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 506–510:

> "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 154–155 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 268–274 of the main text:

> "Using top-down approaches, Park et al. (2021) estimated a 7 ± 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 ± 1.6 kt/yr in 2009 to a peak of 13.9 ± 2.4 kt/yr in 2017, followed by a decline to 10.9 ± 1.7 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 596–602):

> "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 returns to its 1980 level around the year 2086.

As suggested, we have added the following information in lines 399–403 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 469–471 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 215.

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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