CHARACTERISTICS OF PAHs EMITTED FROM INCENSE, CANDLES AND MOSQUITO COILS BURNING

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Abstract
Burning of candles, incense and mosquito coils in indoor environment can release a large number of toxic chemicals.
This paper reviews available literature data concerning the nature of these products and their burning characteristics, with particular reference to the PAHs emissions.
It has been evidenced that PAHs emission factors show large differences among the investigated materials, even for products belonging to the same class; moreover, it was difficult to find a relationship between the waxes and other raw materials characteristics and the PAHs concentrations within the exhausts.
On the other hand, studying the distribution of different PAHs isomers into the exhausts, it was possible to hypothesize if they were released by evaporation and pyrolysis phenomena at relatively low temperature, due to the overheating of the raw materials constituents, or produced by an incomplete combustion at high temperatures.

Introduction
Both indoor and outdoor sources affect the concentration and composition of pollutants in indoor air. There is usually more information available on emission characteristics, such as emission factors or emission rates, of outdoor pollutants sources [1,2] than of indoor ones. However, quantification of emissions from indoor sources is very important for assessing the human exposure to pollutants [3]. Combustion processes are the main indoor sources of smaller particles, being the vast majority of them in the submicrometer range and containing a host of organic and inorganic material [4,5]. However, gaseous pollutants from combustion play also an important role because they can affect directly the human health or they can act as precursors of secondary particles in the indoor environment, through the process of gas-to-particle conversion [6].
The increase of candle and incense use improved the public concern about potential health effects due to the exposure to their emissions; burning of candles, incense or mosquito coils in indoor environments can release a large number of toxic chemicals, including carbon monoxide, nitrogen oxides, aldehydes and unburnt hydrocarbons, such as polycyclic aromatic hydrocarbons [7-14]. It is believed that regular burning of these materials in indoor environments can expose people to

harmful amounts of organic chemicals [13,15]. In particular, incense sticks and mosquito coils typically smolders and combust incompletely, producing smoke that contains both gases and particulates.
Different studies are available in literature concerning the emissions from burning incense, candles and mosquito coils; most of them are focused on particulate matter (PM) emissions [16-18], while less researches have considered the presence of VOC or PAHs into the emissions [7-9, 19-20]. Consequently, in this work a review of available literature data concerning the nature of these products and their burning characteristics, with particular reference to the PAHs emissions, has been performed. Literature data have been analyzed to determine the distribution of different PAHs isomers into the exhausts, because such an approach allows to hypothesize if the PAHs are released by evaporation and pyrolysis phenomena at relatively low temperature, due to the overheating of the raw materials constituents, or produced by an incomplete combustion at high temperatures.

Distribution of PAHs isomers

A fundamental aspect in the determination of the pollutants emissions from burning materials such as incense sticks, candles or mosquito coils is the replication of realistic burning conditions. Several approaches have been proposed in the literature, ranging from sampling ambient air close to the burning material in a real room [9], naturally or mechanically ventilated, or in a fume hood [13,14,21], to sampling exhaust air from a ventilated environmental chamber [8] or a laboratory-scale test equipment [20] where the burning product is located. These methods provide some pro and some contra related to the cost of a real-size instrumented room or to the lack of reproducibility of a non-controlled environment; usually lab-scale and small-scale rooms allow for a better control of the combustion conditions realizing a smooth and constant air flow around the material to be burnt and ensuring well-mixed conditions and uniform concentration of the exhausts at the chamber outlet. In this work, data concerning PAHs emissions obtained under different test conditions have been used to estimate and compare the distribution of PAHs isomers within the exhausts.
As suggested by several authors [22,23], it is possible to define typical isomeric ratios between PAHs in order to evaluate if they are released by evaporation and pyrolysis phenomena at relatively low temperature, due to the overheating of the raw materials constituents, or produced by an incomplete combustion at high temperature. Usually, values of anthracene to anthracene plus phenanthrene Ant/(Ant+Phe) ratio <0.10 are an index of low temperature sources while values larger than 0.10 indicates a dominance of combustion [22]. Concerning Fla/(Fla+Pyr) and B[a]A/(B[a]A+Chr) ratios, values of 0.4 and 0.2 respectively can be assumed as common threshold between low temperature sources and intermediate conditions or combustion sources (liquid and solid fossil fuels); on the other hand, an InP/(InP+B[g,h,i]P) ratio <0.2 is usually associated to a petroleum source, a ratio of 0.2-0.5 refers to liquid fuel combustion, while a ratio >0.5 is

grass, wood and coal combustion. As it is possible that an analysis carried out on the basis of these ratios can lead to discordant results, especially when very low emission factors must be used to compute these parameters, to solve this problem Orecchio [23] proposed a total index, defined as the sum of the above mentioned isomeric ratios, normalized to the limit values reported in literature [20,22]. When the total index is >4, PAHs are mainly originated from combustion processes while lower values indicate prevalently low temperature emissions.

Results and discussion

As previously mentioned, different literature studies were aimed at measuring PAHs emitted from candles burning, but only a few of them report a detailed characterization of all the PAHs collected in the gas phase emissions. For this reason, four literature experimental datasets [9,20,24,25], obtained with candle with different shapes, wicks, raw materials (from paraffin wax to beeswax) and additives (fragrances and dyes), were used to compute the PAH isomeric ratios shown in Table 1. In a similar way, three studies concerning emissions from incense sticks burning were selected [12-14]; in the case of incense burning, PAHs are usually found in the gas phase emissions as well as adsorbed or bonded to particulate matter (PM) particles released by the incomplete combustion of these materials. Light PAHs, such as naphthalene and fluorene, are the most abundant PAHs within the gas phase, while 3-ring to 5-ring PAHs are mainly found onto the PM particles; consequently, the PAH isomeric ratios were calculated only for the solid phase, as summarized in Table 2. Concerning mosquito coils emissions, two literature works were considered; one was aimed at investigating the influence of ventilation conditions on the emissions of PAH-bound to PM [10], the latter evidenced emission factors of PAHs found in both the gas phase and PM particles [11]; a summary of the corresponding isomeric ratios is shown in Table 3.
Generally, among the investigated materials, candles produced the lowest amount of PAHs but once the emission factors are normalized to compute the isomeric ratios only the PAHs distribution is evidenced, so it is possible to compare the behavior of different materials. However, when the available emission factors were too low, very close to the detection limit of the experimental procedure in use for their quantification, it was non meaningful to compute the corresponding isomeric ratios; for this the reason, especially for high-molecular weight PAHs detected in smoke emissions, several isomeric ratios were set to zero.
About PAHs in candles emissions, it is possible to notice that the Ant/(Ant+Phe) ratio is often <0.1. This seems to indicate that these kind of emissions can be associated to vaporization and pyrolysis phenomena that occur at low temperatures but it is not consistent with the results obtained from the other isomeric ratios and for the total index; apart from one sample, which showed a total index of 2.52, all the other candles exhibited Fla/(Fla+Pyr) values in the range 0.43-0.79 and total index values in the range 3.96-12.92, thus confirming that PAHs are emitted by high temperature processes.

Table 1. Isomeric ratios estimated from PAH emission factors available in literature for different kind of candles (scented and scentless, jar and free standing candles).

	Derudi et al. (2012)					Okometric (1997)							Okometric (2007)		
	A[a]	B[a]	C[a]	D[a]	E[a]	1[b]	2[b]	3[b]	4[b]	5[b]	6[b]	7[b]	Paraffin[c]	Soy[c]	Palm[c]
Ant/(Ant+Phe)	0.19	0.33	0.07	0.27	0.25	0.14	0.17	0.21	0.17	0.10	0.22	0.19	0.03	0.05	0.04
Fla/(Fla+Pyr)	0.50	0.50	0.40	0.43	0.60	0.64	0.60	0.59	0.79	0.62	0.79	0.71	0.57	0.59	0.58
B[a]A/(B[a]A+Chr)	0.76	0.67	0.50	0.01	0.50	0.84	0.81	0.86	0.29	0.60	0.29	0.41	0.27	0.27	0.26
InP/(InP+B[g,h,i]P)	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.43	0.41	0.43
Total Index	6.96	7.92	4.18	3.86	6.50	7.14	7.28	7.82	5.12	5.49	5.55	5.74	3.96	4.12	4.00

	Okometric (2007)		Orecchio (2011)											
	Stearin[c]	Beeswax[c]	1[b]	2[b]	3[b]	4[b]	5[c]	6[c]	7[c]	8[b]	9[d]	10[d]	11[d]	12[d]
Ant/(Ant+Phe)	0.03	0.03	0.02	0.06	0.03	0.03	0.05	0.05	1.00	0.02	0.06	0.05	0.00	0.00
Fla/(Fla+Pyr)	0.62	0.61	0.47	0.47	0.46	0.52	0.50	0.60	0.55	0.19	0.50	0.43	0.53	0.49
B[a]A/(B[a]A+Chr)	0.24	0.28	0.62	0.46	0.41	0.61	0.70	0.79	0.12	0.31	0.59	0.48	0.57	0.56
InP/(InP+B[g,h,i]P)	0.38	0.50	0.36	0.17	0.98	0.96	0.50	0.50	0.50	0.16	0.52	0.52	1.00	1.00
Total Index	3.79	4.28	5.23	4.45	5.41	6.58	6.23	6.93	12.92	2.52	5.79	5.05	6.14	6.02

Notes: [a]scented free standing candles; [b]scentless free standing candles; [c]scentless jar candles; [d]scented jar candles.

Table 2. Isomeric ratios estimated from PAH emission factors available in literature for different kind of incense sticks. Only emission factors related to PAH-bound to particulate particles were considered.

	Yang et al. (2007)									Yang et al. (2012)			Yang et al. (2013)		
	A[a]	B[a]	C[a]	D[a]	E[a]	F[a]	G[a]	H[a]	I[a]	A[b]	B[b]	C[b]	21.3[c]	51[c]	90.5[c]
Ant/(Ant+Phe)	0.32	0.34	0.27	0.30	0.33	0.31	0.31	0.31	0.29	0.19	0.17	0.22	0.22	0.21	0.22
Fla/(Fla+Pyr)	0.51	0.49	0.50	0.51	0.50	0.50	0.51	0.50	0.49	0.45	0.45	0.40	0.47	0.47	0.47
B[a]A/(B[a]A+Chr)	0.35	0.39	0.34	0.41	0.37	0.38	0.38	0.39	0.37	0.45	0.47	0.45	0.49	0.49	0.49
InP/(InP+B[g,h,i]P)	0.52	0.50	0.51	0.57	0.53	0.50	0.55	0.51	0.52	0.60	0.54	0.46	0.53	0.53	0.52
Total Index	7.31	7.53	6.68	7.43	7.43	7.27	7.39	7.29	6.99	6.48	6.30	6.32	6.91	6.82	6.87

Notes: [a]estimated for the total PM collected; [b]only for the PM fraction < 0.5 µm; [c]same sample, different RH%.

With reference to Table 2, the considered incense samples evidenced high values of all the isomeric ratios; Ant/(Ant+Phe) was mainly > 0.2, Fla/(Fla+Pyr) in the range 0.4-0.5 and B[a]A/(B[a]A+Chr) index clearly above 0.3. The total index for PAHs-bound to PM particles produced by the incense incomplete combustion was always > 6.3, independently of the PM fraction considered for the estimation of the

PAHs distribution. Consequently, all the computed ratios confirmed that products from combustion of incense are emitted by high temperature processes that resemble those related to wood and coal combustion.
A quite different behavior was highlighted for mosquito coils samples, because for one of the considered study [10] a total index <4 was individuated for all the investigate samples and experimental conditions, while the other one exhibited several isomeric ratios that lead to discordant results. PAHs in gas phase emissions from mosquito coils burning represent always typical vaporization and low temperature generation processes; on the other hand, the results obtained for PAH-bound to PM particles seem to indicate an intermediate behavior, that is between low temperature sources and intermediate combustion sources (liquid fossil fuels).

Table 3. Isomeric ratios estimated from PAH emission factors available in literature for different kind of mosquito coils. Emission factors related to PAH in the gas phase and PAH-bound to PM particles were considered.

	Lin & Lee (1997)				Zhang et al. (2010)		
	Er-Yi (v)[a]	Bai-Gon (v)[a]	Er-Yi (u)[a]	Bai-Gon (u)[a]	Lanju[b]	Lanju[c]	Bison[b]
Ant/(Ant+Phe)	0.14	0.16	0.14	0.16	0.04	0.08	0.03
Fla/(Fla+Pyr)	0.50	0.48	0.46	0.48	0.86	0.65	0.90
B[a]A/(B[a]A+Chr)	0.00	0.00	0.00	0.00	0.00	0.45	0.00
InP/(InP+B[g,h,i]P)	0.00	0.00	0.00	0.00	0.00	0.09	0.00
Total Index	2.68	2.82	2.58	2.83	2.54	4.90	2.57

	Zhang et al. (2010)						
	Bison[c]	Godzilla[b]	Godzilla[c]	Jumbo[b]	Jumbo[c]	Raid[b]	Raid[c]
Ant/(Ant+Phe)	0.12	0.04	0.22	0.02	0.13	0.03	0.11
Fla/(Fla+Pyr)	0.94	0.87	0.94	0.86	0.92	0.86	0.94
B[a]A/(B[a]A+Chr)	0.29	0.00	0.24	0.00	0.65	0.00	0.46
InP/(InP+B[g,h,i]P)	0.09	0.00	0.07	0.00	0.12	0.00	0.00
Total Index	5.14	2.55	5.86	2.37	7.05	2.48	5.76

Notes: [a]only PAH-bound emission factors: (v) ventilated or (u) unventilated test chamber; [b]PAH in the gas phase; [c]PAH-bound to PM particles.

Conclusions

In this paper, available literature data concerning PAHs emitted from candles, incense and mosquito coils burning in indoor environment were analyzed and used to estimate the distribution of different PAHs isomers into the exhausts.
It was found that the distribution of PAHs measured by several authors in candles emissions fairly reproduce the emission pattern which characterize diesel and liquid fossil fuel combustion, while PAHs emitted from burning incense have isomeric ratios more similar to those usually obtained for grass, wood and coal

combustion. Isomeric ratios computed for PAHs emitted by mosquito coils seem to indicate an intermediate behavior between low temperature petroleum sources and conditions typical of liquid fuel combustion sources.

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