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Ultrafine particles and $PM_{2.5}$ in the air of cities around the world: Are they representative of each other?

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ABSTRACT

Can mitigating only particle mass, as the existing air quality measures do, ultimately lead to reduction in ultrafine particles (UFP)? The aim of this study was to provide a broader urban perspective on the relationship between UFP, measured in terms of particle number concentration (PNC) and $PM_{2.5}$ (mass concentration of particles with aerodynamic diameter $< 2.5\,\mu m$) and factors that influence their concentrations. Hourly average PNC and $PM_{2.5}$ were acquired from 10 cities located in North America, Europe, Asia, and Australia over a 12-month period. A pairwise comparison of the mean difference and the Kolmogorov-Smirnov test with the application of bootstrapping were performed for each city. Diurnal and seasonal trends were obtained using a generalized additive model (GAM). The particle number to mass concentration ratios and the Pearson's correlation coefficient were calculated to elucidate the nature of the relationship between these two metrics.

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Results show that the annual mean concentrations ranged from 8.0×10^3 to 19.5×10^3 particles·cm^{-3} and from 7.0 to 65.8 μg·m^{-3} for PNC and $PM_{2.5}$, respectively, with the data distributions generally skewed to the right, and with a wider spread for PNC. PNC showed a more distinct diurnal trend compared with $PM_{2.5}$, attributed to the high contributions of UFP from vehicular emissions to PNC. The variation in both PNC and $PM_{2.5}$ due to seasonality is linked to the cities' geographical location and features. Clustering the cities based on annual median concentrations of both PNC and $PM_{2.5}$ demonstrated that a high PNC level does not lead to a high $PM_{2.5}$, and vice versa. The particle number-to-mass ratio (in units of 10^9 particles·μg^{-1}) ranged from 0.14 to 2.2, > 1 for roadside sites and < 1 for urban background sites with lower values for more polluted cities. The Pearson's *r* ranged from 0.09 to 0.64 for the log-transformed data, indicating generally poor linear correlation between PNC and $PM_{2.5}$. Therefore, PNC and $PM_{2.5}$ measurements are not representative of each other; and regulating $PM_{2.5}$ does little to reduce PNC. This highlights the need to establish regulatory approaches and control measures to address the impacts of elevated UFP concentrations, especially in urban areas, considering their potential health risks.

1. Introduction

The Global Burden of Disease (GBD) studies have demonstrated that exposure to ambient particulate matter (PM) measured as $PM_{2.5}$ (mass concentration of particles with an aerodynamic diameter < 2.5 μm) was the sixth-leading risk factor for premature mortality globally in 2016 Gakidou et al. (2017). $PM_{2.5}$, as well as another mass based metric of ambient particulate matter, PM_{10} (mass concentration of particles with aerodynamic diameter < 10 μm) are regulated by national standards in many countries around the world, with the basis mostly on health guidelines issued by the World Health Organization (WHO, 2006, n.d.). There is a relatively large body of data available globally on their concentrations and characteristics, as well as on epidemiologic evidence of their impacts.

$PM_{2.5}$ and PM_{10} are not, however, the only metrics of ambient particulate matter, and being mass based, account mainly for larger particles with larger mass (Amato et al., 2009; Johansson et al., 2007; Thorpe and Harrison, 2008). Particle number concentration (PNC) is a common metric to measure smaller particles, with little mass. The majority of particles contributing to PNC in typical atmospheric systems are below 0.1 μm (Morawska et al., 1999), and referred to as ultrafine particles (UFP, size < 0.1 μm). UFP originate predominantly from combustion processes and atmospheric new particle formation (de Foy and Schauer, 2015; Kulmala et al., 2004; Seigneur, 2009; Stanier et al., 2004) and are of particular significance in urban air due to traffic being their major source in all cities around the world (Hofman et al., 2016; Kumar et al., 2014; Morawska et al., 1999; Rönkkö et al., 2017). There are much less data available on UFP, or PNC compared with particle mass, with their epidemiology not well established (Tobías et al., 2018), and therefore this particle metric is not used for regulatory purpose.

An unspoken assumption is that, when regulating for $PM_{2.5}$, UFP are also regulated for. Since, however, $PM_{2.5}$ and UFP largely originate from different sources, this is not necessarily true and in fact, a number of studies have shown that the association, if any, is very weak with a few studies being inconclusive (Eeftens et al., 2015; Gomišček et al., 2004; Halek et al., 2010; F. Wang et al., 2010). A number of studies investigating the spatial and temporal variability of PNC and $PM_{2.5}$ have been published; some of these studies did not use actual measurements but only PM concentration estimates from remote sensing and models, while other studies used only short-term data (i.e. less than a month). Most of these studies agree on the seasonality of PNC and $PM_{2.5}$ (i.e. differentiating high and low insolation regions), but differ on many other aspects, such as the presence or absence of correlation between PNC and $PM_{2.5}$. Adding to the complexity is that the spatial distribution of both metrics differs across urban areas (Aalto et al., 2005; Burton et al., 1996; Buzorius et al., 1999; Suh et al., 1997; Tuch et al., 1997). $PM_{2.5}$ is relatively more homogenous within cities, especially in the absence of local sources, with the predominant impact of regional and long-range transport. PNC, on the other hand, are much less homogenous and influenced by the within-city sources mainly traffic. In summary, based on the body of studies available, there is no clear answer to the questions of whether mitigating only particle mass, as the existing air quality measures do, ultimately leads to a reduction in UFP.

There is a mixed epidemiological evidence on the extent to which health impacts associated with the two metrics are similar. A study in Central and Eastern Europe found that an increasing $PM_{2.5}$ directly affects cardiovascular and respiratory hospital admissions, but UFP exposure has delayed and prolonged effects on respiratory hospital admissions (Lanzinger et al., 2016). Viehmann et al. (2015), focusing on systemic blood markers of inflammation and coagulation, reported that $PM_{2.5}$ has a more long-term association with the markers therefore is more likely to contribute to chronic effects of air pollution on cardiovascular disease; while PNC is more inconsistent due to high spatial and temporal variability. On the contrary, Pilz et al. (2018) and Lane et al. (2016) had found an association with PNC and the hs-CRP marker in particular. Further, Strak et al. (2012), in a study performed in the Netherlands, found that changes in PNC are more predictive of acute responses for airway inflammation and impaired lung function, unlike $PM_{2.5}$. The studies by Chung et al. (2015) and Franck et al. (2011), conducted in Boston, MA, USA, and Leipzig, Germany, respectively, reported that cardiovascular risk is associated with elevated PNC ($> 10^4$ particle·cm^{-3}) and not with $PM_{2.5}$. Similarly, Atkinson et al. (2010) found that PNC in London was associated with cardiovascular deaths and admission but $PM_{2.5}$ was more linked to respiratory health outcomes.

Hence, given that the spatial and temporal patterns of PNC and $PM_{2.5}$ differ, and that the sources and processes contributing to each metric differ, it is expected that controlling one will not do much for controlling the other. The different, and probably independent impacts of PNC and $PM_{2.5}$ on human health, is a further support on the need for a good quantitative understanding of both these metrics in urban air. Considering the importance of the relationship between PNC and $PM_{2.5}$ and the lack of complete scientific understanding, this study aimed to provide a broader urban perspective on this matter by addressing the following questions: (1) are there similarities or associations between these parameters in terms of ambient concentrations, and diurnal or seasonal variability in cities around the world?; and (2) would regulating $PM_{2.5}$ assist in controlling PNC?

2. Methodology

2.1. Study areas and sites

Data from a one-year period from sites distributed around the world were required. Identification of cities and acquisition of data was done through convenience sampling by connecting with colleagues on our collaborative network based on our knowledge of data availability. The inclusion criteria applied were: (1) a minimum of 6 months of data covering at least 50% per month in an hourly resolution for both PNC

and $PM_{2.5}$ (i.e. if data acquired covered > 6 months, all available measurements were used); (2) both parameters measured concurrently, in the same location or in close proximity; and (3) data collected within the past ten years from the time of the request. Ten cities, located in four different continents, fulfilled the aforementioned criteria; they are presented on a map in Fig. 1 and listed in Table 1, including information on local climate and pollutant sources in each location. The identified PM sources were those impacting the sites under study, but these sources can also be considered as impacting the cities where the sites are located, because the monitoring sites are representative of the city conditions. For two of the cities, Brisbane and London, $PM_{2.5}$ data from multiple sites were available and used in the analysis.

2.2. Data collection

Data acquired from the cities were from measurements collected between 2012 and early 2016. During this period, the climate characteristics have slightly varied based on the Global Climate Assessment of the National Oceanic and Atmospheric Administration (NOAA, 2013, 2014, 2015, 2016). In 2012, a weak La Niña occurred turning to neutral in 2013 and 2014. Then a weak El Niño started in 2014 and intensified towards 2015. Although these years were identified as among the warmest, the microclimate in each city included in this study were characterised as neutral with no extreme events.

Detailed characteristics of the sites and the instruments used are provided in Table 2 and Section 1 of the supplementary material. Sampling sites were categorised as either urban background (UB) or roadside (RS). According to the European Union Directive on Ambient Air Quality and Cleaner Air in Europe (2008), stations to provide data for urban background should be located such that pollution level is "influenced by the integrated contribution from all sources upwind of the station" and should be representative of air quality within several square kilometres of the urban area. At traffic sites, on the other hand, the sampled air must represent no < 100 m length of a street segment. In the United Kingdom, the Automatic Urban and Rural Network refers to sites as "Roadside" if they are located 2–10 m from the kerb, or "Kerbside" if within 1 m of the kerb. Similarly, for near-road monitoring stations of PM by the United States Environmental Protection Agency (USEPA), siting must be at a distance not > 15 m from roadways to be monitored, based on 40 CFR Part 58 Appendix E – 6.3 (USEPA, 2006).

2.3. Data analysis

Hourly averages of PNC and $PM_{2.5}$ in local time (LT) were used for the analysis. Hourly averages were computed from acquired measurements with a resolution higher than hourly if at least 50% of the hour had available data. Hourly resolution was used since this type of measurement is usually the one used in epidemiological studies relating to hospital admissions. Negative $PM_{2.5}$ due to instrument noise at low particle concentrations and to rapid changes in air humidity were considered valid measurements. Values down to $-5\,\mu g{\cdot}m^{-3}$ were included based on the instrument manuals and government protocols (CEN, 2013; EV-007, 2013). For PNC, the set permissible values were from 1 particle·cm^{-3} to 5×10^5 particle·cm^{-3} (Cheung et al., 2011; Mejía et al., 2007). Non-compliant data were removed based on these criteria (e.g. $PM_{2.5}$ lower than $-5\,\mu g{\cdot}m^{-3}$ and PNC above 5×10^5 particle·cm^{-3}. All data analyses were carried out in R statistical software (RStudio Team, 2016) using the 'ggplot2' package (Wickham, 2009) for data visualization.

2.3.1. Comparison of PNC and $PM_{2.5}$

One fundamental problem when attempting an inter-comparison or use of particle number/size distribution data obtained by different studies is that different studies normally use different instruments for particle number concentration and size distribution measurements, with different size ranges. Although a full scale uncertainty analysis is beyond the scope of this work, and would need more data than those available from the sites compared, we conducted an assessment based on the Augsburg, Brisbane, Los Angeles and Shanghai data to obtain an indication of the magnitude of variation in PNC resulting from choosing different size ranges (see Section 2 of supplementary material). Its outcome demonstrated that the difference in PNC between 9.5 and 13.6 nm as the lower cut-off (based on Brisbane data) was only < 4.0% and < 3.0% of the mean and median concentrations, respectively. The difference between 800 and 2000 nm as the upper cut-off (based on Augsburg data) was < 5.0% and < 4.0% of the mean and median concentrations, respectively. Additionally, the Los Angeles and Shanghai data showed that at the 532.8 nm upper cut-off, 99% of the

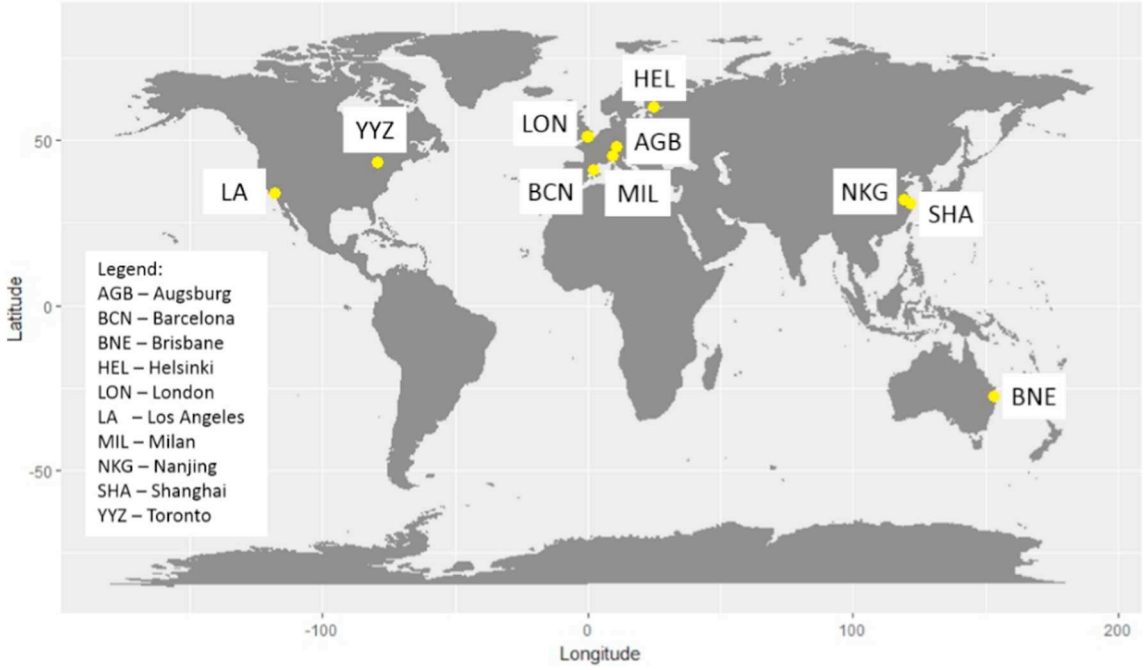

Fig. 1. Cities included in this study.

Table 1
The study areas, their climate classification and the identified PM sources.

City (code[a])	Climate (Köppen classification[b])	PM Sources[c]
Augsburg, Germany (AGB)	Marine West Coast "Cfb"	local traffic, biomass burning (for heating), secondary aerosol (long-range) (Gu et al., 2011; Schäfer et al., 2014)
Barcelona, Spain (BCN)	Mediterranean "Csa"	local traffic, secondary aerosol (harbour emissions), local dust/sea salt, industrial emissions (metallurgy) (Amato et al., 2016; Pérez et al., 2016)
Brisbane, Australia (BNE)	Humid Subtropical "Cfa"	local traffic, secondary aerosol (airport, harbour and industrial emissions), sea salt, biomass burning (for heating and forest fires) (Cheung et al., 2011; Friend and Ayoko, 2009; Friend et al., 2012)
Helsinki, Finland (HEL)	Humid Continental "Dfb"	local traffic, secondary aerosol, biomass burning (for heating), long-range transport, sea salt (S. Carbone et al., 2014; Saarikoski et al., 2008)
London, England (LON)	Marine West Coast "Cfb"	local traffic, secondary aerosol, crustal, sea salt, urban/regional background (Beddows et al., 2015; Charron et al., 2007; Crilley et al., 2017)
Los Angeles, USA (LA)	Mediterranean "Csb"	local traffic, urban background, secondary aerosol (Hasheminassab et al., 2014b; Sowlat et al., 2016)
Milan, Italy (MIL)	Marine West Coast "Cfb"	local traffic, secondary aerosol, biomass burning (Curci et al., 2015; Lonati et al., 2005; Perrone et al., 2012)
Nanjing, China (NKG)	Humid Subtropical "Cfa"	local traffic, secondary aerosol, road dust/sea salt, biomass/coal burning (An et al., 2015; Yang et al., 2005)
Shanghai, China (SHA)	Humid Subtropical "Cfa"	local traffic, industrial emissions, secondary aerosol, aged sea salt (Qiao et al., 2016; Yangjun Wang et al., 2014)
Toronto, Canada (YYZ)	Humid Continental "Dfb"	local traffic, coal burning, secondary aerosol, metallurgy (C. H. Jeong et al., 2011; Lee et al., 2003; Tsai et al., 2004)

[a] City codes (3 letters) are based on the International Air Transport Association's (IATA) identifier of airport location with AGB classified as a regional airport while the rest are international. LON and MIL, which are codes for all airports servicing the metropolitan area, are preferred over LHR and MXP, respectively, to avoid confusion with the air quality studies done for the airports. Similarly, LA is used for Los Angeles and not LAX, which also has a number of published works.
[b] Köppen climate classification – a system to classify climate based on the annual and monthly averages of temperature and precipitation.
[c] Several terminologies are used by different authors to refer to traffic as source but for consistency in this paper, local traffic may mean only vehicle exhaust or may include re-suspended road dust and other non-exhaust emissions.

particles are already accounted for. Therefore, considering the differences as small, and in the absence of size distribution data from all sites to conduct proper uncertainly analysis, we noted this as a limitation of our study, and opted to use the data for our analysis without any modification based on the size ranges. Further, the study of Rodrıguez et al. (2007) also showed that 99% of PNC are within the 10–415 nm range for Milan, London and Barcelona, which are also our study areas.

As mentioned earlier, the category of the monitoring sites in the cities investigated varies. Hence, we tested whether there is a difference in PNC and $PM_{2.5}$ based on the site category (i.e., LON1 [UB site] was tested against LON3 [RS site]). Further, an additional test was performed for $PM_{2.5}$ by comparing measurements from two monitoring sites with the same site classification in the same city (i.e., BNE2 and BNE3 are two RS sites in Brisbane, while LON1 and LON2 are two UB sites in London). Analyses were performed using pairwise mean difference test using the 'boot' function and the Kolmogorov-Smirnov (KS) two-sample test for data distribution comparison. For the comparison between cities, we chose UB sites over RS sites if data from multiple sites were available. This was because the comparison of the urban background sites is considered more informative regarding the differences between the cities, as concentrations at road sites may vary significantly, depending on traffic characteristics, while particles formed from non-traffic NPF and non-traffic are accounted for in UB concentrations (Brines et al., 2014). Thus, for comparison of PNC between cities, AGB, BCN, BNE1, HEL, LA, LON1, MIL, NKG, SHA and YYZ sites were used, while AGB, BCN, BNE2, HEL, LA, LON1, MIL, NKG, SHA and YYZ sites were selected for comparing $PM_{2.5}$. PNC and $PM_{2.5}$ per city were presented using boxplots and density plots. The pairwise mean difference test and the KS two-sample test for data distribution comparison were also used then the Generalized Additive Model (GAM) to obtain the diurnal and seasonal trend.

In order to compare the PNC and $PM_{2.5}$ in the different cities around the world, the annual mean concentrations and the data distributions were analysed. Boxplots and density plots were generated. The boxplots present the mean, median, the interquartile range (IQR), the 5th percentile and the 95th percentile. The kernel smoothing (Wand and Jones, 1995) was applied to generate the density plots. Kernel density plot is a smoothed version of a normalised histogram and is a non-parametric estimate of the probability density. Unlike histograms (frequency plots), the kernel density estimators do not depend on the width of the bins or their endpoints, therefore giving a smoother distribution.

Moreover, to compare the means, the differences in the means between all pairs of cities were tested. The means are reported here because the guideline values are based on means. Then, to compare the data distributions, which capture more information than just means, the Kolmogorov-Smirnov (KS) test (Conover, 1971) was applied. Since the data are non-Gaussian and large (sample sizes up to 8592), that classical statistical tests produce highly significant *p*-values even if the differences are negligible, the non-parametric bootstrap technique (Davison and Hinkley, 1997) was employed. Bootstrapping takes on the concept of the "law of large numbers" whereby, after repeatedly sampling the dataset, approximation of the true population is obtained. The process for the bootstrapped pairwise comparison included numerous resampling of the dataset (bootstrap samples), finding the summary statistic (i.e., in this case, the mean difference) for each sub-sample (bootstrap statistics) then producing the bootstrap distribution and obtaining the *p*-values. For very large samples, data distribution-based statistical tests, e.g., *t*-tests that compare means based on Gaussian data, can produce significant p-values even though the difference between means is, in practice, considered negligible. The non-parametric bootstrap is a distribution-free method that does not require any distributional assumptions around the data, hence, generally, does not lead to a significant p-value when differences are negligible.

The pairwise mean difference test was performed using the 'boot' function of the 'boot' package (Canty and Ripley, 2017; Davison and Hinkley, 1997) in R with 10,000 bootstrapped samples and the p-values were calculated using a Monte Carlo adjustment (Zieffler et al., 2011)). The mean difference of each pair of cities was tested at 0.05 level of significance. Then, the 95% confidence interval (C.I.) for each pairwise mean difference was obtained using the 'smean.cl.boot' function of the 'Hmisc' package (Harrell, 2018). The C.I. is the range that is likely to contain the mean difference between the pairs of cities being compared with 95% level of confidence, and can be used to assess the precision of the estimate since narrower C.I. suggests a more precise estimate of the

Table 2
Location and characteristics of the monitoring sites in the different cities.

Sites	Stations/institutions	Coordinates		Site category[a]	Date of collection	Instruments		
						PNC		$PM_{2.5}$
		Latitude	Longitude			Model	Size range (nm)	Model
Augsburg								
AGB	Fachhochschule Station by the Helmholtz Zentrum München	48.3579	10.9071	UB	Oct 2012 – Sep 2013	CPC TSI 3025	3–3000	TEOM Thermo 1400AB/FDMS 8500B
Barcelona								
BCN	Palau Reial Station by the Institute of Environmental Assessment and Water Research of the Spanish Council for Scientific Research (IDAEA-CSIC)	41.3875	2.1156	UB	Mar 2014 – Feb 2015	CPC TSI 3787	5–1000	GRIMM 180[c]
Brisbane								
BNE1	Queensland University of Technology (QUT)	−27.4774	153.0281	UB	Dec 2013 – Nov 2014	CPC TSI 3787	5–1000	
BNE2	South Brisbane Station by the *Department of Science, Information Technology and Innovation*	−27.4848	153.0321	RS	Dec 2013 – Nov 2014			TEOM 1405-DF
BNE3	Woolloongabba Station by the *Department of Science, Information Technology and Innovation*	−27.4975	153.0350	RS	Dec 2013 – Nov 2014			TEOM 1405-DF
Helsinki								
HEL	Mäkelänkatu Station by the Helsinki Regional Environmental Services Authority (HSY)	60.1964	24.9520	RS	Mar 2015 – Feb 2016	DMPS/CPC Airmodus A20	6–800	TEOM Thermo 1405D
London								
LON1	North Kensington Station by the Automatic Urban and Rural Network (AURN)	51.5211	−0.2135	UB	Jan – Dec 2015	CPC TSI 3775	4–1000	TEOM Thermo 1400AB/FDMS 8500
LON2	Bloomsbury Station by the Automatic Urban and Rural Network (AURN)	51.5223	−0.1259	UB	Jan – Dec 2015			TEOM Thermo 1400AB/FDMS 8500
LON3	Marylebone Station by the Automatic Urban and Rural Network (AURN)	51.5225	−0.1546	RS	Jan – Dec 2015	CPC TSI 3775	4–1000	TEOM Thermo 1400AB/FDMS 8500
Los Angeles								
LA	Particle Instrumentation Unit (PIU) by the University of Southern California	34.0522	−118.2437	UB	Aug 2014 – Jul 2015	SMPS/CPC TSI 3022	13.6–736.5	BAM 1020 Met One Instruments
Milan								
MIL	U9 Site at University of Milano-Bicocca	45.5106	9.2117	UB	Mar 2015 – Feb 2016	CPC TSI 3775	4–1000	GRIMM 1.107
Nanjing								
NKG	Station for Observing Regional Processes of the Earth System (SORPES) by the Nanjing University	32.1206	118.9528	UB	Jan – Dec 2012	DMPS	6–800	SHARP Thermo Sci 5030
Shanghai								
SHA	Shanghai Academy of Environmental Sciences (SAES)	31.1698	121.4297	UB	Jan – Dec 2013	SMPS TSI 3936 L87/ APS	13.6–2500	BAM Thermo Sci FH62C14
Toronto								
YYZ	Southern Ontario Centre for Atmospheric Aerosol Research (SOCAAR)	43.6590	−79.3954	RS	Aug 2014 – Jul 2015	FMPS TSI 3090[b]	8–540	SHARP Thermo Sci 5030

[a] Site Category: UB – urban background, RS – roadside.
[b] FMPS data were adjusted using a correction factor by Jeong and Evans (2009) for the particle size upper limit to 540 nm from 560 nm.
[c] GRIMM 180 were in situ corrected against gravimetric measurements by high volume sample.

data population mean difference than a wider C.I.. For the comparison between the distributions, the 'ks.boot' function of the 'Matching' package (Sekhon, 2011) was used. In a KS two-sample test, the empirical distribution functions (EDF) for both samples were compared and D (KS test statistic) was computed as the distance between these two curves.

The diurnal and seasonal trends of both PNC and $PM_{2.5}$ in the different cities investigated were modelled using a Generalized Additive Model (GAM) (Clifford et al., 2011; Salimi et al., 2017). GAM uses a predictor that involves the sum of smooth functions of covariates. Hence, GAM is considered to be more flexible than a Generalized Linear Model (GLM), which simply uses a linear additive function of the covariates. The GAM equation used in the "stat_smooth" function of the 'ggplot2' package (Wickham, 2009) was y ~ s(x, bs = "cs", k = 8); this uses cubic splines as the smoothing function with 8 knots since some cities had few data points. Knots are cutpoints where fitting must occur, therefore affected by the number of data points and the increments on the x-axis; fewer knots tend to over smooth the curve. In the GAM plots, the line is the fitted GAM while the dots are the actual data; a darker shade means higher count for that particular concentration. Diurnal plots of the data are sorted from the 0th to 23rd hour in local time (LT) while the seasonal plots sort data by month.

2.3.2. Relationship between PNC and $PM_{2.5}$

A scatter plot of the annual median values of the PNC and $PM_{2.5}$ for all the cities, the preferred measure of central tendency for highly skewed data, was prepared to show how these two metrics vary in relation to each other. Further, the annual median PNC was divided by the annual median $PM_{2.5}$ for each city to obtain the particle number-to-mass ratio. These ratios provide quantitative relation between PNC and $PM_{2.5}$ with reference to the sources contributing to their concentrations. When calculating the PNC and $PM_{2.5}$ ratios, we need to consider the extent to which PNC accounts for all the particles within the aerodynamic range $< 2.5\,\mu m$. As we have shown above, the count of particles in the size range $> 1\,\mu m$ is insignificant in contributing to the total PNC $< 2.5\,\mu m$. Hence, the PNC values reported by all the studies and used here for the analysis are reasonably accurate estimates of the actual PNC within the aerodynamic size $< 2.5\,\mu m$, and can therefore be used for computing the particle number to $PM_{2.5}$ mass ratios. Further, because the two metrics were measured separately, the value of one would not have affected the other, and is therefore not biased.

The linear association between PNC and $PM_{2.5}$ was explored through the Pearson correlation coefficients (r) for each city and was tested, again using the non-parametric bootstrap test. The same procedure was also carried out after a log-transformation to both PNC and $PM_{2.5}$. Log-transformation resulted in more apparent linear relationships between the log-transformed data for all cities. Linearity is an assumption of Pearson's correlation; hence, there may be more confidence in the validity of the results around the log-transformed data. The linear relationship was of more interest to this study to determine if the two metrics have a one-to-one correspondence that would demonstrate how control measures may affect concentration. Additionally, pairwise tests of independence between PNC and $PM_{2.5}$, based on the empirical copula process, were carried out using the 'indepTest' of the 'copula' package' (Hofert et al., 2017). Unlike Pearson's correlation, a copula, and hence, the copula test of independence, does not require the relationship between two sets of data to have any particular form (Genest and Rémillard, 2004). Therefore, results from the copula test of independence are valid whether the relationship between PNC and $PM_{2.5}$ is linear or not.

3. Results

3.1. PNC and $PM_{2.5}$ general trends

Mean annual concentrations of PNC and $PM_{2.5}$ and their data distributions in the ten cities were compared by testing for mean differences between pairs of cities. Further, diurnal and seasonal patterns in concentrations were compared. Results are presented using boxplots (Fig. 2 and Fig. 4) and density plots (Fig. 3 and Fig. 5); the tables with the results of pairwise tests and GAM plots are provided in the supplementary material (Table S1a, Table S1b, Fig. S1a, Fig. S1b, Fig. S2a and Fig. S2b). Although the extent of right-skewness was apparent in both the boxplots and density plots, the density plots provide a more complete visual summary of the estimated shape of the data distributions, depicting, for example, skewness ('measure of symmetry') and kurtosis ('measure of tailedness'). The tables of pairwise tests give the bootstrap test results (C.I. and asterisk if significant) for the mean difference in PNC and $PM_{2.5}$ between pairs of cities, and the KS test results (test statistic D and asterisk if significant) for the difference in the distributions of the two particle metrics between pairs of cities. The interpretation of the GAM plots was focused on the trends of the curves.

3.1.1. PNC trends

In general, PNC were lower at UB sites (AGB, BCN, BNE1, LON1, and LA) compared to RS sites (HEL, LON3, and YYZ), except at MIL, NKG and SHA (Fig. 2). In order to understand the extent of variation in PNC between the two types of monitoring sites, LON1 (UB) and LON3 (RS) in London were compared. The mean values of concentrations were 8.6×10^3 and 20.1×10^3 particles·cm^{-3}, for LON1 and LON3, respectively, and the C.I. for the mean difference (UB – RS) was [-12.0×10^3, -11.3×10^3 particles·cm^{-3}] with a bootstrapped p-

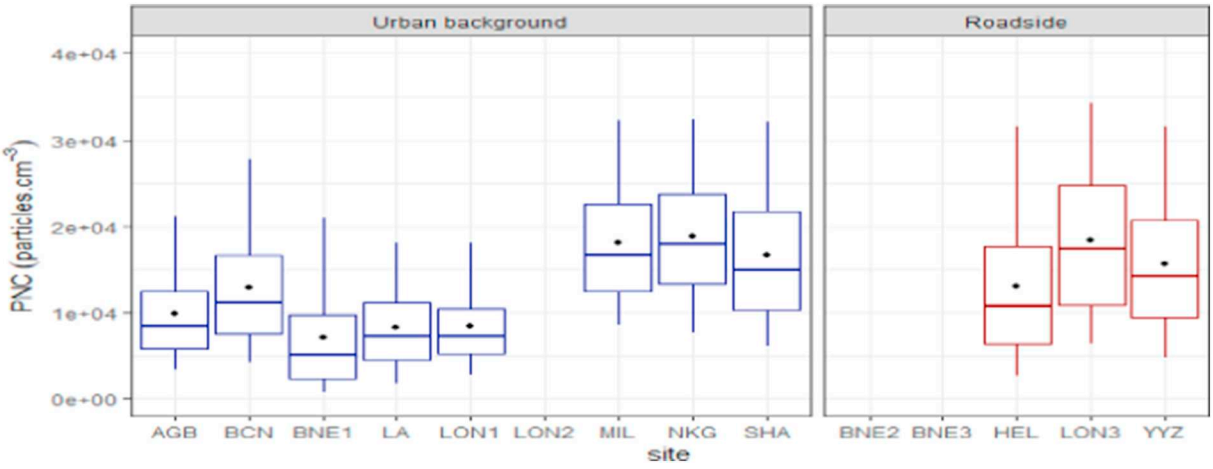

Fig. 2. Boxplot with the mean, median, the interquartile range (IQR), the 5th percentile and the 95th percentile of the hourly PNC in the investigated cities.

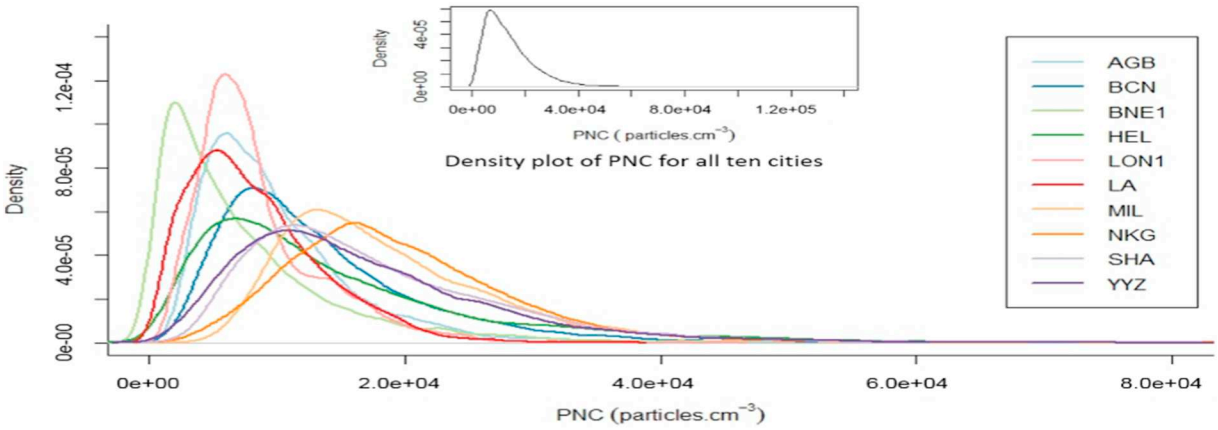

Fig. 3. Density plot of hourly PNC per city.

value of < 0.001 indicating that the means were significantly different. In addition, at 0.05 significance level, the KS test determined that the two sites were different in terms of their PNC data distributions (test statistic D = 0.55, $p \leq 0.001$).

As explained earlier, UB sites were chosen for comparing the cities. The mean PNC (Fig. 2, Table S1a and Table S1b) ranged from 8.0×10^3 to 19.5×10^3 particles·cm^{-3}, which is a relatively small range, considering that the PNC range is by two to three orders of magnitude between different ambient environments (Morawska et al., 2008). Brisbane had the lowest, while Nanjing the highest PNC. Among the European cities, London had the lowest concentration followed by Augsburg, Barcelona, Helsinki then Milan. The high PNC in Helsinki and Toronto can be attributed to the location of the monitoring sites as they were in proximity to a road (RS). Los Angeles had PNC similar to London.

Similar to the boxplot, the density plot (Fig. 3) showed that Helsinki, Toronto, Milan, Shanghai and Nanjing had a wider spread of PNC. Nanjing had an almost normal distribution while Brisbane had the most skewed. Although both boxplots and density plots have shown the differences in the measures of central tendency as well as in the shape of data distribution in cities, it is still necessary to test the difference statistically (Table S2a). At a 0.05 significance level, the means for the pairs of cities differ significantly between each other except between London and Los Angeles ($p = 0.13$). However, the KS test revealed that although the means for these cities are very similar, the data distributions are different (D = 0.09); e.g., London has a higher density peak while Los Angeles has a wider spread (Fig. 3). Further, the data distribution varied between cities. Only Toronto and Shanghai had very similar curves, with the smallest KS test statistic (D = 0.05).

Another comparison done between the cities was in relation to the temporal trends in PNC. In the diurnal plot (Fig. S1a), all ten cities had the lowest PNC early in the morning (02,00 – 04:00 LT). Three concentration peaks can be observed in some cities (BCN, BNE, LA, and NKG), while others only have two (AGB, HEL, LON, MIL, SHA and YYZ). The peaks occurred in the morning around 07:00 LT, at midday about 12:00 LT and at night around 20:00 LT. However, concentration peaks for London, Los Angeles and Nanjing were not so distinct. Helsinki showed the greatest increase (almost 15×10^3 particles·cm^{-3}) in the morning among all the cities followed by Toronto and Barcelona. Further, it is worth noting that, while troughs and peaks appear to occur, PNC varied only by a factor of 5, less than an order of magnitude (i.e., mean PNC ranged from 5×10^3 to 25×10^3 particles·cm^{-3} based on GAM).

The effect of seasonal variability in the PNC was also investigated. In general, across all investigated cities, warm seasons were characterised with lower PNC than cold seasons (Fig. S2a). Brisbane and Shanghai showed the most observable changes in PNC ($> 10 \times 10^3$ particles·cm^{-3}) based on GAM, other cities had relatively small variation in concentrations. PNC in Brisbane were high from May to August, the cooler months being in the southern hemisphere. Although Helsinki and Toronto have the same climate type, seasonal variability in PNC was more distinct in Toronto.

3.1.2. $PM_{2.5}$ trend

In contrast to PNC, the $PM_{2.5}$ were comparable regardless of whether measured at UB or RS sites, except for NKG and SHA based on the

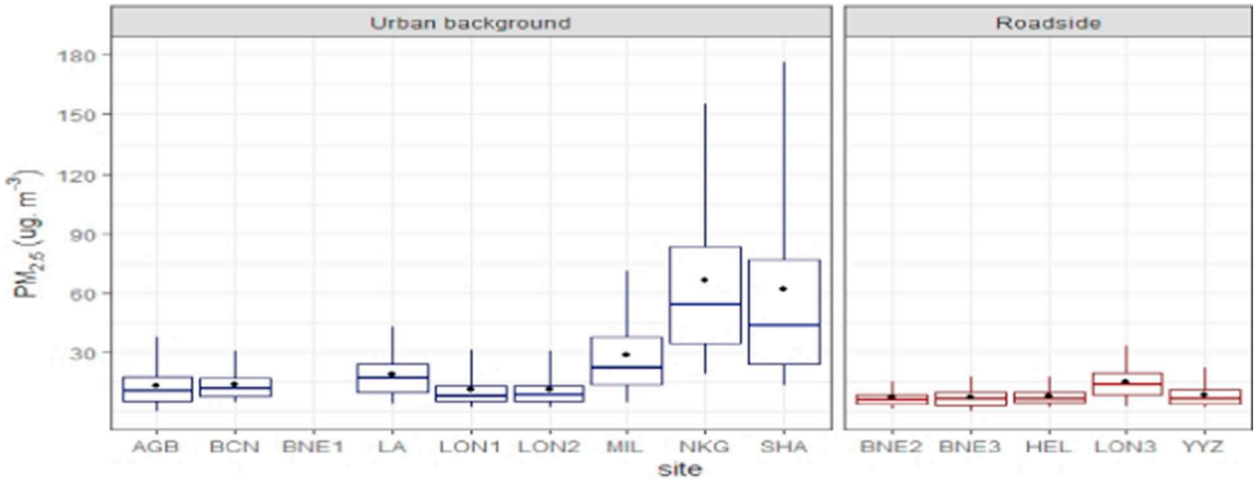

Fig. 4. Boxplot with the mean, median, the interquartile range (IQR), the 5th percentile and the 95th percentile of the hourly $PM_{2.5}$ in the investigated cities.

boxplot (Fig. 4). Nonetheless, the mean difference and data distribution of the $PM_{2.5}$ in LON1 (UB) and LON3 (RS) were tested. The mean concentrations were 10.9 and 15.3 $\mu g \cdot m^{-3}$ for LON1 and LON3, respectively and were significantly different ($p \leq 0.001$), with a C.I. for the mean difference of $[-4.8, -4.2\ \mu g \cdot m^{-3}]$. The KS test also indicated that the data distribution for LON1 and LON3 were significantly different (D = 0.32, $p = 0.001$). However, the D statistic for $PM_{2.5}$ was lower compared with the D statistic obtained for PNC ($D_{PM2.5} = 0.32$ vs $D_{PNC} = 0.55$). This means that the empirical distribution functions (EDF) curves of $PM_{2.5}$ for UB and RS were closer i.e., the differences in the data distribution were smaller. Hence, although site category influences the measured concentrations for both PNC and $PM_{2.5}$, it affects $PM_{2.5}$ less than PNC as observed in this single city where both PNC and $PM_{2.5}$ data were available.

Next, in order to show how $PM_{2.5}$ differ between stations, two sites with the same classification were compared: two UB sites in London (LON1 and LON2) and two RS sites in Brisbane (BNE2 and BNE3). The mean annual $PM_{2.5}$ measured at the UB sites were the same, 10.9 and 10.9 $\mu g \cdot m^{-3}$ for LON1 and LON2, respectively and so not statistically different ($p = 0.66$). For the RS sites, the mean annual $PM_{2.5}$ were comparable, but still statistically different ($p < 0.001$), 7.0 and 7.3 $\mu g \cdot m^{-3}$ for BNE2 and BNE3, respectively. Additionally, the data distribution for both UB (D = 0.04, p = 0.001) and RS (D = 0.08, $p \leq 0.001$) sites differ based on the KS test results but both with very low D statistic.

When comparing the $PM_{2.5}$ of different cities, the same sites for PNC were used except for BNE1, which only measured PNC. Therefore, BNE2 was used and was preferred for its proximity to BNE1. The highest $PM_{2.5}$ annual mean concentrations were measured in Nanjing and Shanghai while the lowest in Brisbane (Fig. 4), which is similar to PNC. Also similar to mean PNC, the mean $PM_{2.5}$ was relatively higher than the medians as can be observed in the boxplot. Nanjing and Shanghai had the widest interquartile range (IQR) with similar data distribution (Fig. 5), while Brisbane and Helsinki had the narrowest IQR, also with similar data distribution. In testing the pairwise mean differences between the cities, the $PM_{2.5}$ means in all cities differ significantly (Table S2b), even those of Augsburg and Barcelona (13.3 and 13.7 $\mu g \cdot m^{-3}$ for AGB and BCN, respectively). The results of the KS test also confirmed that the data distribution between cities were different, with Brisbane, Helsinki, London and Toronto having the most similar shape (i.e., small D statistic, 0.09–0.13).

In the diurnal plot (Fig. S1b), only Los Angeles showed an evident decline (~13 units drop) from the peak concentration around 10:00 LT to the lowest at 16:00 LT. The concentrations in Milan and Nanjing varied somewhat (~7 and 9 units drop, respectively); being elevated in the early morning until around 06:00 LT, decreasing to mid-day then rising again. $PM_{2.5}$ in all other cities did not vary much throughout the day; changes in $PM_{2.5}$ were $< 5\ \mu g \cdot m^{-3}$.

In general, $PM_{2.5}$ was higher during the winter until early spring (Fig. S2b). The peak $PM_{2.5}$ in Shanghai was between December and January. Los Angeles and Nanjing had the peak between January and February, but Nanjing had a second peak in November. Augsburg, Barcelona and London had its peak concentrations between February and March. Milan's data do not cover the whole year, but the peak was only in December. $PM_{2.5}$ concentrations in Helsinki, Toronto and Brisbane were almost constant, and at low levels throughout the year, though the somewhat elevated concentration in Helsinki and Toronto occurred in February while it was in July for Brisbane.

3.2. Relationship between PNC and $PM_{2.5}$

The relationship between PNC and $PM_{2.5}$ is presented in three ways: a scatter plot of the annual city medians (Fig. 6), particle number-to-mass concentration ratio (Fig. 7) and the Pearson's correlation (Fig. 8). The scatter plot of the annual medians shows how the two metrics vary; a low PNC does not always imply a low $PM_{2.5}$ and vice versa. Two main clusters are seen in Fig. 6: the cluster on the left is cities of low $PM_{2.5}$, while the cluster on the right is cities of high $PM_{2.5}$. Helsinki (HEL) and Toronto (YYZ) are presented in a sub-cluster of the left cluster due to very low $PM_{2.5}$ but high PNC. Milan (MIL) at the top right of the left cluster, although an UB site, had very high PNC with a somewhat elevated $PM_{2.5}$. In contrast, Los Angeles (LA) at the bottom right of the left cluster had a low PNC but quite high $PM_{2.5}$. It can also be observed that the right cluster contains both cities in China, Nanjing (NKG) and Shanghai (SHA), which had high PNC and $PM_{2.5}$. Further, a trend line for the UB sites was derived using the nonparametric regression, LOESS.

Next, ratios of median particle number-to-mass concentrations per city were computed (Fig. 7 and Table S2) in order to provide a quantitative measure of the relationship between PNC and $PM_{2.5}$ and to investigate how it varies for different cities. Comparing among the cities and between UB and RS sites, cities with RS sites have PNC:$PM_{2.5}$ of $> 1.0 \times 10^9$ particles·μg^{-1} (HEL and YYZ) while cities with UB sites, have $< 1.0 \times 10^9$ particles·μg^{-1}. Moreover, the RS sites in London (LON3) has a PNC median of 18.3×10^3 particles·cm^{-3} and a $PM_{2.5}$ median of 13.6 $\mu g \cdot m^{-3}$, thus a ratio of 1.3×10^9 particles·μg^{-1}. Though this result is not shown in the plot, this supported that RS sites had > 1 particle number-to-mass ratio. Among the cities with UB sites, NKG and SHA have the lowest ratio of $\sim 0.3 \times 10^9\ \mu g^{-1}$. The ratio for LA is also low ($< 0.50 \times 10^9$ particles·μg^{-1}) compared with the other cities ($> 0.70 \times 10^9$ particles·μg^{-1}).

In order to test statistically the relationship between hourly PNC and $PM_{2.5}$ in the different cities, the non-parametric bootstrap test of the Pearson's correlation was employed (Fig. 8 and Table S3). Based on the calculated Pearson's correlation coefficient (r), the obtained values showed very weak (Barcelona and Los Angeles at 0.08 and 0.07, respectively) to moderate (Shanghai at 0.53) linear relationships. The

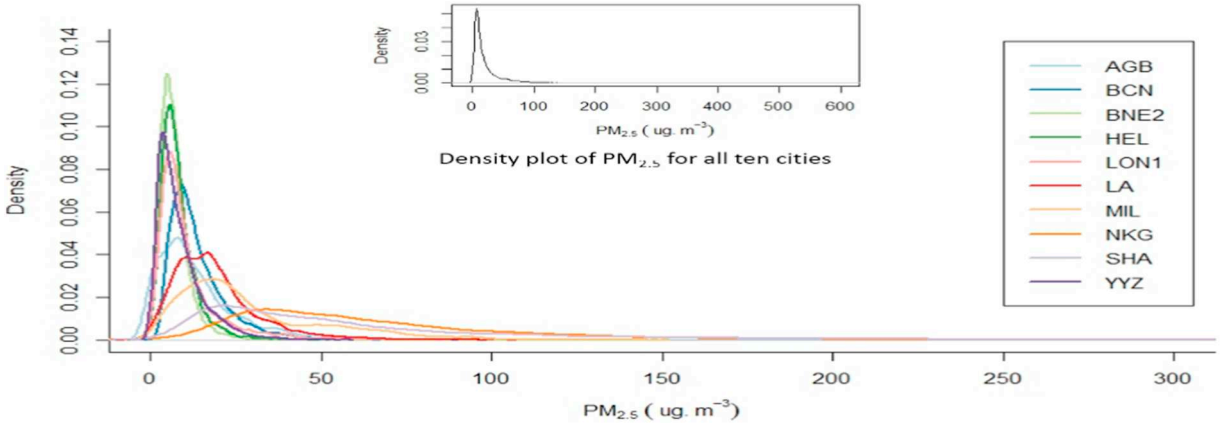

Fig. 5. Density plot of hourly $PM_{2.5}$ per city.

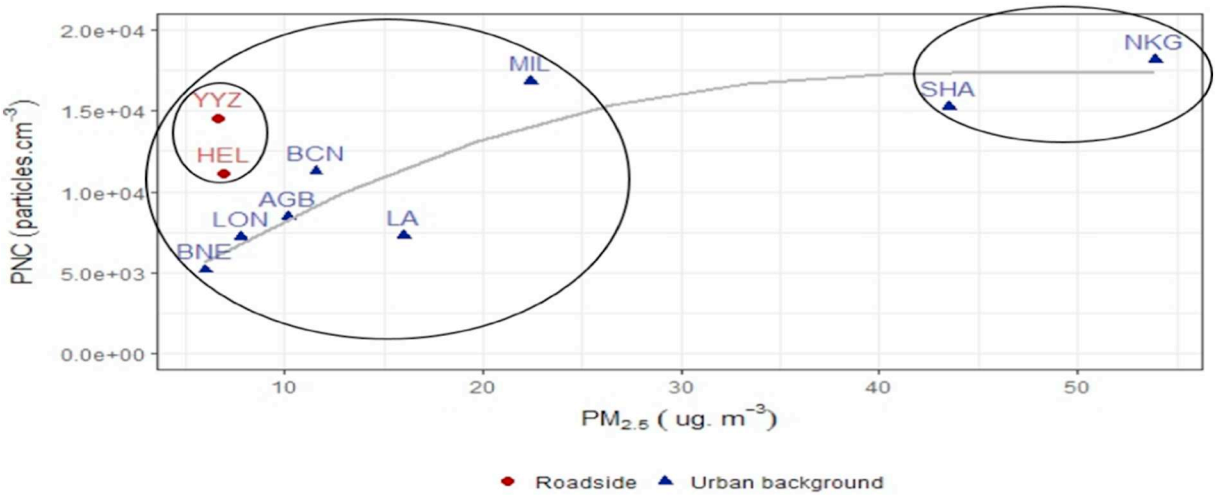

Fig. 6. Annual median of the hourly PNC and $PM_{2.5}$ with the fitted LOESS trend line and clustering of concentration level.

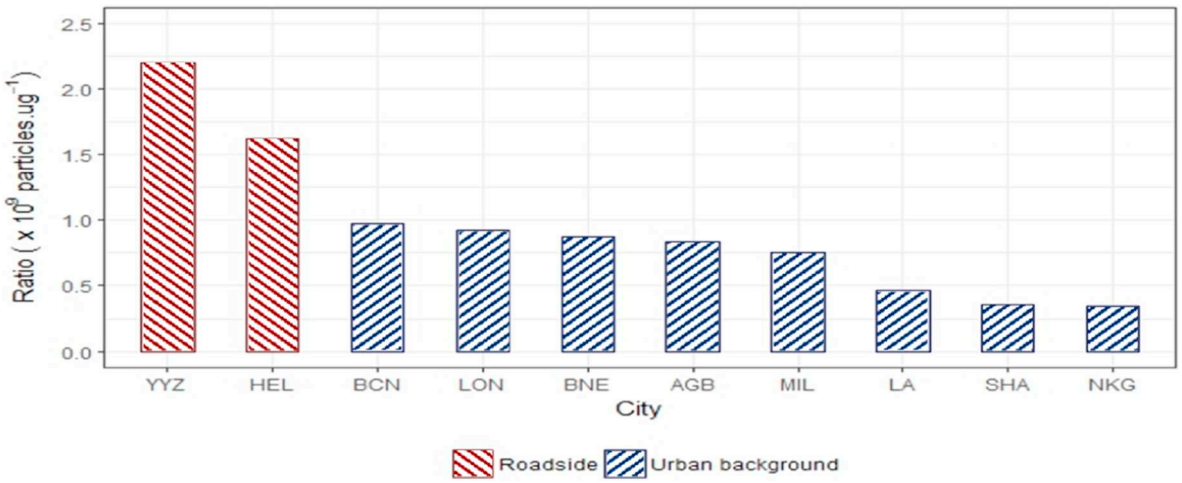

Fig. 7. Particle number-to-mass ratio (PNC:$PM_{2.5}$) of the annual median concentration.

bootstrapped *p*-values were all > 0.05. The log transformation of both metrics provided higher Pearson's *r* for most cities, but similarly indicated only very weak to moderate linear correlations; Shanghai ($r = 0.64$) showing the most linear relationship among the cities. The Spearman's correlation, which does not assume a linear relationship and is more robust to extreme observations in comparison to Pearson's correlation, was also computed for all cities. However, the obtained values for Spearman's correlation were almost equal to the Pearson's correlation after log-transformation so are not additionally reported here. Further, based on the pairwise tests of independence using the empirical copula process, all p-values obtained were < 0.05, meaning the two metrics, PNC and $PM_{2.5}$, were somehow dependent.

4. Discussion

The PNC (Fig. 2 and Fig. 3) and $PM_{2.5}$ (Fig. 4 and Fig. 5) in the different cities under investigation varied. The pairwise comparison of the mean difference and KS test (Table S1a and Table S1b) confirmed that PNC and $PM_{2.5}$ annual means and the data distributions of each city were, in general, significantly different. However, some similarities were observed in the diurnal and seasonal trends of PNC and $PM_{2.5}$ for the cities (Fig. S1a, Fig. S1b, Fig. S2a and Fig. S2b). For example, all PNC diurnal plots showed concentration peaks in the morning, midday and/or night time, unlike most $PM_{2.5}$ diurnal plots, and both PNC and $PM_{2.5}$ were higher during the winter. However, the magnitude of variation differed among cities. Therefore, even though all sites are located in urban areas and subjected to anthropogenic emissions, with transportation, industry and household dominate (Butler et al., 2008; Karagulian et al., 2015), other factors contribute as well. Both anthropogenic (e.g., population density and technology) and natural (e.g., meteorology and geography) factors will be reviewed. In addition, given the dynamics of the PNC and $PM_{2.5}$ in the ten cities, an insight into the relationship between the two metrics can now be provided.

4.1. PNC and $PM_{2.5}$ drivers

Overall, primary and secondary UFP come from combustion processes thus PNC is significantly affected by tail-pipe emissions and NPF from precursors originating from city transport. $PM_{2.5}$, on the other hand, are mostly aged primary or transported secondary aerosols. Hence, the UB sites had lower PNC compared to RS sites (LON3, HEL and YYZ), but the $PM_{2.5}$ was not much affected by the site location (Fig. 2 and Fig. 4). Additionally, $PM_{2.5}$, is more homogenous compared to PNC regardless of site classification (i.e., BNE1 and BNE2 are RS sites, while LON1 and LON2 are UB sites, but the measured concentrations are comparable between the sites). This supports the conclusions from studies conducted by Gomišček et al. (2004) and Puustinen et al. (2007) in Europe demonstrating that PNC varies spatially within a city unlike $PM_{2.5}$. PNC emissions from traffic are more spatially heterogeneous than from stationary combustion sources (Gu

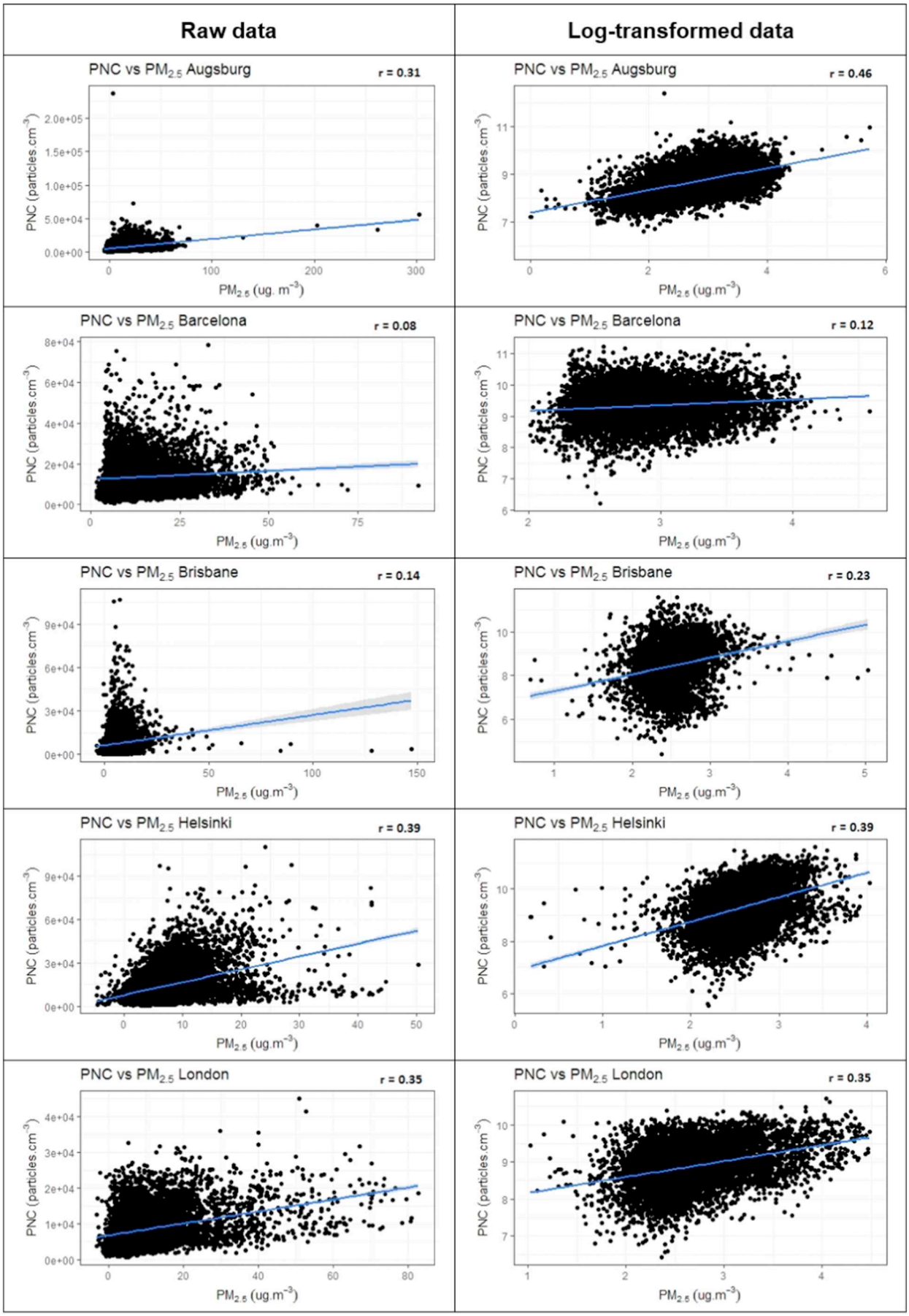

Fig. 8. Pearson's correlation (*r*) of the raw and log-transformed hourly PNC and $PM_{2.5}$ by city.

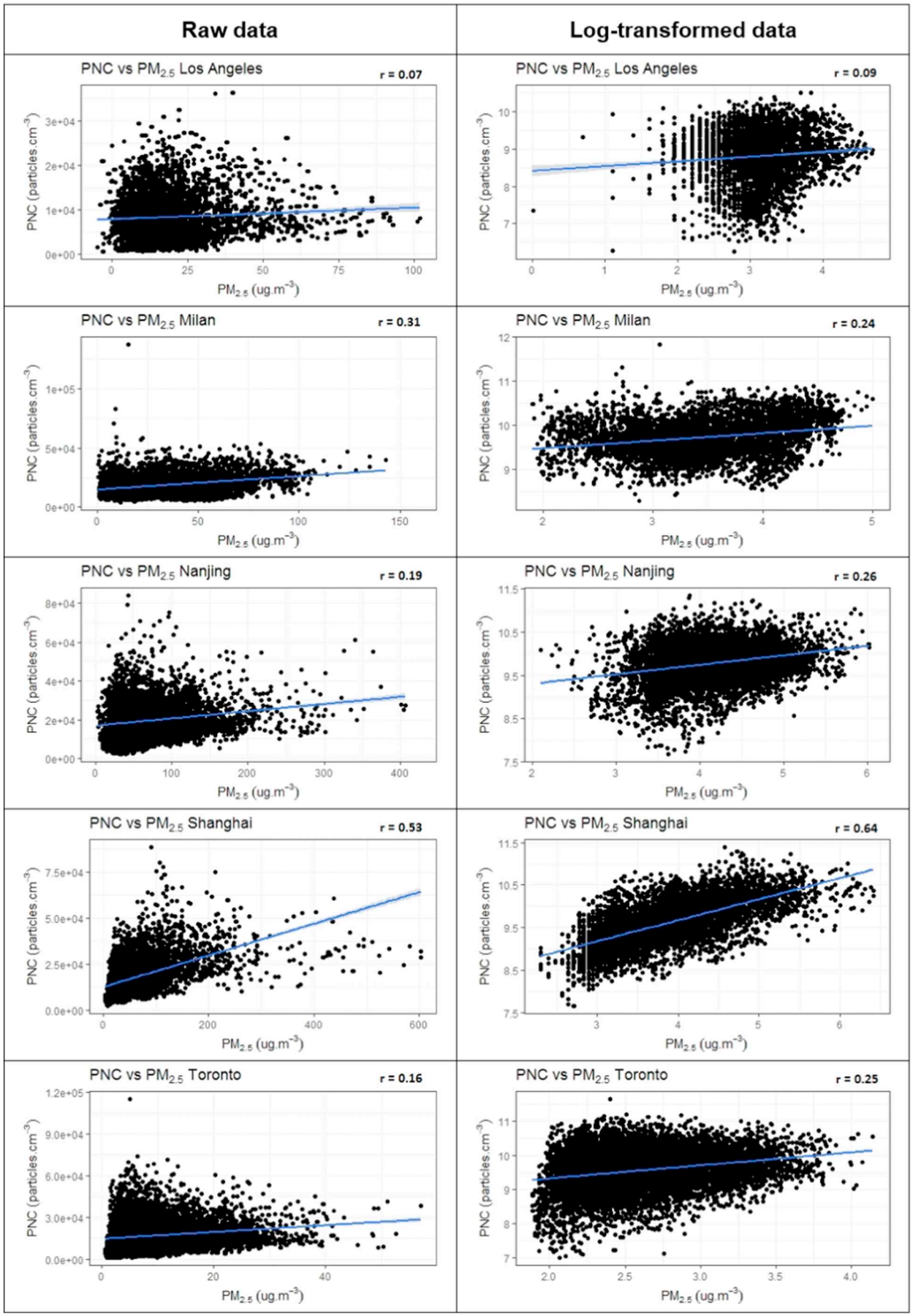

Fig. 8. (*continued*)

et al., 2013).

Fig. 3 and Fig. 5 show how different are the data distributions of PNC and $PM_{2.5}$. PNC are characterised by a wider range (both in counts and concentrations); which implies that emission sources or magnitude of emissions contributing to PNC are diverse and therefore formation and transport of UFP are affected by several factors in most cities. On the contrary, $PM_{2.5}$ is more clustered, except for Nanjing and Shanghai. The data distribution for both PNC and $PM_{2.5}$ are typically skewed due to occasional high and rare, exceptionally high concentrations. Neither the UB sites nor the RS sites showed distinct trend on the occurrence of concentrations above the 95th percentile per city except for the $PM_{2.5}$ in Milan, which mostly occurred from October to December as observed in the time series (Fig. S3). Further, looking at the uppermost concentrations, the peak PNC and $PM_{2.5}$ never occurred at the same day and time in all investigated cities (i.e., further supporting that high PNC does not correspond to high $PM_{2.5}$). However, for this study, determining the cause or source of these extreme concentrations is not in scope since no data on meteorology e.g., temperature, wind speed or wind direction nor source inventory were obtained.

In the diurnal and seasonal plots (Fig. S1a, Fig. S1b, Fig. S2a and Fig. S2b), only PNC shows distinct trend with peak concentrations during traffic rush hours in the early morning and early evening, while the observed third peak in some cities (e.g., BCN, BNE, LA and NKG) is attributed to NPF. Low PM concentrations within the day usually happen with increased mixing layer height. Both PNC and $PM_{2.5}$ exhibit seasonal variation in all investigated cities, with elevated concentrations occurring during winter due to additional fuel burning for heating, low temperatures enhancing NPF, and/or a more stagnant atmosphere (Carbone et al., 2010; Hasheminassab et al., 2014b; Jeong et al., 2011; Li et al., 2015; Liu et al., 2018). High PM concentrations during warm seasons is attributed to increased insolation promoting NPF (Brines et al., 2015) and reduced precipitation. A more detailed description of the factors affecting the PNC and $PM_{2.5}$ in each city, based on the existing literature, is provided in Section 3 of the supplementary material. In summary:

- the low PNC and $PM_{2.5}$ in BNE and the narrow but skewed data distribution are due to limited possible sources and constant atmospheric conditions throughout the year with occasional disturbances leading to extreme PNC and $PM_{2.5}$;
- Helsinki, Milan, Nanjing, Shanghai and Toronto are cities with high PNC and similar broad data distribution (but only Nanjing and Shanghai also have high $PM_{2.5}$)
  - o the elevated PNC in HEL and YYZ is due to station siting and is therefore dependent on traffic volume, while $PM_{2.5}$ levels mainly result from transported primary and secondary emissions with a significant contribution of road dust in spring
  - o MIL, although an UB station, is still much affected by traffic emissions given that it is close to two local roads and that secondary aerosol formation and accumulation are favoured within the Po Valley, contributing to the urban PM concentration
  - o NKG and SHA, both located in the Yangtze River Delta, have significant local emission sources due to continuous urbanisation but are also much affected by regional transport coming from the North China Plain and the Pearl River Delta in the south;
- the somewhat high PNC but average $PM_{2.5}$ in BCN can be attributed to road traffic emissions and the enhanced NPF in the Western Mediterranean Basin especially during summer when a high concentration of O_3 is transported to Barcelona from continental Europe;
- the somewhat high $PM_{2.5}$ but low PNC in LA is due to formation of secondary aerosols influenced by the area's meteorological characteristics with contribution from non-exhaust vehicular emissions during winter and aged sea salt in summer;
- Augsburg and London have average PNC and $PM_{2.5}$ concentrations, although AGB is slightly higher than LON due to meteorological influences – both background stations are influenced by traffic but with only the primary emissions contributing to PM as NPF was observed to be insignificant in these cities (i.e. only significant in summer because of increased solar radiation);
- Sahara dust is another important source for Barcelona and a very minor source for Augsburg and London where contribution can actually occur throughout the year but with episodes mostly occurring from mid-spring to early autumn.

Over the years, there have been significant advances in engine design and fuel formulation to control the PM, NO_x and SO_s in the exhaust. London has introduced the "sulphur free" diesel (< 10 ppm S) in 2007 and within a few months, PNC has decreased dramatically (30–59% less), but with no similar effect on the NO_x concentrations and the regulated mass metrics (Jones et al., 2012). Other cities have observed similar reduction in PNC, especially at kerbsides, upon regulating sulphur content in fuel (Johnson et al., 2009; Wåhlin, 2009; Wang et al., 2011). Another regulation was setting emission standards for vehicles, particularly for particulate matter and NO_x. This took effect in the USA also in 2007, requiring diesel engine manufacturers to reduce emissions by 90%, with 100% compliance by 2010. In addition, the California Air Resources Board in 2012 required all heavy diesel trucks to use diesel particulate filters (DPFs) to attain the 2007 emission standard by 2014. Emissions from the ports of Los Angeles and Long Beach (30 km away from LA site), which are among the busiest in the USA, were also regulated. A study by Hasheminassab et al. (2014a) has shown that these control measures were effective in reducing $PM_{2.5}$ in Los Angeles. A summary of other strategies implemented for Urban Air Quality Management (UAQM) in developed and developing countries were compiled by Gulia et al. (2015).

Both primary and secondary particulate matter may be transported from neighbouring cities or countries, seaports and airports, coal-fired facilities in industrial areas, residential areas or forest fires areas, coastal or arid region (Brook et al., 2007; Buonanno and Morawska, 2015; Chowdhury et al., 2007; Han et al., 2008; Keuken et al., 2012; Squizzato et al., 2017; Vassilakos et al., 2005; Wang et al., 2015). In 2005, the UK airports emitted approximately 3.1×10^5 kg of total $PM_{2.5}$ (i.e., inventory of 20 airports) with the London Heathrow Airport having the highest emissions per air traffic movement at 157 g (Stettler et al., 2011). Brisbane Airport has an annual mean emission of 1.98×10^{24} of PNC and 1.35×10^4 kg of $PM_{2.5}$ (Mazaheri et al., 2011) and gaseous emissions from aircrafts was thought to enhance nucleation in the city (Cheung et al., 2011). Particulate matter generated by aircrafts can increase PNC by 2–5 folds within 60 km^2 area and extend up to 16 km downwind of the runways based on a study done in the Los Angeles Airport which has an emission rate of 1.03×10^{23} particles·yr^{-1} and 7.89×10^3 kg·yr^{-1} for PNC and $PM_{2.5}$, respectively (Hudda et al., 2014; Shirmohammadi et al., 2017). For air pollutants from ports, the contribution from shipping is comparable to road traffic with mean daily concentrations of 12.6 μg·m^{-3} $PM_{2.5}$ and 1.3×10^4 particles·cm^{-3} PNC (Merico et al., 2016). The majority of particles that dominated PNC (99%) were in the size range 9–250 nm, which accounted for only 11.7% of $PM_{2.5}$. Gaseous pollutants from shipping emission may also enhance NPF in urban areas (Viana et al., 2014).

A number of studies have linked primary PM emissions and those of the gaseous precursors for secondary PM to urban population density attributed to high energy demand and therefore increased fuel consumption and combustion emissions (Gurjar et al., 2016; Lin and Zhu, 2018). However, per capita emissions in cities are actually an interplay of several natural environmental and anthropogenic conditions (Han et al., 2016; Larkin et al., 2016; Mayer, 1999; Wang et al., 2017). Focusing on per capita transportation emissions in a city, urbanisation and fuel consumption for transportation are in general inversely related, but activity levels in adjacent localities are greatly influenced, amplifying transportation fuel use (Ergas et al., 2016; Karathodorou et al., 2010; Newman and Kenworthy, 1989). Among the investigated cities,

Barcelona is the most densely populated city at approximately 16,000 inhabitants·km^{-2} followed by Milan at 7000 inhabitants·km^{-2} according to United Nation's 2016 Demographic Yearbook (UN-DESA, 2016). Next are Helsinki, Los Angeles, Shanghai, Toronto, and Greater London, in ascending order, which have 3000 to 5000 inhabitants·km^{-2}. The lowest population densities are for Augsburg, Nanjing and Brisbane at below 2000 inhabitants·km^{-2}.

The populations of Barcelona and Milan (about 1.5 million people in the city centre and around 5 million and 3 million, respectively, in the urban agglomeration) are actually much lower compared to the metropolitan areas of London and Shanghai (above 8 million and 20 million people, respectively) or even Nanjing (above 7 million people). However, the urbanized areas of both Barcelona and Milan are considerably smaller, hence the higher population density. The proximity to other urbanized areas like in the case of Los Angeles (with Riverside – San Bernardino and San Diego) and Toronto (with Hamilton and Oshawa) may elevate emission concentrations. Based on the results, Shanghai and Nanjing had high PM concentrations while Brisbane the lowest. Hence, in our investigated cities, population density did not seem to be the only factor to consider for the differences in ambient PM concentration.

With the understanding that PNC and $PM_{2.5}$ have different sources and drivers, the paradigm of focusing on mass as metric for air quality control needs to be revisited. In summary, only Brisbane and Helsinki have met the WHO guideline of 10 μg·m^{-3}. Brisbane, with its low $PM_{2.5}$ (annual mean of 7 μg·m^{-3}), also meets the Australia's standard of 8 μg·m^{-3} and has low PNC too. For the European cities, all except Milan complied with the EU $PM_{2.5}$ annual mean standard (25 μg·m^{-3}). Milan is located in a big basin valley (the Po Valley) whose atmospheric stability makes the whole region a European hot spot for atmospheric pollution (Ferrero et al., 2014; Rodrıguez et al., 2007). Further, although Helsinki has low $PM_{2.5}$, in fact, has high PNC at roadsides while Milan and Barcelona also have elevated background PNC. Similar to Helsinki, Toronto also has considerably elevated PNC at roadsides, while Los Angeles has low background PNC. However, Los Angeles somewhat exceeded the annual mean $PM_{2.5}$ standard set by the US Environmental Protection Agency of 15 μg·m^{-3} while Toronto complied with Canada's air quality standard for annual mean $PM_{2.5}$ (10 μg·m^{-3}). The annual mean $PM_{2.5}$ standard (35 μg·m^{-3}) set by China is the highest among the national standards but the $PM_{2.5}$ in Shanghai and Nanjing still exceeded and their PNC are also high.

4.2. Relationship between PNC and $PM_{2.5}$

The similarities and differences in the PNC and $PM_{2.5}$ ambient concentrations and data distribution for the ten cities, as well as the diurnal and seasonal variability, showed how several factors affect these two metrics differently. In the above discussion, we demonstrated that PNC and $PM_{2.5}$ have different sources and different drivers. PNC is primarily from vehicular emissions thus depends on available sources with climate and meteorological factors being key, not only for dispersion but also for NPF. On the other hand, $PM_{2.5}$ that is dominated by regional and transported secondary aerosols and highly influenced by atmospheric patterns. It was also demonstrated that low $PM_{2.5}$ does not mean low PNC, as the PNC in most cities were elevated and this was intensified when the measuring sites were situated at roadsides like in Helsinki and Toronto except for Nanjing and Shanghai. The clustering of the cities in Fig. 6 further demonstrated how the two metrics could behave differently wherein PNC varied in the cluster for the low $PM_{2.5}$ cities.

After fitting a curve in the scatter plot of the annual medians of the PNC and $PM_{2.5}$ (Fig. 6), there are a few “outliers” in the UB sites. Based on the trend line, BCN and MIL have PNC above the predicted. Although UB sites, both BCN and MIL are heavily affected by traffic with MIL getting higher contribution from traffic since the site is between Viale dell'Innovazione (total annual traffic of 4,200,000 vehicles and 480 vehicles·hr^{-1} with a peak of 870 vehicles·hr^{-1} during the morning rush hour) and Via Roberto Cozzi (total annual traffic of 1,680,000 vehicles and 191 vehicles·hr^{-1} with a peak of 323 vehicles·hr^{-1} during the morning rush hour). Further, MIL is influenced by biomass burning emissions unlike BCN and by recirculation within the Po Valley, contributing to somewhat elevated $PM_{2.5}$ (Amato et al., 2016). In the LA and SHA sites, on the other hand, the PNC are lower than expected. This is probably (at least partly) caused by much higher cut-off size (14 nm) of the instruments used compared to other sites (typically 3–6 nm). In general, NKG usually has a higher PNC and $PM_{2.5}$ compared to SHA due to the spatial oscillation process in eastern China discussed by Tao et al. (2016) and causing regional accumulation.

The particle number-to-mass concentration ratio (Fig. 7) and the log-transformed Pearson's correlation (Fig. 8) provided a quantitative measure of the relationship between PNC and $PM_{2.5}$. As the ratio is equivalent to the number of particles per unit mass (in 10^9 particles·μg^{-1}), high values suggest dominance of UFP sources in the area, and lower values imply majority of PM sources contribute to $PM_{2.5}$. Correlation between PNC (dominated by UFP) and $PM_{2.5}$, on the other hand, if very low indicate different sources contributing to PNC and $PM_{2.5}$ (Cyrys et al., 2003); UFP originate mainly from local traffic exhaust emissions or NPF and a much lower fraction of $PM_{2.5}$ originate from these sources. However, as mentioned earlier, the use of different instruments for PNC measurements, therefore different size range measured, must be taken into consideration. This can somewhat affect the ratio and the correlation, but still can provide a good estimate. Other factors that have possibly affected the association between PNC and $PM_{2.5}$ in each city are discussed below.

Helsinki and Toronto have high ratios (1.6×10^9 and 2.2×10^9 particles·μg^{-1}, respectively) since both are RS sites with great impact of traffic as the source, but have the reverse trend for the correlations ($r = 0.39$ and 0.25, respectively). The low ratio but high correlation for HEL maybe explained by its location, in a busy street canyon (traffic density of 28,000 vehicles·d^{-1} on weekdays), where dilution process is weakened. Hence, traffic emission might be considered as the dominant source for PNC and $PM_{2.5}$ in the ambient air in this area. On the other hand, YYZ is located at the intersection of local streets in the downtown area and north of a four-lane major arterial road (traffic density of 16,000 to 25,000 vehicles·d^{-1}). The high ratio but low correlation in YYZ indicates difference in contributing sources for PNC and $PM_{2.5}$. Other factors possibly responsible to the low correlation of PNC and $PM_{2.5}$ in YYZ compared to HEL are: (1) the different position of the sampling inlet (3 m above ground for PNC versus ~20 m above ground for $PM_{2.5}$); (2) the lower size range for PNC (8–540 nm only); and (3) the existing vehicle fleets (light versus heavy duty vehicles and diesel versus gasoline fuelled vehicles), which was not explored in this study.

Among the UB sites, Los Angeles, Shanghai and Nanjing have low ratio (0.46×10^9, 0.35×10^9 and 0.34×10^9 particles·μg^{-1}, respectively) but had varying correlation ($r = 0.09$, 0.64 and 0.26, respectively). For all three cities, the bulk of ambient $PM_{2.5}$ are regional secondary aerosols, but only Shanghai had a moderately correlated PNC and $PM_{2.5}$ suggesting partially the same sources (e.g., industrial, domestic and traffic) for both metrics. Los Angeles, on the other hand, was the least correlated, which implies difference in sources for PNC and $PM_{2.5}$; UFP in LA is from local traffic and NPF. Similarly, Barcelona had very weak correlation ($r = 0.12$) but had the highest ratio (0.97×10^9 particles·μg^{-1}). Both BCN and LA are affected by a major roadways (Diagonal Avenue and Interstate Freeway I-110, respectively) located about 200 m away and of traffic density above 100,000 vehicles·d^{-1}. Therefore, the higher ratio in BCN indicates higher concentration of UFP and can be attributed to the substantial contribution of NPF from photochemistry less than in LA. Further, according to Pey et al. (2008), $PM_{2.5}$ in Barcelona is relatively higher compared to other European cities, but PNC is within the range of similar urban environments, indicating a different source of $PM_{2.5}$. Concentration of precursor gases from traffic can be assumed similar due to comparable traffic density,

Table 3
Correlation of PNC and $PM_{2.5}$ from published studies.

Location	No. of sites and Site type[a]	Study duration	Coefficient	Method	Authors
Australia					
Australia	6 (I, R, RS)	Aug, 1996 – Aug, 1997 (six day cycle for 4 weeks per site)	r^2 = 0.05 correlation between PNC and $PM_{0.15}$ (< 150 nm fraction mass)	Regression (daily mean)	Keywood et al. (1999)
Asia					
Guangzhou, CN	1 (UB)	12–13 Jul, 2013 (pollution episode)	r^2 = 0.85 (accumulation mode)	Regression (hourly mean)	Han et al. (2015)
Tehran, IR	5 (UB, R, RS)	Jul – Sep, 2007 (warm season) Jan – Mar, 2008 (cold season)	r^2 = 0.63 (warm season) r^2 = 0.75 (cold season) correlation between number and mass of particles with 1–10 μm size)	Regression (daily mean)	Halek et al. (2010)
Europe					
Augsburg, DE	20 (UB, RB, RS, I)	Mar, 2014 – Apr, 2015 (2 weeks per site cycle, 3 cycles total	r = 0.64	Pearson (annual mean)	Wolf et al. (2017)
Basel, CH	20 (UB, RS)	Jan, 2011 – Dec, 2012 14 days cycle winter, summer, spring	r^2 = 0.43	Regression (bi-annual mean)	Eeftens et al. (2015)
Barcelona, ES	1 (UB)	Nov, 2003 – Dec, 2004	r = 0.05 ($N_{10–20}$) r = 0.13 ($N_{20–30}$) r = 0.19 ($N_{30–50}$) r = 0.28 ($N_{50–100}$) r = 0.37 ($N_{100–200}$) r = 0.43 ($N_{200–300}$) r = 0.54 ($N_{300–415}$) r = 0.57 ($N_{415–800}$)	Pearson (hourly mean) estimated r values from graph	Rodrıguez et al. (2007)
Copenhagen, DK	2 (RS, HW)	23 Mar – 21 Apr, 2008	total concentration r = 0.63 (HW) r = 0.61 (RS) less background concentration r = 0.70 (HW) r = 0.57 (RS)	Pearson (half-hour mean)	Wang et al. (2010)
Erfurt, DE	1 (UB)	1 Sep, 1995–21 Dec, 1998	r = 0.37 (hourly) r = 0.59 (daily)	Spearman	Cyrys et al. (2003)
Erfurt, DE	1 (UB)	Oct, 1991 – May 1992	r^2 = 0.5	Regression (daily mean)	Tuch et al. (1997)
Geneva, CH	18 (UB, RS)	Jan, 2011 – Dec, 2012 14 days cycle winter, summer, spring	r^2 = 0.17	Regression (bi-annual mean)	Eeftens et al. (2015)
Helsinki, FI	1 (SC)	Jan – Jun, 2010	r = 0.59 (weekdays) r = 0.44 (weekends)	Pearson (hourly median)	Dos Santos-Juusela et al. (2013)
London, UK	1 (SC)	28 Apr – 23 May 2003 (exposure study)	r^2 = 0.21 (Regression) r = 0.5 (Pearson)	(18 min measurement at morning, noon and afternoon)	Kaur et al. (2005)
London, UK	1 (UB)	Jan – Dec, 2003	r = 0.04 ($N_{10–20}$) r = 0.08 ($N_{20–30}$) r = 0.21 ($N_{30–50}$) r = 0.4 ($N_{50–100}$) r = 0.54 ($N_{100–200}$) r = 0.56 ($N_{200–300}$) r = 0.48 ($N_{300–415}$)	Pearson (hourly mean) estimated r values from graph	Rodrıguez et al. (2007)
Milan, IT	1 (UB)	Nov, 2003 – Dec, 2004	r = 0.2 ($N_{10–20}$) r = 0.31 ($N_{20–30}$) r = 0.45 ($N_{30–50}$) r = 0.62 ($N_{50–100}$) r = 0.75 ($N_{100–200}$) r = 0.95 ($N_{200–300}$) r = 0.98 ($N_{300–415}$) r = 0.95 ($N_{415–800}$)	Pearson (hourly mean) estimated r values from graph	Rodrıguez et al. (2007)
Rome, IT	2 (RS)	Apr, 2001 – Mar, 2003	r = 0.55	Pearson (daily mean)	Marconi et al. (2007)
North America					
Boston, US	3 (RS, UB)	Sep, 2009 – Aug, 2010 (site 1) Sep, 2010 – Jul, 2011 (site 2) Aug, 2011 – Jul, 2012 (site 3)	r = −0.01 (RS - 1) r = −0.03 (UB - 1) r = 0.26 (RS - 2) r = 0.27 (UB - 2) r = 0.32 (RS - 3) r = 0.19 (UB - 3)	Spearman (hourly median)	Patton et al. (2014)

(*continued on next page*)

Table 3 (*continued*)

Location	No. of sites and Site type[a]	Study duration	Coefficient	Method	Authors
New York, US	1 (UB)	15–16 Aug, 2008 (exposure study)	$r = -0.05$ (underground station) $r = -0.10$ (ground station) $r = 0.14$ (parks) $r = 0.16$ (street side) $r = 0.42$ (subway train) $r = 0.73$ (highway) $r = 0.072$ (overall)	Pearson (continuous measurement every 1 min along different transportation route)	Wang and Gao (2011)
Rochester, US	1 (UB)	01 Jan – 31 Dec, 2003	$r = 0.22$ (winter) $r = 0.31$ (spring) $r = 0.14$ (summer) $r = 0.31$ (fall)	Spearman (hourly mean)	Jeong et al. (2006)
Toronto, CA	1 (RS)	01 Jan – 31 Dec, 2003	$r = 0.31$ (winter) $r = 0.20$ (spring) $r = 0.16$ (summer) $r = 0.06$ (fall)	Spearman (hourly mean)	Jeong et al. (2006)

[a] Site type: I – Industrial, R – Residential, RS – Roadside, UB – Urban Background, RB – Regional Background, SC – Street Canyon, HW – highway.

although type of vehicle fleet must also be considered (i.e. especially type of fuel). Other possible factors affecting PNC and $PM_{2.5}$ are climate type (i.e., both cities are classified as 'Mediterranean' but of different types: 'hot dry-summer' for BCN and 'cool dry-summer' for LA) and availability of other sources (e.g., airport and port emissions).

Augsburg, Brisbane, London, and Milan, the rest of the UB sites, had similarly weak correlations (0.23–0.46) and comparable ratio (0.83×10^9, 0.87×10^9, 0.92×10^9 and 0.75×10^9 particles·μg^{-1}, respectively) of $< 1 \times 10^9$ but $> 0.5 \times 10^9$ particles·μg^{-1}. This means that PNC and $PM_{2.5}$ have relatively the same common sources, possibly due to a high contribution of local emissions and the consistent influence of meteorology. Although, climate classification of Brisbane is 'humid subtropical' characterised by hot and humid summers compared to the mild summers of the 'marine west coast' of Augsburg, London and Milan. The higher ratio for London and Brisbane indicates great contribution of road traffic and NPF to UFP counts. Similar to this result, Wolf et al. (2017) reported that the PNC in Augsburg was moderately correlated with O_3 and $PM_{2.5}$, but highly correlated with NO_x indicating common sources, such as traffic and industry emissions. Further, the work done by Rodrıguez et al. (2007) in Barcelona, Milan and London, investigating PNC and $PM_{2.5}$ in relation to size distribution, chemical composition and trace gases concentrations to determine the evolution of aerosols, found that $PM_{2.5}$ was highly correlated to PNC only for particles > 100 nm, which accounted for just 20% of the total PNC.

Several studies investigated the correlation between PNC and $PM_{2.5}$ in various cities, considering different factors contributing to their spatial and temporal variations, as summarised Table 3. Some key findings are that correlation between PNC and $PM_{2.5}$ increases as particle size increases and correlation varies temporally since it is higher in winter, on weekdays and for daily means compared to summer, weekends and hourly means, respectively. Correlation between PNC and $PM_{2.5}$ also varies spatially as observed across Europe, with sites in the north-western, central and southern regions of the continent. However, "baseline" ratio [$PNC_{>10nm}$ (particles·cm^{-3}) = 250 $PM_{2.5}$ ($\mu g \cdot m^{-3}$) and $PNC_{>100nm}$ (particles·cm^{-3}) = 50 $PM_{2.5}$ ($\mu g \cdot m^{-3}$)] was found for sites not affected by local emissions (clean and rural sites), and this can represent the minimum PNC associated with a given $PM_{2.5}$ loading (Putaud et al., 2010; Van Dingenen et al., 2004). For more polluted sites, increase in PNC (of particles > 10 nm) is no longer proportional to $PM_{2.5}$, implying lower contribution from traffic and photochemically generated UFP to particle mass concentrations. The correlation between PNC and $PM_{2.5}$ calculated for 10 $\mu g \cdot m^{-3}$ wide $PM_{2.5}$ bins was low ($r^2 = 0.16$, $n = 132$). A possible explanation of this is that when the city is highly polluted, the pre-existing particles serve as sinks where the semi-volatile species condense while in a "clean" environment, the semi-volatile species may produce significant amount of nucleation-derived particles (Hamed et al., 2007; Hämeri et al., 1996; Rönkkö et al., 2006).

The limitations of this study include the different size ranges used for the PNC comparison in the ten cities investigated without complete uncertainty analysis apart from the one presented in Supplementary Section 2. This was due to the absence of size distribution data from all cities and not using meteorology and source inventory data to thoroughly discuss the factors contributing to PM ambient concentrations; instead, resorting to the published literature. Given these limitations, we believe that the aims of the study have been addressed. The similarities or differences in ambient concentrations and diurnal and seasonal variability of PNC and $PM_{2.5}$ were quantitatively demonstrated using statistical tests and models. The association between them was illustrated using scatter plots, ratios and linear correlations. Different factors and sources contributed to the disparate ambient concentrations and to the slightly weak linear relationship between the two metrics in the ten cities investigated.

5. Conclusions

PNC, which is dominated by UFP, is particularly affected by local sources, especially combustion processes, with a major contribution from vehicular exhaust and NPF. $PM_{2.5}$, however, dominated by particles > 100 nm in size, is attributed to predominant local meteorological conditions and regional transport (i.e., the mixing layer height that determines occurrence of resuspension and accumulation and the prevailing wind that carries secondary aerosols). Given this main difference between PNC and $PM_{2.5}$, and the varied data distribution for these two parameters across the investigated cities (i.e., the distribution was broader for PNC than for $PM_{2.5}$), variations are attributed to several factors. Similarities exist among the cities in that all diurnal PNC contain concentration peaks related to traffic, and both PNC and $PM_{2.5}$ are high during winter; however, the magnitude of variation differs among cities. Investigation of the relationship between PNC and $PM_{2.5}$ revealed that a high PNC does not mean a high $PM_{2.5}$, and vice versa. The correlation between these two metrics is weak, especially if the sources contributing to each metric are different or contribute to their bulk concentrations in different proportions. Therefore, PNC and $PM_{2.5}$ measurements are not representative of each other.

Considering that that the two metrics quantify PM differently, control measures aiming to reduce $PM_{2.5}$ do not automatically reduce PNC. Consequently, regulating $PM_{2.5}$ does not imply a low level of risk to UFP exposure, especially in zones near roadways, airports or railways. As early as the 1990's, the use of both PNC and $PM_{2.5}$ in epidemiological studies was recommended because of the poor correlation

between the two metrics (Tuch et al., 1997). A more recent study by Harrison et al. (2010), reiterated that using only mass metrics may underestimate the impacts of airborne particle matter exposure. Therefore, monitoring both PNC and $PM_{2.5}$ is essential to fully grasp the health impact associated with the inhalation of ambient PM. However, setting a guideline value or an acceptable standard for PNC could be a complex task considering the spatial and temporal heterogeneity of this metric.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2019.05.021.

References

Aalto, P., Hämeri, K., Paatero, P., Kulmala, M., Bellander, T., Berglind, N., Forastiere, F., 2005. Aerosol particle number concentration measurements in five European cities using TSI-3022 condensation particle counter over a three-year period during health effects of air pollution on susceptible subpopulations. J. Air Waste Manage. Assoc. 55 (8), 1064–1076. https://doi.org/10.1080/10473289.2005.10464702.

Amato, F., Pandolfi, M., Escrig, A., Querol, X., Alastuey, A., Pey, J., Hopke, P.K., 2009. Quantifying road dust resuspension in urban environment by multilinear engine: a comparison with PMF2. Atmos. Environ. 43 (17), 2770–2780. https://doi.org/10.1016/j.atmosenv.2009.02.039.

Amato, F., Alastuey, A., Karanasiou, A., Lucarelli, F., Nava, S., Calzolai, G., Querol, X., 2016. AIRUSE-LIFE+: a harmonized PM speciation and source apportionment in five southern European cities. Atmos. Chem. Phys. 16 (5), 3289–3309. https://doi.org/10.5194/acp-16-3289-2016.

An, J., Duan, Q., Wang, H., Miao, Q., Shao, P., Wang, J., Zou, J., 2015. Fine particulate pollution in the Nanjing northern suburb during summer: composition and sources. Environ. Monit. Assess. 187 (9), 561. https://doi.org/10.1007/s10661-015-4765-2.

Atkinson, W.R., Fuller, W.G., Anderson, R.H., Harrison, M.R., Armstrong, M.B., 2010. Urban ambient particle metrics and health: a time-series analysis. Epidemiology 21 (4), 501–511. https://doi.org/10.1097/EDE.0b013e3181debc88.

Beddows, D.C.S., Harrison, R.M., Green, D.C., Fuller, G.W., 2015. Receptor modelling of both particle composition and size distribution from a background site in London, UK. Atmos. Chem. Phys. 15 (17), 10107–10125. https://doi.org/10.5194/acp-15-10107-2015.

Brines, M., Dall'Osto, M., Beddows, D.C.S., Harrison, R.M., Querol, X., 2014. Simplifying aerosol size distributions modes simultaneously detected at four monitoring sites during SAPUSS. Atmos. Chem. Phys. 14 (6), 2973–2986. https://doi.org/10.5194/acp-14-2973-2014.

Brines, M., Dall'Osto, M., Beddows, D. C. S., Harrison, R. M., Gómez-Moreno, F., Núñez, L., … Querol, X. (2015). Traffic and nucleation events as main sources of ultrafine particles in high-insolation developed world cities. Atmos. Chem. Phys., 15(10), 5929–5945. doi:https://doi.org/10.5194/acp-15-5929-2015

Brook, J.R., Poirot, R.L., Dann, T.F., Lee, P.K.H., Lillyman, C.D., Ip, T., 2007. Assessing sources of PM2.5 in cities influenced by regional transport. J. Toxic. Environ. Health A 70 (3–4), 191–199. https://doi.org/10.1080/15287390600883000.

Buonanno, G., Morawska, L., 2015. Ultrafine particle emission of waste incinerators and comparison to the exposure of urban citizens. Waste Manag. 37, 75–81. https://doi.org/10.1016/j.wasman.2014.03.008.

Burton, R.M., Suh, H.H., Koutrakis, P., 1996. Spatial variation in particulate concentrations within metropolitan Philadelphia. Environmental Science & Technology 30 (2), 400–407. https://doi.org/10.1021/es950030f.

Butler, T.M., Lawrence, M.G., Gurjar, B.R., van Aardenne, J., Schultz, M., Lelieveld, J., 2008. The representation of emissions from megacities in global emission inventories. Atmos. Environ. 42 (4), 703–719. https://doi.org/10.1016/j.atmosenv.2007.09.060.

Buzorius, G., Hämeri, K., Pekkanen, J., Kulmala, M., 1999. Spatial variation of aerosol number concentration in Helsinki city. Atmos. Environ. 33 (4), 553–565. https://doi.org/10.1016/S1352-2310(98)00287-8.

Canty, A., Ripley, B., 2017. Boot: Bootstrap R (S-Plus) Functions. (*R package version 1.3-20)*.

Carbone, C., Decesari, S., Mircea, M., Giulianelli, L., Finessi, E., Rinaldi, M., Facchini, M.C., 2010. Size-resolved aerosol chemical composition over the Italian Peninsula during typical summer and winter conditions. Atmos. Environ. 44 (39), 5269–5278. https://doi.org/10.1016/j.atmosenv.2010.08.008.

Carbone, S., Aurela, M., Saarnio, K., Saarikoski, S., Timonen, H., Frey, A., Hillamo, R.E., 2014. Wintertime aerosol chemistry in sub-Arctic urban air. Aerosol Sci. Technol. 48 (3), 313–323. https://doi.org/10.1080/02786826.2013.875115.

CEN, 2013. PD CEN/TS 16450:2013 - Ambient Air. Automated Measuring Systems for the Measurement of the Concentration of Particulate Matter (PM10; PM2,5). European Committee for Standardization.

Charron, A., Harrison, R.M., Quincey, P., 2007. What are the sources and conditions responsible for exceedences of the 24 h PM 10 limit value (50 μg m −3) at a heavily trafficked London site? Atmos. Environ. 41 (9), 1960–1975. https://doi.org/10.1016/j.atmosenv.2006.10.041.

Cheung, H.C., Morawska, L., Ristovski, Z.D., 2011. Observation of new particle formation in subtropical urban environment. Atmos. Chem. Phys. 11 (8), 3823.

Chowdhury, Z., Zheng, M., Schauer, J.J., Sheesley, R.J., Salmon, L.G., Cass, G.R., Russell, A.G., 2007. Speciation of ambient fine organic carbon particles and source apportionment of PM2.5 in Indian cities. Journal of Geophysical Research: Atmospheres 112 (D15). https://doi.org/10.1029/2007JD008386. n/a-n/a.

Chung, M., Wang, D.D., Rizzo, A.M., Gachette, D., Delnord, M., Parambi, R., Brugge, D., 2015. Association of PNC, BC, and PM2.5 measured at a central monitoring site with blood pressure in a predominantly near highway population. Int. J. Environ. Res. Public Health 12 (3), 2765–2780.

Clifford, S., Low Choy, S., Hussein, T., Mengersen, K., Morawska, L., 2011. Using the generalised additive model to model the particle number count of ultrafine particles. Atmos. Environ. 45 (32), 5934–5945. https://doi.org/10.1016/j.atmosenv.2011.05.004.

Conover, W.J., 1971. Practical Nonparametric Statistics. John Wiley & Sons, New York.

Crilley, L.R., Lucarelli, F., Bloss, W.J., Harrison, R.M., Beddows, D.C., Calzolai, G., Vecchi, R., 2017. Source apportionment of fine and coarse particles at a roadside and urban background site in London during the 2012 summer ClearfLo campaign. Environ. Pollut. 220, 766–778. https://doi.org/10.1016/j.envpol.2016.06.002. *Part B*.

Curci, G., Ferrero, L., Tuccella, P., Barnaba, F., Angelini, F., Bolzacchini, E., Stocchi, P., 2015. How much is particulate matter near the ground influenced by upper-level processes within and above the PBL? A summertime case study in Milan (Italy) evidences the distinctive role of nitrate. Atmos. Chem. Phys. 15 (5), 2629–2649. https://doi.org/10.5194/acp-15-2629-2015.

Cyrys, J., Stölzel, M., Heinrich, J., Kreyling, W.G., Menzel, N., Wittmaack, K., Wichmann, H.E., 2003. Elemental composition and sources of fine and ultrafine ambient particles in Erfurt, Germany. Sci. Total Environ. 305 (1–3), 143–156. https://doi.org/10.1016/S0048-9697(02)00494-1.

Davison, A.C., Hinkley, D.V., 1997. Bootstrap Methods and their Application. Cambridge University Press, Cambridge.

Dos Santos-Juusela, V., Petäjä, T., Kousa, A., Hämeri, K., 2013. Spatial-temporal variations of particle number concentrations between a busy street and the urban background. Atmos. Environ. 79, 324–333. https://doi.org/10.1016/j.atmosenv.2013.05.077.

Eeftens, M., Phuleria, H.C., Meier, R., Aguilera, I., Corradi, E., Davey, M., Tsai, M.Y., 2015. Spatial and temporal variability of ultrafine particles, NO_2, $PM_{2.5}$, $PM_{2.5}$ absorbance, PM_{10} and PM_{coarse} in Swiss study areas. Atmos. Environ. 111, 60–70. https://doi.org/10.1016/j.atmosenv.2015.03.031.

Ergas, C., Clement, M., McGee, J., 2016. Urban density and the metabolic reach of metropolitan areas: a panel analysis of per capita transportation emissions at the county-level. Soc. Sci. Res. 58, 243.

EV-007, 2013. Method 9.13: Determination of suspended particulate matter - $PM_{2.5}$ continuous direct mass method using a tapered element oscilating microbalance monitor. (AS/NZS 3580.9.13:2013). Retrieved from. https://infostore.saiglobal.com/en-au/Standards/AS-NZS-3580-9-13-2013-120034_SAIG_AS_AS_251564/.

Ferrero, L., Castelli, M., Ferrini, B.S., Moscatelli, M., Perrone, M.G., Sangiorgi, G., Cappelletti, D., 2014. Impact of black carbon aerosol over Italian basin valleys: high-resolution measurements along vertical profiles, radiative forcing and heating rate. Atmos. Chem. Phys. 14 (18), 9641–9664. https://doi.org/10.5194/acp-14-9641-2014.

de Foy, B., Schauer, J.J., 2015. Origin of high particle number concentrations reaching the St. Louis, Midwest Supersite. J. Environ. Sci. 34, 219–231. https://doi.org/10.1016/j.jes.2014.12.026.

Franck, U., Odeh, S., Wiedensohler, A., Wehner, B., Herbarth, O., 2011. The effect of particle size on cardiovascular disorders — the smaller the worse. Sci. Total Environ. 409 (20), 4217–4221. https://doi.org/10.1016/j.scitotenv.2011.05.049.

Friend, A.J., Ayoko, G.A., 2009. Multi-criteria ranking and source apportionment of fine particulate matter in Brisbane, Australia. Environ. Chem. 6 (5), 398–406. https://doi.org/10.1071/EN09035.

Friend, A.J., Ayoko, G.A., Jayaratne, E.R., Jamriska, M., Hopke, P.K., Morawska, L., 2012. Source apportionment of ultrafine and fine particle concentrations in Brisbane, Australia. Environ. Sci. Pollut. Res. 19 (7), 2942–2950. https://doi.org/10.1007/s11356-012-0803-6.

Gakidou, E., Afshin, A., Abajobir, A. A., Abate, K. H., Abbafati, C., Abbas, K. M., … Murray, C. J. L. (2017). Global, regional, and national comparative risk assessment of 84 behavioural, environmental and occupational, and metabolic risks or clusters of risks, 19908–2016: a systematic analysis for the Global Burden of Disease Study 2016. Lancet, 390(10100), 1345–1422. doi:https://doi.org/10.1016/S0140-6736(17)32366-8

Genest, C., Rémillard, B., 2004. Test of independence and randomness based on the empirical copula process. Test 13 (2), 335–369. https://doi.org/10.1007/bf02595777.

Gomišček, B., Hauck, H., Stopper, S., Preining, O., 2004. Spatial and temporal variations of PM1, PM2.5, PM10 and particle number concentration during the AUPHEP—project. Atmos. Environ. 38 (24), 3917–3934. https://doi.org/10.1016/j.atmosenv.2004.03.056.

Gu, J., Pitz, M., Schnelle-Kreis, J., Diemer, J., Reller, A., Zimmermann, R., Cyrys, J., 2011. Source apportionment of ambient particles: Comparison of positive matrix factorization analysis applied to particle size distribution and chemical composition data. Atmos. Environ. 45 (10), 1849–1857. https://doi.org/10.1016/j.atmosenv.2011.01.009.

Gu, J., Schnelle-Kreis, J., Pitz, M., Diemer, J., Reller, A., Zimmermann, R., Cyrys, J., 2013.

Spatial and temporal variability of PM10 sources in Augsburg, Germany. Atmos. Environ. 71, 131–139. https://doi.org/10.1016/j.atmosenv.2013.01.043.
Gulia, S., Shiva Nagendra, S.M., Khare, M., Khanna, I., 2015. Urban air quality management-a review. Atmospheric Pollution Research 6 (2), 286–304. https://doi.org/10.5094/APR.2015.033.
Gurjar, B.R., Ravindra, K., Nagpure, A.S., 2016. Air pollution trends over Indian megacities and their local-to-global implications. Atmos. Environ. 142 (C), 475–495. https://doi.org/10.1016/j.atmosenv.2016.06.030.
Halek, F., Kianpour-Rad, M., Kavousirahim, A., 2010. Seasonal variation in ambient PM mass and number concentrations (case study: Tehran, Iran). Environ. Monit. Assess. 169 (1), 501–507. https://doi.org/10.1007/s10661-009-1192-2.
Hamed, A., Joutsensaari, J., Mikkonen, S., Sogacheva, L., Maso, M. D., Kulmala, M., … Laaksonen, A. (2007). Nucleation and growth of new particles in Po Valley, Italy. Atmos. Chem. Phys., 7(2), 355–376. doi:https://doi.org/10.5194/acp-7-355-2007
Hämeri, K., Kulmala, M., Aalto, P., Leszczynski, K., Visuri, R., Hämekoski, K., 1996. The investigations of aerosol particle formation in urban background area of Helsinki. Atmos. Res. 41 (3), 281–298. https://doi.org/10.1016/0169-8095(96)00015-4.
Han, Y.-J., Kim, T.-S., Kim, H., 2008. Ionic constituents and source analysis of PM2.5 in three Korean cities. Atmos. Environ. 42 (19), 4735–4746. https://doi.org/10.1016/j.atmosenv.2008.01.047.
Han, B., Zhang, G., Bi, X., Wang, X., Sheng, G., Fu, J., 2015. Pollution characteristics of atmospheric particle size and number concentrations during summer in urban area of Guangzhou. Res. Environ. Sci. 28 (2), 198–204. https://doi.org/10.13198/j.issn.1001-6929.2015.02.05.
Han, L., Zhou, W., Pickett, S.T.A., Li, W., Li, L., 2016. An optimum city size? The scaling relationship for urban population and fine particulate (PM2.5) concentration. Environ. Pollut. 208, 96–101. https://doi.org/10.1016/j.envpol.2015.08.039. Pt A.
Harrell Jr., F.E., 2018. and with contributions from Charles Dupont and many others. In: Hmisc: Harrell Miscellaneous *R package version 4.1-1*.
Harrison, R. M., Giorio, C., Beddows, D. C. S., Dall, Amp, Apos, & Osto, M. (2010). Size distribution of airborne particles controls outcome of epidemiological studies. Sci. Total Environ., 409(2), 289–293. doi:https://doi.org/10.1016/j.scitotenv.2010.09.043
Hasheminassab, S., Daher, N., Ostro, B.D., Sioutas, C., 2014a. Long-term source apportionment of ambient fine particulate matter (PM2.5) in the Los Angeles Basin: A focus on emissions reduction from vehicular sources. Environ. Pollut. 193, 54–64. https://doi.org/10.1016/j.envpol.2014.06.012.
Hasheminassab, S., Daher, N., Saffari, A., Wang, D., Ostro, B.D., Sioutas, C., 2014b. Spatial and temporal variability of sources of ambient fine particulate matter (PM2.5) in California. Atmos. Chem. Phys. 14 (22), 12085. https://doi.org/10.5194/acp-14-12085-2014.
Hofert, M., Kojadinovic, I., Maechler, M., Yan, J., 2017. copula: Multivariate Dependence with Copulas *R package version 0*. 999-18.
Hofman, J., Staelens, J., Cordell, R., Stroobants, C., Zikova, N., Hama, S.M.L., Roekens, E., 2016. Ultrafine particles in four European urban environments: Results from a new continuous long-term monitoring network. Atmos. Environ. 136, 68–81. https://doi.org/10.1016/j.atmosenv.2016.04.010.
Hudda, N., Gould, T., Hartin, K., Larson, T.V., Fruin, S.A., 2014. Emissions from an international airport increase particle number concentrations 4-fold at 10 km downwind. Environmental Science & Technology 48 (12), 6628–6635. https://doi.org/10.1021/es5001566.
Jeong, C.-H., Evans, G., 2009. Inter-comparison of a fast mobility particle sizer and a scanning mobility particle sizer incorporating an ultrafine water-based condensation particle counter. Aerosol Sci. Technol. 43 (4), 364–373. https://doi.org/10.1080/02786820802662939.
Jeong, C.-H., Evans, G.J., Hopke, P.K., Chalupa, D., Utell, M.J., 2006. Influence of atmospheric dispersion and new particle formation events on ambient particle number concentration in Rochester, United States, and Toronto, Canada. Journal of the Air & Waste Management Association (Air & Waste Management Association) 56 (4), 431–443.
Jeong, C. H., McGuire, M. L., Herod, D., Dann, T., Dabek–Zlotorzynska, E., Wang, D., … Evans, G. (2011). Receptor model based identification of PM2.5 sources in Canadian cities. Atmospheric Pollution Research, 2(2), 158–171. doi:https://doi.org/10.5094/APR.2011.021
Johansson, C., Norman, M., Gidhagen, L., 2007. Spatial & temporal variations of PM10 and particle number concentrations in urban air. Environ. Monit. Assess. 127 (1), 477–487. https://doi.org/10.1007/s10661-006-9296-4.
Johnson, J.P., Kittelson, D.B., Watts, W.F., 2009. The effect of federal fuel sulfur regulations on in-use fleets: on-road heavy-duty source apportionment. Environmental Science & Technology 43 (14), 5358. https://doi.org/10.1021/es8037164.
Jones, A.M., Harrison, R.M., Barratt, B., Fuller, G., 2012. A large reduction in airborne particle number concentrations at the time of the introduction of “sulphur free” diesel and the London Low Emission Zone. Atmos. Environ. 50, 129–138. https://doi.org/10.1016/j.atmosenv.2011.12.050.
Karagulian, F., Belis, C.A., Dora, C.F.C., Prüss-Ustün, A.M., Bonjour, S., Adair-Rohani, H., Amann, M., 2015. Contributions to cities' ambient particulate matter (PM): a systematic review of local source contributions at global level. Atmos. Environ. 120, 475–483. https://doi.org/10.1016/j.atmosenv.2015.08.087.
Karathodorou, N., Graham, D.J., Noland, R.B., 2010. Estimating the effect of urban density on fuel demand. Energy Econ. 32 (1), 86–92. https://doi.org/10.1016/j.eneco.2009.05.005.
Kaur, S., Nieuwenhuijsen, M., Colvile, R., 2005. Personal exposure of street canyon intersection users to PM2.5, ultrafine particle counts and carbon monoxide in Central London, UK. Atmos. Environ. 39 (20), 3629–3641. https://doi.org/10.1016/j.atmosenv.2005.02.046.
Keuken, M.P., Henzing, J.S., Zandveld, P., van Den Elshout, S., Karl, M., 2012. Dispersion of particle numbers and elemental carbon from road traffic, a harbour and an airstrip in the Netherlands. Atmos. Environ. 54 (C), 320–327. https://doi.org/10.1016/j.atmosenv.2012.01.012.
Keywood, M.D., Ayers, G.P., Gras, J.L., Gillett, R.W., Cohen, D.D., 1999. Relationships between size segregated mass concentration data and ultrafine particle number concentrations in urban areas. Atmos. Environ. 33 (18), 2907–2913. https://doi.org/10.1016/S1352-2310(99)00102-8.
Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.M., McMurry, P.H., 2004. Formation and growth rates of ultrafine atmospheric particles: a review of observations. J. Aerosol Sci. 35 (2), 143–176. https://doi.org/10.1016/j.jaerosci.2003.10.003.
Kumar, P., Morawska, L., Birmili, W., Paasonen, P., Hu, M., Kulmala, M., Britter, R., 2014. Ultrafine particles in cities. Environ. Int. 66, 1–10. https://doi.org/10.1016/j.envint.2014.01.013.
Lane, K.J., Levy, J.I., Scammell, M.K., Peters, J.L., Patton, A.P., Reisner, E., Brugge, D., 2016. Association of modeled long-term personal exposure to ultrafine particles with inflammatory and coagulation biomarkers. Environ. Int. 92-93, 173–182. https://doi.org/10.1016/j.envint.2016.03.013.
Lanzinger, S., Schneider, A., Breitner, S., Stafoggia, M., Erzen, I., Dostal, M., Peters, A., 2016. Ultrafine and fine particles and hospital admissions in Central Europe: results from the UFIREG study. Am. J. Respir. Crit. Care Med. 194 (10), 1233–1241. https://doi.org/10.1164/rccm.201510-2042OC.
Larkin, A., van Donkelaar, A., Geddes, J.A., Martin, R.V., Hystad, P., 2016. Relationships between changes in urban characteristics and air quality in East Asia from 2000 to 2010. Environmental Science & Technology 50 (17), 9142. https://doi.org/10.1021/acs.est.6b02549.
Lee, P.K.H., Brook, J.R., Dabek-Zlotorzynska, E., Mabury, S.A., 2003. Identification of the major sources contributing to PM2.5 observed in Toronto. Environmental Science & Technology 37 (21), 4831–4840. https://doi.org/10.1021/es026473i.
Li, B., Zhang, J., Zhao, Y., Yuan, S., Zhao, Q., Shen, G., Wu, H., 2015. Seasonal variation of urban carbonaceous aerosols in a typical city Nanjing in Yangtze River Delta, China. Atmos. Environ. 106 (C), 223–231. https://doi.org/10.1016/j.atmosenv.2015.01.064.
Lin, B., Zhu, J., 2018. Changes in urban air quality during urbanization in China. J. Clean. Prod. 188, 312–321. https://doi.org/10.1016/j.jclepro.2018.03.293.
Liu, Z., Gao, W., Yu, Y., Hu, B., Xin, J., Sun, Y., … Wang, Y. (2018). Characteristics of PM2.5 mass concentrations and chemical species in urban and background areas of China: emerging results from the CARE-China network. Atmos. Chem. Phys., 18, 8849 - 8871.
Lonati, G., Giugliano, M., Butelli, P., Romele, L., Tardivo, R., 2005. Major chemical components of PM2.5 in Milan (Italy). Atmos. Environ. 39 (10), 1925–1934. https://doi.org/10.1016/j.atmosenv.2004.12.012.
Marconi, A., Cattani, G., Cusano, M., Ferdinandi, M., Inglessis, M., Viviano, G., Forastiere, F., 2007. Two-years of fine and ultrafine particles measurements in Rome, Italy. J. Toxic. Environ. Health A 70 (3–4), 213–221. https://doi.org/10.1080/15287390600883174.
Mayer, H., 1999. Air pollution in cities. Atmos. Environ. 33 (24), 4029–4037. https://doi.org/10.1016/S1352-2310(99)00144-2.
Mazaheri, M., Johnson, G.R., Morawska, L., 2011. An inventory of particle and gaseous emissions from large aircraft thrust engine operations at an airport. Atmos. Environ. 45 (20), 3500–3507. https://doi.org/10.1016/j.atmosenv.2010.12.012.
Mejía, J.F., Wraith, D., Mengersen, K., Morawska, L., 2007. Trends in size classified particle number concentration in subtropical Brisbane, Australia, based on a 5 year study. Atmos. Environ. 41 (5), 1064–1079. https://doi.org/10.1016/j.atmosenv.2006.09.020.
Merico, E., Donateo, A., Gambaro, A., Cesari, D., Gregoris, E., Barbaro, E., Contini, D., 2016. Influence of in-port ships emissions to gaseous atmospheric pollutants and to particulate matter of different sizes in a Mediterranean harbour in Italy. Atmos. Environ. 139, 1–10. https://doi.org/10.1016/j.atmosenv.2016.05.024.
Morawska, L., Thomas, S., Jamriska, M., Johnson, G., 1999. The modality of particle size distributions of environmental aerosols. Atmos. Environ. 33 (27), 4401–4411. https://doi.org/10.1016/S1352-2310(99)00217-4.
Morawska, L., Ristovski, Z., Jayaratne, E.R., Keogh, D.U., Ling, X., 2008. Ambient nano and ultrafine particles from motor vehicle emissions: characteristics, ambient processing and implications on human exposure. Atmos. Environ. 42 (35), 8113–8138. https://doi.org/10.1016/j.atmosenv.2008.07.050.
Newman, P.W.G., Kenworthy, J.R., 1989. Gasoline consumption and cities. *American Planning Association*. J. Am. Plan. Assoc. 55 (1), 24.
NOAA, 2013. State of the Climate: Global Climate Report for Annual 2012. Retrieved from. https://www.ncdc.noaa.gov/sotc/global/201213.
NOAA, 2014. State of the Climate: Global Climate Report for Annual 2013. Retrieved from. https://www.ncdc.noaa.gov/sotc/global/201313.
NOAA, 2015. State of the Climate: Global Climate Report for Annual 2014. Retrieved from. https://www.ncdc.noaa.gov/sotc/global/201413.
NOAA, 2016. State of the Climate: Global Climate Report for Annual 2015. Retrieved from. https://www.ncdc.noaa.gov/sotc/global/201513.
Patton, A.P., Perkins, J., Zamore, W., Levy, J.I., Brugge, D., Durant, J.L., 2014. Spatial and temporal differences in traffic-related air pollution in three urban neighborhoods near an interstate highway. Atmos. Environ. 99, 309–321. https://doi.org/10.1016/j.atmosenv.2014.09.072.
Pérez, N., Pey, J., Reche, C., Cortés, J., Alastuey, A., Querol, X., 2016. Impact of harbour emissions on ambient PM10 and PM2.5 in Barcelona (Spain): Evidences of secondary aerosol formation within the urban area. Sci. Total Environ. 571, 237–250. https://doi.org/10.1016/j.scitotenv.2016.07.025.
Perrone, M.G., Larsen, B.R., Ferrero, L., Sangiorgi, G., De Gennaro, G., Udisti, R., Bolzacchini, E., 2012. Sources of high PM2.5 concentrations in Milan, Northern Italy:

Molecular marker data and CMB modelling. Sci. Total Environ. 414, 343–355. https://doi.org/10.1016/j.scitotenv.2011.11.026.
Pey, J., Rodríguez, S., Querol, X., Alastuey, A., Moreno, T., Putaud, J.P., Van Dingenen, R., 2008. Variations of urban aerosols in the western Mediterranean. Atmos. Environ. 42 (40), 9052–9062. https://doi.org/10.1016/j.atmosenv.2008.09.049.
Pilz, V., Wolf, K., Breitner, S., Rückerl, R., Koenig, W., Rathmann, W., Schneider, A., 2018. C-reactive protein (CRP) and long-term air pollution with a focus on ultrafine particles. Int. J. Hyg. Environ. Health 221 (3), 510–518. https://doi.org/10.1016/j.ijheh.2018.01.016.
Putaud, J.P., Van Dingenen, R., Alastuey, A., Bauer, H., Birmili, W., Cyrys, J., Raes, F., 2010. A European aerosol phenomenology – 3: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe. Atmos. Environ. 44 (10), 1308–1320. https://doi.org/10.1016/j.atmosenv.2009.12.011.
Puustinen, A., Hämeri, K., Pekkanen, J., Kulmala, M., de Hartog, J., Meliefste, K., Hoek, G., 2007. Spatial variation of particle number and mass over four European cities. Atmos. Environ. 41 (31), 6622–6636. https://doi.org/10.1016/j.atmosenv.2007.04.020.
Qiao, T., Zhao, M., Xiu, G., Yu, J., 2016. Simultaneous monitoring and compositions analysis of PM1 and PM2.5 in Shanghai: Implications for characterization of haze pollution and source apportionment. Sci. Total Environ. 557, 386–394. https://doi.org/10.1016/j.scitotenv.2016.03.095.
Rodrıguez, S., Van Dingenen, R., Putaud, J.P., Dell'Acqua, A., Pey, J., Querol, X., Gemelli, V., 2007. A study on the relationship between mass concentrations, chemistry and number size distribution of urban fine aerosols in Milan, Barcelona and London. Atmos. Chem. Phys. 7 (9), 2217–2232.
Rönkkö, T., Virtanen, A., Vaaraslahti, K., Keskinen, J., Pirjola, L., Lappi, M., 2006. Effect of dilution conditions and driving parameters on nucleation mode particles in diesel exhaust: Laboratory and on-road study. Atmos. Environ. 40 (16), 2893–2901. https://doi.org/10.1016/j.atmosenv.2006.01.002.
Rönkkö, T., Kuuluvainen, H., Karjalainen, P., Keskinen, J., Hillamo, R., Niemi, J.V., Dal Maso, M., 2017. Traffic is a major source of atmospheric nanocluster aerosol. Proc. Natl. Acad. Sci. 114 (29), 7549.
RStudio Team, 2016. RStudio: Integrated Development for R. RStudio, Inc., Boston, MA Retrieved from. http://www.rstudio.com.
Saarikoski, S., Timonen, H., Saarnio, K., Aurela, M., Järvi, L., Keronen, P., Hillamo, R., 2008. Sources of organic carbon in fine particulate matter in northern European urban air. Atmos. Chem. Phys. 8 (20), 6281–6295. https://doi.org/10.5194/acp-8-6281-2008.
Salimi, F., Md Mahmudur, R., Clifford, S., Ristovski, Z., Morawska, L., 2017. Nocturnal new particle formation events in urban environments. Atmos. Chem. Phys. 17 (1), 521–530. https://doi.org/10.5194/acp-17-521-2017.
Schäfer, K., Elsasser, M., Arteaga-Salas, J. M., Gu, J., Pitz, M., Schnelle-Kreis, J., … Zimmermann, R. (2014). Source apportionment and the role of meteorological conditions in the assessment of air pollution exposure due to urban emissions. Atmos. Chem. Phys. Discuss., 2014, 2235–2275. doi:https://doi.org/10.5194/acpd-14-2235-2014
Seigneur, C., 2009. Current understanding of ultrafine particulate matter emitted from mobile sources. J. Air Waste Manage. Assoc. 59 (1), 3–17. https://doi.org/10.3155/1047-3289.59.1.3.
Sekhon, J.S., 2011. Multivariate and propensity score matching software with automated balance optimization: the matching package for R. J. Stat. Softw. 42 (7), 1–52.
Shirmohammadi, F., Sowlat, M.H., Hasheminassab, S., Saffari, A., Ban-Weiss, G., Sioutas, C., 2017. Emission rates of particle number, mass and black carbon by the Los Angeles international airport (LAX) and its impact on air quality in Los Angeles. Atmos. Environ. 151, 82–93. https://doi.org/10.1016/j.atmosenv.2016.12.005.
Sowlat, M.H., Hasheminassab, S., Sioutas, C., 2016. Source apportionment of ambient particle number concentrations in central Los Angeles using positive matrix factorization (PMF). Atmos. Chem. Phys. 16 (8), 4849–4866. https://doi.org/10.5194/acp-16-4849-2016.
Squizzato, S., Cazzaro, M., Innocente, E., Visin, F., Hopke, P.K., Rampazzo, G., 2017. Urban air quality in a mid-size city — PM2.5 composition, sources and identification of impact areas: from local to long range contributions. Atmos. Res. 186, 51–62. https://doi.org/10.1016/j.atmosres.2016.11.011.
Stanier, C.O., Khlystov, A.Y., Pandis, S.N., 2004. Ambient aerosol size distributions and number concentrations measured during the Pittsburgh air quality study (PAQS). Atmos. Environ. 38 (20), 3275–3284. https://doi.org/10.1016/j.atmosenv.2004.03.020.
Stettler, M.E.J., Eastham, S., Barrett, S.R.H., 2011. Air quality and public health impacts of UK airports. Part I: emissions. Atmos. Environ. 45 (31), 5415–5424. https://doi.org/10.1016/j.atmosenv.2011.07.012.
Strak, M., Janssen, N.A.H., Godri, K.J., Gosens, I., Mudway, I.S., Cassee, F.R., Hoek, G., 2012. Respiratory health effects of airborne particulate matter: the role of particle size, composition, and oxidative potential–the RAPTES project. Environ. Health Perspect. 120, 1183.
Suh, H.H., Nishioka, Y., Allen, G.A., Koutrakis, P., Burton, R.M., 1997. The metropolitan acid aerosol characterization study: Results from the summer 1994 Washington, D.C. field study. Environ. Health Perspect. 105 (8), 826–834. https://doi.org/10.2307/3433700.
Tao, M., Chen, L., Li, R., Wang, L., Wang, J., Wang, Z., … Tao, J. (2016). Spatial oscillation of the particle pollution in eastern China during winter: Implications for regional air quality and climate. Atmos. Environ., 144(C), 100–110. doi:https://doi.org/10.1016/j.atmosenv.2016.08.049
Thorpe, A., Harrison, R.M., 2008. Sources and properties of non-exhaust particulate matter from road traffic: A review. Sci. Total Environ. 400 (1–3), 270–282. https://doi.org/10.1016/j.scitotenv.2008.06.007.
Tobías, A., Rivas, I., Reche, C., Alastuey, A., Rodríguez, S., Fernández-Camacho, R., Querol, X., 2018. Short-term effects of ultrafine particles on daily mortality by primary vehicle exhaust versus secondary origin in three Spanish cities. Environ. Int. 111, 144–151. https://doi.org/10.1016/j.envint.2017.11.015.
Tsai, J., Owega, S., Evans, G., Jervis, R., Fila, M., Tan, P., Malpica, O., 2004. Chemical composition and source apportionment of Toronto summertime urban fine aerosol (PM2.5). J. Radioanal. Nucl. Chem. 259 (1), 193–197. https://doi.org/10.1023/B:JRNC.0000015828.93105.ac.
Tuch, T.M., Brand, P., Wichmann, H.E., Heyder, J., 1997. Variation of particle number and mass concentration in various size ranges of ambient aerosols in Eastern Germany. Atmos. Environ. 31 (24), 4193–4197. https://doi.org/10.1016/S1352-2310(97)00260-4.
UN-DESA, 2016. Demographic Yearbook. Retrieved from. https://unstats.un.org/unsd/demographic-social/products/dyb/dybsets/2016.pdf.
USEPA, 2006. Code of Federal Regulations. Retrieved from. https://ecfr.io/Title-40/pt40.6.58#ap40.6.58_161.e.
Van Dingenen, R., Raes, F., Putaud, J.-P., Baltensperger, U., Charron, A., Facchini, M.C., Wåhlin, P., 2004. A European aerosol phenomenology—1: physical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe. Atmos. Environ. 38 (16), 2561–2577. https://doi.org/10.1016/j.atmosenv.2004.01.040.
Vassilakos, C., Saraga, D., Maggos, T., Michopoulos, J., Pateraki, S., Helmis, C.G., 2005. Temporal variations of PM2.5 in the ambient air of a suburban site in Athens, Greece. Sci. Total Environ. 349 (1–3), 223–231. https://doi.org/10.1016/j.scitotenv.2005.01.012.
Viana, M., Hammingh, P., Colette, A., Querol, X., Degraeuwe, B., Vlieger, I.d., van Aardenne, J., 2014. Impact of maritime transport emissions on coastal air quality in Europe. Atmos. Environ. 90, 96–105. https://doi.org/10.1016/j.atmosenv.2014.03.046.
Viehmann, A., Hertel, S., Fuks, K., Eisele, L., Moebus, S., Möhlenkamp, S., Hoffmann, B., 2015. Long-term residential exposure to urban air pollution, and repeated measures of systemic blood markers of inflammation and coagulation. Occup. Environ. Med. 72 (9), 656–663. https://doi.org/10.1136/oemed-2014-102800.
Wåhlin, P., 2009. Measured reduction of kerbside ultrafine particle number concentrations in Copenhagen. Atmos. Environ. 43 (22), 3645–3647. https://doi.org/10.1016/j.atmosenv.2009.04.023.
Wand, M.P., Jones, M.C., 1995. Kernel Smoothing, 1st ed. Chapman & Hall, London.
Wang, Gao, H.O., 2011. Exposure to fine particle mass and number concentrations in urban transportation environments of New York City. Transp. Res. Part D: Transp. Environ. 16 (5), 384–391. https://doi.org/10.1016/j.trd.2011.03.001.
Wang, F., Ketzel, M., Ellermann, T., Wahlin, P., Jensen, S.S., Fang, D., Massling, A., 2010. Particle number, particle mass and NOx emission factors at a highway and an urban street in Copenhagen. Atmos. Chem. Phys. 10, 2745–2764.
Wang, Y., Hopke, P.K., Chalupa, D.C., Utell, M.J., 2011. Long-term study of urban ultrafine particles and other pollutants. Atmos. Environ. 45 (40), 7672–7680. https://doi.org/10.1016/j.atmosenv.2010.08.022.
Wang, Y., Li, L., Chen, C., Huang, C., Huang, H., Feng, J., Wumaer, A., 2014. Source apportionment of fine particulate matter during autumn haze episodes in Shanghai, China. Journal of Geophysical Research: Atmospheres 119 (4), 1903–1914. https://doi.org/10.1002/2013JD019630.
Wang, L., Wei, Z., Wei, W., Fu, J.S., Meng, C., Ma, S., 2015. Source apportionment of PM2.5 in top polluted cities in Hebei, China using the CMAQ model. Atmos. Environ. 122, 723–736. https://doi.org/10.1016/j.atmosenv.2015.10.041.
Wang, S., Zhou, C., Wang, Z., Feng, K., Hubacek, K., 2017. The characteristics and drivers of fine particulate matter (PM2.5) distribution in China. J. Clean. Prod. 142 (P4), 1800–1809. https://doi.org/10.1016/j.jclepro.2016.11.104.
WHO, 2006. *WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide: Global update 2005* (WHO/SDE/PHE/OEH/06.02). Retrieved from. http://apps.who.int/iris/bitstream/handle/10665/69477/WHO_SDE_PHE_OEH_06.02_eng.pdf?sequence=1.
WHO, n.d., September 2016WHO, n.d. Ambient (Outdoor) Air Quality and Health. Retrieved from. http://www.who.int/mediacentre/factsheets/fs313/en/.
Wickham, H., 2009. ggplot2: Elegant Graphics for Data Analysis. Springer-Verlag, New York.
Wolf, K., Cyrys, J., Harciníková, T., Gu, J., Kusch, T., Hampel, R., Peters, A., 2017. Land use regression modeling of ultrafine particles, ozone, nitrogen oxides and markers of particulate matter pollution in Augsburg, Germany. Sci. Total Environ. 579, 1531–1540. https://doi.org/10.1016/j.scitotenv.2016.11.160.
Yang, H., Yu, J.Z., Ho, S.S.H., Xu, J., Wu, W.-S., Wan, C.H., Wang, L., 2005. The chemical composition of inorganic and carbonaceous materials in PM2.5 in Nanjing, China. Atmos. Environ. 39 (20), 3735–3749. https://doi.org/10.1016/j.atmosenv.2005.03.010.
Zieffler, A.S., Harring, J.R., Long, J.D., 2011. Randomization and permutation tests. In: Comparing Groups. John Wiley & Sons, Inc., pp. 117–137.