Figure S1. Top: Jena Instrument for Greenhouse gases (JIG) installed onboard German research aircraft HALO. Bottom-left: Jena Air Sampler (JAS, bottom) aboard HALO. Bottom-right: JAS instrument in the lab before installation on the aircraft; also visible are two sample flasks used in the CoMet 1.0 campaign, mounted in the instrument.

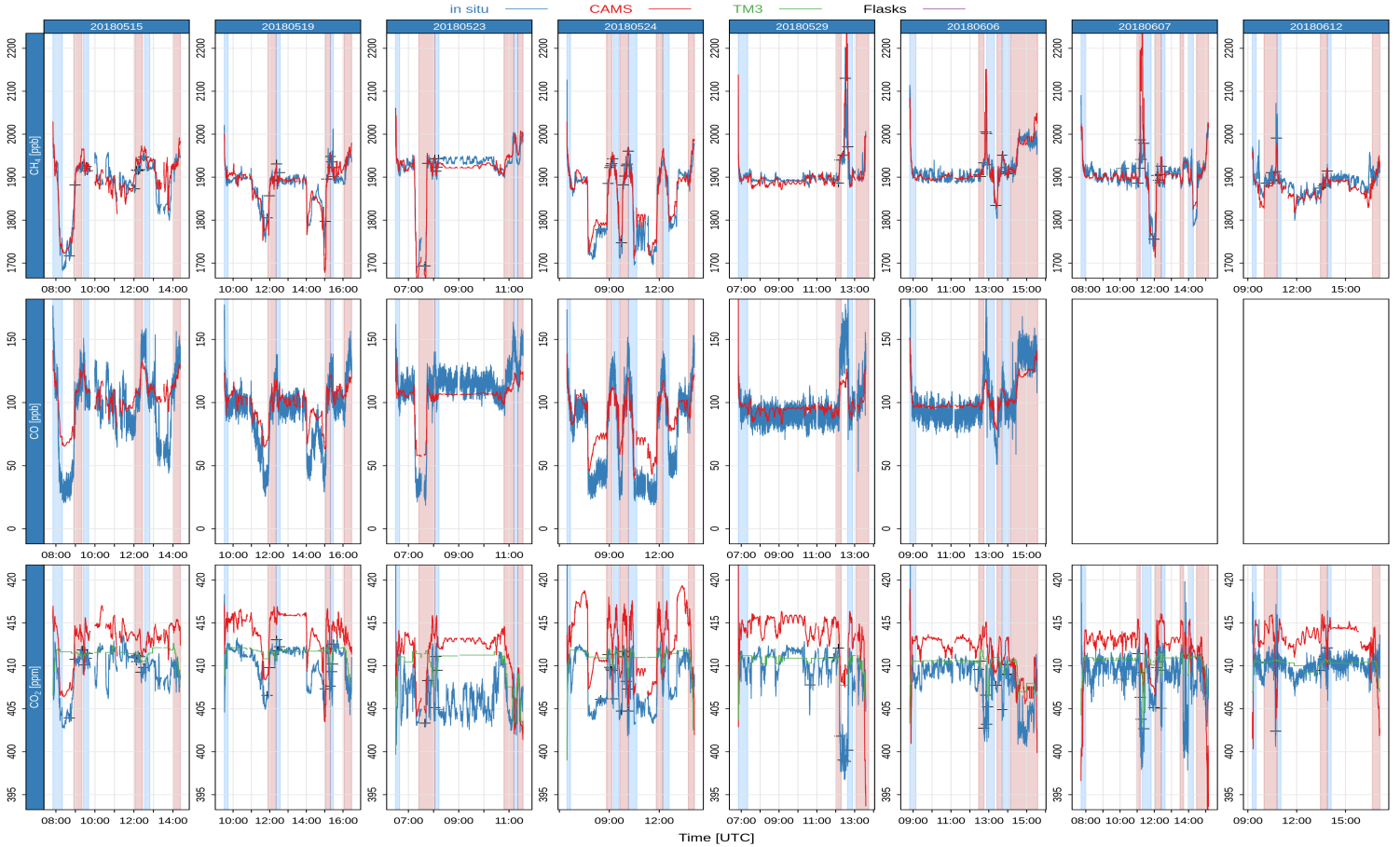

Figure S2. Time-plots of in situ measurements of CH_4, CO and CO_2 performed throughout CoMet campaign aboard HALO aircraft with JIG instrument (blue). Model results from CAMS and TM3 extracted at respective time and aircraft coordinates are given in red and green, respectively. Results from flask samples collected with JAS marked with crosses. Shading denotes data flagged as "vertical profiles" and used for characterization of vertical structure of the atmosphere over the study area. Blue shading - ascending profile. Red shading - descending profile. CO data for flights on June 7th and June 12th discarded due to instrument malfunction. See the manuscript for details.

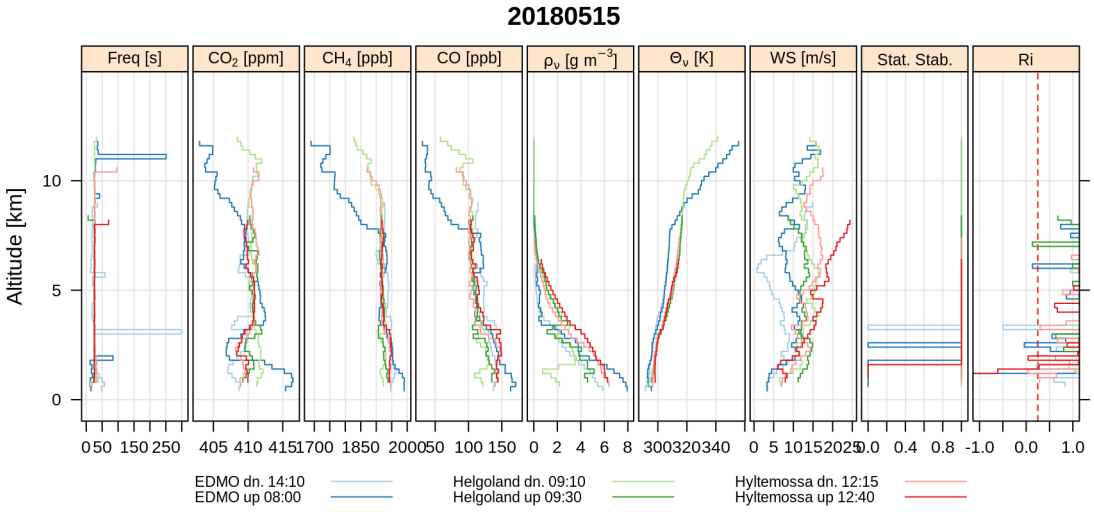

Figure S3. Detailed profile information - flight on May 15th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Locations and their abbreviations can be found in Table S1.

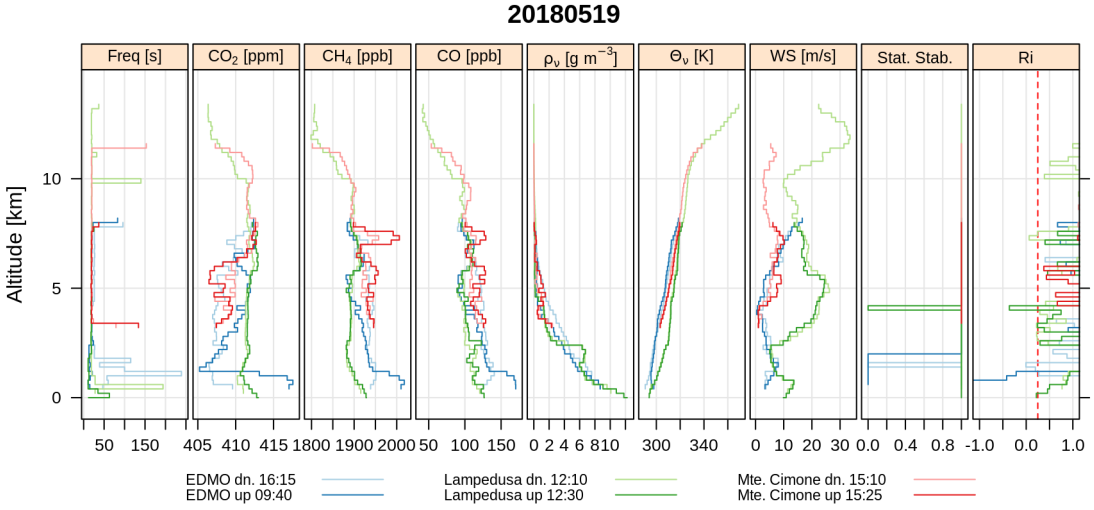

Figure S4. Detailed profile information - flight on May 19th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Locations and their abbreviations can be found in Table S1.

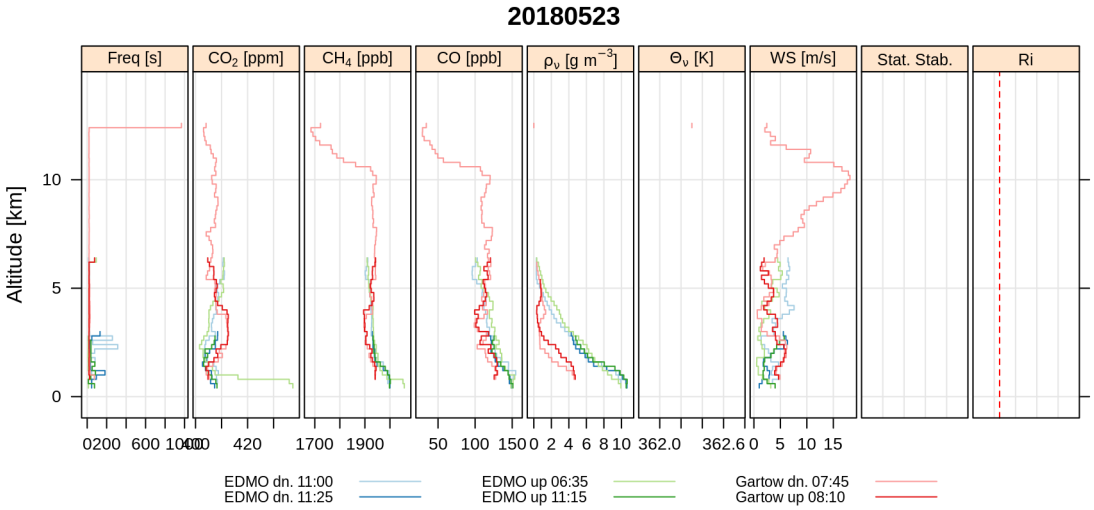

Figure S5. Detailed profile information - flight on May 23rd, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Locations and their abbreviations can be found in Table S1.

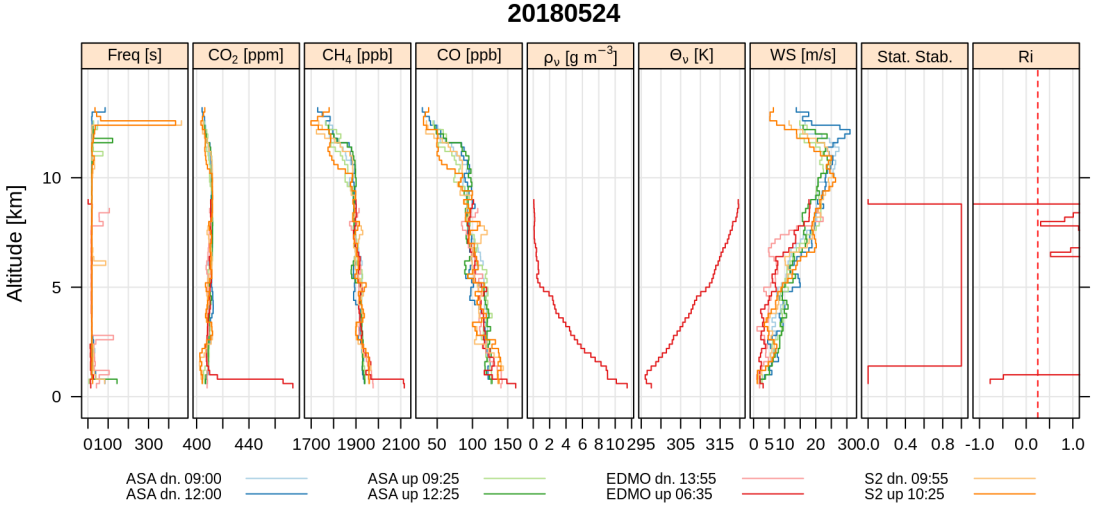

Figure S6. Detailed profile information - flight on May 24th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Locations and their abbreviations can be found in Table S1.

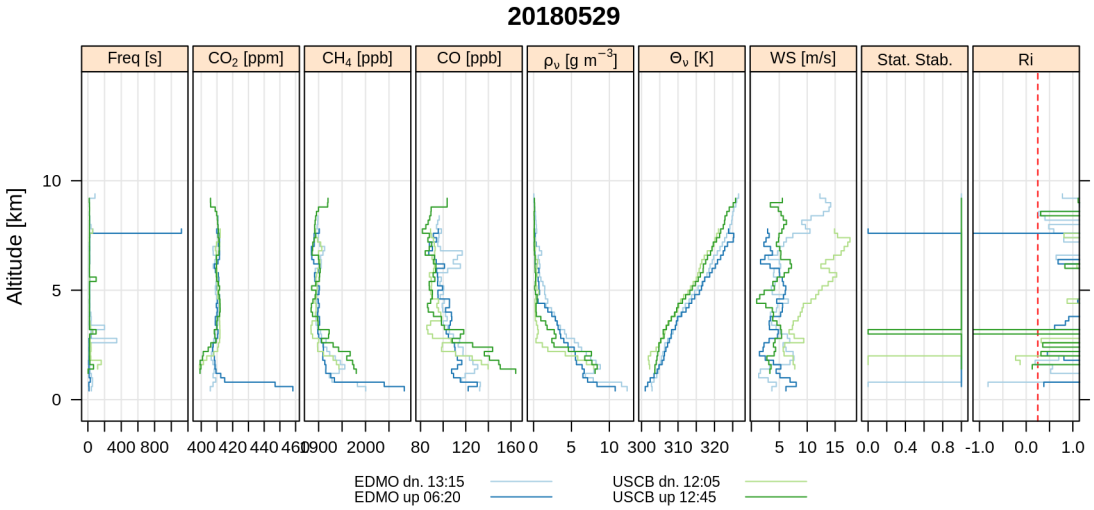

Figure S7. Detailed profile information - flight on May 29th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Locations and their abbreviations can be found in Table S1.

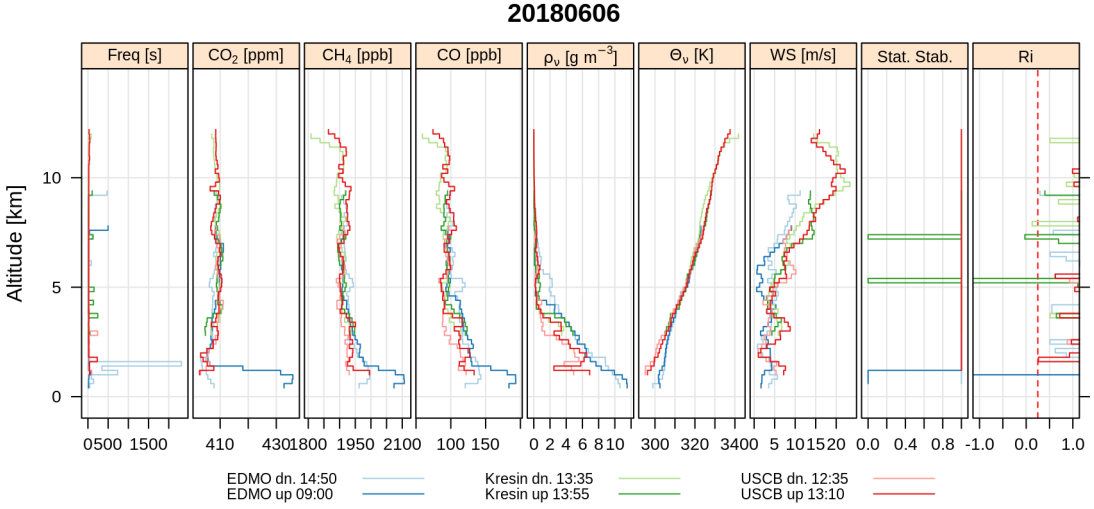

Figure S8. Detailed profile information - flight on June 6th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Locations and their abbreviations can be found in Table S1.

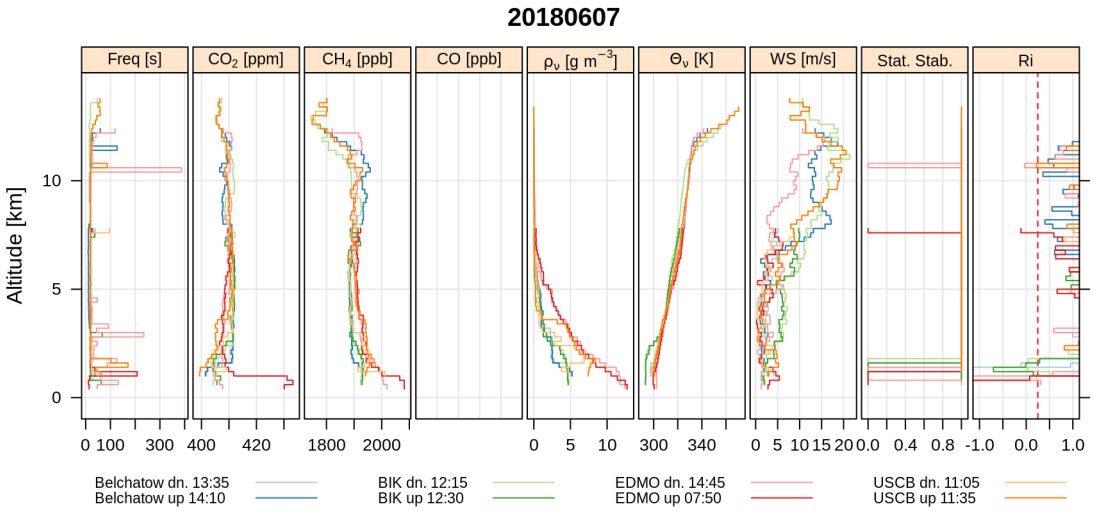

Figure S9. Detailed profile information - flight on June 7th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) an Locations and their abbreviations can be found in Table S1.

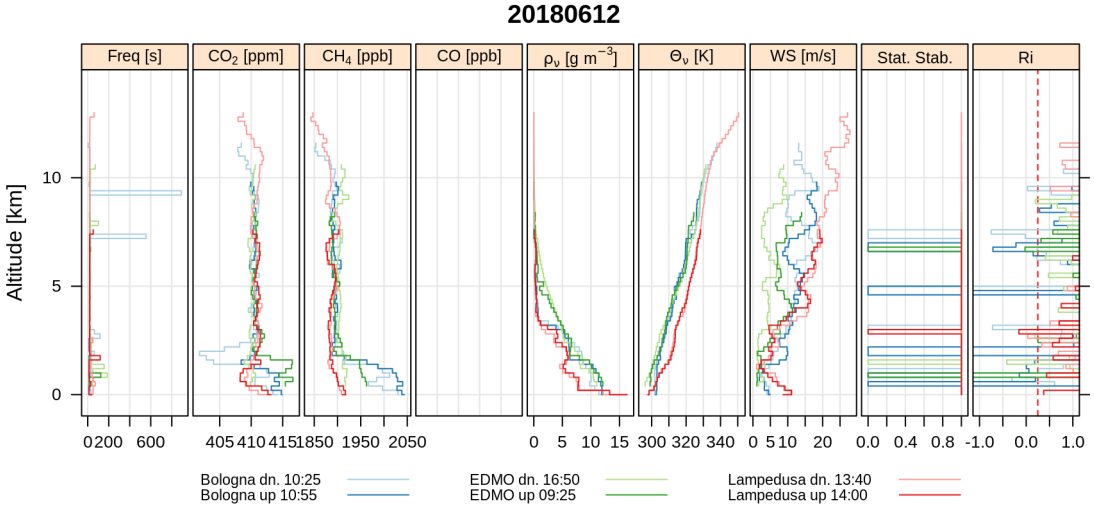

Figure S10. Detailed profile information - flight on June 12th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Locations and their abbreviations can be found in Table S1.

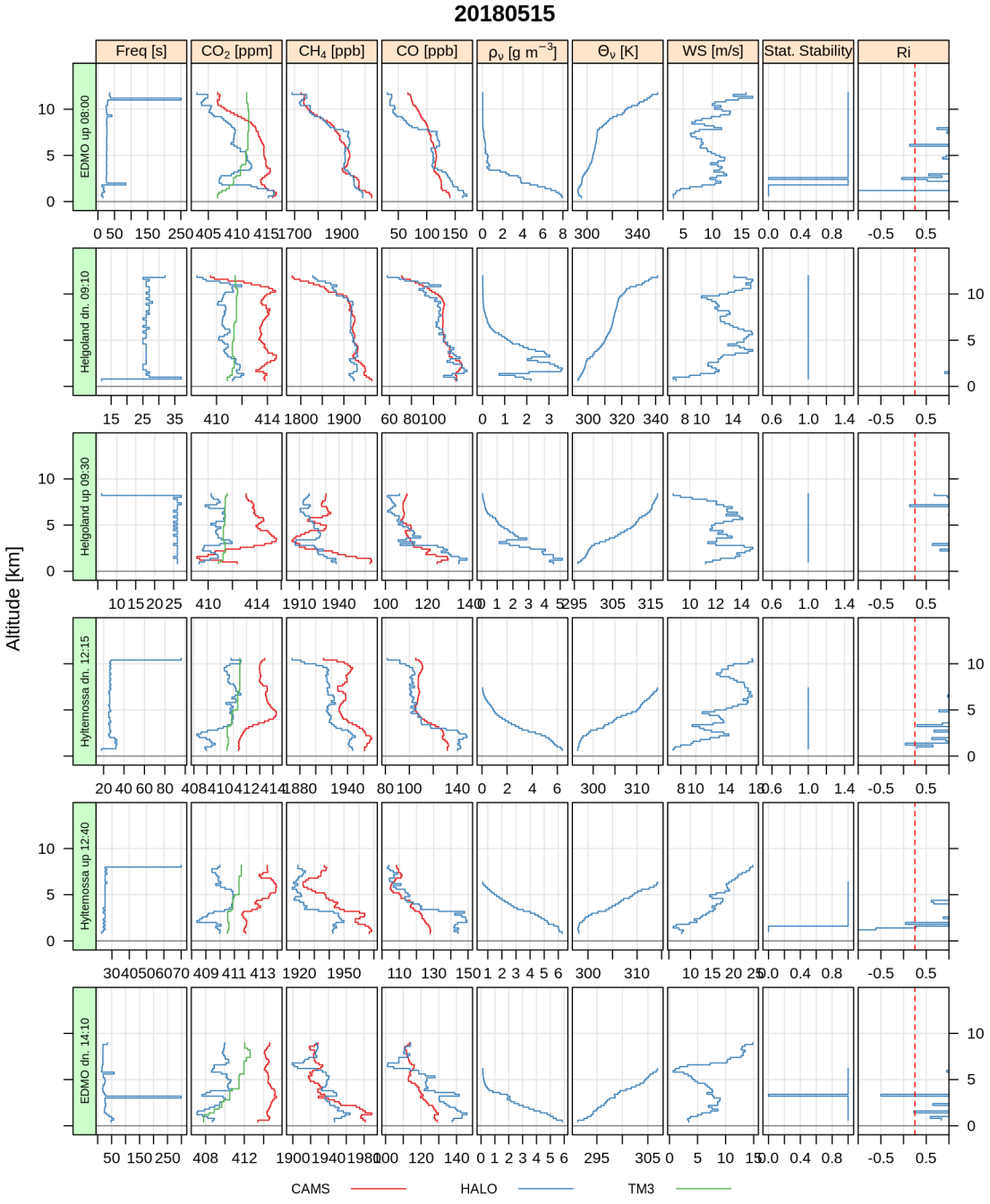

Figure S11. Detailed profile information - flight on May 15th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Modeling results from CAMS and TM3 provided for comparison. See main text for details. Locations and their abbreviations can be found in Table S1.

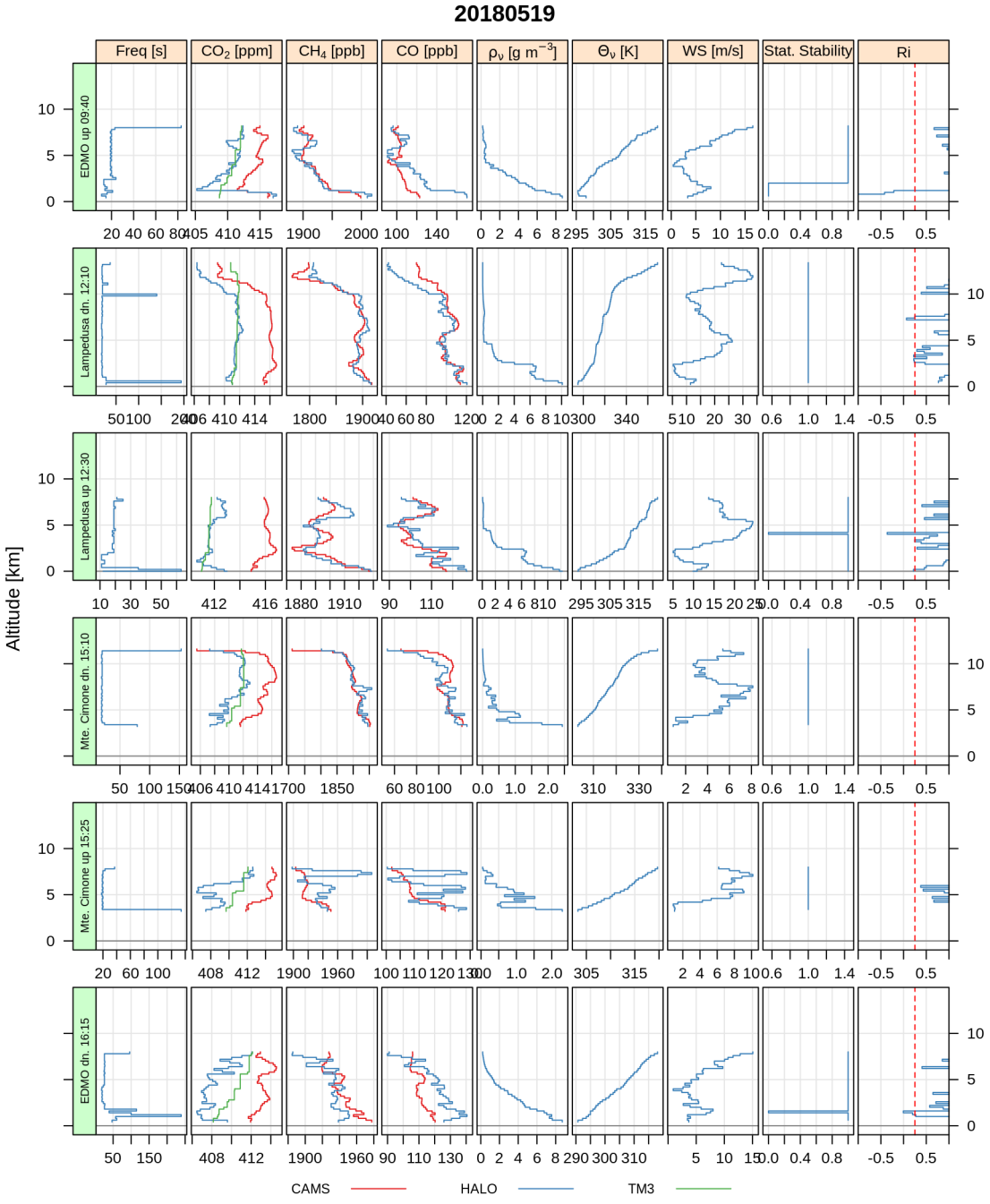

Figure S12. Detailed profile information - flight on May 19th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Modeling results from CAMS and TM3 provided for comparison. See main text for details. Locations and their abbreviations can be found in Table S1.

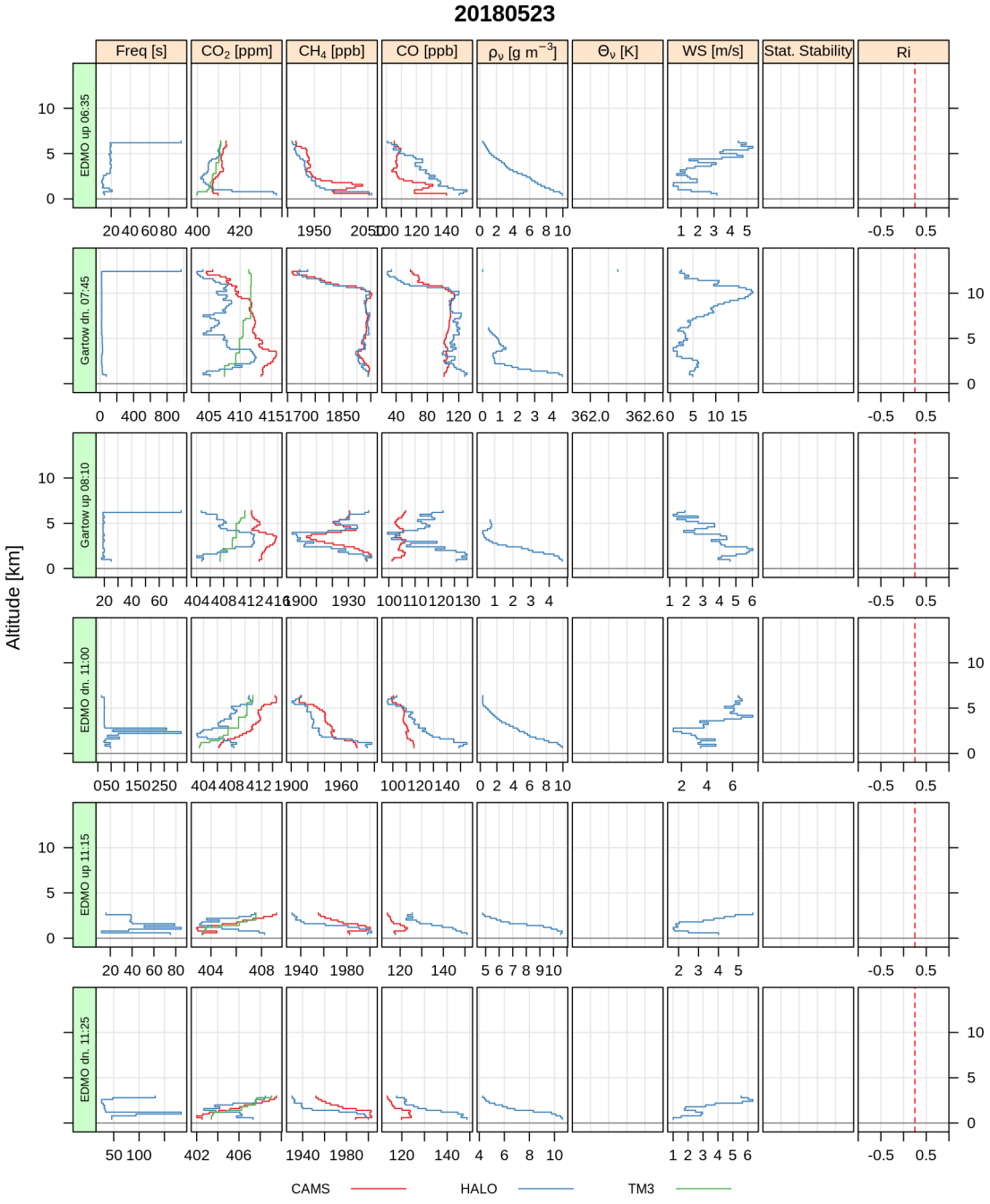

Figure S13. Detailed profile information - flight on May 23rd, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Modeling results from CAMS and TM3 provided for comparison. See main text for details. Locations and their abbreviations can be found in Table S1.

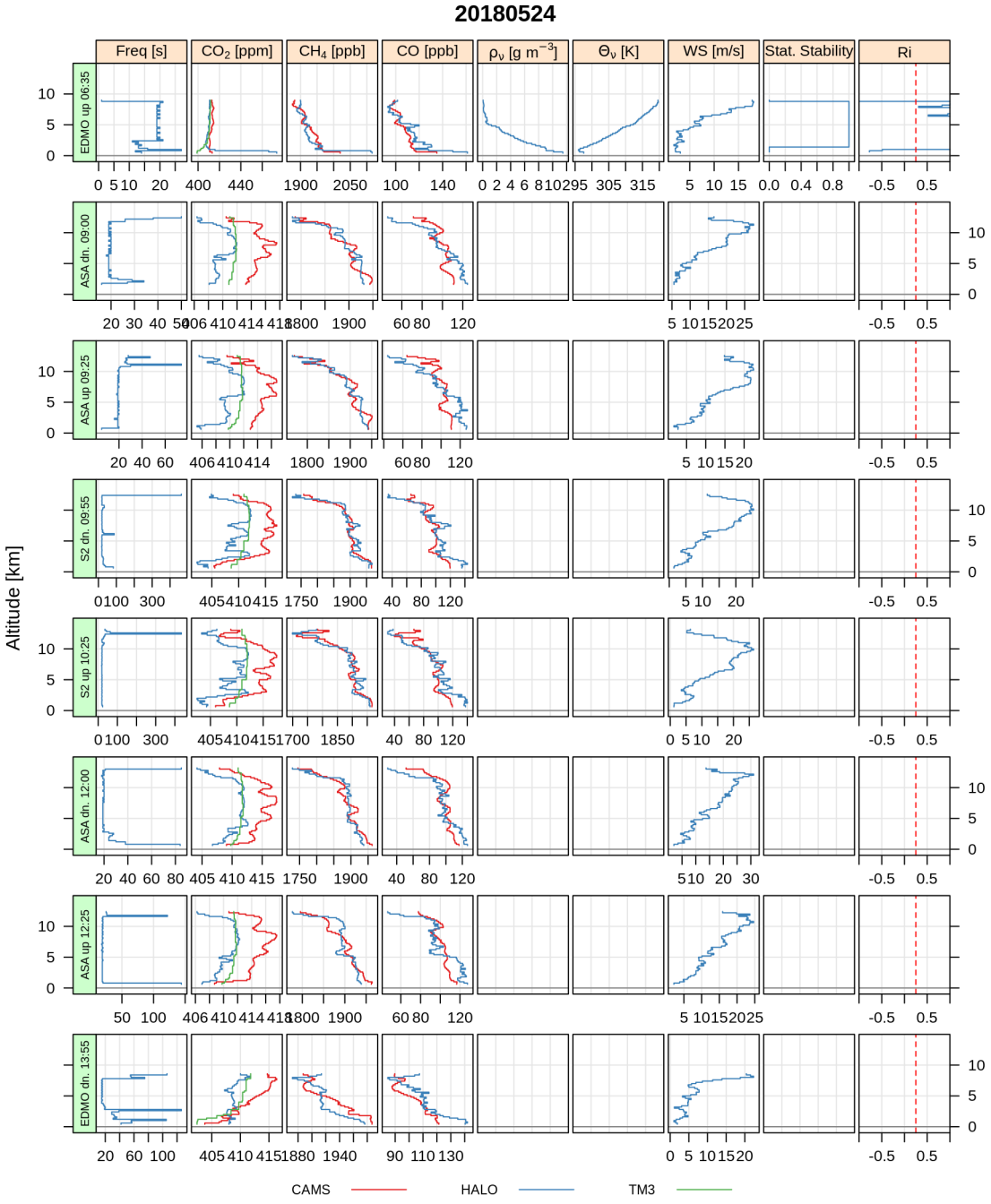

Figure S14. Detailed profile information - flight on May 24th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Modeling results from CAMS and TM3 provided for comparison. See main text for details. Locations and their abbreviations can be found in Table S1.

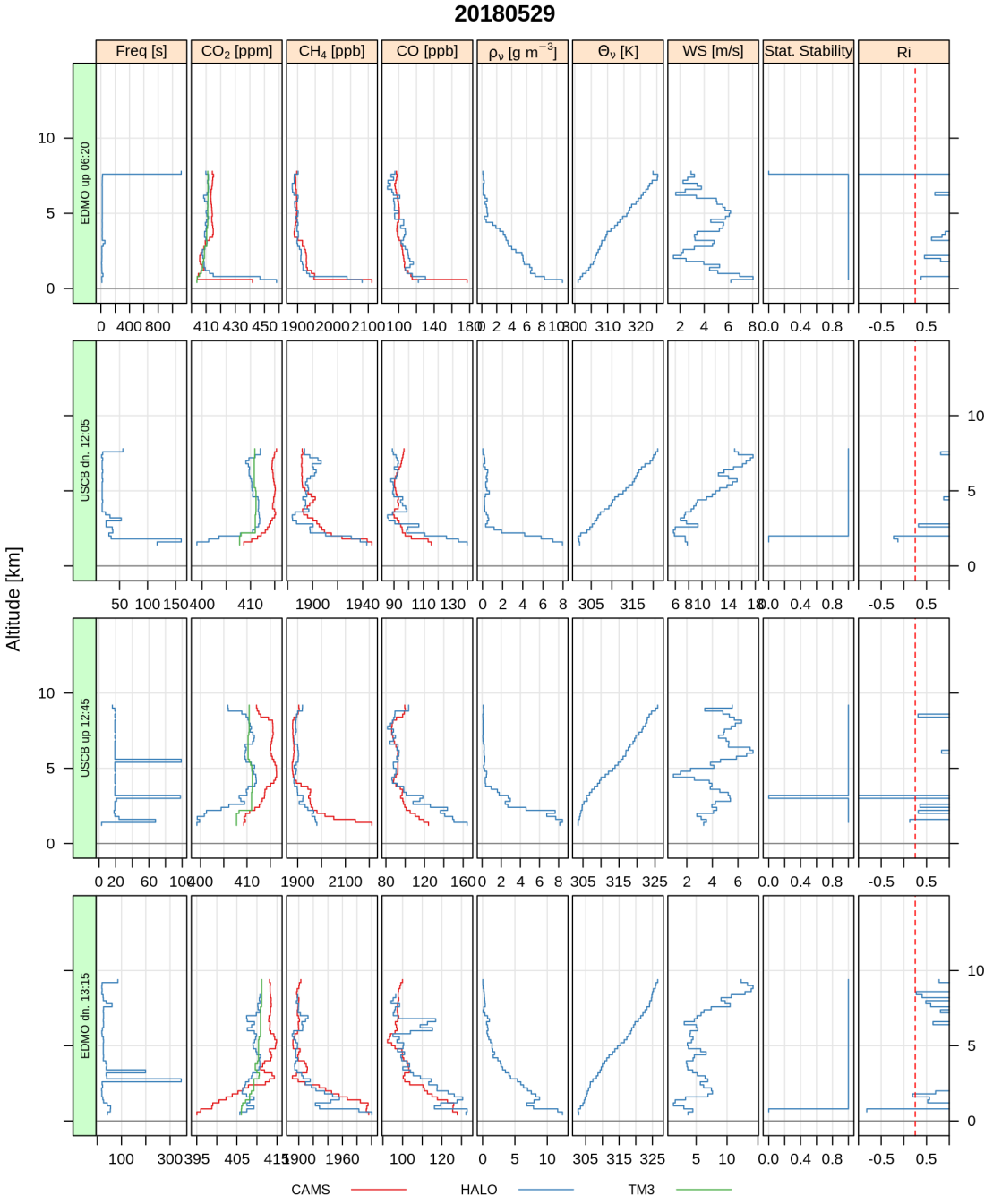

Figure S15. Detailed profile information - flight on May 29th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Modeling results from CAMS and TM3 provided for comparison. See main text for details. Locations and their abbreviations can be found in Table S1.

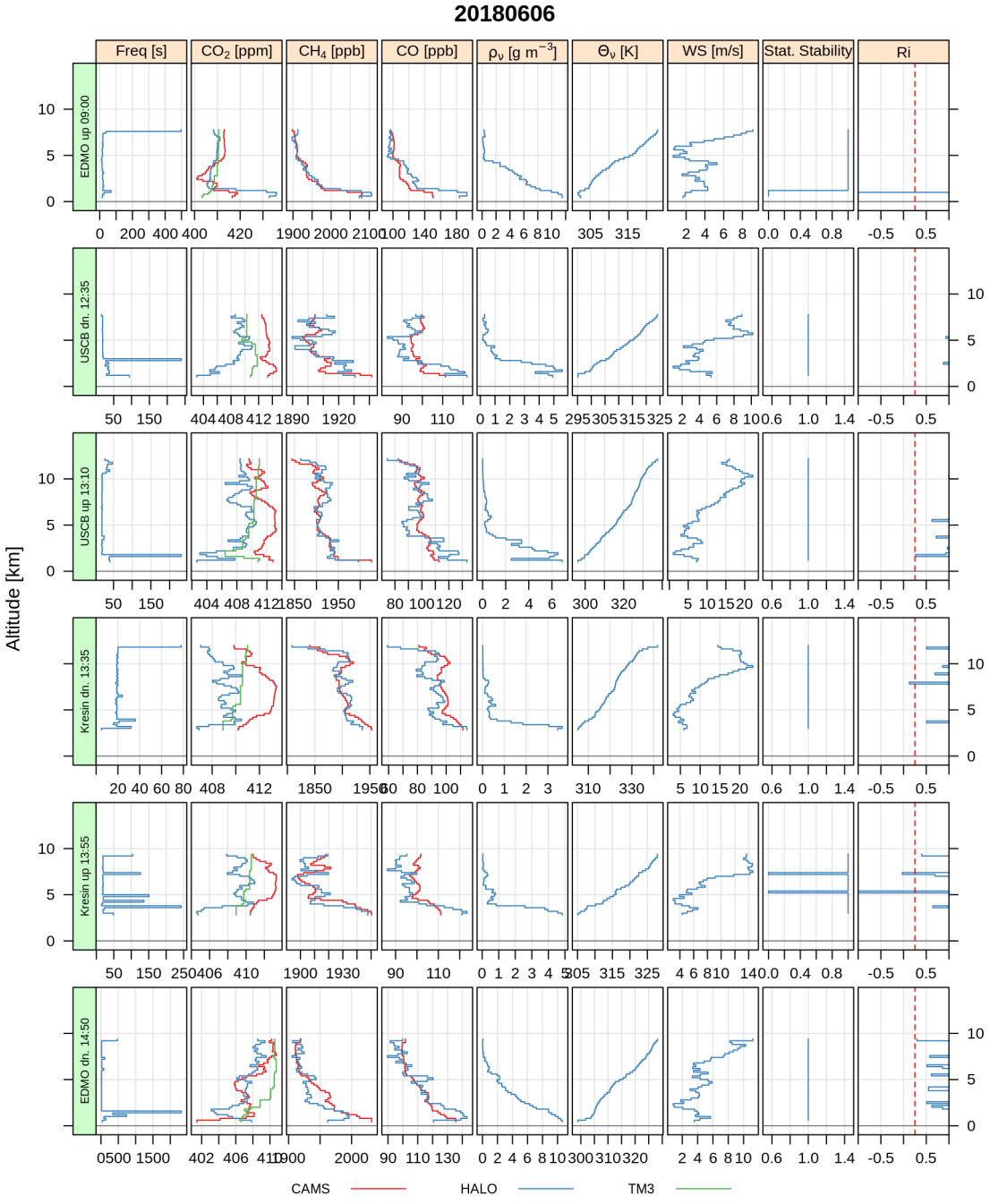

Figure S16. Detailed profile information - flight on June 6th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Modeling results from CAMS and TM3 provided for comparison. See main text for details. Locations and their abbreviations can be found in Table S1.

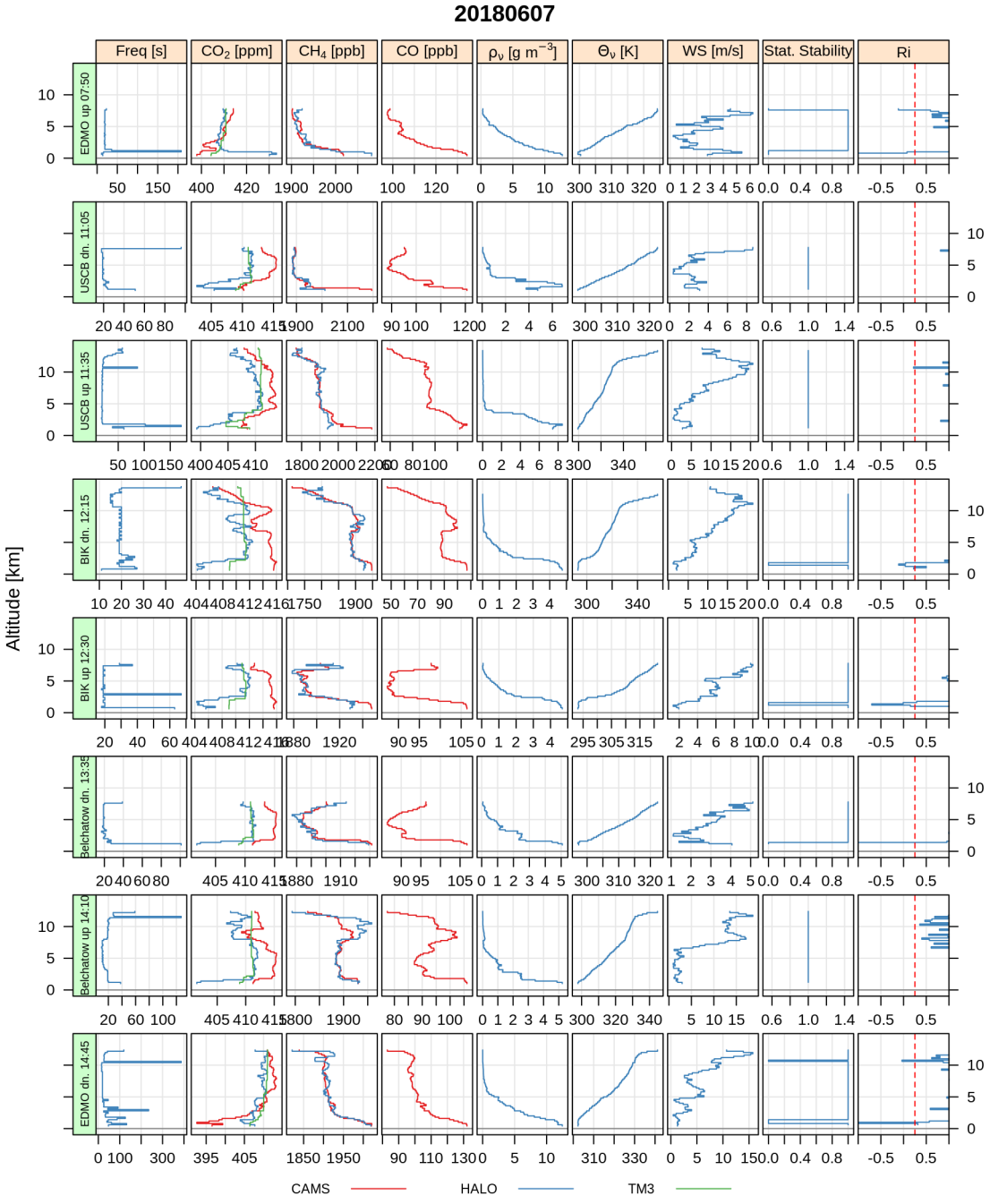

Figure S17. Detailed profile information - flight on June 7th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Modeling results from CAMS and TM3 provided for comparison. See main text for details. Locations and their abbreviations can be found in Table S1.

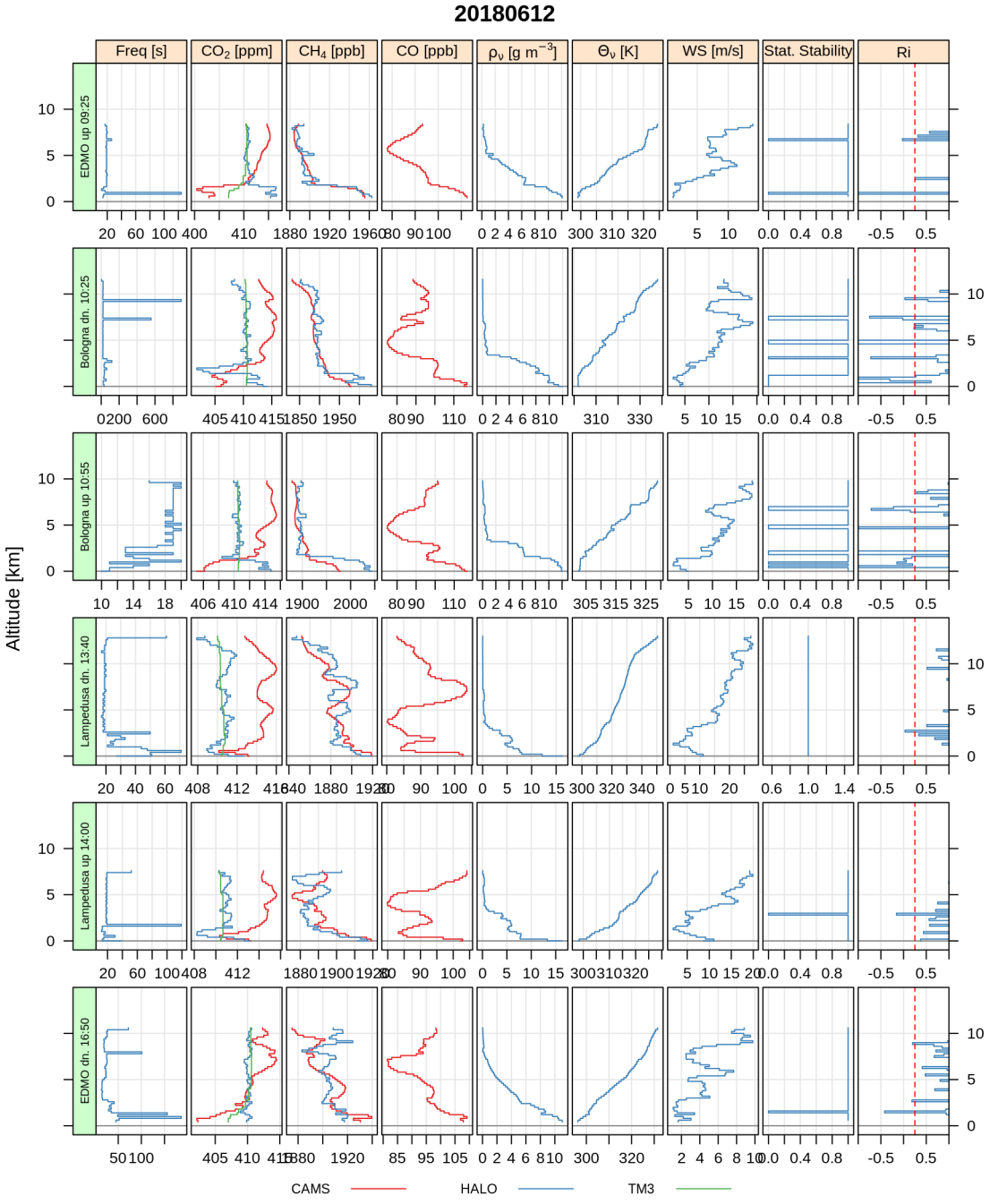

Figure S18. Detailed profile information - flight on June 12th, 2018. Data binned into layers 200 m thick layers, average per layer is plotted. WS - wind speed. Static stability (Stat. Stab.) and Richardson number (Ri) calculated according to Stull (2012). Modeling results from CAMS and TM3 provided for comparison. See main text for details. Locations and their abbreviations can be found in Table S1.

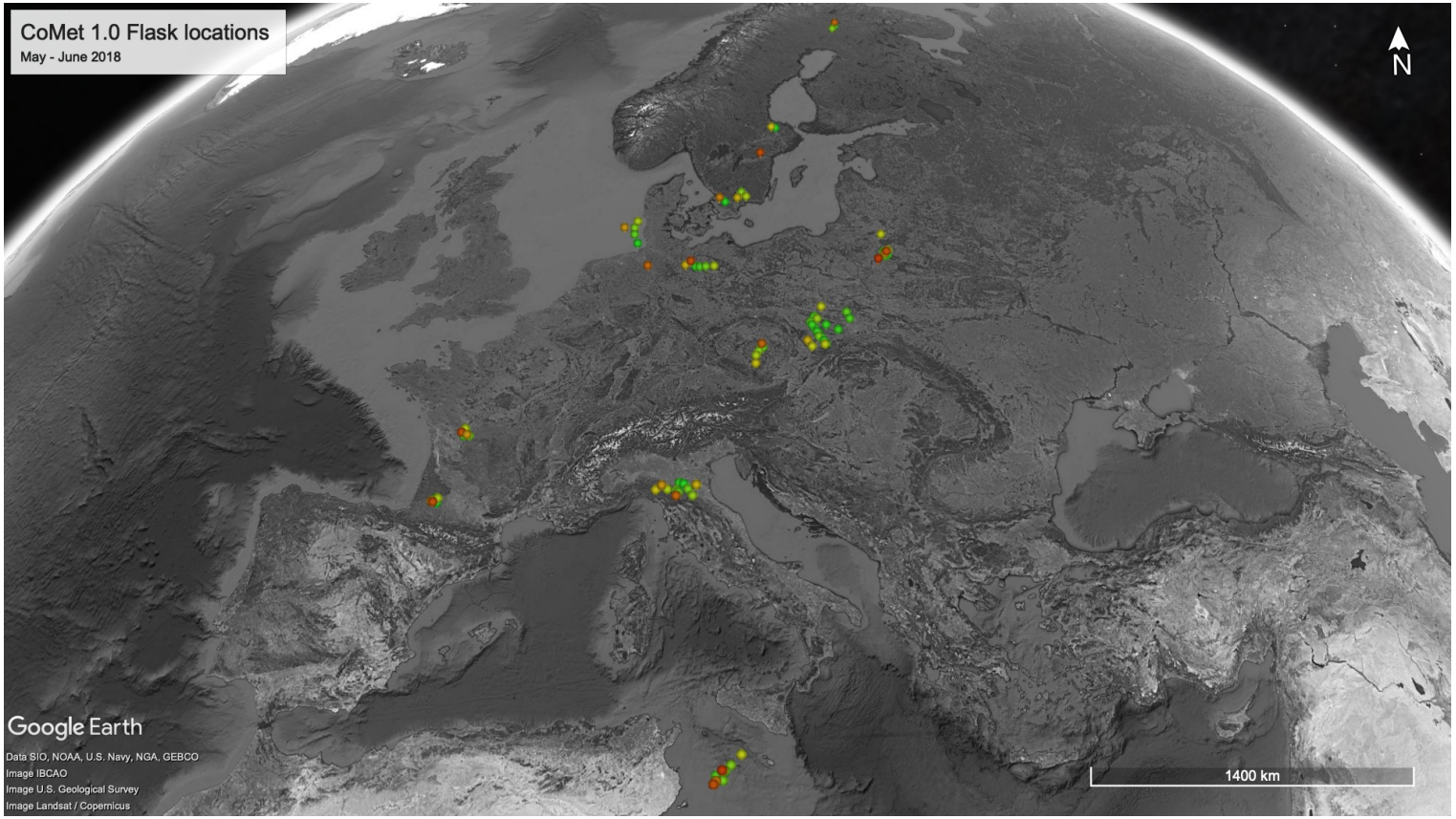

Figure S19. Overview of locations from which flasks were collected with JAS instrument aboard HALO during CoMet 1.0 campaign. Colours mark the average height from which the air was sampled, going from low (green) to high (red) altitudes. Image source: Google Earth v.7.3.2.5776, SIO, NOAA, U.S. Navy, NGA, GEBCO, IBCAO, U.S. Geological Survey, Landsat / Copernicus.

Table S1: Detailed information on vertical profiles sounded for with HALO during the CoMet campaign. Latitudes and longitudes coordinates calculated as an average throughout the profile. Number of flasks is given collectively for ascending and descending profiles.

Date	Time UTC	Dir.	Location			Altitude		In situ			Flasks	Notes
			Name	Lat	Lon	Bottom	Top	CH_4	CO	CO_2		
2018-05-15	07:50 - 08:20	up	EDMO	48.867 N	11.126 E	0.6 km	12 km	✓	✓	✓	0	Home base
	08:55 - 09:20	down	Helgoland	54.307 N	7.677 E	0.8 km	12.2 km	✓	✓	✓	6	ICOS tt.
	09:24 - 09:40	up	Helgoland	54.79 N	7.761 E	1 km	8.6 km	✓	✓	✓	"	"
	12:03 - 12:26	down	Hyltemossa	56.241 N	14.405 E	0.8 km	10.8 km	✓	✓	✓	6	ICOS tt.
	12:32 - 12:49	up	Hyltemossa	55.517 N	13.697 E	1 km	8.4 km	✓	✓	✓	"	"
	14:00 - 14:22	down	EDMO	48.934 N	12.046 E	0.6 km	9.2 km	✓	✓	✓	0	Home base
2018-05-19	09:30 - 09:44	up	EDMO	47.836 N	11.095 E	0.6 km	8.4 km	✓	✓	✓	0	Home base
	11:54 - 12:21	down	Lampedusa	35.536 N	12.78 E	0.4 km	13.6 km	✓	✓	✓	6	ICOS cs.
	12:21 - 12:34	up	Lampedusa	35.766 N	12.966 E	0 km	8.2 km	✓	✓	✓	"	"
	15:02 - 15:19	down	Mte. Cimone	44.511 N	10.402 E	3.4 km	11.8 km	✓	✓	✓	6	ICOS mtn.
	15:19 - 15:29	up	Mte. Cimone	44.351 N	10.552 E	3.4 km	8.2 km	✓	✓	✓	"	"
	16:04 - 16:28	down	EDMO	47.565 N	11.274 E	0.6 km	8.2 km	✓	✓	✓	0	Home base
2018-05-23	06:29 - 06:39	up	EDMO	48.156 N	11.284 E	0.6 km	6.6 km	✓	✓	✓	0	Home base
	07:25 - 08:03	down	Gartow	53.03 N	11.245 E	1 km	12.8 km	✓	✓	✓	6	ICOS tt.
	08:03 - 08:13	up	Gartow	52.986 N	12.1 E	1 km	6.6 km	✓	✓	✓	"	"
	10:48 - 11:11	down	EDMO	48.844 N	12.09 E	0.8 km	6.6 km	✓	✓	✓	6	Home base
	11:11 - 11:20	up	EDMO	47.905 N	11.277 E	0.6 km	3 km	✓	✓	✓	"	"
	11:20 - 11:33	down	EDMO	47.872 N	11.287 E	0.6 km	3.2 km	✓	✓	✓	"	"
2018-05-24	06:27 - 06:40	up	EDMO	47.784 N	10.912 E	0.6 km	9.2 km	✓	✓	✓	0	Home base
	08:50 - 09:09	down	ASA	43.711 N	-0.249 E	1.8 km	12.8 km	✓	✓	✓	6	ICOS tt.
	09:14 - 09:35	up	ASA	44.005 N	-0.182 E	0.8 km	12.8 km	✓	✓	✓	"	"
	09:38 - 10:09	down	S2	46.137 N	0.546 E	0.8 km	12.8 km	✓	✓	✓	6	ICOS tt., Aircore
	10:09 - 10:39	up	S2	46.476 N	0.753 E	0.8 km	13.4 km	✓	✓	✓	"	"
	11:49 - 12:13	down	ASA	43.694 N	-0.225 E	0.8 km	13.4 km	✓	✓	✓	0	ICOS tt.
	12:13 - 12:36	up	ASA	43.803 N	0.907 E	0.8 km	12.6 km	✓	✓	✓	"	"
	13:45 - 14:06	down	EDMO	47.614 N	11.308 E	0.6 km	8.8 km	✓	✓	✓	0	Home base
2018-05-29	06:50 - 07:20	up	EDMO	48.186 N	11.641 E	0.6 km	8 km	✓	✓	✓	0	Home base
	12:00 - 12:17	down	USCB	50.965 N	19.728 E	1.6 km	8 km	✓	✓	✓	6	Plume hunting
	12:37 - 12:53	up	USCB	50.333 N	17.502 E	1.4 km	9.4 km	✓	✓	✓	"	"
	13:05 - 13:45	down	EDMO	48.721 N	12.943 E	0.6 km	9.6 km	✓	✓	✓	0	Home base

(continued)

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Date	Time UTC	Dir.	Location			Altitude		In situ			Flasks	Notes
			Name	Lat	Lon	Bottom	Top	CH_4	CO	CO_2		
2018-06-06	08:48 - 09:08	up	EDMO	48.208 N	11.902 E	0.6 km	8 km	✓	✓	✓	0	Home base
	12:27 - 12:45	down	USCB	49.896 N	18.172 E	1.2 km	8 km	✓	✓	✓	6	Plume hunting
	12:53 - 13:18	up	USCB	49.475 N	17.648 E	1.2 km	12.4 km	✓	✓	✓	"	"
	13:26 - 13:42	down	Kresin	49.552 N	15.112 E	3 km	12.2 km	✓	✓	✓	6	ICOS tt.
	13:44 - 14:06	up	Kresin	49.152 N	14.865 E	3 km	9.6 km	✓	✓	✓	"	"
	14:09 - 15:35	down	EDMO	48.019 N	12.102 E	0.6 km	9.6 km	✓	✓	✓	0	Home base
2018-06-07	07:43 - 07:58	up	EDMO	48.21 N	12.145 E	0.6 km	8 km	✓	X	✓	0	Home base
	10:58 - 11:11	down	USCB	50.503 N	18.115 E	1.2 km	8 km	✓	X	✓	6	Plume hunting
	11:18 - 11:49	up	USCB	50.362 N	19.415 E	1.2 km	14 km	✓	X	✓	"	"
	12:03 - 12:24	down	Białystok	53.227 N	23.093 E	0.8 km	14 km	✓	X	✓	6	MPI tt.
	12:24 - 12:38	up	Białystok	53.131 N	22.854 E	0.8 km	8 km	✓	X	✓	"	"
	13:30 - 13:43	down	Bełchatów	50.886 N	19.138 E	1.2 km	8 km	✓	X	✓	0	Power plant
	13:58 - 14:17	up	Bełchatów	50.419 N	18.384 E	1.2 km	12.6 km	✓	X	✓	"	"
	14:28 - 15:10	down	EDMO	48.281 N	13.075 E	0.6 km	12.6 km	✓	X	✓	0	Home base
2018-06-12	09:16 - 09:30	up	EDMO	47.836 N	10.892 E	0.6 km	8.6 km	✓	X	✓	0	Home base
	09:59 - 10:47	down	Bologna	44.453 N	11.484 E	0 km	11.8 km	✓	X	✓	6	airport m.a.
	10:47 - 11:02	up	Bologna	44.154 N	11.51 E	0 km	10 km	✓	X	✓	"	"
	13:27 - 13:53	down	Lampedusa	35.467 N	12.628 E	0 km	13.2 km	✓	X	✓	6	ICOS cs.
	13:53 - 14:07	up	Lampedusa	35.812 N	13.06 E	0 km	7.8 km	✓	X	✓	"	"
	16:39 - 17:06	down	EDMO	47.373 N	11.324 E	0.6 km	10.8 km	✓	X	✓	0	Home base

Abbrevations: ICOS - Integrated Carbon Observation System; MPI - Max Planck Institute; cs. - coastal site; mtn. - mountain station; tt. - tall tower; EDMO - Oberpfaffenhofen, Germany (HALO home base); ASA - Aire-sur-l'Adour; S2 - Aircore release site; USCB - Upper Silesian Coal Basin

References

Stull, R. B.: An introduction to boundary layer meteorology, vol. 13, Springer Science & Business Media, 2012.