TECHNICAL SUPPORT DOCUMENT FOR EMISSIONS OF HFC-23 FROM PRODUCTION OF HCFC-22:

PROPOSED RULE FOR MANDATORY REPORTING OF GREENHOUSE GASES

Office of Air and Radiation

U.S. Environmental Protection Agency

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Contents

1. Source Description

Trifluoromethane (HFC-23 or CHF_3) is generated as a by-product during the manufacture of chlorodifluoromethane (HCFC-22), which is primarily employed in refrigeration and air conditioning systems and as a chemical feedstock for manufacturing synthetic polymers. Because HCFC-22 depletes stratospheric ozone, its production for non-feedstock uses is scheduled to be phased out by 2020 under the U.S. Clean Air Act.[1] Feedstock production, however, is permitted to continue indefinitely.

HCFC-22 is produced by the reaction of chloroform ($CHCl_3$) and hydrogen fluoride (HF) in the presence of a catalyst, $SbCl_5$. The reaction of the catalyst and HF produces $SbCl_xF_y$, (where $x + y = 5$), which reacts with chlorinated hydrocarbons to replace chlorine atoms with fluorine. The HF and chloroform are introduced by submerged piping into a continuous-flow reactor that contains the catalyst in a hydrocarbon mixture of chloroform and partially fluorinated intermediates. The vapors leaving the reactor contain HCFC-21 ($CHCl_2F$), HCFC-22 ($CHClF_2$), HFC-23 (CHF_3), HCl, chloroform, and HF. The under-fluorinated intermediates (HCFC-21) and chloroform are then condensed and returned to the reactor, along with residual catalyst, to undergo further fluorination. The final vapors leaving the condenser are primarily HCFC-22, HFC-23, HCl and residual HF. The HCl is recovered as a useful byproduct, and the HF is removed. Once separated from the HCFC-22, the HFC-23 may be vented to the atmosphere as an unwanted by-product, captured for use in a limited number of applications, or destroyed.

HFC-23 is also emitted from thermal oxidizers at HFC-23 destruction facilities, which may or may not also be HCFC-22 production facilities.

a. Total U.S. Emissions

Emissions of HFC-23 in 2006 were estimated to be 13.8 million metric tons of CO_2 equivalent ($MMTCO_2e$). This quantity represents a 13 percent decline from 2005 emissions and a 62 percent decline from 1990 emissions despite an 11 percent increase in HCFC-22 production since 1990. Both declines are primarily due to decreases in the HFC-23 emission rate. The ratio of HFC-23 emissions to HCFC-22 production has decreased from 0.022 to 0.0077 since 1990, a reduction of 66 percent. These decreases are primarily attributable to four factors: (a) five plants that did not capture and destroy the HFC-23 generated have ceased production of HCFC-22 since 1990, (b) one plant that captures and destroys the HFC-23 generated began to produce HCFC-22, (c) one plant implemented and documented a process change that reduced the amount of HFC-23 generated, and (d) the same plant began recovering HFC-23, primarily for destruction and secondarily for sale Three HCFC-22 production plants operated in the United States in 2006, two of which used thermal oxidation to significantly lower their HFC-23 emissions. All three plants are part of a voluntary agreement to report and reduce their collective HFC-23 emissions.

2. Options for Reporting Threshold

EPA evaluated a range of threshold options for HCFC-22 production facilities. These included emission-based thresholds of 1,000, 10,000, 25,000 and 100,000 $mtCO_2e$ and capacity-based thresholds equivalent to these. EPA also evaluated a requirement that all HCFC-22 production facilities be required to report.

The capacity-based thresholds are shown in Table 1, and are based on full utilization of HCFC-22 capacity and the emission rate given for older plants (i.e., 0.04 kg HFC-23/kg HCFC-22) in the 2006 IPCC Guidelines. One plant is relatively new, but the emission rate for older plants was used to be consistent and somewhat conservative. Due to the unavailability of facility-specific emissions information, the emission-based thresholds are not presented in a table. (Under the voluntary emission reduction agreement, total emissions from the three facilities are aggregated by a third party, which submits only the total to EPA.)

[1] As construed, interpreted, and applied in the terms and conditions of the *Montreal Protocol on Substances that Deplete the Ozone Layer*. [42 U.S.C. §7671m(b), CAA §614]

Table 1. Capacity-Based Thresholds

Threshold Level (HCFC-22 capacity in tons)	Total National Emissions ($mtCO_2e$)	Total National Facilities	Emissions Covered		Facilities Covered	
			$mtCO_2e/yr$	Percent	Facilities	Percent
2	13,848,483	3	13,848,483	100	3	100
21	13,848,483	3	13,848,483	100	3	100
53	13,848,483	3	13,848,483	100	3	100
214	13,848,483	3	13,848,483	100	3	100

The analysis found that all of the plants, which have capacities ranging from 18,000 to 100,000 metric tons of HCFC-22, exceed the capacity threshold equivalent to 100,000 metric tons CO_2 Eq. by a factor of 85 or more. Two of the three plants currently operating in the United States destroy most or all of their HFC-23 by-product using thermal oxidation, which probably lowers their emissions below at least some of the emission-based thresholds discussed above. However, if the thermal oxidizers malfunctioned, were not operated properly, or were unused for some other reason, emissions of HFC-23 from each of the plants could easily exceed all thresholds. Reporting is therefore important both for tracking the considerable emissions of plants that do not use thermal oxidation and for verifying the performance of thermal oxidation where it is used. This rationale supports either a capacity-based threshold or a requirement that all plants report. A requirement that all plants report would have the additional advantage of simplicity.

EPA is aware of one facility that destroys HFC-23 but does not produce HCFC-22. Although EPA does not know the precise quantity of HFC-23 destroyed by this facility, the Agency believes that the facility destroys a substantial share of the HFC-23 generated by the largest HCFC-22 production facility in the United States. If the destruction facility destroys even five percent of this HFC-23, it is likely to destroy considerably more than 100,000 $mtCO_2e$.

3. Options for Monitoring Methods

a. Review of Existing Reporting Programs and Methodologies

EPA has reviewed a range of protocols for estimating HFC-23 emissions from HCFC-22 production. These include the *2006 IPCC Guidelines*, guidance developed under EPA's voluntary program for HCFC-22 manufacturers, the World Resources Institute/World Business Council on Sustainable Development, the Toxic Release Inventory (TRI), the TSCA Inventory Update Rule, The DOE 1605(b) Voluntary Reporting Program, EPA Climate Leaders, and The Climate Registry.

EPA also considered the findings and conclusions of a recent report that closely reviewed the methods that facilities use to estimate and assure the quality of their estimates of HCFC-22 production and HFC-23 emissions (RTI, 2008). As noted above, the production facilities currently estimate and report these quantities to EPA (across all three plants) under a voluntary agreement. The report is entitled "Verification of Emission Estimates of HFC-23 from the Production of HCFC-22: Emissions from 1990 through 2006" and is available in the docket for this rulemaking.

The 2008 Verification Report (RTI, 2008) found that the estimation methods used by the three HCFC-22 facilities currently operating in the United States were all equivalent to IPCC Tier 3 methods. Under the Tier 3 methodology, facility -specific emissions are estimated based on direct measurement of the HFC-23 concentration and the flow rate of the streams, accounting for the use of emissions abatement devices (thermal oxidizers) where they are used. In general, Tier 3 methods for this source category yield far more accurate estimates than Tier 2 or Tier 1 methods. Even at the Tier 3 level, however, the emissions estimation methods used by the three facilities differed significantly in their levels of absolute uncertainty. The uncertainty of the one facility that does not thermally destroy its HFC-23 emissions dominates the uncertainty for the national emissions from this source category.

Because the methods used by all U.S. facilities are at the Tier 3 level, this document does not discuss the Tier 1 and Tier 2 methods in detail. Instead, it focuses on methods very similar to those actually used by U.S. facilities to estimate HFC-23 emissions. These vary primarily depending on the process architecture of each plant. The methods include two monitoring methods for HCFC-22 production facilities and one for HFC-23 destruction facilities. All the monitoring methods rely on measurements of HFC-23 concentrations in process or emission streams and on measurements of the flow rates of those streams, although the frequency of these measurements varies.

In general, the methods discussed below are very similar to the procedures already being undertaken by the facilities to estimate HFC-23 emissions and to assure the quality of these estimates. The differences (and the rationale for them) are discussed further below in Section b, "Comparison of methods to current practice."

Plants that do not use a thermal oxidizer connected to the HCFC-22 production equipment could be required to:

i) Monitor the concentration of HFC-23 in the product stream containing the HFC-23 (either HCFC-22 or HCl) on at least a daily basis, using equipment and methods with an accuracy of 5% or better.

ii) Monitor the mass flow rate of the product stream containing the HFC-23 either directly (using a flow meter with an error no greater than 1%) or by weighing the other product (e.g., HCFC-22, using scales or load cells with an error no greater than 1%). Plants would be required to make or sum these measurements on at least a daily basis. If the product were measured significantly downstream of the reactor (e.g., at the shipping dock and/or storage tanks), plants would be required to add 1.5% (or another value that can be demonstrated) to the measurement to account for losses between the reactor and the point of measurement.

iii) Estimate the quantity of HFC-23 produced by one of the two calculations below.

1. Where the mass flow of the combined stream of HFC-23 and the other product (e.g., HCl) is measured, multiply the daily (or more frequent) HFC-23 concentration measurement (which may be the average of more frequent concentration measurements) by the daily (or more frequent) mass flow of the combined stream of HFC-23 and the other product. This calculation is summarized in the equation below. To estimate annual HFC-23 production, sum the daily (or more frequent) estimates of the quantities of HFC-23 produced over the year.

$$G_{23} = \sum_{p=1}^{n} c_{23} * F_p * 10^{-3}$$

where:

G_{23}	=	mass of HFC-23 generated annually (metric tons)
c_{23}	=	fraction HFC-23 by weight in HFC-23/other product stream
F	=	mass flow of HFC-23/other product stream during the period p (kg), and
p	=	period over which mass flows and concentrations are measured
n	=	Number of concentration and flow measurements for the year
1×10^{-3}	=	Conversion factor from kilograms to metric tons

2. Where the production of only the other product (e.g., HCFC-22) is measured, multiply the ratio of the daily (or more frequent) measurement of the HFC-23 concentration and the daily (or more frequent) measurement of the other product concentration by the daily mass produced of the other product. This calculation is summarized in the equation below. To estimate annual HFC-23 production, sum the daily (or more frequent) estimates of the quantities of HFC-23 produced over the year.

$$G_{23} = \sum_{p=1}^{n} \left(\frac{c_{23}}{c_{22}} \right) * P_{22} * 10^{-3}$$

where:

G_{23} = mass of HFC-23 generated annually (metric tons)

c_{23} = fraction HFC-23 by weight in HCFC-22/HFC-23 stream

c_{22} = fraction HCFC-22 by weight in HCFC-22/HFC-23 stream

P_{22} = mass of HCFC-22 produced over the period p (kg), and

p = period over which masses and concentrations are measured

n = Number of concentration and mass measurements for the year

1 x 10-3 = Conversion factor from kilograms to metric tons

The mass of HCFC-22 produced over the period p shall be estimated by using the equation below:

$$P_{22} = LF * (O_{22} - U_{22})$$

where:

P_{22} = mass of HCFC-22 produced over the period p (kg)

O_{22} = mass of HCFC-22 that is measured coming out of the production process over the period p (kg)

U_{22} = mass of used HCFC-22 that is added to the production process upstream of the output measurement over the period p (kg)

LF = factor to account for the loss of HCFC-22 upstream of the measurement.

iv) Measure the quantities of HFC-23 sold or sent to other facilities for destruction using flowmeters or scales with an accuracy of 1% of full scale or better. This step will ensure that any fugitive losses of HFC-23 during transport, unloading, or filling of containers were included in the HFC-23 emission estimates for plants that capture HFC-23 for use as a product or for transfer to a thermal oxidation unit for destruction. HFC-23 emitted would be estimated by the following equation:

$$E_{23} = G_{23} - (S_{23} + OD_{23})$$

where:

E_{23} = mass of HFC-23 emitted annually (metric tons)

G_{23} = mass of HFC-23 generated annually (metric tons)

S_{23} = mass of HFC-23 packaged for sale annually (metric tons)

OD_{23} = mass of HFC-23 sent off-site for destruction (metric tons)

This calculation assumes that all production that is not sold or destroyed is emitted. Such fugitive emissions may be the result of the packaging and shipping process; additional emissive losses can be attributed to the number of flanges in a line, and other on-site equipment which is specific to each facility.

Plants using a thermal oxidizer connected to the HCFC-22 production equipment could be required to estimate total HFC-23 emissions as the sum of HFC-23 emissions from equipment 1) leaks upstream of the oxidizer, 2) process vents, and 3) the thermal oxidizer. Emissions from equipment leaks would be estimated using EPA Method 21 and data on the concentration of HFC-23 in the HFC-23/HCFC-22 process stream that is representative of current operating conditions (in terms of capacity utilization, process optimization). Emissions from process vents would be

estimated using the results of annual emissions tests at process vents, adjusting for changes in HCFC-22 production rates since the measurements occurred. Tests would have to be conducted in accordance with EPA Method 18. Finally, emissions from the thermal oxidizer would be estimated by applying the destruction efficiency (DE) of the oxidizer to the mass of HFC-23 fed into the oxidizer.

HFC-23 destruction facilities could be required to:

i. Measure the quantities of HFC-23 fed into the oxidizer using flowmeters or scales with an accuracy of 1% of full scale or better.
ii. Account for any decreases in the destruction efficiency of the oxidizer that occurred when the oxidizer was not operating properly (as defined in state or local permitting requirements and/or oxidizer manufacturer specifications).
iii. Perform annual HFC-23 concentration measurements by gas chromatography to confirm that emissions from the oxidizer are as low as expected based on the rated DE of the device. If emissions are found to be higher, then facilities would have the option of using the DE implied by the most recent measurements or of conducting more extensive measurements of the DE of the device.
iv. The total mass of fluorinated GHGs destroyed would be estimated using the following equation:

$$D = F_D * DE$$

where:

D	=	mass of HFC-23 destroyed annually (metric tons)
F_D	=	mass of HFC-23 fed into the destruction device annually (metric tons)
DE	=	Destruction Efficiency of the destruction device (fraction)

v. The total mass of fluorinated GHGs emitted from destruction devices would be estimated by using the equation below:

$$E_D = F_D - D$$

where:

E_D	=	mass of fluorinated GHG emitted annually (metric tons)
F_D	=	mass of fluorinated GHG fed into the destruction device annually (metric tons)
D	=	mass of HFC-23 destroyed annually (metric tons)

Because emissions that occur as a result of startups, shutdowns, and malfunctions can be both significant and atypical, an accurate accounting of facility emissions must address HFC-23 production and emissions that occur as a result of startups, shutdowns, and malfunctions. To ensure that these emissions are accounted for, facilities could either record HFC-23 production and emissions during these events, or could document that these events do not result in significant HFC-23 production and/or emissions.

b. Comparison of methods to current practice

In general, the high GWP of HFC-23 makes it worthwhile to require accurate and precise measurements and estimates for this source category. The methods discussed above are very similar to the procedures already being undertaken by the facilities to estimate HFC-23 emissions and to assure the quality of these estimates. There are, however, six areas in which these methods go beyond current practice at one or more of the plants:

1. Factor to compensate for fugitive losses of product. As discussed above, facilities that use product (HCFC-22 or HCl) masses or mass flow rates to estimate HFC-23 generation and that measure those masses or mass flow rates significantly downstream of the reactor could be required to add 1.5% to their mass or mass flow rate measurements. This adjustment would account for upstream product losses, which are estimated to range from one to two percent. Without the adjustment, HCFC-22 production and therefore HFC-23 generation at

affected facilities would be systematically underestimated (negatively biased). A one- to two-percent underestimate could translate into an underestimate of HFC-23 emissions of 100,000 mtCO2e or more for each affected facility. In general, the 2006 IPCC Guidelines recommend correcting for biases wherever possible (Volume 1, p 3.22).

EPA's understanding is that one facility measures the product mass significantly downstream of the reactor. This facility confirms that, because of small losses due to fugitive emissions, the amount of HCFC-22 that is recovered and weighed (and used to estimate emissions) is on the order of 98 to 99 percent of the generated HCFC-22 (RTI, 2008). The 1.5% factor is the midpoint of the one-to-two-percent range of product loss rates cited by the affected facility. If the facility could demonstrate a lower fugitive loss rate of HCFC-22 (e.g., through a mass-balance approach using measurements of sufficient precision), a lower loss rate factor (e.g., 1%) may be appropriate.

2. Daily measurements. As discussed above, facilities not using an thermal oxidizer connected to the HCFC-22 production equipment could be required to measure HFC-23 concentrations and product flow rates on at least a daily basis. This requirement would account for day-to-day fluctuations in the rate at which HFC-23 is generated; this rate can vary depending on process conditions. While one affected facility measures these quantities several times an hour, the other measures them twice per week. Thus, the second facility would have to increase the frequency of its measurements.

3. Calibration of gas chromatographs using standards with representative concentrations. As discussed above, HCFC-22 production facilities could be required to calibrate gas chromatographs used to determine the concentration of HFC-23 by analyzing certified standards with known HFC-23 concentrations that are in the same range (percent levels) as the process samples. This requirement would verify the accuracy of gas chromatographs at the concentrations of interest; calibration at other concentrations does not verify this accuracy with the same level of assurance. This requirement would be similar to requirements in protocols for the use of gas chromatography, such as EPA Method 18. While one affected plant currently calibrates its gas chromatograph in this fashion, the other does not. Thus, the second plant would have to change its calibration practices.

4. Accounting for emissions during startups, shutdowns, and malfunctions. As discussed above, facilities could be required to account for HFC-23 production and emissions that occur as a result of startups, shutdowns, and malfunctions, either recording HFC-23 production and emissions during these events or documenting that these events do not result in significant HFC-23 production and/or emissions. Depending on the circumstances, startups, shutdowns, and malfunctions (including both the process equipment and any thermal oxidation equipment) can be significant sources of emissions, and therefore an accurate accounting of plant emissions should address them. This requirement would lead to a change in the practice of one plant.

5. Annual check of thermal oxidizer. As discussed above, facilities using thermal oxidizers could be required to perform annual HFC-23 concentration measurements by gas chromatography to confirm that emissions from the oxidizer are as low as expected based on the rated DE of the device. Although the initial testing and parametric monitoring that facilities currently perform on their oxidizers provides general assurance that the oxidizer is performing correctly, the requirement would provide additional assurance at relatively low cost (e.g., approximately two hours of technician time per year to sample and analyze the vent gases). Even a one- or two-percent decline in the destruction efficiency of the oxidizer could lead to emissions of over 100,000 $mtCO_2e$, making this a particularly important factor to monitor accurately. This requirement would lead to a change in the practice of at least one plant.

6. Annual monitoring of emissions from process vents. As discussed above, HCFC-22 production facilities that use a thermal oxidizer connected to the HCFC-22 production process could be required to conduct annual monitoring of HFC-23 emissions from process vents. Although HFC-23 emissions from process vents are believed to be quite low, this monitoring would ensure that any year-to-year variability in the emission rate was captured by the reporting. Like the annual check on the thermal oxidizer, this requirement would cost relatively little. The requirement would lead to a change in the practice of one plant.

One additional consideration is the accuracy and precision of the flowmeters and scales in use at HCFC-22 production facilities. The accuracy and precision of these flowmeters and scales currently ranges from ±0.15 percent to ±1 percent. EPA's understanding is that scales and flowmeters with good accuracy and precision (e.g., ±0.2 percent or better) have an installed cost of approximately $20,000 per flowmeter. Given the large quantities of

HFC-23 generated by each facility, it may be appropriate to require facilities to consistently use such high-accuracy devices to measure the flows used to estimate HFC-23 emissions.

4. Procedures for Estimating Missing Data

In cases when an upstream flow meter (i.e., at reactor outlet) is ordinarily used but is not available for some period, the plant could compensate by using downstream production measures (e.g., quantity shipped) and adding 1.5%. If HFC-23 concentration measurements are unavailable for some period, the facility could use the average of the concentration measurements from just before and just after the period of missing data.

It may be appropriate to permit an exception to these requirements if either method would result in a significant under- or overestimate of the missing parameter (e.g., because the monitoring failure was linked to a process disturbance that is likely to have significantly increased the HFC-23 generation rate). In this case, the facility could be required to develop an alternative estimate of the parameter and explain why and how it developed that estimate.

5. QA/QC Requirements

Typical QA/QC requirements for measuring devices include initial and periodic verification and calibration. (For example, see the requirements of EPA's Acid Rain regulations at 40 CFR Part 75.) In this case, it would be appropriate to require an initial verification of flowmeters and weigh scales and periodic calibration in accordance with the applicable industry standards. Calibration of flowmeters and scales could be performed prior to the reporting year; after the initial calibration, recalibration could be performed at least annually or more frequent if specified by the manufacturer. Under this approach, producers could perform the verification and calibration of their weigh scales during routine product line maintenance.

For the gas chromatography analytical method described under the monitoring section of this document, monthly calibration using known certified standards would be appropriate to counter instrument drift. The concentrations of the standards should be in the same range (percent levels) as the process samples to ensure accurate measurements at the concentrations of interest (RTI, 2008).

A relatively inexpensive but effective QA/QC requirement for thermal oxidizers would be to verify their performance by conducting annual HFC-23 concentration measurements by gas chromatography to confirm that emissions from the oxidizer are as low as expected based on the rated DE of the device (RTI, 2008). To be representative, such measurements should be performed under conditions that are normal for the production process and oxidizer at the facility.

6. Reporting Procedures

For HCFC-22 production facilities, reporting of the following data would be useful for confirming emissions calculations and/or calculating emission rates that could be compared across facilities and over time for data quality control purposes:

- HCFC-22 production in tons over the year;
- Consumption of reactants (including factors to account for quantities that typically remain unreacted) in tons, by reactant, over the year;
- The mass of materials other than HCFC-22 and HFC-23 (i.e., unreacted reactants, HCl and other byproducts) that are permanently removed from the process over the year; and
- Method for tracking startups, shutdowns, and malfunctions and HFC-23 generation/emissions during these events.

For HCFC-22 production facilities that estimated missing data, the following information would be important for ensuring data quality:

- The reason the data were missing, length of time the data were missing, method used to estimate the missing data, and estimates of those data; and

- If missing data was estimated by a method other than one of those specified, the reason why the specified method would lead to a significant under- or overestimate of the parameter (s) and the rational for the methods used to estimate the missing data.

For HFC-23 destruction facilities, reporting of the following data would be useful for ensuring data quality:

- Results of their annual HFC-23 concentration measurements at the outlet of the oxidizer.

- One-time report similar to that required under EPA's stratospheric protection regulations at 40 CFR 82.13(j):
    1. Destruction unit's DE
        a. DEs may be calculated based on measured feed rates and stack gas emission rates occurring during performance tests under controlled conditions using representative compounds. Facilities can determine that they are achieving the applicable DE by determining that the units are being operated within the permitted range of operating parameters, which is developed based on conditions of the performance tests.

           Therefore, the DE recorded during the performance test can be assumed to apply when the actual supply of HFC-23 is destroyed, provided that the unit was being operated within the permitted range of operating parameters (e.g., waste feed rate, combustion temperature).

    2. Methods used to record volume destroyed and to determine DE
    3. Names of other relevant federal or state regulation that may apply to destruction process
    4. Type of destruction technology used.

- Revision to the one-time report submitted within 60 days of any changes to the unit's destruction efficiency or methods used to record volume destroyed.

These data are useful either because they are necessary to verify facilities' calculations of HFC-23 generation, emissions, or destruction or because the data allow EPA to implement other quality assurance checks (e.g., calculation of an HFC-23/HCFC-22 generation factor that can be compared across facilities and over time).

For HCFC-22 producers, the following records would be important for verifying and documenting emissions monitoring and calculations:

- Data actually used to estimate emissions

- The initial and any subsequent calibrations of the gas chromatographs, scales, and flowmeters used to measure the quantities reported.

For HFC-23 destruction facilities, the following records would be important for verifying and documenting destruction and emissions estimates:

- Information documenting their one-time and annual reports.

7. References

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Coriolis flow meters:

Micro Motion: www.emersonprocess.com/MicroMotion/

Siemens: https://pia.khe.siemens.com/index7625.htm

Weigh Scales / Weigh Cells

Mettler Toledo: www.mt.com or

http://us.mt.com/mt/filters/products-applications_industrial-weighing/

American Weigh Scales: www.americanweigh.com