# Banked CFC-11 contributes to an unforeseen emission rise and sets back progress towards carbon neutrality

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**Abstract.** An unexpected rise of trichlorofluoromethane (CFC-11) emissions has undermined the efforts behind the Montreal Protocol. However, the sources of these increased emissions, from CFC-11 banks to unreported production, remain contentious. Here, we enhanced the bottom-up dynamic material flow analysis model to characterize the stocks and flows of CFC-11, retrospectively and prospectively from 1950 to 2100. We find that dynamic changes in bank-related emissions could have led to an increased CFC-11 emissions from 2014 to 2018, implying an overestimation of unreported production. Under Scenario 2 (mid-range unreported production levels), long-term emission of banked CFC-11 will accumulate to 980 (600–1500) kilotons (kt), equivalent to 4.7 (2.9–7.1) gigatons (Gt) $CO_2$, between 2025 and 2100. Scenario analysis highlights the potential to reduce up to 50% of emissions through optimized end-of-life (EoL) management strategies. Our results call for further investigation into the lifespan and EoL processes of products containing ozone-depleting substances (ODSs) to reconcile emission estimates derived from bottom-up and top-down modeling approaches. The modeling approach could also be applied to estimate and project the bank-related emissions and impacts of other ODSs.

## 1 Introduction

Chlorofluorocarbons (CFCs) are major contributors to ozone depletion and potent greenhouse gases, which trap heat approximately 4,750-13,900 times more effectively than carbon dioxide over a 100-year horizon (Duan et al., 2018; Liu et al., 2024). The Montreal Protocol on Substances that Deplete the Ozone Layer has been a key global effort in phasing out ozone-depleting substances (ODSs), including trichlorofluoromethane (CFC-11; Chipperfield et al., 2020; Pyle et al., 2022; Young et al., 2021). The successful elimination of CFC-11 production has led to a decreasing of its atmospheric concentrations (Lickley et al., 2022; WMO, 2023). However, despite the complete global phaseout of CFC-11 by 2010, emissions unexpectedly increased during 2014–2018 (Montzka et al., 2018; Rigby et al., 2019). It is estimated that 40%–60% of this rise could be attributed to China, but the limited number of monitoring stations hindered the ability to trace the remainder to other regions (Rigby et al., 2019). Furthermore, several developed regions have exhibited elevated CFC-11 emissions after the complete phase-out of production and consumption, with no evidence of unreported production, raising unresolved questions about their sources (Dunse et al., 2019; Redington et al., 2023; Table S1). Clarifying these sources is therefore critical for addressing the emission increase and for projecting future CFC-11 emissions and their impacts on ozone depletion and climate change.

To date, studies have employed top-down and bottom-up material flow analysis (MFA) models to estimate CFC-11 emissions (Table S1). Top-down models rely on atmospheric measurements to infer emissions, though regional emission estimates are limited due to the sparsity of the monitoring networks (Montzka et al., 2021; Park et al., 2021; WMO, 2021). By contrast, bottom-up models rely on reported CFC production and consumption data, and use emission factors (EFs) associated with various CFC-containing products' life stages to estimate emissions (Flerlage et al., 2021; TEAP, 2019; 2021). These models have large uncertainties, due to uncertainties in the completeness of CFC-11 production and consumption reporting, the lifespans of the host products, EFs in various life stages, and end-of-life (EoL) management (TEAP, 2019; 2021). The Montreal Protocol's Technology and Economic Assessment Panel (TEAP) highlighted the major uncertainties in a bottom-up model by evaluating of the variations in production levels, EFs and static lifespans (TEAP, 2019; 2021). Nonetheless, this analysis did not adequately consider the variabilities in lifespans of foam products and their EoL management. For instance, polyurethane rigid (PUR) boardstock and panel foams for building and construction

applications, which used CFC-11 as a blowing agent, were typically assigned lifespan estimates of 25 years (McCulloch et al., 2001; IPCC, 2006), 50 years (Ashford et al., 2004; UNEP, 2002), or 75 years (UNEP, 2021; Table S2). These default values may be obtained from simple assumptions or relevant structural calculation specifications. However, surveys reveal significant temporal and spatial variability in the actual lifespans of buildings (Aktas et al., 2012; Liu et al., 2019; Tsutsumi et al., 2004; Table S3). Furthermore, high levels of uncertainty persist regarding CFC-11 emissions associated with EoL handling of foam products (IPCC, 2005; McCulloch et al., 2001; UNEP, 2021; Table S4). These factors can significantly influence the trends in the release of blowing agents from obsoleted foam products (Liu et al., 2024), a point often overlooked in bottom-up estimates. These gaps necessitate a reassessment of the timing and magnitude of bank-related CFC-11 emissions and their time-lagged impact.

Here, we propose a dynamic MFA (D-MFA) model to characterize CFC-11 emissions from 1950 to 2100 to address these gaps. We incorporate a wide range of uncertainty in product lifespans, EoL handling processes and associated emission factors, and temporal and spatial evolution. This model aims to estimate bank-related emissions and explain the unexpected emission rise observed between 2014 and 2018. Products that were produced in the past and still contain CFC-11 are said to be banks of potential future emissions. By considering both banks and unreported production, we project time-lagged CFC-11 emissions and their climate impacts across different sectors, regions, and lifecycle stages. Finally, we discuss the potential mitigation measures to prevent future emissions.

## 2 Methods

### 2.1 Dynamic material flow analysis model

The end-use applications of CFC-11 are in non-hermetic refrigeration systems, closed-cell foams (CCFs), and prompt emissive uses, such as aerosol propellants, open-cell foams, solvents, and others (TEAP, 2019; 2021). CFC-11-containing CCF products had been applied across a range of insulation applications, mainly including polyurethane rigid (PUR) boardstock, panel, spray, appliance and other insulation products. PUR boardstock foams have been used extensively in residential and commercial roof insulation and walls of metal buildings and agricultural buildings, and PUR panel foams have been used in industrial settings, such as refrigerated warehouses (S1.2). The investigated lifespan of these foam products in building contexts exhibits notable spatiotemporal variability (Table S3).

91     A D-MFA model is used for each end-use application. In this model, the calculation process for

92     CFC-11 emissions is methodically divided into distinct stages, which represent different phases of

93     products containing CFC-11 throughout their lifecycle, from manufacture, use to EoL handling. The first

94     stage involves the quantification of first-fill emissions during the manufacturing stage. Subsequently,

95     emissions during product use are evaluated. The final stage pertains to the release of emissions from the

96     EoL handling of obsolete products, which may involve recovery, recycling, and disposal processes of

97     CFC-11 contained within them.

98     End-use products, such as insulation foams for household refrigerator appliances or constructions,

99     exhibit durability and long lifespans. In this study, the Weibull distribution, which had been widely used

100     in previous research (Liu et al., 2019; 2024; UNEP, 2021), is introduced to simulate the survival rate

101     curves for new products with long lifespans (Eq. 1, Fig. S13).

$$f(y, t) = exp\left\{-\left(\frac{t + 0.5}{u}\right)^{\beta}\right\} \tag{1}$$

102     where $f(y, t)$ refers to the remaining rate of the sales of new products in year $y$; $t$ indicates the product

103     age; $\beta$ corresponds to the shape parameter $(\beta > 0)$; $u$ is the scale parameter ($u > 0$). Parameter estimates

104     for foam products and refrigeration equipment across different geographical regions were derived from

105     our surveys (Duan et al., 2018; Liu et al., 2024) and an extensive literature review (e.g., McCulloch et

106     al., 2001; FTOC, 2002; IPCC, 2006; TEAP, 2021). The quantity of CFC-11 mass in use or at EoL in each

107     end-use application within each geographical region can be determined using Eqs 2–5:

$$q_{use}(y, t) = q_{new}(y, 0) \times f(y, t) \tag{2}$$

$$q_{EoL}(y, t) = q_{use}(y, t) - q_{use}(y, t + 1) \tag{3}$$

$$Q_{use}(Y) = \sum_{y}^{Y} q_{use}(y, t = Y - y) \tag{4}$$

$$Q_{EoL}(Y) = \sum_{y}^{Y} q_{EoL}(y, t = Y - y) \tag{5}$$

108     where $Y$ represents the counted year of CFC-11 emissions; $y$ indicates the sales year of new product

109     containing CFC-11; $q_{new}(y, 0)$ refers to the sales data of CFC-11 in new products in year $y$; $q_{use}(y, t)$

110     denotes the quantity of in-use CFC-11 in the product in year $y$ with age $t$; $q_{EoL}(y, t)$ stands for the

111     quantity of CFC-11 in the product sold in year $y$ with age $t$ entering into the EoL stage; $Q_{use}(Y)$ means

112    the total quantity of in-use CFC-11 at year $Y$; $Q_{EoL}(Y)$ corresponds to the total quantity of CFC-11 in

113    EoL products in year $Y$.

114        The emission mass is derived based on the annual mass of products containing CFC-11 during the

115    first fill (manufacturing), use, and EoL stages. Following the Tier 2a method of the Intergovernmental

116    Panel on Climate Change Guidelines for National Greenhouse Gas Inventories (IPCC, 2006; 2019), the

117    historical and future emissions of CFC-11 from refrigeration systems can be estimated and projected

118    using Eq. 6.

$$E_{ref}(Y) = E_{man}(Y) + E_{use}(Y) + E_{EoL}(Y)$$
$$= Q_{new}(Y) \times Ef_{man}(Y) + Q_{use}(Y) \times Ef_{use}(Y) + Q_{EoL}(Y) \qquad (6)$$
$$\times (1 - \gamma_{EoL}(Y))$$

119    where $E_{ref}(Y)$ refers to the total emissions of CFC-11 from refrigeration systems in year $Y$; $E_{man}(Y)$,

120    $E_{use}(Y)$, and $E_{EoL}(Y)$ correspond to the mass emissions of CFC-11 from the manufacturing (first fill),

121    use, and EoL stages of refrigeration equipment, respectively, at year $Y$; $Q_{new}(Y)$ represents the sales of

122    CFC-11 in new equipment in year $Y$ and equals $q_{new}(y, 0)$ in Eq. (2) when $Y$ equals $y$. $Ef_{man}(Y)$ and

123    $Ef_{use}(Y)$ denote the EFs of CFC-11 during the life cycle stages of equipment manufacturing and use,

124    respectively; $\gamma_{EoL}(Y)$ indicates the recovery efficiency of CFC-11 during EoL handling for obsolete

125    equipment.

126        Similarly, the release of CFC-11 from CCF can occur throughout various life cycle stages, including

127    manufacture, use, EoL handling, and post-life, such as landfilling disposal. Eqs 1–5 are used to calculate

128    the CFC-11 bank and EoL flow from CCF. Eq. 7 is used to aggregate emissions across various life cycle

129    stages originating from CCF.

$$E_{ccf}(Y) = E_{man}(Y) + E_{use}(Y) + E_{EoL}(Y) + E_{post}(Y)$$
$$= Q_{new}(Y) \times Ef_{man}(Y) + Q_{use}(Y) \times Ef_{use}(Y) + Q_{EoL}(Y) \qquad (7)$$
$$\times Ef_{EoL}(Y) + Q_{EoL}(Y) \times l_{EoL}(Y) \times Ef_{post}(Y)$$

130    where $E_{ccf}(Y)$ indicates the total emissions of CFC-11 from an end-use application of CCF in year $Y$;

131    $E_{man}(Y)$, $E_{use}(Y)$, $E_{EoL}(Y)$, and $E_{post}(Y)$ denote the mass emissions of CFC-11 from the

132    manufacture, use, EoL handling, and post-life stages of CCFs, respectively, at year $Y$. $Q_{new}(Y)$, $Q_{use}(Y)$,

133    and $Q_{EoL}(Y)$ refer to the same representation as in Equations (2–6); $Ef_{man}(Y)$, $Ef_{use}(Y)$, $Ef_{EoL}(Y)$,

134    and $Ef_{post}(Y)$ are the EFs of CFC-11 during the life cycle stages of CCF manufacture, use, EoL

135    handling, and landfilling, respectively; $l_{EoL}(Y)$ means the landfilling proportion of CFC-11 during the

136 EoL handling of CCF, represented as $(100\% - Ef_{EoL}(Y))$.

137 The prompt emissive use category includes applications that use CFC-11 in processes that result in

138 immediate or relatively rapid release into the atmosphere (TEAP, 2019; 2021). A 6-month delay after

139 sale is conventionally assumed in the case of aerosol and solvent uses, which results in half of the CFC-

140 11 being released in the year of sale and the remaining half being released in the following year on

141 average (McCulloch et al., 2001). The emission calculations are performed using Eq. 8.

$$E_{emi}(Y) = p \times Q_{emi}(Y) + (1 - p) \times Q_{emi}(Y - 1) \tag{8}$$

142 where $E_{emi}(Y)$ denotes the total emissions of CFC-11 from prompt emissive uses in year $Y$; $Q_{emi}(Y)$

143 refers to the sales quantity of CFC-11 in prompt emissive uses; $p$ is the release fraction. In this study,

144 $p$ is set to 0.5 to simplify the calculations. Given the global phaseout of CFCs since 2010, variations in

145 the $p$ value will not affect the overall emission calculations beyond this point.

146 **2.2 Bottom-up aggregated emissions**

147 In addition to previously mentioned CFC-11 release from products, emissions of CFC-11 can occur

148 during its production and packaging processes. A comprehensive literature review reveals that fugitive

149 emissions resulting from the production of CFC-11 and its supply chain can contribute an additional

150 1.5%–5% to the overall production emission (TEAP, 2019; 2021; Eq. 9).

$$E_{pro}(Y) = r \times Q_{pro}(Y) \tag{9}$$

151 where $E_{pro}(Y)$ denotes the emissions from the CFC-11 production process in year $Y$; $Q_{pro}(Y)$

152 indicates the production quantity of CFC-11 in year $Y$; $r$ refers to the release rate of CFC-11 resulting

153 from its production and supply chain. TEAP's multi-scenario assessments indicate that production-stage

154 release rates have minimal overall impact on total emissions (TEAP, 2019; 2021). Therefore, in this study,

155 the value of $r$ is set at 3%, except for unreported production, which is set at 5%.

156 The emission profiles of each end-use category and its subdivision are characterized using a distinct

157 set of parameters. We integrate these parameters into D-MFA model to estimate and project temporal and

158 spatial evolution of CFC-11 emissions in accordance with Eq. 10.

$$E_{total}(Y) = \sum E_{pro}(Y) + \sum E_{emi}(Y) + \sum E_{ref}(Y) + \sum E_{ccf}(Y) \tag{10}$$

$$= \sum E_{pro}(Y) + \sum E_{emi}(Y) + \sum E_{man}(Y) + \sum E_{use}(Y) + \sum E_{EoL}(Y)$$

$$+ \sum E_{post}(Y)$$

159    where $E_{total}(Y)$ represents the total emission mass of CFC-11; $\sum E_{pro}(Y)$, $\sum E_{emi}(Y)$, $\sum E_{ref}(Y)$,

160    and $\sum E_{ccf}(Y)$ indicate aggregated emission mass of CFC-11 from compound production and supply

161    chain, prompt emissive uses, refrigeration systems, and CCF applications; $\sum E_{man}(Y)$, $\sum E_{use}(Y)$,

162    $\sum E_{EoL}(Y)$, and $\sum E_{post}(Y)$ denote the aggregated emission mass of CFC-11 brought about by the

163    manufacture, use, EoL handling, and landfill stages of products in refrigeration systems or CCFs,

164    respectively. $\sum E_{pro}(Y)$, $\sum E_{emi}(Y)$, and $\sum E_{man}(Y)$ can be classified as direct emissions; $\sum E_{use}(Y)$,

165    $\sum E_{EoL}(Y)$, and $\sum E_{post}(Y)$ can be categorized as bank release (Lickley et al., 2022).

166        The global and regional database for CFC-11 can be found in Figs. S1–S12. We compiled

167    comprehensive consumption data for each relevant end-use application. Global total production and

168    consumption statistics were sourced from well-established references (AFEAS, 2003; McCulloch et al.,

169    2001; TEAP, 2006, 2021). Regional consumption across CCF subsectors, particularly historical

170    polyurethane rigid (PUR) foam production, was quantified. For non-hermetic refrigeration, global

171    demand was distributed according to regional consumption patterns. Finally, emissive uses of CFC-11

172    were derived by subtracting quantities allocated to CCFs and non-hermetic systems (including refill

173    volumes) from total global consumption.

174        In the D-MFA model for estimating CFC-11 emissions, various parameters influence the estimates

175    when consumption data for different end-use applications across regions are fixed. These parameters

176    include EFs in various life cycle stages, and scale and shape parameters in Weibull distribution. The scale

177    parameters of Weibull distribution typically represent the average lifespans of CFC-11-containing

178    products. Uncertainty in the CFC-11 emission inventory was quantified through systematic Monte Carlo

179    simulations. This approach propagates parameter uncertainties through the emission model, generating

180    output distributions. The Weibull scale ($u$) and shape ($\beta$) parameters, manufacturing-stage EF ($Ef_{man}(Y)$)

181    and use-stage EF ($Ef_{use}(Y)$) were modeled as independent normal variables truncated at zero to preclude

182    implausible negative values. These distributions were defined using regional and global mean values

183    (Tables S7-S13), with standard deviations set to 25% of the respective means. For instance, in the global

184    boardstock subsector, the mean Weibull scale ($u$) and shape ($\beta$) parameters are 50 and 6, while the EFs

185    for manufacturing ($Ef_{man}(Y)$) and use ($Ef_{use}(Y)$) are 6% and 1%, respectively. Consequently, their

standard deviations are thus set to 12.5, 1.5, 1.5%, and 0.25%. Following distribution definition for all

parameters, 10,000 random sample sets (e.g., Fig. S14) were drawn per foam subsector. EoL and post-

life EFs were treated as constant values due to poorly constrained ranges. Annual emissions were

computed for each parameter combination, with 10,000 realizations aggregated to construct the emission

probability distribution. Uncertainties are reported as the 0.5th–99.5th percentile range, corresponding

to 99% confidence interval. The dynamically varying ranges reflect physically plausible outcomes within

the defined uncertainty bounds.

**2.3 Atmospheric concentration modelling**

Global CFC-11 emissions and atmospheric mole fractions (expressed as global mean mixing ratios)

were derived using Equation (11):

$$Mix(Y + 1) = Mix(Y) * e^{-1/\tau} + Emis(Y) * F * \tau * (1 - e^{-1/\tau}) \qquad (11)$$

where $Mix$ denotes global mean atmospheric mixing ratios in parts per trillion (ppt), with data

through 2020 sourced from WMO (2023); $\tau$ denotes CFC-11 atmospheric lifetime (52 years; WMO,

2023); F (*ppt/tonne*) is a constant conversion factor for transforming concentration units to emission

units, obtained using Equation (12).

$$F = 5.679 \cdot 10^{-3} * 1.07/M_{CFC} \qquad (12)$$

where $M_{CFC}$ represents the molecular weight of CFC-11 in grams per mole (g/mol), and the constant

1.07 relates the global mean surface mixing ratio to the global mean atmospheric mixing ratio.

**2.4 Scenarios for unreported CFC-11 production**

TEAP (2019; 2021) identified the most likely routes for unreported CFC-11 production: (a) large-scale

carbon tetrachloride (CTC) conversion (≥50 kt/yr capacity), and (b) micro-scale operations (0.1–2 kt/yr)

producing low-grade CFC-11 for foam blowing. China's 2018 enforcement actions revealed no evidence

of large-scale CFC-11 usage within the country (China MEE, 2019). During these actions, 177.6 metric

tons of CFC-11 precursor materials and 29.9 tons of illegally produced CFC-11 were seized. Among

1,172 inspected polyether enterprises, 10 small-scale operations were found to have partially used CFC-

11. Considering these findings, we developed three scenarios. Scenario 1 (S1) serves as a baseline,

representing an extreme situation with no unreported production. Scenario 2 (S2) is designated as an

intermediate scenario with mid-range unreported production levels. For S2, we assume microscale plants

(0.1–2 kt/yr) contributing 25 kt/yr of unreported production during 2014–2018—half of the TEAP large-

213    scale estimate—with 10 kt in 2013. This assumption would yield a 3–8 kt/yr increase in CFC-11

214    emissions, depending on its application in PUR appliance or spray insulation foam. Scenario 3 (S3)

215    indicates an extreme worst case of unreported CFC-11 production, approximately 23±7 kt/yr of emissions

216    caused by the unreported production of CFC-11 during 2014–2018 (Lickley et al., 2021; TEAP, 2021).

217    Given the diverse end uses of foam products, two subscenarios have been formulated under both S2 and

218    S3. Subscenario 1 assumes unreported CFC-11 is used in spray insulation foams (_SIF), which exhibit a

219    substantial release fraction during their manufacturing. Subscenario 2 involves unreported CFC-11 use

220    in appliance insulation foams (_AIF), characterized by a lower release fraction during manufacture and

221    a shorter product lifespan compared to construction spray foams.

222    **2.5 Uncertainty and sensitivity analysis**

223    This section explores the range of effects of influential factors on CFC-11 emission levels. The sensitivity

224    analysis is structured as follows: 1) The scale parameters ($u$), shape parameters ($\beta$), and EFs associated

225    with manufacturing ($Ef_{\mathrm{man}}(Y)$) and usage ($Ef_{\mathrm{use}}(Y)$) are varied in turn based on their respective

226    distributions, and other factors remain constant values as in Table S13; 2) adjustments in EFs pertinent

227    to EoL are examined, with the assumption that either 100% of CFC-11 is emitted during EoL handling

228    or that only 20% is released during this phase, with the remainder being landfilled and continuously

229    emitted; 3) the quantity of CFC-11 used in CCF products is adjusted, scaled down to 90% or up to 110%

230    of its baseline amount, and the discrepancy in CFC-11 consumption quantities generated is allocated for

231    prompt emissive uses. Specifically, we first evaluated the individual impact of the scale parameter ($u$)

232    through 10,000 Monte Carlo simulations. In these simulations, the value of $u$ was randomly sampled

233    from its respective distribution per foam subsector, while all other factors were fixed at their

234    baseline/mean values. Annual emissions were aggregated to derive the global CFC-11 emission

235    distribution, with uncertainty characterized by the 0.5th–99.5th percentile range, representing the 99%

236    confidence interval. Following the same approach, we sequentially performed single-parameter analyses

237    for shape parameter ($\beta$), manufacturing-stage EFs ($Ef_{man}(Y)$) and use-stage EFs ($Ef_{use}(Y)$). For factors

238    lacking probabilistic distributions, specifically EoL EFs and CFC-11 allocation quantities, we

239    implemented bounded analysis. Weibull parameters ($u$, $\beta$) and manufacturing/use-stage EFs ($Ef_{man}(Y)$,

240    $Ef_{use}(Y)$) were independently sampled from subsector-specific distributions, while EoL EFs were set to

241    either 20% or 100%, with other parameters held at baseline/mean values. Similarly, to assess the impact

242 of CFC-11 quantities allocated to CCFs (i.e., 90% or 110% of the baseline value), we integrated

243 simultaneous random sampling of parameters $u$, $\beta$, $Ef_{man}(Y)$, $Ef_{use}(Y)$ with these allocation

244 adjustments, while fixing other parameters at mean values. Emissions under the baseline scenario (S1)

245 served as reference values for comparative assessment. Collectively, the emission levels falling within

246 the range of uncertainty from the outlined cases are considered plausible, which reflects the complexity

247 and variables inherent in the global CFC-11 emission landscape.

**3 Results and discussion**

**3.1 Estimates of banked CFC-11 emissions and implications**

250 Figs. 1a–1b provide our bottom-up results for the temporal dynamics of global and China CFC-11

251 emissions in comparison with previous studies using top-down and combined approaches, under our

252 baseline S1 without unreported production. These results can be calibrated to align with the 2014–2018

253 top-down emission trends by adjusting Weibull scale/shape parameters or EoL handling emission

254 fractions. However, given the spatiotemporal fluctuations in these parameters (Tables S3–S4), we

255 prioritized robust uncertainty quantification over strict calibration of annual emission trends. We estimate

256 that annual global CFC-11 emissions reached 43 (26–56) kt during 2014–2018 under S1. The estimate

257 from TEAP (2019), 31 (20–45) kt/yr, is lower than ours because our model incorporates a more

258 comprehensive treatment of uncertainties across the lifecycle of CCF products. The upper bound of 56

259 (±2) kt for 2014–2018 emissions in our analysis represents a plausible outcome, significantly narrowing

260 the gap with the top-down estimates of 69 (±10) kt CFC-11 (Montzka et al., 2021).

261 In China, the annual emissions of CFC-11 increased from 8 (4–13) kt/yr in 2008–2012 to 11 (5–13)

262 kt/yr in 2014–2018 according to our bottom-up estimate (Fig.1b). This upward trend, from 8 kt/yr (2008–

263 2012) to 11 kt/yr (2014–2018), was derived using region-specific parameter combinations as detailed in

264 Table S11. Our estimated emission trend over this period differs significantly from previous bottom-up

265 estimates for China. Earlier models arbitrarily assumed that 10% of CFC-11 contained within foams were

266 released during manufacturing, and the remaining 90% uniformly emitted over the subsequent two

267 decades (Fang et al., 2018; Wan et al., 2009). Using top-down approaches, Park et al. (2021) estimated a

268 7±4 kt/yr increase in emissions from eastern China during 2014–2017 compared to 2008–2012. Our

269 national-scale bottom-up modelling aligns the upward trend reported by Park et al. (2021), albeit with a

270  slightly smaller magnitude. Yi et al. (2021) reported a national trend that climbed from 8.3±1.6 kt/yr in

271  2009 to a peak of 13.9±2.4 kt/yr in 2017, followed by a decline to 10.9±1.7 kt/yr in 2019. Our

272  independent estimates of 7 (4–14), 11 (5–14), and 10 (4–13) kt/yr for the corresponding years are broadly

273  consistent with these findings when considering overlapping uncertainties.

274
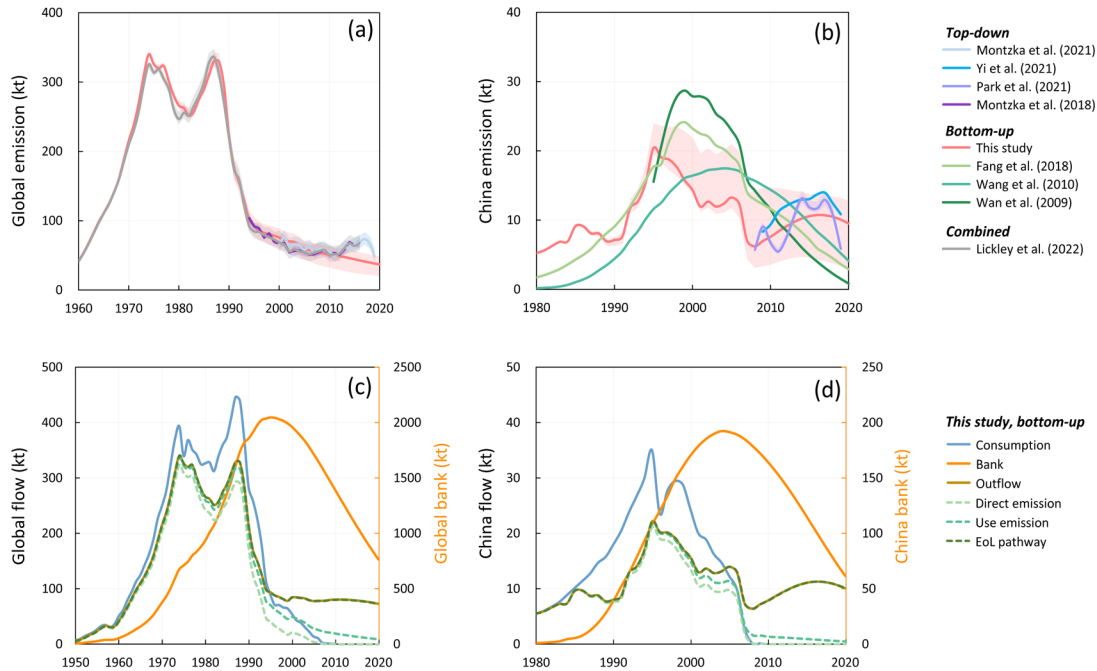
**Figure. 1 CFC-11 emissions comparison through bottom-up, top-down, and combined methods.**
(a) Global emissions. (b) China's emissions. Red shading represents the 99% confidence interval in this
study. (c) Global and (d) China's CFC-11 flows: consumption, banks and outflows. Bank quantities
(orange lines) correspond to the right-hand axis (orange scale). Annual outflows comprise direct
emissions (chemical production/supply chain, emissive uses and end-use product manufacturing), use-
stage emissions, and end-of-life (EoL) flows associated with obsolete products. These EoL flows may
undergo atmospheric release (EoL emissions), landfilling (post-life release), or destruction/reclamation.

282  Figs. 1c–1d depicts our refined assessments of CFC-11 banks and flows at both global and China

283  scales, incorporating a 38-year average lifespan for PUR boardstock and panel foams. This lifespan falls

284  within the distribution range for such foams (Fig. S14). Our analysis yields 2014–2018 use-stage

285  emissions averaging 13±2 kt/yr, while CFC-11 contained in obsolete products reaches up to 65±1 kt/yr.

286  In developing countries, inadequate waste management systems may accelerate CFC-11 release from

287  obsolete product (Gómez-Sanabria et al., 2022; Liu et al., 2024). In addition, the transboundary

288  movement of used appliances (or e-waste) containing CFC-11 (excluded here due to data limitations)

289  from developed to developing economies could redistribute EoL flows (Martínez et al., 2022).

290 Substantial uncertainty surrounds EoL release fractions, with estimates ranging from 20% (UNEP, 2021)

291 to 100% (McCulloch et al., 2001; Liu et al., 2024). Adopting a higher release fraction would bring our

292 global bottom-up CFC-11 emissions into closer alignment with the top-down estimate of 69±10 kt/yr

293 (Montzka et al., 2018; 2021). As shown in Fig. 1d, CFC-11 associated with obsolete products in China

294 rose from 5 kt in 2008 to 10 kt by 2014. Globally, CFC-11 banks peaked at 2,000 kt (1,600–2,200 kt)

295 circa 1995(Fig.1c). This value is comparable with estimates by the TEAP (2006; 2019) but slightly lower

296 than those by Lickley et al. (2022). The discrepancy could be attributed to differing breakdowns of CFC-

297 11 consumption across various end-use applications.

298 **3.2 Banked CFC-11 emissions across regions**

299 Under the Montreal Protocol, parties are classified into developing (Article 5, A5) and developed (non-

300 Article 5, non-A5). In this work, A5 parties are split between China and other A5 parties, while non-A5

301 parties are divided into North America, non-A5 European parties, Japan, and other non-A5 parties (Table

302 S5). Detailed comparisons of CFC-11 emissions from the U.S. and non-A5 European parties are shown

303 in Figs. 2a-c and 2d-f, respectively. Our bottom-up model results indicate that annual CFC-11 emissions

304 in the U.S. have experienced a sharp decline from 40 (38–44) kt in 1990 to 14 (12–19) kt by 1996 (Fig.

305 2a). This reduction is primarily attributed to the phasing out of CFC-11 across the states. Subsequently,

306 emissions originating from banks slightly decreased further to 9 (6–13) kt by 2018. However, with an

307 increasing trend of foam products entering EoL stage, annual CFC-11 emissions are expected to remain

308 around 9 (5–13) kt for several more years. The U.S. Environmental Protection Agency (EPA) has

309 estimated national CFC-11 emissions on an annual basis using bottom-up approach which assigns a fixed

310 value for lifespans; the year-to-year variabilities in these estimates are substantial (U.S.EPA, 2024).

311 According to the latest report, annual CFC-11 emissions in the U.S. declined from 29 kt in 1990 to 5 kt

312 in 2022. When considering the full range of maximum and minimum values from the series of U.S. EPA

313 national estimates (U.S. EPA, 2024), our predicted values align well with their findings. Furthermore, if

314 the lifespan of PUR boardstock and panel foam products changes, i.e., from 50 years (Fig 2b) to 38 years

315 (Fig 2c), CFC-11 emissions are projected to remain a downward trajectory. This trend is consistent with

316 top-down observations and falls within the estimated range of emissions (Hu et al., 2017).
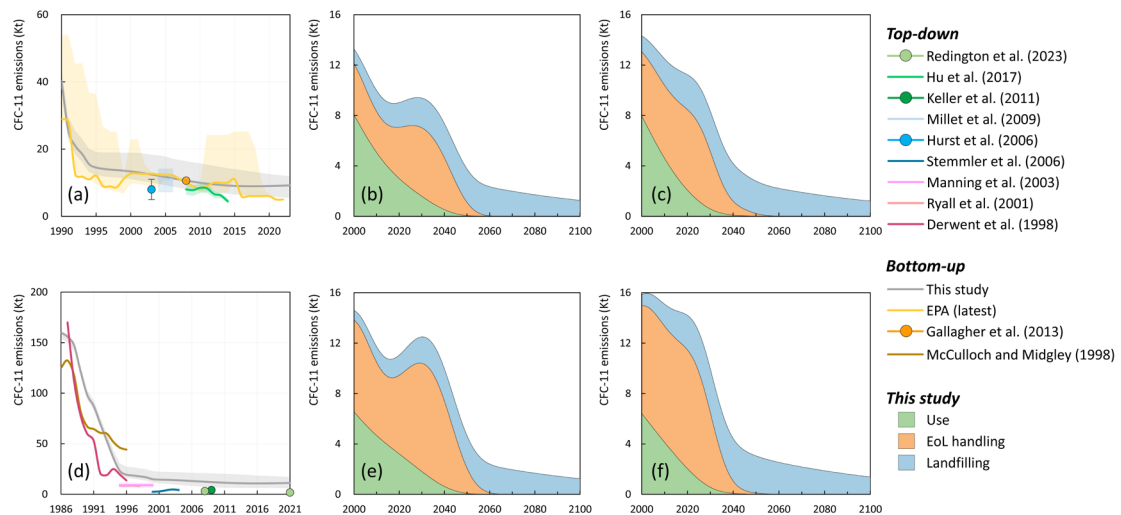
**Figure. 2 Comparison of CFC-11 emission estimates in the U.S. and non-A5 European parties**. (a) CFC-11 emission comparison for the U.S. based on bottom-up and top-down approaches in 1990–2022, respectively. The grey shaded regions indicate the 99% confidence interval in this study. The yellow shaded regions represent the upper and lower bounds of CFC-11 emission estimates conducted by the U.S. EPA. (b) Estimated and projected CFC-11 emissions at various lifecycle stages in the U.S. in 2000–2100. (c), same as (b) but considering the changes of lifespan from 58 to 38 years. (d-f), same as (a-c) but for non-A5 European parties.

Estimates of European CFC-11 emissions exhibit significant geographical variability (Table S1). For non-A5 European parties (Table S5), our analysis indicates emissions declined sharply from 160 (157-165) kt/yr in 1986 to 15 (11-21) kt/yr by 2000, consistent with regional phase-out commitments. This decreasing trend aligns with prior work (McCulloch et al., 1998; Derwent et al., 1998), though emission discrepancies arise from geographical and methodological differences. For instance, McCulloch et al. (1998) applied bottom-up modeling to European Union (EU) emissions, assuming fixed release rates of 10% during manufacturing and 90% uniformly emitted over two decades. Our divergent estimates (Fig. 2d) stem from broader geographical coverage (non-A5 Europe versus EU) and dynamic emission modeling. Post-2000 emissions continued declining to 11 (6–18) kt/yr by 2011, stabilizing near 12 (5–16) kt/yr in subsequent years. Industry data suggest comparable historical CFC-11 usage levels in CCFs across the EU and North America during the 1970s–1980s (Hammitt et al., 1986; FTOC, 1993). Thus, our slightly higher estimates for non-A5 Europe versus the U.S. post-2000 are reasonable. In addition, according to estimates by SKM Enviros, the quantity of obsolete foam products containing CFCs within the EU decreased from 17 kt around 2000 to 12 kt by circa 2011 but subsequently increased again, reaching 15 kt over several years (SKM, 2012). Accordingly, annual CFC emissions remained steady by approximately 6–7 kt during this period. Considering diverse geographical coverage, our

estimates align closely with those reported by SKM (2012). Moreover, based top-down method, Redington et al. (2023) indicated elevated emissions of CFC-11 over Belgium, the southern Netherlands, northern France and west Germany during 2013–2021. Our estimations, as depicted in Fig. 2e, reveal a similarly upward trend in CFC-11 emissions. However, these trends could change. As shown in Fig. 2c and Fig. 2f, persistent downward trends may occur due to shifts in the lifespan of PUR boardstock and panel foam products from 50 to 38 years.

China's distinct emission trajectory, relative to the U.S. and Europe, arises from region-specific factors. First, appliances dominate CFC-11 use in China PUR foams, whereas construction prevails in the U.S. and Europe (Figs. S3/S5 vs. S6). Second, appliances generally have shorter lifespans than construction materials. Third, China exhibits higher release fractions during waste management processes (SI Tables S8 vs. S11). The synergistic interaction of shorter lifespans and elevated EoL release fractions amplified the magnitude of China's emission surge.

In addition, our estimates of banked CFC-11 emissions in Japan and other non-A5 parties also align with previous bottom-up and top-down findings (Fig. S15). In our estimates, banked CFC-11 emissions in Japan decreased from 1.7 (0.8–3.2) kt/yr in 2010 to 0.7 (0.4–1.0) kt/yr by 2022. These values are comparable with those reported in previous estimates (Fig. S15a). For example, the Japan Ministry of Economy, Trade and Industry (METI, 2024), using a bottom-up modeling method, estimated the banked CFC-11 emissions in Japan from 1.3 kt/yr in 2010 to 0.7 kt/yr by 2022. In other non-A5 parties, such as Australia, the emissions of banked CFC-11 decreased from 0.8 (0.6–1.1) kt/yr in 1995 to 0.3 (0.2–0.5) kt/yr by 2022 (Fig. S15b) according to our estimate, which can coincide with the top-down emission level of $0.32 \pm 0.04$ kt/yr in Australia since 2010 (Dunse et al., 2019; Fraser et al., 2020).

To summarize, our analysis suggests that the trajectory of banked CFC-11 emissions can exhibit upward or downward trends, aligning with atmospheric observations recorded for the U.S., Europe, and some other non-A5 parties. These variabilities may be attributed to regional and temporal evolutions in product lifespans and EoL practices (Tables S3–4). A bottom-up methodology necessitates more precise surveys on product lifespans, as well as EFs during EoL handling and landfill disposal.

**3.3 CFC-11 emissions and mitigation potentials considering banks and unreported production**

Although banked CFC-11 likely contributed to the unexpected increase in emissions during 2014–2018, direct emissions from unreported production of CFC-11 also occurred in this period. To assess future

emissions and impacts, we developed three scenarios accounting for emissions from both banks and unreported production. Figs. 3(a–c) present the banks, emissions, and global atmospheric concentrations of CFC-11 across these scenarios. Under S1, global CFC-11 banks declined from 1270 (1000–1500) kt in 2010 to 1040 (720–1250) kt in 2015, and further to 680 (350–950) kt in 2025, with projections indicating a continued decrease to 120 (10–300) kt by 2050. These estimates closely align with those reported by TEAP (2021) and are validated by TEAP's (2019) independent assessment using Ashford et al. (2005) methodology, to estimate 2015 banks at 1070–1292 kt. Cumulative time-lagged global emissions for 2025–2100 under S1 reach 890 (550–1350) kt by 2100, equivalent to 4.2 (2.6–6.4) Gt $CO_2$.

Under S2, unreported production totaled 135 kt during 2013–2018, increasing the 2019 bank by up to 120 (70–130) kt (S2_SIF) and 90 (75–105) kt (S2_AIF). These values are slightly higher than estimated 75 (46–112) kt for China but lower than TEAP assessments of 320–700 kt (Park, et al., 2021; TEAP, 2021). By 2030, this bank increase declines to 75 (45–90) kt (S2_SIF) and 45 (0–105) kt (S2_AIF), falling further to 40 (0–75) kt and 1 (0–15) kt by 2050. Unreported production under S2_SIF may have increased emissions by 7 (4–13) kt during 2014–2018. Cumulative global emissions for 2025–2100 could reach 980 (600–1500) kt, equivalent to 4.7 (2.9–7.1) Gt $CO_2$.

Under S3, cumulative unreported production (390–1100) kt moderately exceeds TEAP's (2021) upper estimate (320–700 kt), primarily due to differences in direct emission rates from foam manufacturing. For S3_AIF, this production increased the 2019 bank by 950 (700–1000) kt, causing a second peak of 1,800 (1500–2,100) kt following the 1995 maximum (Fig.3a). Unreported production may have increased emissions by 22 (12–38) kt (S3_SIF) and 26 (20–35) kt (S3_AIF) during 2014–2018. Emissions from unreported production under S3_AIF are projected to rise from 2020, peaking at 43 (22–67) kt by 2027 (Fig.3b). Cumulative global $CO_2$-equivalent ($CO_2$-eq) emissions for 2025–2100 under S3_AIF could reach 7.5 (4.9–10.5) Gt, corresponding to 18% (12–26%) of global greenhouse gas emissions in 2023 (Friedlingstein et al., 2023). Based on surveys from China (China MEE, 2019) and investigations into illegal production capacity by the TEAP (2019; 2021), S3 is deemed less plausible, suggesting more optimistic outcomes for climate and the ozone recovery.

Using the method outlined in Lickley et al. (2020), polar equivalent effective stratospheric chlorine (EESC) under S1 is projected to return to pre-1980 levels around 2086. This projection is slightly earlier than WMO's estimate of 2087 (WMO, 2023). This discrepancy primarily arises from higher CFC-11 concentrations in WMO assessment, attributed to their larger bank and emission estimates derived using

400 the Lickley approach (Lickley et al., 2022; WMO, 2023).

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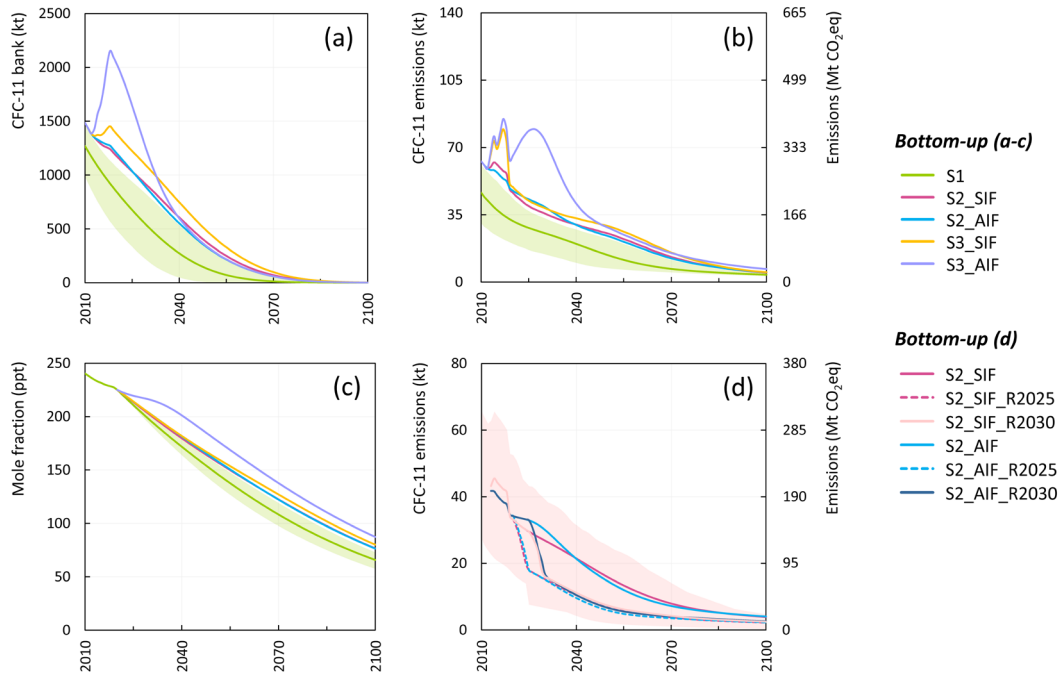
**Figure. 3 CFC-11 banks, emissions and impacts considering unreported production and their**
**mitigation potential**. (a) CFC-11 banks based on scenario analysis. (b) CFC-11 emissions and
corresponding $CO_2$-equivalent ($CO_2$-eq) emissions under various scenarios. (c) CFC-11 global mole
fractions in part per trillion (ppt) under various scenarios. (d) Estimated mitigation potential of CFC-11
emissions and its $CO_2$-eq emissions under scenario 2 (S2). The shaded regions represent the 99%
confidence interval for S1 in panels (a–c) and for S2 in panel (d).

408 Currently, the proportion of emissions resulting from the EoL handling of obsolete CFC-11-

409 containing products is higher than those emitted during the landfilling stage (Figs. 2b-c, and 2e-f).

410 However, by approximately 2050, CFC-11 released from landfilling will become the dominant source.

411 Proper management of EoL handling processes can efficiently reduce CFC-11 emissions. Three different

412 management practices have been considered to evaluate future mitigation potentials: i) a business-as-

413 usual (BAU) scenario, which includes ongoing partial landfilling or recovery of CFC-11 associated with

414 EoL handling; ii) an advanced scenario in which obsolete products will be collected and properly treated,

415 aiming to destroy 85% of CFC-11 in obsolete foam products from 2025 onwards (EU, 2012); iii) a similar

416 scenario as ii but starting from 2030. These cases are aligned with previously defined scenarios S2_SIF

417 and S2_AIF, resulting in a comprehensive matrix of 6 sub-scenarios denoted by unique identifiers.

418 Fig. 3d illustrates the mitigation potential for CFC-11 and its $CO_2$-eq emissions under different S2

419 scenarios. Implementing globally well-managed EoL handling starting in 2025 could reduce cumulative

CFC-11 emissions by 50–52% compared with the BAU scenario between 2025 and 2100. Delaying implementation until 2030 would yield only a 42% reduction. Compared with CFC-11 in building insulation foams, CFC-11 in appliance insulation foams offers greater emission reduction potential through effective EoL handling. In addition, globally well-managed EoL handling from 2025 could reduce cumulative $CO_2$-eq emissions by 2.3 Gt under S2_SIF_2025 and 2.4 Gt under S2_AIF_2025 compared with S2_SIF and S2_AIF, respectively. When comparing scenarios with well-managed EoL handling from 2030 to those from 2025, the latter could further reduce $CO_2$-eq emissions by an additional 0.4–0.5 Gt, underscoring the importance of prompt action. However, this also indicates that advanced strategies will be required to mitigate approximately 2.2–2.7 Gt $CO_2$-eq emissions from the landfilling of CFC-containing foam products.

**3.4 Sensitivity analysis**

Fig. 4 presents the results of sensitivity analysis of the key factors that influence the bottom-up estimation of CFC-11 emissions in this work. According to the analysis, the changes of shape parameters of Weibull distribution and EFs during manufacture and use contribute to a narrow range of uncertainty. Prior research has investigated uncertainties associated with CFC-11 consumption across different end-use patterns. In this study, these uncertainties are assessed in connection with those of product lifespans. Our analysis reveals a large portion of these associated uncertainties attributable to the overall uncertainty in scale parameters of the Weibull distribution, which are highly sensitive to changes in average product lifespan. Moreover, the emissions during the EoL handling of obsolete products exhibit notable disparities. These discrepancies underscore the need for the refinement of the current understanding of emissions from EoL handling. As investigated, regions generating the highest amounts of waste quantities may have the lowest collection rates and the poorest management systems, and unsuitable management, such as dumpsite disposal or burning without air pollution controls, may exist (Gómez-Sanabria et al., 2022). The influence of these unsuitable EoL handling practices on the release of banked CFC-11 remains to be investigated.

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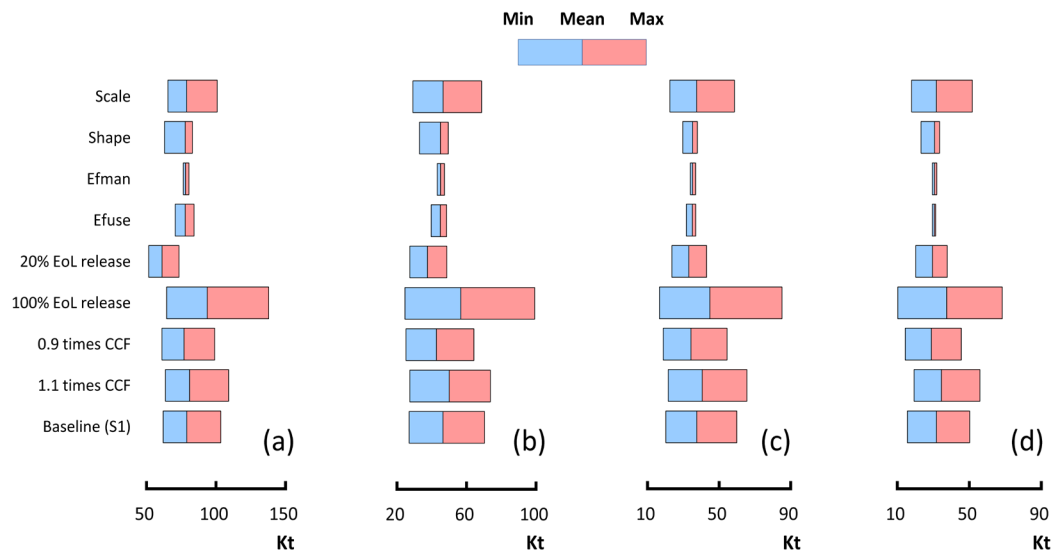
**Figure. 4 Uncertainty and sensitivity analysis of CFC-11 emissions through bottom-up modeling**. (a)–(d) Emission uncertainties induced by various parameters for the year 2000 (a), 2010 (b), 2015 (c) and 2020 (d), respectively. closed-cell foam (CCF)

## 4 Conclusions

In this study, a D-MFA model was developed to quantify CFC-11 banks and emissions at both global and regional scales from 1950 to 2100. Our results indicate that existing banks partially explain the unexpected surge in CFC-11 emissions observed during 2014–2018. By tracking emissions across product lifecycle stages, we further illustrate that the trajectory of bank-derived CFC-11 emissions may exhibit upward or downward trends, driven by uncertainties in foam product lifespans and EoL handling practices. Specifically, through the integration of probabilistic parameter distributions, our model yields an upper-bound emission estimate of 56±2 kt/yr for the 2014–2018 period under the baseline scenario S1. At the regional scale, emissions from China increased moderately, rising from 8 (4–13) kt/yr (2008–2012) to 11 (5–13) kt/yr (2014–2018), primarily driven by EoL product management practices. For other regions, including the U.S., Japan and other non-A5 parties, our model results further support consistency with the regional top-down estimates.

Our findings highlight the important role of product lifespan and EoL handling EFs in estimating long-term trajectories of ODSs and hydrofluorocarbons (HFCs). Bottom-up modeling approaches involve multiple parameters and key processes, which collectively contribute to the overall uncertainty in emission estimates. Rigorous and systematic analyses, particularly those examining temporal and spatial variations in product lifespans and release patterns during EoL handling, are therefore essential

466 to mitigating the inherent uncertainties linked to bottom-up modeling. While this study primarily focuses

467 on CFC-11 emissions, the methodology developed here, which explicitly accounts for uncertainties from

468 underexplored sources, offers broad applicability for estimating emissions of other ODSs and HFCs.

469

470 **Data availability**

471 All data inputs, including regional production statistics, product lifespan datasets, emission factors and

472 end-of-life handling metrics, are available in the Supplementary Information.

473

474 **Author contributions**

475 H.L., H.D. and Y.M. conceptualized and designed the study, conceived the paper, developed the model.

476 H.L. and Y.M. collected the data and conducted the analysis. N.Z. drew the figures. H.L., H.D., N.Z.,

477 T.R.M. and M.X. contributed to the conceptual model development. R.M., M.X. and J.Y. enhanced

478 scenario analysis discussions. H.D., H.L., N.Z., R.M., Y.M., T.R.M., M.X. and J.Y. contributed to

479 discussing the results and writing the paper.

480

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484

485 **Declaration of interests**

486 The authors declare no competing interests.

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