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# 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. Long-term emission of banked CFC-11 will accumulate to accumulate to 1000 (700–1300) kilotons (Kt), equivalent to 4.6 (3.2–6.3) gigatons (Gt) $CO_2$e, between 2025 and 2100. Scenario analysis highlights the potential to reduce up to 50% of emissions through optimized end-of-life 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.

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## 1 Introduction

Chlorofluorocarbons (CFCs) are major contributors to ozone depletion and potent greenhouse gases, which trap heat approximately 4,660-13,900 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, some areas have shown increased emissions without clear evidence of unreported production or consumption, 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, these 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

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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 and landfilling 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, and prompt emissive uses, such as aerosol propellants, open-cell foams, solvents, and others (TEAP, 2019; 2021). A D-MFA model is used for each end-use application. In this model, the calculation process for CFC-11 emissions is methodically divided into distinct stages, which represent different phases of products containing CFC-11 throughout their lifecycle, from manufacture, use to EoL handling. The first stage involves the quantification of first-fill emissions during the manufacturing stage. Subsequently, emissions during product use are evaluated. The final stage pertains to the release of emissions from the

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EoL handling of obsolete products, which may involve recovery, recycling, and disposal processes of CFC-11 contained within them.

End-use products, such as refrigerant and insulation foams for household refrigerator appliances or constructions, have durability and long lifespans. In this study, the Weibull distribution, which had been widely used in previous research (Liu et al., 2019; 2024; UNEP, 2021), is introduced to simulate the survival rate curves for new products with long lifespans (Eq. 1).

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

where $f(y,t)$ refers to the remaining rate of the sales of new products in year $y$; $t$ indicates the product age; $\beta$ corresponds to the shape parameter $(\beta > 0)$; $u$ is the scale parameter ($u > 0$). The quantity of CFC-11 mass in use or at EoL in each end-use application within each geographical region can be determined using Eqs 2–5:

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

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

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

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

where $Y$ represents the counted year of CFC-11 emissions; $y$ indicates the sales year of new product containing CFC-11; $q_{\mathrm{new}}(y,0)$ refers to the sales data of CFC-11 in new products in year $y$; $q_{\mathrm{use}}(y,t)$ denotes the quantity of in-use CFC-11 in the product in year $y$ with age $t$; $q_{EoL}(y,t)$ stands for the quantity of CFC-11 in the product sold in year $y$ with age $t$ entering into the EoL stage; $Q_{use}(Y)$ means the total quantity of in-use CFC-11 at year $Y$; $Q_{EoL}(Y)$ corresponds to the total quantity of CFC-11 in EoL products in year $Y$.

The emission mass is derived based on the annual mass of products containing CFC-11 during the first fill (manufacturing), use, and EoL stages. Following the Tier 2a method of the Intergovernmental Panel on Climate Change Guidelines for National Greenhouse Gas Inventories (IPCC, 2006; 2019), the historical and future emissions of CFC-11 from refrigeration systems can be estimated and projected using Eq. 6.

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

where $E_{ref}(Y)$ refers to the total emissions of CFC-11 from refrigeration systems in year *Y*; $E_{man}(Y)$, $E_{use}(Y)$, and $E_{\mathrm{EoL}}(\mathrm{Y})$ correspond to the mass emissions of CFC-11 from the manufacturing (first fill), use, and EoL stages of refrigeration equipment, respectively, at year *Y;* $Q_{\mathrm{new}}(Y)$ represents the sales of 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 $Ef_{\mathrm{use}}(Y)$ denote the EFs of CFC-11 during the life cycle stages of equipment manufacturing and use, respectively; $\gamma_{EoL}(Y)$ indicates the recovery efficiency of CFC-11 during EoL handling for obsolete equipment.

Similarly, the release of CFC-11 from closed-cell foams can occur throughout various life cycle stages, including manufacture, use, EoL handling, and post-life, such as landfilling disposal. Eqs 1–5 are used to calculate the CFC-11 bank and EoL flow from closed-cell foams. Eq. 7 is used to aggregate emissions across various life cycle stages originating from closed-cell foams.

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

where $E_{ccf}(Y)$ indicates the total emissions of CFC-11 from an end-use application of closed-cell foam in year *Y*; $E_{man}(Y)$, $E_{use}(Y)$, $E_{\mathrm{EoL}}(\mathrm{Y})$, and $E_{post}(Y)$ denote the mass emissions of CFC-11 from the manufacture, use, EoL handling, and post-life stages of closed-cell foams, respectively, at year *Y*. $Q_{\mathrm{new}}(Y)$, $Q_{use}(Y)$, and $Q_{EoL}(Y)$ refer to the same representation as in Equations (2–6); $Ef_{\mathrm{man}}(Y)$, $Ef_{\mathrm{use}}(Y)$, $Ef_{EoL}(Y)$, and $Ef_{post}(Y)$ are the EFs of CFC-11 during the life cycle stages of closed-cell foam manufacture, use, EoL handling, and landfilling, respectively; $l_{EoL}(Y)$ means the landfilling proportion of CFC-11 during the EoL handling of closed-cell foam, represented as (100%-$Ef_{EoL}(Y)$).

The prompt emissive use category includes applications that use CFC-11 in processes that result in immediate or relatively rapid release into the atmosphere (TEAP, 2019; 2021). A 6-month delay after sale is conventionally assumed in the case of aerosol and solvent uses, which results in half of the CFC-11 being released in the year of sale and the remaining half being released in the following year on average (McCulloch et al., 2001). The emission calculations are performed using Eq. 8.

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

where $E_{emi}(Y)$ denotes the total emissions of CFC-11 from prompt emissive uses in year *Y*; $Q_{emi}(Y)$ refers to the sales quantity of CFC-11 in prompt emissive uses; $p$ is the release fraction. In this study, $p$ is set to 0.5 to simplify the calculations. Given the global phaseout of CFCs since 2010, variations in the $p$ value will not affect the overall emission calculations beyond this point.

**2.2 Bottom-up Aggregated Emissions**

In addition to previously mentioned CFC-11 release from products, emissions of CFC-11 can occur during its production and packaging processes. A comprehensive literature review reveals that fugitive emissions resulting from the production of CFC-11 and its supply chain can contribute an additional 1.5%–5% to the overall production emission (TEAP, 2019; 2021; Eq. 9).

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

where $E_{pro}(Y)$ denotes the emissions from the CFC-11 production process in year *Y*; $Q_{pro}(Y)$ indicates the production quantity of CFC-11 in year *Y*; $r$ refers to the release rate of CFC-11 resulting from its production and supply chain. In this study, the value of $r$ is set at 3%, except for unreported production, which is set at 5%.

The emission profiles of each end-use category and its subdivision are characterized using a distinct set of parameters. We integrate these parameters into D-MFA model to estimate and project temporal and spatial evolution of CFC-11 emissions in accordance with Eq. 10.

$$\begin{aligned} E_{total}(Y) &= \sum E_{pro}(Y) + \sum E_{emi}(Y) + \sum E_{ref}(Y) + \sum E_{ccf}(Y) \\ &= \sum E_{pro}(Y) + \sum E_{emi}(Y) + \sum E_{man}(Y) + \sum E_{use}(Y) \\ &\quad + \sum E_{EoL}(Y) + \sum E_{post}(Y) \end{aligned} \quad (10)$$

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

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

### 2.3 Scenarios for Unreported CFC-11 Production

This study has developed three scenarios to address potential gaps in elevated CFC-11 emissions. Scenario 1 (S1) represents a baseline situation- an extreme situation- with no unreported production. Scenario 2 (S2) is the most likely situation, there was indeed unreported production of CFC-11 during 2014–2018. We assume microscale plants producing 0.1–2 Kt/yr CFC-11 as potential sources. According to the gap between upper limit of bottom-up and lower limit of top-down estimates, we hypothesize that the unreported production of CFC-11 during 2014–2018 was approximately 25 Kt annually, with a turning point in 2013 at 10 Kt. This setting can lead to an increase of 3-8 Kt/yr in CFC-11 emissions, depending on its application in PUR appliance or spray insulation foam. S3 indicates an extreme worst case of unreported CFC-11 production, approximately 23 ± 7 Kt/yr of emissions caused by the unreported production of CFC-11 during 2014–2018 (Lickley et al., 2021; TEAP, 2021). Given the various end uses of foam products, two subscenarios have been formulated under S2 and S3. Subscenario 1 assumes the use of unreported CFC-11 production in spray insulation foams (_SIF), which are characterized by substantial release fraction during their manufacturing phase. Conversely, subscenario 2 involves the use of unreported CFC-11 production in appliance insulation foams (_AIF), which results in lower release fraction during manufacture and a shorter product lifespan compared with construction spray foams.

### 2.4 Uncertainty and Sensitive Analysis

This section explores the range of effects of influential factors on CFC-11 emission levels. The factors considered include adjustments to EFs during various lifecycle stages, Weibull distribution parameters,

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and consumption fractions across different end-use sectors. The emission analysis is structured as follows: 1) The scale parameters ($u$), shape parameters ($\beta$), and EFs associated with manufacturing ($Ef_{\text{man}}(Y)$) and usage ($Ef_{\text{use}}(Y)$) are varied in turn based on their respective distributions, and other factors remain constant value 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 closed-cell foam products is adjusted, scaled down to 90%, and up to 110% of its original 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. Collectively, the emission levels falling within the range of uncertainty from the outlined cases are considered plausible, which reflects the complexity and variables inherent in the global CFC-11 emission landscape.

## 3 Results and discussion

### 3.1 Estimates of Banked CFC-11 Emissions and Implications

We estimate that the global annual emissions of CFC-11 steadily decreased in the past three decades (Fig.1a), excluding the unreported production. Figs. 1a–1b provide our bottom-up model results for the temporal dynamics of global and Chinese CFC-11 emissions in comparison with previous studies using top-down and combined approaches. We estimate the annual global CFC-11 emissions reached 43 (26–56) Kt during 2014–2018, significantly narrowing the gap with the top-down estimates of 69 (±10) Kt CFC-11 (Montzka et al., 2021). The estimate from TEAP (2019), 31 (20–45) Kt/yr, is lower than ours because we carefully considered multiple factors throughout the lifecycle of closed-cell foam products. Specifically, the lifespan parameter of closed-cell foam products depends not only on product attributes, but also on cultural, economic, and political factors, which will require further elucidation (Tables S2–3).

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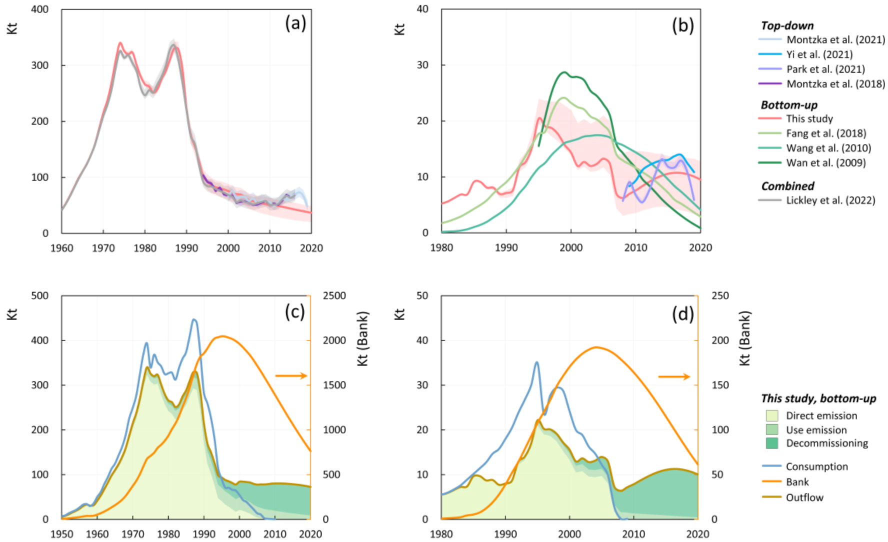


**Figure. 1 CFC-11 emissions comparison through bottom-up, top-down, and combined methods.** (a) Global CFC-11 emissions. (b) China's CFC-11 emissions. The red shaded regions indicate the 99% confidence interval in this study. (c) Global and (d) China's consumption, banks and outflows of CFC-11. Regarding decommissioning phase, CFC-11 contained in obsoleted foam products might either be released into the atmosphere (EoL emissions), disposed of by landfills or destroyed.

Closed-cell foam (CCF) products containing CFC-11 have served diverse insulation applications, mainly including 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 foam products in buildings exhibits significantly varied temporal and spatial trajectories (Table S3). We therefore adopt distributions to model key parameters associated with the lifecycle emissions of foam products over a long-term period (Table S13 and Fig. S13).

In China, the annual emissions of CFC-11 increased from 8 (4–13) Kt/yr in 2008–2012 to 11 (5–13) Kt/yr in 2014–2018 (Fig.1b) according to our bottom-up estimate. This estimate significantly differed from previous bottom-up estimates for China. Previous bottom-up models arbitrarily assumed that 10% of CFC-11 contained within foams were released during manufacturing, and the remaining 90% uniformly emitted over the subsequent two decades (Fang et al., 2018; Wan et al., 2009). This method has been recognized as inadequate since the late 1990s (McCulloch et al., 2001). Our bottom-up estimates have therefore aligned more closely with top-down assessments. For example, using top-down

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approaches, Park et al. (2021) estimated a 7 ± 4 Kt/yr increase in emissions in 2014–2017 compared with 2008–2012 from eastern China; Yi et al. (2019) projected that annual emission of CFC-11 in China started to increase from 8.3 ± 1.6 Kt/yr in 2009, peaked at 13.9 ± 2.4 Kt/yr in 2017, and then declined to 10.9 ± 1.7 Kt/yr by 2019. The TEAP (2019; 2021) studies explored the most likely routes for the unreported production of CFC-11 as follows: a) large-scale conversion of carbon tetrachloride (CTC) to CFC-11/12 in an existing plant with an annual capacity of ≥50 Kt, and b) microscale (0.1–2 Kt/yr) plants using minimal equipment to convert CTC to low-grade CFC-11 for foam blowing uses. In 2018, the Ministry of Ecology and Environment (MEE) of China undertook investigations and imposed penalties regarding the illegal production and use of CFC-11. It was found that only 177.6 metric tons of raw materials intended for CFC-11 production and 29.9 tons of illegally produced CFC-11 were found; a total of 1,172 combination polyether production enterprises were inspected, of which 10 small-sized enterprises were confirmed and punished (China MEE, 2019). There is ample evidence that such activities appear to have occurred at a microscale level (b), with no substantial indication of widespread illegal use for large-scale conversion (a).

The global annual quantity of decommissioned CFC-11 reached approximately 65 (±1) Kt during 2014–2018, when considering an average lifespan of 38-year for PUR boardstock and panel foams. This value falls within the distribution range (Fig. S13). Figs. 1c–1d depicts our refined assessments of CFC-11 banks and flows, both in the global and Chinese scales, between 2014 and 2018. Our estimates of decommissioned CFC-11 for this period were higher than the 45 Kt estimate from TEAP (2021). Some developing countries lack proper waste management systems, which may accelerate the release of CFC-11 from banks (Gómez-Sanabria et al., 2022; Liu et al., 2024). The transboundary movement of CFC-11 containing used appliances (or e-waste) from developed economies to developing countries (Martínez et al., 2022), which has not been included in this study due to insufficient data, might contribute to the flow of banked CFC-11 into developing parties. There is substantial uncertainty regarding CFC-11 release during EoL handling, with estimates varying from as low as a 20% release (UNEP, 2021) to 100% release (McCulloch et al., 2001; Liu et al., 2024). If all decommissioned CFC-11 were released into the atmosphere, our global bottom-up CFC-11 emissions would align with top-down estimates (Montzka et al., 2018; 2021). As shown in Fig. 1d, the decommissioned amount of CFC-11 in China rose from 5 Kt in 2008 to 10 Kt by 2014. Accordingly, China's average contributions to global CFC-11 banks and decommissioning were 10% and 16% during the 2014–2018 period, respectively. Global CFC-11 active

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banks (Fig.1c) peaked at 2,000 Kt (1,600–2,200 Kt) around 1995. This value can be comparable with the estimates by TEAP (2006; 2019) , but is slightly lower than the results by Lickley et al. (2022). The discrepancies in these estimates could be attributed to the differing breakdowns of CFC-11 consumption across various end-use applications. By 2020, the active bank of CFC-11 had decreased to 750 Kt (450–1,200 Kt), which is comparable with TEAP (2021) estimates as well.

### 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 a downward trajectory. This trend is consistent with top-down observations and falls within the estimated range of emissions (Hu et al., 2017).

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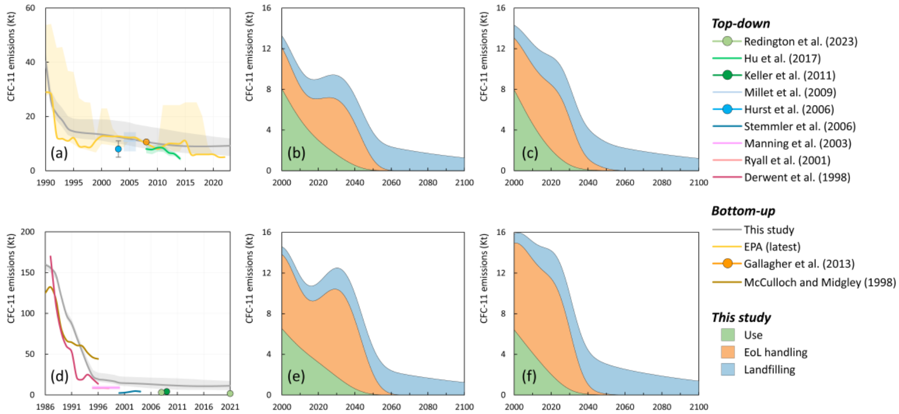


**Figure. 2 Comparison of CFC-11 emission estimates in the U.S. and non-A5 European parties**. (a) CFC-11 emission comparison for the U.S. based on bottom-up and top-down approaches in 1990–2022, respectively. The grey shaded regions indicate the 99% confidence interval in this study. The yellow shaded 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. CFC-11 emissions from end-use applications can be divided into those from prompt emissive uses and those from various lifecycle stages of CFC-11-containing products with long lifespans, including manufacturing, use, EoL handling, and landfilling. Key parameters for our bottom-up model, including lifespan and emission factors across life stages, were shown in Tables S7–8. (c), same as (b) but considering the sensitivity of some parameters, such as changes of lifespan from 58 to 38 years shown in Table S14. (d-f), same as (a-c) but for non-A5 European parties.

Estimates on CFC-11 emissions in Europe tend to cover diverse geographical extents (Table S1). However, non-A5 European parties and the North America show similarities regarding the historical use of CFC-11 in closed-cell foam (Hammitt et al., 1986; UNEP, 2019). Our estimated annual CFC-11 emissions among non-A5 European parties (Table S5) have dramatically decreased from 160 (157–165) Kt in 1986 to 15 (11–21) Kt by the year 2000, reflecting the complete phasing out of CFC-11 within the region. These estimates are comparable with both previous bottom-up (McCulloch et al., 1998) and top-down assessments (Derwent et al., 1998; Manning et al., 2003) within the margin of uncertainty (Fig.2d). Post-2000, emissions from banked CFC-11 have experienced a slight further decline to 11 (6–18) Kt by 2011, with projections suggesting a return to approximately 12 (5–16) Kt that persists for several years. According to SKM Enviros' estimates (SKM, 2012), the quantity of scrap foam products containing CFCs within the European Union (EU) decreased from 17 Kt around 2000 to 12 Kt by circa 2011 but subsequently increased again, reaching 15 Kt over several years. Accordingly, annual CFC emissions remained steady by approximately 6–7 Kt during this period. Considering diverse geographical coverage,

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our estimates align closely with those reported by SKM (2012). Moreover, based top-down method, Redington et al. (2023) reported the consistently increased emissions of CFC-11 from approximately 0.2 Kt in 2011 to 0.4 Kt in 2017 across the regions of northern France, Belgium, the Netherlands, and Luxembourg. 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.

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. S14). 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. S14a). 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. S14b) according to our estimate, which can coincide with the top-down emission level of 0.32 ± 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 Both Banks and Unreported Production

Although banked CFC-11 can probably result in an unexpected increase in emissions in 2014–2018, there are still direct emissions partly from unreported production of CFC-11 during this period. Accordingly, three scenarios have been assumed to assess future potential banks, emissions and impacts of CFC-11 by using bottom-up based model (Methods), considering both banks and unreported production. Figs. 3(a-d) present the banks, emissions, corresponding GWP value and atmosphere concentration of CFC-11 in different scenarios, respectively. As a reminder, scenario 1 (S1) hypothesizes that there was not any

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unreported production - an extreme situation. Scenario 2 (S2) is the most likely situation. According to the gap found between our bottom-up estimate and the top-down estimate during 2014–2018 (Fig.1a), there was indeed unreported production of CFC-11, which resulted in 3-8 Kt/yr direct emissions within this timeframe. Scenario 3 (S3)- the worst-case situation- assumes that approximately 23 ± 7 Kt/yr of CFC-11 emissions were caused by the unreported production of CFC-11 during 2014–2018, aligning with previous estimates by Lickley et al. (2021). Two sub-scenarios have been proposed under S2 and S3 which consider the unreported CFC-11 used in spray insulation foams (S2_SIF and S3_SIF) or in appliance insulation foams (S2_AIF and S3_AIF). Under S3, the estimated cumulative total of unreported CFC-11 production could be 390–1100 Kt. This estimate is slightly higher than the upper limit of 320–700 Kt reported by TEAP (2021), primarily arising from differences in direct emission rates associated with unreported production of CFC-11 and manufacturing of foam products.

Under S1, global time-lagged CFC-11 emissions will accumulate to 890 (550–1350) Kt - equivalent to 4.2 (2.6–6.4) Gt $CO_2$e - till 2100. Regarding S2, the global cumulative emissions of CFC-11 for the period 2025–2100 are projected to be 1000 (700–1300) Kt -equivalent to 4.6 (3.2–6.3) Gt $CO_2$e. However, emissions of CFC-11 under S3_SIF and S3_AIF are substantially higher than those projected for S2. In the S3_AIF scenario, the cumulative unreported production of CFC-11 is projected to result in an increase of 950 (±40) Kt of the banked CFC-11 by 2018, which implies that a second peak -followed the peak around 1995- of CFC-11 banks would occur in 2018 by 1,900 (1400–2,200) Kt (Fig.3a). Consequently, CFC-11 emissions are expected to increase again from 2020, peaking at 68 (45–100) Kt by 2027 (Fig.3b). This implies that the global cumulative GWP values would reach to approximately 7.5 (4.9–10.5) Gt $CO_2$e during 2025–2100 under the S3_AIF_BAU conditions, corresponding to 18% (12–26%) of total greenhouse gas emissions globally in 2023 (Friedlingstein et al., 2023). Our analysis suggests that mean mole fractions of CFC-11 remain below the WMO (2022) estimates, except for S3_AIF (Fig. 3d). However, this scenario (S3) is less likely, offering a more optimistic perspective on both the climate change impact and the recovery trajectory of the ozone layer.

Currently, the proportion of emissions resulting from the EoL handling of obsolete CFC-11-containing products is higher than those emitted during the landfilling stage (Figs. 2b-c, and 2e-f). However, by approximately 2050, CFC-11 released from landfilling will become the dominant source. Proper management of EoL handling processes can efficiently reduce CFC-11 emissions. Three different management practices have been considered to evaluate future mitigation potentials: i) a business-as-

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usual (BAU) scenario, which includes ongoing partial landfilling or recovery of CFC-11 associated with EoL handling; ii) an advanced scenario in which obsolete products will be collected and properly treated, aiming to destroy 85% of CFC-11 in obsolete foam products from 2025 onwards (EU, 2012); iii) a similar scenario as ii but starting from 2030. These cases are aligned with previously defined most likely scenarios S2_SIF and S2_AIF, resulting in a comprehensive matrix of 6 sub-scenarios denoted by unique identifiers.

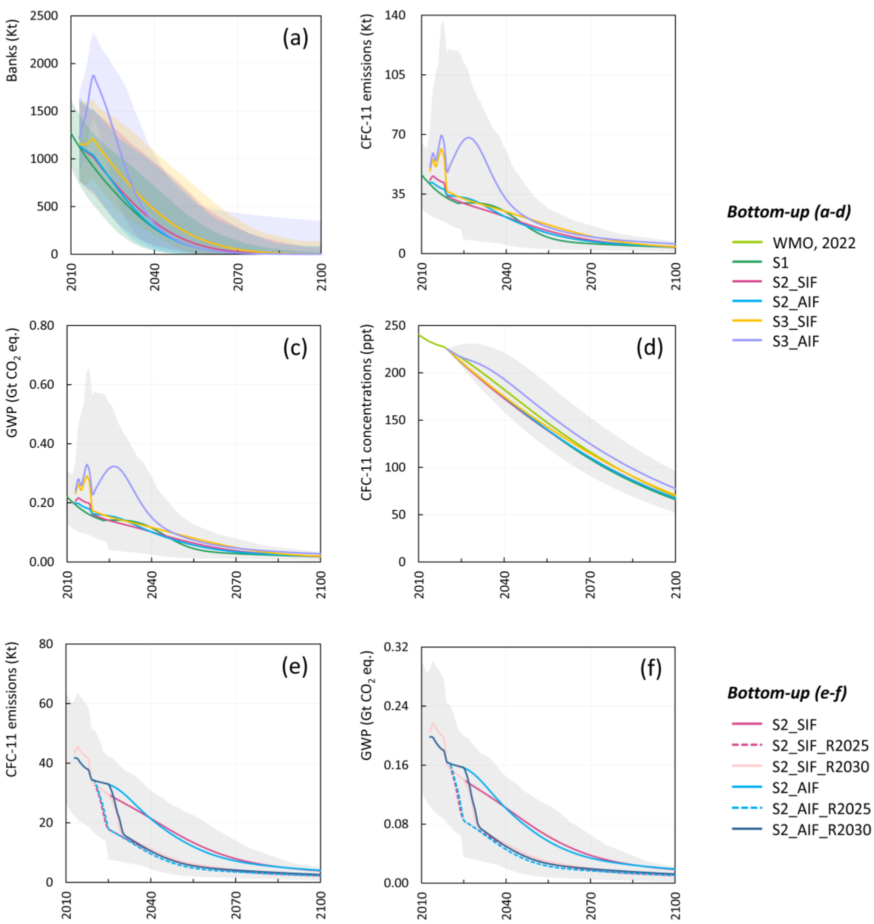


**Figure. 3 CFC-11 banks, emissions and impacts considering unreported production and their mitigation potential**. (a) CFC-11 banks based on scenario analysis. (b)–(c) CFC-11 emissions and their global warming potential (GWP)-weighted emissions in terms of 100-year time horizon subject various scenarios. (d) CFC-11 concentrations in part per trillion (ppt) under various scenarios. The assumed constant lifespan of CFC-11 is 52 years. (e)–(f) Potential mitigation of CFC-11 emissions (e)

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and their GWP-weighted emissions (f) under our scenario 2 (S2). The grey shaded regions indicate the 99% confidence interval.

Fig. 3e-3f illustrates the mitigation potential of CFC-11 and their $CO_2$-equivalent ($CO_2$e) value over a 100-year period under different S2 scenarios. Implementation of globally well-managed EoL handling starting from 2025 has the potential to reduce cumulative CFC-11 emissions by 50–52% compared with the BAU scenario between 2025 and 2100. If well-managed EoL handling is delayed until 2030, cumulative emissions will decrease by 42% in the future. Compared with the CFC-11 used in building insulation foams, the CFC-11 used in appliance insulation foams offers greater potential for emission reduction through effective EoL handling. Specifically, S2_AIF_2025 demonstrates a higher capacity for reducing EoL emissions than S2_SIF_2025. In addition, well-managed EoL handling from 2025 can potentially reduce cumulative $CO_2$e emissions by 2.3 Gt under S2_SIF_2025 and 2.4 Gt under S2_AIF_2025 compared with S2_BAU scenario. Comparing the scenarios with well-managed EoL handling from 2030 to those from 2025, the latter can potentially further decrease $CO_2$e emissions by an additional 0.4–0.5 Gt, which underscores the importance of implementing prompt action as early as possible. However, this condition also indicates that advanced strategies will be required to mitigate approximately 2.2–2.7 Gt $CO_2$e emissions from the landfilling stage 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 (Fig.4b) and EFs during manufacture (Fig.4c) and use (Fig.4d) 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 (Fig.4g-h) attributable to the overall uncertainty in scale parameters of the Weibull distribution (Fig.4a), which are highly sensitive to changes in average product lifespan. Moreover, the emissions during the EoL handling of obsolete products exhibit notable disparities (Fig.4e-f). These discrepancies underscore the need for the refinement of the current understanding of emissions from EoL handling and landfill disposal. 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

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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.

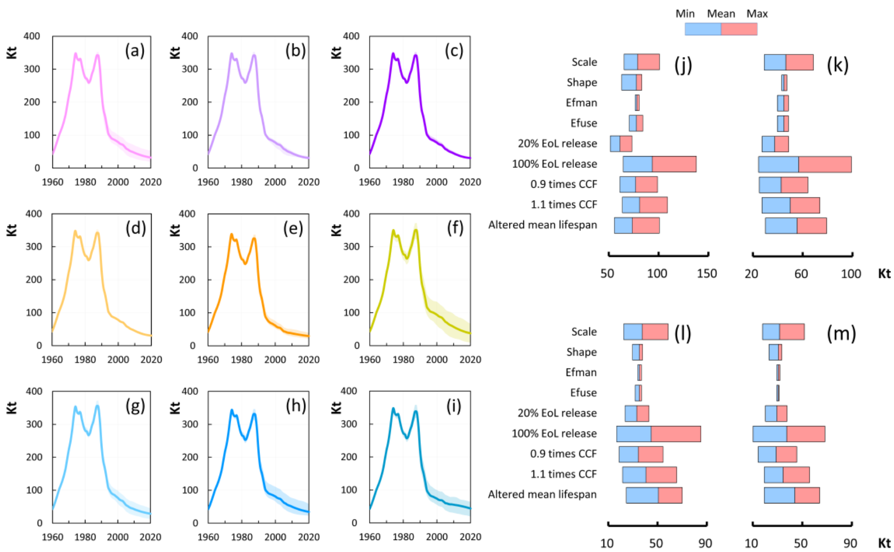


**Figure. 4 Uncertainty and sensitivity analysis of CFC-11 emissions through bottom-up modeling**. (a)–(f) Uncertainty arising from various parameters. (a) Scale parameters, similar to the average lifespan of foam products, vary based on probability distributions, whereas other parameters remain constant. (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. (k)–(m) same as (j) but for the year 2010 (k), 2015 (l) and 2020 (m), respectively. The shaded regions indicate the 99% confidence interval.

## 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. The results indicate that the unanticipated increase in CFC-11 emissions observed between 2014 and 2018 can be partially attributed to existing CFC-11 banks. Through D-MFA tracking of CFC-11 emissions across various lifecycle stages, our analysis further illustrates that the trajectory of bank-derived CFC-11 emissions may exhibit either upward or downward

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trends due to uncertainties surrounding foam product lifetimes and EoL handling practices. These trends are consistent with prior large-scale observations.

Top-down and bottom-up modeling approaches display an inherent association with uncertainties. Bottom-up modeling approaches involve multiple parameters and key processes that collectively contribute to the overall uncertainty in emission estimation. 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 concerning temporal and spatial variations in product lifespans and release patterns during EoL handling and landfill disposal, are imperative to address the inherent uncertainties linked to bottom-up modeling approaches.

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 and accuracy in quantifying emission dynamics. Although this study primarily focuses on CFC-11 emissions, most of the conclusions may also be applicable to other ODS and hydrofluorocarbon emissions.

**Code and data availability**

All data inputs, including regional production statistics, product lifespan datasets, emission factors and end-of-life handling metrics, are available in the Supplementary Information.

**Author contributions**

H.L., H.D. and Y.M. conceptualized and designed the study, conceived the paper, developed the model. H.L. and Y.M. collected the data and conducted the analysis. N.Z. drew the figures. H.L., H.D., N.Z., T.R.M. and M.X. contributed to the conceptual model development. R.M., M.X. and J.Y. enhanced scenario analysis discussions. H.D., H.L., N.Z., R.M., Y.M., T.R.M., M.X. and J.Y. contributed to

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discussing the results and writing the paper.

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**Declaration of interests**

The authors declare no competing interests.

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