

CCGP

coal combustion and gasification products

Coal Combustion and Gasification Products is an international, peer-reviewed on-line journal that provides free access to full-text papers, research communications and supplementary data. Submission details and contact information are available at the web site.

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Web: www.coalcgp-journal.org

ISSN# 1946-0198

Volume# 4 (2012)

Editor-in-chief: Dr. Jim Hower, University of Kentucky Center for Applied Energy Research

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Suggested Citation format for this article:

Long, C.M., Sax, S.N., Lewis, A.S., 2012, Potential Indoor Air Exposures and Health Risks from Mercury Off-Gassing of Coal Potential Indoor Air Exposures and Health Risks from Mercury Off-Gassing of Coal. *Coal Combustion and Gasification Products* 4, 68-74, doi: 10.4177/CCGP-D-12-00006.1

Potential Indoor Air Exposures and Health Risks from Mercury Off-Gassing of Coal Combustion Products Used in Building Materials

Christopher M. Long*, Sonja N. Sax, Ari S. Lewis

Gradient, 20 University Road, Suite 5, Cambridge, MA 02138, USA

ABSTRACT

Coal combustion products (CCPs), including coal fly ash (CFA) and flue gas desulfurization (FGD) gypsum, have gained accepted use as substitutes for traditional substances in building materials. Because of the presence of mercury (Hg) in CCP materials, some concerns have been raised regarding the potential for Hg to off-gas from building materials into indoor air. Risk assessment offers a well-accepted, objective framework in which to determine whether such off-gassing could result in human exposures of health concern. We thus conducted a screening-level human health risk assessment to estimate worst-case inhalation risks from indoor air exposures to Hg for multiple CCP utilization scenarios: (1) FGD-gypsum wallboard used in a school classroom or home, and (2) CFA concrete blocks used in a school classroom. For CFA concrete, published experimental data for Hg emissions during dry curing of concrete were used to calculate a high-end emission rate, and based on extrapolation of these data, an emission rate considered to be more representative of long-term emissions. For wallboard, Hg emission rates were estimated using published data from flux chamber experiments measuring Hg volatilization from FGD-gypsum wallboard samples. Together with the estimated emission rates and conservative values for other parameters (e.g., building characteristics, exposure conditions), a steady-state indoor air model was used to predict indoor air Hg concentrations. Even for parameters intended to overstate potential exposures, predicted indoor air Hg concentrations were typically below background indoor Hg levels. Additionally, estimated Hg air concentrations were well below established inhalation toxicity criteria (noncancer hazard quotients [HQs] ranged from 0.00004 to 0.016). Based on our findings, we conclude that potential indoor air Hg exposures associated with the use of CCPs in concrete and wallboard building materials in either classroom settings or residential homes are associated with negligible human health risks.

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ARTICLE INFO

Article history: Received 16 May 2012; Received in revised form 25 June 2012; Accepted 28 June 2012

Keywords: coal combustion products (CCPs); coal ash; flue gas desulfurization (FGD); mercury; beneficial use; wallboard

1. Introduction

In 2010, approximately 50 million metric tons (or 50 Mt) of coal combustion products (CCPs), about 43% of the total generated nationwide by coal-burning power plants, were recycled in a variety of beneficial uses (American Coal Ash Association [ACAA], 2011). The beneficial use of CCPs not only conserves natural resources and reduces the amount of wastes that must be disposed

of in landfills, but many CCPs have been found to improve the quality and performance of final products. For example, coal fly ash (CFA) has gained usage as a partial replacement for Portland cement in concrete and concrete products due to benefits such as enhanced durability, reduced permeability, improved workability, and reduced greenhouse gas emissions. Flue gas desulfurization (FGD) gypsum is a high-purity substance that can be substituted for mined gypsum in the manufacture of wallboard with little, if any, additional treatment or processing other than drying. The beneficial use of CCPs has increased steadily over the past three decades, reaching a peak of approximately 45% in 2006 (ACAA,

*Corresponding author. Tel.: (617) 395-5532. E-mail: clong@gradientcorp.com

Table 1
Mercury concentrations in ingredients of traditional building materials vs. coal combustion products

Material	Total Hg concn. range (mg/kg)
Mined gypsum	0.0003–0.03 ¹
Traditional concrete ingredients (sand, cement, aggregate)	0.001–0.008 ²
FGD gypsum	0.0075–3.1 ³
Coal fly ash	<0.0025–2.3 ⁴

¹ Based on Sanderson et al. (2008), Shock et al. (2009), and EPRI (2011).

² Based on testing of two sets of concrete ingredients in Golightly et al. (2008, 2009). Golightly et al. (2008) cite other unpublished studies as reporting total Hg levels of 0.06 mg/kg in cement, 0.02–0.1 mg/kg in sand, and 0.005–0.46 mg/kg in aggregate (burnt lime). Due to the limited nature of these data, they should not be assumed to be representative of commercially available concrete ingredients.

³ Based on EPRI (2004, 2010, 2011), Kairies et al. (2006), Hassett et al. (2007), Sanderson et al. (2008), Kosson et al. (2009), Shock et al. (2009), and Gustin and Ladwig (2010).

⁴ Based on Gustin and Ladwig (2004), Golightly et al. (2009), Kosson et al. (2009), and EPRI (2010).

2011). CCP utilization rates have decreased slightly since 2006, remaining below a national goal of 50% that was set by government agencies to be achieved by the year 2010 (U.S. Department of Energy [DOE], 2006; ACAA, 2011).

CCPs contain trace levels of a number of metals that are naturally present in coal, including arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), mercury (Hg), and selenium (Se). The presence of trace metals and other contaminants has led to concerns regarding possible human exposures and potential health risks associated with beneficial use of CCPs (U.S. Environmental Protection Agency [U.S. EPA], 2011a). In response, the U.S. EPA has work under way to evaluate potential human health risks associated with beneficial reuses of CCPs (Chaney, 2011; U.S. EPA, 2011a; Rumelt, 2012).

The Hg content of CCPs is expected to increase as utilities continue to work toward greater Hg removal from flue gases to comply with more stringent air emissions limits (Northeast States for Coordinated Air Use Management [NESCAUM], 2010). As a result, specific attention has been focused on the fate of Hg in CCPs such as fly ash and FGD by-products (Electric Power Research Institute [EPRI], 2009). Mercury is generally present in coal at sub-part per million (ppm) levels as either organic and inorganic components, most commonly associated with pyrite and other sulfur-containing components of coal (Mukherjee et al., 2008). Most of the Hg from coal combustion that is not released in the flue gas ends up in either the CFA or the wet FGD gypsum, either in the form of elemental Hg (also referred to as metallic Hg) or inorganic Hg species, e.g., mercuric chloride (HgCl₂) and Hg-sulfur species (e.g., HgSO₄, HgS, Hg₂SO₄) (Mukherjee et al., 2008; Rallo et al., 2010). Table 1 shows the range in total Hg concentrations reported from testing of CFA and FGD-gypsum samples vs. those reported for traditional building materials and their ingredients. As shown in this table, reported Hg levels have been within the range of levels found in traditional building material ingredients (e.g., sand, cement, aggregates, mined gypsum), but some higher levels have also been recorded.

Due to the high volatility of elemental Hg and the possibility that it can off-gas from CCP building materials, we conducted a screening-level risk assessment focused on potential inhalation exposures and health risks from elemental Hg released from two of the more prevalent types of CCP building materials, namely, CFA concrete and FGD-gypsum wallboard. Risk assessment (hazard

identification, dose-response assessment, exposure assessment, and risk characterization) offers a well-established framework with which to objectively examine whether there is a likelihood of potential exposures and health risks associated with Hg off-gassing from CCP building materials. Indoor air Hg concentrations were modeled for three scenarios: (1) a classroom constructed with CFA concrete; (2) a classroom constructed with FGD-gypsum wallboard; and (3) a residence constructed with FGD-gypsum wallboard. Consistent with U.S. EPA guidance (U.S. EPA, 2009, 2011b), hypothetical Hg exposure levels and potential inhalation risks were predicted for each scenario based on conservative assumptions intended to overstate potential health risks for both high-end and more typical exposure conditions.

2. Methods

2.1. Selection of exposure scenarios

Hypothetical releases of Hg from CCP building materials were evaluated in both a school classroom and a single-family residence due to differences in the building materials used in these two structure types, as well as differences in building characteristics (e.g., air exchange rates, dimensions) and exposure conditions (exposure times [ETs], frequencies, and durations). Specifically, two indoor classroom scenarios were evaluated, one where the floors and walls were constructed of CFA concrete blocks and the other where the walls and ceilings were constructed of FGD-gypsum wallboard. For the residence, all walls and ceilings were assumed to be constructed of FGD-gypsum wallboard; no use of CFA concrete was considered due to the less common use of concrete blocks in single-family home construction (U.S. EPA, 2011b). For each scenario, two cases were evaluated, a case intended to represent high-end exposure conditions and one intended to reflect more typical long-term exposure conditions. For the high-end case, maximum emission rates and “upper percentile” (e.g., 95th percentile) exposure factors were generally used in the indoor air modeling and risk assessment calculations. For the more typical case, central tendency (i.e., average) values for both emission rates and exposure factors were used. EPRI (2009) contains additional details on the assessment methods, although we have since revised the analysis to incorporate new data, such as updated exposure factors based on the 2011 edition of U.S. EPA’s *Exposure Factors Handbook* (U.S. EPA, 2011b).

2.2. Estimation of Hg emission rates

2.2.1. CFA concrete blocks

Data from two recent laboratory studies (Golightly et al., 2005, 2009) that measured Hg emissions during dry curing of CFA concretes were used for estimating the emission rate of elemental Hg from in situ CFA concrete blocks. Golightly et al. (2005) measured Hg releases from curing CFA concretes for intervals of 2, 28, and 56 days at above-ambient temperatures (40°C) and for constant headspace airflow. The concretes included ordinary Portland cement (OPC) concrete and three concretes containing Class F CFA: (1) a 33% fly ash replacement concrete (FA33), (2) a 55% fly ash replacement concrete (FA55), and (3) a concrete with 33% fly ash replacement plus 0.5% Hg-loaded powdered activated carbon (HgPAC). Using a similar experimental setup, Golightly et al. (2009) conducted a follow-up study of four different 55% fly ash concrete samples (in addition to OPC), including one containing

Table 2

Mercury concentrations and release rates for various concrete mixtures

Concrete type ¹	Initial Hg concn. in concrete (mg/kg)	<i>n</i>	Hg release rate (ng/day/kg), 28 days curing	% Hg released from concrete	<i>n</i>	Hg release rate (ng/day/kg), 56 days curing
Golightly et al. (2005)						
OPC	4.1	4	0.10 ± 0.03	0.07	2	0.08 ± 0.05
FA33	9.2	6	0.26 ± 0.04	0.08		
FA55	12.6	3	0.34 ± 0.09	0.08	1	0.10
HgPAC	22.4	4	0.43 ± 0.12	0.06	2	0.11 ± 0.01
Golightly et al. (2009)						
OPC	5.2	3	0.21 ± 0.03	0.11		
CCSI (FA55 w/lignite)	5.6	3	0.62 ± 0.09	0.31		
NRT1019 (FA55 w/subbituminous + PAC)	89.3	2	0.62 ± 0.03	0.02		
MERO32 (FA55)	30.4	3	2.59 ± 0.14	0.24		1.16
MERO357 (FA55 w/subbituminous + PAC)	104.4	3	2.16 ± 0.17	0.06		1.66

¹ OPC = ordinary Portland cement concrete; FA33 = concrete with class F fly ash (33%); FA55 = concrete with class F fly ash (55%); PAC = powdered activated carbon; HgPAC = concrete with class F fly ash (33%) plus 0.5% Hg-loaded PAC; CCSI = concrete with fly ash from lignite coal-burning plant (55%); NRT1019 = concrete with fly ash from subbituminous coal-burning plant that also used PAC (55%); MERO32 = concrete with fly ash from a subbituminous coal-burning plant (55%); MERO357 = concrete with fly ash from subbituminous coal-burning plant that also used PAC (55%).

lignite and three containing subbituminous coal (two of which were from units with powdered activated carbon [PAC] sorbents used for Hg control). Importantly, these experiments demonstrated that replacement of Portland cement with CFA can be a significant source of Hg to concrete, with Golightly et al. (2009) estimating that fly ash contributed 17–96% of the Hg content of the four 55% CFA concretes.

Table 2 summarizes the Hg emission data that were collected by these researchers using headspace air measurements. Golightly et al. (2005) measured daily average Hg release rates ranging from 0.10 to 0.43 ng/kg concrete over the standard first 28 days of curing. Mercury releases followed the order: OPC < FA33 ≈ FA55 < HgPAC. Consistent with their testing of all 55% CFA concretes, Golightly et al. (2009) measured higher release rates for the standard first 28 days of curing of the CFA concretes (range, 0.62–2.59 ng/kg-day), but also a higher release rate of 0.21 ng/kg-day for OPC concrete. Air sampling data beyond the standard 28-day curing period from both sets of experiments provide support for the steady reduction in Hg release rates for FA55 and PAC down to levels exhibited by OPC concrete. Specifically, Golightly et al. (2009) observed a steady decline in Hg release rates for two 55% CFA concretes (MERO32 and MERO357, both with ash derived from subbituminous coals and the latter also containing a brominated PAC) followed over an additional 28 days of curing that they attributed to a loss of porosity and loss of water to hydration reactions. Based on extrapolations beyond the measured interval, these researchers determined that Hg release rates for CFA concretes would be equivalent to levels observed for OPC concrete for times between 10.4 and 12.3 weeks. Importantly, over the initial 28-day curing period, release of Hg from all samples from these two studies was less than 0.3% of the total Hg content, indicating the stability of Hg in the concrete.

Two Hg emission rates based on the Golightly et al. (2009) data were selected for use in the risk assessment calculations. For the high-end case, the highest measured emission rate based on a 56-day curing time (1.66 ng/kg-day, for MERO357; see Table 2) was used. For the more typical case, the OPC emission rate (0.21 ng/kg-day) in Golightly et al. (2009) was used, given that both of these studies provide evidence that Hg emissions from CFA concretes diminish over time to rates consistent with OPC. These Hg release

rates translate to 22 and 2.8 ng/m²-h, respectively, based on the use of 20-kg samples in the laboratory experiments and assuming a surface area of 0.062 m² (Golightly et al., 2005). Although we discuss later the support for the overestimation of Hg off-gassing rates from the experimental concrete curing data, it is important to highlight that both of these emission rates are based on concrete curing experiments conducted at above-ambient temperatures (40°C). Given that studies have shown an exponential increase in Hg emissions from soils with temperature (Gustin et al., 1997), both of these emission rates are likely representative of upper-bound Hg emissions from CFA concretes under typical environmental conditions.

2.2.2. FGD-gypsum wallboard

The FGD systems that are increasingly being installed for sulfur dioxide (SO₂) control in coal-fired power plants can also effectively capture oxidized Hg (NESCAUM, 2010). Although data are available demonstrating the potential for elevated Hg levels in FGD gypsum compared with mined gypsum (Table 1), few data remain available to characterize potential Hg emissions from wallboard products manufactured from FGD gypsum. Instead, recent studies have focused on leaching of Hg and other trace metals from FGD by-products (Kosson et al., 2009; Rallo et al., 2010), as well as Hg releases during the wallboard manufacturing process (Heebink and Hassett, 2003, 2005; EPRI, 2004; Kairies et al., 2006; Sanderson et al., 2008). Data from field and laboratory studies are highly variable regarding the extent of Hg losses from FGD gypsum during the manufacture of wallboard, with some data supporting losses of greater than 50% under certain calcining processes where temperatures can reach as high as 128–163°C (Sanderson et al., 2008).

More limited data were available to characterize Hg releases from FGD-gypsum wallboard, with literature searches identifying only a single study of Hg off-gassing rates from manufactured FGD-gypsum wallboard (Shock et al., 2009). While a few studies have investigated Hg releases from wet unprocessed FGD gypsum (EPRI, 2004; Hassett et al., 2007), data from these studies were not used in this assessment based on the study findings indicating that a major fraction of volatile Hg species is lost during the wallboard manufacturing process (Sanderson et al., 2008), as well as study findings indicating that drying of wet FGD gypsum can greatly

Table 3
Model input parameters for the classroom and residential home scenarios

Parameter	Units	Classroom: high-end	Classroom: typical	Residence: high-end	Residence: typical	Parameter source
Air exchange rate	h^{-1}	1.2	3	0.18	0.45	For classroom, California Energy Commission (1995); for residential home, 10th percentile and central estimate from U.S. EPA (2011b)
Classroom/home volume	m^3	231	231	971	492	For classroom, based on assumed room dimensions of $30 \times 32 \times 8.5$ ft ($9.1 \times 9.8 \times 2.6$ m); for residential home, 90th percentile and central estimate from U.S. EPA (2011b)
Wall area loading ratio	$\text{m}^2 \text{m}^{-3}$	–	–	2.18	0.95	Upper bound and typical home values from U.S. EPA (2011b)
Ceiling area loading ratio	$\text{m}^2 \text{m}^{-3}$	–	–	0.43	0.43	Value representing entire home ceiling area from U.S. EPA (2011b)
Wall and ceiling surface area	m^2	187	187	2534	679	Calculated
Concrete fraction of wall and flooring areas		1	1	–	–	Assumed
Concrete surface area	m^2	187	187	–	–	Calculated
Wallboard fraction of wall and ceiling areas		1	1	1	1	Assumed

reduce Hg release rates (Hassett et al., 2007). Using laboratory chamber flux experiments conducted at room temperature, Shock et al. (2009) determined the release of Hg from three samples of mined-gypsum wallboard and three samples of FGD-gypsum wallboard, where FGD gypsum was collected from a power plant burning blended bituminous coal and using a wet FGD system; a limestone sorbent; forced, in situ oxidation; and no fines blowdown. The maximum emission rate of $8.1 \text{ ng/m}^2\text{-day}$ ($0.34 \text{ ng/m}^2\text{-h}$) measured from the three tested wallboard products was thus used for the high-end case. For the more typical long-term emissions case, the average emission rate calculated by Shock et al. (2009) across all samples tested ($5.9 \text{ ng/m}^2\text{-day}$, or $0.25 \text{ ng/m}^2\text{-h}$) was used.

2.3. Exposure modeling

For both the school classroom and residential home scenarios, the steady-state solution to a well-established indoor air mass balance model was used to estimate the indoor air Hg concentrations corresponding to off-gassing of Hg from the CCP building materials, as shown in the following equation:

$$\text{EPC}_i = \frac{Q_{\text{concrete/wallboard}} \times \text{SA}_{\text{concrete/wallboard}}}{V_{\text{classroom/home}} \times \alpha} \quad (1)$$

where EPC_i is indoor Hg exposure point concentration (ng/m^3); $Q_{\text{concrete/wallboard}}$ is Hg emission rate from concrete ($\text{ng/m}^2\text{-h}$); $\text{SA}_{\text{concrete/wallboard}}$ is surface area of concrete in walls and floor (m^2); $V_{\text{classroom/home}}$ is room air volume (m^3); and α is air exchange rate (h^{-1}).

Such an indoor air model has been used to predict indoor air concentrations of both particles and gases emitted from a variety of different source types for situations where it is assumed that the indoor space is well mixed (Spengler et al., 2001). For this model application, infiltration of outdoor Hg was disregarded, and it was assumed that there were no other indoor Hg sources or sinks, i.e., only the indoor air contributions from off-gassing of CCP building materials were modeled.

Table 3 documents the building parameters used in the indoor air modeling for the school classroom and residential home for both the high-end and more typical exposure calculations. As

shown in this table, loading ratios (i.e., ratios of the surface area-to-volume for walls and ceilings) from U.S. EPA (2011b) were used to estimate high-end and more typical values for the surface area of wallboard in residential homes. For the school classroom, surface areas of building materials were estimated based on the classroom dimensions. For both building types, it was conservatively assumed that CCP building materials were the principal interior construction materials, with modeling of either 100% coverage of floors and walls with CFA concrete or 100% coverage of walls and ceilings with FGD-gypsum wallboard. As illustrated in Table 3, air exchange rates differed considerably between the two building types, as schools typically have mechanical ventilation systems operated to meet minimum building ventilation requirements, such as ANSI/ASHRAE Standard 62.1-2010 (American Society of Heating, Refrigerating and Air-Conditioning Engineers, Inc., 2010). Because there are abundant data showing large variation in building air exchange rates, including data indicating that schools often do not meet standards (Daisey et al., 2003), the high-end exposure calculations relied upon lower-bound air exchange rates, while the long-term, more typical exposure calculations relied on average air exchange rates. More specifically, for the school classroom, a lower-bound air exchange rate based on the mean minimum air exchange rate measured in a study of 14 California schools (1.2 h^{-1} ; as reported by California Energy Commission, 1995) was used, along with a more typical air exchange rate based on the minimum ventilation requirements for school classrooms provided in ASHRAE Standard 62.1-2010 (3 h^{-1} ; following Daisey et al., 2003). For the residential home, U.S. EPA (2011b) recommendations for central estimate and lower percentile air exchange rates (0.45 and 0.18 h^{-1} , respectively), which are based on a national analysis of air exchange rate data, were used.

2.4. Calculation of noncancer health risks

Given that Hg is not considered to be a human carcinogen, only noncancer risks were evaluated in accordance with U.S. EPA inhalation risk assessment guidance. First, an individual's hypothetical chronic exposure was estimated by adjusting the modeled indoor air Hg concentrations to reflect assumptions regarding exposure time (ET, i.e., how many hours a day an individual is

Table 4
Summary of key inputs and risk results

Scenario	EPC _i (ng/m ³) ¹	ET (h/day) ²	EF (days/yr) ³	ED (yr) ⁴	AT (h)	RfC (ng/m ³)	Time-adjusted exposure level (ng/m ³)	HQ	Approximate times lower than RfC
Concrete									
Classroom (high end)	15	9.8	260	16	140,160	300	4.4	1.5E-02	70
Classroom (typical)	0.8	7	200	16	140,160	300	0.12	4.1E-04	2500
Wallboard									
Classroom (high end)	0.23	9.8	260	16	140,160	300	0.07	2.2E-04	4500
Classroom (typical)	0.07	7	200	16	140,160	300	0.01	3.6E-05	28,000
Residence (high end)	4.9	24	350	33	289,080	300	4.7	1.6E-02	60
Residence (typical)	0.76	16	350	12	105,120	300	0.49	1.6E-03	600

¹ Calculated.

² Classroom scenario data from U.S. EPA (2011b; Table 16-17; used age group with the highest value for 50th [typical] and 95th [high end] percentiles); residence scenario data are for assumptions of either continuous residential occupancy 24 h/day (high end) or 16 h/day (typical) that reflect 8 hours spent away from the home at work or in school.

³ Professional judgment; high-end classroom scenario assumes exposure 5 days/week, 52 weeks a year; typical classroom scenario assumes exposure 5 days/week, 44 weeks (10 months) a year; residence scenario assumes 5 days/week, 52 weeks a year minus 2 weeks for vacation.

⁴ Classroom scenario data are based on assumption of entire 16 years of schooling; residence scenario data are from U.S. EPA (2011b; Table 16-5; used values associated with the 50th [typical] and 95th [high end] percentiles).

exposed), exposure frequency (EF, i.e., how many days a year an individual is exposed), and exposure duration (ED, i.e., how many years an individual is exposed), which are provided in Table 4. For both the high-end and more typical exposure cases, these exposure factors were selected based on information presented in U.S. EPA (2011b) or professional judgment when necessary. As examples of the differences between exposure factors used to represent high-end vs. more typical exposure conditions, an ET of 24 h/day and an ED of 33 years were assumed for the high-end case in the residence scenarios, but an ET of 16 h/day and an ED of 12 years were assumed for the more typical case in the residence scenarios. For both cases, where uncertainty existed, values were generally selected to overestimate exposure. The following equation shows the calculation of time-adjusted exposure:

$$\text{Time-adjusted exposure} = \frac{\text{EPC}_i \times \text{ET} \times \text{EF} \times \text{ED}}{\text{AT}} \quad (2)$$

where EPC_i is indoor Hg exposure point concentration; ET is exposure time (h/day); EF is exposure frequency (days/yr); ED is exposure duration (years); and AT is averaging time (hours).

Time-adjusted exposures were compared with the U.S. EPA Reference Concentration (RfC) for elemental Hg, calculating what is known as a hazard quotient (HQ):

$$\text{HQ} = \frac{\text{Time-adjusted exposure (ng/m}^3\text{)}}{\text{RfC (ng/m}^3\text{)}} \quad (3)$$

As defined by the U.S. EPA, an RfC is intended to represent a level of daily human inhalation exposure, experienced over the course of a lifetime, which is likely to be without an appreciable risk of deleterious effects, even for susceptible members of the population (U.S. EPA, 1993). The RfC of 300 ng/m³ for elemental Hg, which is available on U.S. EPA's Integrated Risk Information System (IRIS) website, is based on neurological effects (e.g., hand tremors, increases in memory disturbance, and slight subjective and objective evidence of autonomic dysfunction) observed in several studies of occupational exposures to elemental Hg vapor (U.S. EPA, 1995). Because this RfC is based on a chemical-specific threshold concentration (e.g., a lowest observed adverse effect level, or LOAEL, of 9 µg/m³) and incorporates multiple uncertainty factors (e.g., a 10-fold factor to account for both the protection of sensitive

human subpopulations and the use of LOAEL, and a 3-fold factor for insufficiencies in the data), noncancer health risks for elemental Hg are generally assumed to be negligible for time-adjusted exposures that are less than the 300 ng/m³ RfC, i.e., when the HQ is less than one.

3. Results and Discussion

Table 4 summarizes the predicted indoor air concentrations (EPC_i) corresponding to Hg off-gassing from CFA concrete and FGD-gypsum wallboard building materials for the various scenarios. As shown in this table, the highest indoor Hg concentrations were predicted for the CFA concrete classroom scenario, with concentrations of 15 and 0.8 µg/m³ for the high-end and more typical cases, respectively. In the FGD-gypsum wallboard classroom scenario, the modeled indoor high-end and typical concentrations were 0.23 and 0.07 µg/m³, respectively. Estimated indoor Hg concentrations for the residence using FGD-gypsum wallboard (4.9 µg/m³ for the high-end case and 0.76 µg/m³ for the more typical exposure scenario) were higher than for the classroom, due primarily to the lower air exchange rates characteristic of homes vs. schools.

Mercury is commonly present in both ambient (i.e., outdoor) air and indoor air due to a variety of major ambient sources (e.g., anthropogenic sources such as metal smelting, chlorine chemical plants, medical incinerators, cement plants, and coal-fired power plants, as well as natural sources such as volcanoes, forest fires, and geological deposits) and common indoor sources (e.g., thermometers, fluorescent lighting including compact fluorescent light bulbs [CFLs], electrical switches, barometers, gas regulators, and batteries) (Agency for Toxic Substances and Disease Registry [ATSDR], 1999; Carpi and Chen, 2001; Valente et al., 2007). As a result, additional perspective on the significance of potential Hg exposures from the use of CCP building materials can be gained by comparing the modeled Hg concentrations for the various scenarios with background levels of airborne Hg. As presented in Figure 1, even the highest modeled indoor air Hg concentration of 15 ng/m³ (observed for the high-end case in the CFA concrete classroom scenario) is well within the range of background levels reported in indoor air studies (Carpi and Chen, 2001). In addition,

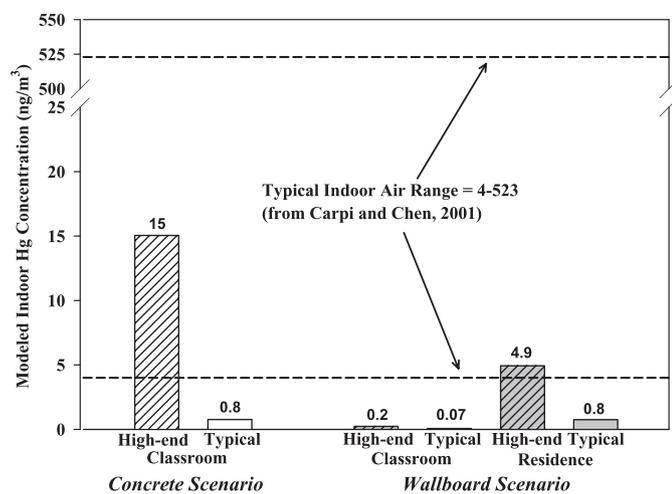


Fig. 1. Modeled indoor concentrations of Hg off-gassed from CCPs in concrete and wallboard compared with measured background indoor Hg concentrations (note the break in the y-axis).

the modeled concentrations are not significantly higher, and are generally lower, than average outdoor Hg levels reported for urban and rural areas, e.g., mean elemental Hg concentrations of 2.7–4.6 ng/m³ as reported by Carpi and Chen (2002) for New York City neighborhoods, and a mean elemental Hg concentration of 1.70 ng/m³ (SD = 0.172 ng/m³) as reported by Valente et al. (2007) for 22 rural/remote sites.

Table 4 also presents the calculation of time-adjusted Hg exposure levels, which were approximately 3 to 6 times lower than the modeled indoor Hg concentrations for the classroom scenarios and up to 1.6 times lower for the residence scenarios. In addition, Table 4 summarizes all estimated noncancer health risks, showing that calculated risks for all exposure scenarios are associated with negligible risk as determined using U.S. EPA risk assessment methods, i.e., all HQs were at least two orders of magnitude below one. This is also illustrated in Figure 2, which shows that time-adjusted exposures are all well below the U.S. EPA RfC.

We generally relied upon model parameters and assumptions (e.g., U.S. EPA-recommended central estimate and 95th percentile exposures factors, U.S. EPA RfC for elemental mercury) deliberately chosen to be conservative (i.e., health-protective) and thus biased towards the overestimation of potential exposures and health risks. However, as with any risk assessment, there are some sources of uncertainty with unknown impacts on the estimated exposures and health risks. For example, exposure calculations considered only the inhalation pathway, although it is unlikely that any significant exposure would result from other exposure pathways (e.g., dermal contact or incidental ingestion) for in situ CCP building materials given the typical presence of protective coatings, such as paint or epoxy, which would effectively limit any direct contact with CCPs in these materials.

Hg off-gassing rates were estimated from limited laboratory data, including two studies of Hg emissions during concrete curing and a single study of Hg fluxes for finished wallboard. While limited in number, these data are for direct measurements of Hg emissions from finished materials that are likely to represent conservative estimates of Hg off-gassing rates due to experimental conditions that served to increase Hg fluxes (e.g., concrete curing at elevated temperature, unpainted wallboard samples, high

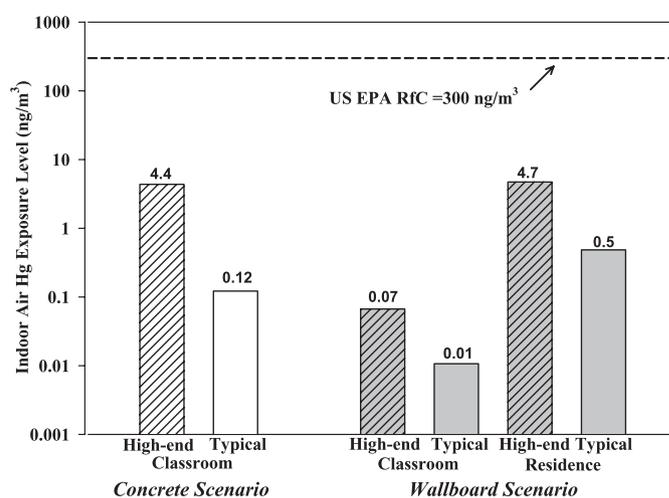


Fig. 2. Comparison of time-adjusted estimates of indoor air exposure levels of Hg off-gassed from CCPs used in concrete and wallboard with the U.S. EPA reference concentration (RfC) for elemental Hg (note the log scale of the y-axis).

airflows). In addition, there were notable differences in the properties of the CFA building materials used in these laboratory studies from those typically used in building construction projects. In particular, the Golightly et al. (2005, 2009) concrete curing experiments used concrete samples with high CFA replacement rates (55% of Portland cement; note that typical CFA replacement rates for Portland cement in concrete are up to 30%, but in some applications can be as high as 50%; U.S. DOE, 2006) and CFA with a relatively high Hg content (1.4 mg/kg). Additionally, the Shock et al. (2009) laboratory study of Hg volatilization from FGD-gypsum wallboard products used newly manufactured wallboard samples that would be expected to have greater Hg off-gassing rates than aged samples. Although this study tested FGD-gypsum wallboard samples having a relatively low total Hg content (0.15 ± 0.02 mg/kg), some difference between the total Hg content of finished FGD-gypsum wallboard products and that of raw FGD-gypsum feedstock (Table 1) is expected due to Hg losses during the wallboard manufacturing process (e.g., during drying, calcining, and forming steps). Lastly, both sets of experimental studies used purified (i.e., Hg-free) air. Although the use of Hg-free air would normally be expected to contribute to enhanced Hg fluxes, it is likely that this purified air was also ozone-free; given that laboratory experiments have shown that ozone is linked with increased Hg emissions from natural Hg-bearing substrates (Engle et al., 2005), the impact of the use of purified air on Hg off-gassing rates is thus unclear.

4. Conclusions

To our knowledge, our screening-level risk assessment of potential indoor air Hg inhalation exposures and health risks associated with CCP-containing building materials is one of few available quantitative risk evaluations for CCP beneficial-use applications. Recognizing the need for such scientific analyses, U.S. EPA announced in late 2011 its plans to conduct a health risk assessment covering various CCP beneficial-use applications (Rumelt, 2012). Our risk assessment indicates that off-gassing of Hg from CCPs in concrete and wallboard building materials is unlikely to result in indoor Hg exposures of potential health concern in either classroom settings or residential homes. This

conclusion is based on high-end exposure scenarios that employed conservative assumptions for modeling indoor air Hg concentrations and calculating time-adjusted exposure concentrations. Moreover, modeled Hg air concentrations for each scenario were within the range of background Hg concentrations in both outdoor and indoor settings. Additional studies would help to further characterize Hg off-gassing rates from CCP-containing building materials and to monitor any changes to Hg levels in CCPs resulting from continued efforts to improve the capture efficiencies of stack gas contaminants. On the other hand, this assessment provides support for a large margin of safety between worst-case indoor air Hg exposures associated with CCP-containing building materials and potential adverse health risks.

Acknowledgments

Funding for this risk evaluation was provided by the Electric Power Research Institute (EPRI), Palo Alto, CA. The authors would like to specifically thank Ken Ladwig, the EPRI Project Manager.

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