# Investigation of East Asian emissions of CFC-11 using atmospheric observations in Taiwan

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19 **ABSTRACT**

20 Recent findings of an unexpected slowdown in the decline of CFC-11 mixing ratios in the
21 atmosphere have led to the conclusion that global CFC-11 emissions have increased over the last
22 decade and have been attributed in part to eastern China. This study independently assesses these
23 findings by evaluating enhancements of CFC-11 mixing ratios in air samples collected in Taiwan
24 between 2014 and 2018. Using the NAME (Numerical Atmospheric Modelling Environment)
25 particle dispersion model we find the likely source of the enhanced CFC-11 observed in Taiwan
26 to be East China. Other halogenated trace gases were also measured and there were positive
27 interspecies correlations between CFC-11 and $CHCl_3$, $CCl_4$, HCFC-141b, HCFC-142b, $CH_2Cl_2$
28 and HCFC-22, indicating co-location of the emissions of these compounds. These correlations in
29 combination with published emission estimates of $CH_2Cl_2$ and HCFC-22 from China, and of
30 $CHCl_3$ and $CCl_4$ from eastern China, are used to estimate CFC-11 emissions. Within the
31 uncertainties, these estimates do not differ for eastern China and the whole of China, so we
32 combine them to derive a mean estimate which we refer to as being from '(eastern) China'. For
33 2014-2018 we estimate an emission of 19 ± 5 Gg $yr^{-1}$ (gigagrams per year) of CFC-11 from
34 (eastern) China, approximately one quarter of global emissions. Comparing this to previously
35 reported CFC-11 emissions estimated for earlier years we estimate CFC-11 emissions from
36 (eastern) China to have increased by 7 ± 5 Gg $yr^{-1}$ from the 2008-2011 average to the 2014-2018
37 average, which is 50 ± 40% of the estimated increase in global CFC-11 emissions and is consistent
38 with the emission increases attributed to this region in an earlier study.

## INTRODUCTION

CFC-11 (trichlorofluoromethane, $CCl_3F$) is presently the second most abundant chlorofluorocarbon in the atmosphere with average global mixing ratios of 231-234 parts per trillion (ppt) in 2018[1]. It is a long-lived ozone-depleting substance (atmospheric lifetime of 52 years) that is controlled under the Montreal Protocol on Substances that Deplete the Ozone Layer[2]. The Montreal Protocol phased out production and consumption of CFCs (including CFC-11) by 1996 in developed countries and by 2010 in developing countries, with a few 'essential' use exceptions[3]. CFC-11 was used primarily as a foam-blowing agent, as an aerosol propellant and as a refrigerant[4]. CFC-11 global emissions peaked at about 350 Gg $yr^{-1}$ in the late 1980s and its tropospheric mixing ratios peaked in the early 1990s at about 270 ppt, after which both began to decline[2].

Excluding 'essential' uses, assuming no new production, there should only be CFC-11 emissions from equipment and products filled with CFC-11 before the ban, referred to as a 'bank' e.g. foam cells in building insulation. CFC-11 emissions are expected to be slowly released from the bank and to decrease over time as the bank diminishes. However, a recent study found an unexpected slowdown in the rate of decline of CFC-11 mixing ratios and an increase in global CFC-11 emissions of 13 ± 5 Gg $yr^{-1}$ from 54 ± 3 Gg $yr^{-1}$ in 2002-2012 to 67 ± 3 Gg $yr^{-1}$ in 2014-2016[5]. Another study also recently found an increase in global CFC-11 emissions of 17 ± 3 Gg $yr^{-1}$ or 11 ± 3 Gg $yr^{-1}$ between 2008-2012 and 2014-2017[6].

There are multiple possible origins of these additional emissions: an increase in the emissions rate from CFC-11 banks; a change in exempt uses of CFC-11; changes in atmospheric dynamics; or

from illegal production. It is unlikely that there would be a large enough increase in emissions from banks[4, 5, 7] or exempt uses of CFC-11[4] to explain the change in CFC-11 emissions and changes in atmospheric dynamics can likely only explain part of the increase in emissions[5].

Therefore, it is likely that since at least 2012 there has been an additional source of CFC-11 from production not allowed under the Montreal Protocol. East Asia[5], specifically eastern mainland China[6], has been identified as a likely source of these new CFC-11 emissions. CFC-11 emissions from eastern mainland China were estimated to be 13.4 ± 1.7 Gg $yr^{-1}$ in 2014-2017, this is 7.0 ± 3.0 Gg $yr^{-1}$ higher than in 2008-2012[6]. The reasons for a potential increase in the illegal production of CFC-11 are a subject of speculation. It has been suggested that reduced availability of HCFC-141b and increased demand for foams in building insulation may have driven demand for new production of CFC-11 for rigid polyurethane foams[4, 8, 9]. During the foaming process for rigid foams approximately 4% (e.g. appliance foams) to 25% (e.g. spray foams) of the blowing agent is immediately released to the atmosphere[4, 8]. Therefore, if CFC-11 was being used for this then that may account for at least some of the recent increase in atmospheric levels of CFC-11. Furthermore, a large amount of the CFC-11 will remain in the foams, thereby increasing the size of the CFC-11 bank and the potential for further emissions of CFC-11 in the future[4, 10]. Continued emissions of the ozone depleting substance CFC-11 could undermine the success of the Montreal Protocol and delay the recovery of the ozone layer[2].

## METHODS

In this study we measured multiple halogenated organic trace gases, including CFC-11, in air samples collected in Taiwan, using gas chromatography coupled with mass spectrometry (GC-MS). Five ground-based air sampling campaigns took place in Taiwan from 2014 to 2018. Between 20 and 33 air samples were collected in the spring of each year (mostly March – April; including May and early June in 2017-18) with a total of 135 samples collected altogether (Table S1). In 2015, samples were collected from a site on the southern coast of Taiwan (Hengchun, 22.0547 °N, 120.6995 °E). In all other years samples were collected at the Cape Fuguei (CAFE) Research Station, operated by Academia Sinica, on the northern coast of Taiwan (25.297 °N, 121.538 °E). Both sampling sites are well located to study the East Asian outflow. During the springtime, Taiwan is typically influenced by strong continental outflow from East Asia, particularly from China[11, 12].

**Analytical technique**

Air samples were collected in 3-litre silco-treated stainless-steel canisters (Restek) using a small 12 VDC diaphragm pump (Air Dimensions, model B161). During sampling they were filled and vented at least 3 times before filling to a final pressure of ~2 bar which takes a few minutes. The samples were then transported to the University of East Anglia (UEA) and analysed for about 50 trace gases including CFC-11. The samples were analysed on an Agilent 6890 gas chromatograph coupled to a high-sensitivity Waters AutoSpec magnetic sector mass spectrometer (GC-MS) using an Agilent GS-GasPro column (length ~50 m; ID: 0.32mm). For more information see the supplement. The samples in 2014 were also measured on a second GC-MS system (Entech-Agilent GC–MS) operating in electron ionisation (EI) mode. This consists of a preconcentration unit

(Entech model 7100) connected to an Agilent 6890GC and 5973 quadrupole MS[13]. In this study the CFC-11, $CCl_4$, $CHCl_3$, HCFC-22, HCFC-141b, and HCFC-142b mixing ratios in 2014 come from the Entech GC-MS measurements as these compounds were not measured on the AutoSpec GC-MS in 2014. The mixing ratios in all other years (2015-2018) come from the measurements on the AutoSpec GC-MS. The $CH_2Cl_2$ mixing ratios come from measurements made on the AutoSpec GC-MS for all five years (2014-2018).

The samples were measured against several clean air standards filled and calibrated by the Global Monitoring Division (GMD) of the National Oceanic and Atmospheric Administration (NOAA) in Boulder, Colorado. Multiple internal comparisons carried out over more than 10 years ensured the reliability and accuracy of the mixing ratios of all trace gases reported here and previous comparisons with NOAA measurements have shown excellent agreement[14]. All CFC-11 results were transferred on to the recent NOAA 2016 GC-ECD calibration scale. The dry-air mole fraction in picomole per mole was measured, and we here report mixing ratios, in parts per trillion (ppt), as an equivalent to the dry-air mole fraction. The uncertainties are calculated the same way for all measurements and represent 1σ standard deviations. They are based on the square root of the sum of the squared uncertainties from sample repeats and repeated measurements of the air standard on the same day.

**Identification of CFC-11 source regions**

The history of air arriving at the sampling sites has been investigated with the Met Office's NAME (Numerical Atmospheric Modelling Environment) Lagrangian particle dispersion model[15].

These histories (hereafter footprints) were calculated by releasing batches of 30000 inert particles over a three-hour period encompassing the collection time of each sample. Over the course of the 12-day travel time, the locations of all particles within the lowest 100 m of the model atmosphere were recorded every 15 minutes on a grid with a resolution of 0.25° longitude and 0.25° latitude. The trajectories of the particles were calculated using three-dimensional meteorological fields produced by the Met Office's Numerical Weather Prediction tool, the Unified Model (UM). These fields have a horizontal grid resolution of 0.23° longitude by 0.16° latitude and 59 vertical levels below ~30 km.

In order to quantify the contribution of various geographical regions to each footprint, the domain was divided into 15 regions using shapefiles produced by ArcGIS, a geographic information system (GIS) (Figure S1). The 15 regions were determined by country boundaries and China was split into regions using province boundaries. The output underpinning the NAME footprints, a mass density residence time (g $m^{-3}$ s) in each model grid cell, is summed across all grid cells within each of these 15 regions. These regional quantities are used to assess the possible relationships between emissions from specific regions and the mixing ratios of CFC-11 observed in Taiwan.

Additionally, the NAME footprints were combined with emission inventories of carbon monoxide (CO) taken from the Representative Concentration Pathway 8.5 (RCP 8.5)[16] for the year 2010 to generate modelled CO mixing ratios at Taiwan resulting only from emissions occurring within the 12-day timescale of the NAME trajectories[12, 17]. The RCP uses decade long averages and 2010 is used as it is the closest to the years of the campaigns in Taiwan. CO is a tracer of anthropogenic emissions and in this study the modelled CO is divided into various anthropogenic

emission sectors e.g. 'industry (combustion and processing)' and 'residential and commercial'. The correlations between the CFC-11 mixing ratios in Taiwan and the modelled CO from the emission sectors in East Asia were then calculated to investigate the spatial distribution of CFC-11 emissions.

**Correlations of CFC-11 with other trace gases**

The relationship between mixing ratios of CFC-11 and other halocarbons were investigated using the Spearman's rank correlation coefficient (R). Spearman's was selected as these data are not normally distributed with a few samples having particularly high halocarbon mixing ratios, including those of CFC-11. Spearman's method gave slightly lower correlation coefficients for these data than the Pearson's method. The significance of the correlations were tested using a two-tailed Student's t-distribution. The background mixing ratios for the months of the campaign were subtracted from each year to account for any long-term trends. For CFC-11, CFC-12 and $CCl_4$ the NOAA Northern Hemisphere background was used (https://www.esrl.noaa.gov/gmd/dv/ftpdata.html). For $CH_2Cl_2$ and $CHCl_3$ NOAA does not provide background values so the 10th percentile of our measurements for each year were used. To calculate the interspecies ratios the enhancements of CFC-11 above its background were plotted against the enhancements of each compound above their respective backgrounds. The slopes were calculated by total least squares regression using the York-Williamson method to account for uncertainties in mixing ratios of both species[18]. These slopes were then used to estimate CFC-11 emissions[19, 20].

**Estimation of CFC-11 emissions from China**

Similar to the approach used in some previous studies of halocarbon emissions from China[19, 20], we estimated emissions of CFC-11 using the slope of CFC-11 mixing ratio enhancements against those of other compounds which had a good correlation with CFC-11 and had published emissions. The compounds chosen were $CCl_4$, $CHCl_3$, $CH_2Cl_2$ and HCFC-22. Equations (1) and (2) were used to calculate emissions of CFC-11 and their uncertainties.

$$E_{CFC-11} = S \; E_x \frac{M_{CFC-11}}{M_x} \quad (1)$$

$$\sigma_{E_{CFC-11}} = E_{CFC-11} \sqrt{\frac{{\sigma_S}^2}{S} + \frac{{\sigma_{E_x}}^2}{E_x}} \quad (2)$$

$E_{CFC-11}$ and $E_x$ represent emissions of CFC-11 and halocarbon $x$ respectively; $M_{CFC-11}$ and $M_x$ represent the molecular weights of CFC-11 and halocarbon $x$ respectively; and $S$ represents the slope of the correlation. $\sigma_{E_{CFC-11}}$ is the uncertainty in the CFC-11 emissions; $\sigma_S$ is the uncertainty in the slope of the correlation; and $\sigma_{E_x}$ is the uncertainty in the emissions of halocarbon $x$.

The $CCl_4$ emissions used in this study were calculated by Lunt et al.[21] for eastern China in 2009-2016 using a top-down approach with atmospheric measurements from Gosan, South Korea, and two atmospheric inversion models, NAME: 17 (11-24) Gg $yr^{-1}$ and FLEXPART: 13 (7-19) Gg $yr^{-1}$. The $CHCl_3$ emissions used in this study were calculated by Fang et al.[22] for eastern China in 2015 using measurements from Gosan and from Hateruma, Japan and the same two atmospheric inversion models, NAME: 82 (70-101) Gg $yr^{-1}$, FLEXPART: 88 (80-95) Gg $yr^{-1}$. The HCFC-22 emissions used were taken from Li et al.[23], who calculated 134 (100-167) Gg $yr^{-1}$ for China in 2016 using an emission-factor based (bottom-up) method. Two reported emission estimates for

200 $CH_2Cl_2$ were used: bottom-up emissions in China of 318 (254-384) Gg $yr^{-1}$ for 2016 were
201 calculated by Feng et al.[24] based on a survey of known consumption and emission factors in
202 industrial sub-sectors; and 455 (410-501) Gg $yr^{-1}$ (2016) were calculated by Oram et al.[17], based
203 on chlorocarbon production and sales information for 2015. The main difference between these
204 two estimates is the amount of $CH_2Cl_2$ produced. Oram et al.[17] estimated Chinese $CH_2Cl_2$
205 production to be 715 Gg using the reported production of HCFC-22, whilst Feng et al.[24] estimated
206 600 Gg of $CH_2Cl_2$ production, based on surveys in the Chinese chloro-alkali industry.
207
208 **Estimation of changes in CFC-11 emissions from China**
209
210 One key question is whether CFC-11 emissions from China have increased in recent times and,
211 if so, by how much. The Taiwan measurements only cover the period 2014-2018 and so to look at
212 CFC-11 emissions in China over a longer period of time, back to 2008, we compared the emissions
213 derived here with previous studies.[6, 19, 20, 25-28] There are some differences in the methods used in
214 these studies to calculate the emissions. All emission estimates from these studies are top-down
215 except those from Wan et al.[25] and Fang et al.[28] which are bottom-up estimates. Wan et al.[25], Fang
216 et al.[19], Wang et al.[20] and Fang et al.[28] are emission estimates for the whole of China. Those from
217 Kim et al.[26], An et al.[27] and Rigby et al.[6] are for eastern China. We have included all these estimates
218 accepting that we are not always comparing like with like as there was no clear difference between
219 CFC-11 emission estimates for the whole of China and eastern China (Figure 3). Also, we decided
220 to include as many studies as possible to increase the confidence in our estimate and in-order to
221 show the possible uncertainty. For further information see Table S2.
222
223 **RESULTS AND DISCUSSION**
224
225 **CFC-11 mixing ratios in Taiwan**

Across all five years the CFC-11 mixing ratios in Taiwan range from 226 ppt to 272 ppt (Figure 1, Table S1). They are on average 3% higher than the northern hemispheric background mixing ratios as represented by Mauna Loa, Hawaii. Some of the measurements are consistent with the background, while many, especially those in years 2016-2018, contain higher mixing ratios than those observed at Mauna Loa implying that CFC-11 is enhanced on a regional scale (Figure 1). Samples with particularly high CFC-11 mixing ratios provide observational evidence of CFC-11 emissions from relatively nearby sources.

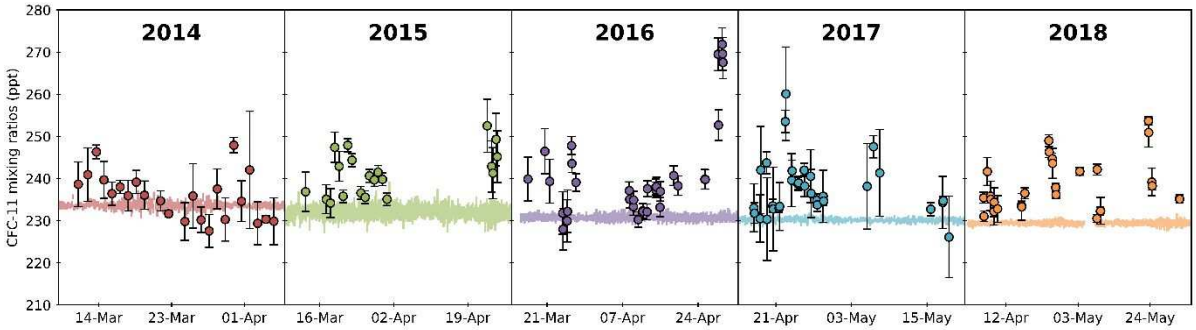



**Figure 1.** CFC-11 mixing ratios in Taiwan 2014-2018. The measurement campaigns lasted for 1-3 months each year. Uncertainties represented by the error bars are described in the text. Hourly in situ measurements of CFC-11 mixing ratios at Mauna Loa, Hawaii from the NOAA/ESRL Global Monitoring Division are included for comparison (ftp://ftp.cmdl.noaa.gov/hats/cfcs/cfc11/insituGCs/CATS/hourly/). The standard deviation error bars of the Mauna Loa measurements are plotted in the same color as the data.

## CFC-11 source regions

For all years combined, the strongest positive correlation is between CFC-11 mixing ratios and contributions to the NAME footprints from the East China region, with a Spearmans correlation

247 coefficient of R = 0.495, p<0.01 (Figure S2). All other regions have a correlation with CFC-11
248 mixing ratios of R < 0.3. East China includes major industrialized areas such as the Yangtze River
249 Delta that have previously been identified as the source region of other chloromethanes: methyl
250 chloride[29] and carbon tetrachloride[21]. Rigby et al.[6] also focused on eastern mainland China but
251 they identified the Shandong and Hebei provinces as the main source of CFC-11 emissions.
252 Shandong is part of our East China region, but Hebei is slightly further north than our East China
253 region.
254
255 While our analysis highlights East China as a potentially important source region for CFC-11 in
256 East Asia, it is possible that other important emission regions exist but have less influence on the
257 observations in Taiwan. Monthly average NAME footprints were used to investigate typical air
258 transport during the sampling period. In the spring air generally travels eastwards across the
259 northern half of China and then curves southwards towards Taiwan[11, 12]. Taiwan is an island and
260 the measurement sites are on the coast, so based on the mass density residence times (g $m^{-3}$ s) of
261 the 12-day NAME footprints, most of the influence on air samples (on average about two-thirds)
262 is from ocean regions: i.e. East China Sea, Pacific Ocean and the South China Sea. When
263 comparing only the land-based source regions, East China and North China typically contributed
264 the most to air sampled in Taiwan. Other potential source regions had much less of an influence
265 on the samples collected in Taiwan, each contributing to about 1-4% of the air in Taiwan based on
266 the mass density residence times. Therefore, CFC-11 emissions from other source regions will
267 have had a small impact on the air samples collected in Taiwan during the times of year of the
268 present study.
269

For all years combined the correlations (Spearmans $p<0.01$) between CFC-11 mixing ratios in Taiwan and modelled CO mixing ratios from a range of sources were found to be very similar: agricultural waste burning on fields ($R = 0.545$); residential and commercial sector ($R = 0.491$); solvent sector ($R = 0.483$); and industry (combustion and processing) ($R = 0.469$) (Figure S3). The correlation between CFC-11 and power plants, energy conversion and extraction was somewhat lower ($R = 0.384$, $p<0.01$). CO tracers that we do not find to significantly correlate ($R < 0.25$) with CFC-11 are waste (landfills, waste water, incineration), forest burning, grassland burning, international shipping, surface transportation, agriculture (animals, rice and soil) and aviation.

Some of the CO emission sectors most likely have very similar correlations because they are generally co-located with each other, so it is not possible to discriminate between the different sources (Figure S4). These sources are predominantly in eastern China, between Shanghai and Beijing, similar to the area identified by Rigby et al.[6] as a major source of CFC-11 emissions.

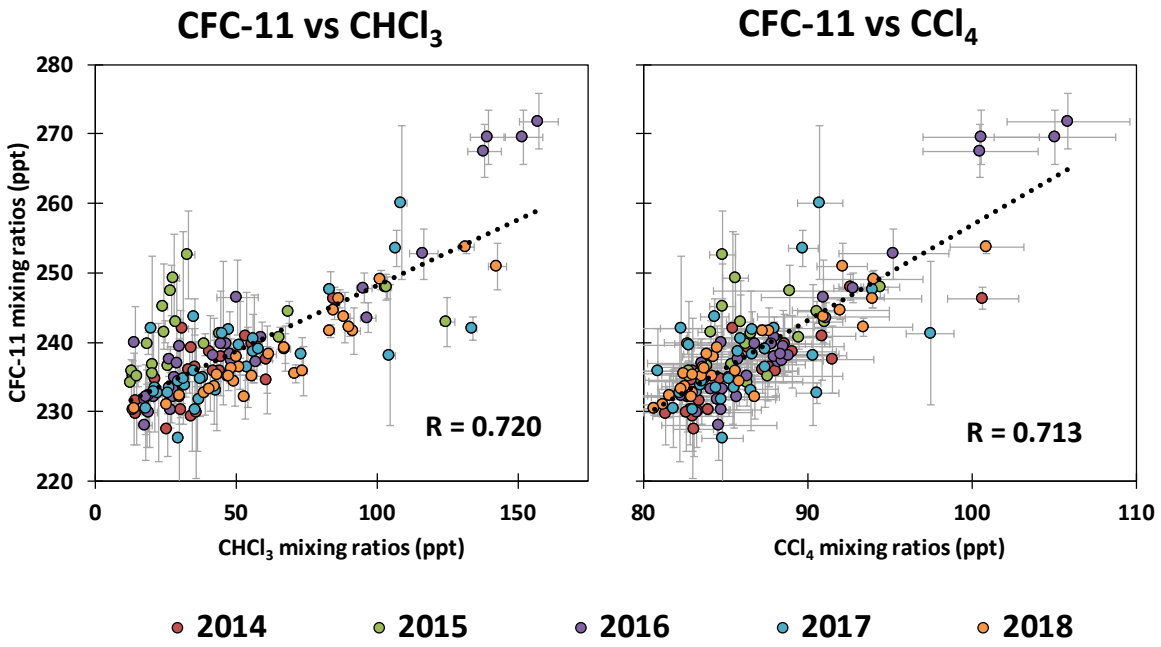



**Figure 2.** Interspecies correlations of CFC-11 mixing ratios with those of $CCl_4$ and $CHCl_3$. For other interspecies correlations with CFC-11 see Figure S5. The dashed line is the trend line calculated by total least squares regression using the York-Williamson method.

## Correlations of CFC-11 with other trace gases

The strongest positive correlations (Spearmans $p<0.01$) between CFC-11 and other halocarbons when measurements from all years are combined are: $CHCl_3$ ($R = 0.720$), $CCl_4$ ($R = 0.713$), HCFC-141b ($C_2H_3Cl_2F$) ($R = 0.671$), HCFC-142b ($CH_3CClF_2$) ($R = 0.667$), $CH_2Cl_2$ ($R = 0.622$) and HCFC-22 ($CHClF_2$) ($R = 0.593$) (Figure 2, Figure S5). These correlations concur with a previous study that also found correlations of HCFC-22 and $CH_2Cl_2$ with CFC-11 in measurements from Hawaii when air masses originated from East Asia.[5]

Compounds generally have correlated mixing ratios in the atmosphere when their emissions are released from a similar location or when atmospheric concentration gradients are present (vertically or horizontally) that are sampled by different wind patterns. CFC-11 emissions are probably found in many locations. The emissions of CFC-11 and other compounds from a production facility are likely to be low as it is not economically viable for a production facility to release their products into the atmosphere[4, 10]. If CFC-11 is used as a foam blowing agent then about 4% (e.g. appliance foams) to 25% (e.g. spray foams) of the CFC-11 emissions would be released from the foam blowing stage when the foam is made[4, 8]. The rest of the CFC-11 emissions would be gradually released from foam degradation or when the foam is broken up e.g. during demolition of buildings[7, 8].

CFC-11 has historically been widely used in polyurethane foam applications[8]. As CFC-11 was phased out HCFC-141b became commonly used as a replacement[4, 8]. HCFC-142b and HCFC-22 are also used in the foam blowing industry in extruded polystyrene production[8]. The correlations between these compounds may be related to them all being used as foam blowing agents in building insulation and co-location of built environments and foam-blowing facilities.

The other compounds that CFC-11 has a good correlation with, $CHCl_3$, $CH_2Cl_2$, $CCl_4$ and HCFC-22, are all involved in the same production chain. $CHCl_3$ and $CH_2Cl_2$ are co-produced through chlorination of methyl chloride ($CH_3Cl$) with a small amount of $CCl_4$ produced as a by-product[17]. Almost all the chloroform ($CHCl_3$) produced is then used as a feedstock in HCFC-22 production[4, 17]. China has a large chloromethanes industry and recent studies have found emissions of $CCl_4$[21], $CHCl_3$[22] and $CH_2Cl_2$[17] from eastern China. Most of the emissions will likely come from the applications of these compounds rather than from production facilities; therefore, these

correlations indicate co-location of the uses of CFC-11 and chloromethanes, possibly in urban areas.

CFC-11 has historically been produced via fluorination of $CCl_4$ to produce a mixture of CFC-11 ($CCl_3F$) and CFC-12 ($CCl_2F_2$)[4]. The production ratio has typically been between 30:70 and 70:30[4]. Therefore, if enhanced mixing ratios of CFC-12 were observed coincident with enhanced mixing ratios of CFC-11, this might suggest the cause of the increased CFC-11 emissions to be new production. However, we found no correlation between mixing ratios of CFC-11 and CFC-12 ($R = 0.285$) in the Taiwan measurements even after removing the decreasing background trend in CFC-12 to focus on enhancements in mixing ratios above the background. CFC-12 mixing ratios in the Taiwan air samples do not show any major enhancements and are similar to the levels at Mauna Loa, Hawaii (Figure S6). This agrees with findings in previous studies that also found no correlation between CFC-11 and CFC-12[5, 6] and adds to the evidence that emissions of CFC-11 from eastern China are not directly associated with emissions of CFC-12. Operating conditions could be controlling the relative proportions of CFC-12 and CFC-11; close to 100% CFC-11 production is difficult to achieve but not impossible[4]. Alternatively CFC-12 may still be being co-produced but is destroyed or used as a refrigerant, which is considered to be a non-emissive source as release of the CFC-12 will take place over a long period of time[4, 7].

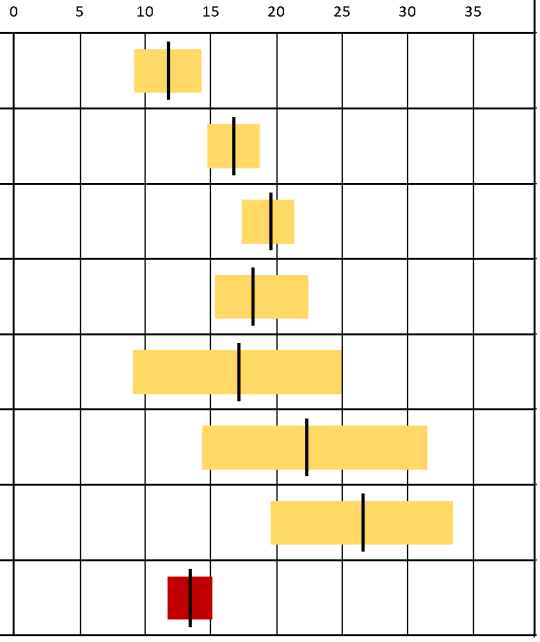
**CFC-11 emissions from China**

The CFC-11 emission estimates in this study are based on emission estimates for the whole of China and eastern China and therefore, when the CFC-11 emission estimates are combined together they are referred to as CFC-11 emissions from '(eastern) China'. CFC-11 emissions from (eastern) China for the period 2014-2018 are estimated based on interspecies correlations of CFC-

11 with other halocarbons for which we found good correlations. There is a large range in the estimates of CFC-11 emissions from (eastern) China derived during this study (Table 1). Most of the uncertainty in the CFC-11 emission estimates is due to the uncertainty in the emissions estimates of the other compounds rather than the uncertainty in the slope of the interspecies ratios. The lowest estimate is 12 (9-14) Gg $yr^{-1}$ using the Feng et al.[24] estimate of $CH_2Cl_2$ emissions. The largest CFC-11 emission estimate is 27 (20-33) Gg $yr^{-1}$, based on HCFC-22 from Li et al.[23]. The two compounds with the strongest correlations with CFC-11 are $CHCl_3$ and $CCl_4$ (Table S3) and the estimates derived from these are in the middle of the range (17-22 Gg $yr^{-1}$) (Table 1). It is important to note that the HCFC-22 and $CH_2Cl_2$ based emissions estimates are for the whole of China, whilst the $CHCl_3$ and $CCl_4$ based emissions are for eastern China only. There is no consistent pattern of higher emissions for the whole of China and lower emissions for eastern China (Table 1). The mean of all the individual estimates is 19 (14-23) Gg $yr^{-1}$. The uncertainties were calculated as the standard deviation of the individual estimates.

**Table 1.** CFC-11 emission estimates and the upper and lower limits based on the interspecies ratios with $CHCl_3$, $CH_2Cl_2$, $CCl_4$ and HCFC-22 in our Taiwan measurements from 2014-2018 and published halocarbon emission estimates. The vertical lines indicate the best estimate. Recently published CFC-11 emission estimates for eastern mainland China are also shown[6]. The emission estimates from this study are colored yellow and the emission estimate from Rigby et al.[6] is red.

| Compound | Location and Years of Emissions[1] | CFC-11 emissions (Gg yr$^{-1}$) |
|---|---|---|
| $CH_2Cl_2$ Feng et al. (2018) | China 2016 | 12 (9-14) |
| $CH_2Cl_2$ Oram et al. (2017) | China 2015 | 17 (15-19) |
| $CHCl_3$ FLEXPART, Fang et al. (2019) | Eastern China 2015 | 19 (17-21) |
| $CHCl_3$ NAME, Fang et al. (2019) | Eastern China 2015 | 18 (15-22) |
| $CCl_4$ FLEXPART, Lunt et al. (2018) | Eastern China 2009-2016 | 17 (9-25) |
| $CCl_4$ NAME, Lunt et al. (2018) | Eastern China 2009-2016 | 22 (14-31) |
| HCFC-22 Li et al. (2016) | China 2016 | 27 (20-33) |
| Rigby et al. (2019) | Eastern China 2014-2017 | 13.4 (11.7-15.1) |




[1] For the estimates from the current study based on interspecies correlations the location and years of emissions are based on the region for the emission estimate of compound x.

Another recent study used CFC-11 measurements at Gosan, Jeju Island, Korea and Hateruma, Japan and two atmospheric inversion models to calculate CFC-11 emissions from eastern mainland China to be 13.4 ± 1.7 Gg yr$^{-1}$ in 2014-2017[6]. These estimates are at the lower end of the range that we calculate here based on the Taiwan samples. The reason for this might be that the Rigby et al.[6] estimates are confined to provinces to which the measurements were most sensitive. Rigby et al.[6] mentioned that including the provinces adjacent to their 'eastern mainland China' region increased their emissions by 15%. The $CHCl_3$ and $CCl_4$ emissions that we use in our calculations[21, 22] are based on observations from the same measurement sites as Rigby et al.[6]. These studies[21, 22] derived emissions for 'eastern China', but a slightly larger area to that used by Rigby et al.[6].

385
386 The region of our CFC-11 emission estimate is defined by the region of the emission estimate
387 for the other compound used in the interspecies ratio (Table 1). So whilst our estimates based on
388 $CHCl_3$ and $CCl_4$ emissions are largely restricted to eastern China, those using HCFC-22 and
389 $CH_2Cl_2$ emission estimates are for the whole of China[17, 23, 24]. It is assumed that the emission ratio
390 of CFC-11 to the other compounds is the same in all regions. Our overall emission estimate (19
391 (14-23) Gg $yr^{-1}$) based on the Taiwan measurements is a combination of estimates for eastern
392 China and the whole of China.
393
394

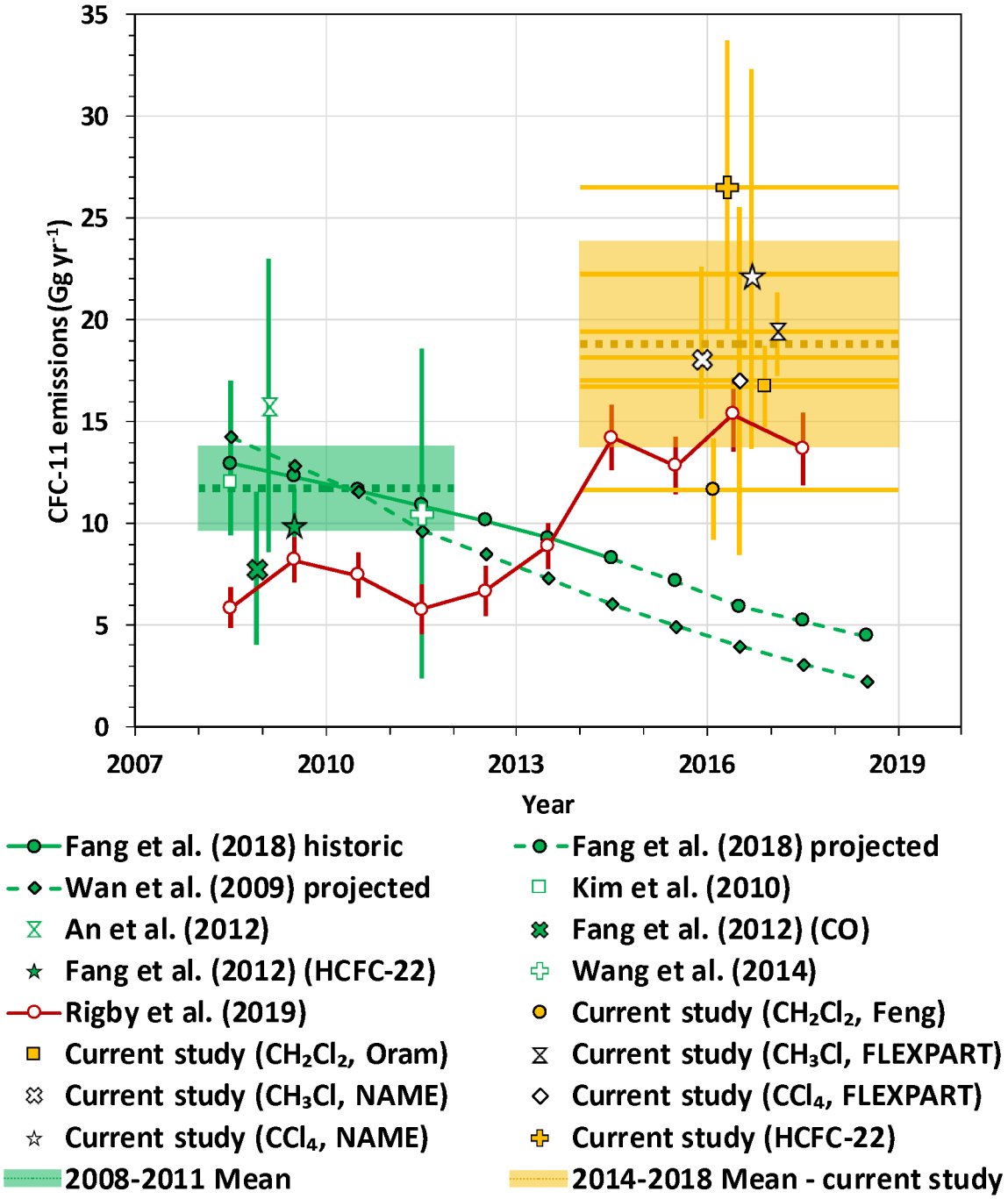



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**Figure 3.** CFC-11 emissions in China and eastern China.[5, 6, 19, 20, 25-28] Data that cover means of several years have horizontal error bars to indicate the periods that they relate to. Vertical error bars show the uncertainties in the emission estimates. Data from the same years are offset slightly so that the error bars are visible. The data points joined by dashed lines are projections. The 2008-2011 mean is the mean of the estimates in Wan et al.[25], Kim et al.[26], An et al.[27], Fang et al.[19], Wang et al.[20] and Fang et al.[28]. The 2014-2018 mean is the mean of the seven interspecies correlation estimates in this study. Emissions for the whole of China have filled data points and emissions for eastern China have clear data points.

## Changes in CFC-11 emissions from China

There is variation in the CFC-11 emission estimates between different studies, but the combined evidence suggests an increase in CFC-11 emissions in (eastern) China from 2008-2011 to 2014-2018. Rigby et al.[6] emission estimates for 2008-11 are lower than the other estimates for this period. Possibly because Rigby et al.[6] estimates are limited to 'eastern mainland China' whereas some of the other estimates cover larger areas (see above). Averaging published emission estimates for 2008-2011, and excluding Rigby et al.[6] gives CFC-11 emissions of 12 (10-14) Gg $yr^{-1}$ (green bar, Figure 3)[19, 20, 25-28]. The uncertainties were calculated using the standard deviation of the individual estimates.

CFC-11 emissions from (eastern) China for the period 2014-2018 are estimated in this study to be 19 (14-23) Gg $yr^{-1}$ (golden bar, Figure 3) by combining the seven interspecies correlation emission estimates (Table 1). This gives an increase of 7 (2-12) Gg $yr^{-1}$ since 2008-2011. The uncertainties are the square root of the sum of the uncertainties for each time period squared.

425 This increase in emissions between 2008-2011 and 2014-2018 that we estimate is similar to the
426 increase estimated by Rigby et al.[6] of 7.0 ± 3.0 Gg $yr^{-1}$ between 2008-2012 and 2014-2017. If we
427 include the Rigby et al.[6] emissions in our analysis, this gives us slightly lower CFC-11 emissions
428 for both the earlier and later time periods, as the Rigby et al.[6] estimates are generally lower than
429 the other estimates we use in our study (Table S2). Including the Rigby et al.[6] estimates in our
430 averages still gives an absolute increase of ~ 7 Gg $yr^{-1}$ in CFC-11 emissions(Table S2). The
431 consistency between our results and those of Rigby et al.[6], obtained by different, independent
432 methods, provides some confidence in this estimated size of the CFC-11 emissions in eastern
433 China, although it is recognized that both estimates have uncertainty in them.
434
435 In our study, the emission estimates are based on measurements of samples collected during
436 springtime each year when Taiwan is consistently impacted by air masses transported from
437 mainland China. This minimises dilution so the observed interspecies concentration ratios will
438 better reflect their emission ratios. Seasonal variations in emission ratios in this region are not well
439 constrained. Kim et al.[26] in their estimates of halocarbons emissions from China, assumed
440 emissions were constant throughout the year. However, their observed ratio between CFC-12 and
441 HCFC-22 enhancements suggest higher values in the summer. Seasonally varying ratios of
442 halocarbon enhancements were also observed in the US in the 1990s[30]. Limited seasonal sampling
443 will therefore introduce some error into our analysis but by using interspecies ratios of CFC-11
444 with four different halocarbons, we aim to reduce this error. Interspecies emission ratios may also
445 vary with location and this approach assumes sources to be perfectly co-located, which is unlikely.
446 This is partly accounted for in the uncertainty of the observed interspecies ratios. Additionally, we
447 are combining emission estimates from multiple studies that used different methods and are for

different time periods and regions. Therefore, our CFC-11 emission estimates will have some unaccounted for uncertainties.

CFC-11 emissions were expected to have decreased since 2012, due to the diminishing size of the banks, assuming compliance with the Montreal Protocol. This means the difference between projected bottom-up emissions and actual emissions may be larger than the increase in CFC-11 emissions from 2008-2011 to 2014-2018[2, 5, 6]. Previous studies projected future CFC-11 Chinese emissions using bottom-up estimates of reported production, estimates of the size of the CFC-11 bank and assumed emission rates[25, 28]. These bottom-up estimates agree with the top-down estimates in 2008-2011 but decrease such that they disagree in 2014-2018 with the top-down estimates (Figure 3). Averaging the estimates for the individual years between 2014 and 2018 from Wan et al.[25] and Fang et al.[28] gives 5 (3-7) Gg $yr^{-1}$. The uncertainties are the standard deviation of the estimates for the individual years. If we subtract 5 (3-7) Gg $yr^{-1}$ from our estimate of 19 (14-23) Gg $yr^{-1}$ this leads to 14 (9-19) Gg $yr^{-1}$ more emissions of CFC-11 in China than projected.

**Comparison to global CFC-11 emissions**

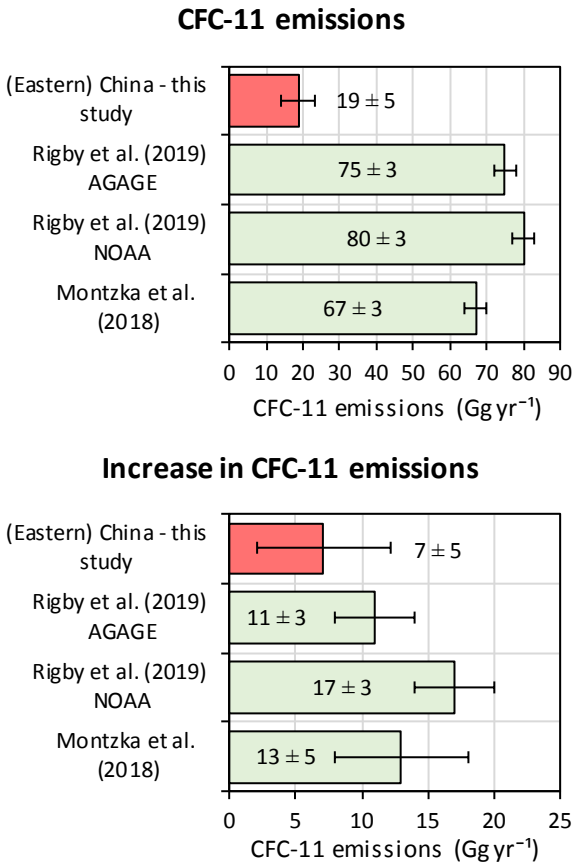


**Figure 4.** Top: Global CFC-11 emissions (green bars) compared to the (eastern) China emissions in this study (red bar). Bottom: Increase in global CFC-11 emissions (green bars) compared to the increase in (eastern) China emissions (red bar). These estimates are for slightly different time periods. The estimates from Montzka et al.[5] are for 2014-2016 compared to the 2002-2012. The Rigby et al.[6] estimates are for 2014-2017 compared to 2008-2012. This study's emission estimates are for 2014-2018 compared to 2008-2011.

Montzka et al.[5] used NOAA observations to calculate global CFC-11 emissions of 67 ± 3 Gg $yr^{-1}$ in 2014–2016 which was an increase of 13 ± 5 Gg $yr^{-1}$ above the 2002-2012 mean. Rigby et al.[6] calculated global CFC-11 emissions in 2014-2017 to be 80 ± 3 Gg $yr^{-1}$ based on NOAA observations and 75 ± 3 Gg $yr^{-1}$ based on AGAGE observations. These are increases since 2008-2012 of 17 ± 3 Gg $yr^{-1}$ (NOAA) and 11 ± 3 Gg $yr^{-1}$ (AGAGE). The NOAA-derived CFC-11

483 emissions in Rigby et al.[6] differ from the NOAA-derived Montzka et al.[5] CFC-11 emissions
484 because Rigby et al.[6] includes an additional year (2017) and uses a shorter atmospheric lifetime
485 for CFC-11. The atmospheric lifetime contributes to the differences in emission estimates given
486 in the different studies but has very little effect on the change in emissions over the short time
487 period.
488
489 In the section above, (eastern) China emissions were estimated to be 19 (14-23) Gg $yr^{-1}$ in 2014-
490 2018 and the increase estimated to be 7 (2-12) Gg $yr^{-1}$ compared to 2008-2011. These regional
491 emissions are 25% (19%-32%) of the global emissions (Figure 4). This is the proportion of our
492 (eastern) China emissions compared to the average of the three global estimates. The uncertainties
493 are based on the square root of the sum of squares of the uncertainty in our (eastern) China estimate
494 and the standard deviation of the three global estimates. The increase in (eastern) China CFC-11
495 emissions are a large proportion of the increase in global CFC-11 emissions but are also highly
496 uncertain (Figure 4). They are 52% (13%-91%) of the increase in global emissions (Figure 4).
497 Where the remaining CFC-11 emissions are coming from is not well known as there are many
498 parts of the world that are not well covered by the global networks, including South America,
499 Africa and the rest of Asia[7]. Note these CFC-11 emission estimates do not consider possible
500 changes in atmospheric dynamics which could slow-down the rate of decline in background CFC-
501 11 mixing ratios and lead to an over estimation of global CFC-11 emissions[5]. Large-scale changes
502 in atmospheric dynamics are likely to have less impact on regional emission estimates than global
503 ones.
504
505

Overall, the current study finds independent evidence of continuing and significant CFC-11 emissions from China, in particular from eastern China. Comparing with past studies this implies a recent increase in (eastern) China CFC-11 emissions, which will have contributed to a substantial proportion of the increase in global CFC-11 emissions. However, the extent of this contribution still has considerable uncertainty and further investigation is needed to better understand the recent changes in CFC-11 emissions.

ASSOCIATED CONTENT

**Supporting Information**

Excel spreadsheet containing mixing ratios and the influence of source regions and emission sectors.

Additional information about the sampling technique and the GC-MS analysis.

The dates and locations of the Taiwan measurement campaigns and the number of samples collected each year.

The previously reported emission estimates used in this study and the methods used to calculate them.

The interspecies correlations and regressions.

Regions for which the contribution to the footprint simulated by the NAME model is quantified.

The distribution of carbon monoxide (CO) emissions taken from the Representative Concentration Pathway 8.5 (2010) inventories.

Figures showing the correlations between CFC-11 mixing ratios and the influence from East China, the CO emission sectors and the mixing ratios of the other compounds of interest.

CFC-12 mixing ratios measured in Taiwan.

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534 **Notes**

535 The authors declare no competing financial interest.

536

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