

A review of intraurban variations in particulate air pollution: Implications for epidemiological research

J. Gaines Wilson^{a,b,*}, Simon Kingham^a, Jamie Pearce^a, Andrew P. Sturman^{a,b}

^a*GeoHealth Laboratory, University of Canterbury, Private Bag 4800, Christchurch, New Zealand*

^b*Centre for Atmospheric Research, University of Canterbury, Christchurch, New Zealand*

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Abstract

Epidemiological studies typically utilise one or few central monitoring stations as a proxy for personal exposure to particulate matter air pollution. However, recent research indicates that central monitoring sites may not accurately characterize the spatial complexities of the particulate field across an urban area. Consequently, intraurban assessment of exposure to air pollution has become a priority area of study. This paper reviews particulate air pollution exposure studies with a focus on monitored-data studies at the intraurban spatial scale. Portions of the literature provide contradictory conclusions as to the homogeneity of intraurban particulate concentrations, due in part to local conditions such as source composition, meteorology, locations of monitoring sites and topography, but which may also be a result of the methods and definitions used to quantify relative and absolute spatial concentration variations. Comparative analyses of the literature by particle size fraction, method of determining heterogeneity, and sampling characteristics were performed. We find that particular attention should be given to local conditions and methods when using one or few monitoring sites to characterise wider population exposures. The utilisation of absolute and relative measures of homogeneity such as the coefficient of divergence which are based on data from several monitoring sites (e.g., $n > 4$) in combination with an appropriate sample size (e.g., $n \geq 50$) may reduce the possibility of misclassification based on incorrect assumptions about heterogeneity. The errors in exposure misclassification based on these assumptions about intraurban concentration variations are especially critical in long-term cohort epidemiological analyses that assess the effects of exposure variations in air pollution upon health.

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1. Introduction

Pronounced air pollution episodes in Europe and the United States during the early to middle twentieth

century brought wide attention to the study of related health effects, including mortality and morbidity (Firket, 1936; Ministry of Health, 1954; Schrenk et al., 1949). By the late 1970s, due mainly to legislative changes, concentrations of air pollutants in developed countries had been reduced greatly and were no longer considered by many to be of a magnitude large enough to cause health concerns. However, more sophisticated and sensitive monitoring techniques and statistical methods applied from the early 1990s established that

*Corresponding author. GeoHealth Laboratory, University of Canterbury, Private Bag 4800, Christchurch, New Zealand. Fax: + 64 3 364 2907.

E-mail address: j.wilson@geog.canterbury.ac.nz (J.G. Wilson).

health effects were present even at levels below ambient guidelines for pollution, especially particulate exposure (Brunekreef et al., 1995; Dockery and Pope, 1994; Mage et al., 1999; Pope et al., 1992). In the last few years, the relationship between particulates and adverse health effects, including mortality, have been established at lesser and lesser levels of exposure (Bell et al., 2004; Vedal et al., 2003). The World Health Organisation has concluded that there is no zero-effect threshold for particulates and that health risks are present at *any level* of exposure (World Health Organisation, 1999).

The linkages and associations between air pollution and health are determined largely through epidemiological studies. The dominant approach for estimating exposure to air pollution in these studies is the application of an exposure value or exposure variation at a central site to the entire population of the study area (e.g., Dockery et al., 1993; Pope et al., 2002; Pope et al., 1992; Samet et al., 2000; Wong et al., 2001; Zanobetti et al., 2003). A common assumption is that the spatial distributions of certain pollutants, especially smaller particulates, are homogeneously distributed within large urban areas and that the concentrations between sites are well correlated. Early studies in the United States supported that assumption, finding homogeneous, well-correlated distributions of particulates across intraurban (*within-city*) areas (Burton et al., 1996; Suh et al., 1997; Wilson and Suh, 1997). However, several recent studies suggest that there may be greater variation within urban areas than previously reported and that the ecological method of particulate exposure assessment may misclassify personal exposures to a larger extent than previously thought (Briggs, 2000; Ito et al., 2004; Kim et al., 2005; Pinto et al., 2004; Zhu et al., 2002). Further, exposure misclassification due to the selection of monitoring sites and sampling frequencies may notably alter the significance, direction and magnitude of health outcomes in epidemiological studies (Ito et al., 2005, 1995). Despite the development of Geographic Information Systems (GIS), dispersion models, advanced spatial statistical techniques, and integrated hybrid models, many epidemiological studies continue to operate under the spatial homogeneous distribution assumption, especially for particulate air pollution (Huang and Batterman, 2000; Jerrett et al., 2005a). The study of air pollution exposures at the intraurban scale presents a challenge in a new era of exposure assessment in epidemiological research, and has been recently identified as a priority area for future work (Brunekreef and Holgate, 2002; Jerrett et al., 2005a; Kukkonen et al., 2001; Sajani et al., 2004).

A review specifically focused on particulate air pollutants is of significant interest as particulates are one of the most widely studied air pollutants linked with health effects in the literature; Both in terms of morbidity and mortality effects (Dockery and Pope,

1994; Moolgavkar and Luebeck, 1996; Pope et al., 1995; Pope et al., 1991; Pope et al., 1992; Samet et al., 2000). A pubmed search (Mar 2005) containing the keywords “health effects” and “health” with combinations of “particulate air pollution”; “pm10” and “pm2.5” yielded approximately 1200 journal articles written in english on the subject since 1991; When Pope et al. (1991) published their seminal time-series analysis on particulates and health (time-series have been performed since the late 1950s). There is a risk that the homogeneous particulate concentration distribution assumption; upon which many subsequent epidemiological studies based their exposure estimates; May significantly misclassify personal exposures; Potentially leading to errors in health risk estimates; Particularly for multi-community and longer-term study designs (Brauer et al., 2002; Dominici et al., 2003; Pinto et al., 2004; Zeger et al., 2000).

The aim of this paper is to examine the particulate air pollution literature with a specific focus on the spatial nature of outdoor monitored particulate exposures at the intraurban scale. We will meet this aim by outlining the literature, and then by comparing elements of the reviewed papers. First, we provide an overview of particulate air pollution and the physical characteristics of PM that contribute to spatial variation. Next, we discuss the concept of measurement error and how heterogeneous concentrations at the intraurban level impact on risk estimates for different types of epidemiological study designs. Third, the intraurban particulate monitoring literature is reviewed in detail by homogeneity conclusions, particle size fractions, methods of characterising homogeneity, and both site and sampling characteristics. Finally, we compare methods, site considerations, and results between studies conducted on the same pollutant particle size fractions in identical urban areas. Based on a comparative review, we comment on the suitability of central monitoring sites in estimating intraurban study area exposures and discuss considerations for reducing the possibility of exposure misclassification due to incorrect assumptions about uniformity.

2. Particulate matter air pollution

Particulate matter (PM) refers to a suspension of solid, liquid or a combination of solid and liquid particles in the air (Hinds, 1999). PM is one of six ‘criteria pollutants’ designated by the US Clean Air Act of 1971, which are measured and reviewed in the development and adjustment of environmental and health standards. Sources of PM originate from both anthropogenic and natural sources, and may be classified as primary or secondary pollutants. Primary particulates, which may be coarse or fine, are formed

directly and are most commonly associated with combustion sources including traffic, industry and domestic heating. Secondary PM are finer and formed in the atmosphere through chemical and physical conversions of gaseous precursors such as nitrogen oxides, sulphur oxides, and volatile organic compounds. Primary particles generally affect local scales, whereas secondary particles affect regional and wider-ranging areas (Blanchard et al., 1999; Ito et al., 2004).

In health and exposure studies, particles are often classified by size fraction, or aerodynamic diameter (d_a), which can range from a few nanometers up to 100 micrometers (μm). Size fraction is a term that aids in the classification of particles and refers more to the physical behaviour of particles rather than their actual size. The more widely used size fractions for PM are the coarse fractions PM_{10} ($d_a \leq 10 \mu\text{m}$) and $\text{PM}_{10-2.5}$ ($2.5 \mu\text{m} \leq d_a \leq 10 \mu\text{m}$). Smaller fractions are $\text{PM}_{2.5}$ ($d_a < 2.5 \mu\text{m}$), PM_1 ($d_a < 1 \mu\text{m}$), also called fine particles, and UFP, or ultrafine particles ($d_a < 0.1 \mu\text{m}$). Total suspended particulate (TSP), or TSP, is used less frequently today, and represents particles $d_a < 100 \mu\text{m}$. Physical size fraction greatly influences the region of deposition of inhaled particles in the respiratory tract (Yeh et al., 1976). PM_{10} , also called ‘thoracic’ particles, can penetrate into the lower respiratory system, while $\text{PM}_{2.5}$, or ‘respirable’ particles, can enter deeper into the gas-exchange portions of the lung. Associations between PM and adverse health effects are expected to be stronger than for coarse particles ($> 2.5 \mu\text{m}$) (Schwartz et al., 1996). PM_{10} and $\text{PM}_{2.5}$ are the most common size fractions considered in epidemiologic studies, although more recent research has been conducted on fine and ultrafine size fractions (Schulz et al., 2005). Particles greater than $10 \mu\text{m}$ in diameter have limited stability in the air, and are seldom considered in exposure studies. For exposure and health studies, PM is most commonly measured according to the mass concentration of the specific particle size range. The most common unit is $\mu\text{g}/\text{m}^3$, which represents the density of aerosol particles in the air and is usually averaged over a defined period of time (Hinds, 1999). Particles may also be measured by number concentration, or the number of specific particles per unit volume, usually expressed as number/ cm^3 . It is unclear whether it is the mass load, or some other physical factor (particle number has been suggested), or the chemical or biological composition that is the causative factor for adverse health effects (Monn, 2001). Number concentration may indeed be more important than mass concentration in terms of health effects, especially for UFP, although little work has been done in this emerging area of interest to date (Englert, 2004; Peters et al., 1997).

Ambient concentrations of particles vary spatially and temporally to differing degrees at the intraurban scale depending on size fraction, largely because the terminal

settling time for particles increases rapidly with particle size. For particles with a diameter larger than $1.0 \mu\text{m}$, terminal settling velocity is proportional to the square of the diameter of the particle (Hinds, 1999), or in other words, the smaller the particle, the more homogeneous its distribution. Stopping distance, or the distance a particle travels before settling when ejected from a source, is one property that provides an explanation as to why larger particle classes travel shorter distances, creating more spatial heterogeneity in an intraurban setting. For a starting velocity of 1000 cm/s , UFP have a stopping distance of $8.8 \times 10^{-5} \text{ cm}$, for PM_1 $3.5 \times 10^{-3} \text{ cm}$, and for PM_{10} 0.23 cm (Willeke and Baron, 1993). Stopping distances and settling times are based on the assumption that meteorological conditions are favourable. A general assumption based on these stopping velocities, settling times, and field evidence is that spatial distributions for finer particles ($\text{PM}_{2.5}$ and PM_1) are relatively more uniformly distributed than for coarser fractions (Brimblecombe, 1986; Monn, 2001; Wilson and Suh, 1997). However, at sizes smaller than fine particles, i.e., UFP, number counts have shown to vary significantly over small areas in the limited work done to date (Harrison et al., 1999; Junker et al., 2000; Noble et al., 2003).

In addition to spatial variation, particle concentrations have been shown to fluctuate over time (Adgate et al., 2002; Bari et al., 2003; Chow et al., 1994; Ito et al., 1995). Many urban areas experience a diurnal cycle in air pollutant concentrations, as anthropogenic sources make up a large portion of measured concentrations (Chow et al., 2002). Over short sampling durations (hourly or daily), concentrations between sites in a city will usually differ to a greater degree than measurements averaged over longer periods (annually) (Monn, 2001).

3. Exposure error implications

Exposure misclassification is a well recognised and inherent limitation of ecological studies involving the environment and disease (Armstrong et al., 1992). The terms ‘measurement error’, ‘exposure error’ and ‘exposure misclassification error’ refer to any discrepancy between the true value of a variable (e.g., personal exposure) and its measured or assumed value (ambient background concentration) (Thomas et al., 1993). The error from using central ambient sites as a proxy for personal exposure may lead to bias in estimates of health effects of air pollution, although the effect of exposure misclassification on risk estimates differs depending on study design and parameters (Armstrong et al., 1992; Dominici et al., 2003; Thomas et al., 1993). Most air pollution and health studies are of one of the following four epidemiological design types: time-series, case-crossover, panel or cohort design (Dominici et al.,

2003). Time-series, case-crossover and panel studies are best suited for measuring short-term (acute) effects of air pollution, while cohort studies are adept at measuring both short and long-term effects. The most common study designs in recent years have been time-series and cohort studies, which we focus on below (Bell et al., 2004).

3.1. Error in time-series studies

The major design feature of time-series studies is quantifying short-term temporal exposure variation and modelling the association between the probability of an outcome (e.g., mortality or morbidity) and the level of air pollution shortly before an event (Bell et al., 2004; Kunzli et al., 2001). This association is calculated while correcting for other variables (e.g., temperature, day of week, etc.) over a given area. In PM daily time-series studies, intra-community variation is not likely to be a source of large error in risk estimates if temporal correlation between sites is high, because it is longitudinal correlation between personal exposures and ambient concentrations that is most important and any bias due to exposure measurement is likely to be negative, i.e., an underestimation of the effect (Janssen et al., 1998; Schwartz, 2000; Schwartz and Levin, 1999; Schwartz et al., 1996; Zeger et al., 2000). However, as highlighted by Zeger et al. (2000), the relatively small bias in time-series studies that is caused by intraurban variations does not indicate the component of exposure error reflected in poor correlations between sites. The relative effects of poorly correlated sites and heterogeneous particulate distributions has not been systematically evaluated to date (Ito et al., 2005). Even when intra-community sites are well-correlated, overlooking spatial variation in particulates and the effects of aggregation across an area may lead to error associated with the *ecological fallacy*, where characteristics of individual exposure are wrongly inferred from characteristics of the aggregate (Berhane et al., 2004; Dominici et al., 2003; Greenland and Morgenstern, 1989; Nurminen and Nurminen, 2000; Nurminen et al., 1999; Piantadosi et al., 1988; Richardson et al., 1987; Selvin, 1958; Sheppard, 2002; Wakefield, 2003; Wakefield, 2004).

3.2. Error in cohort studies

Although time-series study designs have traditionally been applied to issues of air pollution and health with great frequency, cohort designs are being utilised with increasing frequency recently due in part to research that suggests cohort studies may provide more accurate estimates of risk, especially for mortality (Kunzli et al., 2001). A greater opportunity for error due to exposure misclassification under the homogeneous distribution

assumption may be present in cohort studies, which relate long-term average exposures to air pollution with health outcomes. Cohort studies usually involve selecting a group of individuals from multiple communities, to ensure identification of a cohort with significant exposure variation (Dominici et al., 2003). Cohort studies control for age, gender, socio-economic status, smoking and additional personal factors while differentiating exposure variation in space. Exposure calculation assumptions in these studies are dependent on the variation of exposure between cities or communities being greater than the variation *within* communities (Berhane et al., 2004; Greenland, 1992; Houthuijs et al., 2001; Sheppard and Prentice, 1995; Wakefield and Salway, 2001). If particulate concentrations are highly variable within urban areas, and this variability is greater at the intraurban level than between communities, exposure misclassification and downstream risk estimate errors may be of a greater magnitude than the same intraurban variations would manifest in single-community time-series study designs. It should be noted that *multi*-community time-series analyses may be subject to similar error, although markedly less than cohort studies, if inter-community variations are smaller than intra-community variations. Table 1 outlines epidemiological study designs and associated possibility of errors associated with exposure misclassification for the time-series and cohort study designs.

4. Review of monitoring-based studies

4.1. Defining uniformity

What constitutes PM heterogeneity or homogeneity at the intraurban level? Uniformity is not well defined in the published or regulatory literature and, as a consequence, it has been interpreted in a number of different ways. This vagueness also enhances the difficulty in drawing conclusions across the literature as to heterogeneous versus homogeneous distributions. As a means of discerning what constitutes uniformity, we have devised a classification scheme based on the limited definitions of heterogeneity in the literature. In this review, a relative value of twenty percent difference or greater, between intraurban sites is used to indicate a heterogeneous distribution. Blanchard et al. (1999) suggest a value of twenty percent for characterising spatial representativeness of an area based upon considerations that might be deemed significant from a health perspective.

In several of the studies reviewed, the authors actively identify whether or not the pollutant is uniformly distributed across the city (e.g., ‘PM_{2.5} concentrations were uniform between the 5 sites’). These cases are easily identifiable and categorised in accordance with the

Table 1
Study designs and associated possibility of errors in risk estimates from intra-community exposure misclassification

Design	Exposure duration	Communities	Ambient exposure measurement	Examples	Possibility of error
Time-series	Short term (days)	Single	Time varying	Pope et al. (1991), Pope et al. (1992), Schwartz and Dockery (1992)	Very low
Time-series	Short term (days)	Multiple	Time-varying community specific	Katsouyanni et al. (1997), Dominici et al. (2000), Lippman et al. (2000)	Low
Cohort	Long term (years)	Multiple	Space-varying community specific	Dockery et al. (1993), Pope et al. (2002), Gauderman et al. (2002)	Increased

conclusions of each study. However, in some cases authors do not clearly report on homogeneity, but instead present data on concentrations at several sites from which a heterogeneity conclusion may be drawn. Using Blanchard's 20% figure, reviewed studies that do not report or make conclusions on uniformity and show less than 20% variation between sites, are categorised as 'homogeneous', while those that report greater than 20% variation are categorised as 'heterogeneous.' Likewise, coefficients of variation and coefficients of divergence (defined in Section 4.2.2) lower than 0.20 are also classified as homogeneous. The relative guideline for coefficients of divergence is inferred from cities reported to have 'considerable spatial variation' in the literature (Pinto et al., 2004). While this twenty percent guideline is not a perfect measure of spatial heterogeneity, it is sufficient for our purpose here: the classification and comparison of studies. Correlation coefficients are not considered to be representative of spatial heterogeneity in this review, as correlations accurately track temporal similarity of paired sites, but have been shown to have no strong association with the actual spatial homogeneity of concentrations (see Section 5.2) (Pinto et al., 2004; US EPA, 2004).

4.2. Intraurban concentration studies

In the next section of this review, we focus on studies that examine intraurban variations in PM. The following criteria guided our search of the literature. Papers selected were: (i) peer-reviewed journal research published since 1994; with (ii) a specific focus on particulate air pollution concentrations; (iii) some component of intraurban exposure assessment (at least one pair of monitoring sites in an urban area); and (iv) based on monitored, not modelled, data.

Our search yielded thirty-three studies of intraurban particulate concentrations, which are reviewed in the following section and are divided into two categories based on study outcomes and the uniformity classification definition outlined in Section 4.1. Studies resulting in homogeneous (i.e., uniform) spatial concentration distribution within communities are reviewed first, followed by studies that resulted in heterogeneous concentration spatial distribution at the intra-community scale.

4.2.1. Homogeneous studies

Some monitored studies support the homogeneous distribution assumption of particulate concentrations within and across different urban areas. In this subsection reviewing homogeneous studies, we examine the literature by the type of analysis used in quantifying uniformity: correlation, absolute differences, and coefficients of variation.

One popular method of determining spatial uniformity across an urban area is the use of correlations between sites. In one of the early intraurban studies, Burton et al. (1996) measured PM₁₀, PM_{2.5} and coarse particles (PM_{10-2.5}) at eight sites ranging from 0.6 to 28.8 km from the city centre of Philadelphia. Pearson correlation coefficients between sites were high for PM₁₀ (0.62 < *r* < 0.96) and PM_{2.5} (0.70 < *r* < 0.96) but lower for coarse particulates (0.22 < *r* < 0.61). The study concluded that concentrations at a central monitoring site could be used to characterize exposure concentrations across the city, as well as in other similar cities in the north-eastern United States. Similarly, a later study conducted in Philadelphia and St. Louis by Wilson and Suh (1997) found high site-to-site correlations (0.80 < *r* < 0.96) for 24-h PM_{2.5} concentrations at the intraurban scale that were correlated to population density. However, correlations were slightly lower for PM₁₀ (0.79 < *r* < 0.96) and

much lower for $PM_{10-2.5}$ ($0.14 < r < 0.63$), indicating that a central monitoring site was more appropriate as an indicator of population exposure to fine particles than coarse particles. Bari et al. (2003) correlated $PM_{2.5}$ hourly and longer term averages at two monitoring sites in Manhattan and the Bronx, New York City. Correlations between the daily average concentrations of $PM_{2.5}$ at both sites were high ($r^2 = 0.92$), with less correlation for hourly data ($r^2 = 0.62$). Annual absolute concentration levels at the two sites were 15.2 and 15.5 $\mu\text{g}/\text{m}^3$ and monthly concentrations were 13.2 and 21.7 $\mu\text{g}/\text{m}^3$. DeGaetano and Doherty (2004) measured $PM_{2.5}$ in New York City using a high-density monitoring network of 20 stations and found similarly low spatial variation in concentrations across the city. Site correlations between a central site and all but one of the other sites in lower Manhattan were greater than 0.85. Suh et al. (1997) measured $PM_{2.5}$, PM_{10} , and coarse particle ($PM_{10-2.5}$) concentrations at six sites across the Washington, DC metropolitan area. Correlations were high and significant for PM_{10} ($0.64 < r < 0.98$) and $PM_{2.5}$ ($0.69 < r < 0.98$), but lower for $PM_{10-2.5}$ ($0.34 < r < 0.48$). The study concluded that a central stationary monitoring site was sufficient to estimate the ambient exposures for PM_{10} and $PM_{2.5}$.

Buzorius et al. (1999) measured urban aerosol number concentration at four sites in the Helsinki area, and noted that one sampling site could accurately describe changes in a relatively large area of the city with a high correlation coefficient ($r > 0.7$). A recent study of 10 urban environments in the Emilia-Romagna region of Italy, found that correlations between sites for PM_{10} were high (mean $r = 0.89$), but correlation coefficients were lower for TSP ($0.49 < r < 0.91$). The results suggested that a single fixed-site monitoring station might not accurately characterise the spatial nature of air pollution at the intraurban scale, but found evidence that intraurban spatial variability of particulate concentrations were low (Sajani et al., 2004). Ye et al. (2003) conducted a 9-month analysis of $PM_{2.5}$ concentrations at two sites in Shanghai that were 4 km apart, yielding low long-term average variations. Average concentrations over the period at the two sites were 67.6 and 64.6 $\mu\text{g}/\text{m}^3$ with a high correlation value between sites ($r^2 = 0.94$), suggesting regionally homogeneous sources. Researchers in Vancouver measured PM_{10} at 11 monitoring sites and found high temporal correlation between sites and relatively small spatial variation. However, the scale of the monitoring network (7.5 km) may not have sufficiently high spatial resolution to detect more localized and important features of the PM_{10} field (Li et al., 1999). Ito et al. (2005) conducted a meta-analysis of PM_{10} among other pollutants using 1–30 monitoring sites in 225 air quality control regions and found that monitor-to-monitor concentrations were on average well correlated ($\sim 0.6 < r < 0.7$). Both the

region of the country and distance between monitors were found to be significant predictors of temporal correlation between sites, but differences in absolute levels between monitors were not considered in the analysis.

In addition to correlation, some studies that found intraurban PM homogeneity calculated absolute differences in concentrations between monitoring locations. A study in Basel, Switzerland, at six mobile sites and one fixed site within 3.3 km of each other found relatively homogeneous PM_{10} mass concentrations ranging from 27.6 to 32.0 $\mu\text{g}/\text{m}^3$ (Röösli et al., 2000). In a related study in the same city, researchers found PM_{10} mass concentrations to be uniformly distributed ($\pm 10\%$) across the city at seven sites with the exception of a site in a street canyon next to a stoplight (Röösli et al., 2001). Oglesby et al. (2000) compared outdoor concentrations at 28 sites in Basel to a fixed central monitoring site. High Spearman correlations (mean $r = 0.96$) were observed between outdoor concentrations of $PM_{2.5}$ and corresponding weighted averages of fixed-site PM_4 (PM less than 4 μm in diameter) levels. However, outdoor levels were on average 9% lower than the fixed site (20.0 versus 21.9 $\mu\text{g}/\text{m}^3$). The study concluded that for regional air pollution, fixed-site concentrations are valid surrogates for population exposure in the city. He et al. (2001) measured long-term average concentration variations of $PM_{2.5}$ at two sites 10 km apart in Beijing. Annual mean concentrations at the two sites were 115 and 127 $\mu\text{g}/\text{m}^3$, but weekly values were much more variable (37–357 $\mu\text{g}/\text{m}^3$). Results of daily variations between sites were not reported.

Two studies finding homogenous concentrations utilised coefficients of variation in their characterisation of uniformity. A study of 25 metropolitan areas in six Central and Eastern European Countries showed significant coefficients of variation (CV) within study areas for both PM_{10} (24%) and $PM_{2.5}$ (28%), although between-study area CVs were around four times higher. The authors concluded that a single sampling site could be used to characterize exposures of the population in the study area (Houthuijs et al., 2001). Martuzevicius et al. (2004) conducted a spatio-temporal analysis of $PM_{2.5}$ at 11 sites in Cincinnati, a city with particularly high motorway traffic density. Low-concentration variations between sites were reported for total mass $PM_{2.5}$ (median CV = 11.3%), although various elemental concentrations demonstrated larger variations ($38.2\% < CV < 68.7\%$).

4.2.2. Heterogeneous studies

Recent research shows that the intraurban spatial distributions of PM concentrations in some study areas are heterogeneous. The studies that report heterogeneity in this subsection are discussed in four categories: (a) studies that measure uniformity by absolute concentrations; (b) those that apply a coefficient of divergence in

determining uniformity; (c) those that attribute heterogeneity to local sources; and (d) studies attributing lack of uniformity to land use types.

Several studies used absolute concentration differences to make conclusions about intraurban PM heterogeneity. [Nerriere et al. \(2005\)](#) measured seasonal $PM_{2.5}$ and PM_{10} concentrations in four metropolitan areas in France. Each metropolitan area was sampled at a proximity site, a background site, and an industrial site. Maximum average $PM_{2.5}$ ambient air concentration differences between types of site at the intraurban scale ranged from 20% to 64% in the winter, and from 17% to 69% in the summer. Maximum average PM_{10} ambient concentration differences between types of ambient sites ranged from 26% to 34% in the winter and from 15% to 22% in the summer. $PM_{2.5}$ and, to a greater extent, PM_{10} concentrations underestimated population exposures in almost all cities, seasons and age categories. The study concluded that each urban area should undergo a site-specific analysis before making assumptions about population exposures from ambient air monitoring data. [Cyrus et al. \(1998\)](#) also measured PM_{10} concentration level differences between one downtown site and two suburban sites in Erfurt, East Germany. Median concentration differences between sites were 30–40%. Due to the discrepancies between sites, the authors concluded that the use of only one monitoring site as a proxy for population exposure may be biased. [Noble et al. \(2003\)](#) compared $PM_{2.5}$ and PM_{10} absolute mass concentrations at two sites over a 3-week period in El Paso, Texas. Average gravimetrically sampled PM_{10} concentrations between sites varied significantly (61–91 $\mu\text{g}/\text{m}^3$), while $PM_{2.5}$ concentrations were more uniform (17–20 $\mu\text{g}/\text{m}^3$). Continuous variations in the level of particulates were not available at both sites.

Several studies finding heterogeneous distributions of particulates applied a coefficient of variation (CV) or a coefficient of divergence (COD) to describe relative intraurban concentration heterogeneity. Researchers in Athens, where concentrations are routinely higher than other urban areas across Europe, conducted PM_{10} sampling at four sites. Spatial variation between sites was found to be significant. The coefficient of variation was high within the study area ($CV = 0.36$), and correlation coefficients ranged from 0.57 to 0.84. These values were consistent with other studies where ambient PM_{10} concentrations were high ([Grivas et al., 2004](#)). [Pinto et al. \(2004\)](#) conducted a thorough spatial uniformity analysis using US Environmental Protection Agency $PM_{2.5}$ data from over 1000 sites in 27 Metropolitan Statistical Areas (MSAs) across the United States. Various degrees of spatial heterogeneity were observed, using a coefficient of divergence to characterize spatial heterogeneity. A large range of intraurban Pearson correlation coefficients ($0.28 < r < 0.98$) were

found for the 27 MSAs. A COD was applied to sites within study areas as a relative measure of particulate concentration uniformity. The COD is defined mathematically as

$$\text{COD}_{jk} = \sqrt{\frac{1}{p} \sum_{i=1}^p [(x_{ij} - x_{ik}) / (x_{ij} + x_{ik})]^2},$$

where x_{ij} and x_{ik} represent the 24 h average particulate concentration for sampling day i at sampling sites j and k , and where p is the number of observations ([Wongphatarakul et al., 1998](#)). A COD of zero means there are no differences between concentrations at the sites, while a value approaching one indicates maximum differences and absolute heterogeneity. Metropolitan areas in the central and eastern United States showed relative spatial uniformity ($0.082 < \text{max COD} < 0.27$), whereas cities in other areas of the country, especially in the west, showed higher degrees of spatial variation ($0.20 < \text{max COD} < 0.48$). The study concluded that high correlation between sites does not necessarily indicate uniformity, and that spatial data from individual urban areas need to be examined before conclusions about $PM_{2.5}$ homogeneity can be made, especially for exposure application in epidemiological cohort studies.

Particulate source composition is a dominant factor in determining spatial variability ([Monn, 2001](#)). Many of the studies finding heterogeneity concluded that the lack of uniformity was attributable, at least in part, to the nature and density of the sources. [Wongphatarakul et al. \(1998\)](#) studied source-related intraurban spatial variations of $PM_{2.5}$ at five monitoring sites in Los Angeles. While correlations between sites were large ($0.704 < r < 0.928$) for site pairs sharing similar sources, correlations were low ($-0.027 < r < 0.120$) among sites with dissimilar sources. The study concluded that sites sharing similar sources monitored like concentrations, as reflected by the COD. Source-similar sites had a COD of 0.099, while sites measuring concentrations from different types of sources resulted in a COD of 0.230. [Kim et al. \(2005\)](#) used positive matrix formulation to compare spatial variability in $PM_{2.5}$, its elemental components, and source contributions at ten sites in St. Louis, MO. $PM_{2.5}$ concentrations were fairly well correlated between all site pairs (mean $r = 0.79$), but large differences were found in component species (mean $r \leq 0.59$, mean $\text{COD} \geq 0.30$), suggesting that there may be significant potential for exposure misclassification in time-series epidemiologic studies. [Blanchard et al. \(1999\)](#) studied spatial representativeness of PM_{10} mass and chemical composition in the California cities of Corcoran, Bakersfield and Fresno at 12–25 sites in each metropolitan area. Mean concentrations of PM_{10} varied from core-site concentrations by 20% or greater for distances of 4–14 km. Transport of primary pollutants were found at distances of 10–30 km, indicating that one

or few central monitoring sites may not represent PM_{10} concentrations over extended areas. Another study in central California by VanCuren (1999), identified several problems associated with using central monitoring sites to characterise particulate pollution over a larger area. The study asserted that the similarity of concentrations in time and space between widely separated monitoring stations is probably due to the sampling of comparable local environments rather than a uniformly mixed regional air mass. Ito et al. (2004) recently studied source-apportioned $PM_{2.5}$ at three sites in New York City and conclude that source-oriented evaluations of PM associated health effects should consider the uncertainty associated with spatial representative of the species measured at a single monitor.

Several studies finding intraurban heterogeneous PM concentrations attributed lack of uniformity to localised traffic sources. Harrison et al. (1999) measured PM_{10} particle number counts at five sites in Birmingham, United Kingdom. While particle number counts between two stations were different by a factor of three, concentration masses were comparable. The variation was attributed to the influence of vehicle emissions on particle numbers. In one site comparison, particle counts were 7.5 times the background levels, while PM_{10} mass was only double background levels. Hoek et al. (2002b) collected $PM_{2.5}$ 14-day samples four times at 40 or more intraurban sites (not concurrently) in Stockholm, Munich and the Netherlands. Annual average $PM_{2.5}$ concentrations ranged from 11 to $20 \mu\text{g}/\text{m}^3$ in Munich, $8\text{--}16 \mu\text{g}/\text{m}^3$ in Stockholm, and from 14 to $26 \mu\text{g}/\text{m}^3$ in the Netherlands. Concentrations near major roads were on average 17–18% higher than urban background sites. Junker et al. (2000) analysed a number of particulate indicators at three sites in Basel, Switzerland. Average concentrations between sites ranged from 17.8 to $28.8 \mu\text{g}/\text{m}^3$ for particulates less than $4 \mu\text{m}$ in aerodynamic diameter (PM_4) and from 22.4 to $34.8 \mu\text{g}/\text{m}^3$ for PM_{10} . Temporally non-concurring average ultrafine particle concentrations between the three sites in the city ranged in number from 5690 to $19,300/\text{cm}^3$. These differences were also attributed to variations in traffic densities between locations. Goswami et al. (2002) monitored $PM_{2.5}$ at 40 outdoor sites in Seattle, Washington and found significant spatial variability in outdoor concentrations. Spatial characteristics of the sites, such as elevation and distance from major roads, were found to be significant in predicting mass concentrations. Chen and Mao (1998) found vertical and horizontal heterogeneous concentrations of PM_{10} and TSP during the summer and fall of 1991 in Taipei related to traffic sources. PM_{10} concentration levels on a roadside, sidewalk and covered walkways near a busy road were 527.8, 466 and $477 \mu\text{g}/\text{m}^3$, respectively. Although the main street was nearest to the main pollution source, the side street experienced the highest

PM_{10} concentrations due to traffic jams and a lower diffusion level. Smargiassi et al. (2005) recently found that while $PM_{2.5}$ concentrations were relatively uniform between four sites near urban roads and a background site, filter absorption coefficients differed by 40% between traffic sites and the background site, indicating that intraurban spatial variability in $PM_{2.5}$ absorption coefficients was related to traffic intensities, even at very small scales.

Land use is associated with specific sources, and some studies chose to attribute spatial heterogeneity to monitoring site land use characteristics. Sun et al. (2004) collected aerosol samples at an industrial site, a traffic site and a residential land use site in Beijing and a relatively homogeneous distribution of $PM_{2.5}$ chemical species was found. However, PM_{10} levels varied significantly between sites, with the highest concentrations at the residential site in winter and the industrial site in the summer. Chan et al. (1997) measured PM_{10} and its components over 1 year at five sites within 15 km of the city centre of Brisbane, Australia. Average mass concentrations ranged from 18.9 to $37.8 \mu\text{g}/\text{m}^3$ over the period. Major rural dust episodes did not increase seasonal averages of crustal matter, and chemical composition variations were generally explained by the nature of sources, climate and land use. Chan et al. (2001) collected samples in Hong Kong at 11 sites representing four land use categories: urban industrial, new town, urban residential and urban commercial. PM concentrations varied greatly between and within land use categories. The highest PM concentrations were found in urban residential areas, but the differences in concentration levels among these districts were also significant.

The plethora of studies demonstrating clear variability and clear uniformity of intraurban particulate concentrations in various areas of the world underscores the assertion that conclusions about concentration homogeneity are mixed and are not necessarily applicable between study sites. Next, we compare the literature by factors between and within study areas as a means of investigating assumptions about intraurban PM uniformity.

5. Comparison of intraurban studies

There are a number of factors that may contribute to assumptions about concentration uniformity at the intraurban scale. First, we review the selected studies by particle size and methodological issues pertaining to relative and absolute characterizations of intraurban particulate concentrations. Second, we examine aspects of study designs and sampling characteristics that may influence the intraurban homogeneity conclusion. Finally, we compare methodological and sampling

characteristics of studies conducted in the same urban areas, with a focus on what factors lead to alternative conclusions about uniformity.

5.1. Particle size

In total, 33 intraurban particulate studies were reviewed. Of the 33 studies reviewed, nine (27%) of them found a homogeneous distribution of particulates for all particulate size classes in their respective study, while 17 (52%) found heterogeneous distributions, and seven studies (21%) came to a conclusion that were conditional on particulate size fraction (i.e., that $PM_{2.5}$ was uniform and PM_{10} was not). As several of the papers focused on more than one particle size class (i.e., PM_{10} and $PM_{2.5}$ were analysed in the same study), this set of 33 studies actually produced 51 conclusions about particulate uniformity by particle size fraction (Table 2). For the purpose of comparison, we have again separated studies into homogeneous and heterogeneous groups based on their conclusions, as outlined in Section 4.1.

As discussed, the most significant factor governing the spatial uniformity of PM is its aerodynamic diameter, or size fraction. Coarse particles (PM_{10} and $PM_{10-2.5}$) are generally thought to be less uniform than finer particles ($PM_{2.5}$ and PM_1) over urban areas, while UFP ($d_a < 0.1 \mu m$) has been shown to exhibit high spatial variability (Monn, 2001). A larger group of PM_{10} studies concluded that spatial distribution displayed higher heterogeneity (60%) than for $PM_{2.5}$ studies (43%). Intraurban spatial variation of coarse particles ($PM_{10-2.5}$) was high in all four studies reviewed, as well as in all three TSP studies reviewed and all four studies measuring UFP particle numbers. There were no examples in the literature where larger particles (PM_{10} or $PM_{10-2.5}$) were found to be more uniform than $PM_{2.5}$ in the same study area.

5.2. Methodological factors

Statistical methods used in determining absolute and relative intraurban concentration uniformity may also affect conclusions about heterogeneity. The method used to determine spatial variation was compared across studies as shown in Table 2. Seventeen (52%) studies examined absolute concentration differences between sites, 17 (52%) utilised some form of correlation or regression to describe variation, four (12%) employed coefficient of variation, and only three (9%) applied a coefficient of divergence. The calculation and reliance on correlation coefficients as a relative measure for interpreting the degree of intraurban spatial uniformity (used in nearly half of the compared studies) may contribute to incorrect assumptions about spatial homogeneity and ultimately, exposure misclassification. However, depending on the purpose for measuring intraurban

heterogeneity, one method may be more appropriate than another. As highlighted earlier (Section 3), in time-series studies, where longitudinal correlations are important, absolute concentration differences are not as critical as temporal correlations between sites and Pearson correlations may suffice as descriptors of intraurban concentrations. However, in cohort studies, absolute differences and intraurban uniformity are liable to have a greater effect on health effect estimates (Table 1). Therefore, when determining intraurban uniformity for time-series studies, correlations are recommended and when determining uniformity for long-term cohort studies, coefficients of divergence and 90th percentiles of absolute concentration differences are appropriate. A study by the US Environmental Protection Agency (2004) is illustrative of the fact that correlation coefficients and absolute measures of concentration can yield different conclusions about uniformity. Fig. 1 shows that concentrations at paired monitoring sites in three MSAs were highly correlated, yet exhibited various ranges of concentration heterogeneity. Paired sites in Columbia, SC; Chicago, IL; and Detroit, MI all exhibited similar and high Pearson correlations ($0.93 < r < 0.97$), yet daily absolute concentration difference profiles were markedly different. While absolute concentrations for site pairs in Columbia were highly similar, Chicago and Detroit had higher spreads of concentrations between site pairs, indicating greater heterogeneity. In this case, an assumption that intraurban concentration levels were uniform in Chicago and Detroit based on Pearson correlations alone could present error due to exposure misclassification if this assumption was applied in the context of a long-term cohort study.

5.3. Site and sampling characteristics

In addition to the influence of particle size and methodological considerations, site and sampling characteristics of studies may influence intraurban heterogeneity conclusions. In this subsection, we review the 31 studies by the types of sources monitored, the numbers of intraurban monitoring sites, the averaging times, and numbers of concurrent samples between sites (Table 3).

Particulate sources are an important component in determining not only spatial heterogeneity, but also exposure classification and health effects (Ito et al., 2004; Kim et al., 2005; Laden et al., 2000; Oglesby et al., 2000; Wongphatarakul et al., 1998). Sites were designated as being ambient (a), traffic (t) or industrial (i) based on the sources monitored. Of the 31 studies reviewed, 17 (52%) monitored ambient air alone, where three (9%) monitored at sites that were designated as influenced by traffic sources. Thirteen of all studies (39%) compared monitored concentrations between more than one type of source. More than two-thirds

Table 2
Reviewed studies by absolute and relative methodological characteristics

Conclusion/study ^a	Particle d_a^b	Absolute measure (averaging period) ^c	Relative measure ^d
<i>Uniform (homogeneous)</i>			
Bari et al. (2003)	PM _{2.5}	15.2–15.5 (anl); 13.2–21.7 (min)	Mean $r^2 = 0.92$ (hourly); mean $r^2 = 0.62$ (daily)
Burton et al. (1996)	PM ₁₀ , PM _{2.5}	17.7–21.0 (anl) PM _{2.5} ; 24.5–28.4 (anl) PM ₁₀	$0.62 < r < 0.96$ (PM ₁₀); $0.70 < r < 0.96$ (PM _{2.5})
DeGaetano and Doherty (2004)	PM _{2.5}		$r > 0.85$ most sites
He et al. (2001)	PM _{2.5}	115–127 (anl); 37–357 (w)	
Houthuijs et al. (2001)	PM ₁₀ , PM _{2.5}	41–98 (anl) PM ₁₀ ; 29–68 (anl) PM _{2.5}	CV = 24% (PM ₁₀); CV = 28% (PM _{2.5})
Ito et al. (2005)	PM ₁₀		$\sim 0.6 < r < 0.8$
Li et al. (1999)	PM ₁₀		Mean $r = 0.59$
Martuzevicius et al. (2004)	PM _{2.5}		Median CV = 11.3%
Noble et al. (2003)	PM _{2.5}	17–20 (d)	
Oglesby et al. (2000)	PM _{2.5}		Mean $r = 0.96$
Rööslä et al. (2000)	PM ₁₀	27.6–32.0 (d)	
Sajani et al. (2004)	PM ₁₀		Mean $r = 0.89$ (PM ₁₀)
Suh et al. (1997)	PM ₁₀ , PM _{2.5}		$0.64 < r < 0.98$ (PM ₁₀); $0.69 < r < 0.98$ (PM _{2.5})
Sun et al. (2004)	PM _{2.5}	135–182 (d, winter)	
Wilson and Suh (1997)	PM ₁₀ , PM _{2.5}		$0.79 < r < 0.96$ (PM ₁₀); $0.80 < r < 0.96$ (PM _{2.5})
Ye et al. (2003)	PM _{2.5}	64.6, 67.6 (mean anl)	$r^2 = 0.94$
<i>Non-uniform (heterogeneous)</i>			
Blanchard et al. (1999)	PM ₁₀		> 20% variation
Burton et al. (1996)	PM _{10–2.5}	5.2–8.1 (anl)	$0.22 < r < 0.61$
Chan et al. (1997)	PM ₁₀	18.9–37.8 (d)	
Chan et al. (2001)	TSP, PM ₁₀ , PM _{2.5}	94.85–301.63 (d) TSP; 67.67–142.68 (d) PM ₁₀ 50.01–125.12 (d) PM _{2.5}	
Chen and Mao (1998)	TSP, PM ₁₀	54.0–506.0 (d) TSP; 45.9–455.0 (d) PM ₁₀	
Chen et al. (1999)	PM ₁₀ , PM _{2.5}	42.19–77.10 (d) PM ₁₀ ; 23.09–48.47 (d) PM _{2.5}	
Chow et al. (1994)	PM _{2.5}	25.4–63.9 (Summer); 68.9–90.2 (Fall)	
Cyrys et al. (1998)	PM ₁₀		$0.69 < r < 0.94$
Grivas et al. (2004)	PM ₁₀		CV = 0.36 $0.57 < r < 0.84$
Harrison et al. (1999)	PN	1.86×10^4 – 9.60×10^4 (d) cm ⁻³	
Hoek et al. (2002b)	PM _{2.5}	11–20 (anl) Munich; 8–16 (anl) Stockholm; 14–26 (anl) Netherlands	
Houthuijs et al. (2001)	PM _{10–2.5}	12–40 (anl)	CV = 63%
Ito et al. (2004)	PM _{2.5}		$0.26 < r < 0.95$
Junker et al. (2000)	PM ₁₀ , PN	22.4–34.8 (27h) PM ₁₀ ; 5,690–19,200 (27h) PN	
Kim et al. (2005)	PM _{2.5}		$r \leq 0.59$, COD ≥ 0.30 (component species)
Monn et al. (1997)	PM ₁₀		CV = 13%
Nerriere et al. (2005)	PM ₁₀ , PM _{2.5}	34% winter diff. (d) PM ₁₀ ; 69% summer diff. (d) PM _{2.5}	
Noble et al. (2003)	PM ₁₀ , PN	61–91 (d) PM ₁₀ ; 13,600–14,600 (h) PN	
Pinto et al. (2004)	PM _{2.5}	10.5–31.3 (anl), (Los Angeles)	$0.20 < \max \text{COD} < 0.48$ (western US)

Table 2 (continued)

Conclusion/study ^a	Particle d_a ^b	Absolute measure (averaging period) ^c	Relative measure ^d
Sajani et al. (2004)	TSP	184–292 (d, winter)	$0.49 < r < 0.91$
Suh et al. (1997)	PM _{10–2.5}		$0.34 < r < 0.48$
Sun et al. (2004)	PM ₁₀		
Wilson and Suh (1997)	PM _{10–2.5}		$0.14 < r < 0.63$
Wongphatarakul et al. (1998)	PM _{2.5}		$0.09 < \text{COD} < 0.572$ (Los Angeles)

^aUniformity conclusion based on Section 4.1 and study name.

^bParticle aerodynamic diameter: particulate matter (PM_x) or particle number (PN) count.

^cAbsolute measure of concentration difference between sites in $\mu\text{g}/\text{m}^3$ (PM) or cm^{-3} (PN) with averaging period: minutes (min), hours (h), 24-h days (d), weeks (w), years (anl), or season.

^dRelative measure of intraurban concentration variation: correlation (r), regression (r^2), coefficient of variation (CV), or (COD) coefficient of divergence.

of all studies (69%) that found homogeneous concentrations monitored only one type of source, while just over half of studies (54%) finding heterogeneous intraurban concentrations utilised a single monitoring site source-type.

The number of sites that concurrently monitor concentrations across an urban area may also influence uniformity conclusions. Studies finding homogeneous concentrations measured at 2–30 concurrently operating sites with a median of six sites, where studies finding heterogeneous concentrations utilised from 2 to 42 sites with a median of five. Base averaging times for the studies finding homogeneous concentrations were from minutes to weeks, while heterogeneous studies all used sample averaging times of less than or equal to 24-h. The long averaging times in some of the studies which found uniform concentrations may have biased results towards homogeneous concentrations by not accounting for shorter-term variations in pollution.

In addition to methodological factors such as site source-types, number of sites, and averaging times, the number of concurrent samples collected at multiple sites may influence the conclusions of studies interested in intraurban uniformity. Of the studies finding homogeneity, the median sample size was 80 with a range of 19 to >1000, while the studies finding heterogeneous intraurban concentrations of particulates sampled far fewer times, with a median sample size of 48 and a range from 8 to >1000 samples. Seven of the 24 (29%) of the studies finding heterogeneous concentrations had concurrent sample sizes of ≤ 20 , while only two studies finding uniform particulate concentrations conducted 20 or fewer concurrent samples. A large number of concurrent samples ($n \geq 50$) would increase the statistical power of studies investigating uniformity of intraurban particulates.

5.4. Comparison by study area

As a final step in our comparative analysis, aspects of intraurban studies conducted on the same particle types in the same urban areas were compared. The aim of this section is to demonstrate the effect of differing study characteristics on diverse uniformity conclusion outcomes given similar study areas. The factors evaluated were (a) the number of monitoring sites, (b) absolute and relative methods of determining homogeneity, (c) study site and sampling characteristics, and (d) the uniformity conclusion. Selected areas for comparison were required to have at least three intraurban studies conducted in the same metropolitan area based on the same particle size fraction. We selected 12 studies in four metropolitan areas: Basel, Switzerland; Los Angeles, New York City, and Philadelphia (Table 4).

In Basel, three separate studies of PM₁₀ were reviewed. Conclusions about uniformity were mixed, ranging from concentrations varying ‘a great deal’ to ‘relatively constant’ intraurban uniformity. Study A (Junker et al., 2000), which concluded high variability, had three sampling sites, which may not have been a representative sample of the area. Studies B (Monn et al., 1997) and C (Röösli et al., 2001) found similar small scale variations of 10% and 13%, respectively. It should be noted that studies A and C utilised similar methods for characterising uniformity even though study C monitored concurrently at twice as many sites, and had more samples by nearly a factor of three, which may explain the difference in uniformity conclusions. All three studies measuring daily PM_{2.5} in Los Angeles yielded results indicating intraurban variability. Conclusions for study A (Chow et al., 1994) were based on daily concentration differences, but studies B (Pinto et al., 2004) and C (Wongphatarakul et al., 1998) employed a coefficient of divergence. Annual means

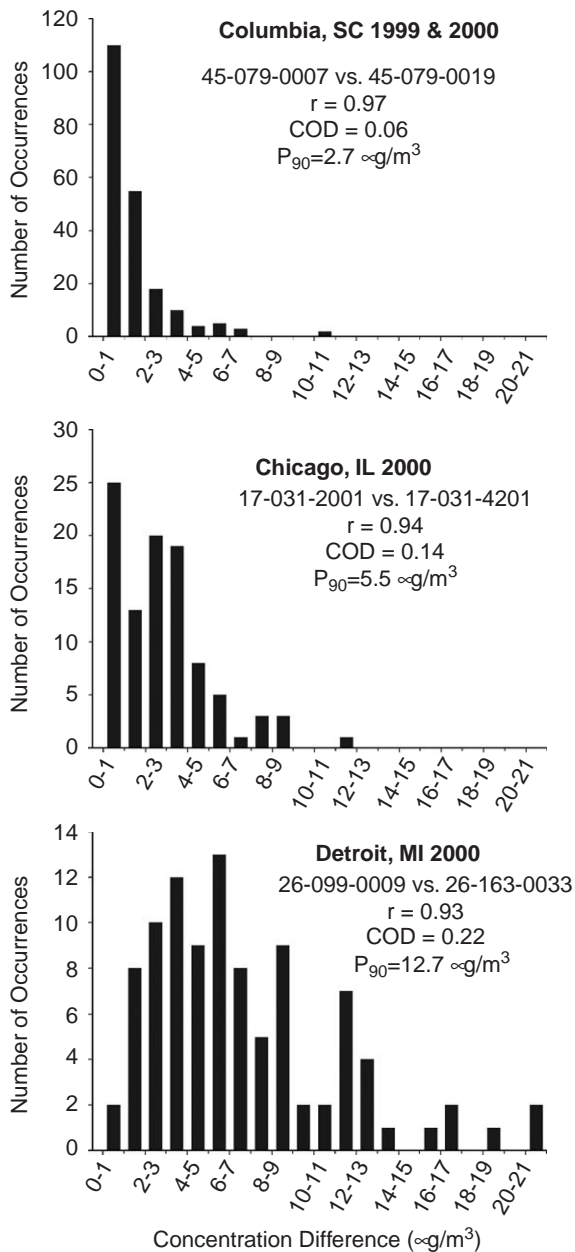


Fig. 1. Three examples of well correlated site-pairs with coefficients of divergence (COD) and 90th percentiles of daily concentration differences (P_{90}). Note that the y-axis scales are not uniform. (Source: US EPA 2004.)

between sites in study B were similar (20.2–24.8 $\mu\text{g}/\text{m}^3$), but COD values ranged from 0.14 to 0.26. Study C also found uniformity between sites with similar sources (COD = 0.099 between downtown and Burbank sites), but high heterogeneity between sites with different source compositions (COD = 0.23). Study A had almost twice the number of monitoring sites ($n = 9$), but a low

number of samples ($n = 11$) compared to 60 or more samples in studies B and C. Of the three studies in Los Angeles, those that utilised CODs reported high to moderate heterogeneity, while a study based on absolute concentration differences with a low number of concurrent samples resulted in moderate heterogeneity. In New York City, three studies were compared. All three papers used correlations to compare $\text{PM}_{2.5}$ concentrations, resulting in 'low' heterogeneity, except for some pollution source-types in study C (Ito et al., 2004), which indicate 'high' heterogeneity. Studies A and B monitored using hourly averaging times, while study C utilised 24-h averages. All three studies had sufficient sample sizes ($n > 170$), but studies A and C monitored at only a few sites ($n \leq 3$) while study B analysed data from 20 sites. New York City pollution is known to be dominated by regional processes and not by local sources, which is a major factor in determining intraurban uniformity (DeGaetano and Doherty, 2004). In addition to differences in averaging time, the high range of correlations found in study C is likely due to the inclusion of both ambient and traffic-related sites, while studies A and B focused on ambient sources alone. All three studies in Philadelphia, spanning a total of 8 years, concluded that daily $\text{PM}_{2.5}$ concentrations were uniform across the city, which is likely reliable as all of the studies were based on a sufficient number of sites ($n \geq 5$) and samples ($n > 60$). Correlations between sites were similar ($r \sim 0.9$) in studies A–C (Burton et al., 1996; Pinto et al., 2004; Wilson and Suh, 1997, respectively). The range of CODs (0.08–0.16) in study B indicates that for the most part, site pairs share similar absolute concentrations, indicating uniformity.

6. Discussion

To date, there have been no published reviews of the literature which have focused specifically on outdoor monitored intraurban particulate air pollution exposures across entire urban areas. This review has determined that particulate concentrations across urban areas are not always uniform, regardless of particles size, and, consequently, caution is advised when using central monitoring sites as proxies for population exposure in epidemiological studies without a prior analysis of spatial uniformity.

Our review of methods used in determining uniformity at the intraurban scale indicates that method may be an important factor in accurately depicting uniformity. Some methods may be more descriptive than others. Correlation coefficients are one of the more commonly used techniques for describing concentration uniformity at the intraurban and other scales. The fact that correlations, when used alone, are poor predictors of actual concentration uniformity has been highlighted

Table 3
Reviewed studies by quantitative site and sampling characteristics

Conclusion/study ^a	Particle d_a ^b	Site type ^c	Site no. ^d	Avg. time ^e	Sample no. ^f
<i>Uniform (homogeneous)</i>					
Bari et al. (2003)	PM _{2.5}	a	2	h	>1000
Burton et al. (1996)	PM ₁₀ , PM _{2.5}	a	8	d	>100
DeGaetano and Doherty (2004)	PM _{2.5}	a	20	h	>1000
He et al. (2001)	PM _{2.5}	a	2	w	52
Houthuijs et al. (2001)	PM ₁₀ , PM _{2.5}	a	3	d	488
Ito et al. (2005)	PM ₁₀	a	81-414	d	>60
Li et al. (1999)	PM ₁₀	a	11	h	>1000
Martuzevicius et al. (2004)	PM _{2.5}	a,t	11	d	219
Noble et al. (2003)	PM _{2.5}	t	2	min	19
Oglesby et al. (2000)	PM _{2.5}	a	44	d	38
Rööslä et al. (2000)	PM ₁₀	a,t	4-6	d	52
Sajani et al. (2004)	PM ₁₀	a,t,i	12	d	356
Suh et al. (1997)	PM ₁₀ , PM _{2.5}	a	6	d	~30
Sun et al. (2004)	PM _{2.5}	a,t,i	3	d	20
Wilson and Suh (1997)	PM ₁₀ , PM _{2.5}	a,i	3-7	d	>140
Ye et al. (2003)	PM _{2.5}	a	2	w	52
<i>Non-uniform (heterogeneous)</i>					
Blanchard et al. (1999)	PM ₁₀	a	12-25	d	~40
Burton et al. (1996)	PM _{10-2.5}	a	8	d	>100
Chan et al. (1997)	PM ₁₀	a,t	5	d	~76
Chan et al. (2001)	TSP, PM ₁₀ , PM _{2.5}	a,t	11	d	~12
Chen and Mao (1998)	TSP, PM ₁₀	a,t	3	d	10
Chen et al. (1999)	PM ₁₀ , PM _{2.5}	a	9	d	8
Chow et al. (1994)	PM _{2.5}	a	9	d	11
Cyrus et al. (1998)	PM ₁₀	a	4	d	23
Grivas et al. (2004)	PM ₁₀	a,i,t	4	d	26
Harrison et al. (1999)	PN	a,t	5	min	>1000
Hoek et al. (2002b)	PM _{2.5}	t	40-42	d	56
Houthuijs et al. (2001)	PM _{10-2.5}	a	3	d	488
Ito et al. (2004)	PM _{2.5}	a,t	3	d	170
Junker et al. (2000)	PM ₁₀ , PN	a	3	min	17
Kim et al. (2005)	PM _{2.5}	a	10	d	364
Monn et al. (1997)	PM ₁₀	t	4	d	176
Nerriere et al. (2005)	PM ₁₀ , PM _{2.5}	a,t,i	4	d	37
Noble et al. (2003)	PM ₁₀ , PN	t	2	d	19
Pinto et al. (2004)	PM _{2.5}	a	4-8	d	>60
Sajani et al. (2004)	TSP	a,t,i	12	d	356
Suh et al. (1997)	PM _{10-2.5}	a	6	d	~30
Sun et al. (2004)	PM ₁₀	a,t,i	3	d	20
Wilson and Suh (1997)	PM _{10-2.5}	a,i	3-7	d	>140
Wongphatarakul et al. (1998)	PM _{2.5}	a,t,i	5	d	~60

^aUniformity conclusion based on Section 4.1 and study name.

^bParticle aerodynamic diameter: particulate matter (PM_x) or particle number (PN) count.

^cMonitoring site types: ambient (a), traffic (t), or industrial (i).

^dNumber of concurrent intra-community sites.

^eAveraging times are minutes (min), hours (h), 24-h days (d), weeks (w), or years (anl).

^fNumber of concurrent intra-community samples.

in the United States, but not elsewhere (Pinto et al., 2004). Epidemiological studies that classify particulate concentrations as 'uniform' based solely on correlation coefficients may increase the risk for error in risk

estimates due to exposure misclassifications, especially in long-term cohort studies relating air pollution concentrations to health. Based on our review, we recommend that correlation coefficients be used in conjunction with

Table 4
Comparison of intraurban studies conducted on same particle size fraction by city

City/study	d_a^a	Measurement (averaging period) ^b	Site type ^c	Site no. ^d	Avg. time ^e	Sample no. ^f	Conclusion ^g
<i>Basel, Switzerland</i>							
(A) Junker et al. (2000)	PM ₁₀	22.4–34.8 (27 h)	a	3	min	17	Heterogeneous
(B) Monn et al. (1997)	PM ₁₀	CV = 13%	t	4	d	176	Heterogeneous
(C) Rööslı et al. (2000)	PM ₁₀	27.6–32.0 (d)	a,t	4–6	d	52	Uniform
<i>Los Angeles</i>							
(A) Chow et al. (1994)	PM _{2.5}	25.4–63.9 (summer); 68.9–90.2 (fall)	a	9	d	11	Heterogeneous
(B) Pinto et al. (2004)	PM _{2.5}	0.07 ≤ COD ≤ 0.48, 4.3 ≤ P ₉₀ ≤ 43.1; 10.5–31.3 (anl)	a	4–5	d	> 60	Heterogeneous
(C) Wongphatarakul et al. (1998)	PM _{2.5}	0.09 ≤ COD ≤ 0.572	a,t,i	5	d	~60	Heterogeneous
<i>New York City</i>							
(A) Bari et al. (2003)	PM _{2.5}	15.2–15.5 (anl); 13.2–21.7 (min); mean $r^2 = 0.92$ (hourly)	a	2	h	> 1000	Uniform
(B) DeGaetano and Doherty (2004)	PM _{2.5}	$r > 0.85$ most sites	a	20	h	> 1000	Uniform
(C) Ito et al. (2004)	PM _{2.5}	0.26 < r < 0.95	a,t	3	d	170	Heterogeneous
<i>Philadelphia</i>							
(A) Burton et al. (1996)	PM _{2.5}	17.7–21.0 (anl); 0.70 < r < 0.96	a	8	d	> 100	Uniform
(B) Pinto et al. (2004)	PM _{2.5}	14.7–16 (anl); 0.82 < r < 0.96; 0.08 ≤ COD ≤ 0.16	a	5	d	> 60	Uniform
(C) Wilson and Suh (1997)	PM _{2.5}	0.80 < r < 0.96	a,i	6	d	> 140	Uniform

^aParticle aerodynamic diameter: ≤ 10 μm (PM₁₀) or ≤ 2.5 μm (PM_{2.5}).

^bAbsolute and relative measures of intraurban concentrations in μg/m³ with averaging period: minutes (min), hours (h), or 24-h days (d).

^cMonitoring site types: ambient (a), traffic (t), or industrial (i).

^dNumber of concurrent intracommunity sites.

^eAveraging times in minutes (min), hours (h), or 24-h days (d).

^fNumber of concurrent intracommunity samples.

^gIntraurban particulate concentration conclusion.

a coefficient of divergence and daily absolute concentration differences between sites when characterising intraurban homogeneity.

The literature reviewed indicates that monitoring a diverse range of sources may be a significant factor in findings of intraurban heterogeneity (Table 3). Studies of uniformity by source in terms of road-proximity have identified both homogeneous and heterogeneous spatial distributions of $PM_{2.5}$ and PM_{10} , depending on traffic source compositions, regional processes, localised sources, and methods of determining spatial uniformity (e.g., Fisher et al., 2002; Janssen et al., 2001; Kim et al., 2004; Kingham et al., 2000; Monn et al., 1997; Roorda-Knape et al., 1998). Source contributions to intraurban uniformity present a critical gap in the literature and more studies are needed to understand the complexities of source interactions at the intraurban scale.

A substantial number of samples (e.g., $n \geq 50$) may provide more accurate approximations of intraurban heterogeneity, especially when the number of concurrently monitored sites is low (e.g., $n < 5$). The potential for exposure misclassification has been shown to be greater in studies utilising a limited number of monitoring sites to represent population exposures (Kim et al., 2005; Monn et al., 1997). It may also be useful to shorten sample-averaging times to ≤ 24 -h as longer averaging times may not capture diurnal patterns that are important in characterising intraurban uniformity.

To date, a very small fraction of the air pollution and health literature employ any sort of intraurban framework for assessing or integrating intraurban exposure variations of particulates (e.g., Hoek et al., 2002a; Jerrett et al., 2005b). The most comprehensive study was performed by Jerrett et al. (2005b), who used geographic information systems to interpolate exposures over small areas in Hamilton, Canada. When socio-economic, demographic, and lifestyle factors were included in the analysis, the health effects of particulate air pollution were reduced but not eliminated, suggesting that intraurban variations in PM were significantly associated with premature, all-cause, cardio-respiratory, and cancer mortality in small areas of Hamilton. There is still much work to be done in this area. A careful analysis of the effects of measurement error in health studies due to intraurban particulate heterogeneity would contribute significantly to our understanding (Ito et al., 2005; Wakefield, 2004).

It should be noted that this review has a number of limitations. First, we examined the spatial representativeness using the outdoor monitoring component of total personal exposure. Exposure in both time and space is a complex problem for modellers and health experts, and outdoor exposure to pollutants represents only a fraction of personal integrated exposure, as individuals move through multiple microenvironments and participate in various activities that may affect their

daily and long-term exposure to particulates. We chose to focus on outdoor levels specifically because: (i) studies conducted using outdoor monitoring are most abundant in the literature, (ii) time series epidemiological research typically uses outdoor exposures for population exposure, (iii) indoor levels are considered personal and not subject to external regulation, and (iv) in many situations, indoor levels can be inferred from outdoor levels (Monn, 2001). When extensive spatial monitoring is not possible, one suggested solution for reducing costs is the use of models. Atmospheric models have recently been used in exposure assessments to predict pollution levels down to the sub-neighbourhood level, but are highly dependent on emission inventories, which may not be available in some areas (Pearce et al., 2005). Jerrett et al. (2005a) recently reviewed intraurban exposure assessment using six different classes of models, concluding that hybrid models incorporate monitoring data may be the best suited for estimating concentrations at the intraurban scale. Based on our review, we suggest that intraurban hybrid models should include relative methods that account for differences in concentrations (e.g., coefficient of divergence) rather than correlations alone, when comparing sites. Finally, the definition of uniformity is somewhat nebulous in the literature, when it is defined at all, complicating the task of categorising study conclusions as uniform or heterogeneous. In several studies reviewed, authors make conclusions about homogeneity or heterogeneity with little attempt to quantify or classify their conclusions (i.e., ‘correlations were high between monitoring sites, indicating uniformity’). There is no formal regulative or scientific definition of what constitutes uniformity at the intraurban spatial scale, or at any other scale. The 20% figure of absolute and relative differences used in this paper was merely a means of classifying studies for comparative purposes. This figure is not applicable in all situations as homogeneity is partially a function of particle size fraction, scales, and types and densities of sources, among other factors.

It is important to note that not all cities exhibit heterogeneous distributions of particulate concentrations. This review does not intend to challenge the exposure assumptions of *all* health studies using central monitoring sites, as the uniformity assumption is valid in urban areas with homogeneous spatial distributions of PM. For example, take the case of Philadelphia, a city with considerable particulate intraurban uniformity. Burton et al. (1996) concluded that “ $PM_{2.5}$ and PM_{10} concentrations were found to be relatively uniform across Philadelphia, suggesting that concentrations measured at a single monitoring site are able to characterize particulate concentrations across Philadelphia and other similar urban areas well.” We do not disagree with Burton et al.’s (1996) initial assertion about the uniformity of PM in Philadelphia, as our

review of other studies conducted in Philadelphia also concluded relative uniformity for the study area (Table 4). The validity of the uniformity assumption in Philadelphia may partially explain the assumption's widespread application elsewhere in the literature, as it was one of the earlier studies on heterogeneity of particles. However, we emphasize that the homogeneous distribution of intraurban PM_{2.5} in Philadelphia and its applicability to population exposures should be applied *only* to Philadelphia, and is not necessarily valid in other 'similar urban areas' as well. Other cities in the region and around the world demonstrate that PM_{2.5} and PM₁₀, as well as other size fractions, may be homogeneous or heterogeneous, depending on the local conditions.

This review highlights several conclusions about the spatial heterogeneity of intraurban particulate concentrations and the resulting implications for epidemiological studies. First, correlation coefficients are not associated with absolute uniformity of particulates between sites at the intraurban scale and should only be applied in conjunction with more descriptive relative measures, such as coefficients of divergence when characterising intraurban concentrations. Second, while larger particle concentrations and ultra-fine particle numbers are generally more heterogeneous, uniformity does not conform to a fixed set of absolute assumptions from one urban area to the next (e.g., 'PM_{2.5} concentrations are uniform across cities'). Third, in order to more accurately characterise the nature of intraurban particulate concentrations, a sufficient number of monitoring sites (e.g., $n > 4$) and concurrent samples (e.g., $n \geq 50$) should be included in any proposed study, with sufficiently short averaging times (≤ 24 -h). Lastly, the intraurban spatial homogeneity of particulates in an area should be ascertained before applying central monitoring site data as a proxy for population exposure in order to minimise exposure misclassifications and relative risk uncertainties, especially in long-term cohort epidemiological study designs.

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