Science of the Total Environment 785 (2021) 147111

Contents lists available at ScienceDirect

Science of the Total Environment

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

Analysis of spatial factors, time-activity and infiltration on outdoor generated $PM_{2.5}$ exposures of school children in five European cities

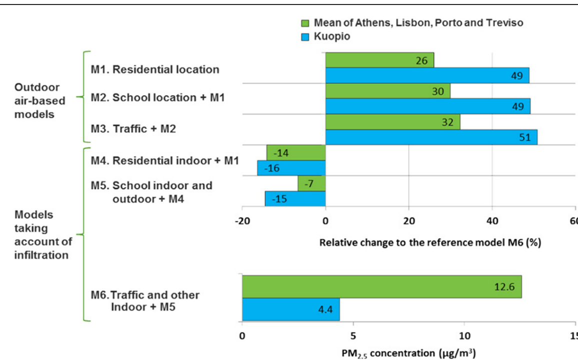
Antti Korhonen [a,b,*], Hélder Relvas [c], Ana Isabel Miranda [c], Joana Ferreira [c], Diogo Lopes [c], Sandra Rafael [c], Susana Marta Almeida [d], Tiago Faria [d], Vânia Martins [d], Nuno Canha [d], Evangelia Diapouli [e], Konstantinos Eleftheriadis [e], Eleftheria Chalvatzaki [f], Mihalis Lazaridis [f], Heli Lehtomäki [a,g], Isabell Rumrich [a], Otto Hänninen [a]

[a] Department of Public Health Solutions, Finnish Institute for Health and Welfare (THL), 70701 Kuopio, Finland
[b] Department of Environmental and Biological Sciences, University of Eastern Finland, 70701 Kuopio, Finland
[c] CESAM, Department of Environment and Planning, University of Aveiro, Aveiro, Portugal
[d] Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Bobadela, Portugal
[e] Institute of Nuclear & Radiological Sciences & Technology, Energy & Safety, N.C.S.R. "Demokritos", Agia Paraskevi, 15310 Athens, Greece
[f] School of Environmental Engineering, Technical University of Crete, 73100 Chania, Greece
[g] Faculty of Health Sciences, School of Pharmacy, University of Eastern Finland (UEF), 70701 Kuopio, Finland

HIGHLIGHTS

- Exposure to outdoor $PM_{2.5}$ varies considerably depending on the modelling approach.
- Exposure occurs mainly indoors, although infiltration decreases the concentrations.
- Inclusion of school and traffic microenvironments increased the exposure estimates.
- Indoor-generated sources potentially important contributors to the total exposure.
- Time-activity, spatial mobility and infiltration important in exposure modelling

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 9 December 2020
Received in revised form 4 March 2021
Accepted 8 April 2021
Available online 16 April 2021

Editor: Lidia Morawska

Keywords:
Exposure
Misclassification
Time-activity

ABSTRACT

Atmospheric particles are a major environmental health risk. Assessments of air pollution related health burden are often based on outdoor concentrations estimated at residential locations, ignoring spatial mobility, time-activity patterns, and indoor exposures. The aim of this work is to quantify impacts of these factors on outdoor-originated fine particle exposures of school children.
We apply nested WRF-CAMx modelling of $PM_{2.5}$ concentrations, gridded population, and school location data. Infiltration and enrichment factors were collected and applied to Athens, Kuopio, Lisbon, Porto, and Treviso. Exposures of school children were calculated for residential and school outdoor and indoor, other indoor, and traffic microenvironments. Combined with time-activity patterns six exposure models were created. Model complexity was increased incrementally starting from residential and school outdoor exposures.
Even though levels in traffic and outdoors were considerably higher, 80–84% of the exposure to outdoor particles occurred in indoor environments. The simplest and also commonly used approach of using residential outdoor concentrations as population exposure descriptor (model 1), led on average to 26% higher estimates (15.7 $\mu g/m^3$)

* Corresponding author at: Department of Public Health Solutions, Finnish Institute for Health and Welfare (THL), 70701 Kuopio, Finland.
E-mail address: antti.korhonen@thl.fi (A. Korhonen).

https://doi.org/10.1016/j.scitotenv.2021.147111

Spatial mobility
Infiltration
Microenvironment
Particulate matter
Health impact assessment

compared with the most complex model (# 6) including home and school outdoor and indoor, other indoor and traffic microenvironments (12.5 μg/m^3). These results emphasize the importance of including spatial mobility, time-activity and infiltration to reduce bias in exposure estimates.

1. Introduction

Air pollution, especially fine particles ($PM_{2.5}$), is a leading environmental health risk worldwide (WHO, 2016). In the European Union around 400 thousand premature deaths per year are associated with $PM_{2.5}$ (EEA, 2020a). Epidemiological studies of air pollution have been conventionally relying on outdoor air concentrations measured at fixed locations or on predicted outdoor concentrations calculated e.g. with land use regression or chemical transport models (Beelen et al., 2014; de Hoogh et al., 2014; Dockery et al., 1993; Im et al., 2018). Observations have been considered more reliable than predicted values, but spatial representativeness of the air quality models are far better than measured concentrations.

Besides poor spatial coverage of observations or air quality models' difficulties to capture variability of concentrations in diverse urban environments, recent studies have shown that relying only on outdoor concentrations as surrogate for population exposure leads to a misclassification in health burden assessments. Kazakos et al. (2020) estimated that ignoring infiltration of outdoor $PM_{2.5}$ indoors, spatial mobility, and time-activity leads to overestimated results. Using their most complex model in London, which included 4 microenvironments (MEs) (residential outdoor, indoor, above- and underground transportation), led to a 24% reduction in mortality estimates in comparison to results obtained only using residential outdoor concentrations ($n = 1541$). Indoors was the largest contributor (83%) to the total $PM_{2.5}$ exposure the fact that. Despite only 0.4% of the time was spent in London Underground was the second largest contributor (15%). Singh et al. (2020) estimated also $PM_{2.5}$ exposures in London using a modelling approach, which integrated time-activity and spatial mobility in three microenvironments (home, work and transport). The exposure (approximately 9 μg/m^3) was 28% lower in comparison to outdoor only exposures at residential locations, with 85% of the exposures occurring in residential and workplaces and 15% in transport MEs.

In general, people in Western countries spend the majority of their time indoors. Hussein et al. (2012) conducted a survey in Helsinki, Finland, and reported that people aged between 2 and 93 years old ($n = 167$) spent on average 82–92% of their time indoors, 5–14% outdoors, and 3–5% in transit. In colder periods (−13 to 2 °C) people spent more time indoors (90–92%) in comparison to warmer period (8–15 °C) when people spent 82 and 88% indoors on weekend and workday, respectively. Faria et al. (2020) reported that school children ($n = 1189$) in Lisbon, Portugal, aged from 5 to 10 years old, on weekdays 86% of their time indoors, mostly at home (56%) and in classroom (27%), 10% outdoors and 3% in transport. On weekends the same period of time (87%) was spent indoors (77% at home and 8% other indoor locations, like shops and restaurants), slightly less time outdoors (9%) and slightly more in transports (4%). Cunha-Lopes et al. (2019) observed similar results in a survey conducted among a small number of Lisbon school children ($n = 9$). During the week children spent more than 80% of their time indoors (55% at home and 22% in classroom), about 9% outdoors and 5% in transports.

Although the time spent in transport is small, a higher concentration during commuting makes it an important contributor to exposures. Fine particulate matter concentrations during commuting are higher compared to the background levels and depend on the type of commuting, route choice and ventilation settings of the vehicle. In Lisbon, Correia et al. (2020) measured highest $PM_{2.5}$ concentrations in the metro (37.8 μg/m^3), followed by car (33.7 μg/m^3), bicycle (30.5 μg/m^3), and bus (28 μg/m^3). Concentrations in all transport modes were approximately 2 times higher in comparison to traffic station and around 3 times higher compared to background station measurements. Lower concentration levels of bicycle rides were linked to route that was in some parts far away from traffic emissions, i.e. roads. Similarly, buses exhibited lower concentrations because they were running on dedicated lanes and were therefore less prone to traffic jams. In addition, filters may have been more efficient to remove particles from the outdoor air entering the vehicle cabin. Results of de Nazelle et al. (2017) report in a review article concerning Europe that pedestrians were consistently least exposed to $PM_{2.5}$ in comparison to all other transport modes. Commuters in bicycle, car and bus modes were exposed on average to 1.3, 1.4 and 1.5 times higher concentrations, respectively, in comparison to pedestrians. Concentrations during commuting were 2–2.5 times higher than the background levels. In undergrounds, exposure levels are considerably higher; e.g. Smith et al. (2020) measured average $PM_{2.5}$ concentration of 88 μg/m^3 in London underground, while outdoor levels were 19–22 μg/m^3 at background and central London roadside environments.

In residences and offices, infiltration of outdoor air pollution is a major factor affecting the indoor air $PM_{2.5}$ levels. Infiltration factors for $PM_{2.5}$ in residences ranged from 0.59 in Helsinki to 0.70 in Athens, with Basle (0.63) and Prague (0.61) in between (Hänninen et al., 2004). Infiltration was higher in buildings with natural ventilation or mechanical ventilation with low or non-existent particle filtration. Kalimeri et al. (2019) study showed that the $PM_{2.5}$ indoor/outdoor (I/O) ratio in offices (0.62) was below 1.0, whereas in schools this ratio was above 1.0 (1.44). These findings revealed the importance of outdoor air particles in indoor air quality, but also that in schools there are significant indoor sources. Schools are often more crowded than offices and students are more active, favouring resuspension of particles. This difference in I/O ratios was also explained by the fact that the majority of the offices were mechanically ventilated, whereas schools were naturally ventilated. Morawska et al. (2017) ended up with similar results in their review. In homes and offices, outdoor air was the major source of $PM_{2.5}$, while in schools or day care present indoor sources, like children's activity and PM resuspension, as well as poor ventilation, were major factors affecting the $PM_{2.5}$ concentrations. In residences, cooking, candle burning and wood stove burning devices are among the most significant indoor $PM_{2.5}$ sources (MacNeill et al., 2012; Siponen et al., 2019).

The overall aim of this paper is to quantify exposures of outdoor-originated $PM_{2.5}$ and bias related to the most common use of outdoor concentrations as population exposure descriptor when using atmospheric modelling as source. Specifically, we estimate how (i) spatial mobility, (ii) time-activity, and (iii) infiltration of outdoor particles indoors affect the $PM_{2.5}$ exposures of school children, selected as target population due to available school locations at community level. Furthermore, (iv) we quantify uncertainties due to modelled particle concentrations and their influence on exposure estimates, health impact assessment and discuss implications for air pollution epidemiology.

2. Material and methods

In this work, annual outdoor-originated atmospheric aerosol, defined as fine particulate matter ($PM_{2.5}$), exposures of school children in 2015 were estimated in the context of LIFE Index-Air project for Athens, Kuopio, Lisbon, Porto and Treviso (Fig. 1). Spatially distributed outdoor concentrations were calculated with the air quality modelling system combining Weather Research and Forecasting (WRF) model

Fig. 1. Four cities in the Mediterranean climate and one in Northern Europe were included in this study.

with Comprehensive Air Quality Model with Extensions (CAMx), resulting in a 3D chemical transportation modelling system for particles (Ferreira et al., 2020). The influence of spatial mobility, time-activity and infiltration of outdoor particles indoors was taken into account incrementally. In total, six exposure models were created. The biases related to each approach using the most complex and presumably most accurate model as the reference point, was evaluated.

The modelling domain sizes of the WRF-CAMx in South European cities were between 2100 and 2300 km^2 and in Kuopio 320 km^2 (Table 1). There was a high variability in domain populations, ranging from 84 thousand in Kuopio to 3.3 million in Athens. Infiltration of outdoor generated particles indoors was estimated to be higher in South European cities.

2.1. Air quality modelling and observations of outdoor-originated $PM_{2.5}$

Outdoor fine particulate matter concentrations were calculated on hourly resolution with the WRF-CAMx air quality modelling system and were averaged to annual level. The model was applied for the urban regions of Athens, Kuopio, Lisbon, Porto and Treviso, for 2015, following a nesting approach starting by a coarse domain over Europe with a 0.25° grid cell size until reaching the domains of interest, at 0.01° resolution. The mesoscale numerical weather prediction system Weather Research and Forecasting model (WRF, version 3.7.1) (Skamarock et al., 2008) simulated the meteorological fields, which were used as inputs for the three dimensional chemical transport model Comprehensive Air Quality Model with Extensions (CAMx version 6.3) (ENVIRON, 2016; Ferreira et al., 2020). The European Centre for Medium-Range Weather Forecasts' (ECMWF) ERA-Interim atmospheric reanalysis data at 6 h temporal and 0.75° spatial resolution was used as meteorological forcing for the WRF model (Dee et al., 2011). For the European domain, initial and boundary conditions with temporal resolution of 6 h were provided by the global chemical model MOZART (Emmons et al., 2010). Anthropogenic emissions reported by the Member States of the European Union were derived from the European emission inventory (EMEP) with a resolution of 0.1° and were spatially disaggregated to 1 km resolution (Ferreira et al., 2020). Hourly concentrations were modelled with a spatial resolution of approximately 1.1 km × 0.5 km in Kuopio and 1.1 km × 0.8 km in the other cities residing in the Mediterranean region.

Moreover, observed outdoor PM concentration data collected in 2015 were compiled from national and European air quality databases (APA, 2020; EEA, 2020b; FMI, 2020; YPEKA, 2020). In total 17 $PM_{2.5}$ measurement stations were included. Six stations were located in Athens of which 3 were classified as suburban background, 2 as traffic and 1 as industrial station. In Lisbon there were 5 (4 urban background and 1 traffic), in Kuopio 3 (1 suburban background, 1 urban background and 1 traffic), in Treviso 2 (1 suburban and 1 urban background) and in Porto one urban background station.

To estimate misclassification related to modelled $PM_{2.5}$ concentrations and discuss its influence to exposure, health impact estimates and epidemiology, predicted and observed annual average concentrations at measurement station locations were compared. Differences were quantified as absolute difference (mean bias, MB), underestimation of variance (UEV) and random error (RE).

2.2. Gridded population, school data, time-activity patterns, infiltration and traffic enrichment factors

The GEOSTAT 2011 population data provided at 1 km × 1 km resolution across Europe (Eurostat, 2016) was obtained from Eurostat and

Table 1
Characteristics of modelling domains, populations, and input parameters of the target cities.

City, country	Athens, Greece	Lisbon, Portugal	Porto, Portugal	Treviso, Italy	Kuopio, Finland
Domain size (grid cell size, area)	0.88 × 1.1km^2 43x54km^2	0.87 × 1.1km^2 42x54km^2	0.84 × 1.1km^2 41x54km^2	0.78 × 1.1km^2 38x54km^2	0.51 × 1.1km^2 12x27km^2
Inhabitants (thousands)[a]	3300	2300	1200	880	84
Pupils (thousands)	300[b]	72	32	81[b]	7.8
Schools (n)	n/a	212	105	n/a	20
School/home $PM_{2.5}$ concentration ratio	1.19	1.19	1.09	1.19	1.02
Infiltration factor, residences	0.66[c]	0.66[c]	0.66[c]	0.66[c]	0.55
±SD, ±SE	±0.41, ±0.023	±0.41, ±0.023	±0.41, ±0.023	±0.41, ±0.023	±0.16, ±0.017
Infiltration factor, schools	0.82[c]	0.82[c]	0.82[c]	0.82[c]	0.55[d]
±SD, ±SE	±0.82, ±0.14	±0.82, ±0.14	±0.82, ±0.14	±0.82, ±0.14	±0.16, ±0.017
Enrichment factor, traffic[e]	1.58	1.35	1.35	1.58	1.30

n/a: not available.
[a] Inhabitants in the domain.
[b] Number of 6–14-year old children.
[c] Parameters estimated from Athens and Lisbon measurements.
[d] Estimated from residences.
[e] Estimated from PM observations using traffic/background ratio (Athens measurements used for Treviso and Lisbon for Porto) (see text for the references).

was used with WRF-CAMx modelled concentrations to calculate residential exposures in all five cities. In addition, Statistics Finland 2015 population data at 1 km^2 resolution containing three age groups (<15 years, 15–64 years, >64 years) was used to test differences between age group specific exposures (Statistics Finland, 2020a).

Outdoor exposures in school locations were assessed using locations of compulsory schools and number of pupils for Kuopio, Lisbon and Porto (República Portuguesa - Ministério da Educação, 2019; Statistics Finland, 2020b). In Portugal and Finland basic education consists of nine grades, primary education including grades from 1 to 6 and secondary education grades from 7 to 9. In Portugal basic education is started at the age of 6 lasting to the age of 14. While in Finland basic educations starts at the age of 7 last until the age of 15 (EC/EACEA, 2015). For Athens and Treviso school locations were not available. Instead the ratio of school to residential outdoor exposure in Lisbon was used. The ratio was calculated by dividing Lisbon school exposure by residential exposure. Residential exposures of Athens and Treviso were then multiplied by the calculated ratio to estimate school outdoor exposures.

An annual time-activity pattern for school children of Lisbon, Porto, Athens and Treviso was derived from a time-activity survey conducted in Lisbon in 2016–2017 among children aged between 5 and 10 years old. A self-report questionnaire including weekday and weekend, and 17 indoor and outdoor microenvironments (ME) was applied in 26 schools. Parents returned 1189 completed questionnaires, representing a response rate of 20% (Faria et al., 2020). Weekend patterns (196 days), which were used also for vacation days and weekday patterns (169 days) of schooldays were used to calculate a combined time-activity pattern at annual level.

In Kuopio time-activity data of school children and students was based on a survey conducted in Finland during 2009 and 2010 (Statistics Finland, 2020c). It included population over 10 years old (3800 people, 7500 days, 41% response rate) who kept a record of their time-activity on one weekday and one Saturday or Sunday. Activities were classified into 146 categories and were aggregated into 26 categories. Compiled time-activity data of the two cities, Kuopio and Lisbon (Lisbon data applied also in Athens, Porto and Treviso), were further allocated to 6 MEs: (i) residential outdoor, (ii) school outdoor, (iii) residential indoor, (iv) school outdoor (v) other indoor, and (vi) traffic.

Based on indoor and outdoor $PM_{2.5}$ concentrations measured in Lisbon and Athens, a linear regression was applied to estimate infiltration factors for South European residences and schools. The regression slope, which corresponds to the fraction of the outdoor generated particles penetrating indoors, was found equal to 0.82 for schools (95% Confidence interval 0.55–1.09) and 0.66 for the residences (95% CI 0.61–0.70) (Table 1). In Lisbon, measurements were conducted in 2017–2018, during the occupied period, in 5 schools and 40 residences, in the context of the LIFE Index-Air project. In schools concentrations were monitored during the school week from Monday to Friday and in residences during 4 weekdays and one day in the weekend (Faria et al., 2020). In Athens measurements were conducted in 7 schools during the occupied period in two winter periods in 2003 and 2004 for 2–5 consecutive schooldays (Diapouli et al., 2008). For the Athens residences, $PM_{2.5}$ concentration measurements were conducted in two separate studies. In 2002 sampling was performed in 3 residences during the cold and warm period (Diapouli et al., 2011). During the years 2005 and 2006, measurements were conducted in additional nine residences (Diapouli, 2008; Diapouli et al., 2010).

For Kuopio, the infiltration factor in 2015 was estimated linearly, by taking into account the annual renewal and renovation rate of building stock (2%), assuming a better filtration of outdoor particles (Geels et al., 2015; Hänninen et al., 2015). An infiltration factor (0.59) calculated by Hänninen et al. (2004) for residences in Helsinki in year 1997 was used as a starting point. The linear regression resulted in a infiltration factor of 0.55, which was used both for home and school indoor MEs (Table 1).

Traffic enrichment factors (EF) were defined by calculating annual PM concentration ratios measured in traffic and urban background stations (Table 1). In Kuopio, the enrichment factor (1.30) was assessed using ratios of PM_{10} measurements. For Lisbon and Porto, the enrichment factor (1.35) was defined from $PM_{2.5}$ observations measured in Lisbon and for Athens and Treviso (EF = 1.58) from $PM_{2.5}$ measurements conducted in Athens.

2.3. Exposure models

Six exposure models were used to estimate annual outdoor generated $PM_{2.5}$ exposures in each city (Table 2). To quantify how ignoring spatial mobility, time-activity and infiltration affects to exposure estimates, complexity of the models were increased by gradually adding microenvironments. School children spend the majority of their time during the year in five major MEs (residential and school indoor and outdoor and traffic). The sixth microenvironment (other indoor) was formed from multiple MEs including for example shops, theatres and restaurants. Models 1 to 3 included outdoor MEs only. In models 4 to 6, the effect of infiltration was incorporated to models. Exposures in each ME were estimated based on modelled outdoor $PM_{2.5}$ concentrations at home or school locations, taking into account infiltration factors for indoor ME, and enrichment factor for traffic ME.

Calculated $PM_{2.5}$ exposures in each ME were integrated with corresponding time-activity data at annual level to calculate time-activity weighted exposures (Eq. 1).

$$E = \sum_{j=1}^{n} C_j t_j \tag{1}$$

where E is the total exposure for school children, C_j is the population or pupil weighted outdoor concentration (PWC) in microenvironment j and t_i is the fraction of time (%) spent by the school children in each ME.

In model 1, annual population-weighted concentration in residential outdoor ME was calculated using modelled $PM_{2.5}$ concentrations. Population grid centroids were used as residential addresses and number of inhabitants as weights. The nearest modelled concentration of population centroid was used as exposure level in each grid cell. Model 2 incorporated residential and school outdoor exposures. School outdoor exposures were computed using number of pupils as weights in each school location and the nearest modelled concentration as exposure level. For Athens and Treviso, school outdoor exposure was calculated by multiplying residential outdoor exposure with the ratio of school to residential outdoor exposure in Lisbon. Traffic exposure was calculated by multiplying residential outdoor exposure with the traffic enrichment factor determined for each city and was integrated to model 3.

Model 4 included outdoor and indoor exposures in residential areas, and school indoor and outdoor exposures were further added to model 5. Outdoor-originated indoor exposure was calculated by multiplying residential and school outdoor exposures with the infiltration factors defined for each city. School indoor exposure with time spent in other indoor microenvironments was used to calculate other indoor exposure in model 6, which included six MEs: (i) residential outdoor, (ii) school

Table 2
Used modelling approaches to assess fine particulate matter exposures.

Exposure model		Microenvironments	No. of MEs
Outdoor air-based models	M1.	Residential outdoor	1
	M2.	School outdoor + model 1	2
	M3.	Traffic + model 2	3
Models taking account of infiltration	M4.	Residential indoor + model 1	2
	M5.	School indoor and outdoor + model 4	4
	M6.	Other indoor and traffic + model 5	6

outdoor, (iii) residential indoor, (iv) school indoor, (v) other indoor and (vi) traffic.

3. Results

Outdoor-originated annual particulate matter ($PM_{2.5}$) exposures of school children were assessed using six exposure models based on atmospheric chemical transport models, starting from residential based outdoor exposures and adding complexity to the model in each step. Relative bias in comparison to the most complex model was quantified. Furthermore, bias and error in modelled $PM_{2.5}$ concentrations were evaluated.

3.1. Outdoor-originated fine particulate matter exposures

The annual residential outdoor exposures ranged from 4.5 μg/m^3 in Porto to 27.3 μg/m^3 in Treviso representing well the high variability of outdoor air pollution levels within the European region (Table 3). Traditional population weighting of residential outdoor concentrations (model 1), lead on average to 26% higher exposure estimates in comparison to model 6. In models 2 and 3 the effect of spatial variability of outdoor school and traffic exposures modestly increased estimated outdoor exposure levels by 3.1% and 5.0%, respectively, compared to model 1. In comparison to outdoor air-based models, the numerical estimates dropped by up to 32% on average when adding infiltration to the models 4–6. Residential based model (# 4) resulted in 14% lower exposures compared to model 6. Spatial supplements to the models 5–6 increased exposure estimates by 8.7% and 16% in comparison to model 4. Accordingly, the underestimation in comparison to model 6 decreased.

In all cities, models relying on outdoor concentrations (# 1–3) highly overestimated the exposures in comparison to model number 6 results, ranging from 3.5 μg/m^3 in Porto to 21.7 μg/m^3 in Treviso. In Kuopio, the residential outdoor exposure overestimation (+49%) was the highest. In South European cities overestimation were smaller, but still considerable, ranging from +26% to +30%. Adding the school outdoor microenvironment (ME) (model 2) had virtually no effect on exposure level in Kuopio (+0.2%), but it increased exposure estimates in the South by 1.5% to 3.2%. Traffic (model 3) increased the exposure estimates with respect to model 1 by 5.3% in Athens and Treviso, by 4.5% in Lisbon, by 2.8% in Porto, and by 1.4% in Kuopio.

Including infiltration (models 4–6) decreased residential outdoor based exposure estimates by up to 32% in South European cities and by up to 44% in Kuopio. In residential based model (# 4) exposures were between 11% and 16% lower than the model 6 estimates. Adding the school ME (model 5) increased exposure estimates in Athens, Lisbon and Treviso (by 8.8%), in Porto by 6.8% and in Kuopio by 2.2%, and decreased the underestimation of exposures, with respect to the reference value (model 6).

3.2. Contribution of microenvironments on outdoor-originated $PM_{2.5}$ exposures

School children were estimated to spent annually 87% of their time indoors and 10% outdoors in Lisbon and other South European cities (Athens, Porto and Treviso) (Fig. 2). In Finland school children spent more time indoors (91%) and less time outdoors (5%) in comparison to South Europe. Time spent in traffic was the same in all cities (4%).

The time spend in residential indoor ME contributed the most to the school children's $PM_{2.5}$ exposure originated from outdoors (55–59%). School indoor contributions were 5–6% points greater in South Europe (15–16%) and 13% of exposures occurred outdoors. In Kuopio outdoor contribution was smaller (8%). Traffic contributed slightly less in South Europe (6–7%) in comparison to Kuopio (8%). The contribution of other indoor ME was higher in Kuopio (14%) than in the South (8–9%). Even though outdoor and traffic exposure levels were up to over 2 times higher in comparison to outdoor-originated indoor exposures, the amount of time spent indoors makes indoor environments the largest contributors to the estimated exposures.

4. Discussion

We assessed outdoor-originated annual fine particulate matter exposures of school children in five European cities using six exposure models. In comparison to the most complete microenvironmental model (# 6; including residential and school in- and outdoor, other indoor and traffic), the common use of residential outdoor $PM_{2.5}$ concentrations resulted on average in 26% higher estimates. Time spent indoors greatly reduced exposure to outdoor generated particles, while adding school and traffic MEs increased the exposure estimates. Most of the exposure to outdoor-originated $PM_{2.5}$ occurred in indoor MEs.

4.1. Estimation of bias and error in modelled $PM_{2.5}$ concentrations and implications on exposure, health impact assessment and epidemiology

Over all modelling domains there was on average minor positive bias (0.9 μg/m^3), slightly more variation in predicted concentrations (underestimation of variance, UEV −2%) and modest random error (2.0 μg/m^3) (Table 4). In the city domains, Athens had the largest mean bias (2.1 μg/m^3) while in other cities bias was smaller (0.1–0.3 μg/m^3). Contrary to all station average, there was more variation in observed concentrations in individual city domains. The highest UEV was in Kuopio (99%) where measurement stations were situated within 1 km radius of each other and lead to negligible variation in predicted concentrations. Also in Treviso UEV of predictions was high (51%), while in Athens (11%) and Lisbon (16%) the variations of predicted and observed concentrations were closest to each other. Highest random errors between predicted and observed concentrations in measurement station locations was in Kuopio (2.7 μg/m^3) and lowest in Treviso (0.7 μg/m^3). In Athens (1.8 μg/m^3) and Lisbon random errors (2.1 μg/m^3) were modest.

Table 3
Annual outdoor generated $PM_{2.5}$ exposures (μg/m^3) calculated with six models. City-specific and combined weighted exposure across all cities and relative bias (%) to the reference model (#6).

	Athens	Kuopio	Lisbon	Porto	Treviso	Average[a]
	μg/m^3 (%)	μg/m^3 (%)	μg/m^3 (%)	μg/m^3 (%)	μg/m^3 (%)	μg/m^3 (%)
M1. Residential outdoor	19.7 (+26)	6.5 (+49)	12.0 (+27)	4.5 (+30)	27.3 (+26)	15.7 (+26)
M2. School outdoor + M1	20.3 (+30)	6.5 (+49)	12.4 (+31)	4.6 (+32)	28.1 (+30)	16.2 (+30)
M3. Traffic + M2	20.7 (+32)	6.6 (+51)	12.6 (+33)	4.6 (+34)	28.7 (+32)	16.5 (+32)
M4. Residential indoor + M1	13.4 (−14)	3.7 (−16)	8.2 (−14)	3.1 (−11)	18.6 (−14)	10.7 (−14)
M5. School (in + out) + M4	14.6 (−7)	3.7 (−15)	8.9 (−6)	3.3 (−5)	20.2 (−7)	11.6 (−7)
M6. Other in and traffic + M5	15.7	4.4	9. 5	3.5	21.7	12.5

[a] Population size weighted five-city mean.

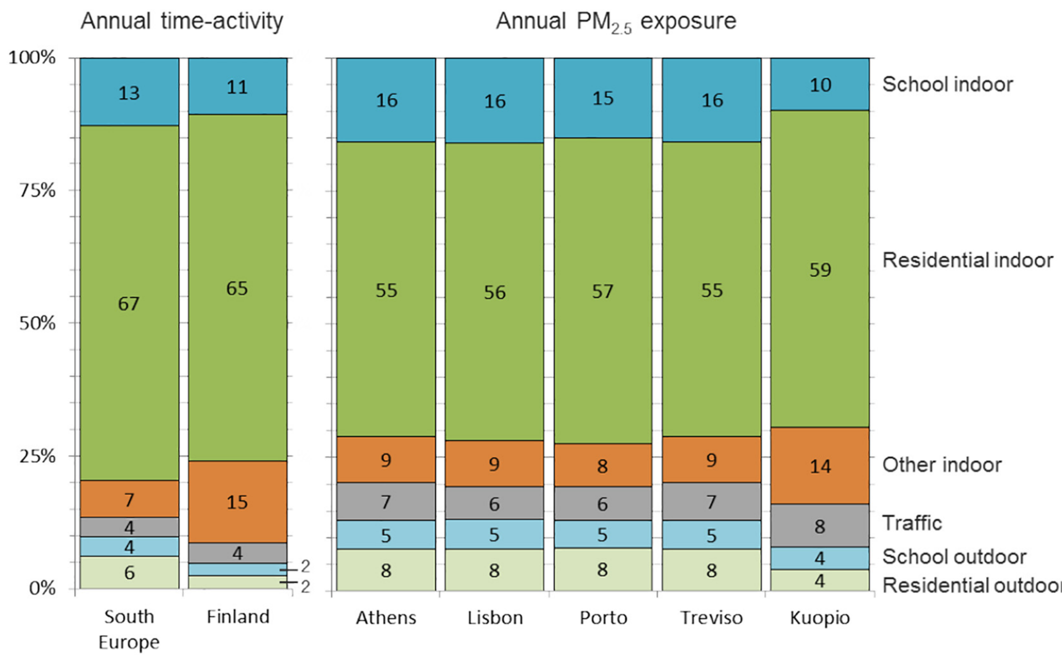


Fig. 2. Time-activity and contribution of each microenvironment to outdoor-originated $PM_{2.5}$ exposures of school children. Exposures are clearly dominated by indoor environments.

The air quality modelling community traditionally uses a rich set of parameters to characterize model performance, including mean and fractional bias, and the corresponding error characteristics (Borrego et al., 2008; Carslaw and Ropkins, 2012; Kukkonen et al., 2018). Interestingly, the two main uses of health related air quality data, health impact assessment and epidemiology, have contrasting behaviour in relationship to bias and random error.

Model bias is important in health impact assessments; any bias in exposures will be directly reflected in similar bias in estimates. In epidemiological analysis, bias in exposure has much less significant role. As the health data is observational, the magnitude of health effects associated with air pollution does not depend on any bias of the latter, even though it may lead to incorrect understanding of excess risk attributable to unit exposures. However, while in health impact assessment random error has no net effect on impact estimates, their detrimental effects in epidemiological analysis are well known (even if less well acknowledged): random error leads to regression attenuation, or bias towards zero and thus underestimation of health responses associated with air pollution (Berkson, 1950). As this regression attenuation effect is proportional to relative standard error, the attenuation becomes more profound at lower exposure levels. The traditional air quality model evaluation parameters are barely used, probably also due to the challenges to propagate them through health effect analyses.

4.2. Uncertainties related to exposure models

People spend most of their time indoors at home which considerably lowers the exposure to outdoor-originated particles, but in the same time makes homes the major ME contributing to overall $PM_{2.5}$ exposure. Besides building envelope, also time of the year and climate affect infiltration of outdoor particles indoors (Canha et al., 2017; Hänninen et al., 2011; Taylor et al., 2014). In summer time and in warmer climate areas, windows are likely kept open more and more time is spent outdoors, leading to higher infiltration and exposures. These are also the reasons for the highest overestimation of exposures in Kuopio. Due to colder climate in North Europe more time is spent inside and the particle infiltration rate indoors is lower in comparison to South. Thus better filtration of outdoor air would most likely have great impact on human exposure. On the other hand, in cities that have high outdoor pollutant levels, primarily mitigating pollution from local sources may be the most efficient solution to reduce exposure to $PM_{2.5}$.

Exposure estimates are sensitive to time-activity patterns, especially if an individual or certain population groups spend significant amount of time outdoors or indoors (e.g. elderly). This effect of lifestyle to the personal exposure was studied in urban-traffic area near city centre of Athens (Assimakopoulos et al., 2018). Personal exposures of pensioner (2nd floor) and student (4th floor) were monitored in 15-day campaign living in the same naturally ventilated building. The exposure was lower for pensioner who spent more than 90% of the time in residential microenvironment in comparison to student who spent more time (around 40%) in other more polluted MEs. On average residential outdoor $PM_{2.5}$ concentrations were higher outdoors (24.2 $\mu g/m^3$) than in pensioner's 2nd floor flat (21.6 $\mu g/m^3$) that was closer to trafficked street compared to student's 4th floor flat (15.8 $\mu g/m^3$). In seven other ME's concentrations ranged from 10.6 $\mu g/m^3$ in car to 126 $\mu g/m^3$ in café where smoking was allowed.

Table 4
Estimate of mean bias, random error and underestimation of variance between observed and predicted (WRF-CAMx) $PM_{2.5}$ concentrations in 17 measurement stations.

		Athens	Kuopio	Lisbon	Porto	Treviso	Total
Monitoring stations	n	6	3	5	1	2	17
Observed mean ± SD	$\mu g/m^3$	16.0 ± 4.1	6.5 ± 2.7	12.3 ± 2.1	5.1	27.5 ± 1.4	13.9 ± 6.9
Predicted mean ± SD	$\mu g/m^3$	18.1 ± 3.7	6.6 ± 0.0	12.6 ± 1.7	5.2	27.7 ± 0.7	14.8 ± 7.1
Mean bias (MB)	$\mu g/m^3$	2.1	0.1	0.3	0.1	0.3	0.9
Random error (RE)	$\mu g/m^3$	1.8	2.7	2.1	[a]	0.7	2.0
Underestimation of variance (UEV)	%	11%	99%	16%	[a]	51%	−2%

[a] Limited data ($n \leq 2$) for estimation.

Time-activity patterns in Kuopio differed from other cities and led to different contributions of MEs to $PM_{2.5}$ exposures. Same time-activity data used for South European cities largely resulted in similar contributions. Nevertheless Lisbon survey results do not necessarily differ drastically from time-activity patterns of school children of those in Greece and Italy. Gatto et al. (2014) monitored time-activity patterns of five children aged between 8 and 11 years in Rome during the spring and summer/autumn campaigns in 2012, each lasting for 15 days. School children spent on average 69% of their time in residences, 18% in schools, 8% outdoors and 5% in other microenvironments including buses, cars and gyms. The results were similar to our annual estimate for time-activity patterns of school children in Lisbon, which was further applied in Porto, Athens and Treviso. Also in Athens time-activity patterns of students were found very similar to the ones of Lisbon. Based on a small-scale (100 elementary school children) survey conducted in the Athens metropolitan area during 2018–2019, children spend on average 83% of their time indoors (66% in residences, 13% in schools and 5% in other indoor ME), 8% in traffic and 8% outdoors (Laskari, 2019).

Our results indicate that when health impacts are analysed in an epidemiological study using outdoor pollution levels, the unit toxicity may be substantially underestimated. This is because the outdoor-originated personal exposure is lower than outdoor concentrations due to major fraction of exposure is happening in indoor ME where the concentrations are lowered by infiltration. According to the most comprehensive model (#6) considered here, the reduction of outdoor-originated exposures and thus underestimation in particulate matter toxicity is on average 21%.

True personal exposures indoors include also particles from indoor sources, which were not considered in the current paper. Chen et al. (2020) in their recent work observed that peak exposures in schools due to indoor sources lead to 10% - 90% higher total exposures for students, in comparison to outdoor levels. They further estimated that 37–89% of the personal exposure could result from indoor sources, which become more dominant when outdoor air gets cleaner. Amato et al. (2014) estimated that about half (47%) of the school children exposures were originated from indoor sources. Unpaved playgrounds in comparison to paved playgrounds, and windows oriented directly to the street rather than to the interior block, were factors that increased exposure to soil particles and road traffic emissions. In residences, the role of indoor sources is usually lower. Azimi and Stephens (2020) tested 4 scenarios to estimate total $PM_{2.5}$ exposures in the United States in 2012. They estimated that residential indoor exposure of outdoor origin was the largest contributor (40 to 60%) to the related mortality ($230-300k$) followed by residential indoor generated sources (20 to 40%). Outdoor concentrations of air pollutants are most often used in epidemiological studies (Evangelopoulos et al., 2020), ignoring exposures to indoor sources in air pollution health impact studies. This also suggests, in addition to possibly underestimated unit toxicity of outdoor-originated particles, that the overall health impacts are consistently underestimated.

The choice of a transport mode (active vs. passive), vehicle window position (closed vs. open) and ventilation settings are among factors that affect the exposure levels which occur during commuting. Active commuters route further away from the roads leads to lower exposure; thus the route choice is an important parameter in the estimation of exposure in traffic. When taking account the higher inhalation rates of cyclists or those who are walking, inhaled doses are commonly higher among active commuters than passive commuters (Correia et al., 2020). Adams et al. (2016) further demonstrated that inhaled dose during walking trip (3.2 μg) for students was greater compared to cycling (2.2 μg). Okokon et al. (2017) estimated that in three European cities (Helsinki, Rotterdam and Athens) highest intakes were found during bicycle rides (24; 28 and 37 μg) in comparison to buses (8; 5 and 21 μg), closed window (3; 5 and 6 μg) and open window car (7; 7 and 13 μg) commuters.

Here we assumed that exposure in traffic happens in outdoors or in other words, all school children were actively commuting near roads where pollution levels are higher than background levels. Although the time school children spent in traffic was low, higher concentrations during commuting increased the exposure estimates up to 5% (between model 1 and 3) and up to 14% (model 4 and 6). Even though significant amount of school children in reality are travelling in vehicle and their exposures and inhaled doses would be lower, the health benefits gained during active commuting (Tainio et al., 2016) likely counterbalance the differences in health impacts, and therefore neglecting the commuting choice may not lead to bias in associated health impacts.

Time spent in traffic increased exposures most in the bigger cities (Athens, Lisbon and Treviso) most likely due to higher traffic volumes. Also the exposures in school locations were higher in comparison to residential areas and were relatively higher in bigger than in smaller cities (Kuopio). This indicates that schools were located in areas where contribution of traffic and other local sources were bigger.

4.3. Spatial and temporal resolution of modelled concentration data and influence of age distributed population data

Use of a higher temporal resolution of the gridded concentration data instead of annual means in exposure models, could have led to bigger differences between residential and school exposure estimates. Population level time-activity impacts $PM_{2.5}$ exposures in some areas, for example during peak traffic hour during morning and afternoon commute and lower activity and corresponding concentrations especially during the night while people are at home (Fig. S 1). Thus using hourly concentrations correlating with the time-microenvironment activity, most likely would have resulted in lower average concentrations in residential areas, and higher in schools.

Spatial resolution of 1 km used in this work was reasonably accurate, because school children spent most time close to home. Fagerholm and Broberg (2011) showed for example that in Turku, Finland, over 50% of the school children's activity happens within a radius of 0.5 km from home. Even though it is shown that moving from coarser to finer resolution have increasing effect on $PM_{2.5}$ exposure and health impact estimates especially in densely populated urban areas, improving model resolution from 1 km to finer resolution does not necessarily always lead to considerable change in results (Fenech et al., 2018; Jiang and Yoo, 2018; Korhonen et al., 2019; Lehtomäki et al., 2020). The choice of the modelling system (e.g. chemical transport model or satellite-based estimates), differences in emission data and in methodology when calculating health burdens may have greater impact to exposure and health impact estimates (Ford and Heald, 2016). Also location of the residence and school plays an important role, since in urban areas $PM_{2.5}$ concentrations originating from local sources are higher compared to rural areas, thus resulting higher overall exposures (Fig. S 2). Combining both spatially and temporally high resolution concentration data with corresponding time-activity and spatial mobility data, could potentially greatly improve exposure assessment accuracy of the individuals.

In residential areas school children's exposures were assessed using total population of Eurostat population grid (1 km^2). To test whether the exposure estimates were sensitive to age group specific population-weighting, we calculated exposures for three age groups (under 15, 15–64 and over 64 years old) in Kuopio with Statistics Finland gridded population data, and found that there were no differences in exposures. Therefore, we assume that use of total population to calculate school children's residential outdoor exposures did not result to any great bias in this work.

5. Conclusions

We created six models to alternative quantification of school children's annual exposures to outdoor-originated atmospheric aerosols defined as fine particles, adding complexity of the model in each step. Using our best exposure estimate (model 6) as a reference point, relative bias of each model was quantified. The most important factor affecting school children's exposure was infiltration of outdoor particles to indoors.

The most common use of residential outdoor concentration as population exposure descriptor led to 26% higher exposure estimates on average in comparison to the most complex model (12.5 μg/m^3), which incorporated outdoor and indoor exposures of outdoor sources in residential areas and schools, other indoor and traffic microenvironments. Adding spatial variability for schools and traffic increased exposures relying only on residential outdoor concentrations (model 1) on average by 3.1% and 5.0%, and when infiltration of outdoor particles indoors was included (model 4) by 8.7% and 16%, reflecting higher exposures in school and traffic microenvironments. Although outdoor exposure levels of PM were up to 1.8 times and traffic exposures up to over 2 times higher in comparison to indoor microenvironments here we estimated that 80–84% of the outdoor-originated $PM_{2.5}$ exposure occurred indoors.

Relying only on outdoor concentrations in assessment of outdoor generated $PM_{2.5}$ exposures leads to considerable overestimations. This underlines the importance of incorporating time-activity, spatial mobility and infiltration of outdoor particles indoors as part of the exposure assessment approach. In epidemiological assessments, overestimation of exposure leads to underestimation of toxicity of outdoor-originated exposures, which occur to a substantial extent indoors at lower levels as demonstrated here. Ignoring infiltration variability also adds to random exposure misclassification with similar implications for outdoor epidemiological studies.

CRediT authorship contribution statement

AK developed and calculated the models, participated in compiling parameters used in exposure models, wrote most of the manuscript with contribution from co-authors. **HR**, **JF**, **DL** and **SR**, coordinated by **AIM**, performed the meteorological and the air quality simulations for all target cities. **SMA** coordinated the Life Index-Air project. **TF**, **VM** and **NC**, coordinated by **SMA**, conducted time-activity study and school campaign measurements used for estimation of infiltration in Lisbon and participated in compiling input data for the models. **LD** and **KE** participated in compiling the input data used in the exposure models, in particular for Athens. **ML** and **EC** reviewed and edited the manuscript. **HL** and **IR** reviewed and edited the manuscript and participated in determining uncertainties for health impact assessments and epidemiology. **OH** conceptualized the work, supervised and coordinated the article drafting process, participated in creation of the models and defining input parameters.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was supported by EU LIFE Index-Air project (LIFE15 ENV/PT/000674), Academy of Finland project APPEAL (322150) and intramural funding by the Finnish Institute for Health and Welfare. For Finland the LIFE Index-Air modelling data was compared with similar data from Nordic WelfAir (NordForsk project #75007) and BATMAN (Academy of Finland, 285672). This work reflects only the authors' views. The authors also thank the FCT/ MCTES financial support to CESAM (UIDP/50017/2020 + UIDB/50017/2020), C2TN (UIDB/04349/2020 + UIDP/04349/2020) and ExpoLIS (LISBOA-01-0145-FEDER-032088).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.147111.

References

Adams, M.D., Yiannakoulias, N., Kanaroglou, P.S., 2016. Air pollution exposure: an activity pattern approach for active transportation. Atmos. Environ. 140, 52–59. https://doi.org/10.1016/j.atmosenv.2016.05.055.

Amato, F., Rivas, I., Viana, M., Moreno, T., Bouso, L., Reche, C., Àlvarez-Pedrerol, M., Alastuey, A., Sunyer, J., Querol, X., 2014. Sources of indoor and outdoor PM2.5 concentrations in primary schools. Sci. Total Environ. 490, 757–765. https://doi.org/10.1016/j.scitotenv.2014.05.051.

APA, 2020. "Base de dados on-line sobre qualidade do ar". Agência Portuguesa do Ambiente-QualAr [WWW Document]. URL http://qualar.apambiente.pt/. (Accessed 8 April 2020).

Assimakopoulos, V.D., Bekiari, T., Pateraki, S., Maggos, T., Stamatis, P., Nicolopoulou, P., Assimakopoulos, M.N., 2018. Assessing personal exposure to PM using data from an integrated indoor-outdoor experiment in Athens-Greece. Sci. Total Environ. 636, 1303–1320. https://doi.org/10.1016/j.scitotenv.2018.04.249.

Azimi, P., Stephens, B., 2020. A framework for estimating the US mortality burden of fine particulate matter exposure attributable to indoor and outdoor microenvironments. J. Expo. Sci. Environ. Epidemiol. 30, 271–284. https://doi.org/10.1038/s41370-018-0103-4.

Beelen, R., Raaschou-Nielsen, O., Stafoggia, M., Andersen, Z.J., Weinmayr, G., Hoffmann, B., Wolf, K., Samoli, E., Fischer, P., Nieuwenhuijsen, M., Vineis, P., Xun, W.W., Katsouyanni, K., Dimakopoulou, K., Oudin, A., Forsberg, B., Modig, L., Havulinna, A.S., Lanki, T., Turunen, A., Oftedal, B., Nystad, W., Nafstad, P., De Faire, U., Pedersen, N.L., Ostenson, C.-G., Fratiglioni, L., Penell, J., Korek, M., Pershagen, G., Eriksen, K.T., Overvad, K., Ellermann, T., Eeftens, M., Peeters, P.H., Meliefste, K., Wang, M., Bueno-de-Mesquita, B., Sugiri, D., Kramer, U., Heinrich, J., de Hoogh, K., Key, T., Peters, A., Hampel, R., Concin, H., Nagel, G., Ineichen, A., Schaffner, E., Probst-Hensch, N., Kunzli, N., Schindler, C., Schikowski, T., Adam, M., Phuleria, H., Vilier, A., Clavel-Chapelon, F., Declercq, C., Grioni, S., Krogh, V., Tsai, M.-Y., Ricceri, F., Sacerdote, C., Galassi, C., Migliore, E., Ranzi, A., Cesaroni, G., Badaloni, C., Forastiere, F., Tamayo, I., Amiano, P., Dorronsoro, M., Katsoulis, M., Trichopoulou, A., Brunekreef, B., Hoek, G., 2014. Effects of long-term exposure to air pollution on natural-cause mortality: an analysis of 22 European cohorts within the multicentre ESCAPE project. Lancet (London, England) 383, 785–795. https://doi.org/10.1016/S0140-6736(13)62158-3.

Berkson, J., 1950. Are there two regressions? J. Am. Stat. Assoc. 45, 164–180. https://doi.org/10.1080/01621459.1950.10483349.

Borrego, C., Monteiro, A., Ferreira, J., Miranda, A.I., Costa, A.M., Carvalho, A.C., Lopes, M., 2008. Procedures for estimation of modelling uncertainty in air quality assessment. Environ. Int. 34, 613–620. https://doi.org/10.1016/j.envint.2007.12.005.

Canha, N., Lage, J., Candeias, S., Alves, C., Almeida, S.M., 2017. Indoor air quality during sleep under different ventilation patterns. Atmos. Pollut. Res. 8, 1132–1142. https://doi.org/10.1016/j.apr.2017.05.004.

Carslaw, D.C., Ropkins, K., 2012. Openair — an R package for air quality data analysis. Environ. Model. Softw. 27–28, 52–61. https://doi.org/10.1016/j.envsoft.2011.09.008.

Chen, L.-W.A., Olawepo, J.O., Bonanno, F., Gebreselassie, A., Zhang, M., 2020. Schoolchildren's exposure to PM2.5: a student club–based air quality monitoring campaign using low-cost sensors. Air Qual. Atmos. Health 13, 543–551. https://doi.org/10.1007/s11869-020-00815-9.

Correia, C., Martins, V., Cunha-Lopes, I., Faria, T., Diapouli, E., Eleftheriadis, K., Almeida, S.M., 2020. Particle exposure and inhaled dose while commuting in Lisbon. Environ. Pollut. 257, 113547. https://doi.org/10.1016/j.envpol.2019.113547.

Cunha-Lopes, I., Martins, V., Faria, T., Correia, C., Almeida, S.M., 2019. Children's exposure to sized-fractioned particulate matter and black carbon in an urban environment. Build. Environ. 155, 187–194. https://doi.org/10.1016/j.buildenv.2019.03.045.

de Hoogh, K., Korek, M., Vienneau, D., Keuken, M., Kukkonen, J., Nieuwenhuijsen, M.J., Badaloni, C., Beelen, R., Bolignano, A., Cesaroni, G., Pradas, M.C., Cyrys, J., Douros, J., Eeftens, M., Forastiere, F., Forsberg, B., Fuks, K., Gehring, U., Gryparis, A., Gulliver, J., Hansell, A.L., Hoffmann, B., Johansson, C., Jonkers, S., Kangas, L., Katsouyanni, K., Künzli, N., Lanki, T., Memmesheimer, M., Moussiopoulos, N., Modig, L., Pershagen, G., Probst-Hensch, N., Schindler, C., Schikowski, T., Sugiri, D., Teixidó, O., Tsai, M.-Y., Yli-Tuomi, T., Brunekreef, B., Hoek, G., Bellander, T., 2014. Comparing land use regression and dispersion modelling to assess residential exposure to ambient air pollution for epidemiological studies. Environ. Int. 73, 382–392. https://doi.org/10.1016/j.envint.2014.08.011.

de Nazelle, A., Bode, O., Orjuela, J.P., 2017. Comparison of air pollution exposures in active vs. passive travel modes in European cities: a quantitative review. Environ. Int. 99, 151–160. https://doi.org/10.1016/j.envint.2016.12.023.

Dee, D.P., Uppala, S.M., Simmons, A.J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M.A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A.C.M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A.J., Haimberger, L., Healy, S.B., Hersbach, H., Hólm, E.V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A.P., Monge-Sanz, B.M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N., Vitart, F., 2011. The ERA-interim reanalysis: configuration and performance of the data assimilation system. Q. J. R. Meteorol. Soc. 137, 553–597. https://doi.org/10.1002/qj.828.

Diapouli, E., 2008. Exposure of Population Subgroups to Airborne Particles - Contribution of Indoor and Outdoor Sources. National Technical University of Athens.

Diapouli, E., Chaloulakou, A., Mihalopoulos, N., Spyrellis, N., 2008. Indoor and outdoor PM mass and number concentrations at schools in the Athens area. Environ. Monit. Assess. 136, 13–20. https://doi.org/10.1007/s10661-007-9724-0.
Diapouli, E., Chaloulakou, A., Spyrellis, N., Campus, Z., 2010. Indoor/outdoor PM levels and EC surrogate, at typical microenvironments in the Athens area. Glob. NEST 12, 12–19. https://doi.org/10.30955/gnj.000693.
Diapouli, E., Eleftheriadis, K., Karanasiou, A., Vratolis, S., Hermansen, O., Colbeck, I., Lazaridis, M., 2011. Indoor and outdoor particle number and mass concentrations in Athens. Sources, sinks and variability of aerosol parameters. Aerosol Air Qual. Res. 11, 632–642.
Dockery, D.W., Pope, C.A. 3rd, Xu, X., Spengler, J.D., Ware, J.H., Fay, M.E., Ferris, B.G.J., Speizer, F.E., 1993. An association between air pollution and mortality in six U.S. cities. N. Engl. J. Med. 329, 1753–1759. doi:https://doi.org/10.1056/NEJM199312093292401.
EC/EACEA, 2015. The Structure of the European Education Systems 2015/2016: Schematic Diagrams. Eurydice Facts and Figures. European Commission/EACEA/Eurydice https://doi.org/10.2797/63725 2015.
EEA, 2020a. Healthy Environment, Healthy Lives: How the Environment Influences Health and Well-being in Europe. European Environment Agency https://doi.org/10.2800/53670.
EEA, 2020b. Air Quality Statistics Viewer. European Environment Agency [WWW Document]. URL. https://www.eea.europa.eu/data-and-maps/dashboards/air-quality-statistics (Accessed 4.8.2020).
Emmons, L.K., Walters, S., Hess, P.G., Lamarque, J.-F., Pfister, G.G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S.L., Kloster, S., 2010. Description and evaluation of the model for ozone and related chemical tracers, version 4 (MOZART-4). Geosci. Model Dev. 3, 43–67. https://doi.org/10.5194/gmd-3-43-2010.
ENVIRON, 2016. User's Guide Comprehensive Air Quality Model with Extensions Version 6.30 (Novato, California).
Eurostat, 2016. GEOSTAT 1km2 population grid 2011 [WWW Document]. URL https://ec.europa.eu/eurostat/web/gisco/geodata/reference-data/population-distribution-demography/geostat. (Accessed 15 April 2020).
Evangelopoulos, D., Katsouyanni, K., Keogh, R.H., Samoli, E., Schwartz, J., Barratt, B., Zhang, H., Walton, H., 2020. PM2.5 and NO2 exposure errors using proxy measures, including derived personal exposure from outdoor sources: a systematic review and meta-analysis. Environ. Int. 137, 105500. https://doi.org/10.1016/j.envint.2020.105500.
Fagerholm, N.C., Broberg, A., 2011. Mapping and characterising children's daily mobility in urban residential areas in Turku, Finland. Fenn. - Int. J. Geogr. 189, 31–46.
Faria, T., Martins, V., Correia, C., Canha, N., Diapouli, E., Manousakas, M., Eleftheriadis, K., Almeida, S.M., 2020. Children's exposure and dose assessment to particulate matter in Lisbon. Build. Environ. 171, 106666. https://doi.org/10.1016/j.buildenv.2020.106666.
Fenech, S., Doherty, R.M., Heaviside, C., Vardoulakis, S., Macintyre, H.L., O'Connor, F.M., 2018. The influence of model spatial resolution on simulated ozone and fine particulate matter for Europe: implications for health impact assessments. Atmos. Chem. Phys. 18, 5765–5784. https://doi.org/10.5194/acp-18-5765-2018.
Ferreira, J., Lopes, D., Rafael, S., Relvas, H., Almeida, S.M., Miranda, A.I., 2020. Modelling air quality levels of regulated metals: limitations and challenges. Environ. Sci. Pollut. Res. 27 (27), 33916–33928. https://doi.org/10.1007/s11356-020-09645-9 32557059.
FMI, 2020. Air Quality. Finnish Meteorological Institute [WWW Document]. URL. https://en.ilmatieteenlaitos.fi/air-quality (Accessed 4.8.2020).
Ford, B., Heald, C.L., 2016. Exploring the uncertainty associated with satellite-based estimates of premature mortality due to exposure to fine particulate matter. Atmos. Chem. Phys. 16, 3499–3523. https://doi.org/10.5194/acp-16-3499-2016.
Gatto, M.P., Gariazzo, C., Gordiani, A., L'Episcopo, N., Gherardi, M., 2014. Children and elders exposure assessment to particle-bound polycyclic aromatic hydrocarbons (PAHs) in the city of Rome, Italy. Environ. Sci. Pollut. Res. 21, 13152–13159. https://doi.org/10.1007/s11356-013-2442-y.
Geels, C., Andersson, C., Hänninen, O., Lansø, A.S., Schwarze, P.E., Skjøth, C.A., Brandt, J., 2015. Future premature mortality due to O3, secondary inorganic aerosols and primary PM in Europe–sensitivity to changes in climate, anthropogenic emissions, population and building stock. Int. J. Environ. Res. Public Health 12, 2837–2869. https://doi.org/10.3390/ijerph120302837.
Hänninen, O., Lebret, E., Ilacqua, V., Katsouyanni, K., Künzli, N., Sram, R., Jantunen, M., 2004. Infiltration of ambient PM2.5 and levels of indoor generated non-ETS PM2.5 in residences of four European cities. Atmos. Environ. 38, 6411–6423. https://doi.org/10.1016/j.atmosenv.2004.07.015.
Hänninen, O., Hoek, G., Mallone, S., Chellini, E., Katsouyanni, K., Gariazzo, C., Cattani, G., Marconi, A., Molnár, P., Bellander, T., Jantunen, M., 2011. Seasonal patterns of outdoor PM infiltration into indoor environments: review and meta-analysis of available studies from different climatological zones in Europe. Air Qual. Atmos. Health 4, 221–233. https://doi.org/10.1007/s11869-010-0076-5.
Hänninen, O., Asikainen, A., Niittynen, M., Geels, C., Brandt, J., 2015. Century-wide energy efficiency road map for buildings and impact on particle size specific PM2.5 infiltration. In: Leskinen, A., Kontkanen, J. (Eds.), Proceedings of the NOSA-FAAR Symposium 2015 - REPORT SERIES IN AEROSOL SCIENCE, N:o 165 (2015). Finnish Association for Aerosol Research FAAR, Helsinki, p. 36.
Hussein, T., Paasonen, P., Kulmala, M., 2012. Activity pattern of a selected group of school occupants and their family members in Helsinki - Finland. Sci. Total Environ. https://doi.org/10.1016/j.scitotenv.2012.03.002.
Im, U., Brandt, J., Geels, C., Hansen, K.M., Christensen, J.H., Andersen, M.S., Solazzo, E., Kioutsioukis, I., Alyuz, U., Balzarini, A., Baro, R., Bellasio, R., Bianconi, R., Bieser, J., Colette, A., Curci, G., Farrow, A., Flemming, J., Fraser, A., Jimenez-Guerrero, P., Kitwiroon, N., Liang, C.-K., Nopmongcol, U., Pirovano, G., Pozzoli, L., Prank, M., Rose, R., Sokhi, R., Tuccella, P., Unal, A., Vivanco, M.G., West, J., Yarwood, G., Hogrefe, C., Galmarini, S., 2018. Assessment and economic valuation of air pollution impacts on human health over Europe and the United States as calculated by a multi-model ensemble in the framework of AQMEII3. Atmos. Chem. Phys. 18, 5967–5989. https://doi.org/10.5194/acp-18-5967-2018.
Jiang, X., Yoo, E., 2018. The importance of spatial resolutions of community multiscale air quality (CMAQ) models on health impact assessment. Sci. Total Environ. 627, 1528–1543. https://doi.org/10.1016/j.scitotenv.2018.01.228.
Kalimeri, K.K., Bartzis, J.G., Sakellaris, I.A., de Oliveira Fernandes, E., 2019. Investigation of the PM2.5, NO2 and O3 I/O ratios for office and school microenvironments. Environ. Res. 179, 108791. https://doi.org/10.1016/j.envres.2019.108791.
Kazakos, V., Luo, Z., Ewart, I., 2020. Quantifying the health burden misclassification from the use of different PM2.5 exposure tier models: a case study of London. Int. J. Environ. Res. Public Health https://doi.org/10.3390/ijerph17031099.
Korhonen, A., Lehtomäki, H., Rumrich, I., Karvosenoja, N., Paunu, V.-V., Kupiainen, K., Sofiev, M., Palamarchuk, Y., Kukkonen, J., Kangas, L., Karppinen, A., Hänninen, O., 2019. Influence of spatial resolution on population PM2.5 exposure and health impacts. Air Qual. Atmos. Health 12, 705–718. https://doi.org/10.1007/s11869-019-00690-z.
Kukkonen, J., Kangas, L., Kauhaniemi, M., Sofiev, M., Aarnio, M., Jaakkola, J.J.K., Kousa, A., Karppinen, A., 2018. Modelling of the urban concentrations of PM2.5 on a high resolution for a period of 35 years, for the assessment of lifetime exposure and health effects. Atmos. Chem. Phys. 18, 8041–8064. https://doi.org/10.5194/acp-18-8041-2018.
Laskari, K., 2019. Assessment of Children Exposure to Air Pollution through the Use of Time-Activity Data. Hellenic Open University https://apothesis.eap.gr/handle/repo/42576.
Lehtomäki, H., Geels, C., Brandt, J., Rao, S., Yaramenka, K., Åström, S., Andersen, M.S., Frohn, L.M., Im, U., Hänninen, O., 2020. Deaths attributable to air pollution in Nordic countries: disparities in the estimates. Atmosphere (Basel). 11, 467. https://doi.org/10.3390/atmos11050467.
MacNeill, M., Wallace, L., Kearney, J., Allen, R.W., Van Ryswyk, K., Judek, S., Xu, X., Wheeler, A., 2012. Factors influencing variability in the infiltration of PM2.5 mass and its components. Atmos. Environ. 61, 518–532. https://doi.org/10.1016/j.atmosenv.2012.07.005.
Morawska, L., Ayoko, G.A., Bae, G.N., Buonanno, G., Chao, C.Y.H., Clifford, S., Fu, S.C., Hänninen, O., He, C., Isaxon, C., Mazaheri, M., Salthammer, T., Waring, M.S., Wierzbicka, A., 2017. Airborne particles in indoor environment of homes, schools, offices and aged care facilities: the main routes of exposure. Environ. Int. 108, 75–83. https://doi.org/10.1016/j.envint.2017.07.025.
Okokon, E.O., Yli-Tuomi, T., Turunen, A.W., Taimisto, P., Pennanen, A., Vouitsis, I., Samaras, Z., Voogt, M., Keuken, M., Lanki, T., 2017. Particulates and noise exposure during bicycle, bus and car commuting: a study in three European cities. Environ. Res. 154, 181–189. https://doi.org/10.1016/j.envres.2016.12.012.
República Portuguesa - Ministério da Educação, 2019. Estatísticas do Ensino Básico e Secundário [WWW Document]. Portal InfoEscolas URL. http://infoescolas.pt/ (Accessed 12.4.2019).
Singh, V., Sokhi, R.S., Kukkonen, J., 2020. An approach to predict population exposure to ambient air PM2.5 concentrations and its dependence on population activity for the megacity London. Environ. Pollut. 257, 113623. https://doi.org/10.1016/j.envpol.2019.113623.
Siponen, T., Yli-Tuomi, T., Tiittanen, P., Taimisto, P., Pekkanen, J., Salonen, R.O., Lanki, T., 2019. Wood stove use and other determinants of personal and indoor exposures to particulate air pollution and ozone among elderly persons in a northern suburb. Indoor Air 29, 413–422. https://doi.org/10.1111/ina.12538.
Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Wang, W., Powers, J.G., 2008. A description of the advanced research WRF version 3. NCAR Technical Note -475+STR.
Smith, J.D., Barratt, B.M., Fuller, G.W., Kelly, F.J., Loxham, M., Nicolosi, E., Priestman, M., Tremper, A.H., Green, D.C., 2020. PM2.5 on the London underground. Environ. Int. 134, 105188. https://doi.org/10.1016/j.envint.2019.105188.
Statistics Finland, 2020a. Population grid data 1 km x 1 km. [WWW Document]. URL. http://www.stat.fi/org/avoindata/paikkatietoaineistot/vaestoruutuaineisto_1km_en.html (Accessed 4.15.2020).
Statistics Finland, 2020b. Providers of education and educational institutions. [WWW Document]. URL. http://www.stat.fi/til/kjarj/index_en.html (Accessed 4.15.2020).
Statistics Finland, 2020c. Time use. [WWW Document]. URL. http://www.stat.fi/til/akay/akay_2014-12-01_men_001_en.html (Accessed 4.8.2020).
Tainio, M., de Nazelle, A.J., Götschi, T., Kahlmeier, S., Rojas-Rueda, D., Nieuwenhuijsen, M.J., de Sá, T.H., Kelly, P., Woodcock, J., 2016. Can air pollution negate the health benefits of cycling and walking? Prev. Med. (Baltim). 87, 233–236. https://doi.org/10.1016/j.ypmed.2016.02.002.
Taylor, J., Shrubsole, C., Davies, M., Biddulph, P., Das, P., Hamilton, I., Vardoulakis, S., Mavrogianni, A., Jones, B., Oikonomou, E., 2014. The modifying effect of the building envelope on population exposure to PM2.5 from outdoor sources. Indoor Air 24, 639–651. https://doi.org/10.1111/ina.12116.
WHO, 2016. Ambient Air Pollution: A Global Assessment of Exposure and Burden of Disease. World Health Organization.
YPEKA, 2020. Air quality Department of the Greek ministry of environment and energy [WWW Document]. URL http://www.ypeka.gr/. (Accessed 8 April 2020).