Analysis of Reported European Emissions Shows Improvement in Containment of Hydrofluorocarbons

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ABSTRACT: Hydrofluorocarbons are greenhouse gases with high Global Warming Potentials that are mainly used in refrigeration and air conditioning. Historically, they were emitted only slowly from such systems and long term containment remains an important engineering and environmental issue.
As part of their commitment under the Rio Convention, nations are required to report annual calculated production and emissions of all greenhouse gases and the reports submitted by European countries have been examined to determine the extent to which containment of HFCs (expressed as the rate of emission from the bank remaining in equipment) has changed with time. Although there is wide variation between countries, the annual emission rates of HFCs have improved significantly over the past ten years and now represent between 5 and 10% of the bank still contained in equipment. This implies that, at the present state of technology, it would take at least 10 to 20 years for the original mass of refrigerant charged into a system to be released into the atmosphere.

1 INTRODUCTION

The F-gases (hydrofluorocarbons, HFCs; perfluorocarbons, PFCs and sulphur hexafluoride, SF_6) are significant but not important contributors to climate change, globally amounting to less than 1% as CO_2 equivalent (Velders et al., 2005). Unlike the other greenhouse gases, which are byproducts of processes such as fuel combustion, these gases are deliberately manufactured and sold for use in a wide range of effects required by society, principally refrigeration and air conditioning. Emissions from these systems vary both in quantity and in time; relatively little material is lost promptly and any leakage that does occur happens when the

[1] Contribution on a personal basis

equipment is serviced or scrapped after it has operated for several years. Consequently emissions cannot be measured easily, either directly or by measuring another activity, like the quantity consumed, and applying a single emission factor. During 2006 emissions of HFCs were 50 mTeCO_2eq, representing 1.2% of all European greenhouse gas emissions. The analysis here concerns European emissions of the principal HFCs used in refrigeration and air conditioning (RAC), specifically: HFC-125 (CF_3CF_2H), HFC-134a (CF_3CFH_2), HFC-143a (CF_3CH_3) and HFC-32 (CF_2CH_2). The contributions of these HFCs to European HFC emissions are shown in Table 1, weighted by CO_2 equivalence.

Table 1. Relative importance of HFC emissions within Europe

Importance of Emissions relative to the total of HFC emissions during 2006

	Aerosol	Foam	RAC	Other uses	Total
HFC-134a	11.5%	4.8%	42.7%		59.0%
HFC-143a			16.9%		16.9%
HFC-125			16.2%	0.1%	16.3%
HFC-32			1.0%		1.0%
HFC-152a		0.9%			0.9%
All Other HFCs					5.9%

2 HFC USES

2.1 *HFC-125 (pentafluoroethane)*

This fluorocarbon is used almost[2] exclusively in blends with HFCs 134a, 143a and 32 which are employed in commercial refrigeration systems, for example display cabinets in supermarkets. Historically, when using CFCs or HCFCs, these were particularly leaky systems, with upwards of 30%/year loss rates (Ashford et al., 2004a; 2004b).

2.2 *HFC-134a (1,1,1,2-tetrafluoroethane)*

HFC-134a is the only fluorocarbon now used in mobile air conditioning. It is also used alone in refrigeration and in blends with HFCs 125, 143a and 32 in commercial refrigeration and air conditioning. The minor uses in aerosol propulsion and in blowing insulating foam are significant because they represent, on the one hand, prompt emissions and, on the other, very long term emissions. However, the general picture for HFC-134a is similar to that of HFC-125.

2.3 *HFC-143a (1,1,1-trifluoroethane)*

Like HFC-125, this is almost exclusively used in refrigerant blends in system that were, historically when using CFCs, very leaky.

2.4 *HFC-32 (difluoromethane)*

Again, this is a component of blended refrigerants.

[2] Minor quantities are used in fire protection systems with unpredictable, but generally very low, emission rates

3 EMISSION PATTERNS

Emissions of material used in aerosols are prompt (within 2 years). By contrast, the fluorocarbon used to blow plastic insulating foam is essentially trapped within the foam matrix so that emission patterns are dominated by initial losses and releases on disposal. The combination of these losses with the small amount lost during service gives a time interval for total emission of the agent of 30 years (mean of all installations)(McCulloch et al., 2001).

Refrigerants have a more complex and variable pattern: initial losses are generally low (less than 1%), subsequently the emissions spread over the service life of the equipment can vary from zero, in the case of a hermetically sealed unit, to 20 or 30% per year where the equipment requires frequent servicing or relies heavily on mechanical seals to contain the refrigerant. Until the 1990s much of the refrigerant remaining in equipment on its disposal was released into the atmosphere. Recovery and re-use (or destruction) of this refrigerant is now much more commonplace and will be a legal requirement under EU legislation (CEC, 2006a). Historically, the statistical emission function for CFC and HCFC refrigerants, averaged over all refrigeration and air conditioning, was equivalent to total release into the atmosphere within 20 years, with the largest annual releases occurring during the first 10 years (Gamlen et al., 1986). Atmospheric concentrations of CFCs and HCFCs were consistent with a global emissions inventory derived using emission functions of this sort and reported sales (Ashford et al., 2004a; 2004b; McCulloch et al., 2003). However, more recent work has shown a trend towards lower emission rates for HCFC refrigerant due to more widespread use of hermetic systems and improvements to engineering to reduce both leakage and the charge size in commercial equipment (McCulloch et al., 2006).

These emission patterns mean that a "bank" of chemical accumulates in equipment, with a total mass equal to the cumulative sales minus cumulative emissions. If these data are available it is relatively simple to calculate the fraction of the bank that is released in any given year.

Leakage rates from refrigeration equipment are generally quoted as the engineering rate, that is the mass rate of leakage divided by the mass of fluid originally charged. For a single refrigeration system, this would result in a bank that is depleted in time and eventually reaches zero when the whole charge has been released. If this constant rate of mass leakage were expressed as a function of the diminishing bank, the values would increase as time progressed (due to the decreasing size of the bank). In the case where there is a steady demand for new systems, the apparent release rate from the bank would increase to a constant value consistent with total loss of material from the oldest systems. This is shown as the solid line in Figure 1 which was constructed as an illustration with a constant addition to the mass of material in use (the bank) each year and a constant loss rate of 10% of the original charge. This stabilises after ten years to a constant loss of 20% of the bank.

Conversely, a rate of emission from the bank that remains constant throughout, actually implies a reducing absolute rate of emission from equipment. The horizontal line in Figure 1 corresponds to the case where the emission rate is not 10% of the original charge, but 10% of what is left in the equipment at any time. An original charge of 1000 grams, leaking at 100 grams/year shows an initial loss rate of 10% of the bank. After 5 years, the bank size would be 500 grams and a 10% loss rate from this would correspond to an absolute loss of only 50 grams/year. Since the absolute loss rate depends on the pressures in the system (which do not change materially while the system is operable), a reducing loss rate is highly unlikely for a single system. However the condition for a constant rate of emission from the bank and a compounding reduction in absolute emission rates could be met if containment over the whole cohort of new systems were to improve year by year.

An emission rate relative to the bank that is falling implies an even greater reduction with time of the absolute rate of leakage and Figure 1 shows the case where losses are reducing by 5%/year. The picture stays largely unchanged if demand for new systems were to increase in time. Although the absolute extent of emissions increases proportionately to the increase in demand, these emissions remain the same relative to the bank.

4 REPORTING OF NATIONAL EMISSIONS AND CONSUMPTION

National data are submitted to UNFCCC under the Rio Convention and Kyoto Protocol using the ñcommon reporting formatò (CRF), comprising information on the consumption of each compound and its emissions from individual categories of end use (refrigeration and air conditioning, foam blowing, aerosol propulsion, solvent and fire extinguishing) (UNFCCC, 2008). The reporting requirements are covered by a comprehensive set of guidelines and guidance for emissions inventories provided by IPCC and recently updated (IPCC, 1997; Ashford et al., 2006). Critically, the Kyoto Protocol is aimed at relative reductions in greenhouse gas emissions[3] and not at establishing the absolute quantities emitted. As a consequence, the underlying principle for good practice guidance was that it should provide a range of appropriate methods to reflect accurately changes in greenhouse gas emissions relative to a baseline with sufficient flexibility to accommodate the circumstances of all parties. Because of this and because countries may leave out intermediate data on the grounds of commercial confidentiality, there is significant variation in the extent of coverage of the databases and in the rates of emission calculated from them. However, there is a fundamental requirement that the same methodology should be used throughout the years.

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[3] Expressed as a percentage of the reported emissions for the baseline year

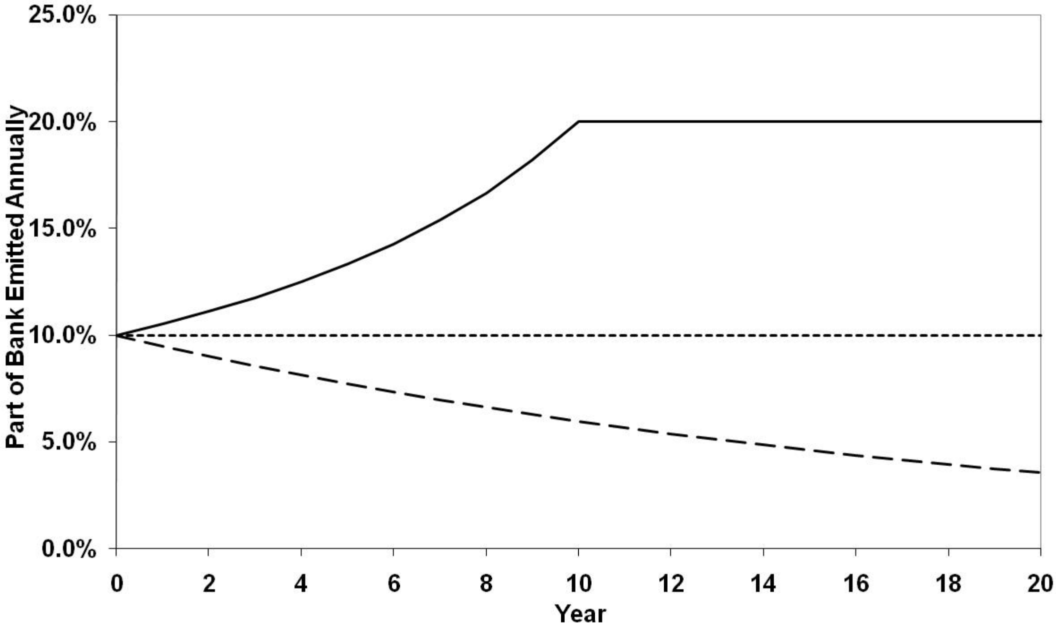


Figure 1. Illustration of the portion of the bank in equipment emitted annually for a case with constant annual consumption. Solid line: emission is 10%/year of the original charge. Dotted line: emission is 10% of the bank per yea. Dashed line: emission rate commences at 10%/year from the bank, subsequently falling at a compound rate of 5%/year.

Each national folder contains a separate file for each annual calculation of emissions from the base year to two years before present. The metadata contained in National Inventory Reports (UNFCCC, 2008) are intended to describe how the calculations in the CRF database have been performed and to authenticate the sources of the parameters used.

The submissions from all European Union countries, together with Norway and Switzerland, were examined. Belgium, France, Greece, Luxembourg and Spain report only emissions (accounting for 20 to 40% of the European total) and so could not be included in the calculation of refrigerant banks. However, because this study was conducted country-by-country, their omission should not affect conclusions. As noted below, consumption data for some of the HFCs in some of the countries was either missing or clearly wrong (for example, it is impossible to have cumulative emissions that are larger than the cumulative consumption). In these cases, the country's data had to be excluded. These exclusions and omissions, coupled with the high variability between national data, mean that a single meaningful European data set cannot be derived.

All data for UK and part of the submission from Austria, Finland and the Netherlands were reported as the total annual quantities of HFCs, expressed as their CO_2 equivalents. These aggregated data were apportioned into individual compounds using the European average annual distributions from the EFCTC database (EFCTC, 2007).

5 RESULTS

Banks were calculated for each year for each of HFCs, 125, 134a, 143a and 32, for each country by subtracting the cumulative emissions from the cumulative sales. All data were obtained from the national submissions under the common reporting format. From this database, the apparent emission function for each year was calculated by dividing the annual emission by the bank accumulated up to that year.

5.1 *Variation between Countries*

Judging from the apparent portions of the banks that are emitted each year, national results fall into two general categories:

- countries where this is falling in the long term, indicating a substantial improvement in containment, and
- countries where the apparent percentage is remaining constant, indicating that containment is at least remaining constant, if not improving at a somewhat lower rate than the first group.

Figures 2a to 2d show the proportion of the bank emitted by countries in the first group that show substantial improvements in containment. These comprise 50% of European emissions of HFC-125, 43% of HFC-134a emissions, 56% of HFC-143a emissions and 23% of emissions of HFC-32. The improvements are not uniform across countries and compounds but improvements have been shown for at least three of the fluorocarbons by Germany, Hungary, Poland, Slovakia, Sweden and United Kingdom.

Figures 3a to 3d show the countries of the second group, where improvement in containment is, for many countries, consistent with a constant rate of loss from the bank (implying improving absolute rates of emission). The principle exception to this is Denmark, where emissions appear to be growing with respect to the banks. The countries in this group account for 15% of HFC-125 emissions, 11% of HFC-134a, 12% of HFC-143a and 8% of HFC-32.

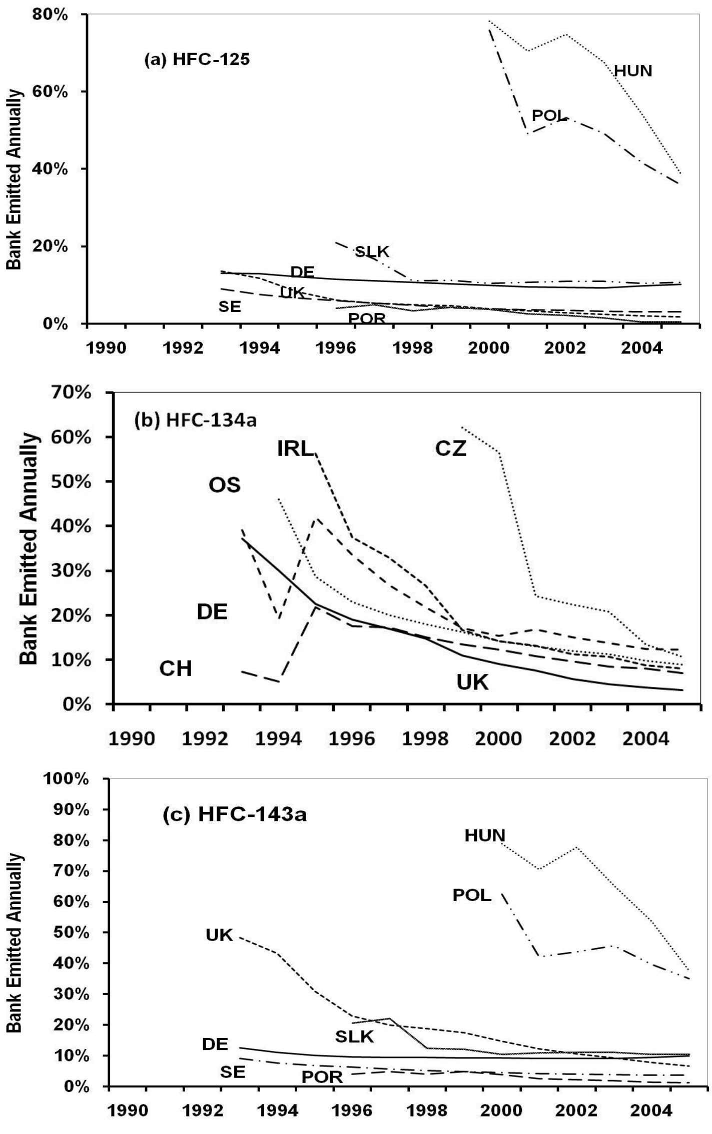
Countries that did not report consumption, so that it was not possible to calculate national banks (principally Belgium, France, Greece and Spain) or that reported unreliable data (for example Italy, where consumption was less than reported emissions) accounted for 35% of the emissions of HFC-125, 46% of HFC-134a, 32% of HFC-143a and 68% of HFC-32.

The very large changes that occur during the early years of consumption, shown particularly in those countries that have improving containment, are associated with small absolute emissions and small bank sizes. Furthermore, consumption did not commence at the same time in all countries.

5.2 *Variation between fluorocarbons*

For HFC-125, while there are substantial differences between the emission rates calculated for each country, most are now below 10%/year, implying a lifetime for the bank greater than 10 years, and some nations with significant emissions (for example, the Netherlands, Sweden and UK), have emissions low enough to sustain bank lifetimes of more than 20 years.

In the case of HFC-134a, the 2005 emission rates show a large range but the weighted mean is less than 10%, indicating a lifetime for the bank in excess of 10 years[4] and HFC-143a shows a similarly low rate of emission from the banks.
For HFC-32, the coverage of national data is heavily influenced by the lack of information on consumption from Belgium, France, Greece and Luxembourg (totalling 16% of emissions) and the questionable data from Italy which, although reporting 52% of European emissions, reports insufficient consumption to sustain these. The average rate of the accounted emissions is between 5 and 10%, giving a containment lifetime of 10 to 20 years.



[4] This is all the more remarkable since this average includes emissions from automotive airconditioning systems which were believed to have substantial emission rates (CEC, 2006b)

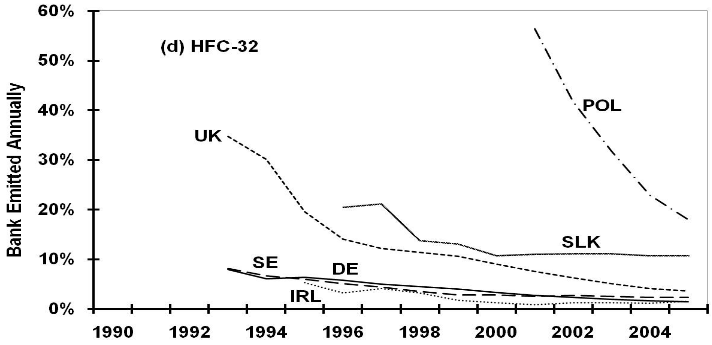
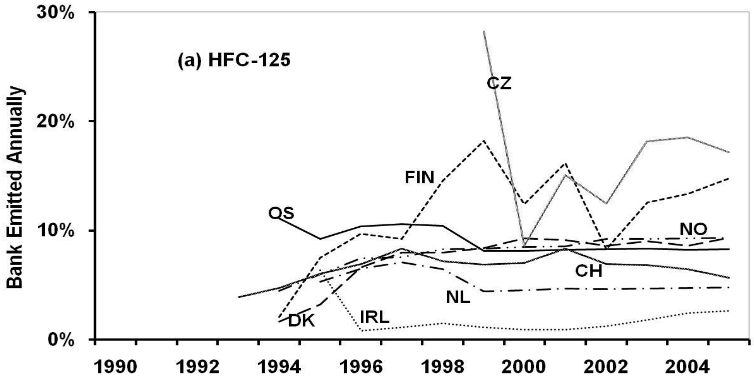
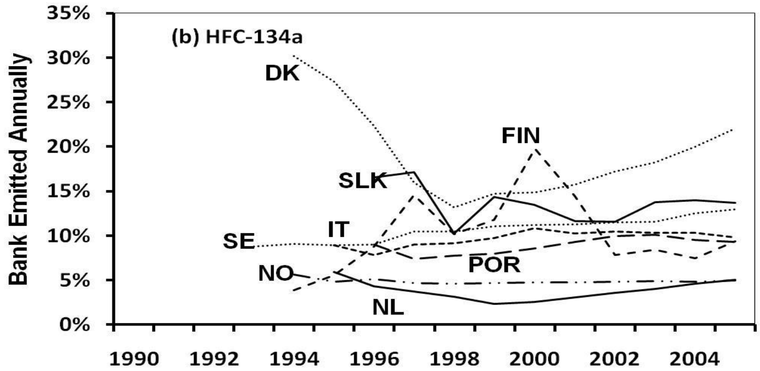
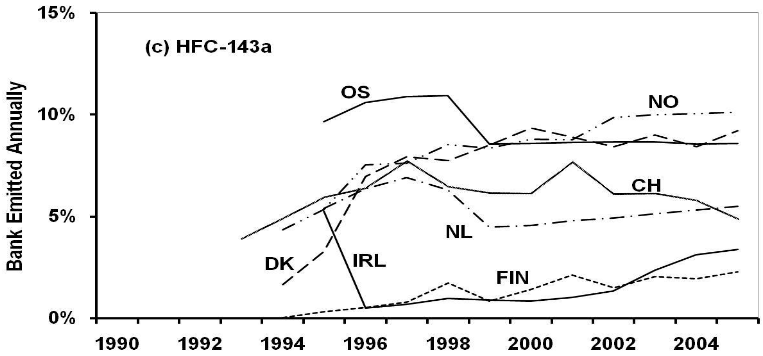


Figure 2 (a) to (d). Proportion of the banks of HFCs emitted by Austria (OS), Czech Republic (CZ), Germany (DE), Hungary (HUN), Ireland (IRL), Poland (POL), Portugal (POR), Slovakia (SLK), Sweden (SE), Switzerland (CH) and UK.







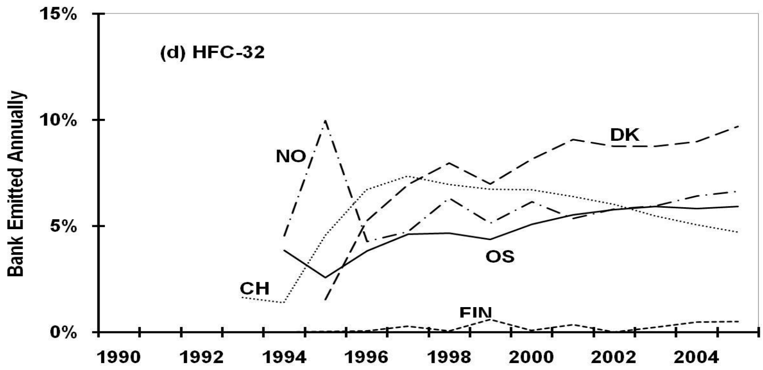


Figure 3 (a) to (d) Proportion of the banks of HFCs emitted by Austria (OS), Czech Republic (CZ), Denmark (DK), Finland (FIN), Ireland (IRL), Italy (IT), Netherlands (NL), Norway (NO), Portugal (POR), Slovakia (SLK), Sweden (SE) and Switzerland (CH).

6 CONCLUSION

From the data provided by countries, it is clear that containment of HFCs in refrigeration and air conditioning applications, has improved. In many cases, the high rates associated with low demand volume early in the product lifetimes have shrunk to between 5 and 10% of the bank . There is no evidence to suggest that this conclusion would change if the lack of consumption data for countries such as France and Spain and the poor quality of consumption data for Italy and Poland were rectified. Emissions from these countries were consistent with those from similar European nations.

It is to be expected that the end uses for individual compounds will be similar across the EU.

However, the discrepancy between individual countriesôcalculations of the emission functions is at least a factor of 2 and can be as high as 10. Although this is not a major problem when the data are used to monitor adherence to the Kyoto Protocol, the differences mean that a European database, calculated by simply adding together the submitted emission data, has no scientific basis, lacks consistency and therefore could be misleading.

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