

# Environmental Release of Mercury from Broken Compact Fluorescent Lamps

Yadong Li\* and Li Jin

*Department of Civil and Environmental Engineering, Jackson State University, Jackson, Mississippi.*

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## Abstract

This article presents an original study on the releases of mercury (Hg) from broken compact fluorescent lamps (CFLs) under various environmental conditions. Leaching of Hg in liquids was examined using the U.S. Environmental Protection Agency's standard procedures Toxicity Characteristic Leaching Procedure (TCLP) and Synthetic Precipitation Leaching Procedure. Emission of Hg in vapor phase from broken CFLs was detected using an emission monitoring system. CFLs of eight different brands and four different wattages were tested. Results show that Hg contents in CFLs varied significantly with brand. Total amount of Hg contained in each CFL ranged from 0.1 to 3.6 mg, and only <4% of the Hg was TCLP-leachable. Hg concentrations in TCLP extracts of all the new CFLs tested were lower than the regulatory level of 0.2 mg/L and thus the discarded CFLs do not fall into the hazardous waste category. Hg concentrations in Synthetic Precipitation Leaching Procedure extracts were lower than those in the TCLP extracts. Hg vapor emission test revealed that the CFLs continuously release Hg vapor once broken and the release can last over 10 weeks. Total amount of Hg vapor released from a broken CFL can exceed 1.0 mg, which can cause Hg level in a regular room to exceed the safe human exposure limit under poor ventilation conditions. Results provide useful implications in guiding the handling and treatment of CFLs during and after use.

**Key words:** compact fluorescent lamp; mercury; leaching; environmental release

## Introduction

**B**ECAUSE OF THE energy-saving attribute and long lifespan, compact fluorescent lamps (CFLs) are widely used in businesses and homes and are rapidly replacing the traditional incandescent bulbs in recent years. CFLs use ~75% less energy than incandescent lamps and last up to 10 times longer. A CFL typically consists of a glass tube, an electronic ballast, and a metal end. The glass tube is filled with low-pressure mercury (Hg) vapor and inert gases, such as argon and krypton, and phosphor coating on the inside wall of the tube. The electronic ballast contains a small circuit board with rectifiers, a filter capacitor, and usually two switching transistors. The shapes of CFLs found on the market include spiral, covered-A-shape, globe, tubed, candle, indoor reflector, and outdoor reflector. The most popular shape is spiral. In fact, all the other shapes except the tubed are different only in their lenses. The insides are also spiral glass tube. When an electric current from the ballast passes the tube, it excites the Hg vapor and generates radiant energy, primarily in the ultraviolet range. The energy causes the phosphor coating on the tube to fluoresce, converting the ultraviolet light into

visible light. Changing the composition of the phosphors powder inside fluorescent tubes changes the spectrum of light produced.

The amounts of Hg contained in CFLs are very small. In April 2007, the National Electric Manufacturers Association set a Hg reduction goal of 5 mg per lamp for CFLs up to 25 W, and 6 mg in CFLs with higher wattages. In 2003, the best practice CFLs already contained as low as 1.4–2.7 mg Hg per lamp (Dunmire *et al.*, 2003). By 2008, many of the largest CFL manufacturers such as GE, Sylvania, Royal Phillips, and Lights of America have achieved the Hg reduction goals with Hg contents of 50% or more below the National Electric Manufacturers Association reduction target (Eckelman *et al.*, 2008). The contribution of fluorescent lamps to the overall Hg emissions is only about 1% in the United States, whereas 99% is from coal-fired power plants (EPA, 1997). However, the emissions of Hg vapor from CFLs are often in direct contact with people when the CFLs are broken during shipping, handling, retailing, and using. The Hg leaching out from CFLs in dissolved forms in the environment can be transformed into methylmercury, which biomagnifies up the food chain and is highly toxic.

The objective of this study was to quantify the releases of Hg from broken CFLs in vapor form and in dissolved forms under various environmental conditions and to evaluate their potential hazards to human health and the environment.

\*Corresponding author: Department of Civil and Environmental Engineering, Jackson State University, 1400 J. R. Lynch St., Jackson, MS 39217-0168. Phone: 601-979-1093; Fax: 601-979-3238; E-mail: yadong.li@jsums.edu

## Materials and Methods

### CFL samples

The CFLs of eight popular brands with two different shapes and five different wattages were chosen as test samples in this study. The two different shapes were spiral and covered-A shapes, which are most commonly found on the market. The wattages of CFLs ranged from 7 to 42 W. They are widely used for lighting in residential houses and commercial buildings. The new CFLs were purchased from the local stores during August 2008 to July 2009. The used CFLs were collected from a residential house and had been used for about 3 years.

### Extraction tests

Three types of extraction tests were performed on the CFL samples. The U.S. Environmental Protection Agency (EPA) SW-846 Method 7471 Mercury in Solid and Semisolid Waste (EPA, 2007) was used for the total Hg extraction from the CFLs. The EPA SW-846 Method 1311 Toxicity Characteristic Leaching Procedure (TCLP) (EPA, 1992) was used to examine the leaching of Hg from broken CFLs under landfill conditions and to determine whether or not the discarded CFLs are hazardous waste. The EPA Synthetic Precipitation Leaching Procedure (SPLP) (EPA, 1994) was employed to evaluate the leaching of Hg from broken CFLs when subjected to rainwater.

To prepare the samples for these extraction tests, the same CFLs were used to achieve a solid mixture a minimum of 140 g. The glass tubes of CFLs were carefully removed from the lamps, weighed, and pulverized in a mortar into particles with a maximum size of 5 mm. The mixture consisted of glass crumbs and phosphor powder originally contained in the tubes. The tubes weighed 9 to 69 g for CFLs with wattages from 7 to 42 W; the higher the wattages were, the heavier the CFL tubes weighed. After being thoroughly mixed, an aliquot of 5.00 g was transferred to a 250 mL glass bottle for total Hg extraction, and two aliquots of 20.0 g were transferred to two separate plastic bottles for TCLP and SPLP tests. Triplicate tests were performed for the same CFLs. Therefore, a total of 135 g of the solid mixture of the same CFLs was used for all the extraction tests. The pulverization of the CFL tubes and transfer of the aliquots were performed in a laboratory fume hood within 30 min.

For the total extraction of Hg, 50 mL of deionized water and 50 mL of aqua regia was added to a 250 mL glass bottle containing the 5.00 g solid sample as described above. The mixture was heated for 30 min at  $95^{\circ}\text{C} \pm 3^{\circ}\text{C}$ . After overnight cooling, the mixture was filtered through Whatman No. 41 filter paper. The filtrate was adjusted to a volume of 100 mL using deionized water, and then preserved in sealed bottles for Hg analysis.

In the TCLP test, the 20.0 g solid sample was placed in a 500 mL high-density polyethylene plastic bottle and 400 mL TCLP extraction fluid #1 was added to meet the 1:20 solid-to-liquid ratio as required by the method. The extraction fluid was a buffer solution of which 1 L contained 5.7 mL of glacial acetic acid and 64.3 mL of 1N sodium hydroxide. The pH was  $4.93 \pm 0.05$ . The bottles were agitated in a rotary extractor at 30 rpm and  $22^{\circ}\text{C} \pm 2^{\circ}\text{C}$  for  $18 \pm 2$  h. After the agitation, the samples were filtered through a  $0.6\text{--}0.8\ \mu\text{m}$  glass-fiber filter.

The filtrates, that is, TCLP extracts, were preserved by adding concentrated nitric acid to a pH below 2.0 for the analysis of Hg.

The SPLP test was performed in the same way as for the TCLP test except that the extraction fluid was different. This SPLP extraction fluid was prepared by adding 60/40 weight percent mixture of sulfuric and nitric acids to deionized water to a final pH of  $4.20 \pm 0.05$ .

### Vapor Hg emission test

This test was to examine the amounts and rates of Hg release from broken CFLs in vapor form. The container used in the test was made of high-density polyethylene plastic and had an effective volume of 21 L. The top cover was made of Plexiglass. Rubber seals were used between the flanges of the top of the container and the cover to ensure that the container was free from air leak. The cover had two openings. One was the air inlet, and the other was connected to Hg sorbent tubes and a flow meter through plastic tubing. A vacuum was applied to the end of the tubing to vent the air from the container. The venting flow rate was set at 300 mL/min. The Hg sorbent tubes were Anasorb C300, which has proven to be very efficient in absorbing vapor Hg from the air (Takaya *et al.*, 2006; Korell, 2009). To insure absorption efficiency, two sorbent tubes were used in series in the venting line of the container. A schematic of this system is shown in Fig. 1. The system was set in a vented hood in the laboratory. The room temperature was  $20^{\circ}\text{C} \pm 2^{\circ}\text{C}$ .

In the beginning of the emission test, the CFL sample was placed in the container and the spiral glass tube was broken into several segments inside the container and the cover was immediately secured. A blank control was produced by pumping the clean air in the same laboratory through the sorbent tubes at the same flow rate (300 mL/min) for 3 days. The two sorbent tubes taken off the venting line after each cycle were crushed and extracted for total Hg analysis using the same methods as for total Hg extraction from solid samples as described in the previous section.

### Analysis of Hg

All the liquid samples produced in the fore-mentioned tests were prepared and analyzed according to the EPA Method

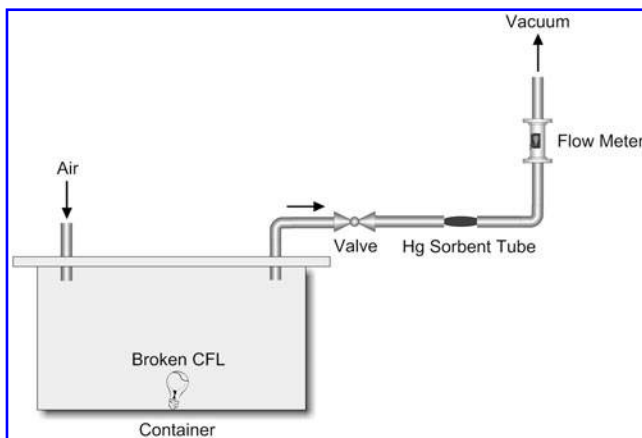


FIG. 1. Schematic of vapor Hg emission test system. Hg, mercury.

245.7 Mercury in Water by Cold Vapor Atomic Fluorescence Spectrometry (EPA, 2005). An Hg analyzer (P S Analytical Millennium Merlin) was used for the analysis. It was calibrated using 0, 0.5, 1, 5, and 20  $\mu\text{g/L}$  standard Hg solutions. Samples with Hg concentrations exceeding the calibration range were properly diluted. The samples from the total Hg extraction test of CFLs normally required 1,000 times dilution, whereas the majority of TCLP and SPLP extracts of the CFLs required no dilution.

All the liquid samples were digested before analysis. An aliquot of 25 mL of the samples was transferred to a 250 mL glass bottle. Then, 7.5 mL of 33% (v/v) HCl and 1 mL of 0.1 N potassium bromate/potassium bromide ( $\text{KBrO}_3/\text{KBr}$ ) were added to the bottle. Deionized water was added to a total volume of 50 mL. The two times dilution of the sample (from 25 to 50 mL) would be factored in the final measurements. The bottle was then closed for a minimum of 30 min digestion. After the digestion, 30  $\mu\text{L}$  of 12% (m/v) hydroxylamine hydrochloride was added to each bottle to remove excess bromine, which was indicated by the disappearance of yellow color because of the destruction of the  $\text{KBrO}_3/\text{KBr}$ . The samples were allowed to react for 5 min before being placed in the analysis rack.

## Results and Discussion

### Extraction test results

The Hg contents (mg/kg) in the solid samples obtained through the total Hg extraction test and the results from the TCLP and SPLP tests for different CFLs are given in Table 1. The Hg content in a CFL glass tube was obtained by multiplying the Hg concentration of the liquid sample from the total Hg extraction by the sample volume (100 mL) and divided by the mass of solid CFL sample in the extraction (5.00 g). Since the mass of each glass tube had been weighed, the total Hg (mg) in the tube was calculated by multiplying the Hg content and the mass of the tube. Some Hg vapor may have been released during the preparation of solid samples for the total Hg extraction. Therefore, the amount of Hg ob-

tained from the total Hg digestion was only the Hg in the phosphor and on the glass. However, the Hg vapor emission test results as presented in next section showed that the Hg vapor release rate was very low. For the three CFLs used in the emission test, the amounts of Hg vapor released in 1 day after they were broken were detected to be 0.058, 0.117, and 0.038 mg for CFL1-13W, CFL4-13W, and CFL5-13W, respectively (Fig. 2), which accounted for 3.6%, 4.5%, and 1.8% of the total Hg obtained in the total extraction test (Table 1). Therefore, the portion of Hg lost in vapor during the preparation of the extraction samples was insignificant in the determination of total Hg content.

It can be seen in Table 1 that the Hg contents varied significantly with CFL brand. Even for the same CFLs, the variations in the results among the triplicate samples were big in many cases, as exhibited in the standard deviations of the measurements. These variations could be mainly caused by the inhomogeneity of the solid samples that consisted of mixtures of phosphor powder and glass crumbs. For the spiral type CFLs produced by Manufacturer 1, there were no significant differences in total amounts of Hg contained for different wattages: 0.92, 1.0, 1.1, and 1.6 mg for 10, 20, 42, and 13 W, respectively. However, the total Hg amounts in CFLs varied with brands or manufacturers. For the most popular 13 W spiral type CFLs, the total Hg ranged from 0.17 to 3.6 mg per bulb for seven different brands. There was a noteworthy difference in total Hg between the new and the used CFLs tested, that is, CFL8-7W and CFL8-7W-used. The new one contained much lower amount of Hg (0.1 mg) than the used one (0.2 mg). This may be attributed to the continuous efforts of the manufacturers in reducing the use of Hg in CFLs in recent years. The used CFLs in this study were 3 years older than the new ones. Also, since much elemental Hg is changed to oxidized form during use, there might be less vapor Hg loss from the used CFLs than from the new ones in the preparation of the total extraction test.

The TCLP and SPLP test results are given as the concentrations of Hg in the TCLP and SPLP extracts as shown Table 1. Since TCLP extraction fluid is more aggressive than SPLP

TABLE 1. MERCURY EXTRACTION RESULTS OF DIFFERENT COMPACT FLUORESCENT LAMPS

Sample ID <sup>a</sup>	Hg content in CFL glass tube (mg/kg) <sup>b</sup>	Hg in TCLP extract ( $\mu\text{g/L}$ ) <sup>b</sup>	Hg in SPLP extract ( $\mu\text{g/L}$ ) <sup>b</sup>	Total mass per tube (g)	Total Hg in CFL (mg)
CFL1-10W	65.3 ± 28.9	5.8 ± 0.8	0.63 ± 0.22	14.0	0.92
CFL1-13W	103 ± 48	5.0 ± 1.6	4.7 ± 2.2	15.5	1.6
CFL1-20W	37.1 ± 1.2	58.6 ± 11.5	16.7 ± 5.9	26.9	1.0
CFL1-42W	15.8 ± 5.2	23.8 ± 11.2	1.1 ± 0.4	69.1	1.1
CFL2-13W	53.4 ± 11.6	4.0 ± 3.4	2.3 ± 1.1	18.7	1.0
CFL3-13W	37.1 ± 24.5	5.0 ± 2.1	1.2 ± 0.1	20.8	0.77
CFL4-13W	127 ± 64	6.7 ± 1.4	2.5 ± 1.3	20.3	2.6
CFL5-13W	105 ± 15	15.5 ± 7.8	0.45 ± 0.16	20.3	2.1
CFL6-13W	171 ± 55	144 ± 21	3.2 ± 2.9	20.9	3.6
CFL7-13W	11.9 ± 5.1	2.0 ± 0.1	0.40 ± 0.19	19.6	0.23
CFL8-7W	11.2 ± 3.5	9.3 ± 2.2	0.47 ± 0.3	8.7	0.10
CFL8-7W-used	23.2 ± 7.1	179 ± 30	17.5 ± 7.7	8.9	0.20

<sup>a</sup>In the CFL sample ID, the first part is for different CFL manufacturers or brands differentiated by the number and the second part is the wattage of the CFL. CFL1 through CFL 7 were all spiral type and CFL8 was Cover-A-Shape.

<sup>b</sup>Data values are average ± standard deviation.

CFL, compact fluorescent lamp; Hg, mercury; TCLP, Toxicity Characteristic Leaching Procedure; SPLP, Synthetic Precipitation Leaching Procedure.

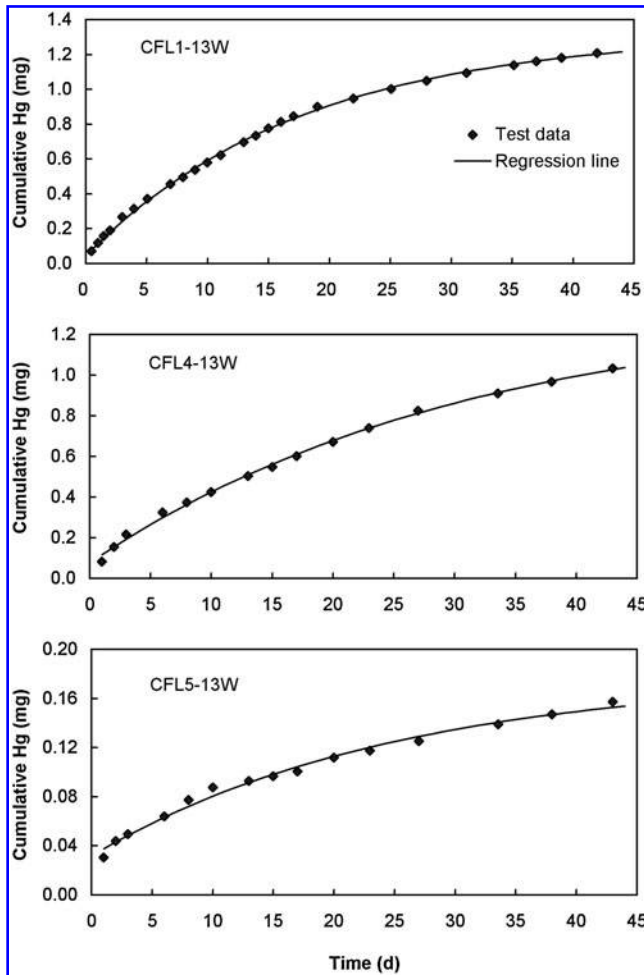


FIG. 2. Hg vapor releases from three different broken CFLs. The regression equations are  $M = 1.32 - 1.28e^{-0.056t}$ ,  $M = 1.35 - 1.27e^{-0.032t}$ , and  $M = 0.18 - 0.15e^{-0.040t}$  for CFL1-13W, CFL4-13W, and CFL5-13W, respectively, where  $M$  is the cumulative amount of Hg vapor (mg),  $t$  is time (d), and  $e$  is Euler's number. The squared correlation coefficients are 0.999, 0.997, and 0.989, respectively. CFL, compact fluorescent lamp.

extraction fluid, the Hg concentrations in TCLP extracts were significantly higher. However, the Hg concentrations in TCLP and SPLP extracts were not necessarily proportional to the Hg content in the same CFLs, indicating differences in the leachabilities of Hg in different CFLs. The ratios of TCLP-leachable Hg and SPLP-leachable Hg verse the total Hg were calculated and summarized in Table 2. The ratios were calculated as

$$\text{Ratio} = \frac{C \cdot V}{1000M \cdot S} \times 100$$

where  $C$  ( $\mu\text{g/L}$ ) is Hg concentration in the TCLP or SPLP extract,  $V$  (mL) is the volume of TCLP or SPLP extract,  $M$  (g) is the mass of the solid sample for TCLP or SPLP test, and  $S$  (mg/kg) is the Hg content in the solid CFL sample. The values show that the TCLP-leachable Hg in all the new CFLs ranged from 0.1% to 4.0%, and SPLP-leachable 0.01% to 0.14%. The leachable Hg in the used CLF (CFL8-7W-used) was significantly higher than the new one (CFL8-7W): 15% versus 1.6%

TABLE 2. RATIOS OF LEACHABLE MERCURY IN TOXICITY CHARACTERISTIC LEACHING PROCEDURE AND SYNTHETIC PRECIPITATION LEACHING PROCEDURE TESTS

CFL Sample	TCLP-leachable Hg (%)	SPLP-leachable Hg (%)
CFL1-10W	0.18	0.02
CFL1-13W	0.10	0.09
CFL1-20W	3.2	0.90
CFL1-42W	3.0	0.14
CFL2-13W	0.15	0.09
CFL3-13W	0.27	0.06
CFL4-13W	0.11	0.04
CFL5-13W	0.30	0.01
CFL6-13W	1.7	0.04
CFL7-13W	0.33	0.07
CFL8-7W	1.6	0.08
CFL8-7W-used	15	1.5

TCLP-leachable and 1.5% versus 0.08% SPLP-leachable for the used and new respectively. This is in agreement with previous studies with the explanation of gradual transformation of elemental Hg to divalent soluble forms through oxidative reaction with phosphor (Dang *et al.*, 1999; Raposo *et al.*, 2003; Lee *et al.*, 2009).

The TCLP results of all the new CFLs tested were below  $100 \mu\text{g/L}$  (Hg in TCLP extracts) except one (CFL6-13W) that had  $144 \mu\text{g/L}$ . The used CFL (CFL8-7W-used) produced the highest Hg concentration in the TCLP extract, that is,  $179 \mu\text{g/L}$  with a standard deviation of  $30 \mu\text{g/L}$ , which makes it in the proximity of the EPA regulatory level of  $200 \mu\text{g/L}$  to be classified as hazardous waste. Note that only the glass tubes of the CFLs where all the Hg is enclosed were used in the TCLP and SPLP tests. If other components (e.g., ballast and metal end) were included, the results would be even lower.

#### Vapor Hg release results

Three different brands of CFLs with the same wattage (13 W) were used for Hg vapor emission test. The sample identifications were CFL1-13W, CFL4-13W, and CFL5-13W on which the total Hg extraction, TCLP test, and SPLP test were also performed. The cumulative amounts of Hg vapor release from each of the three CFLs versus time are plotted in Fig. 2. The graphs reveal that Hg vapor was continuously released over the 43 days test period. The release rates decreased with time as reflected by the decreasing slopes of the cumulative curves with time. The Hg release could have continued for much longer time if the test did not have to be stopped because of time constraint. A regression analysis of the data resulted in three exponential equations as shown in the legend for Fig. 2.

Total amounts of Hg that could be eventually released in vapor phase from the CFLs can be estimated from the regression equations with the time approaching infinity. The estimated amounts are 1.32, 1.34, and 0.18 mg for CFL1-13W, CFL4-13W, and CFL5-13W, respectively. These vaporizable amounts represent the amounts of elemental Hg in the CFLs. In the total Hg extract test, the total amounts of Hg in these CFLs were measured to be 1.6, 2.6, and 2.1 mg, respectively, as presented in Table 1. Therefore, the percentages of the Hg that could be released in vapor are  $\sim 83\%$ ,  $52\%$ , and  $8.6\%$  of the

total Hg contained in these CFLs, respectively. Using the regression equations, it can also be estimated that for 95% of the total vaporizable Hg in CFL1-13W, CFL4-13W, and CFL5-13W to be released under this test condition, 53, 92, and 128 days are needed, respectively. These data suggest that the vaporizable amounts of Hg in different CFLs and the rates of vaporization are significantly different. These differences could be caused by the varied speciations of Hg in the phosphors and glasses.

Studies have indicated that Hg in the initial lamps is primarily in elemental form and is mostly changed to divalent form at the end of lamp life (Dang *et al.*, 1999; Dunmire *et al.*, 2003; Raposo *et al.*, 2003). Therefore, the breakage of unused CFLs during transportation, handling, sales, and installation causes more release of Hg into the air than that of the used ones. According to Johnson *et al.* (2008), the release of 1 mg of Hg vapor into a 500 m<sup>3</sup> room can yield an Hg level 10 times the children's exposure limit (0.2 µg/m<sup>3</sup>) recommended by the Agency for Toxic Substances and Disease Registry. Thus, in most cases, one broken CFL could cause the Hg vapor concentration in such a room to exceed the safe level for children if the room is not vented.

### Summary

The Hg contents in CFLs vary significantly with manufacturers. For the most popular 13 W spiral CFLs, the total amount of Hg contained range from 0.17 to 3.6 mg per bulb for the CFLs of seven different brands or manufacturers. For the same brand CFLs, the Hg contents increase slightly with wattage. Of the total Hg in each CFL, only < 4% is leachable in the TCLP test and < 0.14% in SPLP test. All the CFLs passed the TCLP test and majority of the CFLs tested had a Hg concentration in the TCLP extracts significantly lower than the regulatory level of 0.2 mg/L, indicating that vast majority of CFLs are nonhazardous. However, the TCLP results do not fully reflect the potential hazards of CFLs because the CFLs continuously release Hg vapor once broken. The emission can last weeks even months and the total amount of Hg that can be released in vapor from new CFLs can often exceed 1.0 mg. Since vapor Hg can be readily inhaled by people, rapid removal of broken CFLs and sufficient ventilation of rooms by fresh air are critical to prevent people from potential harms. Effective packaging for preventing the breakage of CFLs and retaining the Hg vapor from broken CFLs is another way to reduce human exposure to Hg in transportation, handling, and storage.

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### Author Disclosure Statement

The authors declare that no conflicting financial interests exist.

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