



Supplement of

Tropospheric observations of CFC-114 and CFC-114a with a focus on long-term trends and emissions

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This supplement contains additional details on firn air data and modelling in Figure S1 as well as numerical results from the Cape Grim air archive in Table S1, from the CARIBIC flight on 10/02/2015 in Table S2, from the 2015 Taiwan campaign in Table S3, from the two firn campaigns in Table S4, and from the emission model in Table S5.

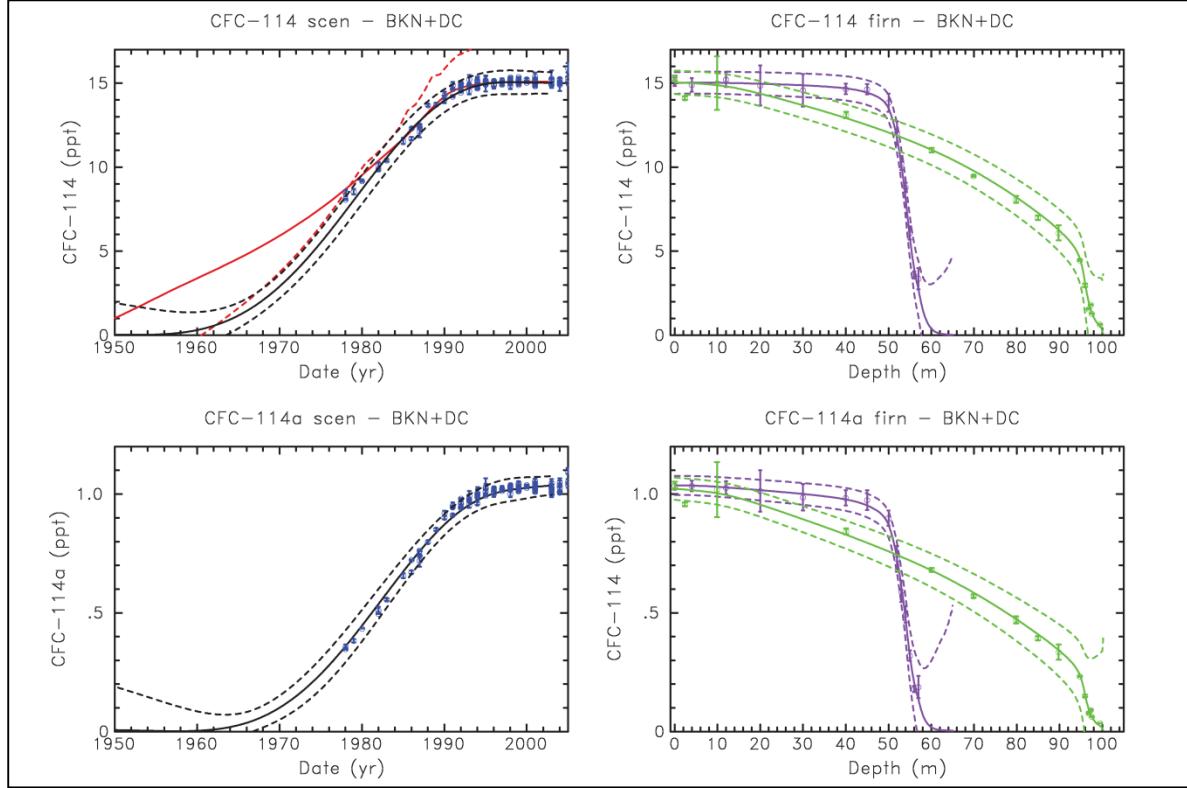


Figure S1. Right: Mixing ratios of CFC-114 and CFC-114a measured in firn air collected at the Berkner (BKN, purple dots) and Dome C (DC, green dots) Antarctic sites and the resulting modelled firn profiles (green and purple lines). Left: Atmospheric time series corresponding to the modelled firn profiles (black lines, scen: scenario) in comparison to Cape Grim air archive data (blue dots). For CFC-114, the atmospheric trends from Sturrock et al. (2002) (firn air based, red dashed line) and Martinerie et al. (2009) (AFEAS emissions based, red continuous line) are also shown.

Table S1. Numerical results from the Cape Grim air archive.

Collection			Mixing ratio [ppt]			
Day	Month	Year	CFC-114	uncertainty	CFC-114a	uncertainty
26	4	1978	7.88	0.05	0.335	0.003
7	7	1978	8.08	0.04	0.348	0.003
30	10	1978	8.47	0.19	0.357	0.010
6	2	1979	8.54	0.16	0.383	0.007
26	8	1980	9.19	0.05	0.435	0.000
23	2	1982	9.86	0.08	0.498	0.002
26	5	1982	10.10	0.20	0.510	0.015
1	8	1983	10.41	0.08	0.556	0.005

8	8	1985	11.55	0.18	0.656	0.010
6	2	1986	11.70	0.09	0.671	0.007
12	11	1986	12.32	0.06	0.723	0.001
8	1	1987	12.48	0.10	0.763	0.003
9	1	1987	12.14	0.37	0.719	0.025
28	5	1987	12.35	0.02	0.741	0.004
21	6	1988	13.69	0.08	0.799	0.007
20	4	1989	13.73	0.10	0.851	0.006
25	5	1990	14.03	0.23	0.901	0.007
18	12	1990	14.28	0.22	0.919	0.010
13	3	1991	14.21	0.04	0.912	0.005
19	6	1991	14.66	0.05	0.945	0.003
4	7	1991	14.89	0.18	0.952	0.014
16	1	1992	14.45	0.00	0.943	0.003
12	2	1992	14.56	0.08	0.945	0.005
8	4	1992	14.85	0.05	0.965	0.010
2	7	1992	14.91	0.06	0.963	0.006
23	9	1992	14.79	0.20	0.961	0.013
24	9	1992	15.00	0.05	0.981	0.002
7	6	1993	14.91	0.07	0.979	0.004
29	6	1993	14.88	0.05	0.973	0.002
11	8	1993	14.54	0.20	0.955	0.012
20	9	1993	15.23	0.25	0.993	0.020
11	2	1994	15.17	0.15	1.000	0.004
15	3	1994	14.70	0.07	0.961	0.006
27	4	1994	15.04	0.00	0.987	0.004
1	6	1994	15.11	0.28	1.000	0.020
9	6	1994	15.09	0.12	0.992	0.009
5	10	1994	14.85	0.12	0.992	0.012
7	12	1994	15.04	0.35	1.006	0.024
8	2	1995	15.01	0.15	1.010	0.013
13	3	1995	15.18	0.07	1.010	0.008
12	4	1995	14.92	0.04	0.994	0.004
13	6	1995	14.98	0.47	1.029	0.037
23	11	1995	15.14	0.26	1.024	0.020
9	2	1996	14.97	0.18	1.011	0.013
4	4	1996	14.86	0.18	1.001	0.012
28	5	1996	14.96	0.28	1.008	0.020
21	8	1996	15.14	0.01	1.021	0.006
20	11	1996	15.15	0.08	1.025	0.008

7	1	1997	15.14	0.14	1.022	0.012
26	2	1997	14.92	0.07	1.014	0.003
30	5	1997	15.16	0.01	1.020	0.002
15	7	1997	15.05	0.05	1.022	0.008
5	3	1998	15.13	0.05	1.032	0.005
15	4	1998	15.02	0.02	1.021	0.004
20	5	1998	15.10	0.22	1.026	0.016
16	6	1998	14.86	0.20	1.002	0.014
1	7	1998	15.24	0.01	1.030	0.003
29	8	1998	15.04	0.05	1.037	0.002
24	9	1998	15.16	0.01	1.020	0.004
15	10	1998	15.20	0.09	1.032	0.007
27	11	1998	15.35	0.04	1.039	0.003
17	2	1999	15.20	0.05	1.027	0.004
4	3	1999	14.97	0.02	1.012	0.002
13	4	1999	14.98	0.03	1.013	0.004
13	5	1999	15.18	0.13	1.045	0.010
21	6	1999	15.24	0.13	1.037	0.000
16	8	1999	15.01	0.11	1.014	0.002
24	9	1999	15.37	0.01	1.045	0.007
18	10	1999	15.09	0.01	1.023	0.001
15	11	1999	14.91	0.00	1.019	0.009
17	12	1999	15.09	0.04	1.025	0.003
20	1	2000	15.24	0.06	1.038	0.003
23	2	2000	15.04	0.04	1.027	0.004
27	4	2001	15.40	0.23	1.055	0.009
15	6	2001	14.83	0.01	1.010	0.005
15	7	2001	15.21	0.04	1.042	0.011
8	8	2001	15.25	0.01	1.042	0.005
4	9	2001	14.97	0.21	1.021	0.012
18	10	2001	15.08	0.20	1.026	0.012
13	11	2001	15.11	0.18	1.029	0.006
11	12	2001	15.14	0.11	1.028	0.005
21	1	2003	15.27	0.01	1.036	0.007
13	3	2003	14.98	0.08	1.011	0.003
15	4	2003	14.91	0.19	1.027	0.012
21	5	2003	15.27	0.49	1.025	0.038
18	9	2003	15.20	0.11	1.045	0.008
8	10	2003	14.92	0.10	1.020	0.001
13	12	2003	15.16	0.14	1.000	0.003

15	1	2004	15.09	0.00	1.037	0.007
19	5	2004	15.02	0.08	1.007	0.002
30	6	2004	14.92	0.38	1.029	0.029
6	8	2004	15.26	0.08	1.050	0.016
1	9	2004	14.98	0.06	1.011	0.003
17	1	2005	15.92	0.27	1.098	0.012
6	4	2005	15.35	0.26	1.052	0.012
5	12	2005	15.13	0.24	1.038	0.012
10	2	2006	14.77	0.24	1.040	0.012
20	2	2006	14.78	0.47	1.031	0.036
23	8	2006	14.89	0.25	1.020	0.012
11	12	2006	14.94	0.47	1.029	0.036
20	6	2007	14.83	0.25	1.026	0.012
20	9	2007	14.99	0.26	1.027	0.011
13	6	2008	14.88	0.47	1.036	0.036
6	3	2009	15.21	0.23	1.036	0.011
10	12	2010	15.19	0.49	1.062	0.038
26	5	2011	14.91	0.11	1.028	0.009
21	6	2011	15.34	0.17	1.057	0.016
27	4	2012	15.32	0.51	1.056	0.039
4	12	2012	15.29	0.31	1.067	0.017
14	3	2013	14.71	0.28	1.032	0.014
5	7	2013	14.71	0.28	1.030	0.014
14	10	2013	14.70	0.34	1.038	0.017
21	2	2014	14.74	0.34	1.035	0.017
5	5	2014	14.89	0.27	1.021	0.013
17	7	2014	14.84	0.27	1.028	0.013
11	9	2014	14.71	0.27	1.026	0.013
10	11	2014	14.76	0.27	1.018	0.013

Table S2. Numerical results from the CARIBIC flight on 10/02/2015.

Latitude [°] (negative: SH)	Mixing ratios [ppt]			
	CFC-114	uncertainty	CFC-114a	
45.94	14.82	0.16	1.058	0.013
42.78	14.70	0.16	1.049	0.013
36.85	15.10	0.16	1.068	0.013
31.27	14.92	0.16	1.066	0.013
25.7	14.74	0.16	1.056	0.013
20.14	14.79	0.16	1.068	0.013
14.45	14.94	0.16	1.059	0.013
8.56	14.95	0.16	1.060	0.013

2.71		14.75	0.16	1.066	0.013
-3.09		14.97	0.16	1.059	0.013
-8.82		14.87	0.21	1.043	0.013
-14.49		14.76	0.21	1.051	0.013
-20.08		14.71	0.21	1.049	0.013
-25.81		14.78	0.21	1.055	0.013
-31.76		14.66	0.21	1.044	0.013

Table S3. Numerical results from the 2015 Taiwan campaign.

Collection			Mixing ratio [ppt]			
Day	Month	Year	CFC-114	uncertainty	CFC-114a	uncertainty
12	3	2015	14.73	0.19	1.062	0.002
17	3	2015	14.98	0.19	1.058	0.002
18	3	2015	14.91	0.19	1.067	0.002
19	3	2015	14.80	0.19	1.080	0.002
20	3	2015	14.75	0.19	1.064	0.004
21	3	2015	14.84	0.20	1.059	0.004
22	3	2015	15.02	0.20	1.210	0.004
23	3	2015	14.93	0.20	1.200	0.004
24	3	2015	14.87	0.20	1.101	0.004
25	3	2015	14.97	0.20	1.097	0.004
26	3	2015	15.15	0.20	1.094	0.004
27	3	2015	14.85	0.18	1.068	0.002
28	3	2015	14.81	0.18	1.146	0.002
29	3	2015	14.77	0.18	1.143	0.002
30	3	2015	14.87	0.18	1.096	0.002
31	3	2015	14.80	0.18	1.062	0.002
22	4	2015	15.01	0.18	1.106	0.002
23	4	2015	15.05	0.18	1.103	0.002
23	4	2015	15.00	0.40	1.111	0.008
24	4	2015	14.98	0.40	1.100	0.008
24	4	2015	15.21	0.40	1.096	0.008
25	4	2015	14.84	0.39	1.093	0.008
25	4	2015	14.85	0.40	1.073	0.008

Table S4. Numerical results from the two firn campaigns.

Depth [m]	Mixing ratios [ppt]			
	CFC-114	uncertainty	CFC-114a	uncertainty
Dome C				
0	15.46	0.11	1.050	0.016

0	15.15	0.11	1.023	0.008
2.38	14.11	1.60	0.957	0.116
9.9	15.00	0.20	1.019	0.013
40.1	13.10	0.13	0.843	0.008
60.1	11.01	0.05	0.681	0.007
69.9	9.48	0.21	0.570	0.015
79.85	8.08	0.13	0.470	0.009
84.95	6.99	0.44	0.393	0.032
89.8	6.09	0.04	0.335	0.002
94.75	4.47	0.04	0.233	0.001
95.96	3.07	0.03	0.155	0.004
95.96	2.88	0.02	0.145	0.003
96.83	1.57	0.01	0.078	0.001
97.3	1.82	0.02	0.091	0.000
97.55	1.29	0.01	0.063	0.001
99.45	0.65	0.00	0.036	0.000
Berkner Island				
0	15.07	0.25	1.040	0.017
0	15.09	0.40	1.029	0.022
4	14.99	0.44	1.031	0.028
4	14.80	1.19	1.039	0.087
12	14.94	0.96	1.003	0.056
12	15.50	0.31	1.051	0.032
20	15.14	0.30	1.027	0.042
20	14.38	0.55	0.983	0.032
20	15.06	0.71	1.028	0.054
30	14.58	0.41	0.987	0.045
40	14.59	0.27	0.960	0.034
40	14.74	0.40	1.009	0.022
45	14.64	0.22	0.975	0.013
50	13.90	0.63	0.906	0.047
50	13.78	0.56	0.892	0.041
52	12.17	0.19	0.734	0.010
52	11.87	0.25	0.719	0.023
53	10.25	0.26	0.591	0.009
54	8.81	0.08	0.493	0.007
55	6.09	0.14	0.320	0.020
56	3.59	0.13	0.179	0.009
57	3.42	0.07	0.192	0.008
57	3.30	0.10	0.185	0.006

57	3.40	0.03	0.188	0.005
61	1.45	0.01	0.072	0.002
63	0.60	0.02	0.037	0.003
63	0.60	0.03	0.034	0.003
63	0.61	0.03	0.035	0.002

Table S5. Numerical results from the emission model based on Cape Grim air archive data.

Year	Emissions [Gg]			
	CFC-114	Uncertainty range	CFC-114a	Uncertainty range
1978	13.58	11.26-15.94	1.330	1.23-1.44
1979	15.11	12.54-17.73	1.346	1.24-1.45
1980	15.58	12.92-18.28	1.358	1.25-1.47
1981	15.23	12.63-17.87	1.369	1.26-1.48
1982	14.57	12.09-17.10	1.389	1.28-1.50
1983	14.98	12.42-17.57	1.441	1.33-1.56
1984	16.34	13.55-19.17	1.533	1.40-1.65
1985	17.65	14.64-20.71	1.606	1.46-1.73
1986	18.25	15.14-21.41	1.716	1.56-1.85
1987	18.13	15.04-21.28	1.785	1.63-1.93
1988	17.60	14.60-20.65	1.772	1.62-1.91
1989	15.12	12.54-17.74	1.445	1.31-1.56
1990	13.23	10.97-15.53	1.238	1.12-1.37
1991	10.40	7.97-13.00	0.948	0.86-1.06
1992	6.50	4.98-8.13	0.646	0.58-0.74
1993	4.23	3.12-5.42	0.554	0.50-0.64
1994	2.39	1.44-3.46	0.520	0.45-0.62
1995	2.10	1.27-3.04	0.485	0.42-0.58
1996	2.00	1.20-2.89	0.451	0.38-0.55
1997	1.94	1.17-2.80	0.423	0.35-0.53
1998	1.93	1.16-2.79	0.386	0.31-0.49
1999	1.83	1.02-2.75	0.356	0.27-0.45
2000	1.82	1.02-2.74	0.334	0.25-0.43
2001	1.81	1.01-2.72	0.325	0.24-0.42
2002	1.80	1.01-2.71	0.323	0.24-0.42
2003	1.80	1.00-2.70	0.323	0.24-0.42
2004	1.80	1.00-2.70	0.323	0.24-0.42
2005	1.80	1.00-2.70	0.317	0.23-0.41
2006	1.80	1.00-2.70	0.307	0.22-0.40
2007	1.80	1.00-2.70	0.297	0.22-0.39
2008	1.80	1.00-2.70	0.287	0.21-0.37

2009	1.80	1.00-2.70	0.277	0.20-0.36
2010	1.80	1.00-2.70	0.267	0.19-0.35
2011	1.80	1.00-2.70	0.257	0.19-0.34
2012	1.80	1.00-2.70	0.248	0.18-0.32
2013	1.80	1.00-2.70	0.248	0.18-0.32
2014	1.80	1.00-2.70	0.248	0.18-0.32