

Assessment of Modeled Indoor Air Concentrations of Particulate Matter, Gaseous Pollutants, and Volatile Organic Compounds Emitted from Candles

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INTRODUCTION

The majority of candles are generally produced using paraffin waxes refined during hydrogenation processes that remove most of the impurities normally present in raw paraffin. The increase in paraffin prices and retail sales over the last few years has led to the market introduction of candles intended for indoor use and made from non-hydrogenated paraffin waxes. These candles may raise some health concerns because the combustion of poorly refined raw waxes could emit high levels of air pollutants, such as particulate matter (PM), polycyclic aromatic hydrocarbons (PAH), and sulphur dioxide (SO₂).

In previous studies, benzene, toluene, ethylbenzene, xylenes (BTEX), PAH, aldehydes, nitrogen oxides (NO_x), carbon monoxide (CO), polychlorinated dibenzop-dioxins (PCDD), polychlorinated dibenzofurans (PCDF), and particulate matter emissions from candle burning were measured. Lau *et al.* (1997) concluded that calculated indoor concentrations of PCDD, PCDF, benzo(a)pyrene (BaP), and aldehydes were well below the recommended limits. Nevertheless, they estimated indoor concentrations under extreme-use scenarios (*i.e.*, the burning of 30 candles) and compared them to occupational exposure limits. Other studies have suggested that predicted indoor BaP concentrations may be within the same order of magnitude as the respective recommended values (Derudi *et al.* 2012). Several studies examined the impact of emissions from candles with metal wick cores on the concentrations of Pb and Ni in the combustion fumes, with conflicting results. In particular, mean Pb emissions from tall-form candles with Pb wick cores were 0.5 µg/h (van Alphen 1999), while another study measured greater Pb emissions (on average, 770 µg/h) and estimated a Pb indoor mean concentration of 9 µg/m³, with a maximum level of 42.1 µg/m³ (Nriagu and Kim 2000), which could pose possible health risks for the general population.

Indoor concentrations of PM_{0.3-10} exceeded the guidelines for PM₁₀ in churches where candle burning is allowed (Loupa *et al.* 2010). Therefore, candle burning may constitute an important contribution to the background concentrations of indoor PM, which can itself be high in residential environments (Cattaneo *et al.* 2011). The emission of PM from candle burning led to a potential exposure of 0.68 mg h m⁻³ in a restaurant scenario (Fan and Zhang 2001). Emissions of PM were affected by different modes of candle burning; they were approximately 20 times greater under unsteady burning conditions compared to steady-state burning (Zai *et al.* 2006). Another study reported low mass emission rates and particles smaller than 100 nm in diameter in the “normal burning” mode, high emission rates in the “sooting” mode, and high emission rates and particles’ diameters between 400 and 800 nm in the so-called smoldering phase (Fine *et al.* 1999). The steady burn phase emits predominantly ultrafine particles that mainly consist of phosphates and alkali nitrates most likely originating from flame retardant additives in the wick. These findings are in agreement with other studies of other indoor sources, such as incense sticks and mosquito coils, whose combustion generates particles with size distribution modes between 80 and 240 nm (Roy *et al.* 2009). Conversely, the “sooting” mode burn emits larger particles predominantly composed of elemental carbon, while particles emitted during the “smoldering” phase are dominated by organic compounds (Pagels *et al.* 2009). Some experimental studies conducted in indoor environments or large environmental chambers have reported PM_{2.5} mass

concentrations between 0.012 and 0.063 mg/m³ (Lee and Wang 2006) and ultrafine particle concentrations between 10⁴ and 10⁶ particles/cm³, depending, again, on the burning mode (Pagels *et al.* 2009; Glytsos *et al.* 2010).

As summarized in a recent study, VOC emissions from scented candles varied from 1 µg/g to dozens and hundreds of µg/g for acetaldehyde and formaldehyde, respectively (Maupetit and Squinazi 2009). Two- to four-ring PAHs (particularly naphthalene) dominated the relative emissions from scented candles, while 5-ring PAHs accounted for the largest fraction of the total PAH emitted from other decorative candles (Orecchio 2011; Derudi *et al.* 2012).

Literature data for NO₂ and SO₂ emissions from candle burning are scarce (Derudi *et al.* 2014), and NO₂ indoor concentrations in medieval churches with massive candle burning can reach approximately 75 ppb (Loupa and Rapsomanikis 2008).

The goal of the present study was to evaluate indoor concentrations of chemical contaminants emitted into atmosphere through the combustion of candles in residential scenarios. A simple risk assessment approach was performed on the basis of the estimation of indoor air pollutant concentrations in theoretical residential premises by using a reasonable worst-case exposure scenario (RWCS). These values were then compared to regulatory or guideline limits.

METHODS

Test Candles and Selected Air Pollutants

Three different raw materials used for the production of candles and five different scented candles (scented with cedarwood, plumeria, rhubarb, opium, and aloe vera fragrances) were selected for study. Among the raw materials, paraffin (50/1), intermediate (55/9), and slack (65/6) waxes were investigated. The first part of the abbreviations refers to the melting point (°C) of the material, whereas the second part indicates the oil content (wt%). These differences in melting point and paraffin oil content are linked to different phases of the refining process.

The emission factors of the following airborne pollutants were determined for each tested candle: BTEX, aldehydes (acetaldehyde, formaldehyde, acrolein and acetone, butyraldehyde, benzaldehyde), PAHs (naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, chrysene, dibenzo(a,h)anthracene, benzo(g,h,j)perylene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, BaP, indeno(1,2,3-cd)pyrene), CO, NO_x, sulphur dioxide (SO₂), and PM. A combustion chamber was purposely designed and developed to determine the emission factors under standardized experimental and steady-state burning conditions. The sampling and analytical methods for the determination of the emission factors were presented in detail in Derudi *et al.* (2012) and Derudi *et al.* (2013).

Risk Assessment

The evaluation of indoor concentrations was performed only on those air pollutants considered toxicologically relevant and of potential concern after a preliminary comparison between estimated indoor concentrations and the guideline limits recommended for indoor or general life environments. A toxicological survey was first

Table 1. Guidelines for the selected pollutants.

Pollutant	Guidance value	Type (averaging time)	Reference
formaldehyde	0.1 mg/m ³	Acute (30 min)	WHO (2010)
acrolein	0.7 µg/m ³	Acute (8-h)	OEHHA (2008)
acetaldehyde	0.2 mg/m ³	Acute (15 min)	Kotzias <i>et al.</i> (2005)
benzene	5 µg/m ³	Chronic (1-y)	EU (2000)
naphthalene	3 µg/m ³	Chronic (1-y)	USEPA (1998)
BaP	1 ng/m ³	Chronic (1-y)	EU (2004)
CO	10 mg/m ³	Acute (8-h)	WHO (2010)
NO ₂	200 µg/m ³	Acute (1-h)	WHO (2010)
SO ₂	350 µg/m ³	Acute (1-h)	EU (2008)
PM _{2.5}	25 µg/m ³	Acute (24-h)	WHO (2006)

conducted to select regulatory and guideline limits (Table 1) for risk assessment purposes, applying the following selection criteria, which are listed by priority. This hierarchy does not correspond with well-established protocols and is proposed based on common sense.

- The regulatory limits or guideline values recommended by international institutions were preferred to those recommended by institutions with national or regional coverage.
- The guidelines for indoor air pollutants were preferred over others.
- Where available, acute (<8 h) and subacute (≤24 h) reference levels were preferred over chronic (lifetime) recommended levels, due to the assumption that candles are sporadically burned in residential homes during a period of a few hours per day, so that exposure is generally limited in time.
- Under the same conditions previously specified, the most conservative limit was selected in compliance with the precautionary principle.

The only exception to the listed selection criteria was the guideline value for acrolein. The U.S. Environmental Protection Agency's (USEPA's) reference concentration (RfC) for acrolein is 0.02 µg/m³ (USEPA 2003). This RfC was derived from the results of an early bioassay in laboratory rats, but it was recently updated by the California Office of Environmental Health Hazard Assessment using current bioassay data (OEHHA 2008) to derive a chronic reference exposure level of 0.35 µg/m³. The same report also indicated a reference exposure level for 8-h acute exposures (0.7 µg/m³), which was used here for risk assessment purposes because chronic reference exposure levels would be highly conservative and not realistic in the case of air emissions from candles, as previously stated.

After this preliminary screening, the following pollutants were selected for further evaluation: formaldehyde, acrolein+acetone, acetaldehyde, benzene, naphthalene, BaP, CO, NO_x, SO₂, and PM. The selected regulatory and guideline limits listed in Table 1 were used for comparison to the predicted indoor concentrations of these airborne pollutants, which were estimated using a single-compartment mass balance tool from previously measured emission factors (Derudi *et al.* 2012, 2013).

A RWCS was assumed, in agreement with the ReACh guidelines, to evaluate the possibility of health risks even in unlikely circumstances (Ahrens and Traas 2007; EChA 2008). This scenario was developed using the following assumptions: simultaneous combustion of four candles for 4 h in an indoor environment with a volume of 25 m³, which corresponds to that of a small bathroom, and ventilation of 0.1 air changes per hour (ACH). It should be noted that a consumer survey revealed that more than 80% of candle users typically burn four or fewer candles at one time for four or fewer hours (Ökometric 2007). Moreover, 0.1 ACH can be assumed as the minimum ventilation of residential homes (Chao *et al.* 2004; Gilbert *et al.* 2008). The RWCS is by definition a rare but not impossible exposure scenario. In most of cases, candles are infrequently burned in domestic environments but multiple burning may quite common. As an example, the simultaneous burning of four candles for some hours may be needed to perform normal domestic activities in black-out conditions. The burning of scented candles could also be a frequent event depending on personal behaviors. Therefore, the choice of the RWCS conditions is somehow subjective but it seems to be in accordance with the ReACh guidelines (“unlikely circumstances”).

The modeling was conducted assuming that particulate and gaseous pollutants are emitted at a constant release rate, the weight fraction of the compound in the total product corresponded to the emission factors measured in Derudi *et al.* (2012) and Derudi *et al.* (2013) and that the total product amount is entirely emitted in 4 hours. The emission rates of air contaminants were determined using an experimental chamber (Derudi *et al.* 2012). The temperature in the sampling stack was in the 30–42°C range during the entire experimental time. The temperature on chamber walls and candle external surfaces was always between 22 and 30°C. The total PM mass was considered to be PM_{2.5}, conforming to previous findings (Derudi *et al.* 2012) and a precautionary approach.

Indoor concentrations resulting from scented candle burning were estimated using the average emission factors and average burning rates measured by Derudi *et al.* (2012) because the difference in emissions among candles was small. Conversely, indoor concentrations that originated from raw material burning were estimated using the specific emission factors and burning rates described in Derudi *et al.* (2013).

Indoor concentrations were estimated using a single-compartment mass balance model assuming uniform mixing (Eqs. (1) and (2)). Equation (1) was applied until the end of candle burning, which was assumed to be 4 h; Eq. (2) was used over the remaining exposure period.

$$t \leq t_{rel}$$

$$C(t) = \frac{E}{V(\alpha + k)} [1 - e^{-(\alpha+k)t}] \quad (1)$$

$$t > t_{rel}$$

$$C(t) = C(t_{rel}) [1 - e^{-(\alpha+k)t}] \quad (2)$$

where $C(t)$: concentration of the compound in the room’s air (g m⁻³), $C(t_{rel})$: concentration of the compound in the room’s air at the end of the emission period (g m⁻³), E : emission rate (g s⁻¹), V : room volume (m³), α : ventilation rate (number of air changes per time unit) (s⁻¹), k : decay constant (s⁻¹), t : time (s).

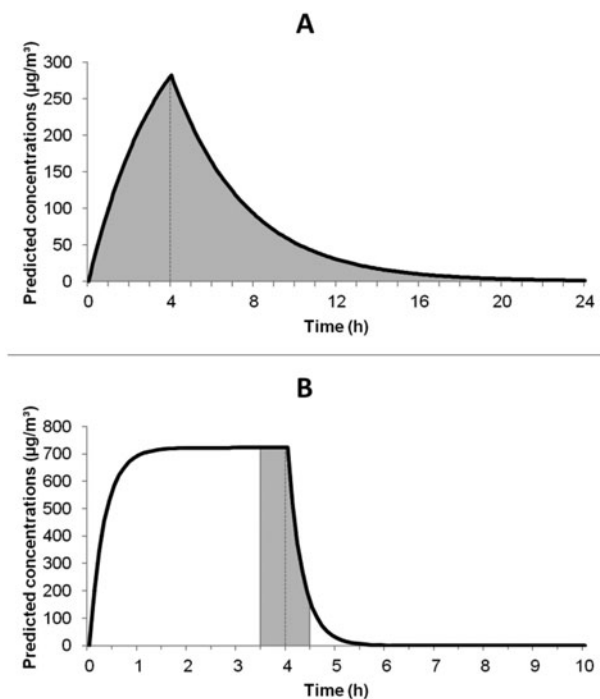


Figure 1. Instantaneous predicted concentrations used to obtain integrated indoor $\text{PM}_{2.5}$ (A) and SO_2 (B) concentrations for comparison with reference values. Worst-case scenario with four burning candles (65/6 type), ventilation of 0.1 Ach and room volume of 25 m^3 . Dashed line = end of emission. Filled area = averaged concentrations (24-h averaging time for $\text{PM}_{2.5}$; 1-h integration for SO_2).

The indoor concentrations that were compared to the limits for the protection of human health were calculated by integrating the instantaneous predicted values over the exposure times reported in Table 1 and centering the integration interval on the end of the emission period, in accordance with a precautionary approach. Provided in Figure 1 are examples for $\text{PM}_{2.5}$ (24-h averaging time) and SO_2 (1-h averaging time).

RESULTS

The estimated concentrations of formaldehyde, acetaldehyde, benzene, BaP, and naphthalene did not exceed the recommended or reference values listed in Table 1, even considering the RWCS. The impact of some behavioral and structural variables on modeled indoor concentrations is depicted in Figures 2 and 3 for pollutants whose indoor concentrations were near or above guideline or regulatory limits. Depicted in Figure 2 are the variations in the estimated concentrations of aldehydes and PM emitted by one and four scented candles as a function of volume and ventilation of rooms. Estimated indoor concentrations of formaldehyde and acetaldehyde

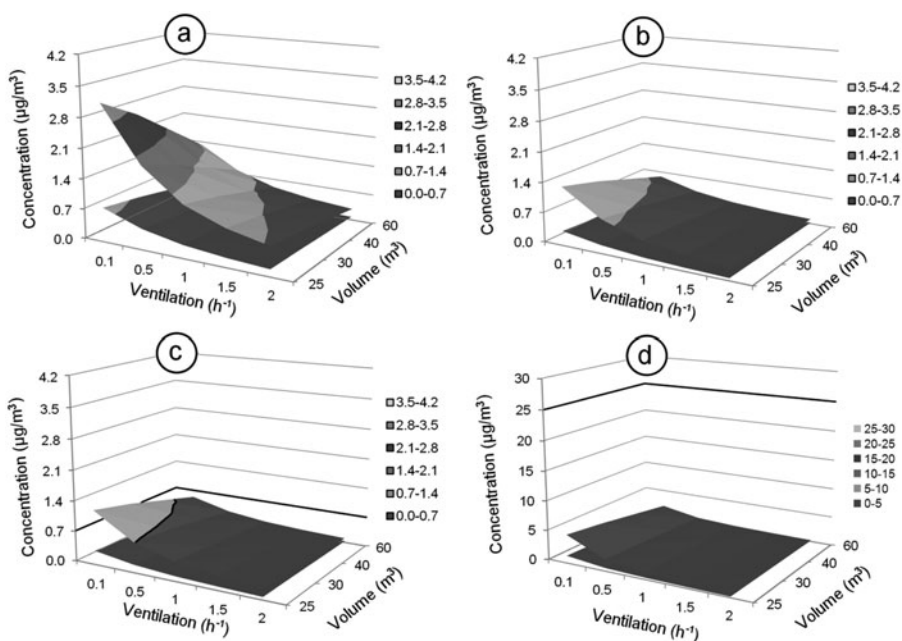


Figure 2. Predicted concentrations of formaldehyde (A), acetaldehyde (B), acrolein+acetone (C) and PM_{2.5} (D) emitted by one (lower surface) and four (upper surface) scented candles. Red lines = guideline values. Acrolein guidance value is not directly comparable to cumulated acetone and acrolein predicted concentrations.

were well below the guideline threshold concentrations (about 30 and 200 times, respectively) (Figures 2A and 2B).

The estimated concentrations of acrolein+acetone in the RWCS exceeded the recommended level for acute exposures to acrolein ($0.7 \mu\text{g}/\text{m}^3$) (Figure 2C). In the RWCS, the average concentration of acrolein+acetone was approximately two times greater (Table 2) than the corresponding recommended level. However, it should be noted that threshold levels have been proposed to protect human health against the irritating effects of acrolein ($0.7 \text{ mg}/\text{m}^3$ for decreased respiratory rate, $0.13 \text{ mg}/\text{m}^3$ for eye irritation, and $0.3 \text{ mg}/\text{m}^3$ for nasal irritation) are three orders of magnitude greater than the estimated RWCS concentration (WHO 1992). Moreover, the emission factors from candle burning seem to be characterized by high variability in the acetone/acrolein ratio, approximately in the range 0.3–3.5 (Lee and Wang 2006). Thus, the predicted indoor concentrations of “true” acrolein should be divided by a factor of 1.3–4.5 with respect to those values reported in Figure 2C.

The predicted indoor concentrations of PM were approximately 5 times less than the World Health Organization’s (WHO) threshold level (Figure 2d). Illustrated in Figure 3 are the estimated concentrations of NO_x, SO₂, and PM, deriving from the combustion of different raw waxes as a function of room volume and ventilation. These contaminants were selected among those reported in Table 1 because they

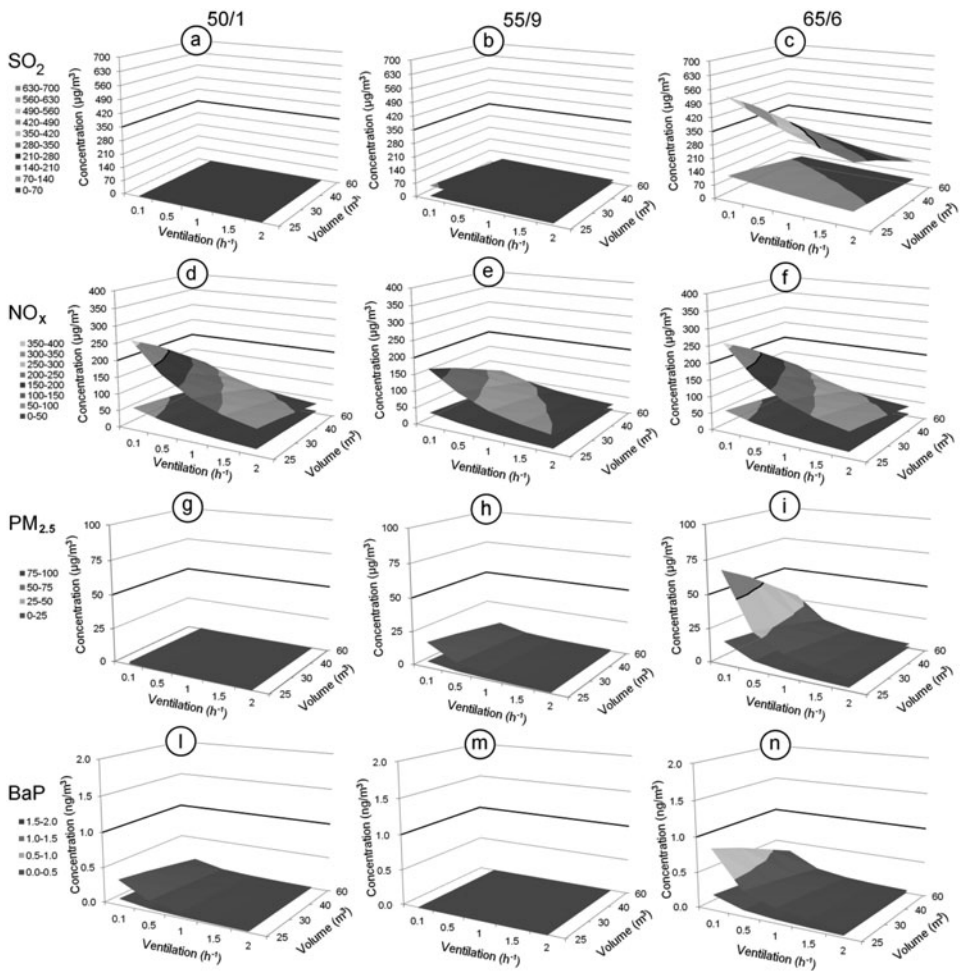


Figure 3. Predicted concentrations of SO₂ (A–C), NO_x* (D–F), PM (G–I), and BaP (L–N) for 50/1 (A, D, G, L), 55/9 (B, E, H, M), and 65/6 (C, F, I, N) waxes as a function of room volume and ventilation. Comparison between scenarios with one (lower surface) and four (upper surface) burned candles. Red lines = guideline values**. *Expressed as NO₂. **Nitrogen dioxide (NO₂) guidance value is not directly comparable to NO_x predicted concentrations because candle burning tends to generate NO_x emissions characterized by a predominant amount of NO with respect to NO₂.

may reach indoor levels that exceed their respective recommended concentrations, while all of the other studied pollutants were at least one order of magnitude less.

The indoor PM 24-h concentrations predicted based on the 65/6 wax burning emissions exceeded the recommended guideline for PM_{2.5} (25 µg/m³) only under RWCS conditions (Figure 3I), while the 24-h indoor concentrations modeled for the 50/1 and 55/9 waxes were largely below the WHO guideline (Figures 3G and

Table 2. Model's parameters.

Pollutant	Emission rates (mg/h)				k(h ⁻¹) —References
	SC	50/1	55/9	65/6	
formaldehyde	11.3×10^{-3}	NC	NC	NC	0.36—Nazaroff and Cass (1986)
acrolein	7.51×10^{-3}	NC	NC	NC	0.33*
acetaldehyde	3.94×10^{-3}	NC	NC	NC	0.23—Liu <i>et al.</i> (2006)
BaP	NC	3.95×10^{-6}	9.5×10^{-9}	9.29×10^{-6}	0.18**
NO _x	NC	1.71	1.12	1.70	0.8—Spicer <i>et al.</i> (1993)
SO ₂	NC	0.0140	2.03	14.9	3.2—USEPA (2009)
PM _{2.5}	51.6×10^{-3}	0.0195	0.201	0.733	0.18—Lai (2002)

*The decay rate of acrolein was assumed to be the mean of those estimated in two recent studies (Liu *et al.* 2006; Seaman *et al.* 2009). **The decay rate of BaP was assumed to be equal to that of PM due to the predominant partitioning in the particulate phase rather than in the gaseous phase. NC = Not considered due to very low concentrations in screening models.

3H). The PM_{2.5} guideline was used as a threshold value for risk assessment for the following reasons:

- The particulates emitted from the selected candles (particles with an aerodynamic diameter generally less than 0.5 μm) are totally included within this size range (Derudi *et al.* 2014);
- No recommended thresholds for PM fractions characterized by aerodynamic diameters less than 2.5 μm have been proposed; and
- The PM_{2.5} guidance value is health-based, accounting for an estimated increase in mortality of approximately 0.5% for each 10 μg/m³ increment in the daily concentration of PM_{2.5} and PM₁₀ (WHO 2006).

Modeled indoor concentrations of CO did not exceed or approach the 8-h limit of 10 mg/m³ recommended by the WHO indoor air quality guidelines (WHO 2010). Conversely, 1-h NO_x concentrations estimated from the emission factors of all three tested waxes were comparable to the WHO guideline value for NO₂ (200 μg/m³) (WHO 2010) only in the worst-case scenarios for all the three tested candle waxes (Figures 3D–3F). Nevertheless, it should be noted that NO₂ usually represents only a small fraction of NO_x emissions, which are mostly composed of nitrogen monoxide (NO), even in the case of candles (Lee and Wang 2006).

The modeled concentrations of SO₂ exceeded the recommended limit of 350 μg/m³ (EU 2008) for 65/6 (Figure 3C) wax, while those deriving from 50/1 and 55/9 waxes (Figures 3A and 3B) and scented candle (Figure 2D) emissions were significantly less than this threshold.

The levels of benzene predicted based on RWCS conditions were substantially less than the regulatory limit of 5 μg/m³ (EU 2000). The estimated indoor concentrations of naphthalene emitted from raw wax and scented candle burning under

Table 3. Indoor concentrations of air contaminants emitted from raw wax burning, estimated by a RWCS approach.

	Raw materials		
	50/1	55/9	65/6
NO _x ($\mu\text{g}/\text{m}^3$)	265	174	222
SO ₂ ($\mu\text{g}/\text{m}^3$)	0.51	73	537
PM ($\mu\text{g}/\text{m}^3$)	1.85	19	69
BaP (ng/m^3)	0.37	0.001	0.88

RWCS conditions were well below (more than 30 times) the limit of $3 \mu\text{g}/\text{m}^3$ recommended by the USEPA for chronic exposures to naphthalene (USEPA 1998).

The RWCS concentration for BaP approached the reference level ($1 \text{ ng}/\text{m}^3$) (Figure 3N) only for 65/6 wax, while BaP in the emission products of 55/9 wax was below the limit of detection for the analytical technique (Derudi *et al.* 2014). In this particular case, an emission factor based on one-half of the analytical detection limit of BaP was used to estimate indoor levels. As previously mentioned, the worst-case concentrations were derived assuming a hypothetical RWCS characterized by specific structural (25 m^3 room), behavioral (simultaneous burning of 4 candles), and environmental conditions (0.1 Ach). Summarized in Table 3 are the values obtained under these assumptions only for the pollutants that exceed or are of the same order of magnitude as the recommended concentrations for the protection of human health.

DISCUSSION

Emission Factors

As previously stated, the emission rates used in the present study are described in two companion papers (Derudi *et al.* 2012, 2013). The BTEX emission factors were comparable between the raw materials and scented candles, while the PAH emissions were markedly greater for raw materials (in particular 55/9 and 65/6 waxes) than for scented candles, which were produced using fully refined paraffin waxes as raw materials. Conversely, scented candles emitted greater amounts of aldehydes compared to raw materials, and their emission factors were also markedly greater (up to 2 orders of magnitude) than those reported in the scientific literature (Lau *et al.* 1997). Aldehydes may arise from processes similar to those described for the heating of animal or vegetable oils at temperatures $>200^\circ\text{C}$ or from pyrolysis or oxidation of other organic chemicals during combustion processes, such as tobacco smoking and fossil fuel burning (Shibamoto 2008).

In agreement with previous findings (Fan and Zhang 2001), the mass of particles that was emitted by scented candles and raw materials was predominantly within the size range below $0.5 \mu\text{m}$ (about 90% of total PM). PM emission factors derived from 50/1 wax and scented candle burning were less or comparable to those measured for steady-burning candles (Zai *et al.* 2006), while the emission rates for 55/9 and 65/6 waxes were significantly greater than those of 50/1 waxes (Derudi *et al.* 2014).

The emission rates of PM from scented candles used in this study (Table 2) were also less than those reported in the literature for unscented candles (Fan and Zhang 2001).

Scented Candles

Among the modeled indoor concentrations of selected air contaminants emitted from burning scented candles, only those of acrolein exceeded the corresponding guideline and only under the worst-case behavioral and structural conditions. The recommended value has been proposed to protect the general population from the risk of developing lesions in the upper respiratory tract based on recent experimental data from rats (Dorman *et al.* 2008). In addition, laboratory analyses have failed to discriminate acrolein and acetone, thus measuring only the sum of the two substances. A previous study (Lee and Wang 2006) reported experimental acetone/acrolein emission ratios varying from approximately 0.3 to 3.5 using different candles. Accordingly, the expected indoor concentrations of acrolein actually released from the burning of scented candles can be roughly assumed to be approximately half of those that also include acetone, and the predicted concentrations of acrolein would fall below the recommended level and be considerably less than the concentrations (2.1–12.2 $\mu\text{g}/\text{m}^3$) normally found in indoor living environments (Seaman *et al.* 2007). Moreover, given that the use of scented candles is usually limited in time, other sources (construction materials, furniture, cooking, and smoking) seem to have a greater impact on indoor acrolein concentrations and dominate the causal relationship with the resulting exposures (Zhang *et al.* 1994; Seaman *et al.* 2007).

The indoor levels modeled for all of the other selected pollutants, resulting from the combustion of scented candles, were greatly less than the corresponding recommended concentrations listed in Table 1.

Raw Materials

Among the indoor concentrations of the selected air pollutants emitted by raw materials, the NO_x , SO_2 , and PM levels exceeded their respective recommended values (Table 1). The emission factors and estimated indoor concentrations appeared to be roughly independent of the raw material in the case of NO_x , while SO_2 and PM indoor levels displayed an increasing trend from 50/1, through 55/9 and up to 65/6 waxes (Figure 3).

Notably, NO_2 is the compound with the highest toxicity; therefore, specific guidance values were not developed for NO or nitrogen oxide mixtures but only for NO_2 . The NO_2 guideline value was developed based on human clinical data indicating a decline in pulmonary function and changes in airway responsiveness following short-term exposures and involving susceptible populations, such as asthmatics (WHO 2000). Other biological, acute effects are reported in literature, such as lung immune and inflammatory responses and changes in blood chemistry, with the latter being associated with higher doses (Hesterberg *et al.* 2009). Because previous literature data indicated a NO_x/NO_2 ratio in the 3–4 range (Lee and Wang 2006), the indoor concentrations of NO_2 in the RWCS can be assumed to be significantly below the recommended limit for acute exposures. It should also be noted

that the average concentrations of NO₂ in Italian households located in urban areas were 62 µg/m³ (Simoni *et al.* 2002), and ambient concentrations in many urban areas of Europe during winter generally range from 20 to 200 µg/m³, tend to persist for several consecutive days and can reach peak concentrations of 2000 µg/m³ (WHO 2006). In accordance with these statements, it can be inferred that the use of candles may contribute, in the worst case use scenarios and in combination with other sources, to exceed the threshold for 1-h acute exposures to NO₂ in living environments.

The 1-h recommended value for SO₂ may be exceeded when burning candles manufactured with 65/6 wax and in non-exceptional micro-environmental conditions (Figure 3C), due to the prevalence of sulphur impurities in this type of wax. In fact, 65/6 waxes are earlier products in the refining processes of crude petroleum, compared to 55/9 and, especially, 50/1 waxes.

Note that the regulatory limit selected here based on the aforementioned criteria is less cautionary than the guideline value proposed by WHO (125 µg/m³) (WHO 2000). Thus, indoor air concentrations of SO₂ arising from 65/6 wax burning could pose some risk for human health when multiple candles are used in small spaces scarcely ventilated. The critical effect of SO₂ acute exposures on human health consists essentially of a possible impairment of airway function, especially in asthmatics (Roger *et al.* 1985; Linn *et al.* 1987). SO₂ indoor concentrations in residential areas are usually approximately 10 µg/m³, and the maximum concentrations for short periods can reach 100 µg/m³ (Lee *et al.* 2004). Thus, the contribution of burning candles made with 65/6 or 55/9 waxes to usual indoor levels can be very important.

Estimated indoor concentrations of BaP from 65/6 wax emission rates were comparable with the recommended level only in the RWCS, which was different from the 55/9 and scented candles. The greater emissions of BaP from 50/1 type wax compared to 55/9 wax were not expected and could be possibly interpreted based on different wick behavior during candle burning (position, length, thickness). However, BaP does not cause any important acute or sub-acute adverse effects on human health as a result of exposures to low doses. Thus, the regulatory limit has been developed to ensure that the level of carcinogenic risk for lung cancer remains acceptable in terms of chronic lifetime exposures, which are not primarily attributable to candle fumes. In addition, BaP in outdoor urban areas (near heavy road traffic) is usually in the 0.5–3 ng/m³ range (EC 2001); ambient concentrations are, therefore, already close to or above the regulatory limit.

The PM concentrations predicted for the combustion of 50/1 and 55/9 waxes, as well as scented candles, were below the 25 µg/m³ guideline recommended by WHO for PM_{2.5}. However, the concentrations estimated from the emission factors of 65/6 wax exceeded the recommended limit in worst-case scenarios. As stated before, the estimated PM concentrations were compared to the PM_{2.5} WHO guideline because the aerodynamic diameters of the particles emitted by the combustion of the selected candles generally fell well below the cutoff diameter of 2.5 µm. No guideline values are currently recommended for PM size fractions with cutoff diameters less than 2.5 µm. However, the exposure of consumers to PM emitted by candles should be characterized by a relevant amount of particles that tend to increase their aerodynamic diameters in time due to coagulation and accumulation processes. This behavior is well documented in the literature (Ingebrethsen 1986)

Table 4. Nominal range sensitivity analysis.

Parameter	Pollutant	Input parameters					Reference	Change vs NL (%)	
		MU	LPV	HPV	NV	Min		Max	
V (m ³)	All	m ³	12	80	25	UK DCLG (2012)	-68.8	108.3	
α (h ⁻¹)	NO ₂ -SO ₂	h ⁻¹	0.06	3.7	0.1	Dimitropolou (2012)	-84.3	4.4	
	PM-BaP	h ⁻¹	0.06	3.7	0.1		-92.8	16.3	
	Formal	h ⁻¹	0.06	3.7	0.1		-88.7	6.1	
	Acetald	h ⁻¹	0.06	3.7	0.1		-89.6	6.7	
	Acrol	h ⁻¹	0.06	3.7	0.1		-88.4	7.8	
k (h ⁻¹)	NO ₂	h ⁻¹	0.2	1.3	0.8	Nazaroff <i>et al.</i> (1993)	-37.1	127.2	
	SO ₂	h ⁻¹	2.2	3.2	26.9	USEPA (2009)	-91.2	53.6	
	PM-BaP	h ⁻¹	0.12	0.31	0.18	Long <i>et al.</i> (2001)	-31.6	26.5	
	Formal	h ⁻¹	0.14	0.58	0.36	Nazaroff and Cass (1986)	-25.7	41.9	
	Acetald	h ⁻¹	0.023	1.4	0.23	Liu <i>et al.</i> (2006)	-71.0	42.9	
	Acrol	h ⁻¹	0	14	0.33	Liu <i>et al.</i> (2006)	-96.7	21.4	
	E Raw materials (mg/g)	50/1	mg/h	1.62	1.79	1.71	Derudi <i>et al.</i> (2013)	-3.4	3.4
		NO _x 55/9	mg/h	1.06	1.18	1.12	Derudi <i>et al.</i> (2013)	-3.4	3.4
		65/6	mg/h	1.65	1.76	1.70		-2.2	2.2
		50/1	mg/h	<DL	<DL	<DL		NA	NA
SO ₂ 55/9		mg/h	1.58	2.49	2.03	-22.4		22.4	
65/6		mg/h	9.92	11.06	10.49	-5.4		5.4	
50/1		mg/h	1.15	6.75	3.95	Derudi <i>et al.</i> (2013)		-70.9	70.9
BaP 55/9		mg/h	<DL	<DL	<DL	NA		NA	
E Scented candles (mg/g)	65/6	mg/h	0.08	13.00	6.54	Derudi <i>et al.</i> (2012)	-98.8	98.8	
	PM	mg/h	9.30	13.30	11.46	Derudi <i>et al.</i> (2012) and unpublished data	-18.8	16.1	
	Formal	mg/h	2.28	2.91	2.51		-9.2	15.9	
	Acetald	mg/h	0.62	1.12	0.88		-29.5	27.3	
n candles	Acrol	mg/h	0.95	2.54	1.67		-43.1	52.1	
	All	n	1	8	4	NA	-75.0	100.0	

MU = Measurement unit; LPV = lowest plausible value; HPV = highest plausible value; NV = nominal value; <DL = below the detection limit; NA = not available.

up to approximately a 1 μm diameter, above which such processes are generally inhibited for thermodynamic reasons (Nazaroff 2004). This effect was confirmed by a study of Cypriot medieval churches that reported the highest PM concentrations in the 0.3–2 μm size fraction (Loupa *et al.* 2010).

STRENGTHS AND LIMITATIONS OF THE STUDY

This work was conducted on the basis of emission factors measured during steady-state and high-temperature candle burning in a standardized and controlled chamber (Derudi *et al.* 2012, 2013). This approach had the advantage of avoiding

possible local interfering variables that could affect indoor concentrations. Conversely, it has been shown that the steady-state combustion of candles emits lower amounts of PM than unsteady burning (sooting and smoldering) modes (Fine *et al.* 1999; Zai *et al.* 2006). Thus, the worst-case burning conditions that could lead to short-term exposure peaks were not considered in the present study and modeled indoor concentrations may be underestimated with respect to some real scenarios. However, the present risk assessment was conducted based on RWCS assumptions (0.1 ACH and 25 m³ room volume) and emission factors were fairly comparable with literature data (Derudi *et al.* 2012).

One of the major limitations of the model is the impossibility of estimating the spatial variations of the predicted concentrations, which could be especially important in the vertical direction when dealing with exposures to coarse particles and gaseous compounds with densities substantially different from air. In such a case, indoor pollutant concentrations should be predicted at heights in keeping with the breathing zone of occupants. The single-compartment mass balance model also did not account for spatial variations from sources as a function of the chemical–physical properties and diffusion factors of air pollutants, or ventilation or other structural peculiarities of real indoor spaces.

A nominal range sensitivity analysis of the model was performed by investigating the effect of parameters on the estimated indoor concentrations (Cullen and Frey 1999). The model's inputs were individually varied across their entire range of plausible values, while holding all other inputs at the RWCS base values. The sensitivity was represented as a positive or negative percentage change compared with the RWCS solution (Table 4). Room volume, NO₂ deposition rates, BaP emission rates, and the number of candles simultaneously burnt were found to be the most important parameters leading to possible underestimations of predicted results. Conversely, overestimations of modeled indoor concentrations were largely expected, as the nominal solution was built using RWCS parameters. The model seems also to be insensitive to variations in emission rates calculated from repeated or continuous measurements. Emission rates for BaP represented an exception as they were affected by high analytical uncertainty (Derudi *et al.* 2014). Some of the details of the data quality for the emission factors used here are described in Derudi *et al.* (2012) and Derudi *et al.* (2013).

CONCLUSION

This study estimated the indoor concentrations of air pollutants emitted from the combustion of different types of scented candles and raw materials used in the production of commercial candles. Such indoor levels were compared to regulatory limits or threshold values recommended by international and national bodies.

The evaluation of indoor concentrations predicted from the emission rates of raw materials revealed that the recommended levels of NO_x, SO₂, and PM_{2.5} could be exceeded in some indoor environmental scenarios. The choice of raw materials in candle production is also crucial for limiting emissions and is, therefore, a pivotal step in the risk management process. In particular, the concentrations of SO₂ exceeded the corresponding recommended levels for 65/6 wax. Also, the

concentrations of NO_x and PM_{2.5} modeled from the 65/6 emissions slightly exceeded the recommended levels, but only in very unlikely scenarios. None of the modeled scenarios arising from the combustion of 50/1 and 55/9 waxes have led to an exceedance of the SO₂ and PM_{2.5} recommended thresholds.

These findings are generally valid for all types of candles, and scented candles were found to be responsible for further emissions of aldehydes into indoor environments (Derudi *et al.* 2012). The estimate of the atmospheric concentrations generated by the burning of scented candles in theoretical indoor residential premises revealed that, in the RWCS, the recommended levels of acrolein may be exceeded. Moreover, future studies should elaborate on the specific impact of acrolein emissions in indoor environments because it was not possible to analytically separate acrolein from acetone in this study. None of the other pollutants emitted by scented candles and selected for study was characterized by predicted concentrations of the same order of magnitude as the correspondent recommended levels for living environments.

In conclusion, the possibility of producing indoor concentrations of acrolein from scented candle burning close to the recommended level was only discovered when modeling estimates with the precautionary worst case criteria.

A future extension of this study could include the determination of air pollutant concentrations and associated exposures arising from the burning of candles in domestic indoor environments to evaluate the present model's estimates in real scenarios and perform a more accurate risk assessment.

In light of the results of the present research, the use of high-quality candles made of pure paraffin and not containing high sulphur and oil amounts is strongly recommended, and candles should be burned steadily, avoiding placement in draughts. Candles should be also burned in holders that allow a proper oxygen supply. In unsteady burning modes in turbulent air conditions and low-oxygen atmospheres, more PM and PAH are emitted as a consequence of incomplete combustion (Ott and Siegmann 2006). It is also preferable to extinguish candles by smothering to contain soot emissions. Moreover, the ventilation of indoor premises after candle extinguishing is important for abating indoor levels of air contaminants that are potentially present at concentrations above or similar to the recommended levels, thus reducing the concentration available for inhalation.

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REFERENCES

- Ahrens A and Traas TP. 2007. Environmental exposure scenarios: Development, challenges and possible solutions. *J Expo Sci Env Epid* 17:S7–S15
- Cattaneo A, Peruzzo C, Garramone G, *et al.* 2011. Airborne particulate matter and gaseous air pollutants in residential structures in Lodi province, Italy. *Indoor Air* 21:489–500
- Chao CJ, Wan MP, and Law AK. 2004. Ventilation performance measurement using constant concentration dosing strategy. *Build Environ* 39:1277–88

- Cullen AC and Frey HC. 1999. Probabilistic Techniques in Exposure Assessment: A Handbook for Dealing with Variability and Uncertainty in Models and Inputs, pp 243–71. Plenum Press, New York, NY, USA
- Derudi M, Gelosa S, Sliepcevic A, *et al.* 2012. Emissions of air pollutants from scented candles burning in a test chamber. *Atmos Environ* 55:257–62
- Derudi M, Gelosa S, Sliepcevic A, *et al.* 2014. Emission of air pollutants from burning candles with different composition in indoor environments. *Environmental Science and Pollution Research* (in press) DOI: 10.1007/s11356-013-2394-2,
- Dimitropolou C. 2012. Ventilation in European dwellings: A review. *Build Environ* 47: 109–25
- Dorman DC, Struve MF, Wong BA, *et al.* 2008. Respiratory tract responses in male rats following subchronic acrolein inhalation. *Inhal Toxicol* 20:205–16
- EC (European Communities). 2001. Ambient Air Pollution by Polycyclic Aromatic Hydrocarbons (PAH). Position Paper. Office for Official Publications of the European Communities. Luxembourg
- EChA (European Chemicals Agency). 2008. Guidance on Information Requirements and Chemical Safety Assessment. Part D: Exposure Scenario Building. Helsinki, Finland
- EU (European Union). 2000. EC Directive relating to limit values for benzene and carbon monoxide in ambient air. *Off J Eur Communities* L163:12–21
- EU. 2004. Directive 2004/107/EC of the European Parliament and of the Council of 15 December 2004 relating to arsenic, cadmium, mercury, nickel and polycyclic aromatic hydrocarbons in ambient air. *Off J Eur Communities* L23:3–16
- EU. 2008. Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe. *Off J Eur Communities* L152:1–44
- Fan CW and Zhang J. 2001. Characterization of emissions from portable household combustion devices: Particle size distributions, emission rates and factors, and potential exposures. *Atmos Environ* 35:1281–90
- Fine PM, Cass GR, and Simoneit BRT. 1999. Characterization of fine particle emissions from burning church candles. *Environ Sci Technol* 33:2352–62
- Gilbert NL, Guay M, Gauvin D, *et al.* 2008. Air change rate and concentration of formaldehyde in residential indoor air. *Atmos Environ* 42:2424–28
- Glytsos T, Ondracek J, Dzumbova L, *et al.* 2010. Characterization of particulate matter concentrations during controlled indoor activities. *Atmos Environ* 44:1539–49
- Hesterberg TW, Bunn WB, McClellan RO, *et al.* 2009. Critical review of the human data on short-term nitrogen dioxide (NO₂) exposures: Evidence for NO₂ no-effect levels. *Crit Rev Toxicol* 39:743–81
- Ingebretsen BJ. 1986. Evolution of the particle size distribution of mainstream cigarette smoke during a puff. *Aerosol Sci Technol* 5:423–33
- Kotzias D, Koistinen K, Kephelopoulos S, *et al.* 2005. Final Report of the INDEX Project, Critical Appraisal of the Setting and Implementation of Indoor Exposure Limits in the EU. In *Communities, OfOPotE* (ed). Luxembourg
- Lai ACK. 2002. Particle deposition indoors: A review. *Indoor Air* 12:211–14
- Lau C, Fiedler H, Hutzinger O, *et al.* 1997. Levels of selected organic compounds in materials for candle production and human exposure to candle emissions. *Chemosphere* 34:1623–30
- Lee K, Bartell SM, and Paek D. 2004. Interpersonal and daily variability of personal exposures to nitrogen dioxide and sulfur dioxide. *J Expo Anal Env Epid* 14:137–43
- Lee SC and Wang B. 2006. Characteristics of emissions of air pollutants from mosquito coils and candles burning in a large environmental chamber. *Atmos Environ* 40:2128–38

- Linn WS, Avol EL, Peng R, *et al.* 1987. Replicated dose-response study of sulfur dioxide effects in normal, atopic, and asthmatic volunteers. *Am Rev Respir Dis* 136:1127–34
- Liu W, Zhang J, Zhang L, *et al.* 2006. Estimating contributions of indoor and outdoor sources to indoor carbonyl concentrations in three urban areas of the United States. *Atmos Environ* 40:2202–14
- Long CM, Suh HH, Catalano PJ, *et al.* 2001. Using time- and size-resolved particulate data to quantify indoor penetration and deposition behavior. *Environ Sci Technol* 35:2089–99
- Loupa G and Rapsomanikis S. 2008. Air pollutant emission rates and concentrations in medieval churches. *J Atmos Chem* 60:169–87
- Loupa G, Karageorgos E, and Rapsomanikis S. 2010. Potential effects of particulate matter from combustion during services on human health and on works of art in medieval churches in Cyprus. *Environ Pollut* 158:2946–53
- Maupetit F and Squinazi F. 2009. Characterization of benzene and formaldehyde emissions from burning incense and scented candles indoors: Development of exposure scenarios and recommendations for use. *Environ Risque Sante* 8:109–18
- Nazaroff WW. 2004. Indoor particle dynamics. *Indoor Air*. 14:175–83
- Nazaroff WW and Cass GR. 1986. Mathematical Modeling of Chemically Reactive Pollutants in Indoor Air. *Environ Sci Technol* 20:924–34
- Nazaroff WW, Gadgil AJ, and Weschler CJ. 1993. Critique of the use of deposition velocity in modeling indoor air quality. In: Nagda LM (ed), *Modeling of Indoor Air Quality Exposure*, pp 81–104. American Society for Testing and Materials, Philadelphia, PA, USA
- Nriagu JO and Kim MJ. 2000. Emissions of lead and zinc from candles with metal-core wicks. *Sci Total Environ*. 250:37–41
- OEHHA (Office of Environmental Health Hazard Assessment). 2008. TSD for Noncancer RELs. Appendix D1. Individual Acute, 8-Hour, and Chronic Reference Exposure Level Summaries. California Government, Oakland, CA, USA
- Ökometric. 2007. Testing of Selected Organic Contaminants in Exhaust Gases from Burning Container Candles. The Bayreuther Institute of Environmental Research. Bayreuth, Germany
- Orecchio S. 2011. Polycyclic aromatic hydrocarbons (PAHs) in indoor emission from decorative candles. *Atmos Environ* 45:1888–95
- Ott WR and Siegmann HC. 2006. Using multiple continuous fine particle monitors to characterize tobacco, incense, candle, cooking, wood burning, and vehicular sources in indoor, outdoor, and in-transit settings. *Atmos Environ* 40:821–43
- Pagels J, Wierzbicka A, Nilsson E, *et al.* 2009. Chemical composition and mass emission factors of candle smoke particles. *J Aerosol Sci* 40:193–208
- Roger LJ, Kehrl HR, Hazucha M, *et al.* 1985. Bronchoconstriction in asthmatics exposed to sulfur dioxide during repeated exercise. *J Appl Physiol* 59:784–91
- Roy AA, Baxla SP, Gupta T, *et al.* 2009. Particles emitted from indoor combustion sources: Size distribution measurement and chemical analysis. *Inhal Toxicol* 21:837–48
- Seaman VY, Bennett DH, and Cahill TM. 2007. Origin, occurrence, and source emission rate of acrolein in residential indoor air. *Environ Sci Technol* 41:6940–6
- Shibamoto T. 2008. Acrolein. In: Stadler RH and Lineback DR (eds), *Process-induced Food Toxicants*, pp 51–73. John Wiley & Sons, Hoboken, NJ, USA
- Simoni M, Carrozzi L, Baldacci S, *et al.* 2002. Acute respiratory effects of indoor pollutants in two general population samples living in a rural and in an urban area of Italy. In: *Proceedings 9th International Conference on Indoor Air Quality and Climate*. Indoor Air 2002, pp 119–24. Monterey, CA, USA
- Spicer CW, Kenny D, Ward GF, *et al.* 1993. Transformations, lifetimes, and sources of NO₂, HONO, and HNO₃ in indoor environments. *J Air Waste Manage Assoc* 43:1479–85

- UK Government Department for Communities and Local Government (DCLG). 2012. English Housing Survey: HOMES Annual Report on England's Housing Stock, 2010. London, UK
- USEPA (US Environmental Protection Agency). 1998. Toxicological Review of Naphthalene (CAS No. 91-20-3) in Support of Summary Information on the Integrated Risk Information System (IRIS). National Center for Environmental Assessment, Cincinnati, OH, USA
- USEPA. 2003. Toxicological Review of Acrolein (CAS No. 107-02-8) in Support of Summary Information on the Integrated Risk Information System (IRIS). EPA/635/R-03/003. Washington, DC, USA
- USEPA. 2009. Risk and Exposure Assessment to Support the Review of the SO₂ Primary National Ambient Air Quality Standards: Final Report. U.S. EPA-452/R-09-007. Office of Air Quality Planning and Standards, Research Triangle Park, NC, USA
- van Alphen M. 1999. Emission testing and inhalational exposure-based risk assessment for candles having Pb metal wick cores. *Sci Total Environ* 243/244:53–65
- WHO (World Health Organization). 1992. Acrolein. WHO press, Geneva, Switzerland
- WHO. 2000. Air Quality Guidelines for Europe, 2nd edit. WHO Regional Office for Europe, Copenhagen, Denmark
- WHO. 2006. WHO Air Quality Guidelines for Particulate Matter, Ozone, Nitrogen Dioxide and sulfur Dioxide: Global Update 2005. Summary of Risk Assessment. WHO press, Geneva, Switzerland
- WHO. 2010. WHO Guidelines for Indoor Air Quality: Selected Pollutants. WHO Regional Office for Europe, Copenhagen, Denmark
- Zai S, Zhen H, and Jia song W. 2006. Studies on the size distribution, number and mass emission factors of candle particles characterized by modes of burning. *J Aerosol Sci* 37:1484–96
- Zhang J, He Q, and Liy PJ. 1994. Characteristics of aldehydes: Concentrations, sources, and exposures for indoor and outdoor residential microenvironments. *Environ Sci Technol* 28:146–52