Supplement of

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

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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

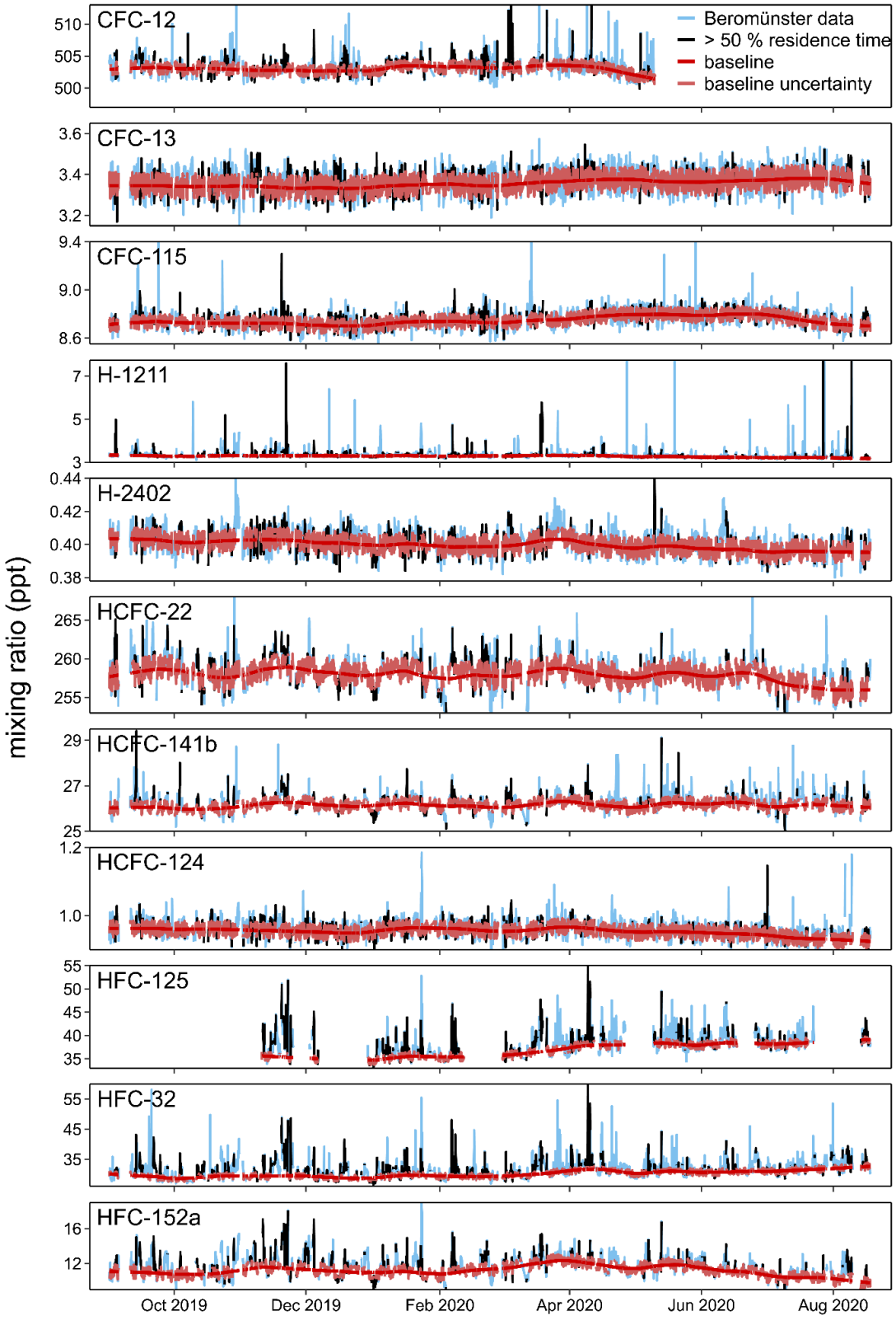
a) (WMO, 2018)

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 2-sigma 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



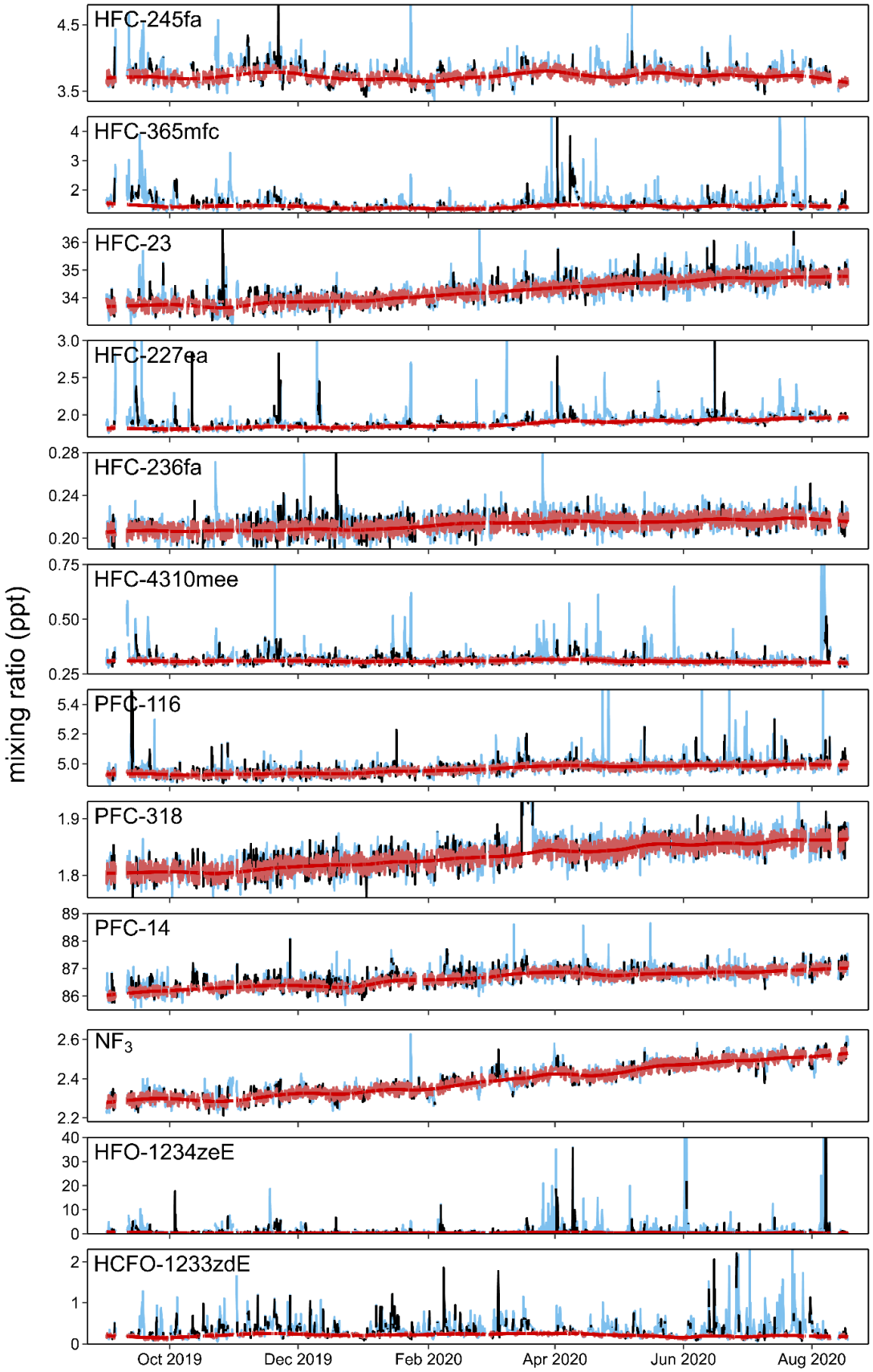


Figure S1: One-year time series of the atmospheric concentrations measured at Beromünster. Air samples with more than 50 % relative residence time in Switzerland are highlighted in black. The baselines (red lines) calculated with a REBS bandwidth parameter of 30 days are shown with their uncertainty bands (light red) derived with the REBS sigma 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 setting of 30 days and a sigma factor of 1.5. In addition, the average calculated background level for the duration of the measurement campaign is given.

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

15 **Distribution of modeled country residence times within the inversion domain**

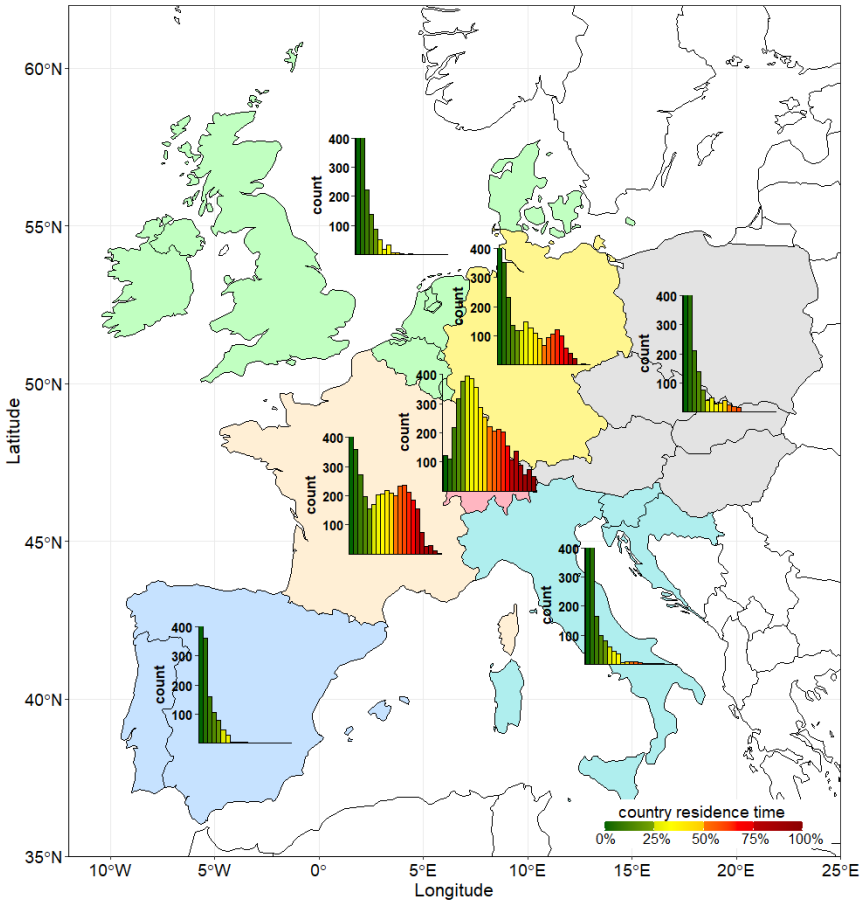


Figure S2: Distributions of the modelled relative country residence times of air masses captured at Beromünster for different European regions.

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
CFC-11	33	339	133
CFC-12	14	140	55
CFC-13	2	78	22
CFC-115	7	138	46
H-1211	24	322	131
H-2402	4	146	46
HCFC-22	7	169	53
HCFC-141b	12	213	74
HCFC-142b	39	353	145
HCFC-124	5	156	49
HFC-134a	44	407	167
HFC-125	30	220	100
HFC-32	45	395	168
HFC-152a	37	315	133
HFC-245fa	10	204	71
HFC-365mfc	17	272	99
HFC-23	3	130	40
HFC-227ea	19	297	113
HFC-236fa	4	89	28
HFC-4310mee	21	233	94
PFC-116	10	155	57
PFC-318	3	122	38
PFC-14	5	162	55
SF_6	14	205	79
NF_3	4	113	37
HFO-1234yf	50	416	177
HFO-1234zeE	43	353	150
HCFO-1233zdE	22	320	124

S5 Emission estimation by Bayesian inverse (BI) modelling

Table S5: Output diagnostics for the Bayesian inverse (BI) modeling, i.e. the Chi index, the degrees of freedom (DF), the a posteriori correlation coefficient (r) 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	Chi index	DF	r	nSD	Reliability
CFC-11	0.9	35	0.6	0.5	yes
CFC-12	0.6	38	0.3	0.3	yes
CFC-13	0.4	21	–0.1	0.6	no
CFC-115	0.3	17	0.1	0.1	no
H-1211	0.9	31	0.3	0.3	yes
H-2402	0.5	17	0.3	0.4	no
HCFC-22	1.0	28	0.4	0.4	yes
HCFC-141b	1.0	34	0.4	0.5	yes
HCFC-142b	1.0	11	0.4	0.6	yes
HCFC-124	0.6	30	0.1	0.1	no
HFC-134a	1.0	52	0.6	0.7	yes
HFC-125	1.0	55	0.6	1.0	yes
HFC-32	1.0	49	0.6	0.9	yes
HFC-152a	1.0	27	0.4	0.5	yes
HFC-245fa	1.1	28	0.3	0.5	yes
HFC-365mfc	1.1	54	0.5	0.8	yes
HFC-23	1.2	25	0.0	0.3	no
HFC-227ea	1.5	15	0.3	0.5	yes
HFC-236fa	0.5	30	0.1	0.4	no
HFC-4310mee	0.6	17	0.3	0.4	yes
PFC-116	0.9	28	0.2	0.6	yes
PFC-318	0.7	15	0.1	0.1	no
PFC-14	0.6	26	0.2	0.2	no
SF_6	0.9	53	0.5	0.8	yes
NF_3	0.7	25	0.4	0.3	yes
HFO-1234yf	1.0	60	0.6	1.1	yes
HFO-1234zeE	1.0	26	0.2	0.2	yes
HCFO-1233zdE	1.1	18	0.3	0.5	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	56	54	56	50	-	-	-	-
CFC-12	36	33	26	15	-	-	-	-
CFC-13	–0.1	0.3	–0.5	–1.4	-	-	-	-
CFC-115	1.2	1.4	1.4	1.7	-	-	-	-
H-1211	4.0	4.3	3.3	2.7	-	-	-	-
H-2402	0.3	0.5	0.4	0.4	-	-	-	-
HCFC-22	58	57	55	46	-	-	-	-
HCFC-141b	15	15	14	9	-	-	-	-
HCFC-142b	25	26	26	26	-	-	-	-
HCFC-124	0.7	0.3	0.3	0.3	-	-	-	-
HFC-134a	323	287	272	292	486	496	502	509
HFC-125	96	105	85	86	120	116	121	125
HFC-32	28	29	29	30	40	42	47	51
HFC-152a	34	32	26	24	0.7	0.5	0.4	0.4
HFC-245fa	9.5	9.6	9.6	9.6	0.0	0.2	0.2	0.2
HFC-365mfc	12	8.8	8.3	8.8	6.6	6.4	4.9	4.7
HFC-23	6.1	5.8	3.0	3.3	0.6	0.6	0.7	0.6
HFC-227ea	3.2	3.2	2.9	3.2	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	1.3	1.5	0.6	1.0	0.8	0.5
PFC-116	1.3	1.3	1.2	0.7	0.3	0.3	0.4	0.5
PFC-318	0.1	0.3	0.1	0.3	0.0	0.1	0.0	0.0
PFC-14	6.5	5.6	4.4	3.0	0.6	0.3	0.6	0.6
SF_6	8.4	7.9	6.6	6.2	12	9.4	8.9	6.9
NF_3	0.3	0.5	0.4	0.4	0.0	0.0	0.1	0.0
HFO-1234yf	-	-	-	-	-	-	-	-
HFO-1234zeE	-	-	-	-	-	-	-	-
HCFO-1233zdE	-	-	-	-	-	-	-	-

a) (UNFCCC, 2021)

b) (Reimann et al., 2021)

S7 Source regions

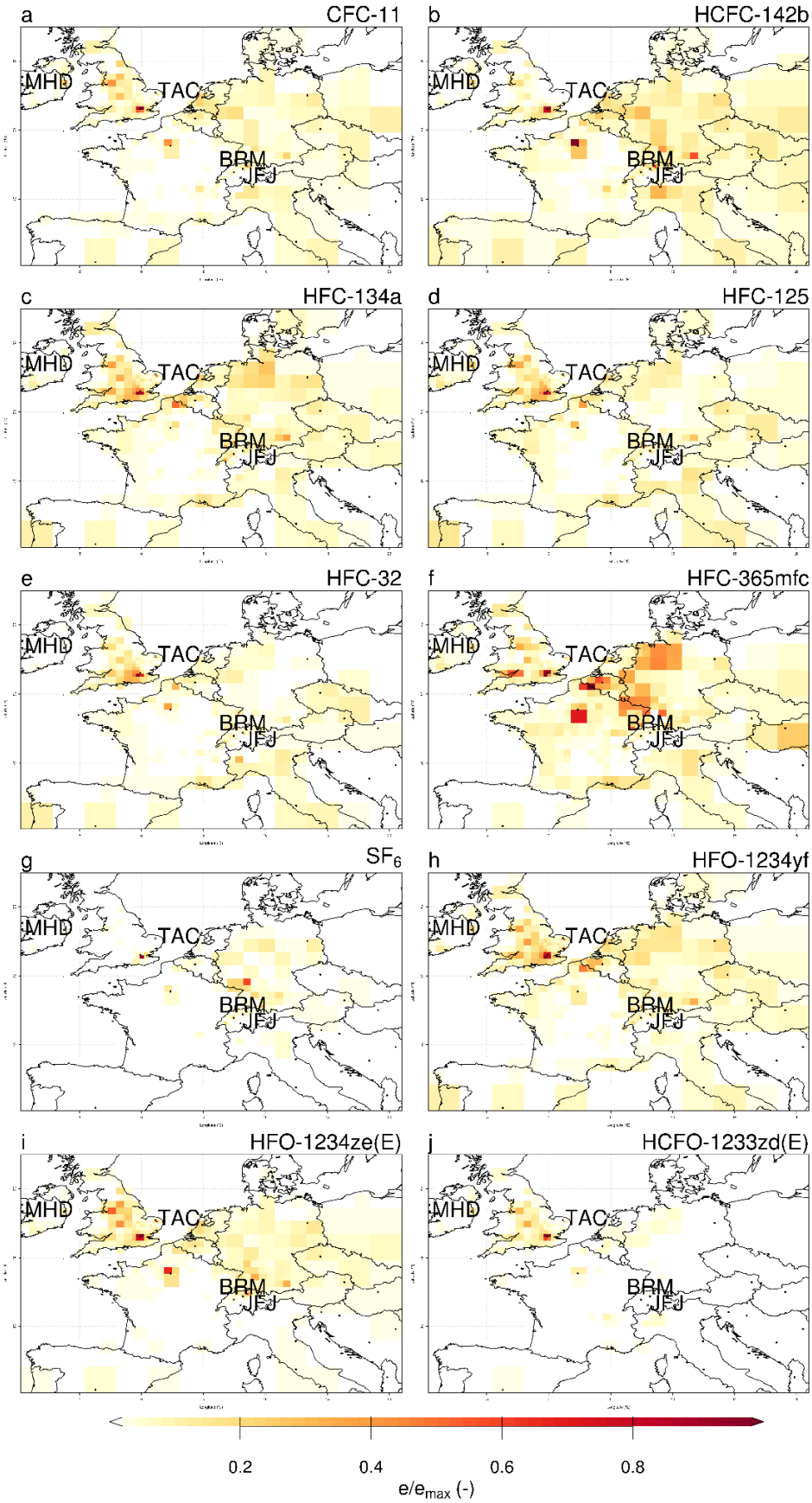


Figure S3: Emission maps for the European domain, generated from the Bayesian inverse (BI) modelling. The sites at Beromünster (BRM), Jungfraujoch (JFJ), Tacolneston (TAC), and Mace Head (MHD) are indicated. Gridded emissions (e), given in µg s^{-1} km^{-2}, are scaled to the global maximum (e_{max}).

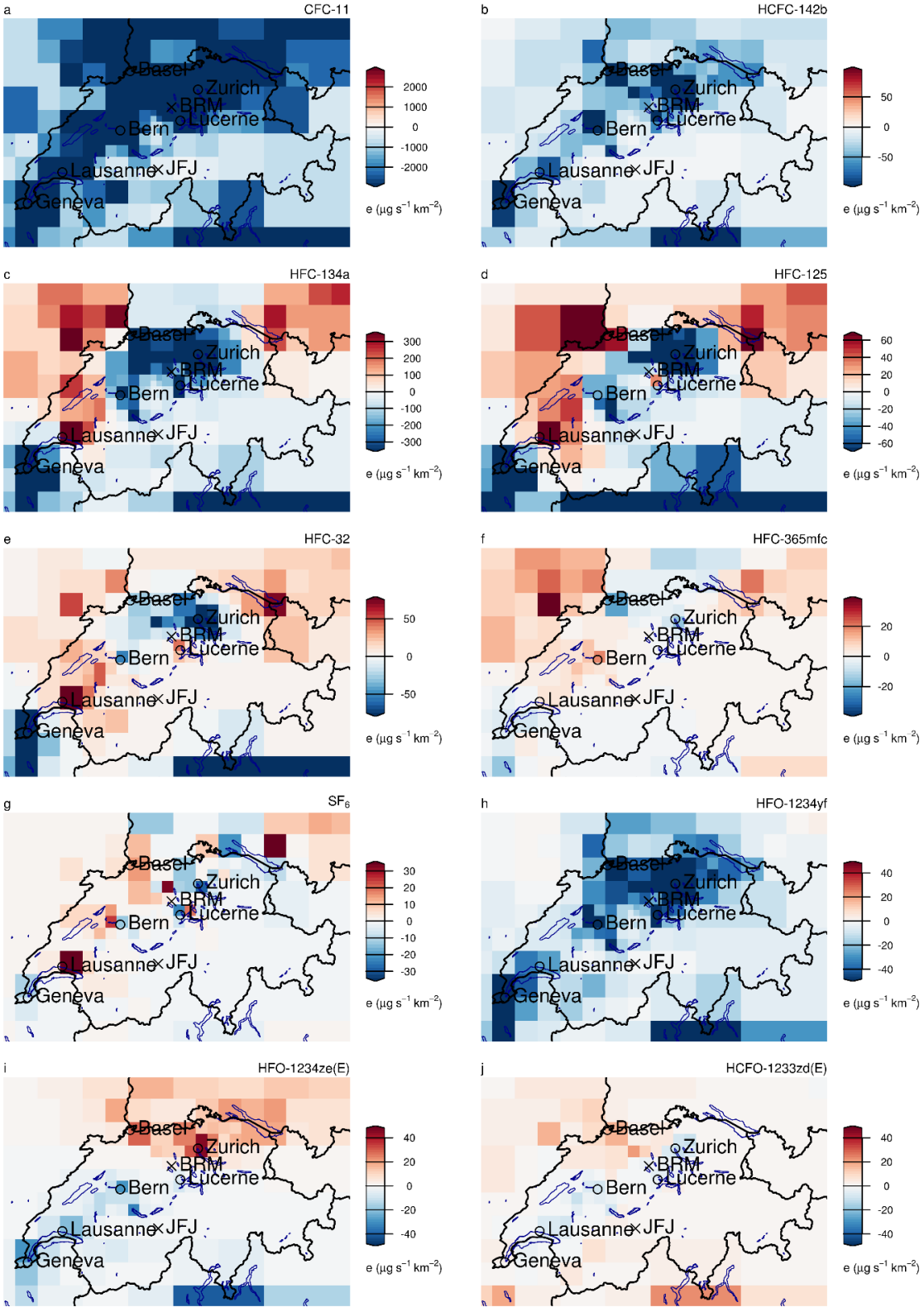


Figure S4: Absolute differences of the a priori and the a posteriori emission values, generated from the Bayesian inversion (BI). Beromünster (BRM), Jungfraujoch (JFJ), and major Swiss cities are indicated. Gridded emissions (e) are given in µg s^{-1} km^{-2}.

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