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Exposure analysis of accidental release of mercury from compact fluorescent lamps (CFLs)

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HIGHLIGHTS

- The critical period for intake covers the first 4 h after the CFL breaks.
- The room air temperature affects significantly the intake rate.
- Potential exposure and uptake are significantly higher for infants/kids due to specific behavior.
- Simple risk reduction measures (if followed in the right order) can minimize the potential health risk.

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ABSTRACT

Mercury release after breakage of compact fluorescent lamps (CFLs) has recently become an issue of public health concern, especially in the case of early life infants. Preliminary, screening type calculations have indicated that there is potential for increased intake of mercury vapor by inhalation after breakage of a CFL.
Several experimental and computational studies have shown that, when modeling the breakage of a CFL, the room space must be segregated into different zones, according to the potential of mercury vapor to accumulate in them after accidental release. In this study, a detailed two-zone model that captures the physicochemical processes that govern mercury vapor formation and dispersion in the indoor environment was developed. The mercury fate model was coupled to a population exposure model that accounts for age and gender-related differences in time–activity patterns, as well as country differences in body weight and age distribution. The parameters above are used to determine the intake through inhalation (gas phase and particles) and non-dietary ingestion (settled dust) for each age, gender group and ethnicity.
Results showed that the critical period for intake covers the first 4 h after the CFL breaks and that room air temperature significantly affects the intake rate. Indoor air concentration of mercury vapor may exceed toxicological thresholds of concern such as the acute Reference Exposure Limit (REL) for mercury vapor set by the Environmental Protection Agency of California. Ingestion intake through hand-to-mouth behavior is significant for infants and toddlers, counting for about 20% of the overall intake. Simple risk reduction measures including increased indoor ventilation followed by careful clean-up of the accident site, may limit dramatically the estimated health risk.

1. Introduction

Compact fluorescent lamps (CFLs) were introduced in the global market in response to the need to reduce energy consumption within the wider framework of rational energy use and minimizing carbon dioxide emissions (Parsons, 2006). The constantly increasing domestic use of compact fluorescent lamps (CFLs) is promoted by approximately 75% reduction in energy usage and 10-fold increase in lifetime relative to incandescent bulbs. As a result, their use is highly recommended as a replacement option for incandescent bulbs by many federal and local government agencies (Johnson et al., 2008). However, an important disadvantage of fluorescent lamps is that CFLs contain milligram (mg) quantities of mercury. Mercury is a human toxicant metal found throughout the environment. It is an essential component for a range of household devices and very fundamental for the operation of fluorescent lights (Carpi and Chen, 2001).

Because of the frequent use of mercury in household equipment there is a considerable amount of research concerning the relevance of the source to indoor air pollution. In order to decrease several environmental issues caused by the use of mercury another chemical should substitute it. As this has not yet been found the amount of

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mercury used in fluorescent lamps is being controlled and eventually has decreased during the years.

CFLs are widely used as their environmental benefits in contrast to incandescent bulbs that outweigh potential health risks. Internationally, concerns have been raised regarding potential mercury releases and consequently human exposures following CFL breaking and more in particular to elemental mercury. According to Nance et al. (2012) the dominant exposure pathway to humans from broken CFLs is inhalation. A percentage of 80–97% of the inhaled elemental mercury which is readily dispersed in the air is absorbed in the body through the lungs.

According to the latest study carried out by Nance et al. (2012), once a CFL breakage event occurs, mercury vapor, liquid mercury (if present) and mercury adsorbed onto the phosphorous powder is released (NJ-MTF, 2002). The amount of the mercury is significantly small (up to 5 mg) and any mercury spilt would form droplets, so it is highly impossible that any spilled liquid mercury will be visible. Once the lamp is broken the phosphorous powder can be separated from the glass (NJ-MTF, 2002). The amount of mercury which will be released in the form of vapor or powder is highly dependent on the initial quantity of the mercury in the lamp and the age of the lamp. Fluorescent lamps contain several forms of mercury according to what is added by the manufacturer and how it has changed throughout the years (UNEP, 2005). As the bulb ages, the elemental mercury in the lamp will be oxidized and will form inorganic mercury compounds, predominantly HgO (Aucott et al., 2004). More than 80% of the mercury becomes a component of the phosphorous powder and approximately 14% can be diffused through the glass matrix (Jang et al., 2005; Rey-Raap and Gallardo, 2011). New lamps release more mercury vapor in contrast to older ones or to spent ones, where the mercury has been oxidized or distributed into the lamp components. There is an initial spike in air-borne mercury concentration after the breakage of a CFL or a linear fluorescent tube as mercury vapor is released (Aucott et al., 2004), followed by slower release of mercury present in solid and liquid forms (amalgams, liquid elemental mercury, inorganic mercury and mercury absorbed onto lamp components).

Several studies are based on toxicological information and worst-case scenarios for exposure to mercury. The report delivered by Groth (2008) for the Illinois Environmental Council, evaluated the risks posed by accidental breakage, re-evaluating the calculations carried out by Chandrasekhar (2007) and all relevant toxicological information available to the date the report was published. Although the report did not provide any original contribution to the field, it was a very comprehensive review describing qualitatively the events following a CFL breakage, the associated risks and prevention measures, based on the current state of the art.

The study carried out by Chandrasekhar (2007) modeled the dispersion of mercury in the air in a room after a CFL was broken. The assumptions taken were that all 5 mg of mercury in the CFL were in vapor state and that the volume of air in the room was about 33 m^3. The mercury vapor concentration in the room a few minutes after a CFL was broken was measured and found to exceed 150 $\mu g/m^3$. Although this represents the ever worst-case scenario, in practice, this is not likely to occur.

According to Aucott et al. (2004) and Johnson et al. (2008), lamp breakage is accompanied by an initial release of about 5% of the overall mercury amount, while the rest is progressively released over time. This assumption is also supported by an extensive research done by Stahler et al. (2008), where the maximum concentration observed was 54 $\mu g/m^3$, but at a vertical distance of 30 cm over the lamp breakage. In addition, experiments carried out by the same investigators (Stahler et al., 2008) proved that lamps working for 1 h (thus being very hot) had no significant differences in the initial amount released. This leads to the conclusion that the ambient temperature is more significant for the vaporization of mercury released after the lamp breakage (Aucott et al., 2004).

Moreover, Stahler et al. (2008) showed that concentrations were significantly lower at the height of 1.5 m, indicating that the assumption of "uniform" concentrations within a room (at least for the first minutes following the breakage) may be misleading, as it tends to underestimate concentrations close to the breakage surface and to overestimate concentrations for the rest of the room. A further implication of the non-uniformity within the room is related to exposure. Different age groups have different breathing zones and respiration rates as a function of their body height. For instance, after the breakage of a CFL on the floor infants and children are more vulnerable to mercury vapor exposures than adults because of their smaller body size and their rapid respiration rates which allow them to inhale a larger dose of mercury.

Although all the above studies provide valuable experimental data of mercury indoor air concentrations following a CFL breakage, their link to actual exposure and intake is limited. This is due to the following facts:

- Different media partitioning is either qualitatively described (particles) or neglected (dust). However, these media provide additional exposure pathways and routes, which are usually not described within these studies, even though they can be significant for exposed individuals at early developmental stages.
- Experimental studies have strong financial and time limitations. As a result, the number of combinations of the affecting parameters such as emissions, and housing conditions (room ambient temperature, volume, air exchange rates) is limited by the actual experimental settings, while the same occurs for the exploitation of prevention measures and prioritization.

Based on the above, in the current study, a refined exposure assessment following a CFL breakage event is carried out, incorporating a more detailed temporal and spatial resolution and especially highlighting the actual intake differences between adults and infants/children. The assessment is greatly facilitated by an integrative computational framework that incorporates all the parameters affecting the intake associated to an accidental release, including emissions (e.g. ambient temperature), concentrations (e.g. room volume, incorporation of zones, air exchange rate), exposure (e.g. breathing zone, time spent indoors, associated pathways) and intake (e.g. breathing rate, bodyweight and non-dietary dust ingestion), within a probabilistic exposure assessment framework.

2. Methodology

The overall study relies on the simulation of exposure scenarios after an accidental breakage of a CFL, based on the emission scenario reported by Aucott et al. (2004). The breakage is assumed to occur within a typical residential room and the assumptions made (with regard to release rates, room volume and ventilation) are rather conservative. They may thus be useful for the preliminary risk assessment that is required to correctly apply the precautionary principle.

2.1. Emissions

The time course of the mercury releases from broken CFLs containing 4.55 mg mercury for three different temperatures used in our assessment are illustrated in Fig. 1.

The results observed by the study which was carried out by Aucott et al. (2004) were used for the simulation of a broken lamp scenario. The study done by Aucott et al. (2004) described the rate of release of elemental mercury from 4-ft spent CFLs which contained either 4.4 or 4.7 mg of mercury. During the experiments the CFLs were broken inside a 32 gallon (146 L) plastic container and the concentration of mercury vapor inside the plastic container was identified. The experiments were done under three room temperatures for each trial which 4.4 °C, 15.6 °C and 29.4 °C respectively. The conclusions

concerning the emissions were similar to those observed by Johnson et al. (2008): a rapid release rate was initially observed, followed by a declining release rate. Aucott et al. (2004) attributed the decline of the release rate to two factors; to a gradual release of less volatile forms of mercury and to the oxidation of mercury. The results showed that the mercury release rate depends on temperature. This was expected because of the higher volatility of mercury at higher temperatures. Although most of the volatile mercury released from the bulbs is elemental, other volatile mercury compounds and powders may also be released (Nance et al., 2012). As these amounts are not well known, they represent a source of uncertainty for the assessment of releases. Aucott et al. (2004) highlighted in his study that the mercury releases which come from disposal and recycling of fluorescent lamps, were reported by other regulatory bodies to vary between 1% and 80%. Specifically, Aucott et al. (2004) reported that a US Environmental Protection Agency (EPA) model indicated 6%; an industry report estimated 1%; and another author estimated a range of 20–80%. In any case, the amount of mercury already contained in fluorescent lamps is an additional source of uncertainty, as this depends on the wattage and the brand of the bulb (Stahler et al., 2008).

For the calculations of the accidental brake scenario, the release curves reproduced by Aucott et al. (2004) will be used, considering a room temperature of 20 °C. The release curve was estimated using data from Aucott et al. (2004) through non-linear regression, with an R^2 of 0.97. A differential equation (Eq. (1)) was derived, where the rate of emission dm/dt is related to vapor pressure V_p (0.00196 mm mercury cooled sub-liquid), ambient temperature T and the initial amount A of mercury in the bulb and m is the amount of mercury emitted.

$$\frac{dm}{dt} = \left(V_p \cdot T^{2.6} \cdot (A-m)^{1.5} / (0.0061 \cdot A + m)\right) \cdot 0.74 \quad (1)$$

Within a very short time after the breakage, 180 μg of mercury was initially released (e.g. 1 s), as stated by Aucott et al. (2004). This initial release was always added to the emission profile derived by the above formula.

The release curves derived by Aucott et al. (2004) are considered conservative enough for calculations related to public health protection, since the release of mercury vapor from the broken lamp is considered to occur until all the amount contained in the lamp is finally vaporized.

2.2. Mercury fate upon release

Emission curves are already described in the previous sector and depicted in Fig. 1 for different room temperatures. A typical room of 27 m^3 was considered as the location of the CFL breakage event. For the needs of the assessment done similarly to the methodology followed by Stahler et al. (2008), the room was divided in two zones: the lower one which has a height of 30 cm from the floor (zone 1) and the higher zone (zone 2) which covers the rest of the height of the room, from 30 cm until the roof. This distinction was done since significantly higher concentrations of mercury releases are expected immediately after the breakage event near the floor. Thus, a two-zone model gives a more realistic depiction of the dispersion process instead of assuming all the room as a homogenous space. Moreover, the lower zone is considered to be the breathing zone for infants and young children. All the calculations used below are carried out taken this differentiation into account. To model these processes, the following formulation (Eqs. (2) and (3)) of a two zone model was used (Pepper and Carrington, 2009):

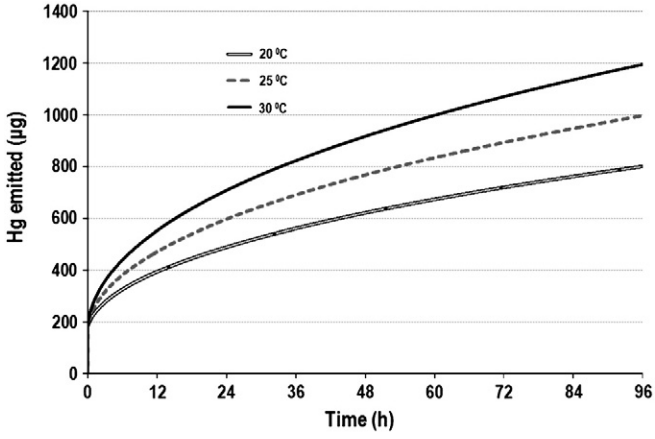

Fig. 1. Amount of mercury released for the first 96 h following the breakage of a fluorescent lamp containing 4.55 mg mercury under different room temperatures.

$$V\frac{dC_1}{dt} = Q(C_a - C_1) + E_1 - kC_1V_1 + Q_{1-2}(C_2 - C_1) \quad (2)$$

$$V\frac{dC_2}{dt} = Q(C_a - C_2) + E_2 - kC_2V_2 + Q_{1-2}(C_1 - C_2) \quad (3)$$

where:

E_1, E_2	the strength of the emission sources (mass/time)
V_1, V_2	the volumes of the respective zones
C_a	the outdoor concentration
C	the indoor concentration of the respective zone
Q	the indoor–outdoor air exchange rate
Q_{1-2}	the air exchange rate between the zones
k	the decay rate of the substance in the examined microenvironment

The air exchange rate between the two zones was considered equal to 60 $m^3 \cdot h^{-1}$. This value matches the experimental data of Stahler et al. (2008). No air exchange was assumed between the low zone and the outdoor environment, while emissions occurred only in the low zone, where the breakage event takes place.

For a more comprehensive analysis, the partitioning of mercury to particles and settled dust was taken into account. Although metallic mercury is characterized by relatively high vapor pressure (0.00196 mm mercury cooled sub-liquid), still it is not so high to consider partitioning to airborne particles and in-house dust negligible. According to Weschler et al. (2008), the equilibrium between the gas phase and the surface of the airborne particles is described by an equilibrium constant referred to as the particle–gas partition coefficient, K_p, which describes the fraction of the compound that is adsorbed on the particles (per mass of particles). K_p is well described by the following empirical formula (Eq. (4)), reported by Naumova et al. (2003), based on over 1800 partition coefficients for PAHs measured outdoors and indoors in three US cities:

$$\log K_p = -0.860 \cdot \log p_L^0 - 4.67. \quad (4)$$

The K_{dust} coefficient which describes the partitioning between the airborne phase (including gaseous phase and particles) and the settled dust is calculated empirically based on actual measurement data. More in detail, the fraction between airborne and dust phase of phthalates from actual environmental samples (Fromme et al., 2004) was correlated to their vapor pressure. These data were fitted by non-linear regression ($R^2 = 0.9759$), and the K_{dust} is estimated by the following formula (Eq. (5)):

$$K_{dust} = 0.0063 \cdot \left(p_L^0\right)^{-0.561} \quad (5)$$

Based on the above, the concentrations of gaseous, particles and dust phase are given by Eqs. (6)–(8) respectively.

$$V\frac{dC_{chem\_gas}}{dt} = E_{chem\_gas} - Q_{ind\_out}\cdot\left(C_{chem\_gas} - C_{chem\_gas\_out}\right)\cdot V - k\cdot C_{chem\_gas}\cdot V - \left(C_{chem\_gas} - \frac{C_{chem\_PM}}{K_p\cdot C_{PM}}\right)\cdot V - \left(\left(C_{chem\_gas} + C_{chem\_PM}\right)\cdot V - \frac{C_{chem\_dust}\cdot m_{\_dust}}{K_{\_dust}}\right) \quad (6)$$

$$V\frac{dC_{chem\_PM}}{dt} = \left(C_{chem\_gas} - \frac{C_{chem\_PM}}{K_p\cdot C_{PM}}\right)\cdot V - Q_{ind\_out}\cdot\left(C_{PM} - C_{PM\_out}\right)\cdot\frac{C_{chem\_PM}}{C_{PM}}\cdot V \quad (7)$$

$$V\frac{dC_{chem\_dust}}{dt} = \left(\left(C_{chem\_gas} + C_{chem\_PM}\right)\cdot V - \frac{C_{chem\_dust}\cdot m_{\_dust}}{K_{\_dust}}\right) \quad (8)$$

Where,

E_{chem_gas} chemical emission rate
Q_{int_out} Indoor/outdoor air exchange rate
K chemical decay coefficient
K_P gas/particle partition coefficient
K_{dust} gas/dust partitioning coefficient
V location volume
C_{PM} *PM concentration indoors*
C_{PM_out} PM concentration outdoors
C_{CHEM_gas} chemical concentration in gas phase
C_{CHEM_PM} chemical concentration in PM phase
C_{CHEM_dusts} chemical concentration in dust phase
$m_{\_dust}$ mass of dust in the location

For the needs of the assessment an average total PM concentration of 40 μg/m^3 was considered as representative for a typical residential dwelling (Sarigiannis et al., 2012), while for dust a typical concentration of 0.000001 g/m^2 was taken into account (Myatt and Macintosh, 2008). With regard to air exchange rates (outdoor to indoor and vice versa), the conclusions of a recent study done for Europe by Dimitroulopoulou (2012) reported that the ventilation rate of the majority of European dwellings is approximately 0.5 h^{-1} and rates greater than 0.5 h^{-1} and up to 1.5 h^{-1}were reported in the Netherlands and in some Mediterranean countries like Greece and Portugal. Air exchange rates used in the study of the typical room are given as follows (Opasnet, 2011):

- 0.5 h^{-1} for the child room until 3 years old for all countries.

For age groups above 3 years:

- 0.6 h^{-1} for UK, Ireland
- 0.8 h^{-1} for Northern, Central and Western Europe
- 1 h^{-1} for France
- 1.3 h^{-1} for Spain, Greece, Malta, Cyprus, Italy and Portugal.

2.3. Exposure and intake

Personal exposure is equal to the average concentration of a pollutant that a person is exposed to over a given period of time. If over the given period of time, T, the person passes through n locations, spending a fraction f_n of the period T in location n where the concentration of the pollutant under consideration is C_n, then the personal exposure for this period T, represented by the concentration C_T, is given by (Ott, 1982) (Eq. (9)):

$$C_T = \sum_n f_n\cdot C_n. \quad (9)$$

Inhalation intake (Eq. (10)) was estimated by the area under the curve of mercury exposure E multiplied by the inhalation rate inh_n for each type of microenvironment n encountered, divided by the bodyweight BW and for the desired simulation time.

$$Intake_{inh} = \frac{\sum_n E_n\cdot inh_n}{BW} \quad (10)$$

Different time activity patterns (Tables 1–4 of the Supplementary material) were used for the several age groups (Opasnet, 2011), while inhalation rates (with respect to age specific differences and intensity according to the activity) were obtained from the ICPR (2002) report (see Supplementary material). Bodyweight values with respect to age and ethnicity (see Supplementary material) were also taken from Opasnet database (Opasnet, 2011).

Intake from dust ingestion was taken into account by non-dietary ingestion, through the hand-to-mouth behavior. Similarly to above (Eq. (11)),

$$Intake_{ing} = \frac{C_{dust}\cdot m_{dust}}{BW} \quad (11)$$

where C_{dust} is the concentration in dust and m_{dust} is the amount of dust daily intake. The daily intake of dust is age dependent. Daily ingestion for dust is estimated to be about 0.05, 0.01 and 0.001 g of dust for infants/toddlers, children and teens/adults respectively (Wormuth et al., 2006).

2.4. Simulation set-up and executions

All simulations were executed using the computational platform INTERA (INTERA, 2011). This computational platform was conceived, designed and developed by the first three authors (Sarigiannis et al., 2011). It is specialized for addressing aggregate and cumulative exposure from indoor sources. The platform allows for dynamic calculations in time, and incorporates probabilistic (Monte-Carlo) and sensitivity analysis. The probabilistic framework allows the incorporation of uncertainty and variability across the source to dose calculation which is reflected at the final outcome. The same occurs for the sensitivity analysis, allowing the identification of the relative importance of the determinants to exposure and overall intake. In the sensitivity analysis module, an input parameter value is changed by 1% and the relative change in outcome is calculated when all other input parameters are fixed.

Intake from inhalation and non-dietary ingestion for the several age groups was estimated by Monte-Carlo simulation. The Monte Carlo method involves a large number of samples (20,000 iterations were executed for each calculation) from the distributions of the input parameters. Prior distributions (Table 1) with regard to bodyweight, inhalation rate, air exchange rate, temperature and initial amount release were propagated, so to derive distributions of intake. For each age group and exposure route four simulations were executed, regarding exposure for 1 to 4 days respectively. Beyond the Monte Carlo simulations, country specific calculations (based on average country representative data) were also executed.

3. Results and discussion

3.1. Concentration of mercury following the breaking event

The concentration time profile for the two zones (with regard to mercury in gaseous phase) within the room is illustrated in Fig. 2. Mercury concentration at the lower zone tends to increase rapidly following the breakage event, up to 60 μg/m^3 and then tends to decline due to mixing with air from the higher zone and an exchange with the outdoor air. In the higher zone the peak is significantly

Table 1
Prior distributions for the Monte Carlo analysis.

Input parameter	Age group	Distr.	Min	Max	Mean	Std. dev.
Bodyweight	0 to 1	*Normal*	6.1	8.1	7.1	0.5
	1 to 3	*Normal*	11.1	12.3	11.7	0.3
	3 to 8	*Normal*	19.4	23.0	21.2	0.6
	8 to 14	*Normal*	38.9	44.7	41.6	1.5
	14+	*Normal*	66.8	75.4	71.3	2.2
Inhalation rate	0 to 1	*Normal*	0.12	0.30	0.20	0.04
	1 to 3	*Normal*	0.23	0.30	0.27	0.02
	3 to 8	*Normal*	0.23	0.67	0.43	0.12
	8 to 14	*Normal*	0.23	1.22	0.69	0.26
	14+	*Normal*	0.34	1.22	0.77	0.24
AER	All age	*LogNormal*	0.23	2.20	0.92	0.21
Temperature	groups	*LogNormal*	15.0	30.0	23.3	4.3
Initial release		*Normal*	2761	6046	4547	495

lower (up to almost 5 μg/m^3) and shortly (within 3 h), the concentrations tend to equilibrate. This behavior is similar to the one reported by Stahler et al. (2008). However, unlike Stahler et al. (2008), in the present simulations an immediate cleanup process is not considered. Instead the mercury concentration profile is examined under the assumption that the broken lamp remains in the room for some time. Concentration of mercury adsorbed in particles remains very low in both zones (about 10 times lower than the gaseous phase), while concentration in dust is up to 120 μg/g_dust, declining at a slower rate than indoor air concentrations.

3.2. Exposure and intake based on the broken lamp scenario

The inhalation exposure profiles follow the concentration profile in the way implied by the activity patterns (Fig. 3). The exposure of infants–children until the age of three is much higher compared to the rest of the age groups during the first hour of the lamp breakage event, since this age group is considered to breathe from the lower zone of the room. After the first hour, concentrations between the zones tend to become equal, thus exposure diurnal variability is determined by the time–activity patterns.

Beyond inhalation exposure (which is the major pathway), overall intake (inhalation and settled dust ingestion) was investigated. The results of cumulative intake (four day exposure scenario) are presented in

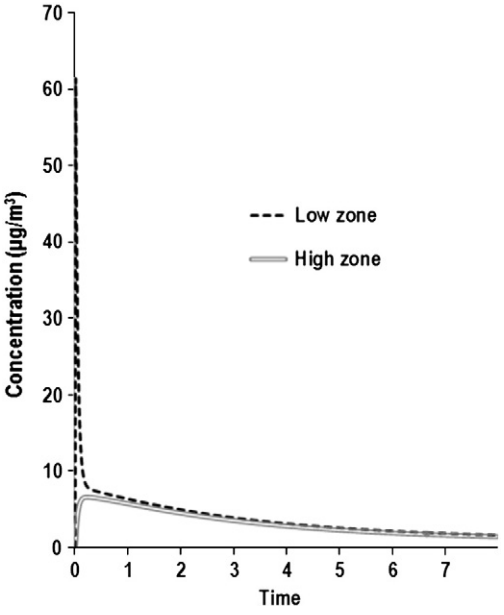

Fig. 2. Gaseous phase mercury concentration time profile for the low and high zones of the room following the breaking event.

the form of probability distribution plots (Fig. 4a to e), for the five distinguished age groups (0 to 1, 1 to 3, 3 to 8, 8 to 14 and 14+) and the relevant exposure routes. Mercury intake through ingestion, was found to be significant mostly for infants and toddlers (about 20% of the overall intake). In total, the overall (bodyweight normalized) intake for infants and toddlers is up to 4 times higher compared to the other groups.

The variability of intake reflects the variability of the prior determinants, indicating that intake might vary up to 5–6 times depending on anthropometric parameters, the amount of vapor mercury initially released and the housing conditions. The relative effect of these parameters was investigated and the results of the sensitivity scores are illustrated in Table 2. Bodyweight, inhalation rate and amount of dust ingested seem to have an almost linear effect in terms of route uptake, while initial amount of mercury released and AER affect in a smaller degree the overall intake.

From the distribution plots, it is easily observed that the rate of intake is declining from day 1 to day 4. The latter is clearly illustrated in

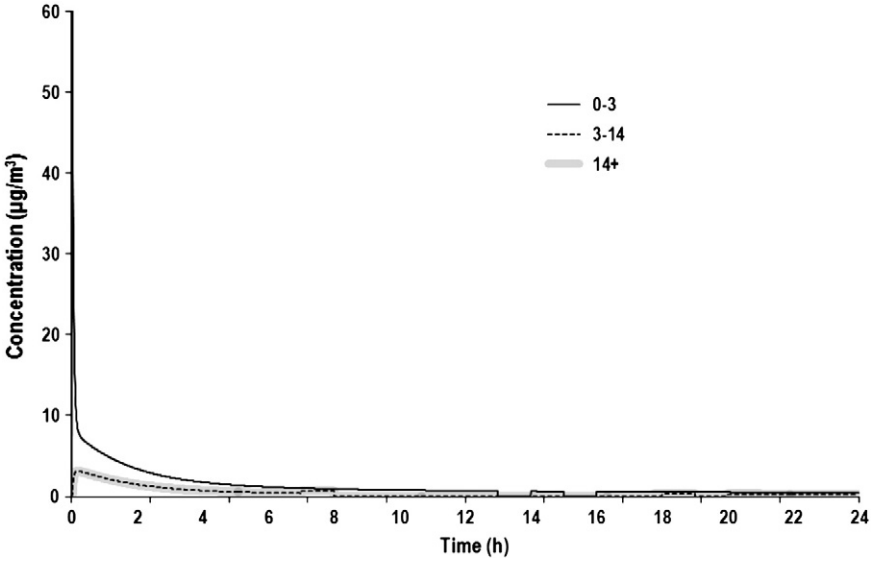

Fig. 3. Exposure profile of the clustered age groups for the first 24 h following the lamp breakage event.

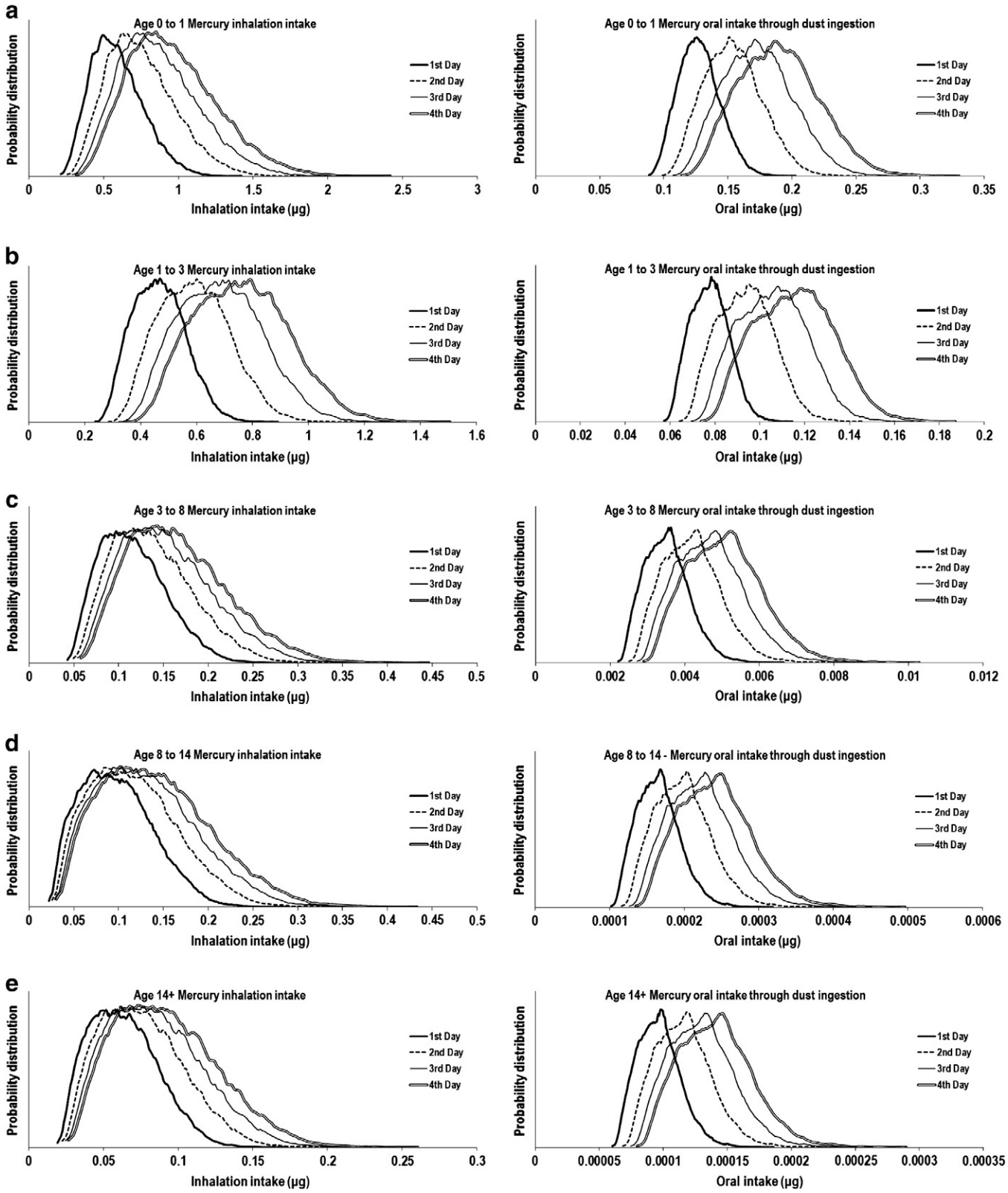

Fig. 4. Probability distribution plots of cumulative intake under a four day exposure scenario for the different age groups (a to e) and exposure routes (inhalation and dust non-dietary ingestion).

Fig. 5 (illustrating the daily mercury intake for infants), the results highlighting the importance of the first day after the breaking event compared to the daily intake of the following days. This is the reasonable consequence of the rapidly declining concentrations after the first moments of the breakage event, followed by the initial release.

Although room temperature was incorporated in the Monte Carlo analysis, the effect was further investigated on a typical infant exposure scenario, under three different temperatures (20, 25 and 30 °C), keeping constant all other model parameters. The results are illustrated in Fig. 6, showing that a rise in room temperature from

Table 2
Sensitivity analysis scores.

BW	Inhalation rate/dust ingestion	Initial release	Air exchange rate	Temperature
−1.0	1.0	0.3	−0.7	0.8
−1.0	1.0	0.3	−0.7	0.8

20 to 30 °C contributes to an increase in mercury intake (at the four day scenario) of about 50% (Fig. 6), the effect being more pronounced right after the lamp breaking event. This is due to the fact that elevated room temperature increases the release of vapor mercury which is attached on the broken CFL pieces. Thus the room temperature is more important, the later the removal of broken CFL pieces occurs. Despite the fact that a temperature of 30 °C is not recommended as a room temperature, the scenario might be occasionally realistic for southern European/Mediterranean countries (e.g. Greece, Cyprus, Southern Italy, Spain, Portugal), if very high ambient temperatures (being a common occurrence during the summertime) are combined with the absence of any air-conditioning equipment in indoor dwellings.

For the same age groups, variability among countries is limited; it is found to be of secondary importance compared to age-specific differences, reflecting the variability of average bodyweight and air exchange rate. The detailed results of cumulative intake assessment (for the 4 day exposure scenario) across EU-27 and for all age groups are illustrated in Fig. 1(a to d) in the Supplementary material.

3.3. Health risk potential assessment

The most relevant toxicological threshold that might facilitate the needs of this assessment is the one proposed by the Office of Environmental Health Hazard Assessment (OEHHA) of the California Environmental Protection Agency. OEHHA has derived an acute Reference Exposure Level (REL) of 1.8 μg/m^3 for a 1-h average exposure to mercury vapor (OEHHA, 2007). The acute REL of 1.8 μg/m^3 is based on behavioral deficits measured in rats following in utero exposure to metallic mercury vapor (Danielsson et al., 1993). The lowest-observed adverse-effect level (LOAEL) was 1.8 mg/m^3 for behavioral deficits in the offspring. The 1-h concentration determined by OEHHA was also 1.8 mg/m^3. The Acute REL (1-h) of 0.0018 mg/m^3 (1.8 μg/m^3) was derived using an uncertainty factor of 1000 (10 for use of a LOAEL, 10 to extrapolate from rats to humans, and 10 for intra-species differences) (Nance et al., 2012).

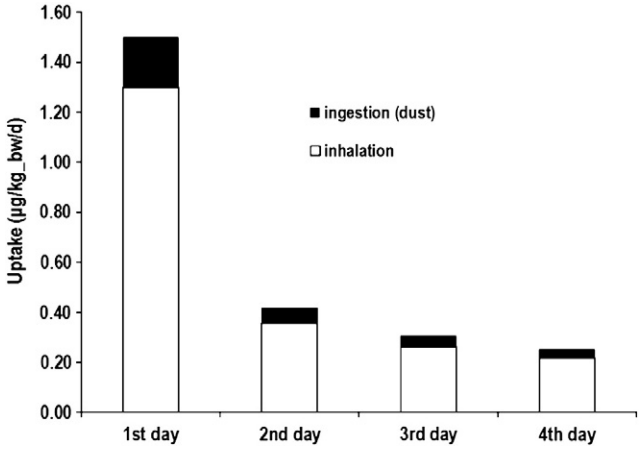

Fig. 5. Daily intake of mercury for a typical infant (7 kg_bw) after the CFL breaking event.

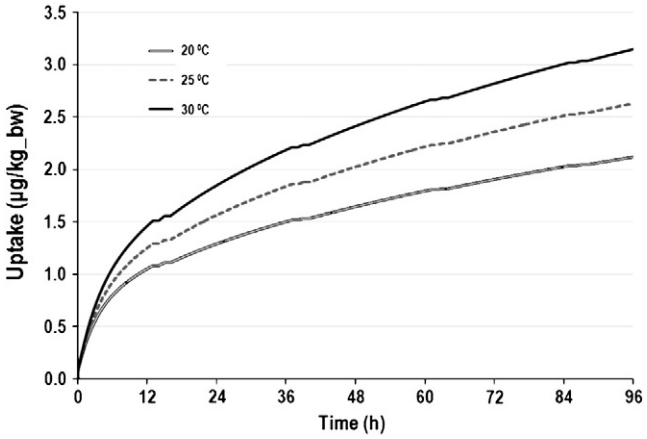

Fig. 6. Cumulative intake for an infant under three different room temperatures (20, 25 and 30 °C).

Comparing the calculated concentrations in the executed scenarios to the above threshold indicates that according to our calculations, average hourly concentrations may remain above the threshold for the first 3–4 h, if no protective measure is taken (e.g. opening of windows in order to increase the indoor/outdoor air exchange rate). Hence, although the calculations are based on the rather conservative mercury release scenario estimated by Aucott et al. (2004), for precautionary reasons, it is found that prompt response for limiting the contamination by increasing the air exchange would be expected to significantly reduce exposure and consequent health risks (Groth, 2008).

3.4. Exploring the preventive scenarios

As shown from the results mercury vapors can remain in the room at least till the 23rd hour in the highest zone and till the 31st hour after CFL breakage at the lower zone in case it is not removed. Therefore it is strongly recommended to remove the mercury as soon as possible. This is the worst case scenario. Major factors governing indoor air decontamination from mercury after a bulb breakage event are the magnitude and the duration of ventilation. Ventilation is very important as it is considered to be the most effective way to clean the indoor atmosphere from pollutants. Therefore, as the air

Table 3
All scenarios investigated for the study.

Scenarios	Air exchange rate (h^{-1})	Removal of pieces of broken glass
1	0.5	No
2	0.5	Immediately
3	0.5	After 3′
4	0.5	After 6′
5	0.5	After 15′
6	4.5 for 120′ from the 3rd min	Immediately
7	4.5 for 30′ from the 10th min	After 30′
8	4.5 for 60′ from the 10th min	After 30′
9	4.5 for 60′ from the 3rd min	After 30′
10	4.5 for 60′ from the 1st min	After 30′
11	4.5 for 120′ from the 3rd min	After 3′
12	1.5 for 60′ from the 3rd min	Immediately
13	1.5 for 60′ from the 3rd min	After 30′
14	1.5 for 120′ from the 3rd min	After 30′
15	1.5 for 120′ from the 3rd min	After 15′
16	1.5 for 120′ from the 3rd min	After 3′
17	1.5 for 120′ from the 6th min	After 3′
18	1.5 for 120′ from the 15th min	After 3′
19	2.5 for 120′ from the 3rd min	After 3′
20	2.5 for 120′ from the 1st min	After 3′
21	3.5 for 120′ from the 3rd min	After 3′

exchange is essential for healthy indoor environments, it is necessary that all buildings are equipped with devices for natural or mechanical ventilation. The air exchange rate differs with respect to the temporal variations, the type of the building and several other factors.

According to Johnson et al. (2004) study to identify the air exchange rate in a residence in Columbus, the 66 hourly-average AERs calculated varied from 0.36 to 15.8 h^{-1}. Thirty nine AER values were measured when all exterior doors and windows were closed and so they varied from 0.36 to 2.29 h^{-1}. The other 27 AER values which varied from 0.50 to 15.8 h^{-1} were calculated while at least one door or window was open. Additional experimental work was carried out by Daghigh et al. (2009) in an office room, where various combinations of window/door openings were investigated. The results from both studies mentioned above are synopsized in the Supplementary material. This review was necessary, in order to build-up scenarios that incorporate realistic AER cases for typical residential building settings.

Preventive actions related to ventilation increase aim to reduce the indoor air concentration of airborne mercury below the Acute Reference Exposure Level (REL) proposed by OEHHA, set to 0.6 μg/m^3. Under this objective, several scenarios related to the magnitude and the duration of ventilation, as well as to the removal of broken CFL pieces were investigated. In total, 22 different scenarios were simulated as shown in Table 3.

The results indicated that the higher the ventilation rate is and the time left (Tables 4 and 5) for the room to be ventilated, the quicker the concentrations of mercury decline. More in detail, when the air exchange rate is equal to 4.5 h^{-1} for 2 h, the concentration of mercury drops below the safety threshold within the second hour. A crucial conclusion derived from the investigated scenarios was that the removal time of the broken pieces does not significantly affect the overall concentrations and consequently exposure and intake. As illustrated in Fig. 7, for a constant ventilation rate of 1.5 h^{-1} for 2 h and different removal times of 30, 15, 3 min or immediate removal respectively, there was no considerable change in the decline rate of mercury concentration within the room. On the contrary, ventilation increase is shown to be much more efficient, as demonstrated in Fig. 8, where in two scenarios, broken CFL piece removal takes place after 30 min, while ventilation rate is increased to 4.5 h^{-1} and 1.5 h^{-1} respectively. In the case of higher ventilation increase, mercury concentration drops below the safety threshold within the 2nd hour, while in the other case within the 5th hour. These observations are very well explained by the mercury emission profile following the CFL breaking event, where a strong emission "pulse" occurs immediately after (Fig. 1). Since this immediate release is linked to the breaking event, thus unavoidable, it is expected that the initial release is determinant for the media concentration profile. Thus, removing (by increasing the ventilation) the mercury already dispersed by the initial release, is far more important than removing the potential continuous but low emission source (remains of broken CFL).

4. Concluding remarks

In this study we simulated computationally the intake dynamics of mercury found in the indoor environment after a CFL breaks. Given the significant differences in physiological and anatomical parameters affecting inhalation patterns, especially when coupled with differences in time–activity patterns among different age groups and nationalities, a detailed exposure analysis was undertaken to appropriately estimate the potential exposure/intake levels. The main results are summarized below:

- Considering that the breathing zone of infants and children is within the lower zone (30 cm), where mercury concentrations are relatively high, there is significantly higher exposure potential for this age group.
- A significant amount of the overall intake occurs within the first hours of the breakage event.
- A 10-degree rise in indoor air temperature could result to up to 50% higher mercury vapor emission during the first 4 h after lamp breakage. This scenario is likely in areas of southern Europe where very high ambient air temperatures can be observed in the summertime.

Cleaning up carefully the lamp remaining and eventually the mercury drops left over and opening the windows (so as to increase the indoor:outdoor air exchange rate) may significantly reduce the potential exposure. Stahler et al. (2008) have evaluated several clean-up and ventilation scenarios related to mercury fluorescent lamp breakage, providing the following suggestions, which are further quantified and strengthened by our simulations with regard to the preventive scenarios:

- To open immediately all windows/outdoor doors possible
- To leave the area/room and wait 15–30 min after breakage and before cleaning up (mercury levels in the air will have fallen from their highest levels by then);
- To use a glass container, metal screw top lid with a seal, such as a canning jar, to contain the lamp pieces, powder, and cleanup materials;
- Immediately remove the lamp breakage from the home once containerized, especially if the homeowner did not have a glass container with a good seal;
- Continue to ventilate the room for several hours.

From the methodological point of view, the current study demonstrates that experimental studies like the ones carried out for assessing the potential risks of CFL breakage (Salthammer et al., 2012; Stahler et al., 2008) could be greatly facilitated by an integrative computational platform that allows detailed exposure scenario analyses. These advantages are summarized as follows:

- Computational studies are a nice complement to environmental media analysis. Most of the experimental studies are carried out by identifying the contamination of indoor air in the gaseous

Table 4
Vapor mercury concentration (μg/m^3) under three scenarios with AER equal to 4.5 h^{-1} for several durations of increased ventilation.

1st scenario		2nd scenario		3rd scenario	
Ventilation	Removal	Ventilation	Removal	Ventilation	Removal
4.5 for 30′ from the 10th min	After 30′	4.5 for 60′ from the 1st min	After 30′	4.5 for 120′ from the 3rd min	After 3′
Time (hours)	High zone	Low zone	High zone	Low zone	High zone	Low zone
1st	2.37	5.82	1.63	4.70	1.73	4.80
2nd	0.94	0.96	0.06	0.07	0.06	0.07
3rd	0.63	0.64	0.005	0.005	0.004	0.005

Table 5
Vapor mercury concentration (μg/m^3) under three scenarios with AER equal to 1.5 h^{-1} for several durations of increased ventilation.

4th scenario		5th scenario		6th scenario	
Ventilation	Removal	Ventilation	Removal	Ventilation	Removal
1.5 for 60′ from the 3rd min	After 30′	1.5 for 120′ from the 3rd min	After 30′	1.5 for 120′ from the 15th min	After 3′
Time (hours)	High zone	Low zone	High zone	Low zone	High zone	Low zone
1st	3.9	7.3	3.9	7.3	4.0	7.0
2nd	1.8	1.9	1.3	1.4	1.3	1.4
3rd	1.2	1.2	0.5	0.6	0.5	0.5
4th	0.8	0.8	0.4	0.4	0.3	0.3

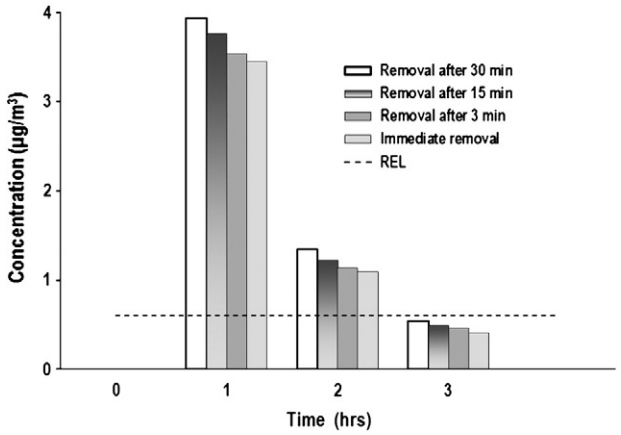

Fig. 7. Comparing three different cases when the ventilation is 1.5 per hour for two consecutive hours and the time of the broken CFL piece removal is different for each event.

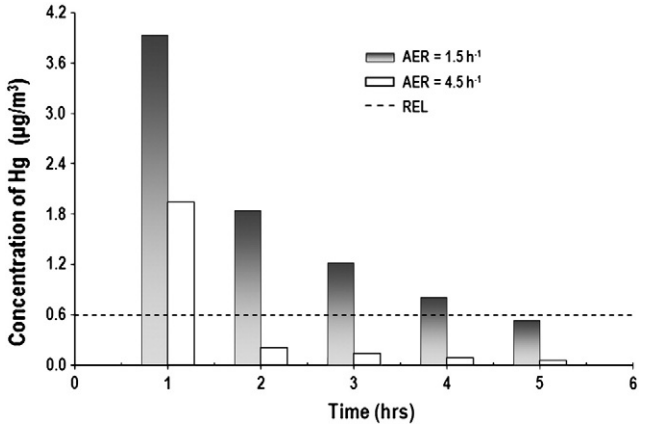

Fig. 8. Two cases with the same time of the broken CFL piece removal and ventilation but different air exchange rates.

phase. The computational platform allows the estimation of concentration in additional environmental media (particles, settled dust), reducing the overall cost of experimental procedures. The later could be further minimized if the combined use of measurements and modeling tools is incorporated in the initial study design. Experimentation should be targeted, so as to check the validity of the predictions made by the computational platform, rather than being the sole assessment tool.

- The accidental release can be translated into actual intake calculations through the assignment of exposure mechanisms and the application of multi-pathway and multi-route exposure models. The latter is very important, as it gives us the capability to refine exposure for different age groups (differences in exposure pathways) and different housing conditions.
- Exploring the prioritization/efficiency of preventive scenarios. A computational platform allows the simulation of a very large number of preventive scenarios, requiring a significantly lower cost (in terms of actual cost, human resources and time consuming) than experimentation. In addition, specific preventive scenarios might not be supported for execution by existing experimental settings.

Thus, either as stand-alone tool, or as a complement to experimentation, a computational platform clearly accelerates the identification of potentially harmful exposure scenarios, increasing thus the cost-efficiency of risk management. Detailed understanding of the accidental conditions that might pose some harm from the CFL use and of the most efficient prevention measures, could be decisive for the future position in the market of CFLs as consumer product. In the case of CFLs, even following a conservative emission/dispersion scenario, it seems that labeling the preventive measures in the product would minimize the risks posed by accidental events, thus increasing the overall safe use of the product.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2012.07.026.

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