Supplement of

Continuous increase in East Asia HFC-23 emissions inferred from high-frequency atmospheric observations from 2008 to 2019

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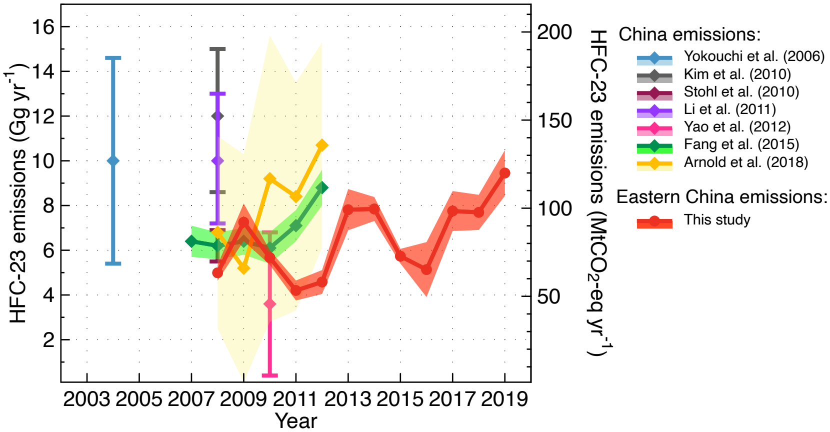

Figure S1: HFC-23 emissions estimate of eastern China in this study, in comparison with previous estimates of Chinese HFC-23 emissions.

Table S1: Information on HCFC-22 production factories in eastern China

#	Company Name	Location	Information
1	Nanjing Jiling Refrigeration Technology Co., Ltd	Jiangsu	Company websites
2	Jiangsu Huafu Poly Environmental Protection Technology Co., Ltd	Jiangsu	Company websites
3	Harbin Sanyi Refrigeration Equipment Co., Ltd	Heilongjian	Company websites
4	Shenyang Guyun Refrigeration Equipment Co., Ltd	Liaoning	Company websites
5	Quzhou saitel Chemical Co., Ltd	Zhejiang	Company websites
6	Shandong Danbu Chemical Co., Ltd	Shandong	Company websites
7	Hangzhou Wula Chemical Co., Ltd	Zhejiang	Company websites
8	Zhejian JuHua Co., Ltd = zhengjiang Quhua Co., Ltd	Zhejiang	TEAP, 2017
9	Changshu Haike_3F Changsu	Jiangsu	TEAP, 2017
10	Shandong Dongyue Chiemical Co., Ltd	Shandong	TEAP, 2017
11	Jiangsu Meilan Chemical Co., Ltd	Jiangsu	TEAP, 2017
12	3F Zonghao_Arkema Changshu (Jiangsu)	Jiangsu	TEAP, 2017
13	Limin Chemical Co., Ltd	Zhejiang	TEAP, 2017
14	Zhejian Dongyang Chemical Co., Ltd	Zhejiang	Company websites
15	Zhonghao Cheguang Research Institute of Chemical Industry	Sichuan	TEAP, 2017
16	China Fluoro Technology Co., Ltd	Shandong	cdm.unfccc.int/Projects/DB/DNV-CUK1182313000.09/view
17	Yingpeng Chemical Co., Ltd	Zhejiang	cdm.unfccc.int/Projects/DB/DNV-CUK1215776483.62/view
18	Shandong ZhongFu	Shandong	TEAP, 2017
19	Zhejiang Sanmei Chemical Incorporated Company	Zhejiang	TEAP, 2017
20	Jingxi YingGuang	Jiangxi	TEAP, 2017
21	Sichuan Zigong Honghe Chemical Co., Ltd.	Sichuan	TEAP, 2017
22	Zhejian Pengyou	Zhejiang	TEAP, 2017
23	Jinhua Yonghe Fluorochemical	Zhejiang	TEAP, 2017
24	Zigong City refrigerant factory	Zigong	Company websites
25	Zigong City reactor Chemical plant	Zigong	Company websites
26	Zhonghao Cheguang Chemical Engineering Research Institute	Sichuan	Company websites
27	Changjiang Chemical Plant	Hebei	Company websites
28	1194 China Fluoro Technology Co. Ltd.	Shandong	Company websites
29	Jinan 3F Fluoro Chemical Co. Ltd.	Shandong	Company websites
30	Zhejiang Yonghe New type Refrigerant Co. Ltd.	Zhejiang	Company websites
31	Shandong Haihua Group Co. Ltd.	Shandong	Company websites
32	Dongyang Chemical	Jinhua	Company websites
33	Jiangxi Sanmei Chemical	Jiangxi	TEAP, 2017
34	Changshu 3 Zhonghao New Chemical Materials Co. Ltd.	Jiangsu	TEAP, 2017
35	Changshu Atofina 3F Fluorine Chemical Co. Ltd.	Jiangsu	TEAP, 2017
36	Ningbo Koman's Refrigeration Industry Co. Ltd.	Zhejiang	Company websites
37	Inner Mongolia Yonghe Fluorochemical Co.	Inner Mongolia	Inside Climate News
38	Zhejiang Jusheng Fluorochemical Co.=Zhejian Quhua	Zhejiang	Inside Climate News
39	Liaocheng Fu'er New Materials Technology Co.	Shandong	Inside Climate News
40	Fujian Shaowu Youghe Jintang New Materials Co.	Fujian	Inside Climate News
41	Fujian Sannong New Materials Co.	Fujian	Inside Climate News
42	Fujian Haidefu New Materials Co.	Fujian	Inside Climate News
43	Zongfu Chemical Material Technology Co.	Sichuan	Inside Climate News
44	Jianxi Liwen Chemical Co.	Jiangxi	Inside Climate News
45	Shandong Hua Fluorochemical Co.	Shandong	Inside Climate News

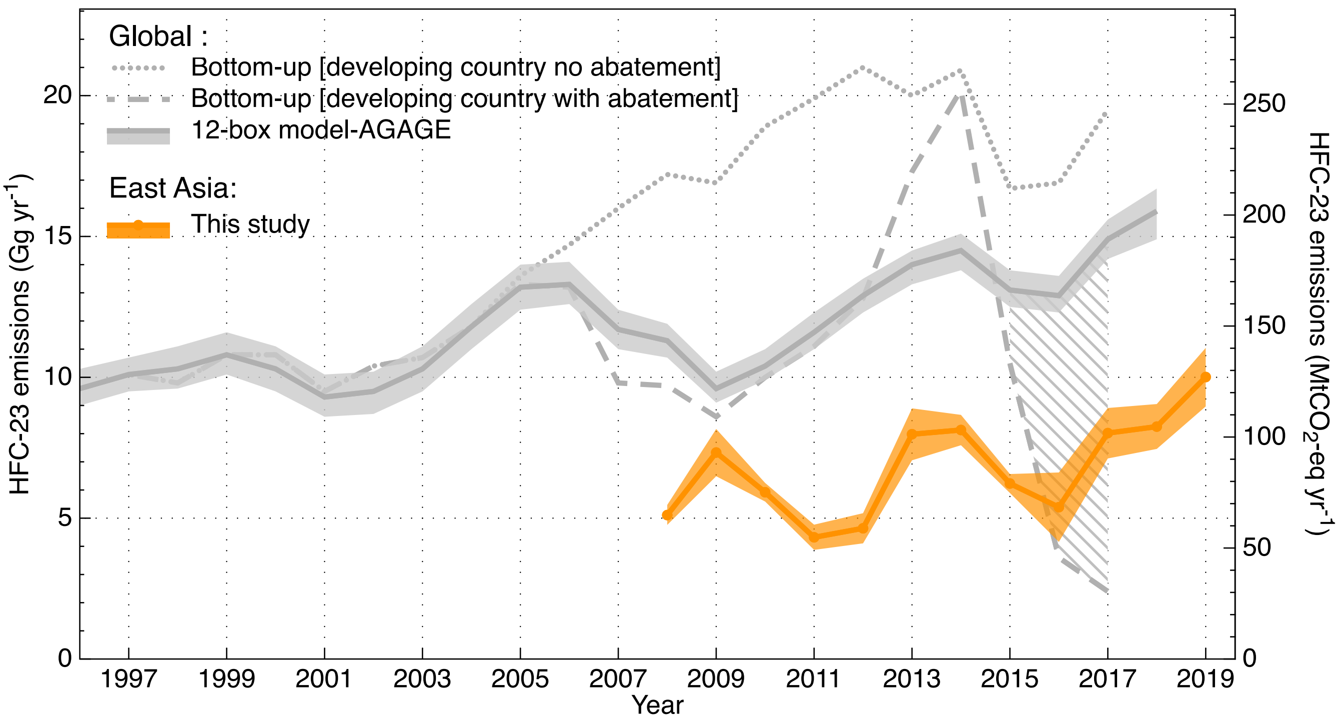

Figure S2: Observation-based HFC-23 emissions from East Asia versus top-down and bottom-up global HFC-23 emissions derived from AGAGE global data (Simmonds et al., 2018; Stanley et al., 2020).

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Three different spatial distributions of *a priori* emissions

Our inverse modelling results represent the mean and 2-σ uncertainty of 27 different model runs, where each set of three different *a priori* distributions (Figure S3) have 9 combinations of different *a priori* emission magnitudes. The first priori distribution is the "Population" *a priori*, determined based on the 2010 World population distribution (Warszawski et al., 2017). Population distribution has often been used as a reasonable first approximation when more specific information is not available (Stohl et al., 2010; Fang et al., 2019). The second priori distribution is the "Asiaflat" *a priori*, where the emissions within each country (whole of China, North Korea, South Korea, and whole of Japan) were evenly spread (Rigby et al., 2019; Park et al., 2021). This flattening may cause large *a priori* emissions to be allocated to western China and eastern Japan, where transport sensitivity was relatively low, while at the same time significantly lowering *a priori* emissions in eastern China and western Japan compared to other *a priori* distributions (Kim et al., 2021). The third priori distribution is the "EastAsiaflat" *a priori*, where the population distribution-based emissions are regionally flattened. Flatting regions are determined as the high sensitivity in the model domain (most of the high sensitivity area has less than or equal to 1° by 1°), i.e., eastern China, South Korea, North Korea, and western Japan. The region denoted "eastern China" contains nine provinces (Anhui, Beijing, Hebei, Jiangsu, Liaoning, Shandong, Shanghai, Tianjin, and Zhejiang) and "western Japan" contains four regions (Chūgoku, Kansai, Kyūshū & Okinawa and Shikoku) (Rigby et al., 2019; Park et al., 2021; Kim et al., 2021). "EastAsiaflat" *a priori* can be unbiased in terms of emission locations, such that the distribution of emissions in the posterior could point to likely emission hot spots, but such inference is reasonable only in regions where the influence of the observations is relatively strong. The *a priori* emissions were kept constants for all years, based on the 2008 emissions. For each *a priori* distribution, we tested 9 different combinations of *a priori* emission magnitudes and uncertainties, which are summarized in Table S2.

Note that our annual HFC-23 results represent the mean of 18 results from two different model runs *a priori* excluding "Asiaflat" *a priori*, because many HCFC-22 factories are located in eastern China, and thus HFC-23 emissions estimates for eastern China using "Asiaflat" *a priori* could be underestimated.

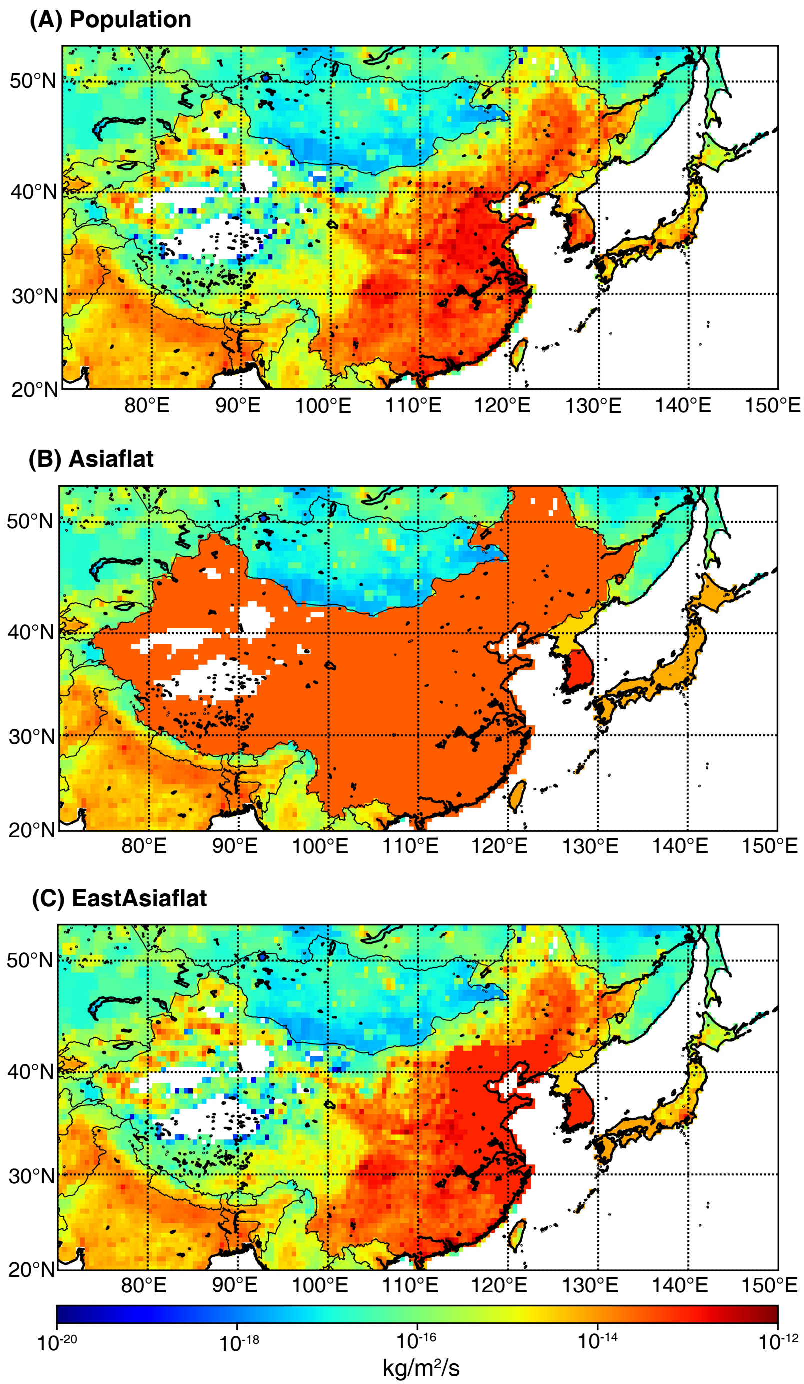

Figure S3: Three different spatial distributions of *a priori* used in this study. (a) *a priori* emissions map based on populations distribution. (b) flat emissions for each country. (c) flat emissions only in the regions with high sensitivity (eastern China, North Korea, South Korea and western Japan).

Table S2: List of 9 *a priori* configurations, with corresponding errors, applied to each *a priori* distribution. For each *a priori* configuration, the initial *a priori* estimate is multiplied by the listed scaling factor.

***A priori* Emissions**								
A priori Dist.	priori	Error (%)	priori Dist.	priori	Error (%)	priori Dist.	priori	Error (%)
	× 0.5	100		× 0.5	100		× 0.5	100
	× 1	50		× 1	50		× 1	50
	× 2	25		× 2	25		× 2	25
	× 0.5	200		× 0.5	200		× 0.5	200
Population	× 1	100	Asiaflat	× 1	100	EastAsiaflat	× 1	100
	× 2	50		× 2	50		× 2	50
	× 0.5	300		× 0.5	300		× 0.5	300
	× 1	150		× 1	150		× 1	150
	× 2	75		× 2	75		× 2	75

Model validation based on CFC-11 emissions estimate from eastern China

To validate the optimization of our inversion framework, we analyzed CFC-11 emissions from eastern China using Gosan CFC-11 concentration data for 2008–2019. Top-down emissions of CFC-11 in East Asia were well-defined in a recent study (Park et al., 2021) which used multiple inversion methods.

Our results (Figure S4) showed good convergence among the runs for the same *a priori* distribution set, but relatively large (not statistically significant) difference between different *a priori* distributions, suggesting that *a priori* distributions have an impact on the a posteriori, and thus uncertainties associated with different *a priori* settings need to be considered to derive the full posteriori uncertainties.

Figure S5 shows that our CFC-11 emission estimates for eastern China are consistent within uncertainties with previously reported results from four different inverse methods that used Gosan observation data (only) (Park et al., 2021). Since the previous study with four different inverse models had used “Asiaflat” *a priori*, our estimation was also made with the same priori for direct comparison.

We took the mean of the a posteriori annual inversion results from 27 independent inversions with different *a priori* settings (see Table S2) as our final estimates of CFC-11 emissions from eastern China for 2008–2019 (Figure S6 and Table S3) and their uncertainties were defined as 2-$\boldsymbol{\sigma}$ of the resulting a posteriori emissions (95 % uncertainty), because the uncertainty of each inversion run can be considered fully correlated with each other. Our final emissions estimates of CFC-11 for eastern China are also consistent with previously reported results from four different inverse methods within uncertainties (Figure S6).

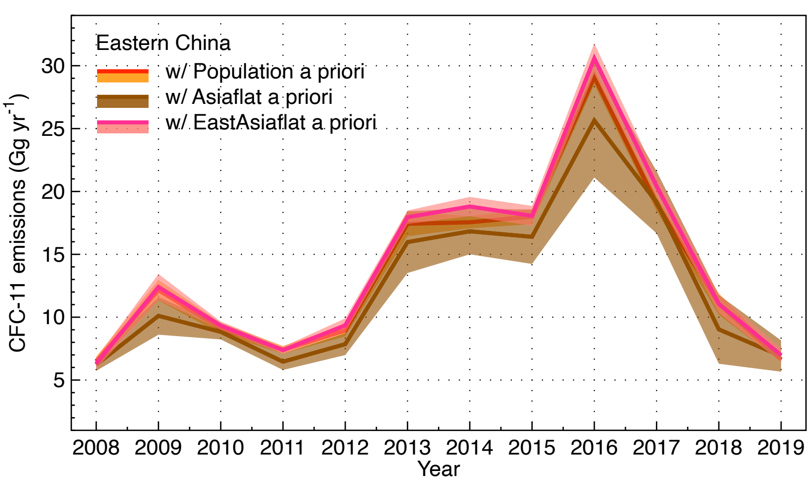

Figure S4: CFC-11 emissions from eastern China derived using three different *a priori* distributions: “Population”, “Asiaflat”, “EastAsiaflat” *a prioris*. Each line represents the annual mean of 9 different model set-ups for each *a priori* distribution. Shading denotes 2-σ uncertainties.

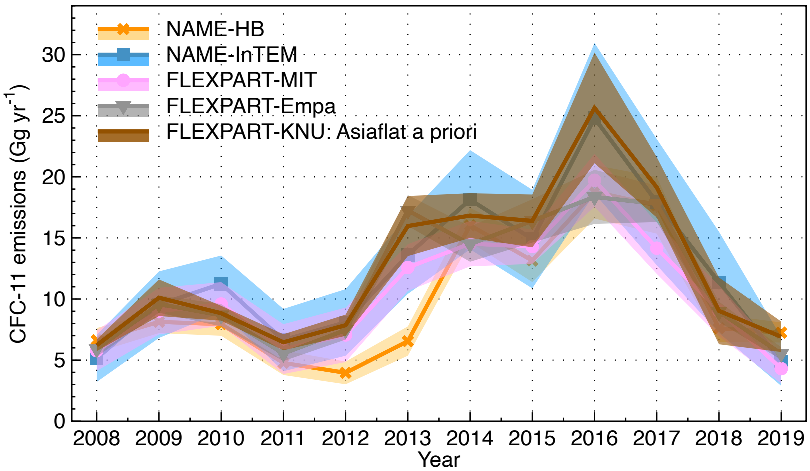

Figure S5: CFC-11 emissions from eastern China derived using the "Asiaflat" *a priori* distribution. Top-down emissions using FLEXPART-KNU (brown line) are compared to previously reported emissions from four different inverse methods using the same Gosan data for 2008–2019: NAME-HB (yellow crosses), NAME-InTEM (blue squares), FLEXPART-MIT (pink circles) and FLEXPART-Empa (gray triangles).

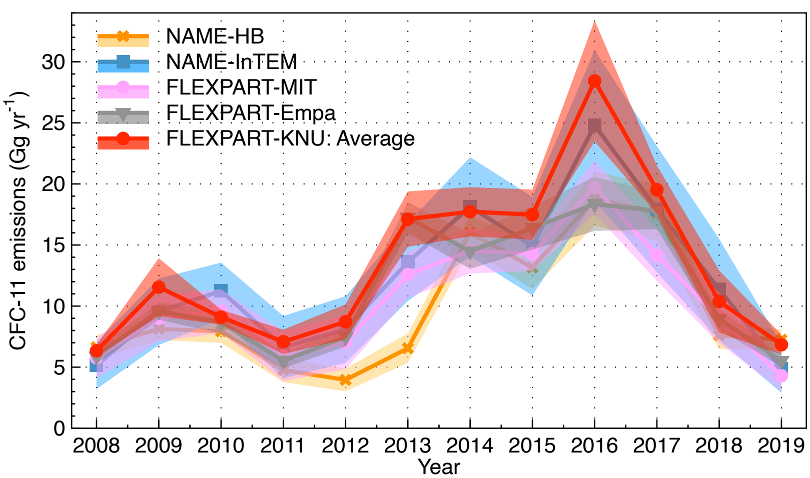

Figure S6: CFC-11 emissions estimate for eastern China derived in this study (FLEXPART-KNU, red circles) compared to previously derived emissions. Our annual results represent the mean of 27 different model runs, where each set of three different *a priori* distributions have 9 combinations of different emission magnitudes and 2-σ uncertainties (shading).

Table S3: Top-down emissions (Gg yr^{-1}) of CFC-11 and HCFC-22 for eastern China

Year	CFC-11			HCFC-22		
	Eastern China			Eastern China		
	mean	max	min	mean	max	min
2008	6.34	6.81	5.86	40.99	45.53	36.44
2009	11.55	13.92	9.18	60.96	76.40	45.51
2010	9.09	9.70	8.48	62.45	73.46	51.44
2011	7.08	8.07	6.09	47.36	59.20	35.52
2012	8.72	10.15	7.29	43.37	51.17	35.56
2013	17.12	19.39	14.85	61.07	70.27	51.87
2014	17.73	19.73	15.72	59.43	68.57	50.30
2015	17.48	19.54	15.43	58.80	66.67	50.93
2016	28.44	33.39	23.48	88.13	111.27	64.98
2017	19.53	21.49	17.56	87.23	102.62	71.84
2018	10.38	12.92	7.84	64.89	83.67	46.12
2019	6.84	7.66	6.01	86.89	100.13	73.64

SI. References

Arnold, T., Manning, A. J., Kim, J., Li, S., Webster, H., Thomson, D., Mühle, J., Weiss, R. F., Park, S., and O'doherty, S.: Inverse modelling of CF4 and NF3 emissions in East Asia, Atmospheric Chemistry and Physics, 18, 13305-13320, 2018.

Fang, X., Stohl, A., Yokouchi, Y., Kim, J., Li, S., Saito, T., Park, S., and Hu, J.: Multiannual top-down estimate of HFC-23 emissions in East Asia, Environmental Science & Technology, 49, 4345-4353, 2015.

Fang, X., Yao, B., Vollmer, M., Reimann, S., Liu, L., Chen, L., Prinn, R., and Hu, J.: Changes in HCFC emissions in China during 2011–2017, Geophysical Research Letters, 46, 10034-10042, 2019.

Inside Climate News: China Moves to Freeze Production of Climate Super-Pollutants But Lacks a System to Monitor Emissions, 2022. https://insideclimatenews.org/news/21012022/china-super-pollutant-emissions/

Kim, J., Li, S., Kim, K. R., Stohl, A., Mühle, J., Kim, S. K., Park, M. K., Kang, D. J., Lee, G., and Harth, C. M.: Regional atmospheric emissions determined from measurements at Jeju Island, Korea: Halogenated compounds from China, Geophysical Research Letters, 37, 2010.

Kim, J., Thompson, R., Park, H., Bogle, S., Mühle, J., Park, M. K., Kim, Y., Harth, C. M., Salameh, P. K., and Schmidt, R.: Emissions of tetrafluoromethane (CF4) and hexafluoroethane (C2F6) from East Asia: 2008 to 2019, Journal of Geophysical Research: Atmospheres, 126, e2021JD034888, 2021.

Li, S., Kim, J., Kim, K.-R., Mühle, J., Kim, S.-K., Park, M.-K., Stohl, A., Kang, D.-J., Arnold, T., and Harth, C. M.: Emissions of halogenated compounds in East Asia determined from measurements at Jeju Island, Korea, Environmental science & technology, 45, 5668-5675, 2011.

Park, S., Western, L. M., Saito, T., Redington, A. L., Henne, S., Fang, X., Prinn, R. G., Manning, A. J., Montzka, S. A., and Fraser, P. J.: A decline in emissions of CFC-11 and related chemicals from eastern China, Nature, 590, 433-437, 2021.

Rigby, M., Park, S., Saito, T., Western, L., Redington, A., Fang, X., Henne, S., Manning, A., Prinn, R., and Dutton, G.: Increase in CFC-11 emissions from eastern China based on atmospheric observations, Nature, 569, 546-550, 2019.

Simmonds, P. G., Rigby, M., McCulloch, A., Vollmer, M. K., Henne, S., Mühle, J., O'Doherty, S., Manning, A. J., Krummel, P. B., and Fraser, P. J.: Recent increases in the atmospheric growth rate and emissions of HFC-23 (CHF_3) and the link to HCFC-22 ($CHClF_2$) production, 2018.

Stanley, K. M., Say, D., Mühle, J., Harth, C. M., Krummel, P. B., Young, D., O’Doherty, S. J., Salameh, P. K., Simmonds, P. G., and Weiss, R. F.: Increase in global emissions of HFC-23 despite near-total expected reductions, Nature communications, 11, 1-6, 2020.

Stohl, A., Kim, J., Li, S., O'Doherty, S., Mühle, J., Salameh, P. K., Saito, T., Vollmer, M. K., Wan, D., and Weiss, R. F.: Hydrochlorofluorocarbon and hydrofluorocarbon emissions in East Asia determined by inverse modeling, Atmospheric Chemistry and Physics, 10, 3545-3560, 2010.

TEAP: Technology and Economic Assessment Panel of the Montreal Protocol: Assessment Report, United Nations Environment Programme, Nairobi, Kenya, 2017.

Warszawski, L., Frieler, K., Huber, V., Piontek, F., Serdeczny, O., Zhang, X., Tang, Q., Pan, M., Tang, Y., and Tang, Q.: Center for International Earth Science Information Network—CIESIN—Columbia University. (2016). Gridded population of the World, Version 4 (GPWv4): Population density. Palisades. NY: NASA Socioeconomic Data and Applications Center (SEDAC). doi: 10. 7927/H4NP22DQ, Atlas of Environmental Risks Facing China Under Climate Change, 228, 2017.

Yao, B., Vollmer, M., Zhou, L., Henne, S., Reimann, S., Li, P., Wenger, A., and Hill, M.: In-situ measurements of atmospheric hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) at the Shangdianzi regional background station, China, Atmospheric Chemistry and Physics, 12, 10181-10193, 2012.

Yokouchi, Y., Taguchi, S., Saito, T., Tohjima, Y., Tanimoto, H., and Mukai, H.: High frequency measurements of HFCs at a remote site in east Asia and their implications for Chinese emissions, Geophysical research letters, 33, 2006.