

Bayesian assessment of chlorofluorocarbon (CFC), hydrochlorofluorocarbon (HCFC) and halon banks suggest large reservoirs still present in old equipment

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Abstract

Halocarbons contained in equipment such as air conditioners, fire extinguishers, and foams continue to be emitted after production has ceased. These ‘banks’ within equipment and applications are thus potential sources of future emissions, and must be carefully accounted for in order to evaluate nascent production versus banked emissions. Here, we build on a probabilistic Bayesian model, previously developed to quantify CFC-11, 12 and 113 banks and their emissions. We extend this model to a suite of the major banked chemicals regulated under the Montreal Protocol (HCFC-22, HCFC-141b, and HCFC-142b, halon-1211, and halon-1301, and CFC-114 and CFC-115) along with CFC-11, 12 and 113 in order to quantify a fuller range of ozone-depleting substance banks by chemical and equipment type. We show that if atmospheric lifetime and prior assumptions are accurate, banks are very likely larger than previous international assessments suggest, and that total production has been very likely higher than reported. We identify that banks of greatest climate-relevance, as determined by global warming potential weighting, are largely concentrated in CFC-11 foams and CFC-12 and HCFC-22 non-hermetic refrigeration. Halons, CFC-11, and 12 banks dominate the banks weighted by ozone depletion potential. Thus, we identify and quantify the uncertainties in substantial banks whose future emissions will contribute to future global warming and delay ozone hole recovery if left unrecovered.

1. Introduction

The Montreal Protocol regulates the production of ozone-depleting substances (ODPs), and its implementation has avoided a world with catastrophic stratospheric ozone depletion (Newman et al., 2009). Globally, there has been a near-cessation of chlorofluorocarbon (CFC) and halon production since 2010, and global production of the replacement hydrochlorofluorocarbons (HCFCs), are scheduled to be phased-out by 2030. Despite production phase-out, these chemicals persist in old equipment produced prior to phase-out, such as refrigeration, air conditioners, foams, and fire extinguishers. These reservoirs of materials (termed ‘banks’) continue to be sources of emissions (e.g., WMO, 2018). Previously published estimates of bank sizes and bank emissions vary widely due to different estimation techniques that incorporate incomplete or imprecise information (TEAP, 2009; WMO, 2003). This uncertainty obscures

47 ongoing emissions attribution and undermines international efforts to evaluate global compliance
48 with the Montreal Protocol. In earlier work, Lickley et al. (2020, 2021) developed a Bayesian
49 probabilistic banks model for CFCs that incorporates the widest range of constraints to date
50 (Lickley et al., 2020, 2021). Here, we extend this model to the suite of major chemicals
51 regulated by the Protocol that are subject to banking.

52 Previously published assessments typically rely on one of three modeling approaches to
53 estimate bank sizes and to then estimate emissions associated with these banks. In the “top-
54 down” approach (e.g. WMO, 2003), banks are estimated as the cumulative difference between
55 reported production and observationally-derived emissions. However, by taking the cumulative
56 sum of a small difference between two large values, small biases in emissions or reported
57 production estimates can propagate into large biases in bank estimates (Velders & Daniel, 2014).
58 Some type of bias is thus expected since total production has very likely been less than reported
59 production both due to under-reporting of production (e.g. Gamlen et al., 1986; Montzka et al.,
60 2018) and due to the exclusion of point of production losses in reported production values.
61 Further, emissions estimates rely on observed concentrations along with global lifetime
62 estimates, which have large uncertainties associated with them (SPARC, 2013).

63 The second approach relies on a “bottom-up” accounting method (Ashford et al., 2004;
64 IPCC/TEAP, 2006), where the inventory of sales by equipment type are carefully tallied along
65 with estimated release rates by application use. The bottom-up approach also relies on sales data
66 from surveys of various equipment types and products as well as estimates of their respective
67 leakage rates (SROC, 2005). These are all subject to uncertainties, which contributes to
68 uncertainties in bottom-up bank estimates as well. A limitation of the bottom-up method is that
69 observed atmospheric concentrations are used only as a qualitative check and are not explicitly
70 accounted for in the analysis. Another important limitation is that data used in the bottom-up
71 accounting method are unobserved but rather rely on estimated processes along with reported
72 data, such as production or sales of equipment, thus bias in reporting could propagate into large
73 biases in bank estimates.

74 The third approach, and the one used in more recent ozone assessments (WMO, 2011, 2014,
75 2018) uses a hybrid approach to calculate banks. Bottom-up banks estimated for 2008 are used
76 as the starting point of the calculations. These banks are taken from SROC (2006) and represent
77 interpolated values from the 2002 and 2015 estimates. The banks are then brought forward to the
78 present time by adding the cumulate reported production and subtracting the cumulative
79 observationally-derived emission from 2008 through the present. This approach is consistent
80 with 2008 bottom-up bank estimates by design, however, as time between 2008 and the present
81 has grown, the cumulative errors associated with the top-down approach have become larger.

82 The modeling approach applied in the present study relies on Bayesian inference of
83 banks (Lickley et al., 2020, 2021) where banks are estimated using an approach called Bayesian
84 parameter estimation. In this approach a simulation model of the bottom-up method is
85 developed, where prior distributions of input parameters are constructed from previously
86 published values, accounting for large uncertainties in production and bank release rates. The
87 simulation model simultaneously models banks, emissions, and atmospheric concentrations.
88 Parameters in the simulation model are then conditioned (or updated) on observed concentrations
89 by applying Bayes’ theorem. The final result is a posterior distribution of banks by chemical and
90 equipment type, along with an updated estimate of production and release rates for each
91 equipment type. This approach incorporates data and assumptions from both the bottom-up and

92 top-down approaches, providing a simulation model consistent with the bottom-up accounting
93 approach while also being consistent with observed concentrations within their uncertainties.

94 The remainder of the paper includes the following: Section 2 presents the Bayesian modeling
95 approach along with data used in the analysis. Section 3 provides a summary of the results of
96 our analysis for each of the chemicals considered here. Finally, Section 4 provides a discussion
97 of our primary findings and limitations of the analysis.

98 99 **2. Methods**

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101 The Bayesian modeling approach from Lickley et al. (2020, 2021) draws on a Bayesian analysis
102 approach called Bayesian melding, designed by Poole & Raftery (2000), that allows us to apply
103 inference to a deterministic simulation model. We employ a version of this method that we
104 henceforth refer to as Bayesian Parameter Estimation (BPE), which allows for input parameter
105 uncertainty (Bates et al., 2003; Hong et al., 2005). The model flow is implemented as follows;
106 first we develop a deterministic simulation model, representing the “bottom-up” accounting
107 method that simultaneously simulates banks, emissions, and mole fractions for each chemical
108 and equipment type. In this analysis, the chemicals considered include CFC-11, 12, 113, 114,
109 and 115, HCFC-22, 141b, and 142b, and halon-1201, and 1311. Prior distributions for each of
110 the input parameters are based on previously published estimates. We then specify the
111 likelihood function as a function of the difference between observed and simulated mole
112 fractions. Finally, we estimate posterior distributions of both the input and output parameters by
113 implementing Bayes’ Rule using a sampling procedure. Each of the steps of the BPE are
114 described in more detail below.

115 116 **2.1 Simulation Model**

117 The simulation model is comprised of equations (1) – (5) which simultaneously models banks,
118 emissions, and mole fractions for each chemical by equipment type for all years with available
119 data up until 2019. Starting dates differ by chemical, see the Supplement for details. The
120 simulation model is specified as follows;

$$121
122 B_{j,t+1} = (1 - RF_{j,t}) \times B_{j,t} + (1 - DE_{j,t}) \times P_{j,t} \quad (1)$$

123
124 where $B_{j,t}$, is banks and $P_{j,t}$ is production of equipment category, j , in year, t . $RF_{j,t}$ reflects the
125 fraction of the bank released and $DE_{j,t}$ reflects the fraction of production that is directly emitted
126 in equipment category, j , year, t . These same parameters are used to simulate emissions, $E_{j,t}$:

$$127
128 E_{j,t+1} = RF_{j,t} \times B_{j,t} + DE_{j,t} \times P_{j,t} \quad (2)$$

129
130 Total banks, $B_{\text{Total},t}$, and total emissions, $E_{\text{Total},t}$, are then estimated as the sum across all N
131 equipment categories;

$$132
133 B_{\text{Total},t} = \sum_{j=1}^N B_{j,t} \quad (3)$$

$$134
135 E_{\text{Total},t} = \sum_{j=1}^N E_{j,t} \quad (4)$$

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137 For chemicals where feedstock usage is reported, an additional term in eq (4) is included that
 138 accounts for feedstock emissions. Emissions are then used to simulate atmospheric mole
 139 fractions, MF_t , along with an assumed atmospheric lifetime, τ_t , taken as the SPARC (2013)
 140 multi-model time-varying mean;

$$141 \quad MF_{t+1} = \exp\left(\frac{-1}{\tau_t}\right) \times MF_t + A \times E_{\text{Total},t} \quad (5)$$

142 where A is a constant that converts units of emissions by mass to units of mole fractions, and
 143 also takes into account a factor of 1.07 that accounts for the discrepancy between surface mole
 144 fraction concentrations to global mean values.

145 2.2 Prior Distributions

146 The input parameters in the simulation model described above require initial values to be
 147 assigned, along with their probability distributions. These prior distributions ('priors') are
 148 developed to estimate mole fractions, emissions, and banks for CFC-11, 12, 113, 114, and 115,
 149 HCFC-22, 141b, and 142b, and halon-1201, and 1311. Categories of bank equipment are
 150 defined by the categorization provided by AFEAS (2001), which varies by compound (shown in
 151 Table 1). For halons, there is a single category of bank (fire extinguishers).

152 AFEAS data reports global annual production up to 2001 categorized by equipment type,
 153 which is generally categorized into short, medium and long-term banks. We use AFEAS data
 154 and categorization to develop our production priors and adopt the WMO (2003) correction where
 155 AFEAS production values are used up until 1989 and then scaled to match UNEP global
 156 production values for all years following 1989. After AFEAS data ends, we assume the relative
 157 production in each category remains constant for all years following 2001. Uncertainty in
 158 production priors is assumed to follow a multivariate log-normal distribution, where temporal
 159 correlation in production reporting bias is estimated in the BPE. Prior distributions differ by
 160 chemical and are developed to be wide enough for atmospheric mole fraction priors to contain
 161 observations. See the Supplement for details on production priors for each chemical.

162 The emissions function by bank equipment type can be characterized by the fraction of
 163 production that is directly emitted during the year of production (DE) and the fraction of the
 164 bank that is emitted in each subsequent year. Prior estimates for emissions functions come from
 165 previously reported data and differ by chemical and equipment type (see the Supplement).
 166 Broadly speaking, it has been estimated that chemicals contained in short-term banks are fully
 167 emitted within the first two years after production, medium-term banks lose about 10 – 20% of
 168 their material each year, and long-term banks can lose as little as 2% of their material each year
 169 (Ashford et al., 2004). We use previously published estimates to develop emissions function
 170 priors specific to each chemical and bank type along with wide uncertainties, as specified in the
 171 Supplement.

172 Amounts of halocarbons used for feedstock production are available annually
 173 (UNEP/TEAP, 2021). A prior mean leakage rate of 2% was assumed during production, which
 174 reflects a medium value between different facilities (MCTOC, 2019).

175 **Table 1:** Application type of halocarbon banks by chemical

176 Chemical	177 Short Bank	178 Medium Bank	179 Long Bank
180 CFC-11	Aerosols	Non-hermetic refrigeration	Closed-cell foam

	Open-cell foam		
CFC-12	Aerosols Open-cell foam	Non-hermetic refrigeration	Refrigeration
CFC-113	solvents		Heat pump
CFC-114			Heat pump
CFC-115	Propellant		Air conditioning
HCFC-22	Open-cell foam	Non-hermetic refrigeration	Foam
HCFC-141b	Open-cell foam	Non-hermetic refrigeration	Foam
HCFC-142b		Non-hermetic refrigeration	Foam
Halon-1211		Fire extinguishers	
Halon-1301		Fire extinguishers	

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2.3 Likelihood function

For each chemical, the likelihood function is a multivariate normal likelihood function of the difference between modeled and observed mole fractions;

$$P(D_{t1}, \dots, D_{tN} | \theta) = \frac{1}{(2\pi)^{\frac{N}{2}} \sqrt{|S|}} \exp \left\{ -\frac{1}{2} \Delta^T S^{-1} \Delta \right\} \quad (6)$$

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Where D_{t1}, \dots, D_{tN} is yearly globally-averaged observed mole fractions for all years where observations are available and θ represents that vector of input and output parameters from the simulation model. Δ is an $N \times 1$ vector of the difference between yearly observed and modeled mole fractions and is assumed to have a mean zero, and covariance function S . S therefore represents the sum of uncertainties between observed and modeled mole fractions. While there are published estimates of uncertainties in observed mole fractions, we do not know the uncertainties in modeled mole fractions. Therefore, we estimate S separately for each chemical, as is done in (Lickley et al., 2020). The off-diagonals in the covariance function incorporate a correlation term, ρ_S , which accounts for our assumption that there is high autocorrelation in the bias between modeled and observed mole fractions. Correlation terms for each chemical are reported in the Supplement along with prior estimates of the uncertainty parameters used for diagonal elements in S . Each column and row in S is therefore populated as;

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$$S_{i,j} = \sigma_i \sigma_j \rho_S^{|i-j|}$$

where σ_i and σ_j represent the sum of the uncertainties in observed and modeled mole fractions at time i and j , respectively, and are inferred in the BPE, whereas ρ_S is prescribed.

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Observations come from the Advanced Global Atmospheric Gas Experiment (AGAGE; <https://agage.mit.edu>) data set (Prinn et al., 2000; Prinn et al., 2018), with the exception of CFC-11 and 12 which, following Lickley et al. (2021), come from the AGAGE and the National Oceanographic and Atmospheric Administration's (NOAA) merged data sets (Engel et al., 2019). Data are aggregated into annual global mean mole fractions. The time frame of availability of observations differs by chemical (see the Supplement).

2.4 Posterior Distributions

214 Following Bayes' Rule, we specify our posterior distribution as;

$$215 \quad 216 \quad P(\boldsymbol{\theta}|D_{t_1}, \dots, D_{t_N}) = \frac{P(\boldsymbol{\theta})P(D_{t_1}, \dots, D_{t_N}|\boldsymbol{\theta})}{P(D_{t_1}, \dots, D_{t_N})} \quad (7)$$

217
218 Where $P(\boldsymbol{\theta})$ represents the joint prior distribution of the input and output parameters described
219 in the simulation model in Section 2.1.

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221 The analytical form of the posterior distribution is intractable. Thus, we estimate the posterior
222 using a sampling procedure (the sampling importance resampling (SIR) method) to estimate the
223 marginal posterior distributions (Bates et al., 2003; Hong et al., 2005; Rubin, 1988). To
224 implement SIR we draw 1,000,000 samples from the priors, run the simulation model, and then
225 resample from the priors 100,000 times using an importance ratio, which is proportional to the
226 likelihood function. These sample sizes were chosen such that multiple iterations of the model
227 produce consistent results.

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229 3. Results

230 Figure 1 shows observed globally averaged mole fractions compared to BPE estimated mole
231 fractions for each chemical. Figure 2 shows BPE estimated and observationally-derived
232 emissions, assuming the SPARC time-varying multi-model mean lifetime for each species.
233 Posterior estimates agree well with observations for the majority of time periods and chemicals.
234 Note, however, that BPE estimates from Lickley et al. (2021) match observed and
235 observationally-derived estimates more closely for CFC-11 than they do in the present analysis.
236 We attribute this difference in consistency to atmospheric lifetimes being assumed in the present
237 analysis, whereas they were inferred in Lickley et al. (2021), which found inferred lifetimes to be
238 somewhat shorter than the SPARC multi-model mean values. Shorter lifetimes would allow
239 modeled mole fractions to decline more quickly following 1990, better matching observations. A
240 notable discrepancy occurs for CFC-115, where modeled mole fractions are increasing
241 throughout the entire simulation period, whereas observed mole fractions from 2000 onwards are
242 relatively constant. This discrepancy could be explained by the large uncertainties in
243 atmospheric lifetimes of CFC-115 (Vollmer et al., 2018), if atmospheric lifetimes are in fact
244 substantially shorter than the SPARC multi-model mean.

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246 Figure 3 provides a comparison of BPE bank estimates alongside previously published bank
247 estimates. BPE bank estimates are generally higher than other published values. This can be
248 explained by production uncertainties that are accounted for in the present analysis. Our analysis
249 suggests that production has very likely been underreported for nearly all chemicals. Table 2
250 provides a summary of our estimated bias in cumulative reported production throughout the
251 simulation period for each chemical type. With the exception of CFC-113 and CFC-115, we find
252 our inferred cumulative production to be significantly higher than reported production (at the 1-
253 sigma level), with our median estimate suggesting that production was as little as 9% higher than
254 reported for CFC-12 and as high as 50% higher than reported for Halon-1211. We would expect
255 any consistent bias in reported production to be a bias low, since consistent undercounting of
256 production is more plausible than overcounting production. The exception for this would be the
257 base year, which reduction targets are made with reference to. In this instance, we would expect
258 overreporting for this year to be more likely. Another possible explanation for the discrepancy in
259 production estimates is that total reported chemical production under the UNEP does not account

260 for leakage during chemical manufacturing, but rather only leakage that occurs during the
 261 application of the chemical. To our knowledge, this potential leakage during chemical
 262 manufacturing has not been well-documented or previously quantified.

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265 **Table 2:** Estimated bias in cumulative reported production. Values indicate the percent
 266 difference between inferred cumulative production from the onset of production to 2019 relative
 267 to reported production, for all uses except for feedstock production. Positive values indicate the
 268 percent by which inferred production is higher than reported.

Chemical Name	CFC-11	CFC-12	CFC-113	CFC-114	CFC-115
Median inferred bias (16th, 84th percentile)	12% (9%, 13%)	9% (7%, 11%)	-1% (-3%, 0%)	11% (9%, 13%)	-1% (-2%, 5%)
Chemical Name	HCFC-22	HCFC-141b	HCFC-142b	Halon-1211	Halon-1301
Median inferred bias (16th, 84th percentile)	10% (6%, 13%)	12% (6%, 19%)	22% (17%, 28%)	50% (41%, 59%)	24% (18%, 32%)

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271 Figure 4 shows the breakdown of emissions by equipment type over time. For CFCs, emissions
 272 from short-term banks tend to peak around 1990, as spray applications were banned earlier than
 273 other applications, after which emissions from medium and long-term banks become more
 274 dominant emission sources. This is to be expected as the phase-out of production after 1990
 275 would lead to more CFC emissions from existing banks rather than new, short-lived equipment.
 276 For HCFC-22, most of the emission throughout the entire time period is from medium banks,
 277 which is largely non-hermetic refrigeration. Long banks (i.e. foams) dominate emissions for
 278 HCFC-141b, and for HCFC-142b, where both foams and non-hermetic refrigeration are
 279 prominent emission sources throughout the simulation period. Estimated feedstock emissions
 280 averaged over 2010 – 2019 are shown in Table 3. HCFC-22 is the largest source of feedstock
 281 emissions by mass, but CFC-113 feedstock emissions are estimated to be larger when weighted
 282 by GWP100 and ODP.

283

284 **Table 3:** Estimated feedstock emissions averaged from 2010 – 2019 from the Bayesian analysis.
 285 Emissions are weighted by mass, global warming potential (GWP100) relative to CO₂ over a
 286 100-year time horizon for a CO₂ concentration of 391ppm, and by ozone depletion potential
 287 (ODP) relative to CFC-11 (WMO, 2018).

Feedstock Emissions	CFC-113	HCFC-22	HCFC-142b
By mass	3.4 Gg/yr	9.3 Gg/yr	2.1 Gg/yr
By GWP100	20, 838 Gg/yr	16,591 Gg/yr	4,302Gg/yr
By ODP	2.8 Gg/yr	0.3 Gg/yr	0.1 Gg/yr

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289 Figure 5 shows the relative quantity of banked materials by chemical type. Banks are weighted
 290 by mass (Figure 5a), by global warming potential (GWP100; Figure 5b), and by ozone depleting
 291 potential (ODP; Figure 5c). Our best estimate is that the sum of the HCFCs currently comprise
 292 about 77% of banks by mass. However, in terms of climate impacts, CFC-11, 12 and HCFC-22
 293 are the largest banked materials weighted by GWP100, accounting for 36%, 14%, and 36% of
 294 current banks, respectively. When banks are weighted by ODP, CFC-11 and 12 represent 46%
 295 and halons also represent 46% of current banked chemicals.

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297 Figure 6 shows the composition of banks by chemical type. This, together with Figure 5,
298 provides insight into the most prominent banked sources of halocarbons with regards to
299 GWP100 and ODP. In terms of GWP100, CFC-11 banks largely reside in foams, whereas CFC-
300 12 and HCFC-22 are largely in non-hermetic refrigeration; the latter may be more readily
301 recoverable. In terms of ODP, CFC-11 foams and CFC-12 non-hermetic refrigeration remain
302 important, along with halons which are all contained in fire extinguishers, a recoverable
303 reservoir.

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306 **4. Discussion and Conclusions**

307 This analysis suggests that if lifetime assumptions are correct, published bank estimates using
308 either the top-down or bottom-up methods were likely underestimating bank sizes for all banked
309 chemicals due to underreporting of production (see Table 2). The Bayesian approach used in this
310 analysis does not assume production is known, but rather jointly infers production along with the
311 other parameters in the simulation model, providing probabilistic estimates of historical
312 production values. Previously published bank estimates (Ashford et al., 2004; TEAP, 2009;
313 WMO, 2003) do not infer production, but rather assume it is known, or consider different
314 scenarios. We argue that production assumptions have been biased low due to underreporting of
315 total production and potentially unaccounted for leakage during chemical manufacturing and
316 thus have led to published bank estimates that were also biased low.

317

318 Discrepancies between observed mole fractions and BPE-derived mole fractions are notable for
319 the suite of chemicals considered here. While the majority fall within the 90% confidence
320 interval throughout most of the time periods, the trends in concentrations between observations
321 and inferred mole fractions do not always agree. This discrepancy could be related to our
322 partitioning of production type following 2003 (i.e. after AFEAS data ends). Another important
323 limitation in this analysis is in the treatment of atmospheric lifetimes, which could also explain
324 some of these discrepancies. The present analysis assumes atmospheric lifetimes are known and
325 equal to the SPARC (2013) time varying multi-model mean lifetimes. However, previous work
326 has indicated potential biases in SPARC lifetimes, for example for CFCs (Lickley et al., 2021).
327 The potential bias in atmospheric lifetimes would result in biased bank estimates in the present
328 manuscript and requires further analysis.

329

330 This modeling approach makes no assumptions about end-of-life emissions. Certain bank
331 estimates assume that applications are dismantled at the end of their lifetime, which would both
332 contribute to decreased banks and increased emissions at fixed years after production (e.g. TEAP
333 progress report, 2021). We do not make this assumption as we believe it would be more realistic
334 for dismantling of equipment to occur over a range of years after production, which would
335 effectively be captured by our bank release fraction estimate. We do, however, test the
336 sensitivity of our bank estimate to end-of-life (EOL) emissions occurring in a single year after
337 production. This we term the EOL scenario and test the sensitivity of banks for CFC-11, CFC-
338 12 and HCFC-22, the three largest banks by global warming potential. The modeling approach
339 is described in the SM and results are shown in Figure SM1. Perhaps unexpectedly, CFC-11
340 posterior bank estimates are ~25% higher in 2020 in the EOL scenario relative to the scenario
341 described in the main text. However, banks in the EOL scenario are decreasing faster than those
342 described in the main text. The larger bank size is due to posterior bank release fractions being ~

343 2% for the EOL scenario relative to 3% for the scenario described in the main text. The faster
344 depletion of the banks in 2020 can be explained by the addition of the EOL decommissioning
345 parameter. These larger bank estimates reflect the consistency of the Bayesian modeling
346 approach where all parameters are jointly inferred. Including an additional process in the model
347 requires that multiple parameters be updated to be consistent with observations. For CFC-12, the
348 EOL scenario produces significantly smaller banks from about 1990 onwards, however, the
349 emissions profile has an artificial dip in emissions relative to observationally-derived emissions,
350 suggesting a set year for decommissioning is not a realistic modeling assumption. For HCFC-22
351 banks are not substantially different between the two scenarios.

352
353 There are important discrepancies between CFC-113 feedstock emissions inferred here and those
354 estimated in the previous analysis (Lickley et al., 2020). In Lickley et al. (2020), feedstock
355 emissions were assumed to be the difference between observationally-derived emissions and
356 inferred bank emissions. In the present analysis, prior distributions of feedstock production and
357 leakage rates are developed and feedstock emissions are then inferred. In the present analysis,
358 observationally-derived CFC-113 emissions are higher than total BPE inferred emissions at the
359 1-sigma level from 2010 onwards. This suggests that either observationally-derived emissions
360 are too high, or our BPE estimates are too low. In Lickley et al. (2021), we find that atmospheric
361 lifetimes of CFC-113 are very likely lower than the SPARC multi-model time varying mean,
362 used in the present analysis. This would imply that the observationally-derived emissions shown
363 in Figure 2 are biased low, suggesting an even larger discrepancy between BPE inferred total
364 emissions and observationally derived emissions. Therefore, it seems plausible that the
365 discrepancy is due to prior feedstock emissions estimates being biased low due to larger leakage,
366 or CFC-113 is being produced for a use that is not allowed under the Montreal Protocol.

367
368 Finally, some important details about production and destruction were not fully accounted for in
369 this analysis. For one, feedstock priors were only included for CFC-113, HCFC-22, and HCFC-
370 142b, which could be limiting our assessment of the sources of emissions for other chemicals.
371 However, published feedstock values for other chemicals are not available and leakage rates in
372 feedstock applications may be uncertain. In addition, we do not account for non-dispersive
373 production in our analysis, namely the production of chemicals as by-products. It is possible, for
374 example, that some of the discrepancies in CFC-115 emissions could be explained by non-
375 dispersive emissions as identified by (Vollmer et al., 2018). Further, we do not consider end-of-
376 life destruction of equipment as there are no published records, to our knowledge, of these
377 processes. Finally, we were not able to account for a more detailed breakdown in production by
378 equipment type than what has been published by AFEAS, which discretizes production into, at
379 most, four categories of equipment, and does not provide data beyond 2003. Without publicly
380 available details of these processes, modeling of banks and emissions will continue to be limited.

381
382 **Code Availability:** All analyses were done in MATLAB. All code used in this work is available
383 at <https://github.com/meglickley/HalocarbonBanks>

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385 **Data Availability:** The datasets generated and/or analyzed during the current study are available
386 at <https://github.com/meglickley/HalocarbonBanks>

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388 **Author Contributions.** All authors contributed to the conceptualization of the manuscript.
389 M.L. conducted the analysis. M.L. prepared the manuscript with contributions from all authors.

390
391 **Competing Interests.** The authors declare that they have no conflict of interest.

392
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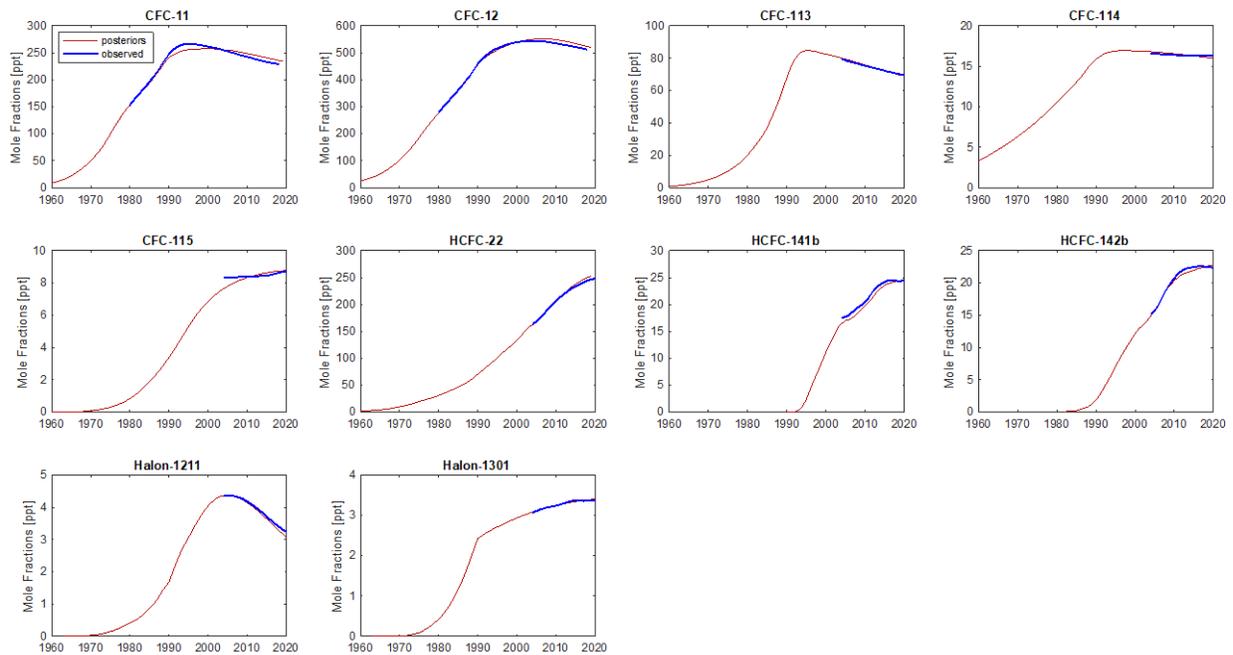
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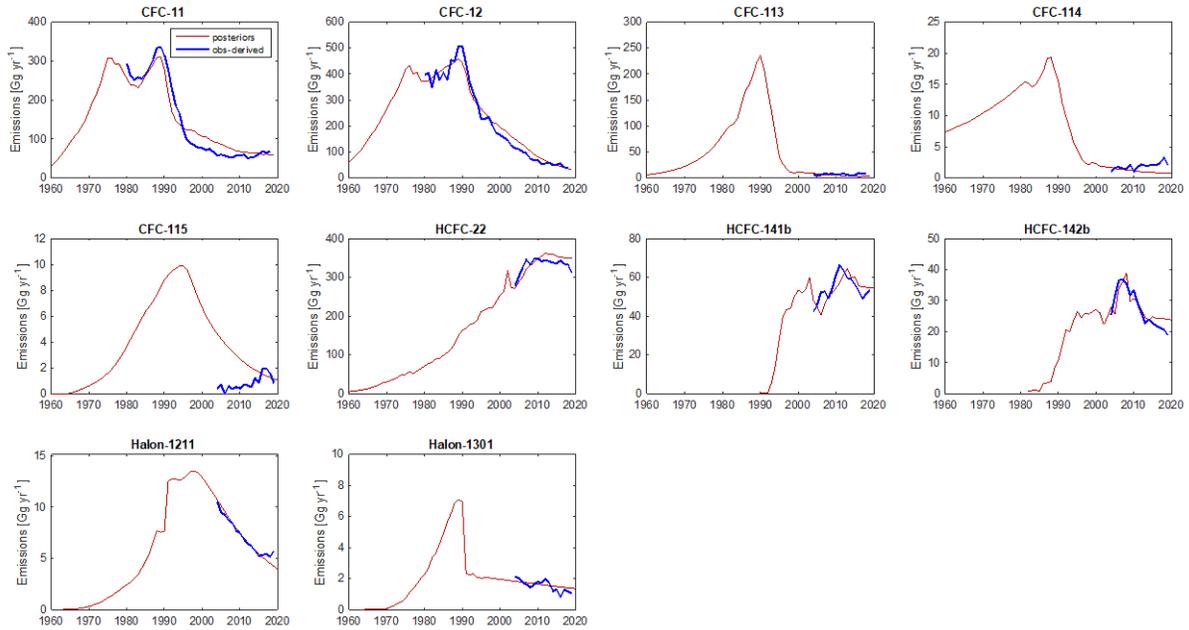
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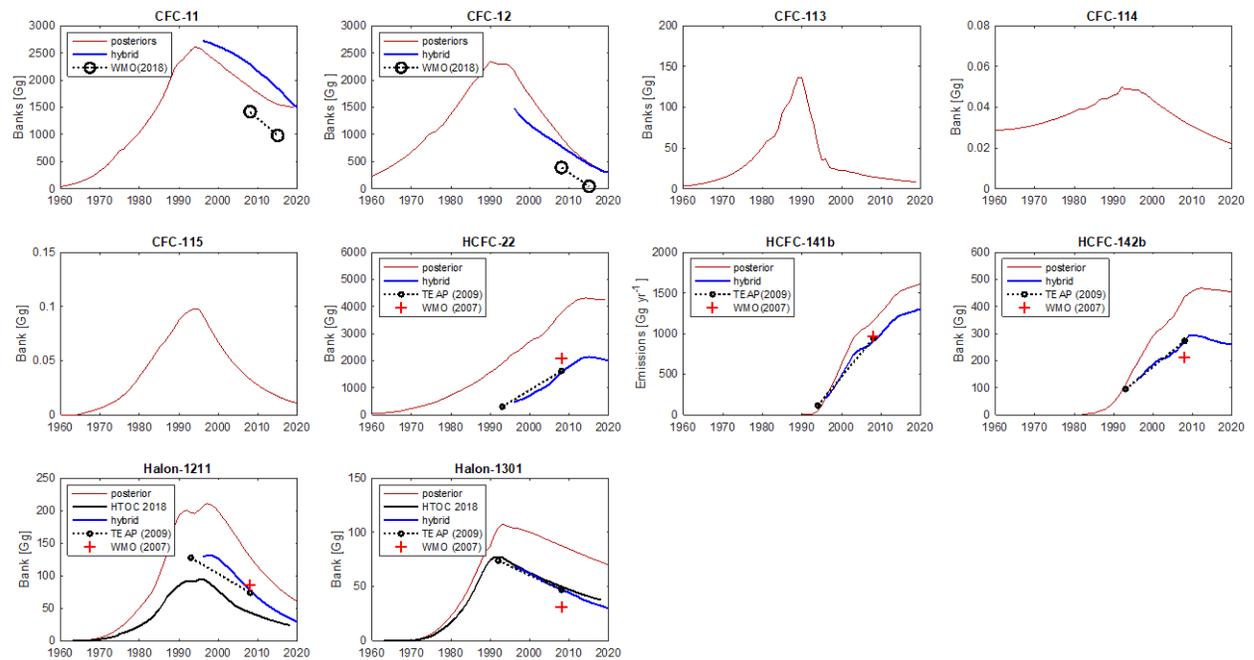
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488 **Figure 1:** Modeled mole fractions versus observed mole fractions. Red lines indicate the
489 posterior median mole fraction estimate from the Bayesian analysis (BPE), with shaded regions
490 indicating the 90% confidence interval. Blue line indicates globally-averaged observed mole
491 fractions.
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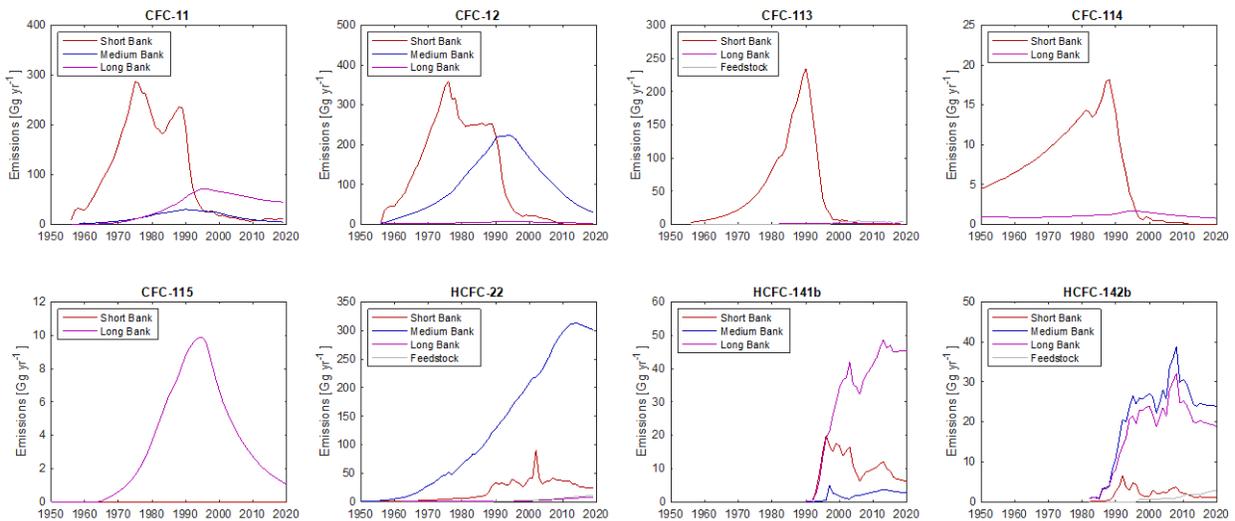


494
 495 **Figure 2:** Modeled emissions versus observationally-derived emissions. Red lines indicate the
 496 posterior median emissions estimate from the Bayesian analysis (BPE), with shaded regions
 497 indicating the 90% confidence interval. Blue line indicates observationally-derived emissions
 498 assuming the SPARC multi-model mean time-varying lifetimes.
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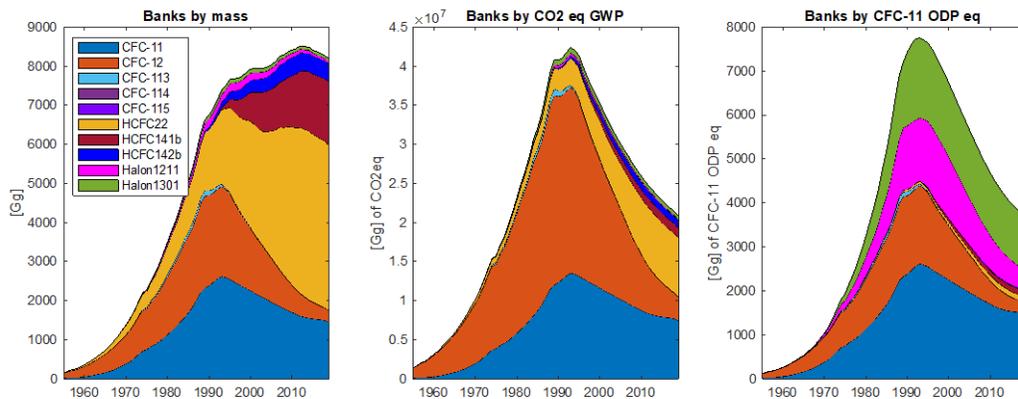


501
 502 **Figure 3:** Magnitudes of Bank estimates. The red line indicates the median posterior estimate of
 503 Banks from the Bayesian analysis, with shading indicating the 90% confidence interval.
 504 Previously published bank estimates are provided for comparison from TEAP (2009), WMO

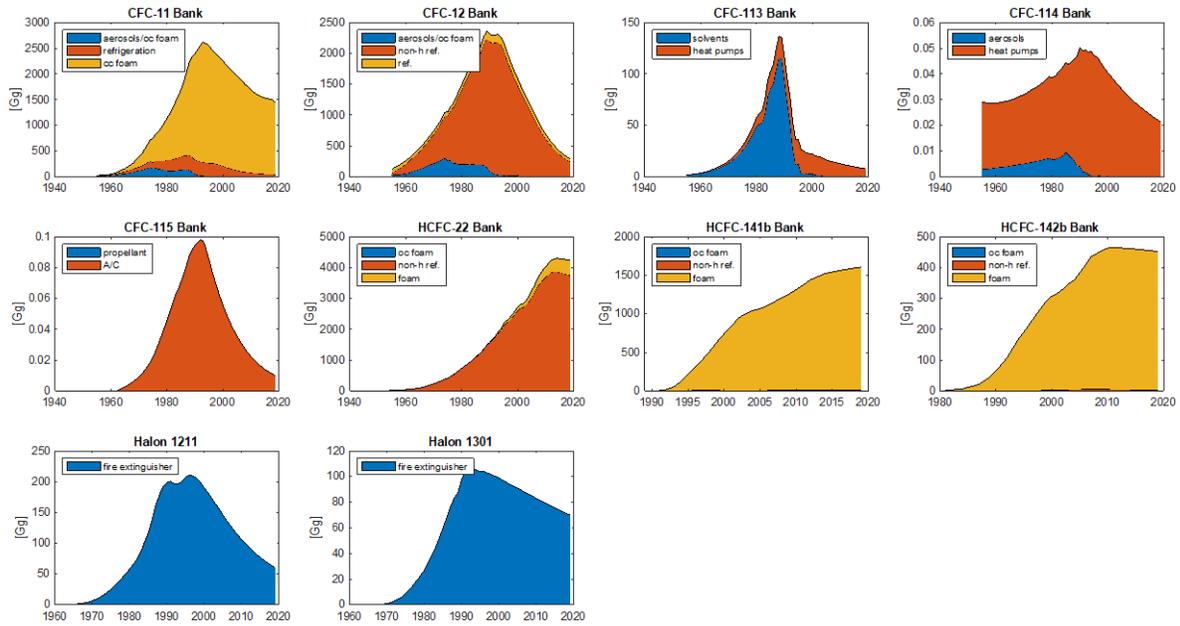
505 (2007), and WMO (2018), along with the hybrid approach updated to current estimated starting
 506 values.
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508
 509 **Figure 4:** Emissions by Source. Emissions estimates by various equipment types, summarized
 510 in Table 1, are shown here along with estimated emissions from feedstock usage. Lines indicate
 511 the median estimate, with the shaded region indicating the 90% confidence interval. Halons are
 512 not included in this figure as 100% of halon emissions come from the same application and are
 513 thus identical to Figure 2 halon totals.



514
 515 **Figure 5:** Total banks by mass, global warming potential (GWP100; WMO, 2018) and ozone
 516 depleting potential (ODP; WMO, 2018). Bank estimates reported in the above figures are the
 517 median estimates from the Bayesian analysis.
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Figure 6: Bank size by equipment type. Bank estimates reported in the above figures are the median estimates from the Bayesian analysis. In the above legends, cc refers to closed-cell foams, non-h ref. refers to non-hermetic refrigeration, ref. refers to refrigeration, and A/C refers to air conditioning.