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A new production-based model for estimating emissions and banks of ODSs: Application to HCFC-141b

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Abstract. The Montreal Protocol on Substances that Deplete the Ozone Layer is a global agreement to protect the stratospheric ozone layer. It requires the phase out of the production of long-lived ozone-depleting substances (ODSs) that are intended for use in emissive applications. The Protocol does not, however, limit the release to the atmosphere of ODSs that currently exist in applications and equipment. Accounting for emissions from these "banked" ODSs (e.g., in insulating foams) is important for monitoring the success of and compliance with the Protocol, for understanding where further mitigation of ODS emissions might be effective, and for estimating future ozone depletion. Here, we present a new bottom-up model for 1,1-dichloro-1-fluoroethane (HCFC-141b), a chemical used primarily in foam insulation and whose production is currently being phased out. Using this refined model, we calculate global emissions that are similar to those derived from atmospheric measurements for the period from 1990 to 2017. After 2017, our modelled emissions are increasingly lower than the observationally based estimates through the end of the comparison in 2021. This discrepancy suggests either a growing additional source of emissions that is inconsistent with reported production or a model deficiency that did not exist or was not apparent before 2017. Our calculations also show that the easily accessible bank will be much smaller in the future than the total bank estimated in other recent work, with important implications for the feasibility of recovering and destroying banks before the release of HCFC-141b to the atmosphere.

1 Introduction

The Montreal Protocol on Substances that Deplete the Ozone Layer went into force on 1 January 1989 with the stated purpose to "protect the ozone layer" (UNEP, 2025). By effectively controlling the supply (e.g., production, import, export, and destruction) of the classes of halogenated chemicals that have been responsible for the most ozone depletion, the Protocol has led to substantially reduced use and emissions of ozone-depleting substances (ODSs) with large benefits to both stratospheric ozone and climate change (Velders et al., 2007). With the Kigali Amendment to the Protocol, it is also expected to substantially reduce future emissions of hydrofluorocarbons (HFCs), which would lead to large further climate benefits (Velders et al., 2009). A potentially extremely large, but speculative, benefit is that without the Protocol, the Earth might have experienced catastrophic ozone loss across most of the globe by the middle of this century (Newman et al., 2009) along with commensurate negative health effects (Slaper et al., 1996).

Parties to the Montreal Protocol are required to report the import, export, and production of controlled substances to the Ozone Secretariat every year under its Article 7. By 2021, the Protocol had led to a reduction in reported global ozone-depletion potential (ODP)-weighted production of ODSs of about 99% when compared with the peak production in the late 1980s (UNEP, 2024). Emissions have also dropped substantially from the peak. Emissions of ODSs are not reported, nor are they regulated under the Protocol; however, global emissions can be estimated from changes in atmospheric concentrations of these controlled substances, such as those measured by global observational networks (Montzka et al., 2015; Prinn et al., 2018). The emissions decline has been somewhat slower than that of production because, for the majority of current uses for ODSs, most emissions occur years to decades after production. This emission lag exists because large quantities of ODSs have been used in applications such as refrigeration and air conditioning (R/AC), insulating foams, and some fire-fighting equipment. ODSs from fire-fighting equipment are emitted only when the contents in the equipment are intentionally deployed or inadvertently released. Emissions from use as a refrigerant in R/AC applications and insulating foam applications occur during active use of the product, upon failure of the product, or when it is decommissioned at end-of-life, and continue once the product containing the controlled chemical is in a landfill. The abundances of chemicals residing in applications actively being used are referred to as “active” banks, and those residing in foams or equipment that have already been decommissioned and landfilled are referred to as “inactive” banks. Without intervention, almost all ODSs amounts that are in active and inactive banks are expected to eventually be emitted into the atmosphere. This eventuality is one of the reasons that the WMO scientific assessments of ozone depletion (e.g., (Daniel et al., 2022)) quantify the future impact of emissions from banks on both ozone depletion and climate forcing. If there were a desire to try to reduce the amount of banked ODSs that would otherwise enter the atmosphere, knowledge of the types of banks is important, as inactive banks are expected to be more difficult and expensive to capture than active banks; furthermore, some active banks (i.e., ODSs used in building insulation) are more expensive to recover than other banks (e.g., ODSs in refrigeration) (Mathis, 2011). An accurate understanding of banks and their magnitudes is also important in assessing whether estimated emissions to the atmosphere are consistent with reported production levels for some ODSs. Such comparisons have important scientific and policy implications for compliance with the Montreal Protocol (Montzka et al., 2018; Chipperfield et al., 2021).

Global bank sizes and future emissions projections have been estimated with at least two different approaches in recent stratospheric ozone assessments and the recent literature. One approach has been to start with a bottom-up estimate of the global bank in 2008 for each long-lived ODS (IPCC/TEAP, 2005) and then to calculate the historical bank for subsequent years by adding annual reported production and subtracting annual emissions estimated from global atmospheric concentration observations (WMO, 2011, 2014, 2018). The average annual fraction of the bank that is released over the past few years (i.e., 5-7) is calculated and then projected into the future, allowing for future estimates of the bank and annual emissions. Most recently (WMO, 2022), a Bayesian analysis was performed (Lickley et al., 2022), that simulates the historical emissions of an ODS as derived from atmospheric concentration observations by allowing actual ODS production to be fit as some factor larger than what was reported and then optimizing the fraction of the ODS bank that is released over all years. Both of these approaches have been used to inform policymakers about the ODS amounts currently in banks and the projections of future emissions if there were no further intervention.

These two approaches have served the Montreal Protocol community well and have provided information that responds to questions related to emissions deviations from projected trends. In particular, they represent straightforward approaches to estimate future emissions and banks, assuming future annual bank release rates remain the same as they have been over some past time period. However, neither method has been applied in a way to include potential changes in bank release rate patterns over time, which are expected as market segmentation changes and the amount of the chemical in each life cycle stage changes. Furthermore, neither methodology has been used to estimate how accessible the bank is at any given time should policymakers wish to take additional action to control ODS emissions. Both of these approaches also rely on derived atmospheric emissions based on chemical concentration observations, which can introduce a bias in the emissions and banks estimates arising from any potential error in the atmospheric lifetime of the chemical as well from systematic errors in the observations.

Product-based approaches are examples of bottom-up methods that represent an alternative way to estimate emissions (e.g., (Gluckman Consulting, 2022; IPCC/TEAP, 2005; Mcculloch et al., 2001; Wang et al., 2015; Zhang et al., 2023). Such approaches begin with knowledge of the various types of applications and products that contain the chemical of interest and the amount of the chemical used in each of these sectors. Knowledge of the expected emission rate during various product life cycle stages and of the distribution of residence times in each life cycle stage allow for estimates to be made of both emissions and bank sizes. Estimating the progression through the various life cycle stages and emissions at each stage can be informed by policy requirements and commercial trends both regionally and temporally. These bottom-up methods can provide information about the accessibility of the banked chemical of interest and have the flexibility to consider changes in the bank release rate over the life cycle of applications, as well as bank release rate changes that occur as the type of equipment/application remaining in service (e.g., air conditioning vs. refrigeration) evolves over time. One of the advantages of these approaches is that they are generally independent of emissions derived from atmospheric observations and can therefore be used as a basis for determining whether actual emissions are consistent with compliance with existing regulations. However, associated with the model flexibility and wide range of model inputs are important data gaps that can result in large uncertainties in results. For example, the emission rate of the primary blowing agents from foams is generally very temperature dependent and depends on the thickness of the foam, on the quality of any facing material covering the foam, and the tightness of any potential cabinet (e.g., in refrigeration) encasing the foam (Andersons et al., 2021; Bomberg et al., 1994; Christian et al., 1991; Hueppe et al., 2020; Makaveckas et al., 2021; Wilkes et al., 2003; Holcroft, 2022). Because of these large sensitivities

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and the assumptions that then must be made to achieve a global or regional average, detailed error analyses are vital to understanding the robustness of such bottom-up approaches.

HCFC-141b (1,1-dichloro-1-fluoroethane, CH_3CCl_2F) is a particularly good compound to use for a model such as described here. It has been primarily used as a foam-blowing agent with minor use as a solvent, and was introduced as a closed-cell foam blowing agent in response to the phase-out of chlorofluorocarbon-11 (CFC-11). Because it was not produced and used in substantial amounts until the 1990s, there is a nearly complete dataset of reported production and atmospheric concentration observations throughout its history of use, unlike datasets for many other ODSs that are significantly banked. Commercial uses of HCFC-141b are also well-understood globally and regionally, as are policies and other issues that can affect markets over time. These factors allow for better validation of this methodology and the associated assumptions than could be performed with many other compounds.

Here, we present a bottom-up model applied to HCFC-141b incorporating knowledge of markets that use this compound and a nearly complete dataset of its production and consumption reported by the Parties of the Montreal Protocol to the Ozone Secretariat. We compare the model's emissions to estimates determined from changes in measured atmospheric chemical concentrations, and we compare bank size results to those of previous studies, while providing a more refined prediction of global banks than has been previously available. The description of the model and underlying data used in it are found in Sect. 2. The results and discussion are found in Sect. 3, and our conclusions about the modeling approach, its applications to HCFC-141b, and opportunities for its application to other compounds are in Sect. 4.

2 Methods

The life cycle stages when HCFC-141b emissions occur are shown in Fig. 1. These include during: (1) production of HCFC-141b, storage, and transport before sale; (2) chemical blending, shipment, and storage of blended systems, and foam blowing and installation; (3) active product use; (4) product decommissioning; and (5) the time after decommissioning when the product is no longer used and is in its final point of disposition (i.e., landfill). Quantifying the transition of a foam blowing agent through the life-cycle stages and estimating the emissions at each stage requires knowledge of numerous parameters that depend on the specific application and can also depend on the geographic region where consumption occurs. These parameters, and their sometimes-sizable uncertainties, are described in the following sections. Parameter values are based on values in the literature, with reference to experimental results when available. With our approach, emissions are calculated at the regional level for each type of product and at each life cycle stage. The regions are defined in Appendix 4 of UNEP (2007), with the only difference imposed here is that Japan is included with Europe so as to not perform any calculation with a region comprised of a single country. These two regions were combined because their use patterns were relatively similar over the time of their peak use. While fundamental model emission parameters and lifecycle lifetime parameters are assumed to be the same in each region (with one exception described below), variations in consumption and the relative sizes of markets in different regions can be large and lead to different total regional emissions and bank characteristics. Regional results are combined to provide annual global emissions and banks estimates. In this work, the only regional parameter variation we consider is a reduced emission associated with the unique decommissioning policy in Europe after 2002, described further in Sect. 2.5. There are some lifecycle lifetime parameters available for China, which differ from the values we use here, and could be applied in future versions of the model (Wang et al., 2015).

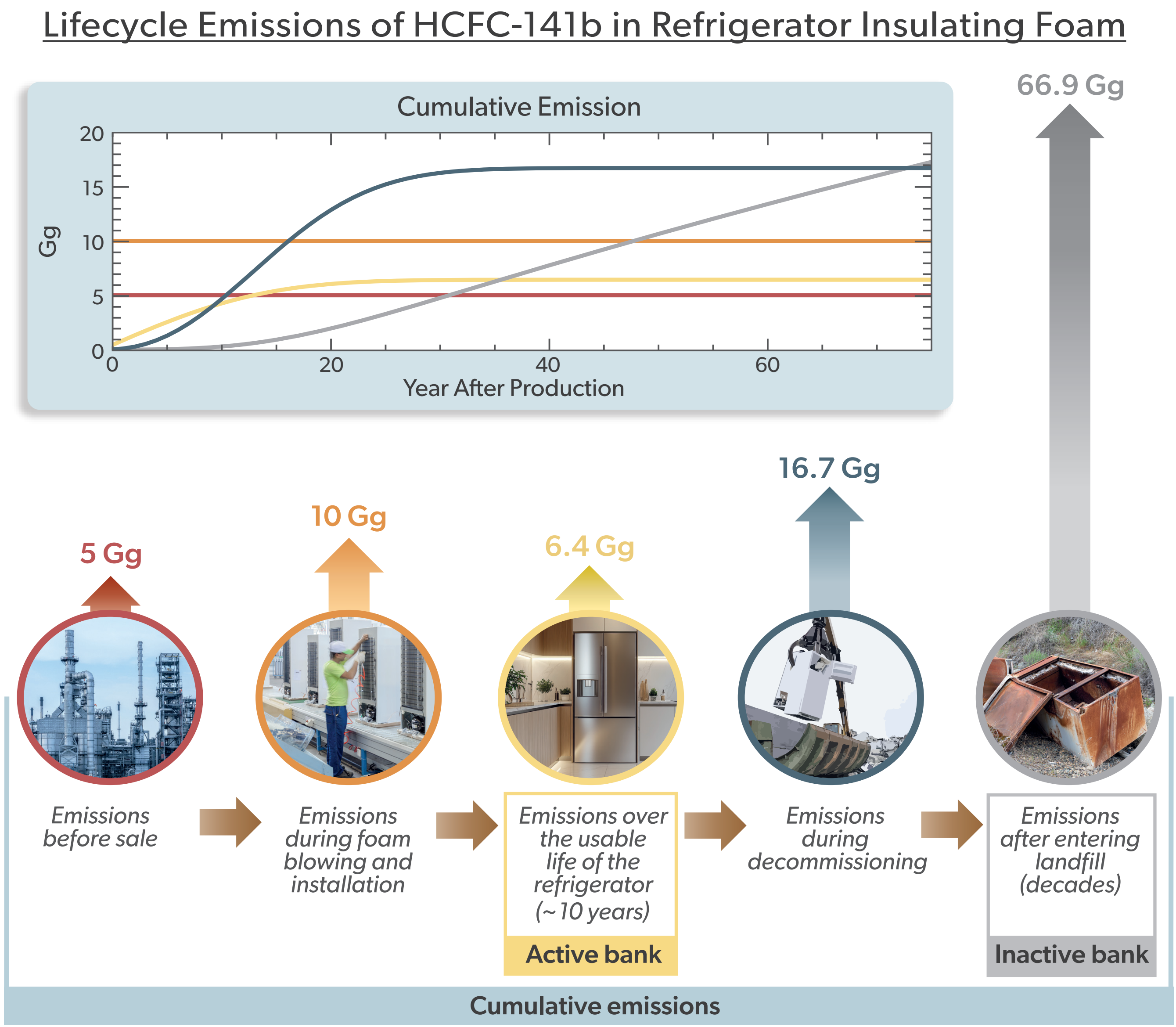

Figure 1. Cumulative emissions over time in the life-cycle stages for refrigeration foams, assuming hypothetical 100 Gg of reported production for refrigerator foam use in year 0. The emissions before sale are assumed to not be included in reported production. The graph

shows the first 75 years after production of the 100 Gg, while the numbers in the graphic represent the amount emitted during each life cycle stage after all the HCFC-141b is emitted. The color-coding of the lines is the same as shown in the graphic of the life cycle descriptions and total emissions.

Parties to the Montreal Protocol annually report chemical production and imported and exported quantities of all controlled ODSs, including HCFC-141b, to the Ozone Secretariat. Calculated consumption, defined as reported production plus imported minus exported chemical is frequently a more appropriate quantity to use for regional calculations as opposed to production, since it better reflects the local supply of chemicals that are ultimately used in manufacturing and thus is generally better representative of local banks and releases from the foam life cycle. Consumption will form the basis of our calculations. It is worth noting that goods assembled in one location can still be exported to another region with no required reporting of import/export of the ODS contained in the finished product, or even in a chemical blend (e.g., foam system).

For HCFC-141b, annual calculated global consumption is more often larger than the reported production since the total global quantity of reported imports is higher than reported exports for most years. If reporting were perfect, consumption and production would be nearly equal in the global sums at least when summing over several years; individual years might have slightly unequal consumption and production if some quantities were to be attributed to the next year due to supply chain delays between the timing of export and import. Cumulatively over 1989-2022, consumption is 2.7% higher than production, although differences can be substantially larger during individual years. Here, regional consumption is never allowed to be negative, and thus, after production ends in non-Article 5 (non-A5) Parties, the small reported exports that are in excess of production, are not considered in our calculations. Regionally based consumption over time is shown in Figure 2. Global consumption is bimodal, with the earlier peak dominated by non-A5 countries (e.g., Australia, Europe/Japan, Canada, United States), and the latter peak dominated by A5 countries. Global consumption from 2023 onward is assumed to follow a linear trend until is drops to 0 in 2028, with it all assumed to be in the Northeast Asia region, consistent with planned phase-out schedules.

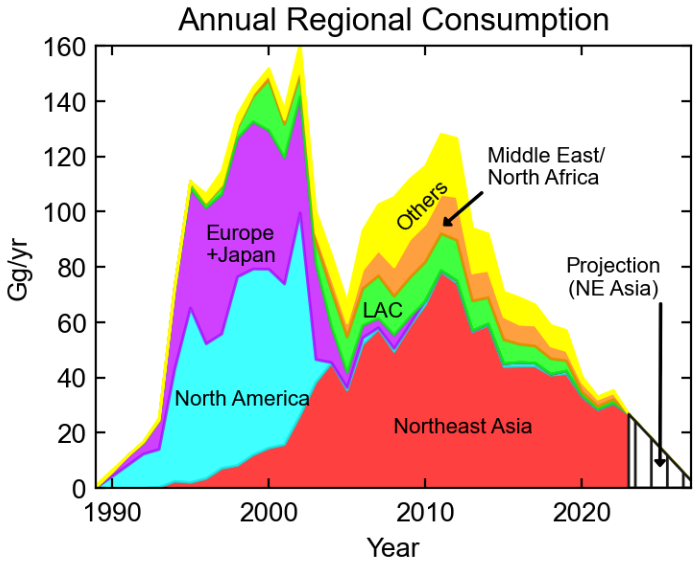

Figure 2. Reported consumption of HCFC-141b by region. "MENA" is the Middle East and North Africa region and "LAC" is the Latin America and the Caribbean region. Regions are stacked from bottom to top in descending order of total consumption over the entire historical period.

2.1 Emissions before sales and when used as a solvent

The reported production and consumption values do not include losses that occur during the initial chemical production of HCFC-141b and before it is sold for use. These losses can occur while filling containers, drums, tanks, etc. in preparation for sale. They have been estimated by the Technical and Economic Assessment Panel (TEAP) to the Montreal Protocol Parties as varying from 0.9-4% percent of the total production for current "heavily regulated sophisticated plants" to 3-5% in "regulated manufacturing plants" over 1960-1980s, as shown in Tables 2.6 and 2.8 of the Medical and Chemical Technical Options Committee (MCTOC) Assessment Report (MCTOC, 2022). The overall emission factor has been estimated as 4% in IPCC (2019), with an extremely large potential variation across individual facilities. Here, we assume that these losses occur before production reporting and represent, on average, 5% of the reported production (4.8% of total production); in our error analysis, this emission factor is assumed to be uniformly distributed between 0 and 10%. The losses are considered to be released to the atmosphere in the same year the production occurred. We do not consider separately any other production that might simply be unreported. Lickley et al. (2022) found emissions were better explained if HCFC-141b production were 12% higher than what was reported.

HCFC-141b has also been used as a solvent. *UNEP* (2003b) estimates that 10% of the produced HCFC-141b was used as a solvent at the time that report was published; more recently, *UNEP* (2019) estimates this has been between 5 and 7.5% from 2011-2018 in A5 countries, while Zhang et al. (2023) has stated that solvent use represented 9.2% of production in 2019 and 8.0% of cumulative production over 2000-2019 in China. We assume that solvent use is 10% of global annual reported production, with a 50% uncertainty on this number (i.e., a uniform range from 5%-15%). Half of the HCFC-141b used as a solvent is assumed to be emitted in the year of being manufactured with the rest emitted in the following year (IPCC, 2006).

2.2 Emissions during foam blowing agent installation

Blowing agent releases at the beginning of and throughout product life depend on the specific application. For example, foam blowing agent (FBA) releases during foam manufacture from spray foam applied in buildings are thought to be greater than releases during a controlled injection process into a mold during the manufacturing of appliances (Aprahamian and Bowman, 2005). To obtain information about the division of HCFC-141b between the various markets (e.g., domestic refrigeration foams, spray foams, etc.), we rely on the three Foams Technical Options Committee (FTOC) Assessment reports that provide this information (UNEP, 2003a, 2007, 2010). These reports have performed a detailed analysis of the markets for each geographic region. The years covered encompass the majority of the transition period from when HCFC use was primarily in developed countries to a time when the majority of HCFC-141b production occurred developing countries. These reports also provide information regarding the fraction of each market that uses HCFCs as opposed to other types of compounds (e.g., HFCs, CFCs, etc.); we used this information about fractional use of HCFCs and further consider only the markets where the HCFC used was HCFC-141b rather than HCFC-142b or HCFC-22. This market information is combined with regional consumption estimates (c.f., Figure 2) to determine the amount of HCFC-141b used in each of the 11 markets in each region. We have applied the same regional market breakdown to consumption data for all years through 2002 as given by UNEP (2003a), with markets from 2003 through 2007 given by UNEP (2007). Values from 2008 onward are given by UNEP (2010), with the added constraint that all use of HCFC-141b in refrigeration foams is linearly phased out over 2010-2015, with the remaining markets scaled up proportionately. This refrigeration phaseout is meant to approximate the policy implemented in many countries mandating a transition away from using HCFC-141b in appliances by 2015 as part of their ODS phasedown.

“Foam blowing and installation” emissions referred to in Fig. 1 include all emissions that occur during the foaming process as well as any excess emission that occurs through the first year of use. These emissions are meant to be globally appropriate values spanning highly controlled large manufacturing facilities to less controlled activities. This incorporates potential losses from the supply chain after sale of the HCFC-141b and prior to the delivery to the manufacturing plants, such as during shipping and handling, when mixing FBAs into polyol blends, during the manufacturing of the foams, and early losses from foams during the first year after manufacturing. Factors such as the volatility of the chemical, its solubility in the polyol, and the extent to which the foam blowing occurs in a controlled environment are important in determining losses during the foaming process. One study found that, given the boiling point of HCFC-141b, a 4% emission at the time of foam blowing would be expected even in the contained environment of a refrigerator-like mold (Aprahamian and Bowman, 2005). To account for all other sources of emission in this category, we assume 10% release rate for foams installed in molds for domestic refrigeration as well as in polyurethane (PU) pipe-in-pipe, panels, and boardstock applications, with higher emissions in other markets. We assume an absolute uncertainty standard deviation of 5% on all installation rates such that the fractional uncertainty proportionately decreases for markets with larger estimated installation emissions.

Table 1. Assumed release rates of HCFC-141b during foam manufacturing and installation and active use, and parameters used to describe the failure rate of products as a function of time. Uncertainties represent the standard deviation of the probability distribution used in the Monte Carlo analysis; installation emissions and emissions during foam use (active bank), are both represented by lognormal distributions while the Weibull scale factors and Weibull lifetime terms are represented by Normal distributions. Color-coding is discussed in Sect. 3.3; green values have uncertainties with the largest influence on global emissions, blue values have uncertainties with an intermediate effect on emissions, with other values contributing least to overall emission uncertainty. The uncertainty associated with market segmentation would be categorized as green if shown in the table. Sources for these values are provided in Appendix Table A1.

Application	Emission During Manufacturing and Installation (Sect. 2.2)	Annual Emissions During Foam Use (ε) (Sect. 2.3)	Failure Rate Parameters for Weibull function: $f(t) = \frac{s}{\tau}\left(\frac{t}{\tau}\right)^{s-1} e^{-(t/\tau)^{s}}$ (Sect. 2.4)	
			Weibull Scale Factor, s (±0.2)	Weibull Lifetime Term (Mean Lifetime), τ (±20%)
Refrigeration				
Domestic refrigeration	**10%**	**0.5%**	**2.34**	**18.1 (16.0)**
Commercial refrigeration	**20%**	**0.5%**	**2.34**	**16.9 (15.0)**
Refrigerated containers	**20%**	**0.5%**	**2.34**	**19.7 (17.5)**
Building Construction				
Continuous panels	**10%**	**0.5%**	**1.97**	**67.4 (60.0)**
Discontinuous panels	**10%**	**0.5%**	**1.97**	**67.4 (60.0)**

Spray foam	**25%**	**1.5%**	**1.97**	**67.4 (60.0)**
PU boardstock	**10%**	**1.0%**	**2.8**	**28.1 (28.0)**
Other Uses				
PU pipe-in-pipe	**10%**	**0.5%**	**3.0**	**33.6 (30.0)[1]**
PU block-pipe	**45%**	**7.5%**	**3.0**	**16.8 (15.0)**
PU block foam slab	**20%**	**1.0%**	**3.0**	**16.8 (15.0)**
PU integral skin	**40%**	**2.0%**	**3.0**	**11.2 (10.0)**

[1] Uncertainty is assumed to be 30% as described in Table A1.

Table 2. Release rates for emissions that are assumed to not depend on market. Using the color-coding of Table 1, if these values were shown by specific market, decommissioning emissions for domestic refrigeration and inactive bank emissions for domestic refrigeration would be green (highest category of uncertainty importance); blue would include: decommissioning emissions for pipe-in-pipe, commercial refrigeration and boardstock; and inactive bank emissions for pipe-in-pipe and boardstock use. Uncertainties represent one standard deviation of the probability distribution used in the Monte Carlo analysis for decommissioning and inactive bank release, which are both represented by lognormal distributions. Emissions before sale and the solvent use are assumed to be described by uniform distributions with the full range represented (*i.e.*, 0-10% and 5-15%, respectively). Sources for these values are provided in Appendix Table A2.

Type of Emission	**Magnitude and Timing**
Before sale (includes loss during production)	5%±5% (uniform distribution function, i.e., 0%-10%) of reported consumption in year produced
During Decommissioning	20%±10% of decommissioned amount in year of decommissioning
From inactive bank (i.e., post-decommissioned bank such as landfill)	0.50%±0.25% of inactive bank, annually
Solvent Use (emitted over 2 years)	10%±5% (uniform distribution function, i.e., 5%-15%) of reported consumption

2.3 Emissions during product use

Emissions during the operational use of foam products depend on the particular application and generally occur gradually (e.g., (Hueppe et al., 2020; Kirpluks et al., 2022; Paul et al., 2021; Wilkes et al., 2001)). Descriptions of how the composition

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of foams changes with aging have been published that estimate changing concentrations of foam blowing compounds from changes in thermal conductivity (Andersons et al., 2021, 2022; Bomberg et al., 1994; Kirpluks et al., 2023) and/or from measuring the gas composition directly (Modesti et al., 2005; Kirpluks et al., 2023). Measurements have shown great variation in diffusivities of the FBA out of foams, with sensitivity to temperature, foam thickness, presence and quality of any facing material on the foam, and the integrity of any casing around the foam. These variations make it unclear how best to extrapolate individual studies to region-wide values. Therefore, our values, with the exception of the block-and-pipe and pipe-in-pipe products, are generally consistent (when uncertainties are considered) with bottom-up values adopted in other work (Table A4-1 in UNEP (2003a), Table 7-7 in IPCC/TEAP (2005), Tables 7.6 and 7.7 in IPCC (2006), and Table A4.3 in TEAP (2019)). Our block-and-pipe emission rate estimate is taken from Table A4.3 in TEAP (2019) (assuming the quoted value is a factor of 10 too large) and is higher than in the other references; we have made this choice due to the expectation that this application would have higher emissions than other types of closed-cell foams. Our pipe-in-pipe emission rate of 0.5% is closer to the 0.25% in the listed references above, excluding TEAP (2019), in which the quoted 25% is thought to be too high. We assume the standard deviation (s.d.) for each emission rates is ±100% of the value used and that the uncertainty follows a lognormal distribution.

The length of time foam products remain in use varies greatly and depends on product type. For example, insulated refrigerated containers exposed to heavy vibration over roads will likely have shorter lifetimes compared to insulating foams installed in buildings. Some foam product lifetimes also vary by region, e.g., as in lifetimes of buildings (Deetman et al., 2020). Here, we calculate emissions during the life cycle stage of active product use by using Weibull survival functions for equipment and buildings to create a probabilistic distribution of the active life stage by foam type (e.g., (Aktas and Bilec, 2012; UNFCCC, 2017; Yazici et al., 2014; UNEP, 2023; Gallagher et al., 2014)). The fraction of equipment that remains in service as a function of time, *t,* after installation is given by

$$F(t) = \exp\left\{-\left(\frac{t}{\tau}\right)^{s}\right\}, \tag{1}$$

which is 1 minus the Weibull function's cumulative distribution function. The Weibull function is described by two parameters. One, *s,* governs the general shape of the distribution of decommissioning timing, with smaller values implying statistically more abrupt decommissioning. The second, *τ*, is related to the length of time the product is used before decommissioning, with the value representing the number of years after being put into service when 63% of the products have been decommissioned. The Weibull parameters are shown in the final two columns of Table 1 for each product type. To account for emission during the active life cycle phase, we assume that leakage emissions from the foam in a particular market is given by

$$\frac{dh}{dt} = -\varepsilon h, \tag{2}$$

so that

$$h(t) = h(0)\exp(-\varepsilon t), \tag{3}$$

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where *h(0)* represents the amount of HCFC-141b in the installed equipment at time 0 and ε is the annual leakage rate. Thus, if we normalize *h(0)* to be 1, the amount of HCFC-141b mass that remains in active equipment at time t is given by

$$M_{active} = h(t)F(t) = \exp\left\{-\varepsilon t - \left(\frac{t}{\tau}\right)^{s}\right\} \tag{4}$$

and the cumulative amount of emission that has occurred from the active bank through time *t* is given by

$$E_{active}(t) = \int_0^t F(t')\frac{dh}{dt}dt' = -\varepsilon h(0)\int_0^t \exp\left(-\varepsilon t' - \left(\frac{t'}{\tau}\right)^{s}\right)dt'. \tag{5}$$

285 Figure 3 shows, as examples, the decommissioning functions for three markets. The curves show the fraction of the installed amount of HCFC-141b remaining in active use as a function of time after installation. Also shown are the cumulative emissions that occurred during use. The difference between the total installed and the sum of cumulative emissions and remaining bank represents the amount of HCFC-141b in applications that have been decommissioned and sent to a landfill. For example, much more of the HCFC-141b in the "Block Foam Slab" market eventually goes to the landfill (>80%) than in the "Block Pipe" market (<50%), since more of the block-pipe HCFC-141b is emitted while the foam is still in use.

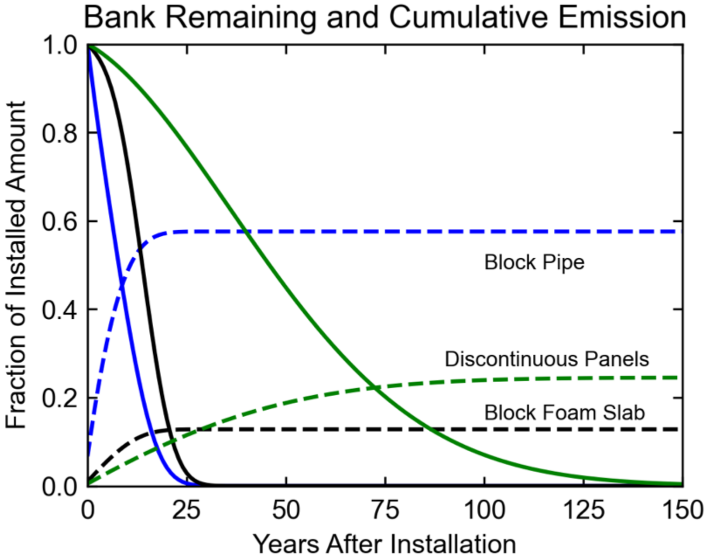

Figure 3. Weibull decommissioning functions assumed for three product types: (1) polyurethane block-pipe, (2) discontinuous panels, and (3) and polyurethane block-foam slab. These three markets were chosen because their rates of emission from foam and service lifetimes provide a wide range of responses. Solid lines represent the amount of HCFC-141b remaining in the equipment as a fraction of what was installed in year 0. Dashed lines represent the cumulative amount emitted while the product is in use. The difference between the total installed and the sum of cumulative emissions and remaining bank represents the amount of HCFC-141b in applications that have been decommissioned and sent to a landfill.

2.4 Emissions during decommissioning

Significant emissions of HCFC-141b and other similar foam blowing chemicals can occur at the end-of-life of products that still contain them due to the dismantling process. In the case of foam products, this primarily results from the partial crushing or shredding of the foam at the time of disposal. Although some foams are recycled or destroyed at this time, most are transported to landfills as waste. In the United States, most foams are crushed or shredded when they enter the waste stream (ICF International, 2011); however, globally, most foams are likely not shredded before being landfilled. Scheutz and Kjeldson (2002) and Kjeldson and Scheutz (2003) measured the immediate and short-term release of FBAs, up to 20% for CFC-11 and 28% for HCFC-141b from shredded foams, finding that the amount of FBA emitted during and soon after shredding is highly dependent on the size of the remaining pieces, with courser (finer) particles results in less (more) emission. They did not measure release during the dismantling process. Because large amounts of landfilled foams are not shredded, we assume a 20% (s.d., 10%, lognormal distribution) release to describe losses during the dismantling, transport and disposal processes and note that this is higher than the amount of 5% used in TEAP (2019) for CFC-11. It is much lower than the 100% used in McCullough et al. (2001) for CFC-11.

Europe mandated that FBAs in refrigeration must be “recovered for destruction … or for recycling …” during decommissioning of appliances beginning in 2002 (E.U. Regulation No. 2037/2000, Article 16 and Directive 2002/96/EC). While evidence of the extent of compliance is unclear, this would reduce this source of emissions. The decommissioning release rate from domestic appliances in the European market has been reduced to zero from 2002 onward and the HCFC-141b in any decommissioned application in Europe from 2002 onward is removed from our calculations, not contributing to the future banks or emissions. While this represents an extreme assumption, it matters little to our global bank and emission calculations.

2.5 Emissions after decommissioning

After foams are brought to the landfill, and after any initial rapid emission due to crushing or shredding of the foams, FBAs generally continue to be emitted slowly over time. We assume annual release rates of 0.50%±0.50% as given in *TEAP* (2022). We neglect any potential for anaerobic degradation (Kjeldsen and Scheutz, 2003; Scheutz et al., 2009).

2.6 Uncertainty analysis

The model described in the previous sections has a total of 79 input parameters, each with uncertainties associated with it, and each with varying degrees of importance to the calculation of emissions and banks of HCFC-141b. All uncertainties are combined to determine their influence on emissions and banks using a Latin-Hypercube Sampling Monte Carlo approach. We perform 1000 simulations to determine the uncertainty ranges in Sect. 3; we find this number is more than sufficient to estimate the influence in a repeatable manner. In each Monte Carlo simulation, the randomized quantities remain fixed for that entire time series and are not allowed any year-to-year variations. The magnitudes of the uncertainties are shown in Tables 1 and 2, and they, themselves, can also be highly uncertain. We will discuss some of the key uncertainties in Sect. 3. Most uncertainties are assumed to follow lognormal distributions around the primary value. Exceptions are that the amount used as a solvent and the amount emitted as "production emissions" are assumed to follow a uniform distribution with the values listed in Table 2 being the full range of the distribution. The uncertainties associated with each variable are assumed to be independent. Market share uncertainties for the 11 different markets are slightly more complicated because all market shares in each simulation must equal 100% and thus are not, by definition, independent. To simulate this, in each Monte Carlo iteration, a 3-step process is carried out: (1) the individual market shares are altered by adding a random number chosen from a standard Normal distribution with a standard deviation equal to 20% of the primary value or 5% in absolute terms, whichever is greater; (2) all negative market shares are raised to 0; and (3) then they are all scaled proportionately so they sum to 100%.

3 Results

3.1 Sectoral breakdown of HCFC-141b use

The market segmentation approach described in Sect. 2.2 yields a market breakdown over time shown in Figure 4. During the first decade of its production, this analysis shows that the majority of HCFC-141b use was in domestic refrigeration foam, spray foam, panels, pipe-in-pipe, and boardstock. The applied linear phaseout of refrigeration uses is apparent from 2010 to 2015 with pipe-in-pipe, spray foam, and discontinuous panel use dominating global markets over the most recent years.

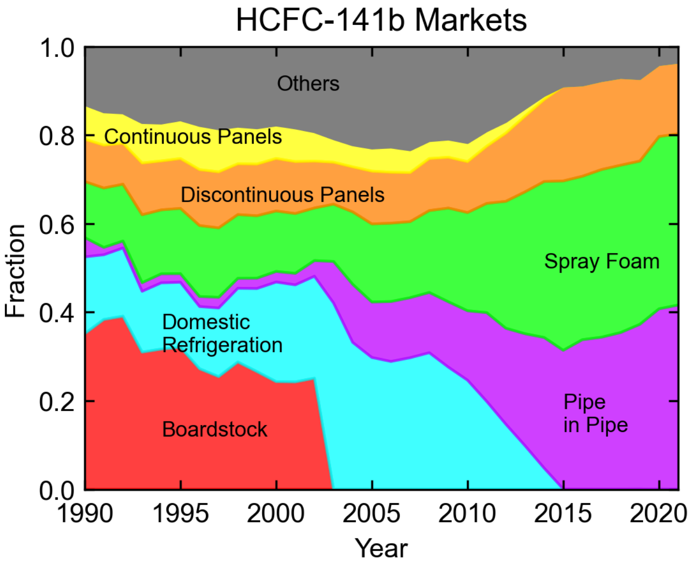

Figure 4. Global breakdown of HCFC-141b markets. Values based on regional market fractions and the amount of HCFC-141b used in each market as described in the text combined with regional consumption shown in Figure 2.

3.2 Annual emissions

The market breakdown is combined with the release parameters of Tables 1 and 2 and annual consumption values (Figure 2) to calculate the emissions and banks of HCFC-141b over time. The calculated total annual emission from the sum of all life cycle stages and all markets is shown in Figure 5, with the 1-sigma range shown. Due to the relatively low release rates, the contribution of emissions from active and inactive banks changes relatively slowly over time, and the higher frequency year-to-year changes are due to rapid emissions such as those associated with production, emissions from solvent use, and emissions associated with installation of various foams.

Figure 5 show a similar temporal shape between our bottom-up emissions and those estimated from observations. The exception to this occurs at the very end of the time period when our calculation suggests a drop in emissions, which seems inconsistent with the atmospheric observations. This supports the finding Western et al. (2022) arrived at using a hybrid bottom-up/top-down model. Barring production over the latest years that is substantially greater than reported, other changes in important parameters such as emission associated with production before sale, emissions associated with installation of foams, market shifts, etc. could lead to better agreement. However, we note that any such changes would be larger than apparently required for the model to be consistent in temporal shape with observations at any other time during the history of HCFC-141b production and use, as is apparent from Figure 5. It is also unlikely that emissions from feedstock uses could explain the recent differences in emissions. Total production for intended feedstock use has been below 20 Gg/yr since the advent of feedstock use (Western et al., 2022) and it is expected that only a few percent of halocarbons used as feedstock will be emitted to the atmosphere (WMO, 2022). The relatively good performance of our model when compared with observationally derived emissions is a particularly informative result since it is a purely bottom-up method. That is, it does not adjust model parameters based on any observations.

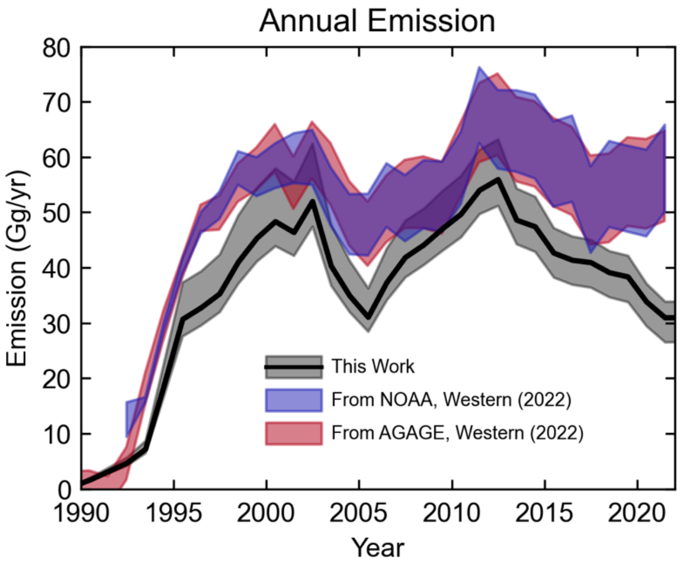

Figure 5. Total calculated global emissions compared with top-down estimates made with a 12-box model using NOAA and AGAGE atmospheric measurement networks, as given in Western et al. (2022). All shaded regions represent the 68% confidence range. Note that the overlap of the NOAA and AGAGE observations appears purple.

3.3 Global life cycle analysis

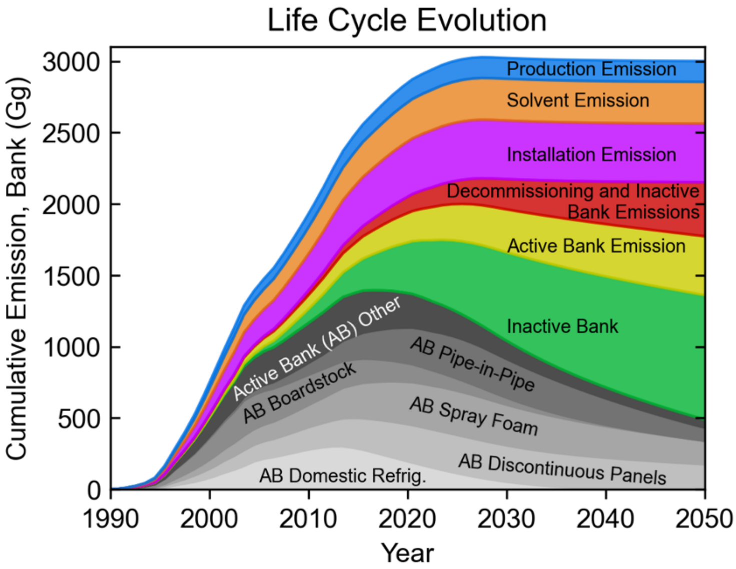

Figure 6. Life cycle analysis of all produced HCFC-141b over time. The top curve represents total cumulative consumption over time, including that which was not reported and was estimated here to be emitted as "Production Emissions", and excluding the small amount of HCFC-141b that is assumed to be captured and destroyed at the time of decommissioning in Europe. The second-to-the-top curve equals the cumulative reported consumption with the same decommissioning exclusionary caveat. The primary three active banks are identified, with the rest grouped together as "Other".

Figure 6 provides one approach to viewing the life-cycle analysis of HCFC-141b over time. It includes the sizes of the largest active banks and of total active and inactive banks, as the cumulative emissions from various emissions sources. It shows that in the year 2020, slightly more than half of all cumulative production to that point is calculated to reside in banks, with less than half having been emitted to the atmosphere. The amount emitted to the atmosphere continues to grow after 2020, coming entirely from the banks after production is assumed to cease from 2028 onward. We do not consider emissions from feedstock production or use in any of these calculations, which is currently believed to be small, as stated above, and is expected to continue after the phaseout of controlled production.

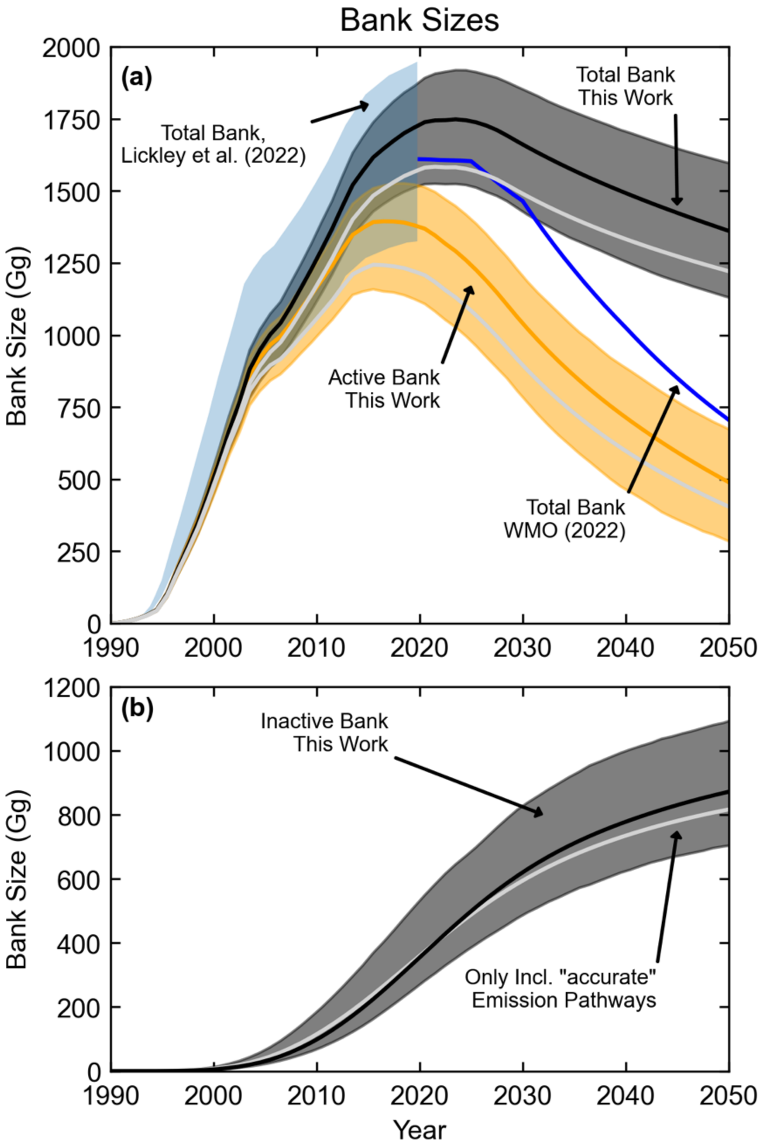

Figure 7. Calculated evolution of HCFC-141b banks. Panel a: The active bank for this work includes the total HCFC-141b found in all applications still in use in all regions and all markets. The total bank (active plus inactive banks) includes banks after decommissioning (i.e., landfills). Also shown are the total banks calculated by Lickley et al. (2022) for past years and by WMO (2022) for the future. The uncertainty range for this work is given as a 90% confidence interval, as is that for Lickley at al. (2022). Solid black and orange lines represent the banks for our baseline parameter values. Light grey lines that track in the lower half of the active, inactive, and total bank ranges represent the average bank sizes if only pathways are considered in which emissions fall within the emissions range estimated from observations (c.f., Figure 5) over most of the time period. Panel b: The inactive bank is separated out from its inclusion in Panel a to show its central value as well as its 90% confidence interval.

Figure 7 provides a more focused comparison of the bank estimates and shows the 2-sigma uncertainty range of our calculated active and total (active plus inactive) banks. The total bank compares well with that of Lickley et al. (2022). This is despite the

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fact that the emissions calculated in this study are slightly lower than those estimated from observations, observations that were used to estimate emissions and then used as constraints in the Lickley et al. (2022) work. If we only consider sets of parameters from our Monte Carlo analysis that lead to good agreement with the observationally-derived emissions, our bank estimates drop to the grey lines in the figure and remain in agreement with Lickley et al. given the size of the error bars. As previously mentioned, Lickley et al. (2022) found that an increase in reported production of 12% led to a better overall fit to emissions estimated from atmospheric concentration observations. In the set of our Monte-Carlo pathways that agree more closely with observationally derived emissions, the average emission associated with production is 8%. While this should not be considered a retrieval, it does show some level of consistency between the two studies in that they find better agreement with emissions estimated from observations when there is additional production relative to what is reported. The comparison of our total bank with the bank projected in WMO (2022) is more complicated. While the starting values are similar, the WMO (2022) bank declines much faster than our total bank. Because the starting point for WMO (2022) was taken from Lickley et al. (2022), our agreement with the starting point is expected. After that, the different approach here leads to differently shaped total bank curves. The WMO (2022) approach to calculated future emissions and banks assumes that the future bank release of HCFC-141b occurs at the same fractional rate of the total bank each year into the future. This approach does not allow for the fact that once in the inactive bank and after decommissioning emissions have occurred, HCFC-141b will almost certainly be released more slowly over time than when averaged over some portion of the previous life cycle stages, which include some periods of rapid release. Thus, it is expected that the total bank here would decline more slowly than that of WMO (2022) after some amount of time. We can also compare our active bank estimates with those shown in Fig. 3.11 of TEAP (2023). When correcting for our low-emission bias as discussed above, while the active bank peaks around the same time, i.e., between 2010 and 2020, our estimate is about 40% higher. Also, it is clear that the active bank in our calculations drops off more slowly than those of TEAP (2023), with ours remaining above 400 Gg in 2050, while theirs is close to 0. The reasons for these differences are unclear.

The separation of active and inactive banks, shown in Figure 7, has the potential to provide more useful information for policymakers regarding any potential climate and/or ozone benefit of mitigating bank emissions than projections of solely total bank values can. This is because capturing foams that are already in landfills in large amounts is particularly challenging. The ozone assessment projection for the total bank of HCFC-141b in 2030 is about 1400 Gg while ours is about 1700 Gg; however, our calculations show that almost 40% of our total bank will have been landfilled at that time and will no longer be in equipment or applications. The specific product/application that contains the foams further affects the feasibility of capturing the HCFC-141b; in fact, the amount of the 2030 bank in all refrigeration foam applications will only be about 75 Gg. We calculate that the primary active banks now and in the future will be in foams used in buildings, most of which have been found to be expensive, perhaps even prohibitively so, to recover (Caleb Management Services Ltd., 2010; ICF International, 2011).

Due to the large variation in the sizes of the various markets as well as in the magnitudes and the uncertainties ascribed to each parameter, there is a large variation in the impact of the uncertainty of each parameter in Tables 1 and 2 on emissions and bank calculations. To identify the key values and uncertainties in the calculations, we have performed Monte Carlo calculations for each parameter individually, with all others fixed, and categorized the results by the maximum standard deviation they cause in the emissions time series. In Tables 1 and 2, the parameters with the most significant uncertainties are categorized in green and represent a maximum standard deviation in emissions of between 1 and 5 Gg yr^{-1}. The second group is categorized in purple and represents a maximum standard deviation between 0.5 and 1 Gg yr^{-1}, while the remaining parameters lead to a standard deviation smaller than 0.5 Gg yr^{-1} in all years. It is worth nothing that these results are highly dependent on both the estimated parameter values as well as their assigned uncertainties.

4 Conclusions

We have presented a new, bottom-up model that tracks the amount of HCFC-141b in different phases of its product life cycles as well as the emissions that occur throughout those life cycles. The calculations are performed for 10 geographic regions and for 11 foam markets. HCFC-141b was chosen for this work primarily because it is characterized by one of the most complete datasets of the controlled ozone-depleting substances. Production was already required to be reported under the Montreal Protocol by the time HCFC-141b began to be used in substantial quantities, and atmospheric observations were also well established by that time. The model is not constrained to atmospheric observations, and thus represents an independent calculation of emissions and banks over time, unlike many other approaches that are constrained to observations in some manner (e.g., (Lickley et al., 2022; Western et al., 2022; WMO, 2011, 2014, 2018, 2022; Velders and Daniel, 2014). Such a bottom-up approach can provide key information regarding compliance with international agreements.

Our bottom-up model provides information about banks in each specific application in which the HCFC-141b has been used, thus allowing for a much better evaluation for the feasibility of capturing banks before they are released to the atmosphere. While the approach presented allows for including changing fundamental emission parameters over time, that has only been done here for recovering banked HCFC-141b in refrigerator foams in Europe from 2002 onward. While other changes have likely occurred, there is not enough information for us to confidently make any other modelling adjustments. We have attempted to be liberal with our uncertainty estimates, however, to account for potential changes.

The comparison of calculated emissions with emissions estimated from global atmospheric measurement networks is quite good in terms of the temporal shape from the beginning of HCFC-141b use through the late 2010s, although our emissions are generally slightly lower. The most straightforward parameter change that would bring the calculated emissions higher across all years is to increase the emissions associated with production. Importantly, however, this is not the only way to improve agreement with observationally derived emissions. For the last few years, our calculated emissions and measurement-derived emissions increasingly diverge, suggesting there may be growing additional sources of emissions not included in the model. It could also be that model parameters changing over this time period may have caused some or all of the discrepancy; however, these changes have not been required to match the shape of observationally derived emissions over the entire previous calculation period.

Historical total bank calculations compare well to those of Lickley (2022) within the rather broad 2-sigma error bars of both studies. Future total bank estimates begin in good agreement with those of WMO (2022) but the size of our bank estimates drops off more slowly over time. This difference is not surprising, given the differing approaches to projecting future banks. Future projections of active bank sizes calculated here fall off more quickly than the total bank. This rapid decline implies that the window of time is shorter than one might expect from the results of Daniel et al. (2022) if one desires to intervene and keep the HCFC-141b in the bank from being released into the atmosphere at some point in the future. Furthermore, our analysis shows that by 2040, the majority of the HCFC-141b in banks will be found in spray foams and discontinuous panels, both used in buildings, and thus likely more expensive to extract before building demolition than capturing an ODS from, for example, refrigeration units. Even in 2030, the more easily accessible foam banks residing in domestic and commercial refrigeration and refrigerated containers (reefers) comprise less than 10% of the total active bank and less than 5% of the total bank.

In this work, we have applied the model to the ODS HCFC-141b, which is primarily used in foams; it could also be used to explore other ODSs and hydrofluorocarbon (HFC) banks and emissions from foam applications as well as from other non-foam applications, such as fire-fighting equipment and use as refrigerants. We have performed all calculations by starting with values for each of the input parameters along with their uncertainties. It would also be possible to use the model in combination with emissions estimated from atmospheric measurements to constrain some of the key parameters so the fit to the observations would be improved. Doing so would mean that the estimated emissions would no longer be independent of the observationally derived emissions. This would imply that calculated emissions would no longer be as clear of an indication of compliance with the Montreal Protocol. On the other hand, this approach would yield bank estimates that are at least more consistent with the observationally derived emissions even if not necessarily more accurate by specific application. A challenge to overcome would be that with so many parameters, many of which lead to similar emission shape changes over time, the correlations affecting parameter retrieval would have to be explored in a careful way.

Code availability. The code used for this paper is publicly available and can be found at https://csl.noaa.gov/groups/csl8/modeldata/

Data availability. The data used for this paper is partially restricted. For further information and requests, please contact the Ozone Secretariat, with contacts found at https://ozone.unep.org

Author contribution. HWT developed the model on which the manuscript is based. JD wrote the software implementing the model. HWT and JD wrote the manuscript draft. CT designed the Fig. 1 schematic, and reviewed and edited the manuscript. LW provided emissions estimates from observations, and reviewed and edited the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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Appendix A

Table A1. Sources used to determine parameters presented in Table 1. TEAP is the Technology and Economic Assessment Panel of the Montreal Protocol.

Application	Emission During Manufacturing and Installation	Annual Emissions During Foam Use	Failure Rate Parameters for Weibull function	
			Weibull Scale Factor	Weibull Lifetime Term
Refrigeration				
Domestic refrigeration	TEAP assessment based on foam installation practices and what is given in the "FTOC" column in Table A4.3 of TEAP (2019). Values are greater than or equal to measurements for panel installation for domestic refrigerators (Aprahamian and Bowman, 2005). All values are identical to the referenced table except for discontinuous panels, which is assumed to be 10% rather than 20%.	Same as Table A4.3 of TEAP (2019). Values are greater than or equal to the change in conductivity measured in the Wilkes et al. studies (Wilkes et al., 2001; Wilkes et al., 2003)	(UNFCCC, 2017)	(UNFCCC, 2017)
Commercial refrigeration				(Doe, 2014; TEAP, 2019)
Refrigerated containers				(Epa, 2011)
Building Construction				
Discontinuous Panels			From global residential results in Deetman et al. (2020); other relevant references are Aktas and Bilec (2012) and Andersen and Negendahl (2023)	From global residential results in Deetman et al. (2020); other relevant references are Aktas and Bilec (2012) and Andersen and Negendahl (2023)
Continuous panels				
Spray foam				
PU boardstock			(TEAP, 2019)	(TEAP, 2019)
Other Uses				
PU block and pipe		(TEAP, 2019), assuming value in table is a factor of 10 too large	(TEAP, 2019)	(TEAP, 2019)
PU pipe in pipe		Treated like discontinuous and continuous panels in *TEAP* (2019) due to unexpectedly large value in TEAP table		15 yrs was used in the calculations of TEAP (2019) owing to unpublished suggestions that previous lifetime assumptions were too large. Table A4.3 provides values

				of 50 and 75 yrs., so here, we compromise and use 30 yrs with a larger 1-s.d. uncertainty of ±30%
PU block foam slab		(TEAP, 2019)		(TEAP, 2019)
PU integral skin	Taken as smaller than many references to account for the nuance that some integral skin manufacturing results in open cells and some results in closed cells; however, due to small market for HCFC-141b, our assumption is rather insignificant.	Assumed based on skin sealing the cells and allowing for only slow release	Assumed to be the same as "Other Uses" in Table 1	Same as in TEAP (TEAP, 2019) although not listed in Table A4.3

Table A2. Sources used to determine parameters presented in Table 2.

Type of Emission	**Magnitude and Timing**
Before sale (includes loss during production)	Consistent with TEAP (2019), although arguably on the lower end when considering both advanced and less sophisticated production plants
During Decommissioning	Drawn from publications that have evaluated impact of foam shredding at time of decommissioning (Kjeldsen and Scheutz, 2003; Scheutz and Kjeldsen, 2002) and considering that much foam is not shredded across the global on disposal
From inactive bank (i.e., post-decommissioned bank such as landfill)	Used lower value of 0.5% from Table A4.3 (TEAP, 2019) for all applications
Solvent Use	See text of Sect. 2.1

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