-Sep-2016	09:14:19	09:43:12	-13.777	-11.4069	11.2054	11.5	5201	1111
18-Sep-2016	10:38:51	10:44:51	-10.8458	-10.787	11.4998	11.464	477	1856
18-Sep-2016	11:18:40	11:42:53	-10.649	-10.8582	11.5001	11.4998	1129	5820
24-Sep-2016	08:50:27	09:22:43	-13.9012	-11.4865	11	11.0002	4880	206
24-Sep-2016	09:51:49	09:55:05	-9.7382	-9.5416	10.9999	10.9999	1414	474
24-Sep-2016	10:00:43	10:24:22	-9.203	-9.1643	11.0001	10.9876	477	6448
24-Sep-2016	10:41:13	10:51:27	-10.0496	-9.1688	11.0092	11.0001	6435	3855
24-Sep-2016	11:05:07	11:18:31	-7.9816	-7.9164	11.0004	10.8992	3856	144
24-Sep-2016	11:58:37	12:07:51	-10.2521	-10.9441	10.9997	11.0001	2075	4847
24-Sep-2016	12:34:10	12:39:59	-12.9787	-12.5574	10.9935	10.9999	4824	6426
24-Sep-2016	12:43:25	13:03:46	-12.2839	-12.3613	11.0018	11.0757	6437	153

Table S2. Continued from Table S1.

date	time		latitude (°)		longitude (°)		altitude (m)	
	start	end	start	end	start	end	start	end
Zone 3								
31-Aug-2016	10:35:38	10:38:59	-15.9804	-15.8023	5.7642	5.5859	74	921
31-Aug-2016	12:35:21	12:50:14	-14.9252	-15.762	4.7151	5.5452	1235	5374
04-Sep-2016	10:28:21	10:39:06	-14.7051	-14.1143	4.4993	3.921	4201	1010
08-Sep-2016	09:25:47	09:34:52	-15.8069	-15.2742	5.763	5.2584	3719	5825
08-Sep-2016	12:12:35	12:26:18	-14.16	-14.9951	4.1698	4.995	5167	1203
08-Sep-2016	12:30:01	12:34:59	-15.1931	-15.4367	5.1923	5.4364	1175	69
10-Sep-2016	09:58:50	10:10:33	-15.9913	-15.2998	5.9386	5.2826	4154	518
10-Sep-2016	10:38:51	10:43:04	-14.1875	-14.366	4.1133	4.3421	1356	170
10-Sep-2016	11:09:28	11:26:48	-14.4579	-14.453	4.4561	4.4526	1307	5161
10-Sep-2016	11:34:14	11:52:14	-14.5431	-14.0152	4.5299	3.992	5129	539
12-Sep-2016	12:58:03	13:07:20	-14.7356	-15.2648	4.7376	5.2638	4520	1915
12-Sep-2016	13:17:22	13:23:27	-15.8326	-16.1396	5.8311	6.1352	1919	463
25-Sep-2016	14:12:51	14:28:13	-15.3605	-16.2562	5.3534	6.253	1278	5799
Zone 4								
02-Sep-2016	08:59:35	09:11:40	-16.2265	-15.2585	9.9997	9.9999	4532	898
02-Sep-2016	09:39:00	09:52:35	-14.9263	-15.6429	9.9506	9.1708	1357	5471
02-Sep-2016	10:13:21	10:28:40	-15.8814	-16.0238	8.9271	8.8474	5492	950
02-Sep-2016	11:38:10	11:52:35	-15.7431	-14.7557	8.8624	8.5356	83	5429
02-Sep-2016	12:11:44	12:26:08	-14.9693	-15.0161	8.6037	8.5387	5432	1052
06-Sep-2016	09:36:41	09:40:14	-15.9555	-15.6823	9	9.0004	874	154
06-Sep-2016	12:10:56	12:17:46	-14.6011	-15.1355	10.0841	10.2742	4362	2267
24-Sep-2016	13:35:10	13:51:35	-14.2117	-15.461	11.0002	11	155	5179

Table S3. Continued from Table S2.

date	time		latitude (°)		longitude (°)		altitude (m)	
	start	end	start	end	start	end	start	end
Zone 5								
31-Aug-2016	09:58:01	10:03:52	-17.9285	-17.6115	7.7431	7.4169	4310	5946
31-Aug-2016	10:05:50	10:25:50	-17.5	-16.4711	7.3027	6.2571	5966	109
31-Aug-2016	13:14:40	13:45:00	-17.2077	-17.1987	7.0033	7.0118	5790	110
04-Sep-2016	09:33:28	09:48:42	-17.7157	-16.9429	7.5241	6.7344	121	4609
08-Sep-2016	09:03:10	09:11:53	-17.1241	-16.5987	7.0276	6.52	5812	3228
10-Sep-2016	09:20:38	09:27:11	-18.0898	-17.7198	8.0082	7.6267	1139	157
10-Sep-2016	09:27:12	09:45:31	-17.7189	-16.772	7.6257	6.6866	155	5106
10-Sep-2016	12:54:16	13:23:03	-15.937	-17.5813	5.9344	7.5795	524	5117
12-Sep-2016	13:56:15	14:01:59	-17.5973	-17.8316	7.596	7.8318	1019	56
14-Sep-2016	12:06:57	12:14:30	-16.8616	-16.2759	7.4951	7.5004	3214	844
14-Sep-2016	13:06:30	13:22:01	-16.1799	-16.5603	7.5002	7.4995	920	5814
14-Sep-2016	13:33:35	13:41:48	-17.4325	-17.1307	7.5003	7.9419	5792	3189
25-Sep-2016	14:48:34	14:57:02	-17.5897	-18.0968	7.5877	8.0948	5782	7578
Zone 6								
02-Sep-2016	08:41:08	08:51:15	-17.6616	-16.8957	10	9.9999	949	3911
02-Sep-2016	12:50:50	13:05:37	-15.6301	-16.7724	8.8255	9.1311	695	5113
02-Sep-2016	13:05:38	13:21:21	-16.7738	-17.9995	9.1314	9.4577	5113	181
06-Sep-2016	12:28:51	12:36:54	-15.9705	-16.5099	10.5728	10.7674	2257	72
14-Sep-2016	09:45:08	10:06:18	-17.4391	-16.4302	9.0002	8.9995	84	5966
14-Sep-2016	10:06:23	10:24:33	-16.4366	-17.9344	8.9996	8.998	5978	792
14-Sep-2016	11:09:27	11:11:09	-17.0586	-17.0066	9.0002	9.0716	348	775
14-Sep-2016	11:20:56	11:29:09	-17.4257	-17.1612	9.0001	8.9999	778	3223
14-Sep-2016	11:33:21	11:40:50	-16.9169	-17.4929	9.0506	8.9999	3207	747
14-Sep-2016	11:43:42	11:51:00	-17.6615	-17.433	9.0004	8.6017	802	3229
14-Sep-2016	14:00:56	14:14:16	-16.6264	-17.6201	8.7888	8.9413	3508	454
18-Sep-2016	08:32:55	08:41:38	-17.2134	-16.466	11	10.9999	5184	2601
20-Sep-2016	08:27:14	08:47:49	-17.515	-15.8821	10.4999	10.5001	5835	81
20-Sep-2016	08:53:33	09:09:10	-15.6696	-16.702	10.504	10.4998	82	4544
20-Sep-2016	09:09:11	09:22:21	-16.7032	-16.7263	10.4998	10.5093	4548	621
20-Sep-2016	10:15:27	10:30:39	-15.9302	-16.9028	10.5024	10.5	526	4357
20-Sep-2016	10:45:33	10:57:44	-16.6074	-16.4061	9.9747	8.9987	4346	643
20-Sep-2016	11:52:23	12:17:49	-16.673	-16.936	8.9997	9.0004	367	7111
20-Sep-2016	12:25:30	12:36:57	-17.5463	-17.4521	9.0001	8.9977	7081	3700
20-Sep-2016	12:47:19	12:58:42	-16.7612	-16.7499	8.9994	8.9429	3687	214
20-Sep-2016	13:10:00	13:13:20	-17.2842	-17.4774	8.9996	8.9998	237	1093

Table S4. Continued from Table S3.

date	time		latitude (°)		longitude (°)		altitude (m)	
	start	end	start	end	start	end	start	end
Zone 7								
02-Sep-2016	08:11:40	08:25:00	-19.9902	-18.9255	10.0022	9.9998	4616	851
02-Sep-2016	13:21:22	13:37:52	-18.0004	-19.2451	9.458	9.7935	176	5405
02-Sep-2016	13:40:01	13:43:20	-19.4362	-19.7287	9.8455	9.9258	5411	4461
04-Sep-2016	08:49:40	09:04:49	-19.9512	-19.1492	9.8926	9.0303	4798	800
06-Sep-2016	07:55:11	08:17:07	-23.0025	-22.0098	10.0194	8.9998	6385	80
06-Sep-2016	09:01:51	09:08:20	-18.6094	-18.131	8.9986	8.9999	555	2487
12-Sep-2016	14:09:14	14:30:44	-18.0997	-19.2671	8.1009	9.2841	86	5817
14-Sep-2016	14:14:17	14:27:33	-17.6213	-18.5055	8.9415	9.412	453	4431
20-Sep-2016	13:25:05	13:41:45	-18.1472	-19.0937	9.1206	9.8954	791	5827
25-Sep-2016	14:57:03	15:17:54	-18.0978	-19.4052	8.0958	9.3978	7579	1222
Zone 8								
31-Aug-2016	08:26:40	08:45:59	-22.3478	-21.4678	12.5055	11.5319	5791	163
31-Aug-2016	08:55:25	08:59:59	-21.0348	-20.814	11.0592	10.8207	83	1084
06-Sep-2016	07:23:10	07:34:04	-22.9802	-22.9907	12.6261	11.8288	3871	6383
06-Sep-2016	13:25:30	13:33:51	-19.5226	-20.0817	11.8769	12.0821	162	2595
08-Sep-2016	13:58:20	14:12:09	-20.3579	-21.1365	10.3287	11.1698	4175	191
08-Sep-2016	14:17:17	14:32:09	-21.3776	-22.2566	11.433	12.4028	178	4820
10-Sep-2016	08:29:25	08:47:09	-20.9431	-19.9548	10.9988	9.9552	5795	400
14-Sep-2016	08:59:31	09:15:06	-20.626	-19.5658	10.6348	9.933	3248	761
25-Sep-2016	15:20:53	15:35:51	-19.5652	-20.4699	9.5589	10.4943	1240	5794