Supplement of Atmos. Chem. Phys., 22, 2447–2466, 2022
https://doi.org/10.5194/acp-22-2447-2022-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

Swiss halocarbon emissions for 2019 to 2020 assessed from regional atmospheric observations

Dominique Rust et al.

Correspondence to: Stefan Reimann (stefan.reimann@empa.ch)

S1 Overview of the measured halocarbons

Table S1: Measured halocarbons with chemical formula, main sources, atmospheric lifetimes, ozone-depletion potentials (ODP) and global warming potentials (GWP) in a 100-year time frame.

substance	chemical formula	main sources	atmospheric lifetime[a)] (yr, unless indicated otherwise)	ODP[a)]	GWP (100 yr)[a)]
CFC-11	CCl_3F	foam blowing agent, propellant	52	1	5160
CFC-12	CCl_2F_2	cooling agent	102	0.73 to 0.81	10300
CFC-13	$CClF_3$	cooling agent	640	1	13900
CFC-115	$CClF_2CF_3$	cooling agent	540	0.26	7310
H-1211	$CBrClF_2$	fire extinction	16	3	1750
H-2402	$CBrF_2CBrF_2$	fire extinction	28	15.7	2030
HCFC-22	CHF_2Cl	cooling agent, foam blowing	11.9	0.024 to 0.034	1780
HCFC-141b	CH_3CCl_2F	foam blowing agent, solvent	9.4	0.069 to 0.102	800
HCFC-142b	CH_3CClF_2	foam blowing agent	18	0.023 to 0.057	2070
HCFC-124	$CHClFCF_3$	cooling agent	5.9	0.022	530
HFC-134a	CH_2FCF_3	cooling and foam blowing agent	14	0	1360
HFC-125	CHF_2CF_3	cooling agent	30	0	3450
HFC-32	CH_2F_2	cooling agent	5.4	0	705
HFC-152a	CH_3CHF_2	foam blowing agent	1.6	0	148
HFC-245fa	$CHF_2CH_2CF_3$	foam blowing agent	7.9	0	880
HFC-365mfc	$CH_3CF_2CH_2CF_3$	foam blowing agent, solvent	8.9	0	810
HFC-23	CHF_3	byproduct of HCFC-22 production	228	0	12690
HFC-227ea	CF_3CHFCF_3	fire extinction, propellant, foam blowing agent	36	0	3140
HFC-236fa	$CF_3CH_2CF_3$	fire extinction, cooling agent	213	0	7680
HFC-4310mee	$CF_3CHFCHFCF_2CF_3$	cleaning agent in electronic industry	17	0	1470
PFC-116	C_2F_6	semiconductor industry, aluminum industry	10000	0	11100
PFC-318	$c\text{-}C_4F_8$	semiconductor industry	3200	0	9540
PFC-14	CF_4	geogenic, aluminum industry	50000	0	6630
SF_6	SF_6	electrical insulation, magnesium/aluminum industry	3200	0	23500
NF_3	NF_3	electronics industry	569	0	15750

HFO-1234yf	$CF_3CF=CH_2$	cooling agent	12 days	0	< 1
HFO-1234ze(E)	(E)-$CF_3CH=CHF$	cooling agent and foam blowing agent	19 days	0	< 1
HCFO-1233zd(E)	(E)-$CF_3CH=CHCl$	cooling and foam blowing agent	42.5 days	< 0.0004	3.7

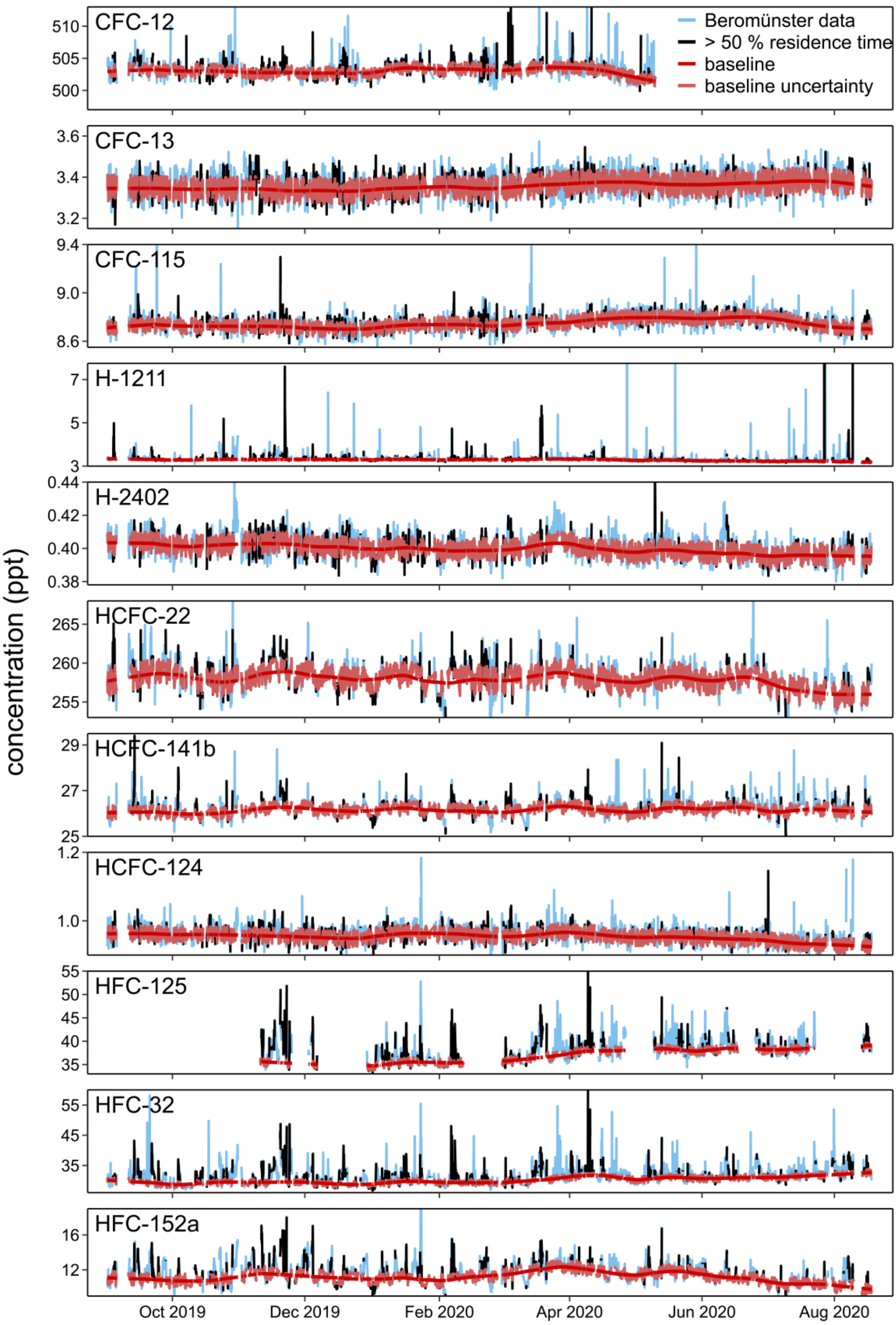
a) (WMO, 2018)

S2 Sampling and analysis

Table S2: Primary calibration scales with accuracies, the uncertainties for the propagation through the three succeeding levels of calibration standards, and the mean measurement precisions. All uncertainties are given at 2σ confidence level.

substance	primary scale	primary scale accuracy (%)	scale propagation uncertainty (%)	mean measurement precision (%)
CFC-11	SIO-05	0.58	2.0	0.47
CFC-12	SIO-05	0.50	2.0	0.41
CFC-13	METAS-2017	4.0	4.2	2.5
CFC-115	SIO-05	0.94	6.0	0.98
H-1211	SIO-05	1.0	6.0	1.0
H-2402	SIO-14	2.0	3.4	2.0
HCFC-22	SIO-05	0.54	2.0	0.62
HCFC-141b	SIO-05	0.28	4.0	0.58
HCFC-142b	SIO-05	0.42	4.0	0.63
HCFC-124	UB-98	4.0	5.0	2.9
HFC-134a	SIO-05	0.56	3.0	0.63
HFC-125	SIO-14	0.40	6.0	3.1
HFC-32	SIO-07	0.98	6.0	0.97
HFC-152a	SIO-05	1.1	6.0	1.1
HFC-245fa	SIO-14	7.0	10	1.7
HFC-365mfc	SIO-14	1.0	6.0	2.2
HFC-23	SIO-07	0.78	6.0	1.1
HFC-227ea	SIO-14	1.5	6.0	1.1
HFC-236fa	SIO-14	1.4	6.0	5.2
HFC-4310mee	SIO-14	3.4	10	4.7
PFC-116	SIO-07	0.58	6.0	0.82
PFC-318	SIO-14	0.46	2.4	1.4
PFC-14	SIO-05	0.46	3.0	0.44
SF_6	SIO-05	0.82	4.0	1.2
NF_3	SIO-12	1.0	4.0	1.5
HFO-1234yf	METAS-2017	4.0	4.6	2.7
HFO-1234ze(E)	EMPA-2013	4.0	3.2	1.8
HCFO-1233zd(E)	EMPA-2013	4.0	3.0	1.7

S3 Measured time series



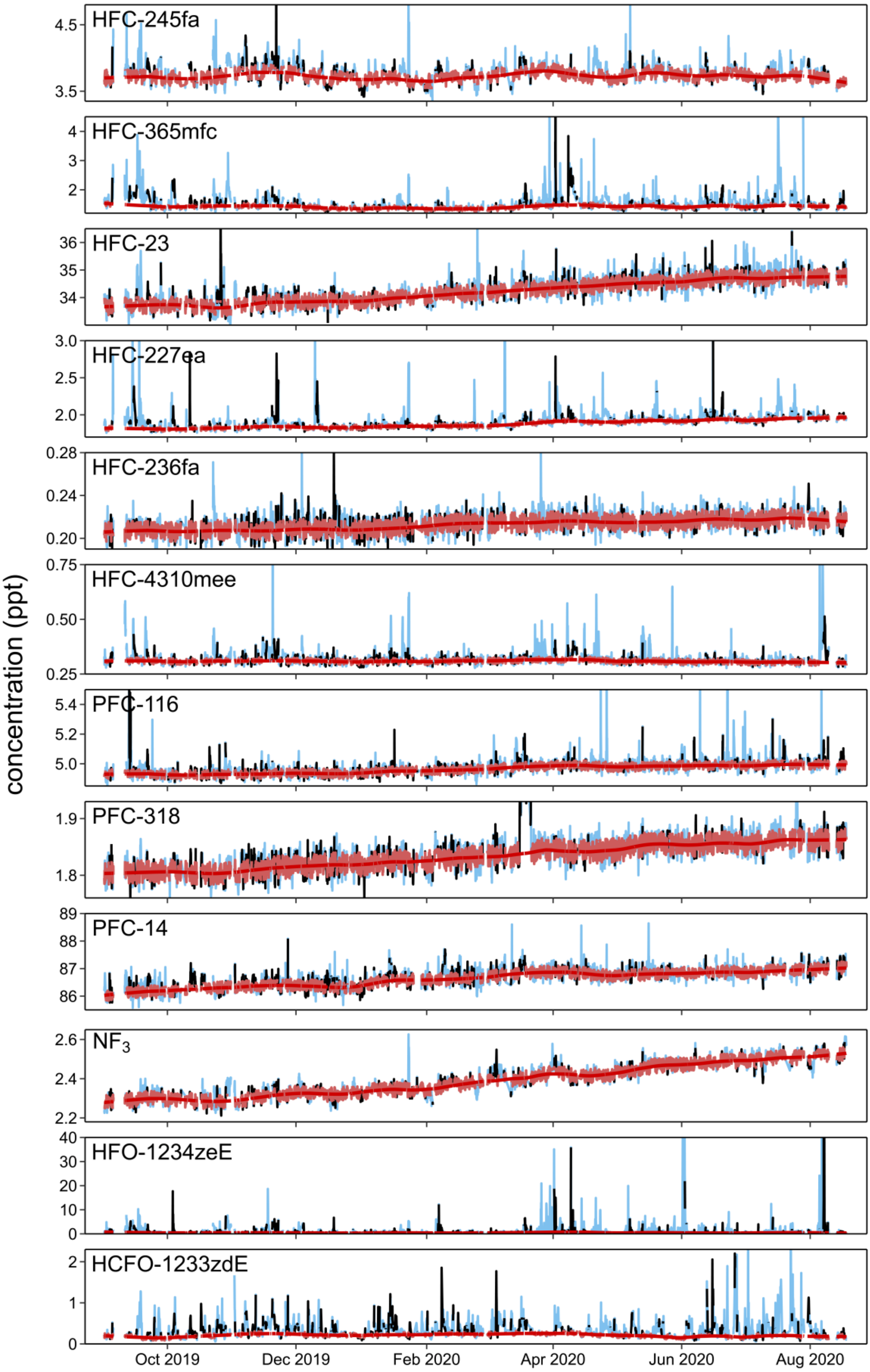


Figure S1: One-year time series of the atmospheric concentrations measured at Beromünster. Air samples with more than 50 % relative residence time in Switzerland are highlighted in black. The baselines (red lines) calculated with a REBS bandwidth parameter of 30 d are shown with their uncertainty bands (light red) derived with the REBS multiplication factor set to 1.5.

S4 Emission estimation by the tracer-ratio method (TRM)

Background evaluation

Table S3: Total number of measurement points considered for the background calculation and the resulting percentage of data assigned as background concentration with the REBS bandwidth parameter set to 30 d and the multiplication factor set to 1, 1.5, and 2. In addition, the average calculated background level for the duration of the measurement campaign is given.

substance	total number of observations	multiplication factor = 1	multiplication factor = 1.5	multiplication factor = 2	
		fraction of background observations (%)			average background concentration (ppt)
CFC-11	3984	54	70	80	226 ± 2
CFC-12	2737	60	78	87	503 ± 1
CFC-13	4022	59	82	93	3 ± 0.1
CFC-115	3998	62	83	92	9 ± 0.07
H-1211	4002	47	63	72	3 ± 0.05
H-2402	4020	61	80	91	0.4 ± 0.01
HCFC-22	3944	69	85	93	258 ± 2
HCFC-141b	3962	66	82	90	26 ± 0.3
HCFC-142b	3986	49	62	72	23 ± 0.3
HCFC-124	4019	60	81	91	1 ± 0.03
HFC-134a	3941	41	57	67	124 ± 4
HFC-125	2256	41	57	69	37 ± 1
HFC-32	4008	43	59	70	30 ± 1
HFC-152a	4005	59	74	83	11 ± 1
HFC-245fa	4005	61	77	86	4 ± 0.1
HFC-365mfc	4015	45	60	71	1 ± 0.1
HFC-23	3995	60	80	90	34 ± 0.3
HFC-227ea	4016	46	62	72	2 ± 0.03
HFC-236fa	4013	61	81	92	0.2 ± 0.01
HFC-4310mee	4017	53	70	80	0.3 ± 0.01
PFC-116	3996	59	78	87	5 ± 0.03
PFC-318	4021	62	82	92	2 ± 0.02
PFC-14	3969	59	78	88	87 ± 0.2
SF_6	3999	50	66	74	10 ± 0.1
NF_3	4012	67	85	93	2 ± 0.04
HFO-1234yf	4003	35	50	60	0.4 ± 0.2
HFO-1234ze(E)	4002	33	47	57	0.5 ± 0.2
HCFO-1233zd(E)	4019	37	51	59	0.2 ± 0.06

Distribution of modeled country residence times within the inversion domain

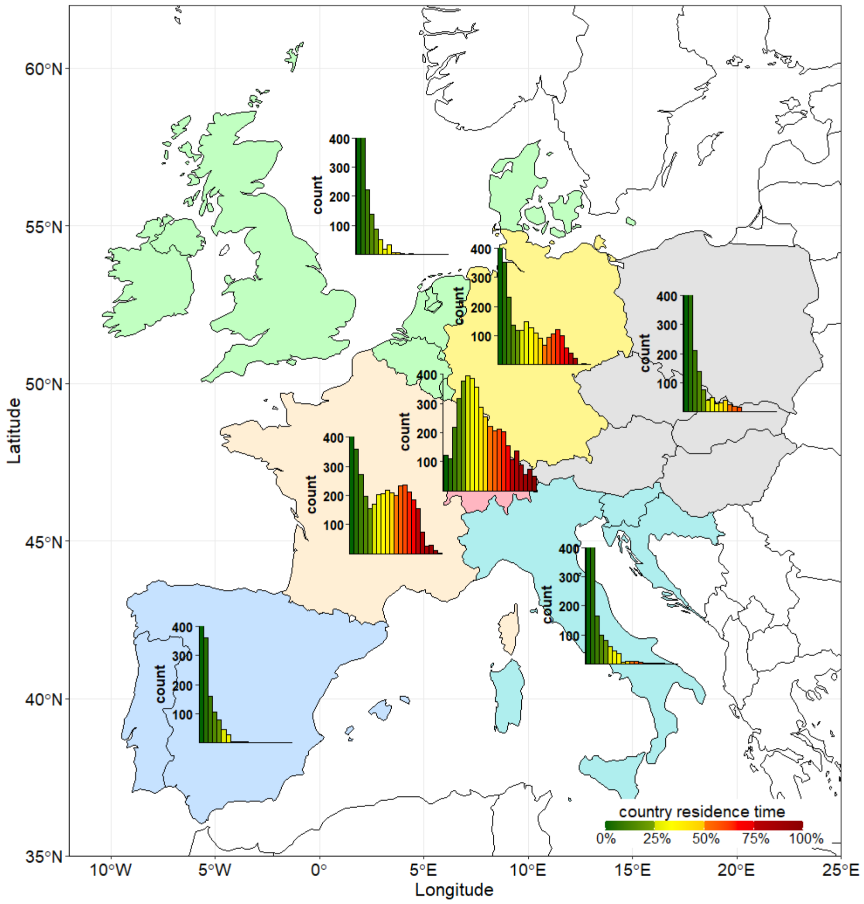


Figure S2: Distributions of the modelled relative country residence times of air masses captured at Beromünster for different European regions.

Variation of REBS and country residence time settings

Table S4: Summary of the minimum, maximum, and average number of data points included in the tracer-ratio method (TRM) calculation for the different REBS and country residence time settings.

substance	minimum number of data points	maximum number of data points	average number of data points	reliability
CFC-11	33	339	133	yes
CFC-12	14	140	55	yes
CFC-13	2	78	22	no
CFC-115	7	138	46	no
H-1211	24	322	131	yes
H-2402	4	146	46	no
HCFC-22	7	169	53	no
HCFC-141b	12	213	74	yes
HCFC-142b	39	353	145	yes
HCFC-124	5	156	49	no
HFC-134a	44	407	167	yes
HFC-125	30	220	100	yes
HFC-32	45	395	168	yes
HFC-152a	37	315	133	yes
HFC-245fa	10	204	71	yes
HFC-365mfc	17	272	99	yes
HFC-23	3	130	40	no
HFC-227ea	19	297	113	yes
HFC-236fa	4	89	28	no
HFC-4310mee	21	233	94	yes
PFC-116	10	155	57	yes
PFC-318	3	122	38	no
PFC-14	5	162	55	no
SF_6	14	205	79	yes
NF_3	4	113	37	no
HFO-1234yf	50	416	177	yes
HFO-1234zeE	43	353	150	yes
HCFO-1233zdE	22	320	124	yes

S5 Emission estimation by Bayesian inverse (BI) modelling

Table S5: Output diagnostics for the Bayesian inverse (BI) modeling, i.e. the χ^2 index, the degrees of freedom (DOF), the a posteriori correlation coefficient (r^2) between simulated and observed signal above baseline at Beromünster, the normalized standard deviation (nSD; ratio of the simulated standard deviation and the observed standard deviation). In addition, an appraisal regarding the reliability of the emission results is given.

substance	χ^2 index	DOF	r^2	nSD	reliability
CFC-11	0.87	59	0.55	0.68	yes
CFC-12	0.60	31	0.11	0.24	yes
CFC-13	0.31	32	0.01	0.03	no
CFC-115	0.29	28	0.06	0.24	no
H-1211	0.87	21	0.11	0.25	yes
H-2402	0.48	32	0.30	0.54	yes
HCFC-22	1.01	27	0.05	0.25	no
HCFC-141b	0.98	28	0.31	0.55	yes
HCFC-142b	0.99	21	0.22	0.67	yes
HCFC-124	0.52	30	0.06	0.22	yes
HFC-134a	1.05	82	0.92	0.77	yes
HFC-125	1.05	69	0.59	1.03	yes
HFC-32	1.01	61	0.46	0.89	yes
HFC-152a	1.01	20	0.43	0.74	yes
HFC-245fa	1.14	51	0.27	0.71	yes
HFC-365mfc	1.06	56	0.34	0.68	yes
HFC-23	0.77	23	0.02	0.25	no
HFC-227ea	1.30	16	0.14	0.50	yes
HFC-236fa	0.44	23	0.04	0.31	no
HFC-4310mee	0.54	27	0.15	0.50	yes
PFC-116	0.94	27	0.27	0.46	yes
PFC-318	0.61	10	0.01	0.16	no
PFC-14	0.63	30	0.04	0.30	no
SF_6	0.86	38	0.33	0.77	yes
NF_3	0.78	12	0.08	0.21	no
HFO-1234yf	1.02	55	0.61	1.06	yes
HFO-1234zeE	1.03	47	0.11	0.34	yes
HCFO-1233zdE	1.01	26	0.12	0.77	yes

S6 Jungfraujoch and national inventory emission records

Table S6: Jungfraujoch (JFJ)-based emission estimates and the Swiss national inventory estimates for the years 2015 to 2018.

	JFJ-based emission estimates[a)]				Swiss national inventory emission estimates[b)]			
substance	2015 ($Mg\ yr^{-1}$)	2016 ($Mg\ yr^{-1}$)	2017 ($Mg\ yr^{-1}$)	2018 ($Mg\ yr^{-1}$)	2015 ($Mg\ yr^{-1}$)	2016 ($Mg\ yr^{-1}$)	2017 ($Mg\ yr^{-1}$)	2018 ($Mg\ yr^{-1}$)
CFC-11	58	57	59	52	-	-	-	-
CFC-12	38	35	27	16	-	-	-	-
CFC-13	–0.1	0.3	–0.5	–1.5	-	-	-	-
CFC-115	1.3	1.4	1.5	1.7	-	-	-	-
H-1211	4.1	4.4	3.4	2.8	-	-	-	-
H-2402	0.3	0.5	0.4	0.4	-	-	-	-
HCFC-22	60	59	58	48	-	-	-	-
HCFC-141b	15	16	15	10	-	-	-	-
HCFC-142b	26	28	27	28	-	-	-	-
HCFC-124	0.7	0.3	0.3	0.3	-	-	-	-
HFC-134a	336	299	285	308	486	496	502	509
HFC-125	100	110	89	90	120	116	121	125
HFC-32	29	30	31	32	40	42	47	51
HFC-152a	36	34	28	25	0.7	0.5	0.4	0.4
HFC-245fa	9.9	10.0	10.0	10.1	0.0	0.2	0.2	0.2
HFC-365mfc	12.6	9.2	8.7	9.2	6.6	6.4	4.9	4.7
HFC-23	6.4	6.0	3.1	3.5	0.6	0.6	0.7	0.6
HFC-227ea	3.3	3.4	3.0	3.4	1.1	1.4	1.4	1.9
HFC-236fa	0.2	0.4	0.4	0.4	0.5	0.5	0.5	0.5
HFC-4310mee	1.0	1.1	1.4	1.6	0.6	1.0	0.8	0.5
PFC-116	1.4	1.3	1.3	0.8	0.3	0.3	0.4	0.5
PFC-318	0.1	0.3	0.1	0.4	0.0	0.1	0.0	0.0
PFC-14	6.7	5.9	4.6	3.2	0.6	0.3	0.6	0.6
SF_6	8.7	8.2	6.9	6.5	12	9.4	8.9	6.9
NF_3	0.3	0.5	0.5	0.5	0.0	0.0	0.1	0.0
HFO-1234yf	-	-	-	-	-	-	-	-
HFO-1234zeE	-	-	-	-	-	-	-	-
HCFO-1233zdE	-	-	-	-	-	-	-	-

a) (Reimann et al., 2021) and (Reimann, 2021)

b) (UNFCCC, 2021)

S7 Source regions

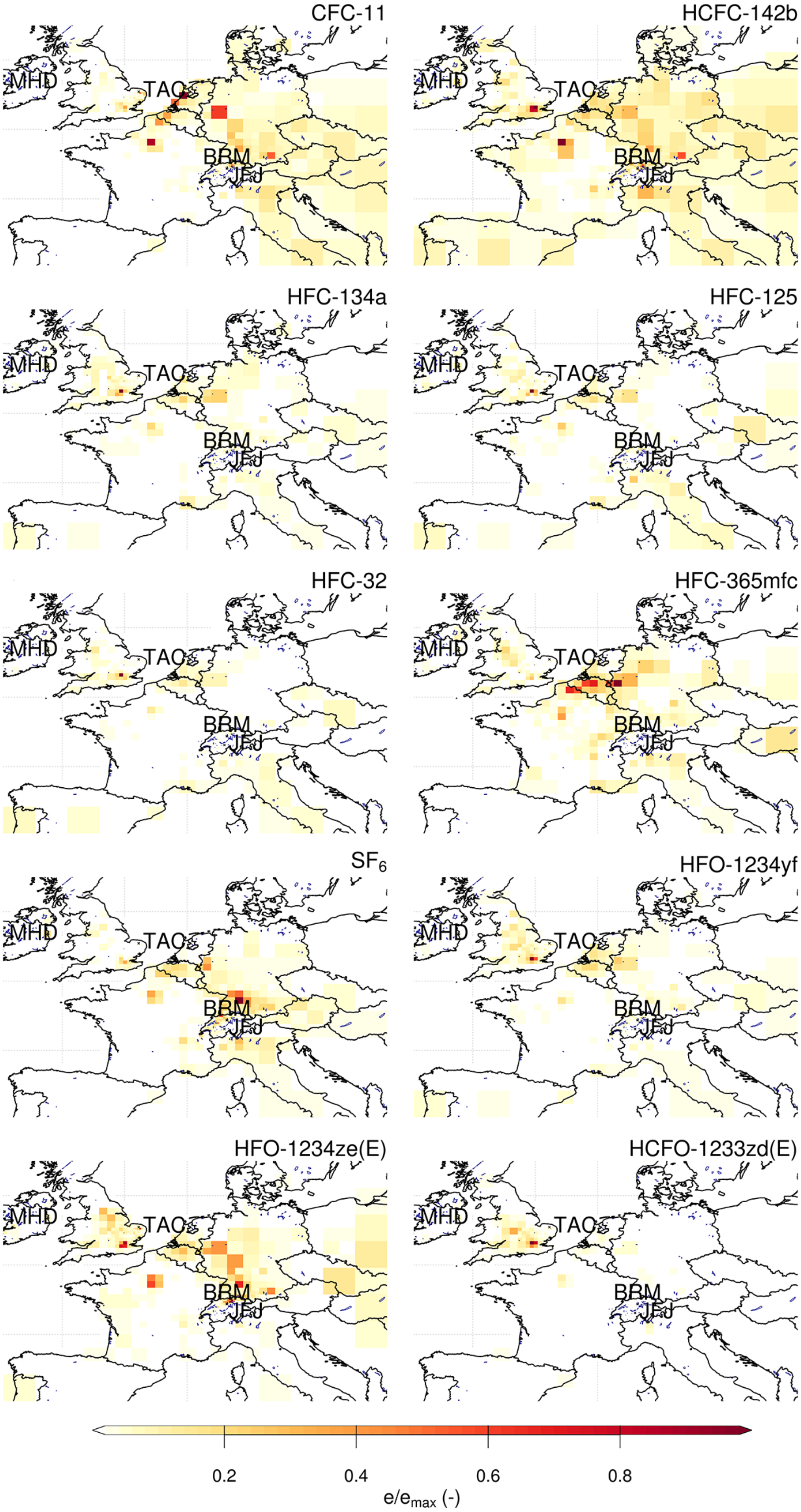


Figure S3: Emission maps for the European domain, generated from the Bayesian inverse (BI) modelling. The sites at Beromünster (BRM), Jungfraujoch (JFJ), Tacolneston (TAC), and Mace Head (MHD) are indicated. Gridded emissions (e), given in µg s^{-1} km^{-2}, are scaled to the global maximum (e_{max}).

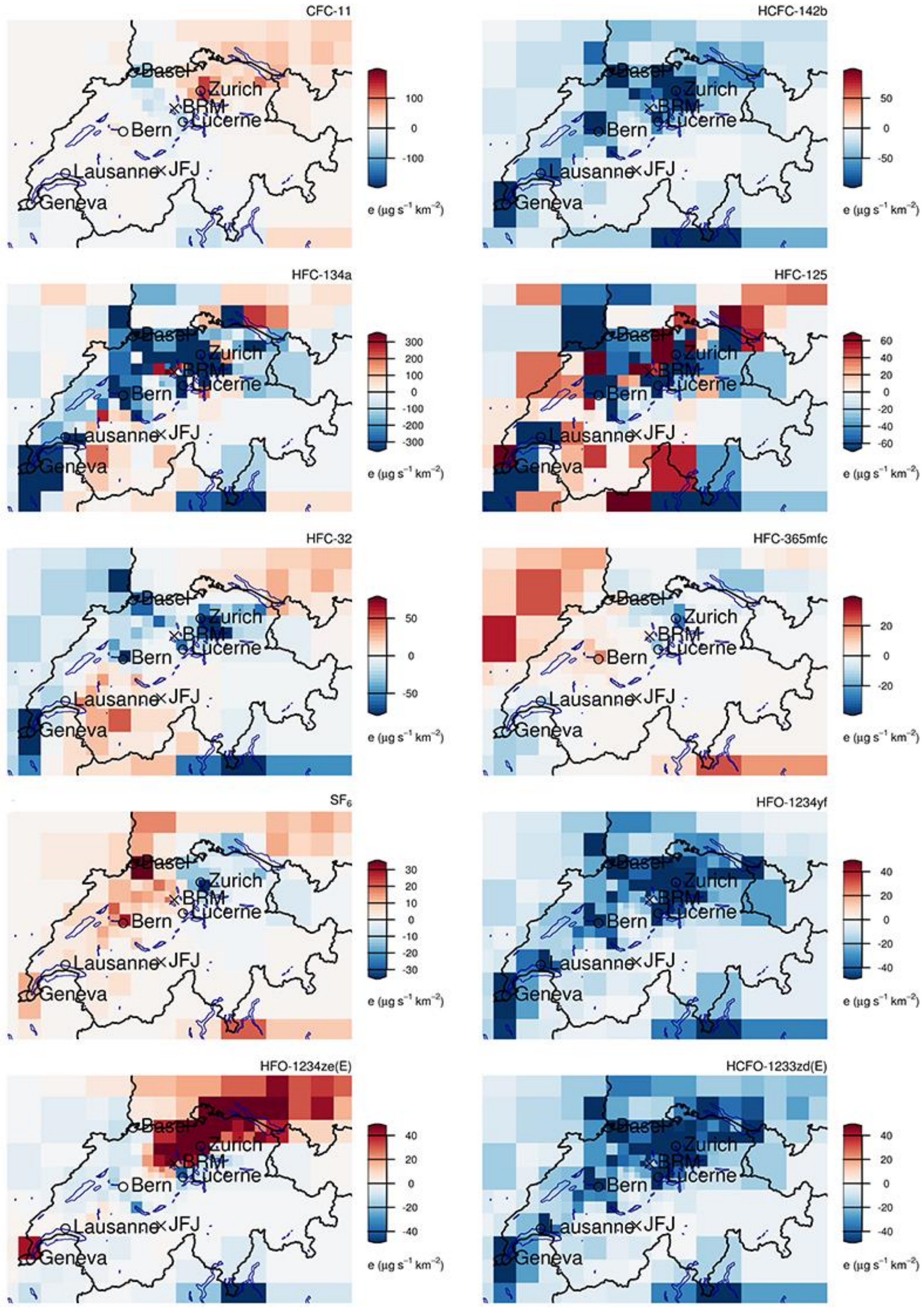


Figure S4: Absolute differences of the a priori and the a posteriori emission values, generated from the Bayesian inversion (BI). Beromünster (BRM), Jungfraujoch (JFJ), and major Swiss cities are indicated. Gridded emissions (e) are given in µg s^{-1} km^{-2}.

References

Reimann, S., Vollmer, M. K., Henne, S., Brunner, D., Steinbacher, M., Schlauri, P., Hill, M., and Emmenegger, L.: CLIMGAS-CH Kontinuierliche Messung der Nicht-CO_2-Treibhausgase auf dem Jungfraujoch und in Beromünster, Laboratory for Air Pollution/ Environmental Technology, Empa, Swiss Federal Laboratories for Materials Science and Technologies, Dübendorf, Switzerland, 94 pp., 2021.

Reimann, S.: CLIMGAS-CH Kontinuierliche Messung der Nicht-CO_2-Treibhausgase auf dem Jungfraujoch und in Beromünster, Swiss Federal Laboratories for Materials Science and Technologies, Dübendorf, data set upon request, 2021.

UNFCCC (United Nations Framework Convention on Climate Change): National Inventory Submissions 2021, https://unfccc.int/ghg-inventories-annex-i-parties/2021: last access 11 May 2021.

WMO: Scientific Assessment of Ozone Depletion: 2018, Global Ozone Research and Monitoring Project–Report No. 58, Geneva, Switzerland, 588 pp., 2018.