Environmental Research 111 (2011) 87–93

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Predictors of indoor fine particulate matter in infants' bedrooms in Denmark

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A R T I C L E I N F O

Article history:
Received 8 February 2010
Received in revised form
18 October 2010
Accepted 20 October 2010
Available online 6 November 2010

Keywords:
Indoor air
Air pollution
Particulate matter
$PM_{2.5}$
Black smoke

A B S T R A C T

Background: Particulate matter (PM) in ambient air is responsible for adverse health effects in adults and children. Relatively little is known about the concentrations, sources and health effects of PM in indoor air.
Objective: To identify sources of fine PM in infants' bedrooms.
Methods: We conducted 1122 measurements of fine PM ($PM_{2.5}$ and black smoke) in the bedrooms of 389 infants and registered indoor activities and characteristics of the house. We used mixed models to identify and quantify associations between predictors and concentrations.
Results: The concentration of $PM_{2.5}$ was 2.8 times (95% confidence interval [CI], 1.4–5.5 times) higher in houses where people smoked; the concentration increased by 19% (95% CI, 15–23%) per doubling of the amount of tobacco smoked and decreased by 16% (95% CI, 9–27%) per 5-m increase in the distance between the smoking area and the infant's bedroom. Frying without a range hood was associated with a 32% (95% CI, 12–54%) higher $PM_{2.5}$ concentration per time per day, whereas frying with use of a range hood did not increase the concentration in the infant's bedroom. Use of a fireplace, stove, candles or vacuum-cleaner, interior rebuilding or renovation, local traffic, inner city residence and cold season increased the fine PM concentration. Open windows decreased the $PM_{2.5}$ concentration in homes with smokers but increased the concentration in non-smoking homes.
Conclusions: We identified several sources of fine PM in infants' bedrooms. The concentrations can be reduced by use of a range hood for frying, by not using candles, a fireplace or a stove, by increasing the distance between the bedroom and the smoking area and by opening windows in houses of smokers. Smoking is a strong predictor of fine PM in infants' bedrooms and should be avoided.

1. Introduction

Particulate matter (PM) in ambient air has been convincingly associated with adverse health effects, in both adults and children (Brunekreef and Holgate, 2002; World Health Organisation, 2005). In a recent study, we used data from a fixed-site station for monitoring ambient levels of PM in Copenhagen to show an association between ambient air pollution and wheezing in infants (Andersen et al., 2008). Previously identified predictors of indoor PM include indoor combustion, with smoking and cooking the most consistently identified sources (Ozkaynak et al., 1996; Jones et al., 2000; Koistinen et al., 2001; Wallace et al., 2003; He et al., 2004; Lanki et al., 2007); outdoor PM concentrations, measured or indicated by urban location or proximity to traffic (Leaderer et al., 1999; Brauer et al., 2000; Jones et al., 2000; Rojas-Bracho et al., 2000; Rojas-Bracho et al., 2002; Wallace et al., 2003; He et al., 2004; Sorensen et al., 2005; Lanki et al., 2007); markers of the air exchange rate, such as open windows (Wallace et al., 2003; Sorensen et al., 2005; Lanki et al., 2007); and season (Leaderer et al., 1999; Liu et al., 2003; Sorensen et al., 2005). Nevertheless, although populations spend most of their time indoors, there is still limited information about the concentrations and determinants of indoor PM. In particular, quantitative information on associations between sources and the concentrations and details of indoor environments and activities is lacking. In addition, PM has not been measured in the bedrooms of infants, who can be expected to be particularly susceptible to the adverse health effects of lung toxicants such as PM in the air (World Health Organisation, 2005). Identifying the sources of fine PM in infants' bedrooms might help prevent adverse health effects.

The aim of the present study was to measure PM in the bedroom air of infants in a Danish birth cohort and to identify and quantify associations between sources and concentrations.

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doi:10.1016/j.envres.2010.10.007

2. Materials and methods

2.1. Study population

The Copenhagen Prospective Study on Asthma in Childhood is a birth cohort study of 411 infants of mothers with asthma, with the aim of investigating gene–environment–phenotype interactions in the origins of asthma and allergy (Bisgaard, 2004; Bisgaard et al., 2006; Bisgaard et al., 2007). As part of the environmental exposure assessment, indoor air pollution was measured in the houses of the participants (Raaschou-Nielsen et al., 2010). The participants mostly lived within the Greater Copenhagen area, i.e. approximately 15 km from the city centre, but some lived up to 90 km from Copenhagen.

2.2. Measurements of fine particulate matter

Fine PM, i.e. PM with an aerodynamic diameter $\leq 2.5\,\mu$m, was sampled continuously over 1 week in the infants' bedrooms, away from windows and doors, preferably at about 1.5 m above the floor. Measurements were repeated up to four times for each child. Because of the complicated logistics, we could not follow a systematic schedule for these PM measurements, which were therefore initiated at different ages and repeated at different intervals. The median ages at which the first and last measurements were conducted were 9 and 22 months, respectively. The median delay between repeated measurements was 6 months, and all measurements were concluded before the fourth birthday of the child.

Trained personnel initiated and finalised each measurement, using a KTL $PM_{2.5}$ cyclone and a BGI400 pump (flow 4 l/min) (Kenny and Gussman, 1997; Jantunen et al., 1998). The equipment was covered with noise-absorbing material and placed in a plastic box; the cyclone was also placed inside the box, with the inlet protruding through a hole, allowing free air intake. The pumps were plugged into the electric mains. Sampling was performed on 37-mm Teflon filters (Biotech Line, Denmark), which were weighed before and after sampling on a micro weight (MST, Mettler-Toledo). The filters were conditioned for 24 h in a climate chamber (20 °C; 52% relative humidity) before each weighing. The pumps were calibrated before each sampling, and airflow was measured at the beginning and end of sampling.

The blackness of the $PM_{2.5}$ filters, which is an indicator of the black carbon content of the collected PM, was measured as reflectance. These measurements are denoted 'black smoke'. Reflectance was measured on a Model 43 Smokestain Reflectometer (Diffusion System Ltd., United Kingdom), with 30 determinations at three different spots on each filter. The 90 measurements were averaged, and the resulting value was transformed into an absorption coefficient (a, m^{-1}) with the formula $a=(A/2V)*\ln(R_0/R)$, where A is the area of the stain on the filter paper (m^2), V the air volume sampled (m^3), R the intensity of light reflected from the exposed filter and R_0 the intensity of light reflected from a clean filter.

PM measurements were related to the registered airflow of the pump (all registered flows were between 3.5 and 4.5 l/min): higher airflows were associated with lower concentrations. This was expected, as a higher flow implies a displacement of the cut-off curve for the cyclone towards smaller PM. Therefore, the results were normalised to the standard airflow (4 l/min) with coefficients obtained by linear regression of log-transformed PM concentration on the registered airflow.

2.3. Potential predictors

The parents of the infants were asked to complete a questionnaire about activities in the house during the sampling period and about specified household characteristics with a potential effect on the indoor air PM concentration. Cleaning activities in the bedroom included vacuum-cleaning, dusting and sweeping (times per day). Information on cooking in the kitchen included use of a toaster, oven and grill (in the oven), pan-frying (times per day) and associated use of a range hood (always, often, rarely, never). Information on indoor combustion sources in the residence included use of a stove or open fireplace (hours per day), burning candles (hours per day) and tobacco smoking. Tobacco smoking was registered as numbers of cigarettes, cheroots, cigars and pipes smoked, and these data were converted into grams of tobacco by equating one cigarette to 1 g, one cigar to 4.5 g, one cheroot to 3 g and one pipe to 3 g of tobacco. Our staff also noted whether the residence was undergoing interior rebuilding or renovation during the measurement period. Information was also collected on open windows in the bedroom (hours per day) as an indicator of the indoor–outdoor air exchange rate.

Our staff also noted certain characteristics of the residence: distance between the bedroom and the usual smoking area and the amount of traffic on streets within 20 m of the residence in four categories: little local traffic; some traffic, e.g. central streets in small towns or streets leading to local residential areas; heavy traffic, e.g. central streets in towns with 20 000–80 000 inhabitants; and very heavy traffic, e.g. a freeway, an approach road or a major street in the centre of a town with more than 80 000 inhabitants. Each street was allocated to one of the four categories, with account taken of the function of the street in the street network and the size of the town and by visual inspection of the traffic. Finally, the location was characterised as either within 5 km of the centre of Copenhagen, 5–10 km from the centre of Copenhagen, provincial town or rural area. The month when the measurement was started was categorised as: winter: November, December, January, February; spring–autumn: March, April, September, October; or summer: May, June, July, August.

2.4. Statistical analyses

We used a mixed linear model (PROC MIXED in SAS, version 9.1), with individuals as random intercepts, for analysing predictors of indoor $PM_{2.5}$ and black smoke. The covariance structure was specified as compound symmetry, assuming constant variance and a constant correlation between two measurements for the same child regardless of the time between the measurements. The variance components were estimated by the restricted maximum likelihood method (REML). The concentrations of fine PM were right-skewed and therefore transformed by the natural logarithm before analysis. The continuous predictor variables were modelled as indicators of the presence or absence of an activity, e.g. whether smoking occurred in the house during the measurement period, and as 'intensity', e.g. grams of tobacco smoked in the house per day. The indicator and intensity variables were included simultaneously in the model, so that the estimate for the indicator variable was interpreted as the effect of the presence compared with the absence of the activity (e.g. the particle concentration in smoker houses compared with non-smoker houses), and the estimate for the intensity of the activity was interpreted as the increase in particle concentration per increase in the activity. Between- and within-individual variations in $PM_{2.5}$ and black smoke were estimated without covariates in the mixed model and were based on log-transformed PM values.

In post-hoc analyses, we investigated possible interactions between opening windows in the bedroom and the four variables smoking, traffic on a street within 20 m of the residence, area of residence and season. In these interaction analyses, smoking was entered as a simple dichotomous variable (yes/no), and the effect of opening windows was investigated for different levels of the four predictors.

We performed 54 parallel measurements, i.e. two samplers next to each other at the same time, to determine the replication uncertainty of the $PM_{2.5}$ and black smoke measurements. For each set of parallel measurements, the coefficient of variation was calculated as the standard deviation divided by the mean (on the basis of flow calibrated PM values).

3. Results

3.1. Descriptive statistics

In this study, 1120 measurements of $PM_{2.5}$ and 1122 of black smoke were made in the bedrooms of 389 infants. Two $PM_{2.5}$ measurements were discarded because of errors in pre-exposure weighing of the filter. One measurement was performed for 18 infants, two for 19 infants, three for 342 infants and four for 10 infants; 59% of the variance in $PM_{2.5}$ measurements was between individuals and 41% was within individuals, whereas the values for the black smoke measurements were 11% and 89%, respectively. The median concentrations of $PM_{2.5}$ and black smoke were 13 $\mu g/m^3$ and $0.86\times10^{-5}\,m^{-1}$, respectively, and both were approximately 40–50% higher in the centre of Copenhagen than in rural areas and were higher in winter than summer (Table 1). The mean coefficient of variation for 54 parallel measurements was 11% for $PM_{2.5}$ and 10% for black smoke.

Table 2 shows the household activities of potential importance for PM concentrations in the bedrooms during the sampling periods. Frying, use of an oven, toasting, burning candles, vacuum-cleaning, dusting and opening windows were recorded throughout most of the measurement periods; smoking was reported during 25% of the periods, and the other activities occurred more rarely. The intensity of the combustion-related activities (tobacco smoking, burning candles and use of a stove) varied widely, the 95th percentiles being 56–80 times higher than the 5th percentile. The kitchen activities also showed substantial, although less, variation, the 95th percentiles being 5–9 times higher than the 5th percentiles; the 95th percentiles for the cleaning activities were 2–4 times higher. Opening windows in the bedroom showed high variability.

3.2. Linearity of the covariates

We tested the linearity of the association between continuous predictors and outcomes in several steps. First, we included a

Table 1
Descriptive statistics of 1122 measurements of particulate matter in infants' bedrooms.

Type of particulate matter	No. of measurements	Mean	Median	5–95% percentiles
$PM_{2.5}$ ($\mu g/m^3$)				
Total	1120[a]	19	13	5.7–58
Area				
Rural	333	17	12	5.1–51
Provincial town	341	18	12	5.8–61
5–10 km from centre of Copenhagen	174	19	13	5.7–48
≤5 km from centre of Copenhagen	272	22	17	6.7–70
Season[b]				
Winter	368	23	15	6.4–78
Spring–autumn	368	18	13	5.5–50
Summer	384	16	12	5.4–47
Black smoke (10^{-5} m^{-1})				
Total	1122	0.97	0.86	0.32–2.1
Area				
Rural	333	0.88	0.72	0.28–2.1
Provincial town	342	0.89	0.76	0.29–1.8
5–10 km from centre of Copenhagen	175	1.0	0.94	0.40–2.0
≤5 km from centre of Copenhagen	272	1.2	1.1	0.52–2.4
Season[b]				
Winter	368	1.3	1.2	0.47–2.7
Spring–autumn	370	0.94	0.84	0.36–1.9
Summer	384	0.74	0.66	0.27–1.4

[a] Two $PM_{2.5}$ measurements were discarded due to error in the pre-exposure weighing.
[b] Winter: November, December, January, February; spring–autumn: March, April, September, October; summer: May, June, July, August.

squared term in the model, which indicated deviation from linearity ($p < 0.05$) for "amount of tobacco smoked" in the $PM_{2.5}$ model ($p = 0.0002$), "use of fireplace or wood-burning stove" ($p = 0.02$), "frying" ($p = 0.04$) and "use of candles" ($p = 0.02$) in the black smoke model. Secondly, we modelled these four variables as linear splines (Greenland, 1995) with boundaries placed at the 25th, 50th and 75th percentiles of the distribution of each variable, and tested whether the linear spline model fitted better than a linear model, which would indicate deviation from linearity. For "frying" and "candles" there was no deviation from linearity in the linear spline model ($p = 0.20$ and 0.18, respectively) and no systematic deviation according to graphical inspection and we, thus, modelled these variables as linear. "Amount of tobacco" and "use of fireplace or wood-burning stove" deviated from linearity in the linear spline model ($p < 0.0001$ and 0.09, respectively) and graphical inspection showed systematic leveling off of the effect at higher exposures. Therefore, we transformed these variables by applying the logarithm to the base 2. After the log-transformation, neither inclusion of a squared term (both $p > 0.47$), test of the linear spline function (both $p > 0.34$) nor graphical inspection indicated deviation from linearity. Because of this transformation, the effect estimates are interpreted as increases in particle concentration per doubling of, respectively, "tobacco smoking" and "use of a fireplace or wood-burning stove".

3.3. Predictors of $PM_{2.5}$ concentration in the bedroom

Table 3 shows the estimated effects of household characteristics and activities on indoor $PM_{2.5}$ concentrations. Tobacco smoking was associated with higher $PM_{2.5}$ concentrations in the infants' bedrooms, and a greater distance between the smoking area and the bedrooms was associated with lower concentrations. Pan-frying in the kitchen increased the $PM_{2.5}$ concentration in the bedrooms if a range hood was rarely or never used. Use of an oven, grilling in an oven and toasting bread also increased the $PM_{2.5}$ concentrations in the bedrooms, but not significantly. Vacuum-cleaning in the bedrooms increased the $PM_{2.5}$ concentration, whereas the results for dusting and sweeping in the bedrooms and interior rebuilding or renovation were not statistically significant. Two indicators of outdoor air pollution, heavy local traffic and living in the inner city, were both associated with higher $PM_{2.5}$ concentrations, and the concentrations were generally higher in the colder seasons.

3.4. Predictors of black smoke in the bedroom

Higher concentrations of black smoke were found in the children's bedrooms in association with three indoor sources of combustion: smoking, burning candles and use of a fireplace or stove (Table 4). Frying was also associated with a higher black smoke concentration. The associations with the other cooking and cleaning activities were not statistically significant. Interior rebuilding or renovation, heavy local traffic, living close to the centre of Copenhagen, colder season and opening windows were all associated with higher black smoke concentrations in the bedrooms.

3.5. Post-hoc interaction analyses

The analyses of possible interactions between open windows and predictors of PM concentration in the bedrooms indicated interactions with smoking. Open windows were associated with a 15% (95% CI, 8–22%) lower concentration of $PM_{2.5}$ per 10 h with open windows per day in houses with smokers but 4% (95% CI, −1% to 10%) higher concentrations in houses without smokers (p for interaction, < 0.0001). Open windows for 10 h per day was associated with 6% higher levels of black smoke (95% CI, 1–12%) in houses without smokers, but virtually no effect was seen in houses with smokers, where 10 h with open windows per day was associated with a 2% higher concentration (95% CI, −7% to 11%; p for interaction $= 0.36$) (data not shown).

Open windows for 10 h/day was associated with 3% lower $PM_{2.5}$ concentrations (95% CI, −8 to 2) during summer, 7% (95% CI, −4% to 20%) higher concentrations during spring–autumn and 7% (95% CI,

Table 2
Descriptive statistics of potential predictors of particulate matter concentration in an infant's bedroom, based on 1122 measurements.

	N[a]	Mean	Median	5–95% percentiles
Tobacco smoking				
No	804	–	–	–
Yes (g per day[b])	263	8.2	5.8	0.43–25
Distance from monitor to smoking area (m)	262	6.9	6	2–14
Use of fireplace or wood-burning stove				
No	984	–	–	–
Yes (hours per day)	129	8.1	5.1	0.43–24
Burning candles				
No	505	–	–	–
Yes (hours per day)	614	1.1	0.57	0.05–4.0
Frying				
No	33	–	–	–
Yes (times per day)	1068	0.57	0.57	0.14–1.0
Use of oven				
No	110	–	–	–
Yes (times per day)	1008	0.40	0.32	0.14–0.85
Grilling in oven				
No	972	–	–	–
Yes (times per day)	147	0.22	0.14	0.12–0.57
Toasting bread				
No	411	–	–	–
Yes (times per day)	708	0.53	0.43	0.14–1.3
Vacuum-cleaning in bedroom				
No	136	–	–	–
Yes (times per day)	982	0.24	0.14	0.14–0.57
Dusting in bedroom				
No	327	–	–	–
Yes (times per day)	788	0.14	0.14	0.14–0.30
Sweeping in bedroom				
No	984	–	–	–
Yes (times per day)	129	0.14	0.14	0.14–0.57
Window open in bedroom				
No	51	–	–	–
Yes (hours/day)	1064	4.7	1.00	0.08–24
Interior rebuilding or renovation				
No	1011	–	–	–
Yes	109	–	–	–
Traffic[c] < 20 m from the residence				
Local	895	–	–	–
Some	147	–	–	–
Heavy	60	–	–	–
Very heavy	20	–	–	–

[a] Numbers do not add up to 1122 because of missing answers in the questionnaire.
[b] Conversion factors for different types of smoking: 1 cigarette = 1 g, 1 cigar = 4.5 g, 1 cheroot = 3 g, 1 pipe = 3 g.
[c] See Methods section for definition of traffic categories.

–8% to 25%) higher concentrations during winter (*p* for interaction = 0.15). The association between open windows and black smoke concentration did not differ significantly in summer, spring–autumn and winter, with 4%, 10% and 6% higher concentrations, respectively, in association with 10 h with open windows (*p* for interaction = 0.65) (data not shown).

No interaction was found between open windows in the bedroom and local traffic or area of residence with respect to either $PM_{2.5}$ or black smoke, with *p* values for these interactions above 0.34 (data not shown).

4. Discussion

The study showed that indoor concentrations of fine PM are due to contributions from indoor combustion sources, cooking, interior rebuilding or renovation, vacuum-cleaning, nearby traffic, inner city residence, season and opening windows.

The indoor $PM_{2.5}$ concentrations measured (mean, 18.8 μg/m^3; median, 12.8 μg/m^3) were higher than those measured in Seattle, USA (Liu et al., 2003) and Helsinki, Finland (Lanki et al., 2007), similar to those measured in Virginia (Leaderer et al., 1999), Boston (Rojas-Bracho et al., 2000), Tucson and Seattle (Wallace et al., 2003), USA, and lower than those measured in Amsterdam, the Netherlands (Lanki et al., 2007), Santiago, Chile (Rojas-Bracho et al., 2002) and New York, Dallas and Chicago, USA (Wallace et al., 2003).

Our study population comprised only the infants of mothers with asthma, and the activities of importance for the PM concentration in houses might be different for families of these mothers than for others. For example, smoking may be less prevalent in the houses of mothers with asthma (Raaschou-Nielsen et al., 2010). Nevertheless, the estimated association between e.g. smoking intensity (g tobacco smoked in the house per day) and PM

Table 3
Predictors of $PM_{2.5}$ in Danish infants' bedroom based on 1015 measurements with no missing values in any of the variables included in the multiple regression analysis.

Predictor	Effect estimate[a]	95% CI
Tobacco smoking (yes versus no)	2.80	1.44–5.45
Tobacco smoking intensity (per doubling of amount)	1.19	1.15–1.23
Distance from monitor to smoking area (per 5 m)	0.84	0.77–0.91
Use of fireplace or wood-burning stove (yes versus no)	1.00	0.87–1.14
Use of fireplace or wood-burning stove (per doubling of time)	1.02	0.97–1.06
Burning candles (yes versus no)	1.04	0.98–1.12
Burning candles (per 1 h per day)	1.00	0.97–1.03
Frying (per 1 time per day) when range hood used[b]		
Always	1.00	0.88–1.13
Often	0.92	0.78–1.08
Rarely	1.17	0.89–1.54
Never	1.32	1.12–1.54
Oven (per 1 time per day)	1.11	0.99–1.26
Grilling in oven (per 1 time per day)	1.15	0.69–1.91
Toasting bread (per 1 time per day)	1.05	0.95–1.16
Vacuum-cleaning in bedroom (per 1 time per day)	1.31	1.05–1.64
Dusting in bedroom (per 1 time per day)	1.12	0.77–1.63
Sweeping in bedroom (per 1 time per day)	1.06	0.73–1.52
Interior rebuilding or renovation (yes versus no)	0.94	0.85–1.04
Traffic[c] <20 m from the residence		
Local (reference)	1.00	–
Some	1.19	1.07–1.32
Heavy	1.22	1.03–1.43
Very heavy	1.77	1.35–2.31
Area		
Rural (reference)	1.00	–
Provincial town	0.96	0.87–1.05
5–10 km from centre of Copenhagen	1.02	0.91–1.15
≤ 5 km from centre of Copenhagen	1.12	1.00–1.25
Season		
Summer (reference)	1.00	–
Spring–autumn	1.22	1.13–1.32
Winter	1.41	1.30–1.53
Windows open in the bedroom (per 10 h per day)	0.99	0.94–1.04

[a] Examples of interpretation of the effect estimates (β-values): 2.80 for smoking is interpreted as 2.8 times higher $PM_{2.5}$ concentrations in houses with smoking than in houses with no smoking during the measurement period; 1.19 for smoking intensity is interpreted as a 19% higher $PM_{2.5}$ concentration per doubling of the amount of tobacco smoked per day; 0.84 for distance between monitor and smoking area is interpreted as a 16% lower $PM_{2.5}$ concentration per 5 m distance; 1.32 for "frying/never range hood" is interpreted as a 32% higher $PM_{2.5}$ concentration per time frying per day in homes where range hood was never used; 1.11 for oven is interpreted as 11% higher $PM_{2.5}$ concentration per time the oven was used per day; 1.41 for winter is interpreted as 41% higher $PM_{2.5}$ concentration during winter than during summer. The estimates are mutually adjusted for the other predictors in the table.

[b] The interaction between frying and use of range hood was statistically significant ($p=0.0002$).

[c] See Methods section for definition of traffic categories.

concentrations in this study would probably be identical to that in houses of mothers without asthma. We therefore consider that the associations between these predictors and PM concentrations estimated in our study can be generalised to other study populations.

Previous studies have consistently shown an association between smoking in the house and fine PM concentrations measured at various locations in the house (Ozkaynak et al., 1996; Jones et al., 2000; Koistinen et al., 2001; Wallace et al., 2003; He et al., 2004; Lanki et al., 2007; Stranger et al., 2009). Our study reinforces this finding and extends it to concentrations measured in the bedrooms of infants, where people do not usually smoke; the lowest concentrations in the infants' bedrooms were those farthest from the usual smoking area. We are not aware of previous studies of this aspect.

We also showed that use of candles in the house was associated with higher concentrations of black smoke in infants' bedrooms. It has been shown that candles emit fine PM (Fine et al., 1999). An association with the indoor concentration of fine PM was shown in one study (He et al., 2004) but not in another (Wallace et al., 2003); a study of personal exposure showed associations between use of candles at home and exposure to both $PM_{2.5}$ and black smoke (Sorensen et al., 2005). Our results also show an association between use of a stove or fireplace and black smoke but not $PM_{2.5}$ indoors. Wallace et al. (2003) showed no association between use of a woodstove or fireplace and $PM_{2.5}$, but these appliances were used in only 1% of the 294 houses studied. Koistinen et al. (2001) found that the presence (not the use) of a wood or gas stove resulted in only slightly increased personal exposure to $PM_{2.5}$, but only 9 of 201 houses had a stove. A study in a Rocky Mountain valley community in the USA showed that replacement of old stoves with new woodstoves certified by the United States Environmental Protection Agency decreased the indoor $PM_{2.5}$ concentration significantly (Ward et al., 2008).

Cooking has consistently been identified as an important source of PM in indoor air (Ozkaynak et al., 1996; Jones et al., 2000; Wallace et al., 2003; He et al., 2004; Baxter et al., 2007; Lanki et al., 2007; Abt et al., 2000). Our study confirmed an association between frying and $PM_{2.5}$ concentration, but we also showed that use of a range hood when frying effectively prevented higher $PM_{2.5}$ concentrations in the infant's bedroom. Previous studies have shown that use of a grill is associated with particularly high PM

Table 4
Predictors of black smoke in Danish infants' bedroom based on 1016 measurements with no missing values in any of the variables included in the multiple regression analysis.

Predictor	Effect estimate[a]	95% CI
Tobacco smoking (yes versus no)	1.93	1.00–3.71
Tobacco smoking intensity (per doubling of amount)	1.05	1.01–1.09
Distance from monitor to smoking area (per 5 m)	0.91	0.84–0.99
Use of fireplace or wood-burning stove (yes versus no)	1.06	0.93–1.21
Use of fireplace or wood-burning stove (per doubling of time)	1.04	1.00–1.09
Burning candles (yes versus no)	1.16	1.08–1.24
Burning candles (per 1 h per day)	1.05	1.02–1.08
Frying (per 1time per day) when range hood used[b]		
Always	1.14	1.01–1.29
Often	1.00	0.85–1.16
Rarely	1.11	0.85–1.46
Never	1.11	0.95–1.29
Oven (per 1 time per day)	0.99	0.88–1.12
Grilling in oven (per 1 time per day)	1.24	0.75–2.05
Toasting bread (per 1 time per day)	1.06	0.96–1.16
Vacuum-cleaning in bedroom (per 1 time per day)	1.15	0.92–1.43
Dusting in bedroom (per 1 time per day)	0.96	0.65–1.40
Sweeping in bedroom (per 1 time per day)	1.42	0.98–2.07
Interior rebuilding or renovation (yes versus no)	1.12	1.01–1.23
Traffic[c] <20 m from the residence		
Local	1.00	–
Some	1.02	0.93–1.12
Heavy	1.21	1.05–1.40
Very heavy	1.50	1.18–1.90
Area		
Rural	1.00	–
Provincial town	0.98	0.91–1.06
5–10 km from centre of Copenhagen	1.21	1.10–1.34
≤ 5 km from centre of Copenhagen	1.36	1.23–1.49
Season		
Summer	1.00	–
Spring–autumn	1.28	1.19–1.39
Winter	1.60	1.47–1.74
Windows open in bedroom (per 10 h per day)	1.05	1.00–1.10

[a] See note to Table 3 for interpretation of effect estimates.
[b] The interaction between frying and use of range hood was insignificant ($p=0.32$).
[c] See Methods section for definition of traffic categories.

concentrations indoors (He et al., 2004). We also found an association between use of grill in the kitchen and the PM concentration in the infant's bedroom, although the association was not statistically significant.

Vacuum-cleaning was associated with higher PM concentrations. One previous study of the association between vacuum-cleaning and indoor particles found no significant association (Wallace et al., 2003), and another study reported an association between unspecified cleaning activity and personal exposure to $PM_{2.5}$ in one of two study areas (Lanki et al., 2007). Vacuum-cleaning implies disturbance of PM deposited on surfaces, and, although large PM is trapped by the air stream and deposited on the filter of the vacuum cleaner, fine PM might not be trapped by the filter and would then be resuspended in the air, thereby increasing the PM concentration in the room. This might explain the association between vacuum-cleaning and the concentrations of fine PM in the present study. Other cleaning activities (dusting, sweeping) were not statistically significantly associated with $PM_{2.5}$ concentrations, in accordance with the results of previous studies, which showed no association with dusting (Wallace et al., 2003) or with unspecified cleaning activity (Jones et al., 2000; Lanki et al., 2007). A recent study showed an association between human activity level and indoor $PM_{2.5}$ (Chen and Hildemann, 2009).

Many studies have shown that the measured ambient PM concentration (Ozkaynak et al., 1996; Jones et al., 2000; Rojas-Bracho et al., 2000, 2002; Wallace et al., 2003; Sorensen et al., 2005; Baxter et al., 2007; Molnar et al., 2007) and proxy measures such as local traffic (Lanki et al., 2007) and urban residence (Brauer et al., 2000) are associated with indoor air PM concentrations. In line with these results, we found higher concentrations of both $PM_{2.5}$ and black smoke in infants' bedrooms in houses in the inner city area than in houses in more rural settings and less than 20 m from a road with heavy traffic.

Ambient PM concentrations are typically higher in cold than in warm weather. We found a seasonal pattern of indoor PM concentrations, consistent with two previous reports (Liu et al., 2003; Sorensen et al., 2005), although two other studies showed no significant differences in $PM_{2.5}$ concentration by season (Brauer et al., 2000; Rojas-Bracho et al., 2000). Leaderer et al. (1999) found a higher indoor PM concentration during the heating season, but only in houses with a kerosene heater. Higher indoor PM concentrations during winter could be due to greater indoor emissions, greater contributions of ambient PM or both. In our study, candles, stoves or open fireplaces were used more frequently in cold weather, but even after mutual adjustment for indoor sources in the multiple regression analyses, cold season was a significant predictor of indoor air PM, indicating that PM in ambient air contributes to the indoor PM concentration.

The results indicate that open windows in a child's bedroom during the summer decreases the concentration of $PM_{2.5}$, whereas open windows during the winter results in a higher concentration. Open windows was also beneficial in houses with smokers.

Although these results appear intuitively correct, given the higher indoor PM concentrations in houses with smokers and the higher ambient PM concentrations in winter than in summer, the balance between indoor and outdoor concentration (which determines whether open windows will increase or decrease the PM concentration in indoor air) depends on the geographical location (the ambient concentration) of the house and indoor sources.

Many of the predictors of $PM_{2.5}$ and of black smoke in this study were identical (smoking, local traffic, area and season), but the sources of these two measures of fine PM also differed. Indoor combustion sources (burning candles and use of wood-burning stoves) predicted black smoke but not $PM_{2.5}$, due perhaps to the high carbon black content of low-mass PM from candles and stoves. The identification of specific sources of PM depends on the method of measurement. The methods used in this study (weight and blackness) were quite general, resulting in similar source profiles for $PM_{2.5}$ and black smoke. More specific methods, such as quantification of elemental carbon and organic carbon (He et al., 2004) or other chemical components (Leaderer et al., 1999), might have improved the detection of some PM sources, whereas others might have been impossible to detect.

There is persuasive evidence that air pollution causes respiratory symptoms, aggravates asthma and impairs lung function development in children (World Health Organisation, 2005). Asthma exacerbation is a common cause of hospitalisations of young children in westernised countries (Newacheck and Stoddard, 1994) and makes major demands on both the involved families and the health care system. Identifying sources of fine PM in infants' bedrooms might help prevent asthma exacerbations.

Our findings show that indoor combustion sources (tobacco smoking, candles, stoves and fireplaces), cooking, vacuum-cleaning, interior rebuilding or renovation, local traffic, inner city residence and cold season contribute to the fine PM concentrations in infants' bedrooms. In particular, we showed that a shorter distance from the smoking area was associated with a higher fine PM concentration in the infant's bedroom and that use of a range hood when frying effectively prevented an increase in the $PM_{2.5}$ concentration.

Acknowledgments

We thank laboratory technician Birgit Thomsen at the Danish National Environmental Research Institute for assistance with air pollution measurements. This study was funded by the Danish Ministry of the Interior and the Health Research Centre for Environmental Medicine.

References

Abt, E., Suh, H.H., Allen, G., Koutrakis, P., 2000. Characterization of indoor particle sources: a study conducted in the metropolitan Boston area. Environ. Health Perspect. 108, 35–44.

Andersen, Z.J., Loft, S., Ketzel, M., Stage, M., Scheike, T., Hermansen, M.N., et al., 2008. Ambient air pollution triggers wheezing symptoms in infants. Thorax 63, 710–716.

Baxter, L.K., Clougherty, J.E., Laden, F., Levy, J.I., 2007. Predictors of concentrations of nitrogen dioxide, fine particulate matter, and particle constituents inside of lower socioeconomic status urban homes. J. Expo. Sci. Environ. Epidemiol. 17, 433–444.

Bisgaard, H., 2004. The Copenhagen prospective study on asthma in childhood (COPSAC): design, rationale, and baseline data from a longitudinal birth cohort study. Ann. Allergy Asthma Immunol. 93, 381–389.

Bisgaard, H., Hermansen, M.N., Buchvald, F., Loland, L., Halkjaer, L.B., Bonnelykke, K., et al., 2007. Childhood asthma after bacterial colonization of the airway in neonates. N. Engl. J. Med. 357, 1487–1495.

Bisgaard, H., Hermansen, M.N., Loland, L., Halkjaer, L.B., Buchvald, F., 2006. Intermittent inhaled corticosteroids in infants with episodic wheezing. N. Engl. J. Med. 354, 1998–2005.

Brauer, M., Hruba, F., Mihalikova, E., Fabianova, E., Miskovic, P., Plzikova, A., et al., 2000. Personal exposure to particles in Banska Bystrica, Slovakia. J. Expo. Anal. Environ. Epidemiol. 10, 478–487.

Brunekreef, B., Holgate, S.T., 2002. Air pollution and health. Lancet 360, 1233–1242.

Chen, Q., Hildemann, L.M., 2009. The effects of human activities on exposure to particulate matter and bioaerosols in residential homes. Environ. Sci. Technol. 43, 4641–4646.

Fine, P.M., Cass, G.R., Simoneit, B.R.T., 1999. Characterization of fine particle emissions from burning church candles. Environ. Sci. Technol. 33, 2352–2362.

Greenland, S., 1995. Dose-response and trend analysis in epidemiology: alternatives to categorical analysis. Epidemiology 6, 356–365.

He, C.R., Morawska, L.D., Hitchins, J., Gilbert, D., 2004. Contribution from indoor sources to particle number and mass concentrations in residential houses. Atmos. Environ. 38, 3405–3415.

Jantunen, M.J., Hanninen, O., Katsouyanni, K., Knoppel, H., Küenzli, N., Lebret, E., et al., 1998. Air pollution exposure in European cities: the 'EXPOLIS' study. J. Expo. Anal. Environ. Epidemiol. 8, 495–518.

Jones, N.C., Thornton, C.A., Mark, D., Harrison, R.M., 2000. Indoor/outdoor relationships of particulate matter in domestic homes with roadside, urban and rural locations. Atmos. Environ. 34, 2603–2612.

Kenny, L.C., Gussman, R.A., 1997. Characterization and modelling of a family of cyclone aerosol preseparators. J. Aerosol Sci. 28, 677–688.

Koistinen, K.J., Hänninen, O., Rotko, T., Edwards, R.D., Moschandreas, D., Jantunen, M.J., 2001. Behavioral and environmental determinants of personal exposures to PM2.5 in EXPOLIS–Helsinki, Finland. Atmos. Environ. 35, 2473–2481.

Lanki, T., Ahokas, A., Alm, S., Janssen, N.A., Hoek, G., De Hartog, J.J., et al., 2007. Determinants of personal and indoor PM2.5 and absorbance among elderly subjects with coronary heart disease. J. Expo. Sci. Environ. Epidemiol. 17, 124–133.

Leaderer, B.P., Naeher, L., Jankun, T., Balenger, K., Holford, T.R., Toth, C., et al., 1999. Indoor, outdoor, and regional summer and winter concentrations of PM10, PM2.5, SO4(2)–, H+, NH4+, NO3–, NH3, and nitrous acid in homes with and without kerosene space heaters. Environ. Health Perspect. 107, 223–231.

Liu, L.J., Box, M., Kalman, D., Kaufman, J., Koenig, J., Larson, T., et al., 2003. Exposure assessment of particulate matter for susceptible populations in Seattle. Environ. Health Perspect. 111, 909–918.

Molnar, P., Bellander, T., Sallsten, G., Boman, J., 2007. Indoor and outdoor concentrations of PM2.5 trace elements at homes, preschools and schools in Stockholm, Sweden. J. Environ. Monit. 9, 348–357.

Newacheck, P.W., Stoddard, J.J., 1994. Prevalence and impact of multiple childhood chronic illnesses. J. Pediatr. 124, 40–48.

Ozkaynak, H., Xue, J., Spengler, J., Wallace, L., Pellizzari, E., Jenkins, P., 1996. Personal exposure to airborne particles and metals: results from the particle TEAM study in Riverside, California. J. Expo. Anal. Environ. Epidemiol. 6, 57–78.

Raaschou-Nielsen, O., Hermansen, M.N., Loland, L., Buchvald, F., Pipper, C.B., Sorensen, M., et al., 2010. Long-term exposure to indoor air pollution and wheezing symptoms in infants. Indoor Air 20, 159–167.

Rojas-Bracho, L., Suh, H.H., Koutrakis, P., 2000. Relationships among personal, indoor, and outdoor fine and coarse particle concentrations for individuals with COPD. J. Expo. Anal. Environ. Epidemiol. 10, 294–306.

Rojas-Bracho, L., Suh, H.H., Oyola, P., Koutrakis, P., 2002. Measurements of children's exposures to particles and nitrogen dioxide in Santiago, Chile. Sci. Total Environ. 287, 249–264.

Sorensen, M., Loft, S., Andersen, H.V., Raaschou-Nielsen, O., Skovgaard, L.T., Knudsen, L.E., et al., 2005. Personal exposure to PM2.5, black smoke and NO_2 in Copenhagen: relationship to bedroom and outdoor concentrations covering seasonal variation. J. Expo. Anal. Environ. Epidemiol. 15, 413–422.

Stranger, M., Potgieter-Vermaak, S.S., Van Grieken, R., 2009. Particulate matter and gaseous pollutants in residences in Antwerp, Belgium. Sci. Total Environ. 407, 1182–1192.

Wallace, L.A., Mitchell, H., O'Connor, G.T., Neas, L., Lippmann, M., Kattan, M., et al., 2003. Particle concentrations in inner-city homes of children with asthma: the effect of smoking, cooking, and outdoor pollution. Environ. Health Perspect. 111, 1265–1272.

Ward, T., Palmer, C., Bergauff, M., Hooper, K., Noonan, C., 2008. Results of a residential indoor PM2.5 sampling program before and after a woodstove changeout. Indoor Air 18, 408–415.

World Health Organization, 2005. Effects of Air Pollution on Children's Health and Development—A Review of the Evidence. WHO Regional Office for Europe, Copenhagen.