# Response to reviewers for preprint egusphere-2025-2277:

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

## RC#2

#### Comment #1

General Comments: This study presents an updated bottom-up inventory of CFC-11 emissions globally, in an effort to better understand the cause of the well-documented, 'unexpected' rise in global emissions in the period 2014-2018. It employs a dynamic material flow analysis model to estimate aggregated emissions from the entire lifespan of CFC-11 containing products, such as closed-cell foams and refrigeration systems, as well as direct emissions from other uses. The authors estimate emissions that are broadly in line with previous work, although discrepancies are found in some places. This is attributed to a more thorough consideration of the different stages in the lifecycle of CFC-11 products. They also conclude that some of the unexpected increase between 2014 and 2018 could be attributed to increased emissions from existing banks, although unreported production also plays a role.

The model that the authors have developed is sound, and the application of Weibull survival functions to estimating global banks and emissions of CFC-11 is a sensible approach. However, I see a number of issues with this manuscript.

- Firstly, the method requires more rigorous analysis of uncertainties, and many of the parameters used to estimate emissions are not justified sufficiently.
- In addition, it is not clear to me, based on the evidence presented, how the authors justify their primary conclusion, that the global emissions rise seen between 2014 and 2018 can be attributed in part to emissions from banks.
- Finally, the results and discussion section is poorly structured and confusing. This makes it hard to determine the validity and significance of the authors' conclusions.

Given these major concerns, I don't think the current version warrants publication in ACP. However, if the authors can fully address the above issues, a revised version may be publishable, subject to further review.

Reply: We sincerely grateful your detailed and constructive feedback on our manuscript. Your time and effort in providing such valuable insights are greatly appreciated.

Regarding your first point concerning the analysis of uncertainties and the justification of parameters, we acknowledge the need for enhanced clarity and rigor in the Monte Carlo

simulations. We have carefully refined the methodology section to include more detailed explanations regarding the selection of parameters and their associated uncertainty ranges (please refer to our response to Comments #6, #7, #8, and #18).

For your second concern about attributing the 2014–2018 global emission rise in part to banks, we have expanded the discussion to provide additional clarification and further rationale supporting this conclusion (see our response to Comments #14).

Regarding the organization of the results and discussion section, we have restructured the content to enhance clarity and logical coherence, ensuring the significance and robustness of our findings are clearly presented (see our response to Comments #19).

Each of your recommendation has been carefully reviewed and integrated into the revised manuscript with due consideration. We are dedicated to refining the manuscript under your expert guidance and are confident that these revisions significantly enhance its overall quality and strengthen its suitability for consideration in *Atmospheric Chemistry and Physics* (ACP).

#### Comment #2

Specific comments

L95: The description of the Weibull distribution does not make clear the significance of the shape and scale parameters. Details about what values are chosen are given in the supplementary information, but these are not cited or justified anywhere. In addition, it isn't clear the role of 'y' in this function. Are the shape and scale parameters functions of the year? If they are, this should be explained clearly, and if not, then f isn't a function of 'y' at all.

Reply: Thank you for your valuable comments on the description of the Weibull distribution. In the main text, we kept the exposition concise, as this distribution is well established for modelling the lifespan behavior of durable products (e.g., Liu et al., 2024; Mueller et al., 2007; TEAP, 2021). However, we fully agree that further mythological detail would be beneficial, and we have accordingly expanded the relevant content.

As suggested, we have added the following text in lines 104–107 of the main text:

"Parameter estimates for foam products and refrigeration equipment across different geographical regions were derived from our surveys (Duan et al., 2018; Liu et al., 2024) and an extensive literature review (e.g., McCulloch et al., 2001; FTOC, 2002; IPCC, 2006; TEAP, 2021)."

The following text has been added in the SI, lines 484–498:

"Reliable lifespan distribution data are essential for quantifying banks and flows. While TEAP (2019) relied on fixed mean lifespans for modeling CFC-11 emissions, TEAP (2021) subsequent adopted the Weibull distribution to better represent lifetime variability in the retirement patterns of refrigeration units and foam products. In this study, we similarly employ the Weibull distribution to characterize lifespan patterns for these categories. The Weibull parameters offer critical insights into the dynamic scrapping behavior of durable products. Specifically, the shape parameter defines the distribution skewness, indicating whether scrapping is more likely early (shape < 1), random (shape = 1), or delayed (shape > 1). The scale parameter, in turn, serves as an estimate of the average lifespan. Regional parameter estimates for foam products and refrigeration equipment were obtained from our surveys (Duan et al., 2018; Liu et al., 2024) and an extensive literature review (e.g., McCulloch et al., 2001; FTOC, 2002; IPCC, 2006; TEAP, 2021). Figure S13 illustrates the resulting remaining rate of refrigeration equipment over their lifespan, using the region-specific Weibull parameters listed in Table S7.

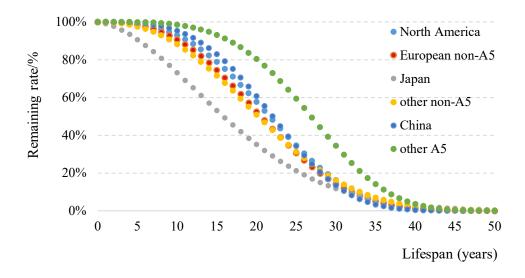


Figure S13 Curves depicting the correlation between remaining rate and lifespan of nonhermetic refrigeration equipment across different geographical regions"

#### Comment #3

L145: No justification is given for the choice of values for the release rate from production, nor why the illegal and legal production are different.

Reply: Thank you for your attention to this detail. As noted in TEAP (2019), a release rate of 1.5% during CFC-11 production may better reflect realistic conditions, with potential

additional emissions of 1–3% from the supply chain. TEAP's multi-scenario analyses demonstrate that production-stage release rates have a negligible effect on total emissions (TEAP, 2019; 2021). Their model evaluated emission rates across a wide range (0.5–5%, and even higher values), finding that variations within this range do not significantly alter overall emission estimates. Therefore, to maintain model parsimony, we adopted fixed release rates of 3% for reported production and 5% for unreported production.

As suggested, we have strengthened the rationale for using uniform rates by adding the following text in lines 154–155 of the revised main text:

"TEAP's multi-scenario assessments indicate that production-stage release rates have minimal impact on total emissions (TEAP, 2019; 2021). Therefore, in this study..."

#### Comment #4

L157: None of the data used to drive the model is presented in the main text. A summary of sources or perhaps a visualisation could be helpful.

Reply: Thank you for your suggestion.

As recommended, we have revised the main text, lines 167–174:

"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 closed-cell foam 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 closed-cell foams and non-hermetic systems (including refill volumes) from total global consumption."

# Comment #5 and Comment #10

L168: Claiming that unreported production is the 'most likely' scenario seems inappropriate, since the abstract claims that at least some of the increase is due to bank emissions. While there is a need for an intermediate scenario, there seems to be no justification for choosing 25 kt/yr as a value for the unreported production of CFC-11. Indeed, later on the authors go on to say that it is the highest emissions scenario (S3) that aligns with previous work.

L231: Again, the discussion of the likely sources of unreported emissions feels out of place here. It might be better placed in the discussion, or in the section where the authors model potential scenarios of unreported production.

Reply: We sincerely appreciate this insightful critique regarding scenario development and aim to address these related points comprehensively.

We agree that labeling Scenario 2 (S2) as the "most likely" scenario was inappropriate. Consequently, all such claims have been removed throughout the manuscript. S2 is now explicitly designated as our intermediate scenario—a designation justified by the systematic lack of mid-range estimates for unreported CFC-11 production in existing literature.

The discussion of potential sources of unreported emission, originally starting at Line 231, has been relocated to the 'Scenarios for unreported CFC-11 production' section. This repositioning enhances analytical continuity.

As suggested, revisions have been implement in the main text, lines 204–215:

"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. 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)..."

## Comment #6

L185: The 'distributions' referred to can only be found in the SI, and it is not clear how they are varied. What sort of sampling is done? A more sophisticated approach for sampling across parameter distributions, such as a Monte Carlo method, might be appropriate here.

Reply: Thank you for this valuable critique. Our study applies the Monte Carlo method to simulate the distributions of key parameters—specifically, Weibull distribution variables and emission factors (EFs) for foam manufacturing and use—that significantly influence CFC-11

emission levels and bank estimates, in alignment with TEAP (2019, 2021). The following detailed description has been added to the revised main text, lines 179–192:

'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  $(\beta)$  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 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  $(\beta)$  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 were 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. Uncertainty ranges are reported as the 0.5th to 99.5th percentile interval, corresponding to 99% confidence interval.'

## Comment #7, Comment #8, and Comment #18

L187: Why is 20% chosen? Could a range of values or a distribution be sampled instead? The same goes for 90% and 110% in L190

L191: I don't follow what has been done here. Does it mean that the parameters are simply set to the values in Table S14? Are these global averages?

L398: It is not clear what is being varied in some cases. In the 20%/100% EoL case and 1.1/0.9x CCF cases, this is clear – but are the other parameters being simultaneously varied? If not, how are a mean, minimum and maximum calculated? For the shape/scale parameters, are all the different parameters (for different products and regions) varied simultaneously? As mentioned above, this seems like a case in which a Monte Carlo method would be suitable. The nine graphs in figure 4 are not a clear way of explaining the impact of each of the parameters, as they are all very similar in shape and the subtle differences are hard to spot. The right hand side of the figure is a more intuitive way of understanding the impact of varying these parameters. As with previous figures, the meaning of the 99% confidence interval is not clear, either.

Reply: We have consolidated our responses to Comments #7, #8, and #18 regarding uncertainty and sensitivity analysis below for coherence.

## i) Lower bound for end-of-life (EoL) emission factors

The 20% lower bound for EoL emission factors is derived from TEAP's global assessment of CFC-11 emissions from foam products (TEAP, 2021). The 100% upper bound represents the maximum plausible emissions from retired products in unmanaged waste streams—a scenario widely adopted in previous studies (e.g., McCulloch et al., 2001; Duan et al., 2018).

Regional compilations (Tables S8–S13) provide intermediate values. For example, sector-specific factor in the U.S. range from 35% for appliance insulation to 100% for spray insulation foams (Table S8), derived from the U.S. Environmental Protection Agency (EPA) 2024 Greenhouse Gas Inventory Annual Report. Similarly, in Japan, EoL EFs range from 10% (panel insulation foams) to 100% (appliance insulation foams; Table S10), based on the annual report from the Japan Ministry of Economy, Trade and Industry (Japan METI). Comparable regional variations, based on literature synthesis, field surveys, and assumptions, were applied to other regions. Global estimates reflect weighted regional averages, inherently incorporating intermediate levels (Table S13). The variation of this factor is quite significant, making it challenging to define a specific value distribution for sampling purposes.

## ii) Uncertainty in the historical CFC-11 allocation to closed-cell foams

Historical production and consumption totals provided by AFEAS and TEAP are considered as robust. However, the sectoral split of CFC-11 among closed-cell foams, refrigeration, aerosol and other emissive uses carries a recognized uncertainty. Following established practice, we adopt 90% and 110% of baseline as representative uncertainty bounds for the quantity allocated to closed-cell foams.

# iii) Uncertainty and sensitivity analysis framework

To quantify parameter-specific contributions to emission uncertainty, we designed a stepwise framework analyzing the following factors: Weibull scale parameter (u), shape parameter  $(\beta)$ , manufacturing-stage EF  $(Ef_{man}(Y))$ , use-stage EFs  $(Ef_{use}(Y))$ , EoL handling EF (set to 20% or 100% of CFC-11 retained in obsolete foam products), and CFC-11 quantities allocated to closed-cell foams (set to 90% or 110% of baseline).

For parameters with defined distributions, we conducted strict single-factor uncertainty analyses. 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 values. Annual emissions were aggregated to derive the global CFC-11 emission distribution, with uncertainty characterized by the 0.5th–99.5th percentile range (representing the 99% confidence interval). Following the same approach, we sequentially performed single-parameter analyses for shape parameter ( $\beta$ ), manufacturing-stage EF ( $Ef_{man}(Y)$ ), and use-stage EFs ( $Ef_{use}(Y)$ ).

For factors lacking probabilistic distributions—specifically EoL handling EFs and CFC-11 allocation quantities—we implemented bounded uncertainty analyses. Within these, Weibull parameters  $(u, \beta)$  and manufacturing/use-stage EFs  $(Ef_{man}(Y), Ef_{use}(Y))$  were independently sampled from their subsector-specific distributions, while EoL EF was systematically set to either 20% or 100%, with other parameters fixed at baseline values. Similarly, to assess the impact of variations in CFC-11 quantities allocated to closed-cell foams (i.e., 90% or 110% of baseline), we integrated simultaneous random sampling of parameters u,  $\beta$ ,  $Ef_{man}(Y)$ ,  $Ef_{use}(Y)$  with these allocation adjustments, while fixing other parameters at their baseline values.

We considered the 38-year global average lifespan for boardstock/laminate and panel products (Table S14) as plausible. However, this value falls within the range of the lifespan distribution (Figure S14). Thus, this factor has been removed from the uncertainty and sensitive analysis. Instead, emissions under the baseline scenario (S1) served as the reference values for comparative assessment.

We have expand the 'Uncertainty and sensitivity analysis' section with additional methodological detail. Please refer to the revised main text, lines 232–246:

"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 emission distribution, with uncertainty characterized by the 0.5th–99.5th percentile range (representing the 99% confidence interval). Following the same approach, we sequentially performed single-parameter analyses for shape parameter ( $\beta$ ), manufacturing-stage EF ( $Ef_{man}(Y)$ ), and use-stage EFs ( $Ef_{use}(Y)$ ). For factors lacking

probabilistic distributions, specifically EoL EFs and CFC-11 allocation quantities, we implemented bounded analysis. Weibull parameters  $(u, \beta)$  and manufacturing/use-stage EFs  $(Ef_{man}(Y), Ef_{use}(Y))$  were independently sampled from subsector-specific distributions, while EoL EFs were set to either 20% or 100%, with other parameters held at baseline/mean values. Similarly, to assess variations in CFC-11 quantities allocated to closed-cell foams (i.e., 90% or 110% of baseline), we integrated simultaneous random sampling of parameters u,  $\beta$ ,  $Ef_{man}(Y)$ ,  $Ef_{use}(Y)$  with these allocation adjustments, while fixing other parameters at mean values. Emissions under the baseline scenario (S1) served as reference values for comparative assessment."

We have also revised Figure 4 to retain exclusively the original right-hand content:

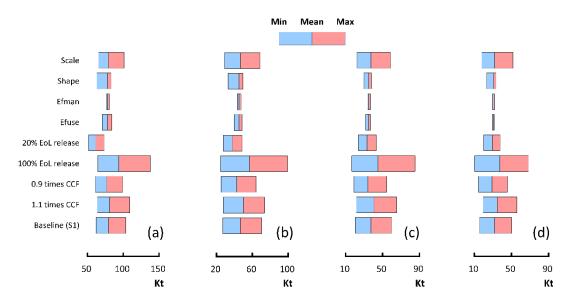


Figure 4 Uncertainty and sensitivity analysis of CFC-11 emissions through bottom-up modeling. (a)–(d) Emission uncertainties induced by various factors for the years 2000 (a), 2010 (b), 2015 (c) and 2020 (d), respectively, closed-cell foam (CCF)

## Comment #9

L213: This paragraph (L213-220) feels like it should be in the Methods section. The 'temporal and spatial trajectories' are clearly important to the results, as they lead to the divergence from other bottom-up inventories. It would be good to see these discussed or tabulated in the Methods section.

Reply: We appreciate this constructive suggestion. Relocating the description (original Lines 213–220) to the Methods section is well-justified, as it provides essential context for subsequent divergence from other bottom-up inventories.

Following your recommendation, we have integrated this paragraph into the Methods section of the main text, lines 86–91:

"CFC-11-containing CCF products have served diverse insulation applications, primarily polyurethane rigid (PUR) boardstock, panel, spray, appliance, and other insulation foam products (S1.2). PUR boardstock foams have been widely used in residential and commercial roof insulation, as well as walls of metal and agricultural buildings. PUR panel foams have been commonly used in industrial settings, such as refrigerated warehouses. The investigated lifespan of these foam products in building contexts exhibits notable spatiotemporal variability (Table S3)."

#### Comment #11 and Comment #12

L242-L262: I find this section slightly confusing. The term 'decommissioned' CFC-11 has not been explained in the introduction or methods section, and is only mentioned in reference to figure 1c. Showing both banks and emissions on the same figures (1c and 1d) make them hard to interpret, and it is not clear which lines/shaded areas should be read off the left and right axes. The orange arrow is not explained, either.

L253: '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)'. It is unclear what time period this refers to, but if it refers to 2014-2018 then this appears to be the primary conclusion of this paper. However, it is not given any further development in this section nor in any of the figures.

Reply: We have consolidated our responses to Comments #11 and #12 regarding figure 1(c) and (d) below for coherence.

The term 'decommissioned' refers to retained CFC-11 in obsolete products. To enhance conceptual clarity, this term has been consistently replaced throughout the manuscript, and figure 1 has been revised accordingly. Specifically, the orange arrow has been removed from Figure 1. In panels (c) and (d), bank quantities (orange lines) correspond to the right-hand axis (orange scale). Annual outflows comprise direct emissions from chemical production/supply chain, emissive uses, end-use product manufacturing, as well as product use-stage emissions and residual CFC-11 from obsolete products. This residual fraction undergoes multiple EoL pathways, covering immediate release during dismantling/shredding/compacting, gradual release flowing landfilling, recovery for reclamation/reuse or intentional destruction. Total annual outflow represents CFC-11 emitted or transferred within the reporting year and, by mass balance, equals or exceeds annual

emissions. Our analysis yields 2014–2018 use-stage emissions averaging 13±2 kt/yr, while CFC-11 contained in obsolete products reaches up to 65±1 kt/yr.

# Please refer to the revised figure 1 below:

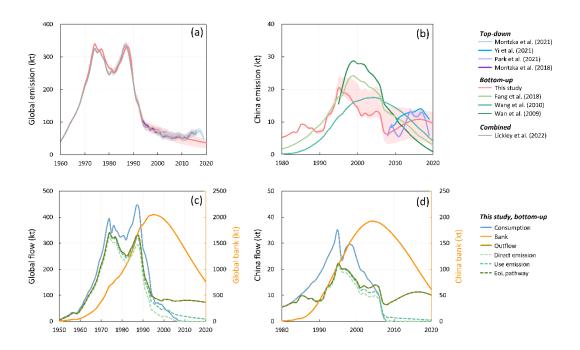


Figure. 1 CFC-11 emissions comparison through bottom-up, top-down, and combined methods.

(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 EoLflows associated with obsolete products. These EoL flows may undergo atmospheric release (EoL emissions), landfilling (post-life release), or destruction/reclamation.

For clarity, we have revised the relevant paragraph as follows (lines 283–294):

"Figs. 1c–1d depicts our refined assessments of CFC-11 banks and flows at both global and Chinese scales, incorporating a 38-year average lifespan for PUR boardstock and panel foams. This lifespan falls within the distribution range for such foams (Fig. S13). Our analysis yields 2014–2018 use-stage emissions averaging 13±2 kt/yr, while CFC-11 contained in obsolete products reaches up to 65±1 kt/yr. In developing countries, inadequate waste management systems may accelerate CFC-11 release from obsolete product (Gómez-Sanabria et al., 2022; Liu et al., 2024). In addition, the transboundary movement of used appliances (or e-waste) containing CFC-11 (excluded here due to data limitations) from developed to developing

economies could redistribute EoL flows (Martínez et al., 2022). Substantial uncertainty exists regarding 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 would bring our global bottom-up CFC-11 emissions into closer alignment with the top-down estimate of  $69 \pm 10$  kt/yr (Montzka et al., 2018; 2021)."

## Comment #13

L302: The text says that the new estimates are comparable with the McCulloch, Derwent and Manning estimates within the margin of uncertainty. However, figure 2 suggests that this is only true for a small number of years. The McCulloch estimates fall within the uncertainties of the present study for only one or two years between 1986 and 1996, the Derwent estimates for only two or three of those ten years, and the Manning estimates don't appear to overlap at all with the present study. This is in contrast to the China estimates in figure 1b, where the authors describe the estimates as 'significantly differing' from the current work despite greater overlap with the plotted confidence intervals.

Reply: We appreciate the opportunity to enhance clarity regarding estimates of European CFC-11 emissions, which exhibit geographical variability across studies.

Please refer to the revised text in the main text, lines 328–339:

"Estimates of European CFC-11 emissions exhibit significant geographical variability (Table S1). For non-A5 European parties (Table S5), our analysis indicates emissions declined sharply from 160 (157-165) kt/yr in 1986 to 15 (11-21) kt/yr by 2000, consistent with regional phase-out commitments. This trend aligns with prior studies (McCulloch et al., 1998; Derwent et al., 1998), though emission discrepancies arise from differences in geographical scope and methodology. For instance, McCulloch et al. (1998) applied bottom-up modeling to European Union (EU) emissions, assuming fixed release rates of 10% during manufacturing and 90% uniformly emitted over two decades. Our divergent estimates (Fig. 2d) stem from broader geographical coverage (non-A5 Europe versus EU) and dynamic emission modeling. Post-2000 emissions continued declining to 11 (6–18) kt/yr by 2011, stabilizing near 12 (5–16) kt/yr in subsequent years. Industry data suggest comparable historical CFC-11 usage levels in closed-cell foams across the EU and North America during the 1970s-1980s (Hammitt et al., 1986; FTOC, 1993). Thus, our slightly higher estimates for non-A5 Europe versus the U.S. post-2000 are reasonable. In addition, according estimates by SKM Enviros..."

Please refer to the revised text in the main text, lines 265–266:

"Our estimated emission trend over this period differs significantly from previous bottom-up estimates for China."

#### Comment #14

L333: 'banked CFC-11 can probably result in an unexpected increase in emissions in 2014-18'. What does 'probably' mean in this case? As mentioned above, this has not been developed and I'm not convinced that it is a valid conclusion, especially given the trend shown in figure 1a. That figure shows emissions on a steady downward trend from 1995 onwards, with no suggestion of an increase after 2014 globally.

Reply: We have removed the term 'probably'. Banked CFC-11 constitutes a persistent emission source capable of driving unexpected increases, as evidenced by regional studies: the area including northern France, Belgium, the Netherlands, and Luxembourg (Benelux) showed consistently elevated emissions of CFC-11 (Redington et al., 2023); Australia recorded a 33% increase, from  $0.48 \pm 0.04$  kt/yr (1996–2003) to  $0.64 \pm 0.07$  kt/yr during 2004–2008 (Dunse et al., 2019). Both regions are non-A5 parties that ceased CFC production prior to 2000.

Our bottom-up modeling reveals that regional emission trajectories—such as those in the U.S. and Europe—are influenced by the lifespan of CFC-11-containing foam products. Moreover, the lifespans of polyurethane (PUR) foams exhibit considerable spatiotemporal variability, which may lead to either an upward or downward shift in estimated emission trends. This explains the observed increase in emissions in the Benelux region and Australia following the phase-out of CFC production and consumption.

Globally, our results can be calibrated to align with the 2014–2018 top-down emission trends through adjustments to Weibull scale/shape parameters or EoL emission fractions. However, given the annual regional fluctuations in these parameters (SI Table S3–4), we prioritize uncertainty quantification over strict calibration of annual trend. The dynamically fluctuating emission ranges depicted in Figure 1a represent physically plausible outcomes. Within the uncertainty ranges in Figure 1, divergent trends—either an upward or downward shift—may also be reflected globally. Specifically, as illustrated in Figure 1c, our analysis indicates 2014-2018 use-stage emissions averaging  $13\pm2$  kt/yr, while CFC-11 contained in obsolete products reaches up to  $65\pm1$  kt/yr. An increased EoL emission factor shifts bottom-up emissions closer to the top-down estimate of  $69\pm10$  kt/yr (Montzka et al., 2021). Meanwhile, historical emissions and bank magnitudes remain consistent with previous assessment.

Please refer to the revised version in the main text, lines 371–372:

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

## Comment #15 and Comment #17

L342: It appears that limited assessment is done of how well each scenario would account for the unexpected emissions, in combination with varying the parameters of the main model. This seems to me to be a crucial element of the analysis that is missing.

L376: There are a lot of issues with this figure. The overlapping uncertainty intervals make subfigure (a) unreadable. Subfigures (b) and (c) are identical – the only difference being a simple rescaling of the y-axis, and the same goes for (e) and (f). There is no discussion of how the 99% confidence interval is calculated, either.

Reply: Thank you for your construction comments. Since Comments #15 and #17 pertain to scenarios and their visual presentation, we would like to provide a consolidated response. We have revised Figure 3 to include more detailed information regarding unexpected emissions. Efforts have been made to enhance the readability of subfigure (a). Panels (b) and (c) have been consolidated into a single optimized subfigure; a similar approach has been applied to subfigures (e) and (f). As previously explained, uncertainties in the manuscript are characterized using the 0.5th–99.5th percentile range (99% confidence interval).

## Please refer to the revised Figure 3:

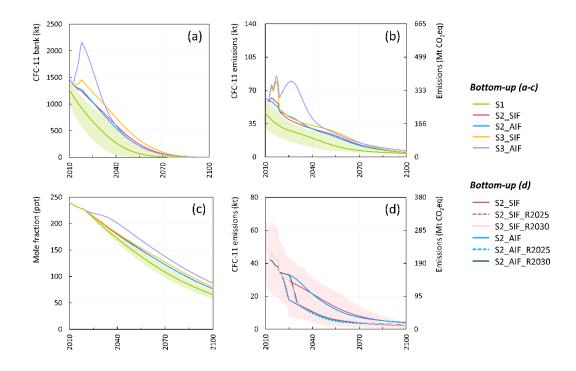


Figure 3 CFC-11 banks, emissions and impacts considering unreported production and mitigation potential. (a) CFC-11 banks based on scenario analysis. (b) CFC-11 emissions and corresponding CO<sub>2</sub>-equivalent (CO<sub>2</sub>-eq) emissions under various scenarios. (c) CFC-11 global mole fractions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

#### Please refer to the revised version in the main text, lines 375–398:

"Under S1, global CFC-11 banks declined from 1270 (1000–1500) kt in 2010 to 1040 (720–1250) kt in 2015, and further to 680 (350–950) kt in 2025, with projections indicating a continued decrease to 120 (10–300) kt by 2050. These estimates closely align with those reported by TEAP (2021) and are validated by TEAP's (2019) independent assessment using Ashford et al. (2005) methodology, to estimate 2015 banks at 1070–1292 kt. Cumulative time-lagged global emissions for 2025–2100 under S1 reach 890 (550–1350) kt, equivalent to 4.2 (2.6–6.4) Gt CO<sub>2</sub>.

Under S2, unreported production totaled 135 kt during 2013–2018, increasing the 2019 bank by up to 120 (70–130) kt (S2\_SIF) and 90 (75–105) kt (S2\_AIF). These values are slightly higher than estimated 75 (46–112) kt for China but lower than TEAP assessments of 320–700 kt (Park, et al., 2021; TEAP, 2021). By 2030, this bank increase declines to 75 (45–90) kt (S2\_SIF) and 45 (0–105) kt (S2\_AIF), falling further to 40 (0–75) kt and 1 (0–15) kt by 2050. Unreported production under S2\_SIF may have increased emissions by 7 (4–13) kt during 2014–2018. Cumulative global emissions for 2025–2100 could reach 980 (600–1500) kt, equivalent to 4.7 (2.9–7.1) Gt CO<sub>2</sub>.

Under S3, cumulative unreported production (390–1100) kt moderately exceeds TEAP's (2021) upper estimate (320–700 kt), primarily due to differences in direct emission rates from foam manufacturing. For S3\_AIF, this production increased the 2019 bank by 950 (700–1000) kt, causing a second peak of 1,800 (1500–2,100) kt following the 1995 maximum (Fig.3a). Unreported production may have increased emissions by 22 (12–38) kt (S3\_SIF) and 26 (20–35) kt (S3\_AIF) during 2014–2018. Emissions from unreported production under S3\_AIF are projected to rise from 2020, peaking at 43 (22–67) kt by 2027 (Fig.3b). Cumulative global CO<sub>2</sub>-equivalent (CO<sub>2</sub>-eq) emissions for 2025–2100 under S3\_AIF could reach 7.5 (4.9–10.5) Gt, corresponding to 18% (12–26%) of global greenhouse gas emissions in 2023 (Friedlingstein et al., 2023). Based on surveys conducted in China (China MEE,

2019) and investigations into illegal production capacity by the TEAP (2019; 2021), S3 is deemed less plausible, suggesting more optimistic outcomes for climate and the ozone recovery."

#### Comment #16

L360: How are the mole fractions calculated? There is no mention of any atmospheric modelling in the manuscript.

Reply: Thank you so much for your insightful comments.

We have added following methodological details in the main text, lines 194–202:

## "2.3 Atmospheric concentration modeling

Global CFC-11 emissions and atmospheric mole fractions (expressed as global mean mixing ratios) were derived using Equation (11) following Velders et al. (2015):

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

Where Mix denotes global mean atmospheric mixing ratios in parts per trillion (ppt), with data through 2020 sourced from WMO (2022);  $\tau$  denotes CFC-11 atmospheric lifetime (52 years; WMO, 2022); F (ppt/tonne) is a constant conversion factor for transforming concentration units to emission units, obtained using Equation (12).

$$F = 5.679 \cdot 10^{-3} * 1.07 / M_{CEC} \tag{12}$$

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."

## Comment #19

L434: The final two paragraphs of the conclusion do not add much to the manuscript. They are a generic restatement of the introduction, with very little development.

Reply: Thank you so much for your critical comments. We have restructured the conclusions.

Please refer to the revised text of the main text, lines 458–471:

"Specifically, through the integration of probabilistic parameter distributions, our model yields an upper-bound emission estimate of 56±2 kt/yr for the 2014–2018 period under the baseline scenario S1. At the regional scale, emissions from China increased moderately, rising from 8 (4–13) kt/yr (2008–2012) to 11 (5–13) kt/yr (2014–2018), primarily driven by EoL product management practices. For other

regions, including the U.S., Japan and other non-A5 parties, our model results further support consistency with the regional top-down estimates.

Our findings highlight the important role of product lifespan and EoL handling EFs in estimating long-term trajectories of ODSs and hydrofluorocarbons (HFCs). Bottom-up modeling approaches involve multiple parameters and key processes, which collectively contribute to the overall uncertainty in emission estimates. Rigorous and systematic analyses, particularly those examining temporal and spatial variations in product lifespans and release patterns during EoL handling, are therefore essential to mitigating the inherent uncertainties linked to bottom-up modeling. While this study primarily focuses on CFC-11 emissions, the methodology developed here, which explicitly accounts for uncertainties from underexplored sources, offers broad applicability for estimating emissions of other ODSs and HFCs."

## Comment #20

Technical corrections:

L25: 'accumulate to' is repeated.

Reply: We appreciate this observation and apologize for the redundancy. The repetitive phrase has been removed, and we have conducted a thorough review of the manuscript to to ensure consistency and eliminate any unnecessary duplication.

Please refer to the revised text of the main text, lines 25–26:

"...long-term emission of banked CFC-11 will accumulate to 890 (550–1350) kilotons (kt)..."

#### Comment #21

L25 and throughout: the abbreviation for 'kilotonnes' is kt, not Kt.

Reply: Thank you for pointing this out. We have corrected the abbreviation "kt" throughout the manuscript.

## Comment #22

L28: 'EoL' is not yet defined

Reply: We have added (EoL) in lines 27–28:

"Scenario analysis highlights the potential to reduce up to 50% of emissions through optimized end-of-life (EoL) management strategies."

## Comment #23

L44: over what period have emissions increased in other regions?

Reply: The area including northern France, Belgium, the Netherlands, and Luxembourg (Benelux) showed consistently elevated emissions of CFC-11 (Redington et al., 2023). In Australia, CFC-11 emissions rose from  $0.48 \pm 0.04$  kt/yr (1996–2003) to  $0.64 \pm 0.07$  kt/yr (2004–2008, Dunse et al., 2019).

We have added more detail for the period in line 44–46:

"Furthermore, several developed regions have exhibited elevated CFC-11 emissions after the complete phase-out of production and consumption, with no evidence of unreported production..."

#### Comment #24

L59: 'this analysis', not 'these analysis'.

Reply: We have revised it accordingly in the revised main text, line 60:

"Nonetheless, this analysis..."

## Comment #25

L181: Should read 'sensitivity analysis'

Reply: We have revised it accordingly. Please refer to line 223 in the revised main text:

"2.5 Uncertainty and sensitivity analysis"

#### Comment #26

L375: In addition to the issues with this figure mentioned above, there is no label for subfigure (b) and the y-axis label on subfigures (c) and (f) should be emissions. The GWP of CFC-11 does not vary with time, and does not have units of GtCO2eq.

Reply: We have refined this figure 3 in the revised version, please refer to the response to Comment #15 and Comment #17.

## Reference

- TEAP (Technology and Economic Assessment Panel), Report of the Technology and Economic Assessment Panel, September 2019, Volume 1: Decision XXX/3 TEAP Task Force Report on Unexpected Emissions of CFC-11, Final Report (2019).
- TEAP (Technology and Economic Assessment Panel), volume 3: decision XXXI/3 TEAP Task Force Report on Unexpected Emissions of Trichlorofluoromethane (CFC-11, 2021).
- McCulloch, A., P. Ashford, and P. M. Midgley. Historic emissions of fluorotrichloromethane (CFC-11) based on a market survey. Atmos. Environ. 35 (26): 4387-4397 (2001).
- Duan, H., Miller, T.R., Liu, G., Zeng, X., Yu, K., Huang, Q., Zuo, J., Qin, Y., and Li, J.: Chilling prospect: climate change effects of mismanaged refrigerants in China, Environ. Sci. Technol., 52, 6350–6356, https://doi.org/10.1021/acs.est.7b05987, 2018.
- United States Environmental Protection Agency (U.S.EPA). Inventory of Greenhouse Gas Emissions and Sinks: 1990-2022. https://www.epa.gov/ghgemissions/inventory-us-1376 greenhouse-gas-emissions-and-sinks-1990-2022 (accessed on August 11, 2024).
- Japan Ministry of Economy, Trade and Industry (Japan METI). Documents pertaining to methods for estimating discharge, https://www.meti.go.jp/policy/chemical\_management/law/prtr/6.html (in Japanese, accessed on August 30, 2024).
- SKM ENVIROS: Further Assessment of Policy Options for the Management and Destruction of Banks of ODS and F-Gases in the EU: Prepared for the European Commission, Final report, https://climate.ec.europa.eu/document/download/89673572-7c70-4ed3-a9aa-de31f3e58fb8\_en?filename=ods\_f-gas\_destruction\_report\_2012\_en.pdf (last access: 10 March 2025), 2012.
- Gómez-Sanabria, A. et al. Potential for future reductions of global GHG and air pollutants from circular waste management systems. Nat. Commun. 13(1), 1-12 (2022).