
Volume 127 November 2013 ISSN 0013-9351
ELSEVIER
environmental
research

Environmental Research 127 (2013) 49–55

Contents lists available at ScienceDirect

Environmental Research

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

Short Communication

Variability in the correlation between nicotine and $PM_{2.5}$ as airborne markers of second-hand smoke exposure ☆

Marcela Fu [a,b,c], Jose M. Martínez-Sánchez [a,b,d], Iñaki Galán [e], Mónica Pérez-Ríos [f,g,i], Xisca Sureda [a,b,c], María J. López [h,i,j], Anna Schiaffino [a], Albert Moncada [k], Agustín Montes [g,i], Manel Nebot [h,i,j,l,1], Esteve Fernández [a,b,c,*]

[a] *Tobacco Control Unit, Cancer Control and Prevention Programme, Institut Català d'Oncologia—ICO, Av. Granvia de L'Hospitalet 199-203, 08908 L'Hospitalet de Llobregat, Barcelona, Spain*
[b] *Cancer Control and Prevention Group, Institut d'Investigació Biomèdica de Bellvitge—IDIBELL, Av. Granvia de L'Hospitalet 199-203, 08908 L'Hospitalet de Llobregat, Barcelona, Spain*
[c] *Department of Clinical Sciences, School of Medicine, Universitat de Barcelona—Pavelló de Govern, c/Feixa Llarga s/n, 08907 L'Hospitalet de Llobregat, Barcelona, Spain*
[d] *Department of Basic Sciences, Universitat Internacional de Catalunya, c/Josep Trueta s/n, 08195 Sant Cugat del Vallès, Barcelona, Spain*
[e] *National Centre for Epidemiology, Instituto de Salud Carlos III, c/Monforte de Lemos 5, 28029 Madrid, Spain*
[f] *Epidemiology Unit, Galician Directorate for Public Health, Galician Health Authority, Xunta de Galicia, Edif. Admtvo, San Lázaro s/n, 15703 Santiago de Compostela, Spain*
[g] *Department of Preventive Medicine and Public Health, School of Medicine and Odontology, Universidade de Santiago de Compostela, R/San Francisco s/n, 15782 Santiago de Compostela, Spain*
[h] *Agència de Salut Pública de Barcelona, Pl. Lesseps 1, 08023 Barcelona, Spain*
[i] *Biomedical Research Centre Network for Epidemiology and Public Health (CIBERESP), Parc de Recerca Biomèdica de Barcelona, c/Doctor Aiguader 88 Planta 1, 08003 Barcelona, Spain*
[j] *Sant Pau Institute of Biomedical Research, c/Sant Antoni Maria Claret 167—Pavelló de Sant Frederic 16 Planta 1, 08025 Barcelona, Spain*
[k] *Community Health Unit, Ajuntament de Terrassa, Ctra. Montcada 596, 08223 Terrassa, Spain*
[l] *Department of Experimental and Health Sciences, Universitat Pompeu Fabra, c/Doctor Aiguader 88, 08003 Barcelona, Spain*

ARTICLE INFO

Article history:
Received 12 February 2013
Received in revised form
20 September 2013
Accepted 26 September 2013
Available online 28 October 2013

Keywords:
Air nicotine
Particulate matter
Second-hand smoke
Correlation

ABSTRACT

The aim of this study was to assess the relationship between particulate matter of diameter ≤ 2.5 μm ($PM_{2.5}$) and airborne nicotine concentration as markers of second-hand smoke exposure with respect to the setting studied, the intensity of exposure, and the type of environment studied (indoors or outdoors). Data are derived from two independent studies that simultaneously measured $PM_{2.5}$ and nicotine concentrations in the air as airborne markers of second-hand smoke exposure in public places and workplaces, including health care centres, bars, public administration offices, educational centres, and transportation. We obtained 213 simultaneous measures of airborne nicotine and $PM_{2.5}$. Nicotine in the air was measured with active samplers containing a sodium bisulphate-treated filter that was analysed by gas chromatography/mass spectrometry. $PM_{2.5}$ was measured with a SidePak AM510 Personal Aerosol Monitor. We calculated Spearman's rank correlation coefficient and its 95% confidence intervals (95% CI) between both measures for overall data and stratified by setting, type of environment (indoors/outdoors), and intensity of second-hand smoke exposure (low/high, according to the global median nicotine concentration). We also fitted generalized regression models to further explore these relationships. The median airborne nicotine concentration was 1.36 $\mu g/m^3$, and the median $PM_{2.5}$ concentration was 32.13 $\mu g/m^3$. The overall correlation between both markers was high (Spearman's rank correlation coefficient = 0.709; 95% CI: 0.635–0.770). Correlations were higher indoors (Spearman's rank correlation coefficient = 0.739; 95% CI: 0.666–0.798) and in environments with high second-hand smoke exposure (Spearman's rank correlation coefficient = 0.733; 95% CI: 0.631–0.810). The multivariate analysis adjusted

☆ Funding sources: This study was partly funded by the *Ministerio de Sanidad, Servicios Sociales e Igualdad*, the *Departament de Salut, Generalitat de Catalunya*, and the *Centro Nacional de Epidemiología, Instituto de Salud Carlos III*. EF, JMMS, AS, XS, MF, MJL, and MN also received funding from the *Department of Universities and Research*, Government of Catalonia (Grants 2009SGR192 and 2009SGR1345) and the *Instituto de Salud Carlos III* (RTIC Cancer RD06/0020/0089 and RD12/0036/0053, and project PI1102054).

* Corresponding author at: Tobacco Control Unit, Cancer Control and Prevention Programme, Institut Català d'Oncologia—ICO, Av. Granvia de L'Hospitalet 199-203, 08908 L'Hospitalet de Llobregat, Barcelona, Spain. Fax: +34 932607956.

E-mail addresses: mfu@iconcologia.net (M. Fu), jmmartinez@iconcologia.net (J.M. Martínez-Sánchez), igalan@isciii.es (I. Galán), monica.perez.rios@usc.es (M. Pérez-Ríos), fsureda@iconcologia.net (X. Sureda), mjlopez@aspb.cat (M.J. López), aschiaffino@gmail.com (A. Schiaffino), Albert.Moncada@terrassa.cat (A. Moncada), agustin.montes@usc.es (A. Montes), efernandez@iconcologia.net (E. Fernández).

[1] Dr. Manel Nebot died in October 18, 2012. He was pioneer and leader on tobacco control research and evaluation of public health interventions.

0013-9351/$ - see front matter
http://dx.doi.org/10.1016/j.envres.2013.09.003

for type of environment and intensity of second-hand smoke exposure confirmed a strong relationship (7.1% increase in geometric mean $PM_{2.5}$ concentration per μg/m^3 nicotine concentration), but only in indoor environments in a stratified analysis (6.7% increase; 95% CI: 4.3–9.1%). Although the overall correlation between airborne nicotine and $PM_{2.5}$ is high, there is some variability regarding the type of environment and the intensity of second-hand smoke exposure. In the absence of other sources of combustion, air nicotine and $PM_{2.5}$ measures can be used indoors, while $PM_{2.5}$ should be used outdoors with caution.

1. Introduction

Second-hand smoke is a complex chemical mixture derived from combustion compounds in tobacco smoke. Its inhalation causes adverse health outcomes, particularly cancer and cardiovascular and pulmonary diseases (US Department of Health and Human Services, 2010). Because involuntary exposure to tobacco smoke is recognised as a cause of disease and death, it is of relevance to assess it using objective measures.

Nicotine in the air and particulate matter of diameter ≤ 2.5 μm ($PM_{2.5}$) are reliable indicators of second-hand smoke exposure, and a correlation between them has been described in some studies (Apelberg et al., 2013; Avila-Tang et al., 2010). However, we are unaware of studies describing the association between these airborne markers that take into account some characteristics of the exposure, such as its intensity, the setting studied, and whether the measurements were taken indoors or outdoors. This study aims to assess the relationship between airborne nicotine and $PM_{2.5}$ as markers of second-hand smoke exposure considering these contextual variables.

2. Materials and methods

Data are derived from two independent studies (López et al., 2013; Sureda et al., 2012) that simultaneously measured both airborne nicotine and $PM_{2.5}$ as markers of second-hand smoke exposure in a variety of different settings: health care centres, bars, public administration offices, educational centres, and transportation. From the first study (López et al., 2013), conducted in 2010–2011, we included 185 paired measures in bars from three regions of Spain. The study followed a multistage design, first involving random selection of a sample of districts and census tracts weighted by population size, and then a selection of a random sample of venues located in the selected census tracts. Fast food venues, musical venues with bar service that were open at night, and restaurants without bar service were excluded. In addition, hospitality venues where smoking was already banned before the enactment of Spanish law 42/2010 and venues with fewer than 3 clients at the time of measurement were also excluded from the study. These venues were substituted by the closest venue fulfilling the inclusion criteria. The second study (Sureda et al., 2012), conducted in 2010, included a convenience sample of 47 venues with second-hand smoke exposure at entrances of some public buildings in the metropolitan area of Barcelona, Spain. Twenty-eight out of the 47 venues had paired measurements of both airborne nicotine and $PM_{2.5}$. These venues were public administration buildings, educational places, public transport stations, and healthcare centres. The criteria for inclusion were: an interior space adjacent to an outdoor area, separated by a doorway providing direct access; at least one room physically separated from the hall; places with cooking facilities must be physically separated from the hall and from the other interior room. Moreover, at least two lit cigarettes were to be observed at main entrances (outdoors) during the time of the measurement.

The studies' designs and specific methods used for the measurements were similar. In brief, $PM_{2.5}$ was measured during 30 min with SidePak AM510 Personal Aerosol Monitors at 1-second sampling intervals; the median of all values in each location was computed. A calibration factor of 0.52 derived from an experiment with a BAM-1020 instrument was applied to all $PM_{2.5}$ values (Ruprecht et al., 2011). In addition, active airborne nicotine samples were simultaneously taken using a sampler device of 37 mm in diameter containing a filter treated with sodium bisulphate. The sampler device was connected to a pump (flow 3 L/min) with a Tygon tube. Nicotine concentration in μg/m^3 was analysed by gas chromatography/mass spectrometry in the Laboratory of the Public Health Agency of Barcelona. The quantification limit was 5 ng per filter, equivalent to 0.06 μg/m^3 of nicotine per 30 min of exposure. Analysis procedure was accredited by the Spanish Accreditation Body (ENAC) according to the ISO 17025. More specific details are provided in a supplement.

For statistical analyses, we calculated medians and interquartile ranges of nicotine and $PM_{2.5}$ measurements. We computed Spearman's rank correlation coefficient and its 95% confidence intervals (95% CI) to assess the relationship between both types of measurements, stratifying by variables of interest: setting studied (health care centres, bars, public administration offices, educational centres, transportation), type of environment (indoors or outdoors, i.e., the area within a radius of 5 m over the door being accessed by the public), and intensity of second-hand smoke exposure (low or high, depending on whether the values were below or above the median value of all nicotine measurements). We computed correlations when there were more than 10 paired observations. Finally, we fitted a generalized linear regression model with the Gaussian family, with log-$PM_{2.5}$ concentration as the dependent variable and nicotine concentration, type of environment, and intensity of second-hand smoke exposure as the independent variables.

3. Results

The pooled data included 213 paired measurements of airborne nicotine and $PM_{2.5}$. The median concentration of nicotine was 1.36 μg/m^3 and the corresponding value of $PM_{2.5}$ was 32.13 μg/m^3 (Table 1). The overall correlation between both markers was high (Spearman's rank correlation coefficient = 0.709; 95% CI: 0.635–0.770; Table 1). By setting, the correlation was higher in health care centres (Spearman's rank correlation coefficient = 0.857; 95% CI: 0.448–0.969) and in bars (Spearman's rank correlation coefficient = 0.739; 95% CI: 0.666–0.798), while a poor non-significant correlation was observed in educational centres (Spearman's rank correlation coefficient = 0.231; 95% CI: −0.366 to 0.694). The correlation was higher indoors and in environments with high second-hand smoke exposure (Table 1). When these two variables were considered together, we observed a relevant and significant correlation only in indoor high-exposure conditions (Spearman's rank correlation coefficient = 0.709; 95% CI: 0.594–0.795; n = 98).

Generalized linear regression analyses confirmed the relationship between $PM_{2.5}$ and airborne nicotine concentration adjusting for type of environment and intensity of second-hand smoke exposure (Table 2). Overall, geometric mean $PM_{2.5}$ concentration increased significantly by 7.1% (95% CI: 4.9–9.4%) per 1 μg/m^3 increase of nicotine concentration. When we stratified by type of environment, this significant association was only confirmed in indoor venues, with a 6.7% increase (95% CI: 4.3–9.1%) in geometric mean $PM_{2.5}$ concentration per 1 μg/m^3 increase of nicotine concentration (Table 2).

4. Discussion

We observed an overall high correlation between airborne nicotine and $PM_{2.5}$ measurements. As summarised in Table 3, previous studies also found good correlations between them, with 61% of the values over 0.5. Nevertheless, very low correlation was observed in non-smoking game rooms in Korea (Kim et al., 2010). Low correlations were also reported in another study, in which high variability was observed in data from restaurants in different countries (Bohanon et al., 2003). A low correlation of 0.365 was observed outdoors (Sureda, et al., 2012, data included in this work); in contrast, highest correlations (over 0.7) were commonly

Table 1
Median nicotine and $PM_{2.5}$ concentrations (and interquartile ranges), and Spearman's rank correlation coefficients between these airborne markers according to selected variables. Spain, 2010–2011.

	n	Nicotine (μg/m^3) median (interquartile range)	$PM_{2.5}$ (μg/m^3) median (interquartile range)	Spearman's rank correlation coefficient (95% confidence interval)
All paired measurements	213	1.36 (0.43–4.33)	32.13 (16.32–193.04)	0.709 (0.635–0.770)
By setting				
Health care centre	9	1.07 (0.81–2.41)	16.12 (12.74–21.06)	–
Bar	185	1.54 (0.41–4.83)	44.88 (17.60–226.19)	0.739 (0.666–0.798)
Public administration	4	1.56 (0.59–2.75)	16.38 (11.05–17.03)	–
Educational centre	13	0.52 (0.21–1.04)	17.68 (8.32–22.62)	0.231 (−0.366 to 0.694)
Transportation	2	2.05 (1.30–2.79)	20.54 (17.68–23.40)	–
By type of environment				
Indoors	185	1.54 (0.41–4.83)	44.88 (17.60–226.19)	0.739 (0.666–0.798)
Outdoors	28	0.81 (0.54–1.52)	16.64 (9.75—20.67)	0.366 (−0.008 to 0.650)
By intensity of second-hand smoke exposure[a]				
Low exposure	106	0.43 (0.16–0.73)	17.00 (12.62–29.24)	0.142 (−0.050 to 0.324)
High exposure	107	4.31 (2.16–10.46)	148.92 (33.15–320.79)	0.733 (0.631–0.810)

n: Paired observations.

[a] According to the median of all nicotine measurements.

Table 2
Average increase in PM2.5 concentration per unit of airborne nicotine adjusted for type of environment and intensity of second-hand smoke exposure. Spain, 2010–2011.

Variables	Coefficient	Percentage change[a] (95% confidence interval)
Overall		
Nicotine (μg/m^3)	0.069	7.1 (4.9–9.4)
Indoor environment[b]	0.908	148.0 (69.8–262.1)
High intensity of exposure[c]	1.144	214.0 (133.9–321.3)
By type of environment		
Indoors		
Nicotine (μg/m^3)	0.064	6.7 (4.3–9.1)
High intensity of exposure[c]	1.285	261.5 (159.8–403.0)
Outdoors		
Nicotine (μg/m^3)	0.187	20.5 (−10.3 to 61.9)
High intensity of exposure[c]	0.096	10.1 (−42.2 to 109.6)

[a] Percentage change in geometric mean PM2.5 concentration.
[b] Outdoor environment as reference.
[c] Low intensity as reference.

observed in indoor venues such as pubs, bars, and restaurants, where second-hand smoke exposure used to be very high (Agbenyikey et al., 2011; Bolte et al., 2008; Ellingsen et al., 2006). This fact could explain the differences observed in our study between indoor and outdoor environments, because available data on indoor venues only included bars.

Airborne concentrations depend on some physical factors, such as the volume of the environment measured and the ventilation (Apelberg et al., 2013). In a study that measured respirable particles in indoor environments, it was observed that second-hand smoke levels remained relatively stable and decayed over time until ventilation was produced, while in outdoor environments second-hand smoke levels dropped immediately to background levels once the source was extinguished (Klepeis et al., 2007). This observation might partially explain the low correlations that we observed outdoors. Particularly in environments with low second-hand smoke exposure, airborne nicotine is non-detectable in the absence of tobacco smoke, while background levels of $PM_{2.5}$ from other sources are always present (Leaderer and Hammond, 1991).

Most studies that calculated a correlation between airborne nicotine and particulate matter assessed only indoor settings; only two of them included both indoor and outdoor assessments of second-hand smoke exposure, although they did not calculate a correlation for each (Baek et al., 1997; Sureda et al., 2012). Although the studies included in Table 3 described the average concentration of these markers according to different context variables, only the study by Kim et al. (2010) calculated a correlation with respect to those variables, including a correlation with respect to the type of environment (smoking and non-smoking rooms). To our knowledge, this is the first study to assess the correlation between these two airborne markers according to various contextual variables. Additional strength of this investigation derives, in our view, from the methodology used in both studies to collect the data: nicotine and $PM_{2.5}$ were measured simultaneously, and the same protocol and the same devices were used for all measurements. As a counterpart, a possible limitation of this work relates to the lack of generalizability, because only one study employed random selection of the venues. Nevertheless, the main focus of this investigation was to examine how these markers behave in different environments. Another potential limitation of this study is that available data from indoor measurements only included bars, which used to be high-exposed places, and this could partially explain the differences observed between indoor and outdoor venues. Previous research on second-hand smoke exposure has been mostly focused on hospitality venues, and studies generally had limited sample sizes (Table 3). While we were able to include an important number of paired measures ($n=213$), the relatively small sample size in some settings precluded stratification according to other characteristics of interest, such as the number of smokers, type of enclosure, or meteorological variables. Taking into account that the type of setting and other contextual variables should be considered for stratification, larger, prospective studies are needed to assess the correlation between $PM_{2.5}$ and airborne nicotine, especially in outdoor settings.

In conclusion, the association between airborne nicotine and $PM_{2.5}$ is high, particularly indoors and when high second-hand smoke levels are observed, confirming the reliability of both measures to estimate second-hand smoke exposure. The observed correlations suggest that both measures can be used indoors when other sources of combustion are absent. Nevertheless, the current data suggest that assessment of second-hand smoke outdoors cannot be based solely on $PM_{2.5}$ measurements, given the limited correlation observed. Additional research on second-hand smoke exposure assessment outdoors still seems necessary (Sureda et al., 2013).

Table 3
Studies reporting a correlation between airborne nicotine and particulate matter (PubMed search and cross-references, 1980–2012).

First author, publication date (country)	Setting	Environment	Time of measurements	Nicotine (μg/m^3)	PM type[a] and value (μg/m^3)	N° of paired measurements	Correlation coefficient[b]
Weber and Fischer (1980) (Switzerland)	Workplaces	Indoors	2 days	$n=160$ Mean = 1.1	$n=489$ PM_{10} mean = 170	158	*Pearson's* $r=0.41$
Thompson et al. (1989) (USA)	An unoccupied office	Indoors	Nicotine: 1 h Minimum PM: NA	$n=10$ Mean = 16.91[c] Median = 6.50[c]	$n=10$ PM mean = 93.36[c] PM median = 59.10[c]	10	*Pearson's* $r=0.988$[c] *Spearman's* $r=0.952$[c] (Both $p<0.01$)
Miesner et al. (1989) (USA)	Public facilities: bars/restaurants, transport stations, office buildings, healthcare centres	Indoors	3–16 h < 1 h for real-time measures	$n=16$ Mean = 6.28[c] Median = 3.15[c]	$n=16$ $PM_{2.5}$ mean = 93.79[c] $PM_{2.5}$ median = 61.90[c]	16	*Pearson's* $r=0.884$[c] *Spearman's* $r=0.841$[c] (Both $p<0.0$ 1)
Coultas et al. (1990) (USA)	Homes	Indoors	24 h	$n=10$ mean values Range = 0.6–6.9	$n=10$ mean values $PM_{2.5}$ range = 32.4–76.9	99	*Spearman's* $r=0.54$ ($p<0.0001$)
Leaderer and Hammond (1991) (USA)	Homes	Indoors	1 week	$n=96$ Mean = 1.1	$n=96$ $RSP_{2.5}$ mean = 29.4	96	*Pearson's* $r=0.842$[d]
Kado et al. (1991) (USA)	Casinos	Indoors	40 min–6 h	$n=6$ Median = 8.02	$n=6$ Total PM median = 200	12	*Spearman's* $r=0.54$
	Bingo parlours	Indoors	40 min–6 h	$n=6$ Median = 65.5	$n=6$ Total PM median = 482		
Turner et al. (1992) (USA)	Offices: general commercial offices, banking offices, cafeterias, newspaper offices, institutional (church, hospital, correctional or educational)	Indoors	1 h	Total $n=585$ Mean = 3.8 Non-smoking activity $n=254$ Mean = 0.2 Smoking activity $n=331$ Mean = 6.7	Total $n=585$ $RSP_{3.5}$ mean = 35 Non-smoking activity $n=254$ $RSP_{3.5}$ mean = 20 Smoking activity $n=331$ $RSP_{3.5}$ mean = 46	585	*Pearson's* $r=0.7345$ ($p<0.01$)
Baek et al. (1997) (Korea)	Homes	Indoors	2 h	$n=$ NA Mean = 1.8 Median = 0.6	$n=$ NA $RSP_{3.5}$ mean = 100 $RSP_{3.5}$ median = 91	26	*Pearson's* $r=0.42$ ($p<0.01$) Computed only for smoking indoor environments ($n=104$)
		Outdoors	2 h	$n=$ NA Mean = 0.3 Median = 0.3	$n=$ NA $RSP_{3.5}$ mean = 80 $RSP_{3.5}$ median = 68	NA	
	Offices	Indoors	2 h	$n=$ NA Mean = 2.5 Median = 1.1	$n=$ NA $RSP_{3.5}$ mean = 99 $RSP_{3.5}$ median = 92	30	
		Outdoors	2 h	$n=$ NA Mean = 0.4 Median = 0.3	$n=$ NA $RSP_{3.5}$ mean = 78 $RSP_{3.5}$ median = 70	NA	
	Restaurants	Indoors	2 h	$n=48$ Mean = 4.8 Median = 3.0	$n=48$ $RSP_{3.5}$ mean = 171 $RSP_{3.5}$ median = 159	48	
		Outdoors	2 h	$n=$ NA Mean = 0.4 Median = 0.3	$n=$ NA $RSP_{3.5}$ mean = 72 $RSP_{3.5}$ median = 67	NA	
Bohanon et al. (2003) (Multicountry)	Restaurants (France)	Indoors	3–4 h	$n=15$ Mean = 30.3	$n=15$ $RSP_{3.5-4}$ mean = 188	15	*Pearson's* $r=0.054$ *Spearman's* $r=-0.002$

				Median = 24.1	$RSP_{3.5-4}$ median = 194		
	Restaurants (Japan)	Indoors	3–4 h	*n* = 16 Mean = 11.7 Median = 11.1	*n* = 16 $RSP_{3.5-4}$ mean = 242 $RSP_{3.5-4}$ median = 194	16	*Pearson's r* = −0.260 *Spearman's r* = −0.294
	Restaurants (Korea)	Indoors	3–4 h	*n* = 47 Mean = 5.72 Median = 3.95	*n* = 50 $RSP_{3.5-4}$ mean = 109 $RSP_{3.5-4}$ median = 107	NA	*Pearson's r* = 0.134 *Spearman's r* = 0.100
	Restaurants (Switzerland)	Indoors	3–4 h	*n* = 32 Mean = 7.81 Median = 3.98	*n* = 31 $RSP_{3.5-4}$ mean = 92.0 $RSP_{3.5-4}$ median = 74.6	NA	*Pearson's r* = 0.930 *Speaman's r* = 0.923
	Restaurants (UK)	Indoors	3–4 h	*n* = 20 Mean = 9.78 Median = 10.1	*n* = 12 $RSP_{3.5-4}$ mean = 195 $RSP_{3.5-4}$ median = 201	NA	*Pearson's r* = −0.150 *Spearman's r* = −0.063
Ellingsen et al. (2006) (Norway)	Restaurants, pubs, nightclubs	Indoors	NA	Pre-ban *n* = 58 Mean = 28.3 Post-ban *n* = 96 Mean = 0.6	Pre-ban *n* = 71 Total dust mean = 262 Post-ban *n* = 93 Total dust mean = 77	48	*Pearson's r* = 0.86 ($p < 0.001$)
Rumchev et al. (2008) (Australia)	Homes	Indoors	24 h	Non-smoking home *n* = 53 Median = 0.10 Smoking home *n* = 39 Median = 0.75	Non-smoking home *n* = 53 PM_{10} median = 27.0 Smoking home *n* = 39 PM_{10} median = 48.9	92	*Spearman's r* = 0.7 ($p < 0.01$)
Bolte et al. (2008) (Germany)	Cafés/restaurants	Indoors	4 h	*n* = 11 Mean = 21.3 Median = 15.0	*n* = 11 $PM_{2.5}$ mean = 205.9[e], 224.3[f] $PM_{2.5}$ median = 178.0[e], 163.8[f] PM_1 mean = 205.6[f] PM_1 median = 155.6[f] PM_{10} mean = 260.3[f] PM_{10} median = 199.2[f]	28	*Spearman's r* = 0.932[e] *Spearman's r* = 0.930[f]
	Pubs/bars	Indoors	4 h	*n* = 7 Mean = 53.7 Median = 31.0	*n* = 7 $PM_{2.5}$ mean = 382.6[e], 397.5[f] $PM_{2.5}$ median = 192.0[e], 202.8[f] PM_1 mean = 370.7[f] PM_1 median = 195.0[f] PM_{10} mean = 422.2[f] PM_{10} median = 218.9[f]		
	Discotheques/clubs	Indoors	4 h	*n* = 10 Mean = 226.6 Median = 192.5	*n* = 10 $PM_{2.5}$ mean = 1406.1[e], 1210.1[f] $PM_{2.5}$ median = 807.5[e], 869.3[f] PM_1 mean = 948.4[f] PM_1 median = 819.3[f] PM_{10} mean = 1344.5[f] PM_{10} median = 1014.3[f]		

Table 3 (*continued*)

First author, publication date (country)	Setting	Environment	Time of measurements	Nicotine (μg/m^3)	PM type[a] and value (μg/m^3)	N° of paired measurements	Correlation coefficient[b]
Chiu et al. (2009) (USA)	Truck cabs	Indoors	NA	$n=16$ Mean=8.20[e], 7.10[f]	$n=16$ $PM_{2.5}$ mean=56.5	16	*Pearson's* $r=0.92$[g] *Pearson's* $r=0.93$[h]
Sureda et al. (2010) (Spain)	Hospitals	Indoors	Nicotine: 7 days PM: 15 min	$n=28$ Median=0.05	$n=33$ $PM_{2.5}$ median=15.60	28	*Spearman's* $r=0.644$
Kim et al. (2010) (Korea)	Computer game rooms	Indoors	Nicotine: 7 days PM: 20 min	Smoking areas $n=18$ Median=0.41 Non-smoking areas $n=19$ Median=0.12	Smoking areas $n=28$ $PM_{2.5}$ median=69.5 Non-smoking areas $n=22$ $PM_{2.5}$ median=34.0	1818	*Spearman's* $r=0.44$ ($p=0.06$) *Spearman's* $r=0.08$ (NS)
Butz et al. (2011) (USA)	Homes	Indoors	NA	$n=110$ Mean=1.43 1 smoker at home Mean=0.99 >1 smoker at home Mean=1.98 Non-smoker caregiver Mean=0.71 Smoker caregiver Mean=1.79 No air conditioner Mean=1.20 Air conditioner Mean=1.60	$n=109$ $PM_{2.5}$ mean=NA 1 smoker at home $PM_{2.5}$ mean=33.96 >1 smoker at home $PM_{2.5}$ mean=44.71 Non-smoker caregiver $PM_{2.5}$ mean=28.88 Smoker caregiver $PM_{2.5}$ mean=44.16 No air conditioner $PM_{2.5}$ mean=38.39 Air conditioner $PM_{2.5}$ mean=40.10	NA	*Spearman's* $r=0.76$
Agbenyikey et al. (2011) (Ghana)	Restaurants, bars, nightclubs, and casinos	Indoors	nicotine: 7 days PM: ≥ 30 min	Smoking venues $n=8$ Median=1.83 Non-smoking venues $n=2$ Median=0.03	Smoking venues $n=8$ $PM_{2.5}$ median=905 Non-smoking venues $n=2$ $PM_{2.5}$ median=26.5	10	*Spearman's* $r=0.76$ ($p<0.001$)
Sureda et al. (2012) (Spain)	Public offices, educational centres, transportation, healthcare centres	Outdoors	30 min	$n=28$ Median=0.81	$n=47$ $PM_{2.5}$ median=17.16	28	*Spearman's* $r=0.365$

NA: not available.
NS: not significant.

[a] PM: particulate matter; RSP: respirable suspended particles.
[b] Including the level of significance when available.
[c] Computed from the data available in the paper.
[d] Computed from the square root of $R^2=0.71$.
[e] Measured cumulatively by gravimetry.
[f] Measured continuously with a laser aerosol spectrometer.
[g] Measured with a "stand-alone" filter.
[h] Measured with a filter behind a PM filter ("in-line sampler").

Acknowledgments

The authors would like to thank Fernando Agüero, Ester Basart, Marta Bosch, Eugenio Calciati, Gisèle Contreras, Elena García, Jordi García, Oleguer Gispert, Teresa Hernández, Matilde López, Sandra Manzanares, Joana Martín, Miquel Molist, Carles Mundet, Anna C. Osanz, Magda Pagès, Angel Rodríguez, Montse Salat, Meritxell Serres, and Martí Tantinyà (Catalonia); Gestaly Valencia (Galicia); and Elga Mayo (Madrid) for data collection. We thank Marina Julià and Esteve Saltó for fieldwork coordination in Catalonia. We also thank Francesc Centrich and Glòria Muñoz (Laboratory of Public Health, Agència de Salut Pública de Barcelona) for nicotine analyses.

Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.envres.2013.09.003.

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