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Research Note

Source Terms for Routine UF_6 Emissions

R. Scott Kemp

Program on Science and Global Security, Princeton University, Princeton, New Jersey, USA

INTRODUCTION

Models for the detection of gas-centrifuge facilities and conversion plants by their emission of uranium-hexafluoride to the atmosphere rely critically on a presumed source term.[1]

To date, no experimentally derived source terms have been published for routine releases from either gas-centrifuge or conversion plants. The most widely cited source term for conversion plants is based on a dose reconstruction estimate done for 1960s-era releases from building K-1131 at the U.S. Atomic Energy Agency's K-25 site. However, that facility released tens of kilograms per month and is not representative of modern-day conversion plants.[2]

This research note gives measured source terms for UF_6 emissions for both commercial centrifuge plants and commercial conversion facilities. Emission for conversion plants are given in Table 1. These are normalized on a gram uranium released per tonne uranium in product basis. Data are given for three conversion facilities along with statistical confidence intervals.

Emissions for centrifuge plants were normalized on a gram released per tonne-SWU (separative work unit) basis. Data from three plants at one site (Capenhurst, UK) are given in Tables 2 and 3. Although a statistical confidence interval is given, the reader is cautioned that generalizing from one site carries with it high systematic error. Much of the Capenhurst facility employs

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Address correspondence to R. Scott Kemp, Program on Science and Global Security, Princeton University, 225 Nassau Street, 2nd Floor, Princeton, NJ 08542. E-mail: rskemp@Princeton.edu

Table 1: Summary release statistics for UF_6 conversion plants.

Facility Name	Median release rate (grams/tonne)	Observations*	95% confidence interval (grams/tonne)
Honeywell Metropolis	16.45	7	13.43–19.47
Cameco Port Hope	3.65	20	3.03–4.27
Comurhex Pierrelatte[1]	0.11	5	0.06–0.17

[1]Comurhex Pierrelatte only converts UF_4 to UF_6, whereas Honeywell Metropolis and Cameco Port Hope also produce UF_4 from concentrated uranium oxides.

basic negative-pressure controls and HEPA filtration to remove significant amounts of the emissions from the air before vented to the atmosphere. Furthermore, the emissions rate depends heavily on the type of centrifuge, with high-performance centrifuges emitting far less. A detailed description of centrifuge-plant estimates is given in the discussion below.

RECOMMENDED SOURCE TERMS FOR COMMERCIAL FACILITIES DISCUSSION OF DATA SOURCES FOR CONVERSION PLANTS

Honeywell Metropolis

Data for Honeywell's Metropolis plant are based on semi-annual measures of uranium-activity released from the facility between 2000 and 2003, as measured by the operator and reported to the U.S. Nuclear Regulatory Authority.[3] Data were normalized using the nameplate capacity for the facility during each measurement period. The Honeywell facility is notoriously leaky and has been investigated by the U.S. Nuclear Regulatory Commission for excessive releases on several occasions. Such a badly operated facility is probably not representative of the state of the art, but might better represent a facility in a developing country.

The Honeywell facility receives raw yellowcake (mainly U_3O_8 and other oxides), which is first passed through a calciner to remove carbonates, water, and

Table 2: Summary release statistics for a gas-centrifuge enrichment plant.

Facility name	Median release rate (grams/tSWU)	Observations*	95% confidence interval (grams/tSWU)
Urenco Capenhurst Site	0.015	51	0.011–0.019

*See text for a description of the observational periods.

Table 3: Detailed Emissions for Facilities at Capenhurst

Plant name	Median release rate (grams/tSWU)	Observations	95% confidence interval (grams/tSWU)
E22 cascade operations	0.021	51	0.016–0.027
A3 cascade operations	0.0081	34	0.0066–0.0097
E23 cascade operations	0.00072	33	0.00037–0.00110
E23 auxiliary facilities	0.00017	12	0.00014–0.00019

other volatile materials. The calcined material is blended and agglomerated in equipment specially designed by Honeywell to obtain an optimum particle size for fluid operations. The agglomerates are screened and classified. The prepared feed is sent to a fluidized-bed reactor where it is contacted at elevated temperature with hydrogen. Certain impurities are reacted and evolved out of the system in the waste gas stream which is filtered, incinerated, and discharged without further treatment. The reduced concentrated uranium dioxide (UO_2) is sent directly to the hydro-fluorination fluidized-bed reactors where reaction with anhydrous hydrofluoric acid (HF) produces uranium tetrafluoride (UF_4). Again, certain impurities are removed in the form of gases which are filtered and scrubbed prior to venting to the atmosphere. The UF_4 is contacted with elemental fluorine in the fluidized-bed fluorinators. Most of the metallic impurities remain as the UF_6 is volatilized. As the UF_6 leaves the fluorination step, it is filtered to separate elutriated particulate material and is then desublimed in heat exchangers. The heat exchangers are periodically cycled by heating the UF_6 under pressure to its melting point whereupon it is sent to the distillation unit. After distillation, the high purity UF_6 liquid is drained into 14-ton product cylinders.[4]

Cameco Port Hope

Data for Cameco's Port Hope facility are based on time-averaged monthly release measurements made at the primary stack between 2005 and 2006, as measured by the operator and reported in its Quarterly Environmental Compliance Reports.[5] The Port Hope facility is an example of a well operating plant, despite a large accidental release in 2007.

The Port Hope facility receives tote-bins containing high-purity uranium from Camco's refinery at Blind River. Each contains 9.5 tonnes of uranium in the form of uranium tri-oxide (UO_3). The tri-oxide is pulverized into a fine powder and reacted with hydrogen gas to reduce the feedstock into uranium-dioxide (UO_2). After reduction, uranium-dioxide is reacted with anhydrous hydrofluoric acid (HF) to produce uranium tetrafluoride (UF_4), which is then calcined to remove water and volatile impurities. Calcined UF_4 is then contacted with elemental fluorine to produce UF_6 gas. The gas is filtered and desublimed in cold traps. The cold traps are periodically cycled to liquefy the UF_6, which is then used to fill 10- and 14-ton product cylinders.[6]

Comurhex Pierrelatte

Data for Comurhex's Pierrelatte facility are from annual environmental reports published by its parent corporation, AREVA.[7] The emissions data are for periods covering 2003 to 2007 at the *Installation Classée pour la Protection de l'Environnement* (ICPE) facility at Pierrelatte, which converts UF_4 into UF_6. Because the steps involved in purifying uranium oxide, the production of di- or tri-oxide and of UF_4 occur at other facilities, the emissions are much lower in comparison to Honeywell Metropolis and Cameco Port Hope. A detailed process description for the Comurhex ICPE plant was not available.

DISCUSSION OF DATA SOURCES FOR GAS-CENTRIFUGE ENRICHMENT PLANTS

URENCO Capenhurst Site Description

Unless otherwise noted, all information for Urenco's Capenhurst site are from a report produced by Urenco in response to an inquiry from Britain's Environment Agency, covering various periods spanning 1993 to 2005.[8]

Three centrifuge plants are operated on the Capenhurst site. The oldest, the E22 plant, dates to 1982 and contains one block of LEC centrifuges, with the remainder being 3LC centrifuges.[9] The second oldest, the A3 plant, was commissioned in 1985 as a British military facility, but was bought by Urenco for commercial use in the first quarter of 1995 and is believed to contain late-generation 3LC centrifuges.[10] The newest of these plants, the E23 plant, was commissioned in 1997 using TC-11 centrifuges.[11] Additional modules of the E23 plant were undergoing installation and commissioning during the reporting period in 2006, probably with TC-12 centrifuges.[12]

General Sources of Emissions

For a centrifuge plant, a primary source of aerosol effluent is the connection and disconnection of UF_6 cylinders. These releases are proportional to the plant's throughput divided by the size of the cylinders. For all three plants, natural UF_6 is fed from 12.5-tonne cylinders and tails are removed using the same type of cylinder. A mix of cylinder types are used for collecting enriched product, although product is not the dominant flow. Cylinder contents are gasified for feeding by heating the cylinder in a sublimer, and processed material is collected by desublimation into cooled cylinders (details are below). At Capenhurst, these processes happen at near-atmospheric pressure.

In addition to feed-and-withdrawal operations, additional material handling steps also produce emissions. Prior to feeding, each cylinder of UF_6 undergoes a purification step in which its contents are completely sublimed and transferred to another cylinder, allowing highly volatile contaminants (e.g., HF) to escape in the process. This operation can be an additional source of effluent. Although UF_6 is not processed through the plant in liquid form, as had been done in some older enrichment plants, all product cylinders are placed into freestanding autoclaves and the contents liquefied for homogenization and quality-control sampling once enrichment is complete. Blending of different product stocks is also done to meet customer requirements. These post-enrichment operations also produce aerosol effluents.

Finally, the centrifuge cascades can release aerosol effluent, especially through the cascade's vacuum system. The level of release is specific to the centrifuge and cascade design. In general, the leakage is proportional to the number of centrifuges, and to per-centrifuge emissions.

Plant-Specific Variations

The type of sublimer used to gasify UF_6 affects the nature of the effluent coming from the feed and withdraw areas. The older E22 and A3 plants use steam to heat the feed cylinders. Because of the humid environment, discharges of UF_6 from these plants will immediately hydrolyze to UO_2F_2 aerosols, which are then entrained in high moisture air. The E23 plant uses electric heating, so its emissions are drier and hydrolyze more slowly.

For the dry emissions of the E23 plant, particulates are effectively scrubbed from plant off-gasses by keeping the entire plant slightly below atmospheric pressure circulating air through High Efficiency Particulate (HEPA) filters. HEPA filters, by definition, are required to remove at least 99.97% of aerosols at 0.3 μm, and physical phenomena cause HEPA filter efficiency to improve as particles become either small or larger.[13]

The steam chests of the older E22 and A3 plants produce an effluent stream that is too wet to scrub using HEPA filters. In the E22 plant, wet venturi scrubbers are used. These spray a fine mist of water at high velocity into the effluent stream scavenging particulate matter into liquid collection tanks. The efficiency of these scrubbers drops off sharply as particle sizes drop below approximately 0.3 μm.[14]

For the A3 plant, Urenco reports that the emissions are monitored, but not scrubbed.

In addition to each plant's cascade operations, the Capenhurst site has several auxiliary facilities that produce UF_6 emissions. The E23 plant includes a vacuum pump disassembly and decontamination facility, which handles pumps from all three plants. Although most of the uranium from these pumps is removed in a liquid process, some escapes as atmospheric effluents. These emissions are processed through HEPA filters. Approximately 50% of the atmospheric effluent from the whole of the E23 plant can be attributed to its vacuum pump disassembly and decontamination activities. Since this and other auxilliary facilities handle decontamination for equipment coming from all three plants, their contribution is normalized not to the capacity of the E23 plant, but to the total site-wide enrichment capacity.

A detailed breakdown of the emissions for each of the Capenhurst plants is given in Table 3. These estimates demonstrate the large range of potential emissions and the effect of plant-specific effects. Omitted from Table 3 are minor emissions from dedicated container handling facilitates, research facilities, and a chemistry laboratory. These emissions are included in the overall site-wide emissions for Capenhurst given in Table 2.

REFERENCES

1. R.S. Kemp, "Initial Analysis of the Detectability of UO_2F_2 Aerosols Produced by UF_6 Released from Uranium Conversion Plants," *Science and Global Security*, 16 (2008), 115–125; International Atomic Energy Agency, "IAEA Use of Wide Area Environmental Sampling In the Detection of Undeclared Nuclear Activities," Report of Member State Support Program, STR-321, 27 August 1999.

2. Building K-1131 was estimated to release 0.00024 grams per gram (grams uranium effluent per gram of uranium in UF_6 produced). See: "Table-D: Mean Estimate of the Uranium Released at K-25," in *Master Release List.* ChemRisk for Oak Ridge Health Studies, Oak Ridge Dose Reconstruction. DRAFT Task 6 Report, February 1997, as cited in: D. Albright and L. Barbour, "Source Terms for Uranium Enrichment Plants," appendix to STR-321.

3. Letters from Plant Manager, Honeywell Specialty Chemicals, Metropolis, IL to Region III, U.S. Nuclear Regulatory Commission, dated: 7 March 2000, 30 October 2000, 18 April 2001, 27 September 2001, 13 August 2002, 3 July 2002, 6 September 2002, 25 August 2003. Acquired through the Freedom of Information Act, NRC FOIA 2005–0216.

4. Nuclear Regulatory Authority Decommissioning Site Status Summary, http://www.nrc.gov/info-finder/decommissioning/fuel-cycle/honeywell.html (accessed 2 February 2010).

5. "Quarterly Environmental Compliance Report for Port Hope Facility, Fuel Facility Operating License FFOL-3631.0/2007," for 2005 Quarter 4, and 2006 Quarters 1, 2, and 3. An annual summary of the daily variance of Port Hope emissions from 2002 to 2006 is available at: "Cameco Port Hope Conversion Facility, Supplemental Submission in Support of Application for License Renewal, Attachment 2: The minimum and maximum values for environmental monitoring data."

6. Cameco Port Hope Fact Sheet.

7. AREVA, "Rapport environmental, social, et societal 2005: Tricastin," 2005; AREVA, "Rapport environmental, social, et sociétal, du sûreté nucléaire et de radioprotection: Tricastin," 2006; AREVA, "Rapport environmental, social, et sociétal, du sûreté nucléaire et de radioprotection: Tricastin," 2007.

8. Urenco (Capenhurst) Ltd., "Information in Response to the Environment Agency's Review of the Urenco (Capenhurst) Limited Radioactive Disposal Authorisations," Document No: HSE/2006/0017, August 2006.

9. The LEC centrifuge, or Light End Cap, is a subcritical fiberglass overwrap aluminum centrifuge. It is believed to operate at approximately 475 m/s. The 3LC was manufactured at Capenhurst from late 1982 to 1987. The design is reportedly based on the German G3, which is estimated by the author to have had a peripheral velocity in the range of 485–540 m/s.

10. It is possible that the plant contains TC-11 centrifuges (in whole or in part), because BNFL began manufacturing TC-11 machines within about one year after the plant's construction commenced. By March 1987, BNFL manufactured only TC-11 machines.

11. The TC-11 is Urenco's first carbon-fiber centrifuge. It is believed to have a peripheral velocity of about 600 m/s. Its successor, the TC-12, is believed to operate around 620 m/s.

12. A history of the plants is given in, BNFL Capenhurst, "An Information Submission in Support of the Environment Agency's Review of RSA 93 Authorisations at BNFL Capenhurst," April 2003. See also, R.B. Kehoe, *The Enriching Troika: A History of Urenco to the Year 2000* (Urenco, 2002).

13. U.S. Department of Energy, "Specification for HEPA Filters used by DOE Contractors," DOE Standard: DOE-STD-3020–97 (1997).

14. M. Lehner, "Aerosol Separation Efficiency of a Venturi Scrubber Working in Self-Priming Mode," *Aerosol Science and Technology*, 28 (1998), 389–402.