Release of Mercury from Broken Fluorescent Bulbs

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ABSTRACT
Mercury is a persistent, bioaccumulative toxin. Although the primary human exposure is from ingestion of fish contaminated with methyl mercury (HgCH₃), exposures to elemental mercury vapor and mercury compounds via inhalation and dermal contact may also occur. Fluorescent bulbs contain mercury. Estimates of the amount of this mercury released when the bulbs are discarded and broken have varied widely.

A new method was developed for measuring mercury released from broken bulbs. With the assumption that all mercury released is elemental vapor, it was found that between 17 and 40% of the mercury in broken low-mercury fluorescent bulbs is released to the air during a two-week period immediately following breakage, with higher temperatures contributing to higher release rates. One-third of the mercury release occurs during the first 8 hr after breakage. Many bulbs contain more mercury than the low-mercury bulbs tested. A typical discarded bulb releases between 3 and 8 mg of elemental mercury vapor over two weeks. Approximately 620 million fluorescent bulbs are discarded annually in the United States, and many are broken during disposal. Based on the estimated release rate of 3–8 mg per broken bulb developed in this study, discarded bulbs release approximately 2–4 tons of mercury per year in the United States. Waste management systems that minimize breakage before final disposal could reduce this total, as would the reduction of mercury content of bulbs. Elevated airborne levels of mercury could exist in the vicinity of recently broken bulbs, and under certain conditions, mercury concentrations could exceed occupational exposure limits.

INTRODUCTION
Mercury exists in elemental form (Hg⁰) and also is found in numerous chemical compounds, including toxic, bioaccumulative, methylated species, such as methyl mercury (HgCH₃⁺). Many forms of mercury, including elemental, are volatile enough so that a significant portion can exist in the gaseous state. All forms of mercury are toxic to humans to some degree and can lead to a variety of nervous system effects. Any form of mercury in the environment is cause for concern because a portion of this mercury is converted to HgCH₃, which accumulates in fish to levels that can harm humans and wildlife that consume the fish. Also, high concentrations of elemental mercury or mercury compounds in air, usually caused by spills or other releases of mercury in indoor environments, and mercury contamination of groundwater may be of concern.

Recent studies have shown that much of the anthropogenic release of mercury to the environment is from coal combustion and the incineration of wastes. Also, the use of mercury in some industrial processes, notably the production of chlorine, has been the source of much release of mercury. The New Jersey Mercury Task Force has identified the manufacturing of iron and steel as a source of mercury emissions; it is believed that recycled metals used as feedstocks by these plants are contaminated with mercury found in switches and other measuring and electrical apparatus that end up in scrap metal streams. Other product-related releases of mercury occur. The New Jersey Mercury Task Force estimates that more mercury is contained in products than is released through coal and other fuel combustion. Much of the mercury in products can be expected to be eventually released to the environment. Discarded fluorescent bulbs are an example of a product that can release mercury to the environment.

Fluorescent bulbs contain mercury; its function is to convert electrical energy to radiant energy in the UV range, which is then re-radiated in the visible spectrum by the “phosphor” compounds that coat the inside of the
bulb. The average mercury content of a 4-ft-long bulb manufactured today is approximately 12 mg. Bulbs manufactured in the mid-1980s and earlier contained 40 mg or more. Low-mercury bulbs, containing 4 mg or less, are currently produced by at least one manufacturer. A typical fluorescent bulb discarded today is likely to contain an average of approximately 20 mg mercury. Because it is very likely that discarded fluorescent bulbs will be broken during conventional waste handling, there is a concern that occupational exposures to workers handling waste materials may occur.

How much mercury will be released from broken fluorescent bulbs has been the subject of some debate. The U.S. Environmental Protection Agency (EPA) has estimated, based on a model, that 6% of the mercury in broken bulbs is released to the air. An industry report states that only about 1% of the mercury in the bulbs is released during disposal and recycling operations. Release of a much higher percentage of the mercury in discarded bulbs has been suggested by a study for Oak Ridge National Laboratory by Lindberg et al. These researchers have indicated that mercury emissions from broken fluorescent bulbs persist for at least a week and may represent 20–80% of the mercury in the bulbs.

The amount released is a function of the form and quantity of mercury in the bulbs and of other factors, especially temperature. The form of mercury depends on a number of factors, such as the age of the bulb, the type of bulb, and, perhaps, whether the bulb is operated continuously or intermittently. Elemental mercury is introduced into the bulb during manufacture as a single charge. A certain amount of this mercury vaporizes each time the bulb heats up during operation and then condenses as the bulb cools. Over time, elemental mercury becomes dispersed throughout the bulb and exists as very small particles or beads that are typically too small to be seen with the naked eye. It is likely that bulbs near the end of their useful life contain a greater portion of mercury in this dispersed form compared with unused bulbs. This dispersion of mercury can be expected to result in a significant increase in surface area and a corresponding increase in the volatilization rate. Liquid mercury in a contained space will volatilize only until a saturated atmosphere has been attained. However, for broken bulbs, an increased volatilization rate associated with more finely dispersed mercury should result in an increase in mercury flux.

At room temperature, a portion of the mercury in an intact bulb will be present in the vapor state; a saturated atmosphere of elemental mercury within the bulb is necessary for the bulb to function. The concentration of a saturated vapor of mercury at standard temperature and pressure (25 °C, 1 atm) is 13 mg/m³. Fluorescent bulbs contain a partial vacuum; however, less than 0.02 mg are reported to be present in the elemental vapor state, and about 0.1 mg are in the form of solid chemical compounds such as mercury oxide (HgO). The balance is present as elemental liquid mercury distributed on the surface of the phosphor and other internal parts. At bulb operating temperatures (approximately 40 °C), the amount of elemental mercury vapor increases but does not exceed 0.05 mg.

The exact fate of mercury within fluorescent bulbs has not been well described. As the bulb ages, an increasing amount of elemental liquid mercury is converted to solid mercury compounds (principally HgO). According to industry reports, the quantity converted to HgO is between 1 and 4 mg in typical bulbs at the end of the bulb’s rated life, and a certain amount of the elemental mercury originally present binds to the glass as the bulb ages.

To gain a better understanding of the amount of mercury released from broken bulbs, an experiment was conducted to measure the amount of mercury actually released from low-mercury fluorescent bulbs. The experiment was designed to mimic a typical solid waste disposal scenario, in which a discarded bulb will be broken during handling and then stored in an uncovered container, such as a dumpster or trash can, for a period of time before final disposal.

**Sampling Methodology**

**Equipment**

In this experiment, burned-out low-mercury fluorescent bulbs were broken in the bottom of a new 32-gal (121-L) high-density polyethylene cylindrical plastic barrel, and then the contained space was sampled at various intervals to determine the release rate of elemental mercury vapor. The bulbs broken were Philips 4-ft Econ-o-watt F40 CW/RS/EW, 0 8E bulbs, which are reported to contain either 4.4 or 4.7 mg of mercury, depending on exact date of manufacture. A midpoint value of 4.55 mg was assumed to represent the amount of mercury in the bulbs broken in this experiment.

A Jerome 411 Gold Film Mercury Vapor Analyzer was used to detect mercury vapor emissions from the fluorescent bulbs. This direct reading instrument is designed to detect elemental mercury vapor. Readings were collected using the meter’s 10-sec sample mode in which an internal pump draws a precise volume of air into the instrument across a gold foil. Mercury vapor present in the sample forms an amalgam on the gold foil. Mercury concentrations, in mg/m³, are determined by the resulting change in electrical potential across the gold foil. The integrated meter readings are displayed digitally. Precision of the meter is ±5% at 0.107 mg/m³ within the range of >0.0005–1.999 mg/m³.

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A 0.5-in. diameter (1.25-cm) port was installed in the side of the barrel, through which a 0.5-in. o.d. rubber probe connected to the analyzer’s intake port could be inserted. The rubber material of the probe is believed to be neoprene. The port was kept closed when the probe was not inserted.

Procedure
The procedure consisted of the following steps:

1. The mercury concentration inside the barrel was recorded. At the start of the experiment, the concentration was typically “non-detectable,” reflecting the fact that normal atmospheric background levels, which are <10 ng/m³, are below the detection limit of the Jerome 411 mercury analyzer.

2. One or more spent fluorescent bulbs were broken and crushed inside the barrel, and the barrel was immediately covered. Shards of broken bulbs ranged from <1 cm to 20 cm in length. The area outside and around the perimeter of the barrel immediately adjacent to the lid seal was sampled with the meter in its 10-sec mode. Such sampling usually resulted in non-detect levels (with several readings in the range of 0.001–0.004 mg/m³), revealing that a significant amount of mercury vapor did not escape from the closed container.

3. After a period of time, which varied during the course of the experiment from 2 min to more than 10 hr, the concentration of mercury vapor inside the covered barrel was recorded by inserting the sampling probe through the port in the side of the barrel.

4. After the concentration was recorded, the lid was removed to allow mercury vapor from the barrel to escape to the outside atmosphere. It was found that a time of approximately 5 min was long enough to allow most of the mercury vapor inside the barrel to escape.

5. After purging, the probe was inserted into the barrel from the top, the post-purge concentration was recorded, and the barrel was immediately covered with its lid.

Steps 3, 4, and 5 were repeated numerous times over several days. As expected, mercury vapor concentrations in the closed drum were always found to be greater than concentrations after venting, with the difference between these two values representing the amount of mercury vapor that volatilized from the broken tube(s) during that time period. It was found that within a few minutes after the initial breaking of the bulbs, the measured mercury concentration inside the barrel reached a level near the analyzer’s upper detection limit of 1.999 mg/m³. To avoid saturating the gold foil and over-ranging the instrument, the mercury concentrations were measured at short intervals early in the experiment. After the first hour, the flux of mercury from the broken bulbs decreased to the point where the risk of over-ranging the instrument was less of a threat; accordingly, the interval between the closed-drum and vented-drum stage was increased. At no time did the measured concentration inside the barrel exceed the upper detection limit of the analyzer, 1.999 mg/m³.

Because this level is well below the concentration of saturated mercury vapor, 13 mg/m³, it is unlikely that the release of mercury from the broken bulbs would have been appreciably inhibited by the internal concentrations of mercury in the air inside the barrel. However, it is possible that concentration gradients inside the barrel could have resulted in some limitation of mercury volatilization.

RESULTS
Results for experiments run at three different temperatures with low-mercury Philips Alto (green label) Econ-o-watt F40 CW/RS/EW, 0 8E bulbs are shown in Tables 1, 2, and 3.

In each experiment, the amount of mercury released in the first interval (i.e., the time from the initial breaking of the tube(s) to collection of the first pre-purged sample) was adjusted to reflect the fact that some of the mercury released in the initial period after breakage was the mercury already existing as vapor in the tube. The bulbs were 4 ft long and 1.5 in. in diameter, corresponding to a volume of 84.78 in³, or 0.00139 m³. Because a saturated mercury vapor at standard temperature and pressure (25 °C and 1 atm) has a concentration of 13 mg Hg/m³, approximately 0.018 mg can be expected to exist in the vapor form within the volume of a 4-ft tube. This amount was released in one burst immediately upon breakage (because the bulbs are in a partial vacuum, the internal concentration is likely to be somewhat higher). The released amount shown in Tables 1, 2, and 3 was adjusted by subtracting the amount of the initial burst of saturated vapor so that the initial reading would correspond to the later readings representing just the mercury flux from the sides of the tube and other surfaces. To calculate the cumulative release of mercury from broken fluorescent bulbs, the quantity of mercury released in the initial burst was added back in to obtain the total quantity of mercury released.

The amount of mercury released was translated into an amount released per time period, and then into a release rate, in milligrams per hr, as shown in Tables 1, 2, and 3. These three different tables represent releases for the same type of bulbs at three different temperatures. The approximately 40 °F (approximately 5 °C) sampling run was performed at ambient temperature. The
approximately 60 °F (approximately 15 °C) and approximately 85 °F (approximately 30 °C) temperatures were maintained by performing the experiment in an enclosed space heated with an electric heater.

The release rate data were plotted, and a best-fit curve was obtained using Microsoft Excel’s curve-fitting algorithm. R$^2$ values, representing the fit of the curves to the data, ranged from 0.92 to 0.97. These plots are shown as Figures 1, 2, 3, and 4, representing the runs at the three different temperatures (Figures 1–3) and all data combined (Figure 4). The equations that represent the best-fitting curves were then used to integrate the area under each curve and estimate the total amount of mercury released per given time period.

The equations fit to a plot of release rate versus time were in the form of eq 1

$$y = at^b$$  \hspace{1cm} (1)

where $y$ is the release rate in g/hr, $t$ is time in hours and $a$ and $b$ are constants generated by the curve-fitting algorithm. Different values for these constants were generated for each of the three different temperature regimes.

Therefore, the total amount released from time $t_1$ to time $t_2$ is expressed with eq 2

$$\int_{t_1}^{t_2} at^b dt$$  \hspace{1cm} (2)

### Table 1. Mercury release from two broken fluorescent tubes at 40 °F. *Initial reading was .651 mg/m$^3$; adjusted by subtracting portion that was from initial puff of saturated vapor.*

| Date and Time | Action | Closed Drum Cycle Duration (hr) | Meter Reading (mg/m$^3$) | Change in Conc. Before and After | Calc. Release Based on Drum Vol. (g) | Calc. Release Rate (mg/hr) for One Tube | Elapsed Time (hr) to Midpoint of Measuring Period |
|---------------|--------|---------------------------------|---------------------------|----------------------------------|--------------------------------------|-------------------------------------------|-----------------------------------------------|
| 11/19/00 11:11 | Bulbs broken, drum covered | 0.03 | 0.000 | 0.353 | 4.27E-05 | 0.641 | 0.02 |
| 11/19/00 11:13 | Concentration measured through port, adjusted | | 0.353 | | | | |
| 11/19/00 11:13 | Concentration measured through port | 0.08 | 0.941 | 0.290 | 3.51E-05 | 0.211 | 0.06 |
| 11/19/00 11:18 | Concentration measured through port, then drum opened to vent | | | | | | |
| 11/19/00 11:40 | Concentration in open drum measured, then drum closed | | | | | | |
| 11/19/00 11:50 | Concentration measured through port | 0.17 | 0.320 | 0.296 | 3.59E-05 | 0.108 | 0.55 |
| 11/19/00 11:50 | No action | | | | | | |
| 11/19/00 12:21 | Concentration measured through port, then drum opened to vent | 0.52 | 0.749 | 0.429 | 5.20E-05 | 0.060 | 0.89 |
| 11/19/00 13:05 | Concentration in open drum measured, then drum closed | | | | | | |
| 11/19/00 14:01 | Concentration measured through port, then drum opened to vent | 0.93 | 0.673 | 0.661 | 8.01E-05 | 0.043 | 2.35 |
| 11/19/00 15:30 | Concentration in open drum measured, then drum closed | | | | | | |
| 11/19/00 17:43 | Concentration measured through port, then drum opened to vent | 2.22 | 0.696 | 0.668 | 8.09E-05 | 0.018 | 5.41 |
| 11/19/00 23:10 | Concentration in open drum measured, then drum closed | | | | | | |
| 11/20/00 7:46 | Concentration measured through port, then drum opened to vent | 8.60 | 0.670 | 0.658 | 7.97E-05 | 0.005 | 16.27 |
| 11/20/00 21:01 | Concentration in open drum measured, then drum closed | | | | | | |
| 11/21/00 7:35 | Concentration measured through port, then drum opened to vent | 10.57 | 0.511 | 0.499 | 6.04E-05 | 0.003 | 39.10 |
| 11/21/00 23:35 | Concentration in open drum measured, then drum closed | | | | | | |
| 11/22/00 9:10 | Concentration measured through port, then drum opened to vent | 9.58 | 0.345 | 0.343 | 4.15E-05 | 0.002 | 65.18 |

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The total amount released between time $t_1$ and $t_2$ is then calculated from

$$a \left( \frac{t_2^{b+1}}{(b+1)} - \frac{t_1^{b+1}}{(b+1)} \right)$$

(3)

With the assumption that discarded bulbs might reside in a dumpster or trash can for a week before trash pickup, and then reside on the tipping floor of a transfer station or incinerator for another several days, the maximum time that broken bulbs would exist in the waste management system was judged to be two weeks (340 hr). The cumulative amount released from 0 to 1 hr, 0 to 2 hr, 0 to 3 hr, and so on, was then calculated as described. Cumulative total releases up to 340 hr at three different temperatures are shown in Figure 5.

Earlier experiments were also run with other brands of bulbs, but the results are not reported here because the method had not been refined. However, it was clear from the results of these experiments that the amount of mercury released is greater with bulbs that contain more mercury. There is no reason to suspect that the amount released is a function of factors other than initial quantity of mercury contained in the tube, the temperature, and, perhaps, the age of the bulb and whether it has been operated continuously or intermittently.

**DISCUSSION**

It was found that the mercury release rate varies proportionally with temperature, which would be expected because of the greater volatility of mercury at higher temperatures (see Figure 6). Also, the release rates are consistent with the range reported by Lindberg et al. 17 The results are also consistent with a preliminary analysis of data collected during the operation of a commercial bulb-crushing system in Illinois, which suggests that approximately 18% of the mercury in fluorescent bulbs is released over an 8-hr period during crushing operations. 18

This study found that at temperatures ranging from approximately 40 to 85 °F (approximately 5–30 °C), between approximately 17 and 40% of the mercury contained in a broken fluorescent tube will volatilize during a two-week period, with higher volatilization rates corresponding to higher temperatures. One-third of the total amount of mercury released is released during the first 8

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Table 2. Mercury release from two broken fluorescent tubes at 60 °F. *Initial reading was 1.152 mg/m³; adjusted by subtracting portion that was from initial puff of saturated vapor.

| Date and Time | Action                                      | Closed Drum Cycle Duration (hr) | Meter Reading (mg/m³) | Change in Conc. Before and After | Calc. Release (g) Based on Drum Vol. | Calc. Release Rate (mg/hr) for One Tube | Elapsed Time (hr) to Midpoint of Measuring Period |
|---------------|---------------------------------------------|---------------------------------|------------------------|----------------------------------|--------------------------------------|------------------------------------------|-----------------------------------------------|
| 12/2/00 17:00 | Bulbs broken, drum covered                  | 1.000                           |                        |                                  |                                      |                                          |                                               |
| 12/2/00 17:01 | Concentration measured through port, adjusted, then drum opened | 0.02                           | 0.854*                 | 0.854                            | 1.03E-04                            | 3.102                                    | 0.01                                          |
| 12/2/00 17:08 | Concentration measured through port, then drum opened | 0.091                          |                        |                                  |                                      |                                          |                                               |
| 12/2/00 17:11 | Concentration measured through port, then drum opened to vent | 0.05                           | 0.161                  | 0.070                            | 8.48E-06                            | 0.085                                    | 0.15                                          |
| 12/2/00 17:15 | Concentration measured, then drum closed    | 0.047                           |                        |                                  |                                      |                                          |                                               |
| 12/2/00 17:34 | Concentration measured through port         | 0.32                           | 0.598                  | 0.551                            | 6.67E-05                            | 0.105                                    | 0.40                                          |
| 12/2/00 17:34 | No action                                   |                                 |                        |                                  |                                      |                                          |                                               |
| 12/2/00 18:14 | Concentration measured through port, then drum opened to vent | 0.67                           | 1.260                  | 0.662                            | 8.02E-05                            | 0.060                                    | 0.89                                          |
| 12/2/00 18:19 | Concentration measured, then drum closed    | 0.198                           |                        |                                  |                                      |                                          |                                               |
| 12/2/00 19:49 | Concentration measured through port, then drum opened to vent | 1.50                           | 1.042                  | 0.844                            | 1.02E-04                            | 0.034                                    | 2.06                                          |
| 12/2/00 20:17 | Concentration measured, then drum closed    | 0.080                           |                        |                                  |                                      |                                          |                                               |
| 12/2/00 23:08 | Concentration measured through port, then drum opened to vent | 2.85                           | 1.146                  | 1.066                            | 1.29E-04                            | 0.023                                    | 4.70                                          |

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hr after breakage. Many bulbs contain more mercury than the low-mercury bulbs tested. A typical discarded bulb, containing approximately 20 mg of mercury, may release between 3 and 8 mg over two weeks. Older bulbs, such as those manufactured in the mid-1980s or earlier, which contained 40 mg Hg or more, can be expected to release proportionately more mercury after breakage.

The pattern of emission, with an initially rapid release declining to a slowly decreasing rate, suggests that at least some of the mercury in the bulbs is in a form that can quickly vaporize, such as tiny droplets of elemental mercury. If this is the case, agitation of the broken pieces would speed up the release. The slowly decreasing rate that appears to result after the first 8 hr or so may reflect the gradual release of forms of mercury less subject to volatilization, such as mercury adsorbed to surfaces within the bulb. It is also possible that the decline in emission rate reflects oxidation of mercury. If significant amounts of mercury are oxidizing over time, mixture of broken bulbs with other wastes might reduce the mercury emission rate.

Approximately 620 million fluorescent bulbs are discarded annually.\textsuperscript{19} Despite the existence of recycling programs, it is estimated that, nationally, only about 20\% of discarded bulbs are recycled.\textsuperscript{20} It is expected that most of the bulbs that are not recycled are likely broken during disposal. Extrapolation of the results of this study, with the assumptions that the conditions in the disposal system mimic the study conditions, suggests that discarded fluorescent bulbs release approximately 2–4 t/yr of mercury in the United States.

However, use of the results of this study to estimate mercury emissions from broken bulbs must note several sources of uncertainty. First, burial, either by soil or by

| Date and Time  | Action                             | Closed Drum Cycle Duration (hr) | Meter Reading (mg/m$^3$) | Change in Conc. Before and After | Calc. Release Rate (g) Based on Drum Vol. | Calc. Release Rate (mg/hr) for One Tube | Elapsed Time (hr) to Midpoint of Measuring Period |
|----------------|------------------------------------|---------------------------------|---------------------------|----------------------------------|-------------------------------------------|------------------------------------------|-----------------------------------------------|
| 12/3/00 11:20  | Bulbs broken, drum covered         |                                 | 0.000                     |                                  |                                           |                                          |                                               |
| 12/3/00 11:22  | Concentration measured through port, adjusted, then drum opened | 0.03                           | 1.142                     | 1.142                            | 1.38E-04                                  | 2.074                                    | 0.02                                         |
| 12/3/00 11:25  | Concentration in open drum measured, then drum closed          |                                 | 0.484                     |                                  |                                           |                                          |                                               |
| 12/3/00 11:27  | Concentration measured through port, then drum opened to vent | 0.03                           | 0.747                     | 0.263                            | 3.19E-05                                  | 0.478                                    | 0.08                                         |
| 12/3/00 11:32  | Concentration in open drum measured, then drum closed          |                                 |                            |                                  |                                           |                                          |                                               |
| 12/3/00 11:54  | Concentration measured through port, then drum opened to vent | 0.37                           | 1.674                     | 1.514                            | 1.83E-04                                  | 0.250                                    | 0.37                                         |
| 12/3/00 12:02  | Concentration in open drum measured, then drum closed          |                                 |                            |                                  |                                           |                                          |                                               |
| 12/3/00 12:22  | Concentration measured through port, then drum opened to vent | 0.33                           | 0.527                     | 0.350                            | 4.24E-05                                  | 0.064                                    | 0.85                                         |
| 12/3/00 15:03  | Concentration in open drum measured, then drum closed          |                                 |                            |                                  |                                           |                                          |                                               |
| 12/3/00 16:05  | Concentration measured through port, then drum opened to vent | 1.03                           | 0.734                     | 0.726                            | 8.79E-05                                  | 0.043                                    | 4.22                                         |
| 12/3/00 16:25  | Concentration in open drum measured, then drum closed          |                                 |                            |                                  |                                           |                                          |                                               |
| 12/3/00 17:47  | Concentration measured through port, then drum opened to vent | 1.37                           | 0.696                     | 0.633                            | 7.67E-05                                  | 0.028                                    | 5.75                                         |
| 12/3/00 18:11  | Concentration in open drum measured, then drum closed          |                                 |                            |                                  |                                           |                                          |                                               |
| 12/3/00 21:35  | Concentration measured through port, then drum opened to vent | 3.40                           | 1.521                     | 1.498                            | 1.81E-04                                  | 0.027                                    | 8.53                                         |

Table 3. Mercury release from two broken fluorescent tubes at 85 °F. *Initial reading was 1.440 mg/m$^3$; adjusted by subtracting portion that was from initial puff of saturated vapor.
other waste, soon after breakage could be expected to reduce the total emission from broken bulbs. Therefore, waste management systems that minimize breakage before final disposal, which is assumed to minimize subsequent release of mercury,\textsuperscript{21} could reduce this total (as could the reduction of mercury content of bulbs).

Second, because in this study the barrel was not flushed continuously as in a dynamic flux chamber, artificial gradients could have occurred because of lack of mixing. In this situation, concentrations near the surfaces of the broken bulbs could have actually become sufficiently high that overall emissions became limited by molecular (as opposed to turbulent) diffusion. Thus, the results reported herein might underestimate actual emissions.

Third, it is known that both plastic and rubber materials adsorb mercury. In this study, the sorption properties of the rubber used for the probe into the barrel, and the barrel itself, were not determined. This is another reason that the measured emission estimates may underestimate actual emissions.

It should also be noted that the Jerome 411 analyzer measures elemental mercury vapor only. Although it is believed that most of the volatile mercury in fluorescent bulbs is elemental, it is possible that, besides elemental mercury, volatile mercury compounds (e.g., oxidized gaseous species) exist and are also released when bulbs are broken. Also, some mercury or mercury compounds may be bound to glass or to particles of the powder coating the inside of fluorescent bulbs. Particles of this powder are released and can be dispersed into the environment when bulbs are broken, and any mercury or mercury compounds bound to the particles would similarly be dispersed. Because this study measured only elemental mercury, the total amount released may be underestimated. Further research should address the species of mercury released.

A significant source of uncertainty in the estimated emission rate is the calculation method itself. As described, power law functions were fit to the data, and then the area under these curves was integrated over a period extending to two weeks. Although the fit of the functions to the data is good, extrapolation of these curves over several orders of magnitude nevertheless introduces uncertainty. To estimate this uncertainty, the data were linearized by taking the log of both the elapsed time and the

**Figure 1.** Release of mercury from single broken fluorescent bulb containing approximately 4.55 mg mercury: Rate vs. time at 40 °F (5 °C) and best-fitting power function, not including initial release of $\sim1.8E-05$ g vapor.

**Figure 2.** Release of mercury from single broken fluorescent bulb containing approximately 4.55 mg mercury: Rate vs. time at 60 °F (15 °C) and best-fitting power function, not including initial release of $\sim1.8E-05$ g vapor.

**Figure 3.** Release of mercury from single broken fluorescent bulb containing approximately 4.55 mg mercury: Rate vs. time at 85 °F (30 °C) and best-fitting power function, not including initial release of $\sim1.8E-05$ g vapor.

**Figure 4.** Release of mercury from single broken fluorescent bulb containing approximately 4.55 mg mercury: Rate vs. time at three temperatures and best-fitting power function, not including initial release of $\sim1.8E-05$ g vapor.
emission rate, and the 95% confidence intervals of the intercept and slope for the linearized (log-log) functions were determined. These values were used to generate additional power law functions, which were in turn integrated over the time period. Nine different combinations of slope and intercept were used, representing the mean intercept/mean slope, mean intercept/minimum slope, mean intercept/maximum slope, minimum intercept/mean slope, and so on. In all cases, the minimum slope/minimum intercept combination generated the smallest total 340-hr emission estimate, and the maximum slope/maximum intercept combination generated the largest. In the case of the 60 °F run, the largest estimate actually exceeded 100% of the possible emissions, and the smallest estimate was that only 10% of the total amount of mercury in the bulbs would be emitted over two weeks. With the 85 °F run, the range of uncertainty was from 88 to 25%, and for the 40 °F run, the range was 37–12%. When all data were combined and a similar analysis was performed, the best estimate was that 25% of the total was emitted, with a range from 17 to 40%.

The high uncertainty associated especially with the runs at 60 and 85 °F is likely a function of the relatively limited amount of data collected at those temperatures.

Despite the uncertainty, it appears that higher temperatures do lead to higher emissions, as would be expected based on the known relationship of temperature to volatilization rate. To attempt to shed more light on the temperature dependence of the emission rates, subsets of the data sets for each temperature run were analyzed by excluding the first three readings, corresponding to the initial short time intervals. Removing the first three data points from the 40 °F run and all runs lumped together makes the slope more negative, implying a quicker decrease in emissions than predicted from the best-fit line derived from all data. However, the opposite is true for the 60 and 85 °F runs. This suggests that, at higher temperatures, relatively high rates of emission may exist for a longer time.

Another aspect of these findings should be noted. This study suggests that elevated airborne levels of mercury, exceeding EPA’s reference concentration of 300 ng/m³, can exist in the vicinity of recently broken bulbs, and under certain conditions, mercury concentrations may exceed occupational exposure limits. The American Conference of Governmental Industrial Hygienists (ACGIH) and the National Institute for Occupational Safety and Health (NIOSH) recommend an 8-hr time weighted average exposure limit of 0.025 and 0.05 mg/m³, respectively, for elemental mercury vapor. The Occupational Safety and Health Administration has set a mercury permissible exposure limit of 0.1 mg/m³. ACGIH and NIOSH both list a “skin” notation for elemental mercury suggesting that exposure, either through direct contact or vapor contact with skin, eyes, and mucous membranes, may contribute significantly to overall exposure. Potential occupational exposure may exist for sanitation workers and employees involved in recycling fluorescent bulbs in the presence of broken bulb residue.

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