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), ~~L~~long-term emission of banked CFC-11 will accumulate to ~~accumulate to~~ 980 (600–1500) kilotons (~~Kt~~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, ~~some areas have shown increased emissions~~ with no evidence of unreported production, raising unresolved questions about their sources (Dunse et al., 2019; ~~Manning et al., 2022;~~ 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, th~~ese~~is 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 various 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

91 in industrial settings, such as refrigerated warehouses (S1.2). The investigated lifespan of these foam

92 products in building contexts exhibits notable spatiotemporal variability (Table S3).

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

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

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

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

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

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

99 CFC-11 contained within them.

100   End-use products, such as ~~refrigerant and~~ insulation foams for household refrigerator appliances or

101 constructions, exhibit durability and long lifespans. In this study, the Weibull distribution, which had

102 been widely used in previous research (Liu et al., 2019; 2024; UNEP, 2021), is introduced to simulate

103 the survival rate curves for new products with long lifespans (Eq. 1, Fig. S13).

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

115 EoL products in year Y.

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

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

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

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

120 using Eq. 6.

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

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

122 $E_{use}(Y)$, and $E_{\mathrm{EoL}}(Y)$ correspond to the mass emissions of CFC-11 from the manufacturing (first fill),

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

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

125 $Ef_{\mathrm{use}}(Y)$ denote the EFs of CFC-11 during the life cycle stages of equipment manufacturing and use,

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

127 equipment.

128   Similarly, the release of CFC-11 from CCF~~closed-cell foams~~ can occur throughout various life cycle

129 stages, including manufacture, use, EoL handling, and post-life, such as landfilling disposal. Eqs 1–5 are

130 used to calculate the CFC-11 bank and EoL flow from CCF~~closed-cell foams~~. Eq. 7 is used to aggregate

131 emissions across various life cycle stages originating from CCF~~closed-cell foams~~.

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

132 where $E_{ccf}(Y)$ indicates the total emissions of CFC-11 from an end-use application of CCF~~closed-cell~~

133 ~~foam~~ in year Y; $E_{man}(Y)$, $E_{use}(Y)$, $E_{\mathrm{EoL}}(Y)$, and $E_{post}(Y)$ denote the mass emissions of CFC-11 from

134 the manufacture, use, EoL handling, and post-life stages of CCF~~closed-cell foams~~, respectively, at year

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

136    $Ef_{\text{use}}(Y)$, $Ef_{EoL}(Y)$, and $Ef_{post}(Y)$ are the EFs of CFC-11 during the life cycle stages of CCF~~closed~~

137   ~~cell foam~~ manufacture, use, EoL handling, and landfilling, respectively; $l_{EoL}(Y)$ means the landfilling

138   proportion of CFC-11 during the EoL handling of CCF~~closed-cell foam~~, represented as (100%-$Ef_{EoL}(Y)$).

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

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

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

142   11 being released in the year of sale and the remaining half being released in the following year on

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

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

(8)

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

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

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

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

148   **2.2 Bottom-up aggregated emissions**

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

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

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

152   1.5%–5% to the overall production emission (TEAP, 2019; 2021; Eq. 9).

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

(9)

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

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

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

156   release rates have minimal overall impact on total emissions (TEAP, 2019; 2021). Therefore, Iin this

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

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

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

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

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

(10)

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

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

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

chain, prompt emissive uses, refrigeration systems, and CCF~~closed-cell foam~~ applications; $\sum E_{man}(Y)$,

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

by the manufacture, use, EoL handling, and landfill stages of products in refrigeration systems or

CCFs~~closed-cell foams~~, respectively. $\sum E_{pro}(Y)$, $\sum E_{emi}(Y)$, and $\sum E_{man}(Y)$ can be classified as

direct emissions; $\sum E_{use}(Y)$, $\sum E_{EoL}(Y)$, and $\sum E_{post}(Y)$ can be categorized as bank release (Lickley

et al., 2022).

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

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

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

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

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

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

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

volumes) from total global consumption.

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

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

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

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

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

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

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

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

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

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

boardstock/laminate subsector, the mean Weibull scale (u) and shape (β) parameters are 50 and 6, while

the EFs 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., Figure 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. ~~To address the temporal and spatial variability, the parameters of Weibull distribution (scale parameter u and shape parameter β) as well as EFs ($Ef_{man}(Y)$ and $Ef_{use}(Y)$) are described using distributions. Relevant information regarding parameters can be found in Tables S7–S14.~~

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); τ 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-scale estimate—with 10 kt in 2013 the most likely situation, there was indeed unreported production of CFC-11 during 2014–2018. We assume microscale plants producing 0.1–2 Kkt/yr CFC-11 as potential sources. According to the gap between upper limit of bottom-up and lower limit of top-down estimates, w. This assumption would yield a 3–8 kt/yr increase in CFC-11 emissions, depending on its application in PUR appliance or spray insulation foam. Scenario 3 (S3) indicates an extreme worst case of unreported CFC-11 production, approximately 23 ± 7 Kkt/yr of emissions caused by the unreported production of CFC-11 during 2014–2018 (Lickley et al., 2021; TEAP, 2021). Given the diverse end uses of foam products, two subscenarios have been formulated under both S2 and S3. Subscenario 1 assumes unreported CFC-11 is used in spray insulation foams (_SIF), which exhibit a substantial release fraction during their manufacturing. Subscenario 2 involves unreported CFC-11 use in appliance insulation foams (_AIF), characterized by a lower release fraction during manufacture and a shorter product lifespan compared to construction spray foams.

2.54 Uncertainty and ~~Sensitive Analysis~~ sensitivity analysis

This section explores the range of effects of influential factors on CFC-11 emission levels. The emission analysis is structured as follows: 1) The scale parameters (u), shape parameters (β), and EFs associated with manufacturing $(Ef_{\mathrm{man}}(Y))$ and usage $(Ef_{\mathrm{use}}(Y))$ are varied in turn based on their respective distributions, and other factors remain constant values as in Table S13; 2) adjustments in EFs pertinent to EoL are examined, with the assumption that either 100% of CFC-11 is emitted during EoL handling or that only 20% is released during this phase, with the remainder being landfilled and continuously emitted; 3) the quantity of CFC-11 used in CCF closed-cell foam products is adjusted, scaled down to 90% or up to 110% of its baseline amount, and the discrepancy in CFC-11 consumption quantities generated is allocated for prompt emissive uses; 4) the average scale parameters of the Weibull distribution are recalibrated based on parameters in Table S14. Specifically, we first evaluated the individual impact of the scale parameter (u) through 10,000 Monte Carlo simulations. In these simulations, the value of u was randomly sampled from its respective distribution per foam subsector, while all other factors were fixed at their baseline/mean values. Annual emissions were aggregated to derive the global CFC-11

244 emission distribution, with uncertainty characterized by the 0.5th–99.5th percentile range, representing

245 the 99% confidence interval. Following the same approach, we sequentially performed single-parameter

246 analyses for shape parameter (β), manufacturing-stage EFs ($Ef_{man}(Y)$) and use-stage EFs ($Ef_{use}(Y)$).

247 For factors lacking probabilistic distributions, specifically EoL EFs and CFC-11 allocation quantities, we

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

249 $Ef_{use}(Y)$) were independently sampled from subsector-specific distributions, while EoL EFs were set to

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

251 of ±10% variations in CFC-11 quantities allocated to CCFs (i.e., 90% or 110% of the baseline value), we

252 integrated simultaneous random sampling of parameters u, β, $Ef_{man}(Y)$, $Ef_{use}(Y)$ with these

253 allocation adjustments, while fixing other parameters at mean values. Emissions under the baseline

254 scenario (S1) served as reference values for comparative assessment. Collectively, the emission levels

255 falling within the range of uncertainty from the outlined cases are considered plausible, which reflects

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

257 **3 Results and discussion**

258 **3.1 Estimates of banked CFC-11 emissions and implications**

259 Figs. 1a–1b provide our bottom-up model results for the temporal dynamics of global and Chinese CFC-

260 11 emissions in comparison with previous studies using top-down and combined approaches, under our

261 baseline S1 with no unreported production. These results can be calibrated to align with the 2014–2018

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

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

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

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

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

267 comprehensive treatment of uncertainties across the lifecycle of CCF~~closed cell foam~~ products. The

268 upper bound of 56 (±2) kt for 2014–2018 emissions in our analysis represents a plausible outcome,

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

270 2021).~~Specifically, the lifespan parameter of closed cell foam products depends not only on product~~

271 ~~attributes, but also on cultural, economic, and political factors, which will require further elucidation~~

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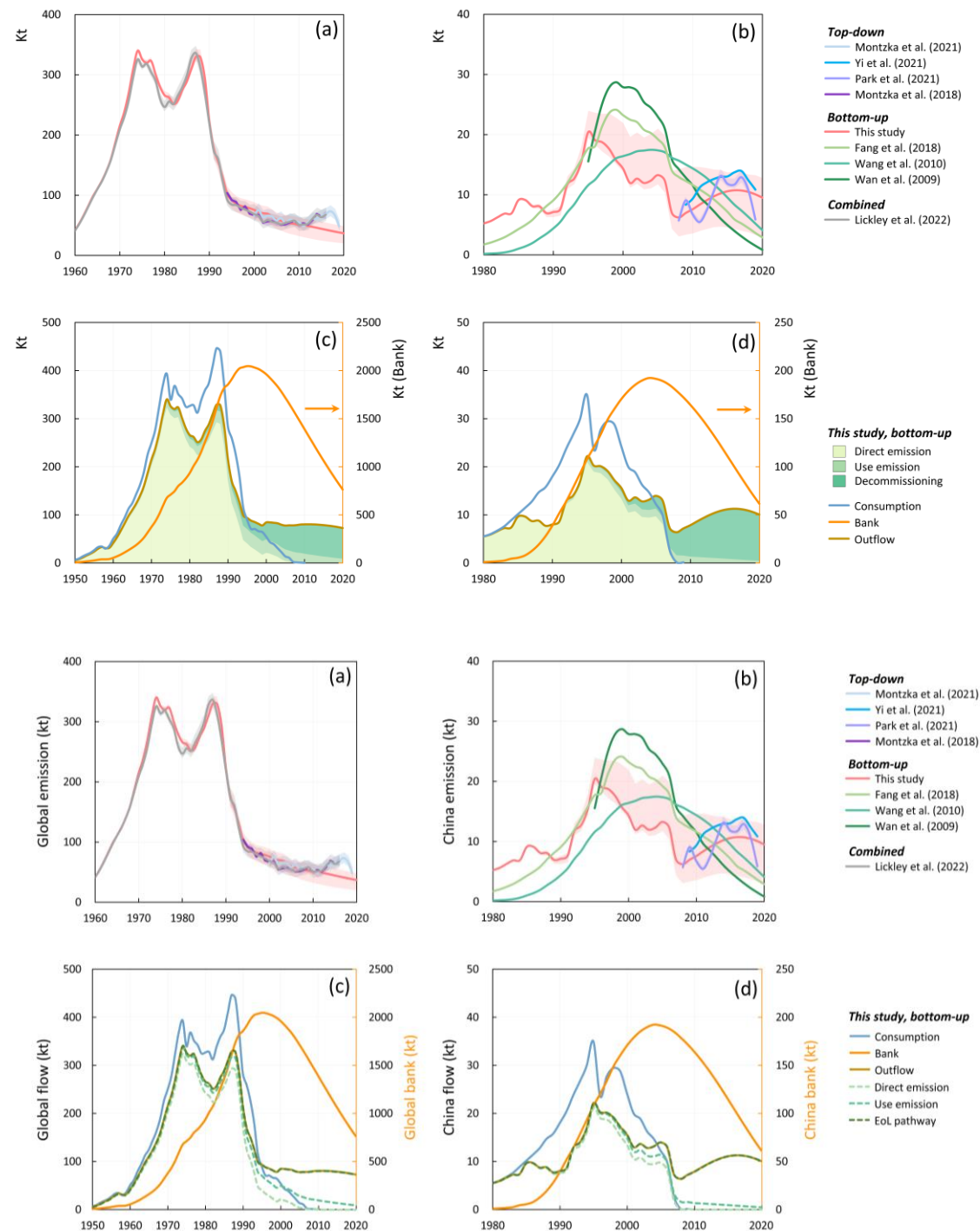
Figure. 1 CFC-11 emissions comparison through bottom-up, top-down, and combined methods.
(a) Global emissions. (b) China's emissions. Red shading represents the 99% confidence interval in this study. (c) Global and (d) China's CFC-11 flows: consumption, banks and outflows. Bank quantities (orange lines) 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

282   destruction/reclamation~~Regarding decommissioning phase, CFC-11 contained in obsoleted foam~~
283   ~~products might either be released into the atmosphere (EoL emissions), disposed of by landfills or~~
284   ~~destroyed~~.

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

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

287   (2008–2012) to 11 kt/yr (2014–2018), was derived using region-specific parameter combinations as

288   detailed in Table S11. Our estimated emission trend over this period differs significantly from previous

289   bottom-up estimates for China. Earlier models arbitrarily assumed that 10% of CFC-11 contained within

290   foams were released during manufacturing, and the remaining 90% uniformly emitted over the

291   subsequent two decades (Fang et al., 2018; Wan et al., 2009).~~Our bottom-up estimates have therefore~~

292   ~~aligned more closely with top-down assessments. For example, u~~Using top-down approaches, Park et al.

293   (2021) estimated a 7 ± 4 ~~K~~kt/yr increase in emissions from eastern China during 2014–2017 compared

294   to 2008–2012. Our national-scale bottom-up modelling aligns the upward trend reported by Park et al.

295   (2021), albeit with a slightly smaller magnitude. Yi et al. (2021) reported a national trend that climbed

296   from 8.3 ± 1.6 kt/yr in 2009 to a peak of 13.9 ± 2.4 kt/yr in 2017, followed by a decline to 10.9 ± 1.7

297   kt/yr in 2019. Our independent estimates of 7 (4–14), 11 (5–14), and 10 (4–13) kt/yr for the corresponding

298   years are broadly consistent with these findings when considering overlapping uncertainties. ~~There is~~

299   ~~ample evidence that such activities appear to have occurred at a microscale level (b), with no substantial~~

300   ~~indication of widespread illegal use for large-scale conversion (a)~~.

301   ~~The global annual quantity of decommissioned CFC-11 reached approximately 65 (±1) Kkt during~~

302   ~~2014–2018, when considering an average lifespan of 38-year for PUR boardstock and panel foams. This~~

303   ~~value falls within the distribution range (Fig. S13).~~ Figs. 1c–1d depicts our refined assessments of CFC-

304   11 banks and flows at both global and Chinese scales, incorporating a 38-year average lifespan for PUR

305   boardstock and panel foams. This lifespan falls within the distribution range for such foams (Fig. S14).

306   Our analysis yields 2014–2018 use-stage emissions averaging 13±2 kt/yr, while CFC-11 contained in

307   obsolete products reaches up to 65±1 kt/yr~~decommissioned CFC-11 for this period were higher than the~~

308   ~~45 K~~kt~~estimate from TEAP (2021)~~. In developing countries, inadequate waste management systems may

309   accelerate CFC-11 release from obsolete product (Gómez-Sanabria et al., 2022; Liu et al., 2024). In

310   addition, the transboundary movement of used appliances (or e-waste) containing CFC-11 (excluded here

311   due to data limitations) from developed to developing economies could redistribute EoL flows (Martínez

312   et al., 2022). Substantial uncertainty surrounds EoL release fractions, with estimates ranging from 20%

(UNEP, 2021) to 100% (McCulloch et al., 2001; Liu et al., 2024). Adopting a higher release fraction all decommissioned CFC-11 were released into the atmosphere, our global bottom-up CFC-11 emissions would bring our global bottom-up CFC-11 emissions into closer alignment with the top-down estimate of 69 ± 10 kt/yr (Montzka et al., 2018; 2021). As shown in Fig. 1d, the decommissioned amount of CFC-11 associated with obsolete products in China rose from 5 kt in 2008 to 10 kt by 2014. Globally, CFC-11 bank peaked at 2,000 kt (1,600–2,200 kt) circa 1995(Fig.1c). This value is comparable with the estimates by the TEAP (2006; 2019) but slightly lower than those by Lickley et al. (2022). The discrepancy could be attributed to differing breakdowns of CFC-11 consumption across various end-use applications.

3.2 Banked CFC-11 emissions across regions

Under the Montreal Protocol, parties are classified into developing (Article 5, A5) and developed (non-Article 5, non-A5). In this work, A5 parties are split between China and other A5 parties, while non-A5 parties are divided into North America, non-A5 European parties, Japan, and other non-A5 parties (Table S5). Detailed comparisons of CFC-11 emissions from the U.S. and non-A5 European parties are shown in Figs. 2a-c and 2d-f, respectively. Our bottom-up model results indicate that annual CFC-11 emissions in the U.S. have experienced a sharp decline from 40 (38–44) kilotons (kt) in 1990 to 14 (12–19) kt by 1996 (Fig. 2a). This reduction is primarily attributed to the phasing out of CFC-11 across the states. Subsequently, emissions originating from banks slightly decreased further to 9 (6–13) kt by 2018. However, with an increasing trend of foam products entering EoL stage, annual CFC-11 emissions are expected to remain around 9 (5–13) kt for several more years. The U.S. Environmental Protection Agency (EPA) has estimated national CFC-11 emissions on an annual basis using bottom-up approach which assigns a fixed value for lifespans; the year-to-year variabilities in these estimates are substantial (U.S.EPA, 2024). According to the latest report, annual CFC-11 emissions in the U.S. declined from 29 kt in 1990 to 5 kt in 2022. Similar U.S. EPA studies underestimated CFC-12 emissions from older refrigeration and air conditioning units by 42% (Gallagher et al., 2014), and this phenomenon may also be evident in their recent estimates of CFC-11 emissions. When considering the full range of maximum and minimum values from the series of U.S. EPA national estimates (U.S. EPA, 2024), our predicted values align well with their findings. Furthermore, if the lifespan of PUR boardstock and panel foam products changes- from 50 years (Fig 2b) to 38 years (Fig 2c), CFC-11 emissions are projected to remain

342 a downward trajectory. This trend is consistent with top-down observations and falls within the estimated

343 range of emissions (Hu et al., 2017).

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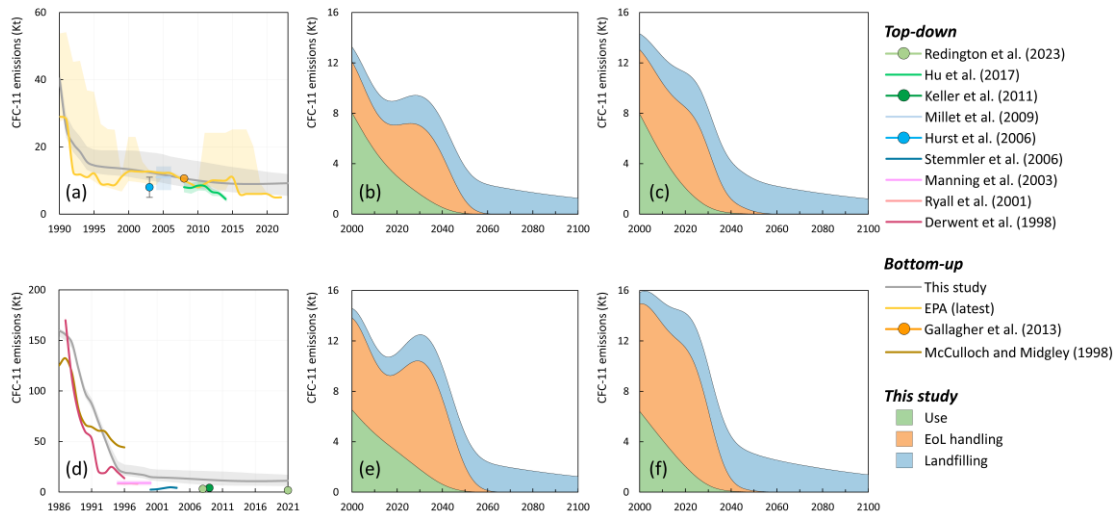
345 **Figure. 2 Comparison of CFC-11 emission estimates in the U.S. and non-A5 European parties**. (a)

346 CFC-11 emission comparison for the U.S. based on bottom-up and top-down approaches in 1990–

347 2022, respectively. The grey shaded regions indicate the 99% confidence interval in this study. The

348 yellow shaded regions represent the upper and lower bounds of CFC-11 emission estimates conducted

349 by the U.S. EPA. (b) Estimated and projected CFC-11 emissions at various lifecycle stages in the U.S.

350 in 2000–2100. ~~CFC-11 emissions from end-use applications can be divided into those from prompt~~

351 ~~emissive uses and those from various lifecycle stages of CFC-11-containing products with long~~

352 ~~lifespans, including manufacturing, use, EoL handling, and landfilling. Key parameters for our bottom~~

353 ~~up model, including lifespan and emission factors across life stages, were shown in Tables S7–8.~~ (c),

354 same as (b) but considering the ~~sensitivity of some parameters, such as~~ changes of lifespan from 58 to

355 38 years ~~shown in Table S14~~. (d-f), same as (a-c) but for non-A5 European parties.

356 Estimates of European~~on~~ CFC-11 emissions exhibit significant geographical variability (Table S1).

357 For non-A5 European parties (Table S5), our analysis indicates emissions declined sharply from 160

358 (157-165) kt/yr in 1986 to 15 (11-21) kt/yr by 2000, consistent with regional phase-out commitments.

359 ~~These estimates are comparable with both previous bottom-up (McCulloch et al., 1998) and top-down~~

360 ~~assessments (Derwent et al., 1998; Manning et al., 2003) within the margin of uncertainty (Fig.2d).~~ This

361 decreasing trend aligns with prior work (McCulloch et al., 1998; Derwent et al., 1998), though emission

362 discrepancies arise from geographical and methodological differences. For instance, McCulloch et al.

363 (1998) applied bottom-up modeling to European Union (EU) emissions, assuming fixed release rates of

364 10% during manufacturing and 90% uniformly emitted over two decades. Our divergent estimates (Fig.

365 2d) stem from broader geographical coverage (non-A5 Europe versus EU) and dynamic emission

366 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, Aaccording to estimates by SKM Enviros, the quantity of obsolete foam products containing CFCs within the European Union (EU) decreased from 17 Kkt around 2000 to 12 Kkt by circa 2011 but subsequently increased again, reaching 15 Kkt over several years (SKM, 2012). Accordingly, annual CFC emissions remained steady by approximately 6−7 Kkt 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 Germanytapproximately 0.2 Kkt in 2011 to 0.4 Kkt in 2017 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 (Figures 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. S154). In our estimates, banked CFC-11 emissions in Japan decreased from 1.7 (0.8–3.2) Kkt/yr in 2010 to 0.7 (0.4–1.0) Kkt/yr by 2022. These values are comparable with those reported in previous estimates (Fig. S154a). 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 Kkt/yr in 2010 to 0.7 Kkt/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) Kkt/yr in 1995 to 0.3 (0.2–0.5) Kkt/yr by 2022 (Fig. S154b) according to our estimate, which can coincide with the top-down emission level of 0.32 ± 0.04 Kkt/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 ~~probably~~ 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–cd) 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) ~~K~~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) ~~K~~kt, ~~—~~equivalent to 4.7 (2.9–7.1) Gt CO_2. ~~However, emissions of CFC-11 under S3_SIF and S3_AIF are substantially higher than those projected for S2.~~

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) ~~K~~kt, causing a second peak of 1,800 (1500–2,100) kt following the 1995 maximum (Fig.3a). Unreported production

425 may have increased emissions by 22 (12–38) kt (S3_SIF) and 26 (20–35) kt (S3_AIF) during 2014–2018.

426 Emissions from unreported production under S3_AIF are projected to rise from 2020, peaking at 43

427 (22–67) (4–100) Kkt by 2027 (Fig.3b). Cumulative global CO_2-equivalent (CO_2-eq) emissions for 2025–

428 2100 under S3_AIF could reach Th cumulative CO_2-equivalent (CO_2-eq) emissions 7.5 (4.9–10.5) Gt,

429 corresponding to 18% (12–26%) of global greenhouse gas emissions in 2023 (Friedlingstein et al., 2023).

430 Based on surveys from China (China MEE, 2019) and investigations into illegal production capacity by

431 the TEAP (2019; 2021), S3 is deemed less plausible, suggesting more optimistic outcomes for climate

432 and the ozone recovery.

433   Using the method outlined in Lickley et al. (2020), polar equivalent effective stratospheric chlorine

434 (EESC) under S1 is projected to return to pre-1980 levels around 2086. This projection is slightly earlier

435 than WMO's estimate of 2087 (WMO, 2023). This discrepancy primarily arises from higher CFC-11

436 concentrations in WMO assessment, attributed to their larger bank and emission estimates derived using

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

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

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

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

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

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

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

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

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

446 scenario as ii but starting from 2030. These cases are aligned with previously defined most likely

447 scenarios S2_SIF and S2_AIF, resulting in a comprehensive matrix of 6 sub-scenarios denoted by unique

448 identifiers.

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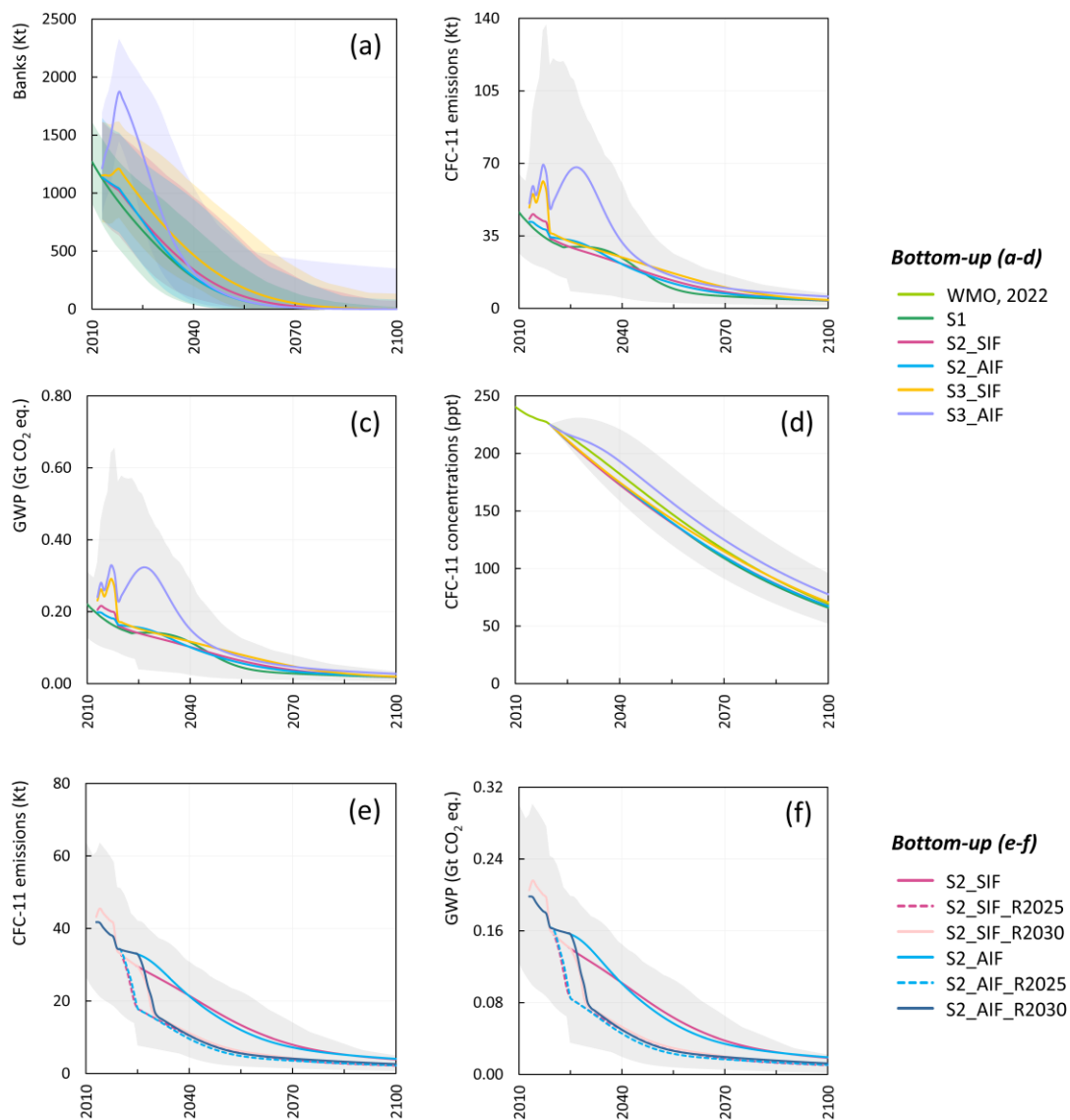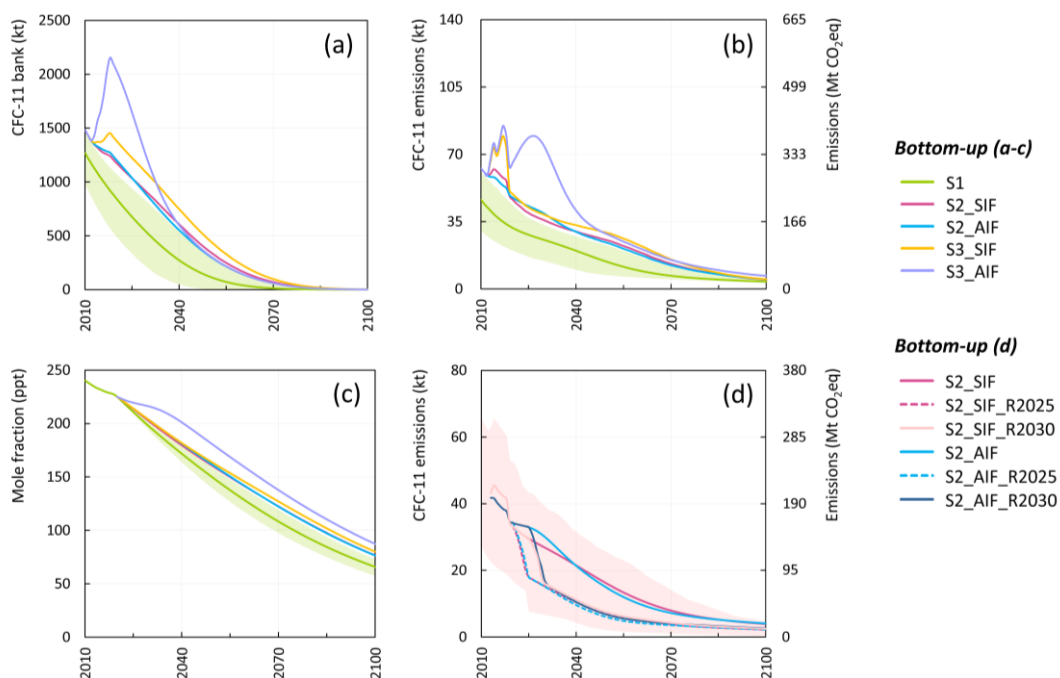
Figure. 3 CFC-11 banks, emissions and impacts considering unreported production and their mitigation potential. (a) CFC-11 banks based on scenario analysis. (b) ~~(c)~~ CFC-11 emissions and corresponding CO$_2$-equivalent (CO$_2$-eq)~~global warming potential (GWP) weighted~~ emissions ~~in terms of 100-year time horizon~~ under various scenarios. (c~~d~~) CFC-11 global mole fractions ~~concentrations~~ in part per trillion (ppt) under various scenarios. ~~The assumed constant lifespan of CFC-11 is 52 years.~~ (d~~e~~) ~~(f)~~ Estimated mitigation potential of CFC-11 emissions ~~(e)~~ and its CO$_2$-eq~~GWP-weighted~~ emissions ~~(f)~~ under scenario 2 (S2). The shaded regions represent the 99% confidence interval for S1 in panels (a–c) and for S2 in panel (d)~~The grey shaded regions indicate the 99% confidence interval~~.

Fig. 3d~~e 3f~~ illustrates the mitigation potential for CFC-11 and its CO$_2$-eq emissions under different S2 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. ~~Specifically, S2_AIF_2025 demonstrates a higher capacity for reducing EoL emissions than S2_SIF_2025.~~ 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~~BAU scenarios~~. 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

471   emissions from the landfilling of CFC-containing foam products.

472   **3.4 Sensitivity analysis**

473   Fig. 4 presents the results of sensitivity analysis of the key factors that influence the bottom-up estimation

474   of CFC-11 emissions in this work. According to the analysis, the changes of shape parameters of Weibull

475   distribution (Fig.4b) and EFs during manufacture (Fig.4c) and use (Fig.4d) contribute to a narrow range

476   of uncertainty. Prior research has investigated uncertainties associated with CFC-11 consumption across

477   different end-use patterns. In this study, these uncertainties are assessed in connection with those of

478   product lifespans. Our analysis reveals a large portion of these associated uncertainties (Fig.4g h)

479   attributable to the overall uncertainty in scale parameters of the Weibull distribution (Fig.4a), which are

480   highly sensitive to changes in average product lifespan. Moreover, the emissions during the EoL handling

481   of obsolete products exhibit notable disparities (Fig.4e f). These discrepancies underscore the need for

482   the refinement of the current understanding of emissions from EoL handling. As investigated, regions

483   generating the highest amounts of waste quantities may have the lowest collection rates and the poorest

484   management systems, and unsuitable management, such as dumpsite disposal or burning without air

485   pollution controls, may exist (Gómez-Sanabria et al., 2022). The influence of these unsuitable EoL

486   handling practices on the release of banked CFC-11 remains to be investigated.
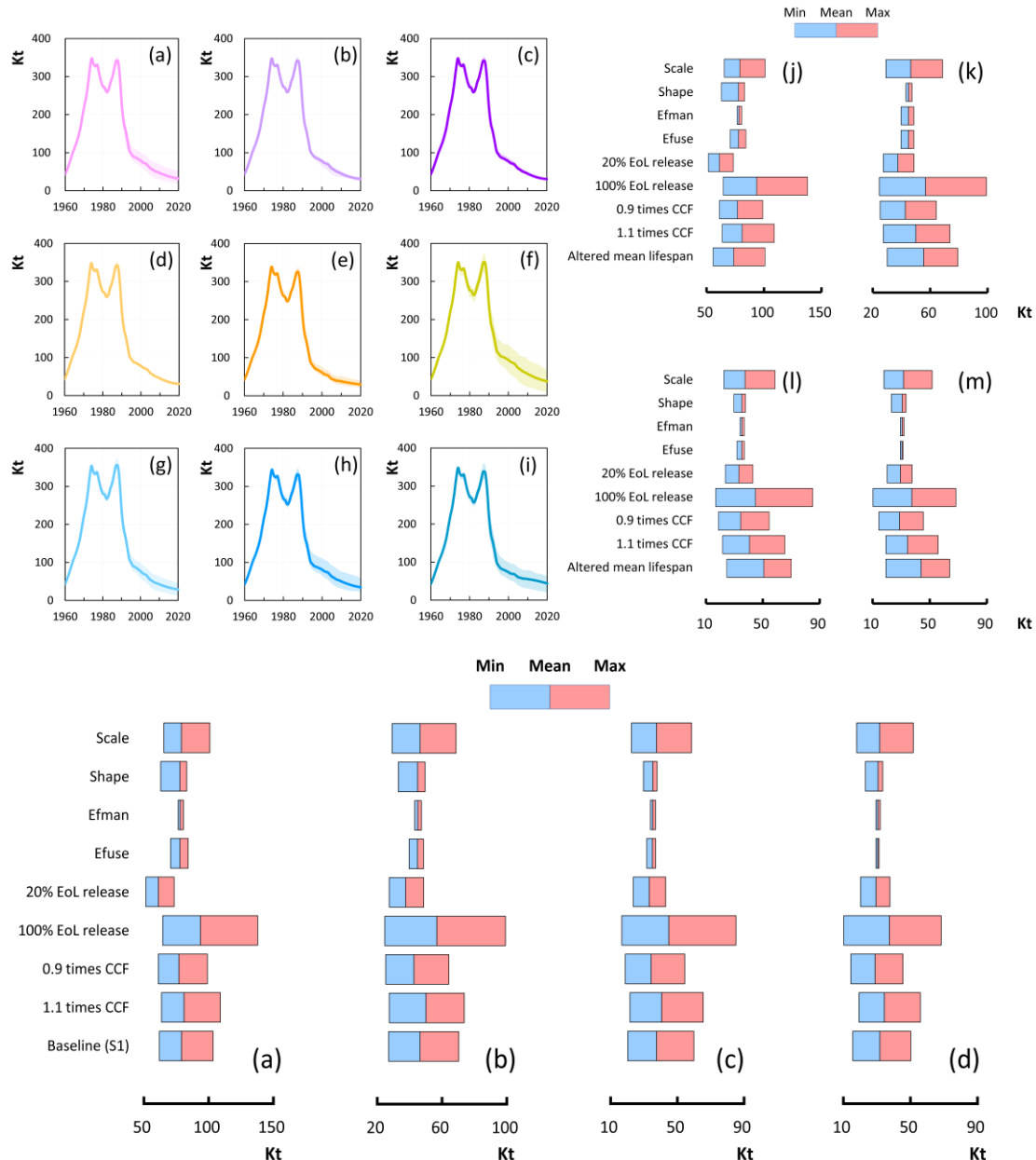
Figure. 4 Uncertainty and sensitivity analysis of CFC-11 emissions through bottom-up modeling. (a)–(d~f~) ~Emission uncertainties induced by~ various parameters. ~(a) Scale parameters, similar to the average lifespan of foam products, vary based on probability distributions, whereas other parameters remain constant. (b) Similar to (a) but for shape parameters. (c) Similar to (a) but for emission factors (EFs) during the manufacturing stage. (d) Similar to (a) but for EFs associated with product usage. (e)~ ~(f) Changes in EFs related to EoL handling and landfilling. (e) A total of 20% of CFC-11 is released during EoL handling, with the remainder being landfilled and continuing to contribute to emissions. (f) Exactly 100% of CFC-11 is emitted during EoL handling. (g)~ ~(h) The quantity of CFC-11 in closed-cell foam (CCF) products is scaled down to 0.9 times (g) and up to 1.1 times (h) its original amount. The resulting gap in CFC-11 quantities is allocated for prompt emissive uses. (i) The mean lifespan has been adjusted. (j) Emission ranges resulting from the factors outlined in (a-i)~ for the year 2000~.~ (a~k~)~ (m) same as (j) but for the year,~ 2010 (b~k~), 2015 (c~l~) and 2020 (d~m~), 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)Top-down and bottom-up modeling approaches display an inherent association with uncertainties. Bottom-up modeling approaches involve multiple parameters and key processes, which collectively contribute to the overall uncertainty in emission estimates. Key factors include, but are not limited to, production quantity, end-use consumption fraction, EFs during various lifecycle stages, lifespans of CFC-11-containing products, and processes involved in EoL handling, such as recovery, recycling, shredding, compacting, landfilling, and other activities related to obsolete products. The interplay among these factors introduces significant uncertainties in emission estimation. Our analysis underscores the importance of considering the broader context of factors affecting CFC-11 emissions and highlighting the role of accurate data in diminishing uncertainties. Rigorous and systematic analyses, particularly those examining temporal and spatial variations in product lifespans and release patterns during EoL handling, are therefore essential to mitigating the inherent uncertainties linked to bottom-up modeling.

Studies have demonstrated the complementary potentials of bottom-up and top-down approaches. Bottom-up methods provide valuable insights into specific source contributions and aid in the development of mitigation strategies, while top-down methods offer independent verification for bottom-up findings. The integration of these analytical frameworks is essential for enhancing our comprehension

531 ~~and accuracy in quantifying emission dynamics.~~ While this study primarily focuses on CFC-11 emissions,

532 the methodology developed here, which explicitly accounts for uncertainties from underexplored sources,

533 offers broad applicability for estimating emissions of other ODSs and HFCs.

534

535 **Data availability**

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

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

538

539 **Author contributions**

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

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

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

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

544 discussing the results and writing the paper.

545

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549

550 **Declaration of interests**

551 The authors declare no competing interests.

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