# 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** #1

#### Comment #1

General comments: This manuscript presents a bottom-up dynamic material flow analysis (D-MFA) to quantify the historical and projected emissions of banked CFC-11, aiming to reconcile these with observed atmospheric increases, highlighting the role of bank emissions and end-of-life (EoL) handling. This is a timely and important contribution given the unexpected rise in CFC-11 emissions after 2014 despite the Montreal Protocol controls. The study is well-structured and employs a robust methodological framework with extensive parameterization and uncertainty analysis. However, several aspects require clarification, additional quantitative support, or broader discussion before it can be recommended for publication.

Reply: We sincerely appreciate your thorough review of our manuscript and your insightful scientific and technical comments. These valuable insights have been instrumental in significantly improving the quality of our work.

Our team has carefully and systematically addressed each of your recommendations, with each comment meaningfully integrated into the revised manuscript. We are confident that, guided by your expert input, the refined version more effectively conveys the significance of our findings on CFC-11 bank emissions and their broader implications.

We deeply appreciate the time and expertise you dedicated to reviewing our manuscript and hope that the revised version aligns with the journal's high standards.

## Comment #2

Specific comments:

L112-113: Equation (6) models manufacturing, use, and end-of-life (EoL) stages emissions. However, whether the model explicitly accounts for CFC-11 servicing or maintenance activities is unclear. Could the authors clarify whether this servicing phase was considered and how it was incorporated? If not, please discuss whether the omission might underestimate

the lifetime size of the in-use stock and how significant this could be relative to the total bank. A brief quantitative or qualitative assessment would strengthen the manuscript.

Reply: Thank you for highlighting this important issue. We have carefully considered the use of CFC-11 as a refrigerant in servicing and maintenance activities. Our analysis assumes that CFC-11 released during the use stage can be replenished through refilling. Based on Equation (6) and the parameters detailed in Supplementary Information (SI) Table S7, we estimated that approximately 190 kt of CFC-11 was used as the initial refrigerant charge in non-hermetic refrigeration systems, with an additional 600 kt used as refill quantities during servicing and maintenance over the study period. China's National Plan for the Phase-out of Ozone-Depleting Substances (1993) reports a ratio of approximately 1:3 between initial fills for new products and refills (China MEE, 1993). These consistencies support the validity of our calculations.

CFC-11 usage in non-hermetic refrigeration systems accounts for a relatively small proportion (approximately 8%) of total CFC-11 consumption (AFEAS, 2003). Study conducted by the Technology and Economic Assessment Panel (TEAP) indicates that emissions from refrigeration systems do not make a significant contribute to overall CFC-11 emissions (TEAP, 2019). For these reasons, we did not assess its uncertainties to the same extent as those of foam sectors.

Following your suggestion to enhance the quantitative rigor of our findings, we have included the following text in SI, lines 503–507:

"It is assumed that the release of CFC-11 during the use stage can be refilled. Using this methodology, we estimated that approximately 190 kt of CFC-11 was used as the initial refrigerant charge in non-hermetic refrigeration systems, with an additional 600 kt applied as refill quantities during the usage stage for servicing and maintenance. These estimates align with previous records (AFEAS, 2003; China MEE, 1993)."

#### Comment #3

L145: The uniform 3% release rate may overlook differences between A5 and non-A5 parties, given historically higher fugitive emissions in developing countries. I would recommend either: (1) implementing region-specific r-values that reflect the technological disparities between A5 and non-A5 countries, or (2) providing robust justification for the uniform rate along with a comprehensive uncertainty analysis regarding this parameter.

Reply: Thank you for your attention to this detail. The 3% release rate applied to the production stage is based on two key considerations. First, CFC-11 production and

consumption were predominantly concentrated in non-A5 countries. According to the TEAP, these regions accounted for 94% of global CFC-11 production (TEAP, 2021). Secondly, TEAP's multi-scenario analyses demonstrate that the selection of this release rate has a negligible impact on total emissions (TEAP, 2019; 2021). Their model evaluated a wide range of production emission rates, from 0.5% to 5%, and even higher values, yet found that variations within this range do not significantly alter the overall emissions profile. For these reasons, we opted not to complicate the release rate setting and instead prioritized refining understudied aspects of previous bottom-up models, such as lifespan parameters and other critical variables.

As suggested, to further justify the use of a uniform rate, we have incorporated the following text in lines 153–154 of the revised main text:

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

## **Comment #4 and Comment #6**

L188: Why does the study assume only two discrete EoL emission scenarios of 20% or 100%? What is the rationale or literature basis for these specific choices? It is recommended to consider a more nuanced range of fractions (e.g., 60%, 80%) or perform a more continuous sensitivity analysis to capture the spectrum of plausible emission pathways better.

L247: The authors mention that inadequate waste management systems may accelerate CFC-11 emissions (which is also mentioned in L328-329), yet the model does not appear to incorporate regionally differentiated EoL emission rates. Were regional differences (e.g., higher recovery/incineration rates in developed countries vs. more landfilling in developing countries) considered? If not, might the use of a globally uniform EoL emission fraction underestimate actual emissions in areas lacking effective collection and destruction systems, thereby affecting the regional attribution of emissions? If region-specific assumptions are not yet implemented, I would suggest discussing this as a limitation, along with the potential implications for the results and how future work could incorporate differentiated EoL management levels to improve realism and policy relevance.

Reply: Thank you for your constructive feedback. As these comments (#4 and #6) pertain to end-of-life (EoL) emission rates, we have consolidated our responses below to provide a comprehensive and coherent explanation.

The selection of 20% and 100% as the lower and upper bounds for EoL emission parameters in our uncertainty analysis is based on existing literature. The upper bound of 100%

represents the maximum plausible emissions from retired foam products within unmanaged waste streams—a scenario that has been widely adopted in previous studies (McCulloch et al., 2001; Duan et al., 2018). Conversely, the lower bound of 20% reflects conservative estimates for controlled waste management systems, as assumed in the TEAP's global assessment of CFC-11 emissions from foam products (TEAP, 2021). These bounds were deliberately chosen to encompass a broad range of potential outcomes while ensuring consistency with established scientific references.

While discrete intermediate values (e.g., 60%, 80%) were not explicitly modeled as standalone scenarios, our regional and global calculations implicitly incorporate intermediate emission levels by integrating diverse regional datasets (detailed in SI Tables S8–S13). Specifically, we applied region-specific emission factors. For example, in the United States, sector-specific factors range from 35% (appliance insulation foams) to 100% (spray insulation foams; Table S8), derived from the U.S. Environmental Protection Agency (EPA) 2024 Greenhouse Gas Inventory Annual Report (U.S.EPA, 2024). Similarly, in Japan, emission factors 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, 2024). Comparable regional variations in EoL emission rates were applied to other regions, based on a synthesis of literature review, field surveys, and assumptions (Duan et al., 2018; Gómez-Sanabria et al., 2022). Global estimates were derived from weighted averages of regional values, thereby naturally incorporating intermediate emission levels (Table S13). The variation of this factor is quite significant, making it challenging to define a specific value distribution for sampling purposes.

#### Comment #5

L227-L231: When comparing the consistency of this study's results with other published estimates, it would strengthen the discussion to quantify these comparisons (e.g., percentage differences or correlation metrics), rather than only describing them qualitatively. This would more clearly illustrate the degree of agreement or discrepancy.

Reply: Thank you for this valuable suggestion.

In the revised manuscript, we have incorporated the following quantitative comparison in lines 267–273 of the main text:

"Using top-down approaches, Park et al. (2021) estimated a  $7 \pm 4$  kt/yr increase in emissions from eastern China during 2014–2017 compared to 2008–2012. Our national-scale bottom-up modelling aligns the upward trend reported by Park et al.

(2021), albeit with a slightly smaller magnitude. Yi et al. (2021) reported a national trend that climbed from  $8.3 \pm 1.6$  kt/yr in 2009 to a peak of  $13.9 \pm 2.4$  kt/yr in 2017, followed by a decline to  $10.9 \pm 1.7$  kt/yr in 2019. Our independent estimates of 7 (4–14), 11 (5–14), and 10 (4–13) kt/yr for the corresponding years are broadly consistent with these findings when considering overlapping uncertainties."

## Comment #7

L320: As can be seen from Fig. S14a, the results for 2010 differ significantly from those of METI. The differences between the two results could be described quantitatively, with additional explanations. Furthermore, the results before 2010 vary considerably from those of METI. What is the reason for this?

Reply: Many thanks for highlighting this point. The discrepancies observed between our results and those reported by Japan METI prior to 2010 are primarily attributable to significant methodological changes in the calculation of foam sector emissions. From 2000 to 2012, METI estimated fluorocarbon emissions from closed-cell foam in construction based on a 30-year average lifespan and a 3.3% annual release rate. Before 2010, emissions were calculated by applying the 3.3% release rate to the remaining CFC-11 bank in installed foams, which was adjusted yearly by subtracting a 3.3% loss. Starting in 2010, METI instead applied the 3.3% rate to the total cumulative initial charge, not the adjusted bank. This change increased the calculation base and led to a notable rise in reported emissions.

Starting in 2013, METI updated its methodology to align with the 2006 Intergovernmental Panel on Climate Change (IPCC) Guidelines. These guidelines incorporate product-specific parameters, including distinct annual emission factors for various foam applications (e.g., spray foam, panels, and laminated boards). This methodological update explains the smoother emission trajectories observed from 2013 onwards compared to earlier periods.

As suggested, to further clarify the differences between our results and METI's, we have added the following note in the revised SI (lines 597–603):

"Japan METI revised its emission calculation methodology over time. Prior to 2010, CFC-11 emissions from closed-cell foam in the construction industry were calculated by applying a 3.3% annual release factor to the bank of CFC-11 in installed foams, which was adjusted yearly by subtracting a 3.3% loss. Beginning in 2010, the calculation base shifted to the cumulative initial charge, resulting in a marked increase in reported emissions. Starting in 2013, METI adopted the 2006 IPCC Guidelines,

which use product-specific emission factors, leading to smoother and more consistent emission trajectories after 2013."

#### Comment #8

L350-351: In the results for Scenario S1 (with no unreported production), the cumulative emissions still reach approximately 4.2 Gt CO2e over 2025–2100. This suggests that legacy banks alone could substantially impact atmospheric CFC-11 levels even without ongoing illegal production. Could the authors comment on what this implies regarding the timing of ozone layer recovery? Although this study focuses on climate metrics (CO2e), it would be valuable to briefly discuss (at least qualitatively) how such sustained emissions from historical banks might delay the return of stratospheric ozone. This could help place the findings in the broader context of both climate and ozone protection goals.

Reply: Many thanks for pointing this out. Using the method described in Lickley et al. (2020), we estimated the potential delay in stratospheric ozone recovery caused by sustained emissions from historical banks. Our analysis projects that under Scenario 1 (S1), polar equivalent effective stratospheric chlorine (EESC) would return to its 1980 level around the year 2086.

As suggested, we have added the following information in lines 396–400 of the main text:

"Using the method outlined in Lickley et al. (2020), polar equivalent effective stratospheric chlorine (EESC) under S1 is projected to return to pre-1980 levels around 2086. This projection is slightly earlier than WMO's estimate of 2087 (WMO, 2022). This discrepancy primarily arises from higher CFC-11 concentrations in WMO assessment, attributed to their larger bank and emission estimates derived using the Lickley approach (Lickley et al., 2022; WMO, 2022)."

## Comment #9

L451: The authors state that their findings "may also be applicable to other ODS and HFCs." However, this generalization is not entirely accurate given the scope of the present study. This work emphasizes explicitly banked emissions from closed-cell foams, whereas most HFCs are used in direct refrigeration and air conditioning systems, which are very different. I would suggest that the authors explicitly qualify this statement in the conclusions to clarify the boundaries of applicability.

Reply: Thank you so much for your insightful comments. Given the discrepancies between atmospheric top-down modeling and bottom-up estimates derived from production and consumption data for some ozone-depleting substances (ODSs) and hydrofluorocarbons (HFCs), we propose that our bottom-up approach—which systematically incorporates

uncertainties from underexplored factors—can be applied to the estimation of emissions of other ODSs and HFCs.

We have revised the relevant statement in the conclusion, lines 466–468 of the main text:

"While this study primarily focuses on CFC-11 emissions, the methodology developed here, which explicitly accounts for uncertainties from underexplored sources, may be broadly applicable to emission estimates of other ODS and hydrofluorocarbon."

## Comment #10

Technical corrections:

L111:In some formulas, there seems to be an extra space between symbols, such as Eq.6. Please check the whole text.

Reply: Thank you very much for your meticulous observation and valuable suggestion. We fully agree with your comment and sincerely appreciate your attention to technical details. We have thoroughly reviewed the entire text to correct any extraneous spaces between symbols and ensure consistency and accuracy in the presentation of all equations.

#### Comment #11

L172: The manuscript uses hyphens (-) extensively to indicate numeric ranges (e.g., "3-8 Kt/yr"), which should be replaced with standard en dashes (-) to conform to scientific publishing conventions. Please check the whole text.

Reply: We have replaced the hyphens (-) with standard en dashes (-). A thorough review of the entire manuscript has been conducted to implement the necessary revisions, ensuring consistency in this formatting aspect.

#### Comment #12

L173: S3 lacks the statement "Scenario 3."

Reply: Scenario 3 has been added to the revised main text, line 214.

## Comment #13

L357: The panel (b) label is missing in Fig. 3b.

Reply: Fig. 3 has been revised, and the label for panel (b) has been added.

#### Comment #14

L480: O'doherty shoud be O'Doherty. Please check the references for additional details.

Reply: The name "O'doherty" has been consistently corrected to "O'Doherty" throughout the manuscript.

#### Reference

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- Alternative Fluorocarbons Environmental Acceptability Study (AFEAS). Production and atmospheric release data through 2003.
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- 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.
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- Lickley, M., Solomon, S. Fletcher, G.J.M. Velders, J. Daniel, M. Rigby, S.A. Montzka, L.J.M. Kuijpers, and K. Stone, Quantifying contributions of chlorofluorocarbon banks to emissions and impacts on the ozone layer and climate, Nat. Comm., 11(1), 1380, doi:10.1038/s41467-020-15162-7, 2020.

## 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 103–106 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 484–496:

"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.

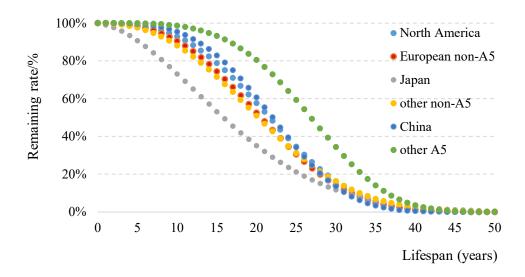


Figure S13 Curves depicting the correlation between remaining rate and lifespan of non-hermetic refrigeration equipment across different geographical regions"

## 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 153–154 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 166–173:

"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 203–214:

"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 178–191:

"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 231–245:

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:

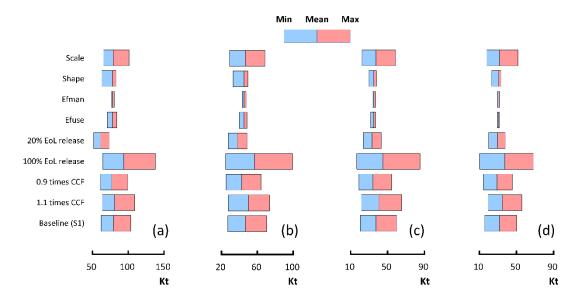


Figure 4 Uncertainty and sensitivity analysis of CFC-11 emissions through bottom-up modeling. (a)–(d) Emission uncertainties induced by various factors for the years 2000 (a), 2010 (b), 2015 (c) and 2020 (d), respectively. closed-cell foam (CCF)

## 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 85–90:

"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.

For clarity, we have revised the relevant paragraph as follows (lines 282–293):

"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 ± 10 kt/yr (Montzka et al., 2018; 2021)."

## Please refer to the revised figure 1 below:

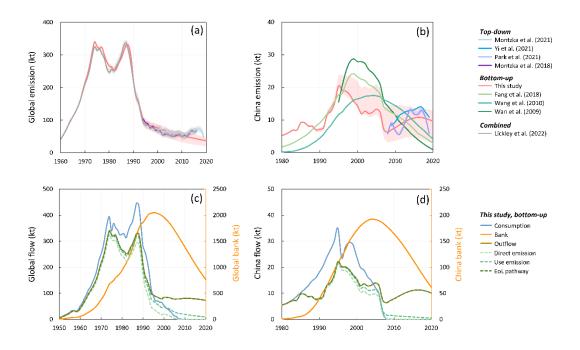


Figure. 1 CFC-11 emissions comparison through bottom-up, top-down, and combined methods.

(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.

## 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 325–337:

"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 264–265:

"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±2 kt/yr, while CFC-11 contained in obsolete products reaches up to 65±1 kt/yr. An increased EoL emission factor shifts bottom-up emissions closer to the top-down estimate of 69 ± 10 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 368–369:

"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:

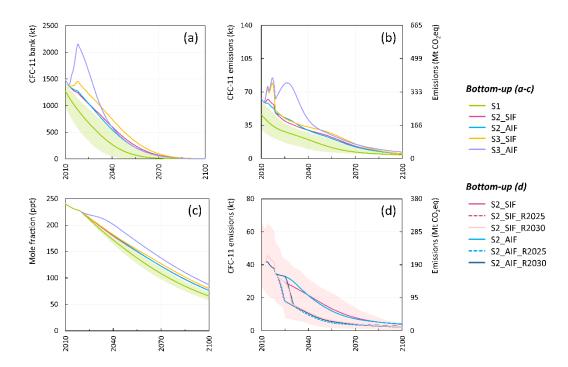


Figure 3 CFC-11 banks, emissions and impacts considering unreported production and mitigation potential. (a) CFC-11 banks based on scenario analysis. (b) CFC-11 emissions and corresponding CO<sub>2</sub>-equivalent (CO<sub>2</sub>-eq) emissions under various scenarios. (c) CFC-11 global mole fractions 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 372–395:

"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 193–201:

## "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).

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

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 455–468:

"Specifically, through the integration of probabilistic parameter distributions, our model yields an upper-bound emission estimate of  $56\pm2$  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 222 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

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- 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).

## EC#1 (on behalf of the 3rd reviewer)

#### Comment #1

This is an analysis of CFC-11 emissions on global and regional scales through recent years when measured atmospheric changes suggested global emissions increased and subsequently decreased. These changes were described in previous papers as having been associated primarily with increased production (and emission) in part in and from China. With a dynamic material flow model to estimate emissions to the atmosphere from the relevant processes, the authors study the sensitivity of calculated emissions from a range of different parameters in an overall useful analysis. They suggest that some increase in emissions might be expected during recent years based on how materials containing CFC-11 were handled during decommissioning and "end-of-life". This suggestion isn't particularly novel, as it was explored to some extent in those initial 4 papers from 2018-2021, was mentioned in Chapter 7 of the 2005 IPCC/TEAP special report (led by P. Ashford et al., although the expected increases were anticipated to be slow and to occur in future years), and is worth repeating, as is the importance of management of these chemicals during decommissioning for minimizing future emissions and their adverse impacts. However, a number of other aspects of the work prevent me from suggesting that it is publishable in its current form, see below.

The abstract is overly general and vague on the main point emphasized there: "bank related emissions could have led to an increased CFC-11 emissions from 2014 to 2018". Figures in the paper actually show global totals declining through this period, and regional emissions from multiple regions that are also decreasing or essentially unchanged if one considers the uncertainties associated with the analysis. Ignoring the range of uncertainties and focusing on only their mid-range estimates, the results can be used to suggest that bank-related emissions increased from only China during this period, but this raises important questions: 1) why is the potential emission increase in China quite different from the decreases derived for the other regions? 2) What are the specific region-based factors/assumptions that cause this uniqueness for China in the model, and what is the supporting information that backs up these choices? Is the uniqueness of China related to the timing and fraction of CFC-11 in different sub-sectors or perhaps of assumptions on lifespan of foam product applications (Table S3)? Many input parameters are provided in tables, but no clarity on the specific ones that make China unique with respect to the time evolution of CFC-11 bank emissions is discussed. 2) Should an increase in Chinese bank-related emissions actually have occurred, the global decreasing trend would imply that the increases in China were offset by unexpectedly rapid decreases in other areas. Is this reasonable?

Reply: We extend our heartfelt thanks for your incisive and constructive appraisal. Your expertise has provided invaluable guidance, and we have meticulously incorporated every suggestion into the revised manuscript to enhance its clarity and rigor.

We fully acknowledge that the potential for increased CFC-11 emissions during decommissioning was previously explored in the initial four studies (2018–2021) and noted in the 2005 IPCC/TEAP Special Report (Chapter 7). However, our work advances the understanding through a systematic, uncertainty-driven analysis of emission trajectories. Specifically, our study quantifies the impact of parameter variability on emissions, contributing to a more robust assessment of potential outcomes. This analytical focus represents a nuanced departure from previous research, offering a more comprehensive understanding of the variability in emissions.

In practice, our results can be calibrated to align with 2014–2018 top-down emission trends by adjusting Weibull scale/shape parameters (product lifespans) or end-of-life (EoL) emission fractions. However, given the spatiotemporal fluctuations in these parameters (SI Tables S3-S4), we prioritized robust uncertainty quantification over strict calibration of annual trends. To model CFC-11 emissions, we employed a systematic Monte Carlo simulation framework. This approach propagates parameter uncertainties through the emission model, generating output distributions. We treated the Weibull scale (u) and shape  $(\beta)$  parameters, manufacturing-stage emission factor (EF,  $Ef_{man}(Y)$ ) and use-stage EF ( $Ef_{use}(Y)$ ) as independent normal variables truncated at zero to exclude non-physical negative values. Distributions were defined using regional/global means (Tables S7-S13), with standard deviations set to 25% of respective means to reflect plausible variability. For instance, in the global boardstock/laminate subsector, mean Weibull scale (u) and shape ( $\beta$ ) are 50 and 6, while the EFs for manufacturing  $(Ef_{man}(Y))$  and use  $(Ef_{use}(Y))$  are 6% and 1%, respectively, yielding standard deviations of 12.5, 1.5, 1.5%, and 0.25%. After defining distributions for all parameters, we generated 10,000 random samples per foam subsector (e.g., Figure S14). EoL and post-life EFs were treated as constant values due to poorly constrained ranges in literature. Annual emissions were computed for each parameter combination, and realizations were aggregated to construct the probability distribution. Uncertainty ranges are reported as the 0.5th to 99.5th percentile interval, corresponding to a 99% confidence interval. The dynamically fluctuating emission ranges in Figure 1a represent physically plausible outcomes within these uncertainty bounds.

Our bottom-up modeling reveals that 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. These divergent

trends could also manifest within global emission trajectories, falling within the uncertainty ranges depicted in Figure 1.

China's emission trajectory diverges from those of the U.S. and Europe due to the differences in 'the timing and fraction of CFC-11 in different sub-sectors', 'assumptions on lifespan of foam product applications', and higher EoL release fractions. First, the timing and sectoral distribution of CFC-11-containing PUR foam differ markedly. In the U.S. and Europe, construction applications dominated (SI Figs. S3, S5), whereas in China, appliance insulation accounted for the majority of use, with peak consumption occurring several years later (SI Fig. S6). Second, appliances generally have shorter lifespans compared to construction materials (SI Table S8 vs. S11). Third, China's EoL release fractions exceed those in developed economies, a discrepancy attributed to less stringent waste management practices (SI Table S8 vs. S11). The combination of shorter product lifespans and higher EoL release fractions amplified the magnitude of the emission increase. These factors collectively explain China's unique peak in bank emissions.

This study investigates uncertainties from previously underexplored sources to enhance the understanding of CFC-11 emission trajectories. The broader uncertainty ranges established here capture plausible variability in the estimates. To improve clarity and provide a more comprehensive interpretation, we have expanded the methodological description and elaborated on the results in the revised manuscript. We address the specific concerns point by point in the responses below.

#### Comment #2

The main message of the abstract is written so that it is easy for the reader to be misled into thinking that perhaps CFC-11 production didn't increase. This is inconsistent with the main text of the paper, discussions of scenarios, and the mismatch on a global scale between expected and atmosphere-derived emission in Figure 1a that would seem to require post-2010 production as an explanation.

Reply: Thank you for your valuable comments on the description of the abstract regarding unreported production.

As suggested, we have revised the abstract, lines 18–32:

"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 may have contributed to the increased CFC-11 emissions from 2014 to 2018, implying potential overestimation of unreported production. Under Scenario 2 (mid-range unreported production levels), long-term emission of banked CFC-11 will accumulate to 980 (600–1500) kilotons (kt), equivalent to 4.7 (2.9–7.1) gigatons (Gt) CO<sub>2</sub>, 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."

#### Comment #3

Some points are also difficult to reconcile: inventory-based model-derived emissions are argued to be consistent with atmosphere-based results in the US, Europe, and China, yet the authors argue that there was substantial unreported production. Are we to conclude that it must have occurred outside these regions despite the contrary evidence provided elsewhere?

Reply: Thank you for your attention to this detail. We appreciate the opportunity to clarify the consistency between inventory-based and atmospheric-based emissions data. Our analysis indicates bank-related emissions partially explain the observed increase in CFC-11 emissions during 2014–2018. While previous studies show declining emissions in China, our analysis suggests a rise from 8 (4–13) kt/yr in 2008–2012 to 11 (5–13) kt/yr in 2014–2018. This upward trend aligns Park et al. (2021), who reported a  $7 \pm 4$  kt/yr increase from eastern China during 2014–2017 versus 2008–2012, albeit with a slightly lower magnitude.

Redington et al. (2023) confirmed declining emissions in western Europe (2008–2021) through top-down modeling, while identifying persistent regional hotspots that the area including northern France, Belgium, the Netherlands, and Luxembourg (Benelux) showed consistently elevated emissions of CFC-11. Our model supports this spatial variability through plausible lifespan parameter adjustments.

Our findings do not suggest that unreported production must have occurred outside these regions. Increased bank-related emissions in China demonstrate that growth during 2014—2018 could originate within these areas.

#### Comment #4

On the regional analyses.

Any revision should be sure to reflect on the uncertainties associated with the assumptions required to perform the analysis in the main text and be more circumspect about the conclusions. For example, China's emissions are said to have increased from 8 (4-13) kt/yr during 2008-12 to 11 (5-13) kt/yr during 2014-18, but given these large uncertainties justifying a conclusion that emissions actually increased is problematic. Discussion of the very small increases reported by Redington et al. are mentioned without consideration of their uncertainties and that they are very small. Did atmosphere-based emissions from these regions actually increase above detectability? The manuscript doesn't indicate that Dunse et al and Manning et al. actually suggested emissions increases through this period, although a reading of lines 43-46 would suggest otherwise. This very slight change in the Redington et al. study is very different from the much larger increase that the inventory model derives for Europe, in apparent contradiction to the wording and assertion on lines 313-314.

These points are central with respect to the stated purposes of this study, which is related to assertions that previous analyses "did not adequately consider the variabilities in lifespans of foam products and their EoL management" and that "surveys reveal significant temporal and spatial variability in the actual lifespans of buildings". Many of the more recent inventory-based studies did provide an analysis of a range of parameter values. Providing more clarity on how the new model adds clarity and understanding to the situation is needed but currently lacking.

Reply: Thank you for your critical feedback. A core methodological distinction of our study is its explicit focus on emission variability arising from plausible parameter fluctuations. Rather than deriving a single best-fit trend calibrated to atmospheric data, we quantify potential bounds of emissions and identify key drivers of variability to enhance the interpretation of regional dynamics. This explains our emphasis on ranges over point estimates.

For instance, regarding China's emissions, the shift from 8 kt/yr (2008–2012) to 11 kt/yr (2014–2018) represents a statistically plausible scenario emerging from region-specific parameter combinations (SI Table S11), with all parameter values remaining within empirical and literature-supported constraints.

Extending this logic to European emissions, we recognize that Redington et al. (2023) reported small, localized emission increases with their own associated uncertainties—findings we do not contradict. Our model, which allows for greater parameter flexibility, captures both increasing and declining trends as well as larger potential fluctuations. This does not introduce contradiction but rather highlights that regional emission trajectories are sensitive to

inputs, with observed small increases representing one end of a broader plausible spectrum. Similarly, while Dunse et al. and Manning et al. do not explicitly emphasize emission increases, our analysis contextualizes their findings by showing how parameter variability can reconcile seemingly divergent trends across studies.

#### Comment #5

#### Details:

Further, the juxtaposition in Figure 1b of top-down and bottom-up estimates for "China" may not be appropriate, given that I believe some (or all) of the top-down estimates represent emissions from only portions of China, whereas I'm guessing the bottom-up estimates are for all of China.

Reply: We sincerely appreciate this important nuance regarding the geographic scope of Figure 1b. To address this, we have added explicit clarification of the spatial coverage of each dataset. This ensures transparency about scale differences and enables accurate interpretation of the figure.

#### Please refer to lines 267–273 of the main text:

"Using top-down approaches, Park et al. (2021) estimated a  $7 \pm 4$  kt/yr increase in emissions from eastern China during 2014–2017 compared to 2008–2012. Our national-scale bottom-up modelling aligns the upward trend reported by Park et al. (2021), albeit with a slightly smaller magnitude. Yi et al. (2021) reported a national trend that climbed from  $8.3 \pm 1.6$  kt/yr in 2009 to a peak of  $13.9 \pm 2.4$  kt/yr in 2017, followed by a decline to  $10.9 \pm 1.7$  kt/yr in 2019. Our independent estimates of 7 (4–14), 11 (5–14), and 10 (4–13) kt/yr for the corresponding years are broadly consistent with these findings when considering overlapping uncertainties."

## Comment #6

Following up on the comment related to the abstract: the authors don't address in the main text why the relative increase suggested in decommission- or EOL-related emissions for China, the US and the EU are so different. Were different parameters used for these different regions based on the unsupported suggestion that they depend on "cultural, economic, and political factors"? Why is different language used to describe the processes for these different regions? More clarity is needed here as to the cause for these differences.

Reply: Thank you for this valuable critique. As addressed in our response to Comments #1, we would like to summarize the key points again.

Please refer to the revised main text, lines 347–352:

"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."

## Comment #7

With respect to the scenarios, some values for emission and unreported production are provided, with uncertainties, but no indication of how those numbers were arrived at and what constraints were used to allow those values to be estimated. Again, further clarity is needed here.

Reply: Thank you for your critical comments.

We have expanded the scenarios development with additional methodological detail. Please refer to the revised main text, lines 203–221:

"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 largescale 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) with unreported production of 25 kt/yr during 2014–2018—half 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) indicates an extreme worst case of unreported CFC-11 production, approximately  $23 \pm 7$  kt/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."

## Please refer to the revised Figure 3:

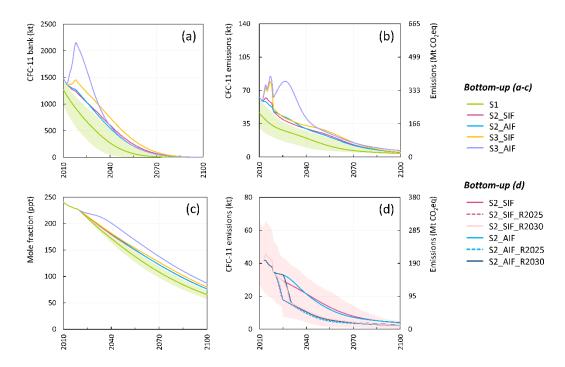


Figure 3 CFC-11 banks, emissions and impacts considering unreported production and mitigation potential. (a) CFC-11 banks based on scenario analysis. (b) CFC-11 emissions and corresponding CO<sub>2</sub>-equivalent (CO<sub>2</sub>-eq) emissions under various scenarios. (c) CFC-11 global mole fractions 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 372–395:

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

## **Comment #8**

Lines 240-241, what is the evidence for this assertion and what do "(b)" and "(a)" refer to?

Reply: Thank you for your insightful comments. The discussion of unreported production has been relocated to the section titled 'Scenarios for unreported CFC-11 production'. The text previously located in lines 240–241 has been removed as it was deemed unnecessary.

#### Comment #9

Lines 253-255, "if all decommissioned..." that's a striking assertion without any clear demonstration and the time dependence of resulting emissions (and how they might compare to atmosphere-based if this were true) is not mentioned.

Reply: We appreciate this constructive suggestion. The statement that "if all decommissioned..." was indeed inappropriate.

We have revised the relevant paragraph as follows (lines 282–293):

"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. S14). 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 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 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)"

## Comment #10

Lines 277-279, why would errors related to emissions estimated in one application (CFC-12 in AC and refrig) apply to CFC-11 whose emissions are from different processes? Gallagher estimates for CFC-11 look reasonably accurate.

Reply: We appreciate your insightful feedback. CFC-11 and CFC-12 are emitted through fundamentally different processes: CFC-11 is primarily from foam-containing products, whereas CFC-12 is largely associated with refrigeration and air-conditioning equipment. Prior EPA methodology assumed 100% retirement of equipment at average lifetime. Gallagher et al. (2013) demonstrated this approach to be overly simplistic, resulting in significant underestimation of CFC emissions in 2008. To address this limitation, they developed a more refined methodology to better capture equipment age distributions at end-of-life. While Gallagher et al. (2013) eventually achieved an accurate estimate for CFC-11, this followed

correction of a systematic bias initially identified in the modeling of CFC-12. Our original reference to CFC-12 was intended solely to illustrate the methodological implications of lifespan assumptions, not to imply mechanistic similarities between the two compounds.

Recognizing that this comparison could be misinterpreted, we have removed the relevant sentence. This deletion does not affect the core arguments or conclusions of the manuscript.

#### Comment #11

Consider projecting the results in panels a-h of Figure 4 out to 2030, as the differences between the bottom-up approaches and assumption should become even larger through this period, and could enable an assessment of their reliability with the atmosphere-based results as they are updated.

Reply: Thank you for this constructive suggestion. Given that Figure 4 focuses specifically on emission sensitivity to various parameters, we have retained its temporal scope up to 2020 to maintain this focused analysis. However, we fully recognize the value of extending projections to facilitate future comparisons with updated atmosphere-based results. To address this, we have revised Figure 3, which presents projections of CFC-11 banks, emissions, and associated impacts—including scenarios that account for unreported production—extending out to 2100. This revised Figure 3 provides a more comprehensive view of the long-term implications and supports the assessment of reliability against atmospheric-based results.

Please refer to the revised Figure 3 in our response to Comment #7.

#### Comment #12

IPCC/TEAP study reference mentioned above: Ashford, P. et al. in Safeguarding the Ozone Layer and the Global Climate System: Issues Related to Hydrofluorocarbons and Perfluorocarbons (eds Metz, B. et al.) Ch. 7 (Cambridge Univ. Press, Cambridge, 2005).). How do you results compare to this (while this could be referred to in the abstract, it perhaps is more a topic of discussion for the main text)?

Reply: Thank you for your suggestion. In the IPCC/TEAP study 'Safeguarding the Ozone Layer and the Global Climate System: Issues Related to Hydrofluorocarbons and Perfluorocarbons' Ch.7 (Ashford, P. et al., 2005), foam products are indeed a significant topic. Chlorofluorocarbons (CFCs) were used as foam blowing agents. CFC - 11, which is the focus of our current research, was among the CFCs used in the production of foam products.

According to the IPCC/TEAP study, the increase in CFC emissions is projected to peak at nearly 100 Mt CO<sub>2</sub>eq during the period between 2030 and 2050, reflecting the impact of

decommissioning CFC-containing products from buildings. Our analysis yields emissions of 115 (70–150) Mt CO<sub>2</sub>eq in 2030, declining to 65 (33–105) Mt CO<sub>2</sub>eq by 2050. These dynamically fluctuating emission ranges represent physically plausible outcomes and are broadly consistent with the projections reported by the IPCC/TEAP study.

TEAP (2019) applied the bottom-up methodology outlined in the IPCC/TEAP study to assess CFC-11 banks and emissions over the period 2002–2015. Subsequently, TEAP (2021) refined and updated the modeling framework for CFC-11 banks and emissions. Our analysis results have been systematically compared with those reported by TEAP (2019, 2021), as detailed in our response to Comment #7.

We have added the comparison with the IPCC/TEAP study as follows (lines 372–376):

"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 further validated by TEAP's (2019) independent assessment using Ashford et al. (2005) methodology, to estimate 2015 banks at 1070–1292 kt."