SPATIAL VARIABILITY OF INTRAURBAN PARTICULATE AIR POLLUTION: EPIDEMIOLOGICAL IMPLICATIONS AND APPLICATIONS

A thesis

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by

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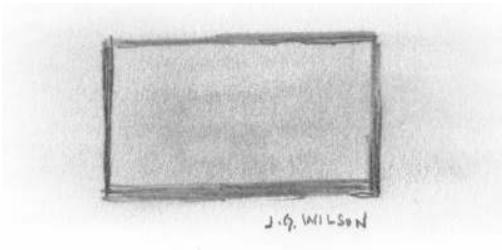
Frontispiece

What is the colour of the wind?

(11th c. Japanese Zen Koan)

…



...the colour of the wind...

(21st c. interpretation by the candidate)

Contents

Abstract

The past twenty years of research that has associated air pollution with health outcomes has brought remarkable advance in statistical techniques that effectively tease out the intricacies of the relationship. However, while statistical techniques progressed, an assumption based on seminal work in the field persisted: that concentrations of particulate matter (PM) air pollution are spatially homogeneous within urban areas, and consequently, that personal exposures could be based on central monitoring site data alone. Although this assumption went unaddressed for years, it has now come to researchers' attention that it may be flawed and that the assumption may induce exposure misclassification error under certain conditions. This thesis explores intraurban spatial variability in PM through a systematic review of the literature, experimental field testing, modelling, and new methodological approaches. The key outcomes of the thesis are as follows: (i) the publication of the first systematic review of the intraurban particulate literature, challenging the widely-held assumption that PM concentrations are spatially uniform; (ii) an experimental test was conducted in Christchurch, New Zealand, revealing that the homogenous assumption was false for a city with high wintertime particulate matter concentrations; (iii) an integrated meteorological-emission model was evaluated for the first time at the intraurban level for PM and a new study design was suggested; and (iv) the spatial modification effect of social and ecological confounders was analysed with respect to respiratory hospital admissions and PM. Collectively, these outcomes provide a new body of knowledge informing researchers focused on assessing the relationship between air pollution and health in applications ranging from small-area exposure assessment to the wider field of environmental epidemiology.

Chapter 1: Introduction

1.1. Introduction

Let us begin by stating an assumption: that concentrations of particulate matter air pollution are spatially homogeneous within urban areas. Most epidemiological studies that statistically associate particulate matter air pollution exposures with health outcomes make this assumption, and by inference assume that, within a given city, exposures to particulate air pollution are also without significant variability. This inference forms an exposure framework for some of the most significant research in this field published in the last 20 years. For example, the American Cancer Society (ACS) cohort study, widely recognised as among the most influential and widely cited pieces of work in this area (Jerrett *et al.* 2005b), states the exposure assumption without critique:

> …personal exposures to combustion source particulate air pollution [were estimated by taking the] median fine particulate concentration … of the average of values from all monitoring sites in a city (Pope *et al.* 1995b).

Another seminal cohort study, the *Harvard Six Cities Study,* utilises a similar approach:

> …ambient (outdoor) concentrations of total suspended particulate matter…were measured in each community at a centrally located air-monitoring station.
> (Dockery *et al.* 1993b).

Although these two papers have been cited over 2,000 times, the effects of their exposure assumptions on risk estimates have rarely been questioned and even less often been objectively critiqued (Hoek *et al.* 2002a, Dominici *et al.* 2003, Jerrett *et al.* 2005b, Wilson *et al.* 2005). It is the objective of this thesis to investigate why this assumption may be questioned and further, to explore the implications and applications of intrauban particulate matter spatial variability related to the field of epidemiological research.

This thesis will critically assess the validity of the homogeneous concentration assumption in the following ways: (i) the literature that both support and refute this assumption will be systematically reviewed and the implications of the assumption will be discussed; (ii) the assumption will be empirically tested with field experiments; (iii) new study designs based on intraurban modelling approaches will be evaluated and tested; and (iv) the effects of intraurban particulate matter variability on spatial analysis of disease will be quantified. We begin by placing this work in its wider context.

1.1.1. A brief history of air pollution

Since the first human ancestors used solid fuel for domestic cooking and heating, anthropogenic air pollution has existed. It is known that early people were exposed to combustion from biomass material used for cooking and heating as the evidence can be found in mummified lung tissues (Brimblecombe 1999). As civilisation organised over the course of the millennia, the first great cities emerged and with them the outdoor urban air pollution problem. The residents of early Rome were acutely aware of air pollution, what they referred to as *gravioris caeli* (heavy heaven) or *infamis aer* (infamous air) (Brimblecombe 1999). Throughout early history, wood and dung were the primary sources of biomass fuel for heating and cooking (Brimblecombe 1999). Then, in the mid 13th century, an important shift occurred. London had grown to a point where it had consumed its forest resources to the point of depletion and shifted to coal as the primary source of energy, launching what would become 700 years of pollution (The Royal College of Physicians of London 1970, Brimblecombe 1999). In 1257, Queen Eleanor of Provence was forced to vacate Nottingham Castle as a result of the 'fouls of the air' caused by the coal fires, and by 1285 King Edward I had established a commission to remedy pollution (Markham 1994). The problem persisted for centuries, and was even chronicled in popular literature of the day:

> I asked him whether there was a great fire anywhere?
> For the streets were so full of dense brown smoke that scarcely anything was to be seen.
> 'O dear no, miss,' he said. 'This is London particular.'
> 'A fog, miss,' said the young gentleman.
> 'O indeed,' said I. -*Bleak House* (Dickens 1881)

The so-called 'fog' in London, often romanticised, developed a more sinister connotation in December 1952, when a temperature inversion set up over the city for several days, leading to the most infamous modern-day air pollution episode – now widely known as the 'killer fog' (Ministry of Health 1954). Levels of air pollution (black smoke) in the County Hall district of London in December 1952 measured as high as 4,460 μg m^{-3} for a 24-hr period, almost 90 times modern-day ambient air health guidelines for particulate matter (Anderson 1999, WHO 2000).

The air pollution episode in London and other similar occurrences in the Meuse Valley of Belgium and in Donora, Pennsylvania (United States) in the early to middle 20th century collectively provided the impetus for what would be clean air legislation (Firket 1936, Schrenk *et al.* 1949, Ministry of Health 1954). The United States Congress introduced the Clean Air Act in 1963 (amended in 1971 and in 1990), one of many early coordinated attempts to measure, manage, and reduce urban air pollution. As a result of widespread legislation and management of emissions, levels of outdoor air pollution from the 1970s decreased substantially in major developed-country cities, but problems persist today in both the developed and developing world. It is now estimated that almost a quarter of the global urban population (1.4 billion persons) are exposed to air pollution exceeding health guidelines every day (WHO 2005). The World Health Organisation, the United Nations, and other multinational development organisations have come to terms with addressing the air pollution problem, which they have deemed a global priority. The reduction of indoor air pollution was hence a component of the United Nations' Millennium Goals, alongside such aims as halting the spread of HIV/AIDS and halving world hunger (United Nations 2005).

1.1.2. Air pollution and health

From the earliest times, urban air pollution was understood to be a potential factor associated with health problems. The first known descriptions of associations between air pollution and health are found in the *Hippocratic Corpus*, as described in the book *Air, Water and Places* (Brimblecombe 1999). Despite the early recognition of the adverse health effects of air pollution, fossil fuel-based transport and industrialization during and after the industrial

revolution led to the sharp rise of air pollution concentrations in many urban areas (Bell *et al.* 2004). The acute air pollution events and associated mortality in Donora, Pennsylvania, the Meuse Valley, and London in the early to middle twentieth century mark the beginning of widespread public health concerns caused by high levels of air pollution (Samet & Jaakkola 1999). The great black smog of London in early December 1952 had a most severe health impact: approximately 4,000 excess deaths were reported, with the figure quoted as high as 12,000 when long-term effects were considered (Anderson 1999, Bell & Davis 2001).

Whilst punctuated episodes of poor air quality are clearly harmful, it is now increasingly accepted that relatively low levels of air pollutants – to which people are exposed outdoors and indoors, during and between pollution episodes – play a significant role in population health in addition to higher levels of exposure incurred during air pollution episodes (Samet 1987, Jones 1999). In the 1950s, statistical epidemiological studies began to emerge that would assess the more subtle linkages and associations between air pollution and health at lower levels of air pollution (Bell *et al.* 2004). As a result of increasingly sophisticated techniques, a robust body of literature on the health effects of air pollution has amassed. A recent PubMed search (February 2006) on the key words 'air pollution' and 'health' returned nearly 10,000 peer-reviewed journal articles, including 1,200 review articles, on the subject.

1.1.3. Particulate matter air pollution

Although many air pollutants are known to be associated with adverse health effects (e.g., NO_x, SO_2, O_3, Pb), particulate matter (PM) air pollution is the most commonly studied air pollutant with respect to health effects (Brunekreef & Holgate 2002). PM is not a specific chemical entity but refers to a suspension of solid, liquid or a combination of solid and liquid particles in the air (Hinds 1999) and is one of six 'criteria pollutants' designated by the U.S. 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 (mostly combustion-related) and natural sources (e.g., sea salt, dust, spores).

Guidelines for PM, as with most monitored air pollutants, are based on health effects. The United States Environmental Protection Agency's (USEPA) model for setting National Ambient Air Quality Standards (NAAQS) is similar to those used in many areas of the world, including Europe and Australasia. Specifically, regulating bodies such as the US EPA, or in New Zealand, the Ministry for the Environment (MfE), are required to list air pollutants that reasonably may be expected to endanger public health or welfare, to issue air quality criteria for them that take into account the latest available scientific information, and to set guidelines to protect human health with adequate margin of safety (Ministry for the Environment 2002, US EPA 2004). As hundreds of papers are published every year on PM, the guidelines are reviewed and accordingly adjusted periodically. The US EPA has recently revised its guidelines for PM for differing particle diameter size classes. For particulate matter smaller than 10 micrometers in aerodynamic diameter (PM_{10}), which is the focus particle size range for this thesis, the US EPA has set a 24-hr maximum exposure guideline value of 50 $\mu g\ m^{-3}$, which is also the guideline value in New Zealand (Ministry for the Environment 2002).

While policy-making bodies strive to reduce ambient concentrations to levels below guidelines, it should be noted that these guidelines are not equated with 'safe levels' of particulate air pollution. Research has shown that there is no established low-end threshold for particulate air pollution, and accordingly, the WHO has set no zero-effect guideline for health effects associated with ambient long-term exposure to PM (Schwartz 1994, WHO 1999). The cumulative global health impacts of PM exposures continue to be significant: recent research estimates that 800,000 annual premature deaths globally are associated with PM air pollution exposures in excess of health guidelines (Cohen *et al.* 2005).

1.1.4. Human exposure to air pollution

Human exposure may be defined as 'an event that occurs when a person comes in contact with a pollutant' (Ott 1982). Pragmatically, epidemiological studies usually calculate an 'average exposure' value by dividing total pollutant exposures by a given period of time (e.g., expressed as a daily or annual mean) (Monn 2001). In air pollution epidemiology, the unit 'concentration' is most commonly used when speaking of average exposures (Monn 2001). Of critical

importance to any environmental epidemiological work is determining an accurate exposure estimate for the population or individual. The almost universal approach for estimating exposure to air pollution in these studies involves an exposure value or exposure variation measured at a central site that is applied to the entire population of the study area (e.g., Dockery *et al.* 1993b, Samet *et al.* 2000, Pope *et al.* 2002). A necessary assumption, then, is that PM, especially smaller particulates, are homogeneously distributed within large urban areas and that the concentrations between sites are well correlated. It has been suggested that the use of data from a central ambient monitoring site as a proxy for personal exposure may present an exposure misclassification that can lead to bias in estimates of health effects of air pollution (Dominici *et al.* 2003). Exposure misclassification, the discrepancy between the true value of a personal exposure and its measured or assumed estimate, is a well recognised and inherent limitation of ecological studies involving the environment and disease (Armstrong *et al.* 1992, Thurston 1996).

Despite the development of new spatial and analytical techniques such as Geographic Information Systems (GIS), dispersion models, advanced spatial statistics, and integrated hybrid models, many epidemiological studies continue to operate under the spatial homogeneous distribution assumption, especially for particulate air pollution (Huang & Batterman 2000a, 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 (Kukkonen *et al.* 2001, Brunekreef & Holgate 2002, Sajani *et al.* 2004, Jerrett *et al.* 2005a). This thesis will examine the homogeneous assumption at the intraurban level as well as the possible implications and applications of intraurban variability.

1.1.5. Origins of the intraurban homogeneity assumption

The fact that the intraurban homogeneous assumption has persisted presents a bit of a mystery, given the assumption would, at least upon initial consideration, appear clearly flawed. The appearance and persistence of the assumption is likely based on several factors – both general methodological issues and an explicit conclusion made in a specific article on intraurban

variability. The first set of factors might be described as 'methodological precedence.' Although time series analyses of air pollution and health have existed since the 1950's with the work of John W. Tukey (Brillinger *et al.* 2002), the multi-city cohort analyses did not emerge to widespread prominence until the early 1990s with the Harvard Six Cities and American Cancer Society Studies (Dockery *et al.* 1993b, Pope *et al.* 1995b). The methodological precedent – that of utilising central sites – was initially proposed in these seminal cohort studies (as described in Section 1.1), and has persisted throughout the 1990s and into the current era. The strength of the argument for the assumption was enhanced by an article published in the journal *Environmental Science and Technology* in 1996 by a team of researchers from the US EPA and the Harvard School of Public Health (Burton *et al.* 1996). The study 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** (Burton *et al.* 1996:400 - emphasis added).

Further, in the conclusion, the paper states that:

> A single stationary ambient monitor may be sufficient to estimate outdoor PM_{10} and $PM_{2.5}$ concentrations for urban populations living in the northeastern United States (Burton *et al.* 1996: 406).

Subsequent work in Philadelphia (Wilson and Suh 1997, Pinto *et al.* 2004) has shown that the uniformity assumption is indeed valid in Philadelphia. The fact that concentrations are homogeneous in Philadelphia likely contributed to the further expansion of the assumption in the literature. However, it should be emphasized that the fact that spatial concentrations of intraurban PM_{10} and $PM_{2.5}$ *are* homogeneous in Philadelphia, the applicability of local assumptions to other urban areas, even regionally in the north-eastern United States, is highly questionable.

1.2. Thesis hypotheses and objectives

While the overall objective of this thesis is to evaluate the implications and applications of intraurban particulate matter concentrations, this thesis tests several hypotheses related to intraurban spatial variability PM:

1.2.1. Hypothesis one – that PM air pollution may be spatially variable at the intraurban scale and that this variability has significant implications for epidemiological studies;

Epidemiological studies typically utilise one or few central monitoring stations as a proxy for personal exposure to particulate matter air pollution over extensive areas. However, recent research indicates that central monitoring sites may not accurately characterize the spatial complexities of the particulate field across an urban area (Pinto *et al.* 2004). This hypothesis will be tested with a focus on monitored-data studies at the intraurban spatial scale. Comparative analyses of the literature by particle size fraction, method of determining heterogeneity, and sampling characteristics will be performed in order to ascertain the validity of the homogeneous particulate matter distribution assumption. It is expected that portions of the literature will provide contradictory conclusions as to the homogeneity of intraurban particulate concentrations. It is further anticipated that the error resulting 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 may differ depending on study design and parameters. The review will also ascertain the types of study designs most at risk of misclassification error, as well as addressing techniques for mitigating the risk of false assumptions about intraurban PM variability.

1.2.2. Hypothesis two – that intraurban variability of PM may be empirically examined in the field;

There has been little work testing the hypothesis that central fixed site PM concentrations are not representative of wider intraurban variability using actual monitored data in cities with high levels of pollution (Pinto *et al.* 2004). Hypothesis two will examine daily concentration variations in particulate matter less than 10 micrometers in diameter (PM_{10}) at the intraurban

scale in Christchurch, New Zealand, a city with high winter pollution concentrations. Daily concentrations of PM_{10} data will be collected for two winter months at ten background monitoring sites and a central fixed monitoring site typically used for estimating exposure in local epidemiological studies (e.g., McGowan *et al.* 2002). Innovative methods, including a coefficient of divergence, will supplement correlation coefficients in order to ascertain whether absolute concentration differences are significant. It is expected that significant differences in particulate concentrations between the central site and background sites will be found, and that these variations may have implications for epidemiological studies conducted in the area.

1.2.3. Hypothesis three – that PM intraurban variability may be estimated using atmospheric dispersion models and that these models present new opportunities for epidemiological study designs;

Many study areas do not have spatially dense monitoring data available. In these areas, advanced integrated meteorological-emission (IME) models may offer one solution to the problem of accurately characterising intraurban particulate concentrations across an area. PM_{10} concentrations will be modelled and evaluated using a dense intraurban monitoring network in Christchurch, New Zealand. It is expected that despite the area's high intraurban concentration variability, and meteorological and topographical complexity, the model will perform satisfactorily. It is also expected that, given that if the model performs satisfactorily, that IME models may be a viable alternative to central monitoring sites for estimating personal exposure in longer-term (monthly or annual) cohort epidemiological studies. This method would allow the reduction of error associated with differing particle composition profiles between cities in multi-city cohort studies.

1.2.4. Hypothesis four – that methodological approaches may be developed to effectively analyse the spatial modification of risk related to PM intraurban variability.

Ecological studies that examine the relationship between exposure and health outcomes often address confounding factors by controlling for them as covariates. Although many studies have

explored how confounding variables such as social deprivation modify the exposure-response relationship and risk outcomes, there has been limited investigative work on spatial modifications of risk at the sub-urban scale. As epidemiological studies continue to focus on increasingly smaller scales through the use of geographic information systems, it is important that these spatial modifications be well understood. It is expected that analysis of spatial modification of relative risk and clusters of respiratory disease may be a useful tool for informing researchers as to the nature of social confounders.

1.3. Thesis intent, implications, structure and overview

1.3.1. Intent and focus of thesis

The focus of this thesis is explicitly on methodologies for identifying patterns of particulate matter air pollution and not necessarily the underlying processes contributing to those patterns. Various studies in the literature have used regression and other methods to ascertain contributing factors to pollution concentration, such as pollutant traffic sources (Briggs et al. 2000) and land use type (Chan et al. 1997). However, the intent here is to describe methods for understanding the nature of spatial patterns in particulate matter air pollution and/or related disease surveillance (see Chapter 5) – not the underlying processes or causes of the spatial patterns and variability.

1.3.2. Theoretical relationship to health geography and environmental justice

This thesis, although arguably placed in the wider theoretical context of exposure assessment and environmental epidemiology, may have implications and applications in the subdiscipline of health geography as well as the study of environmental justice issues. Over the past 15 years, health geography has evolved from a focus on disease and the interests of the medical world to a subdiscipline towards a focus on broader social models of health and health care (Kearns 1993, Kearns and Moon 2002). One of the social-place topics of interest related to health geography that has received much attention lately is the exploration of differential distribution of exposures by socioeconomic position, or environmental justice. More succinctly, environmental justice may be defined as the 'equal access to clean environment and equal protection of issues of environmental harm irrespective of race, income, class or any other differentiating feature of socioeconomic status' (Cutter, 1995). Several studies have recently assessed particulate matter

air pollution as a modifier on health outcomes along social exposure gradients. O'Neill *et al.* (2003) recently reviewed the topic along a three-pronged hypothesis framework: (i) that groups with higher deprivation may receive higher exposure to air pollution; (ii) that because highly deprived groups already experience compromised health status due to material deprivation and psychological stress, they may be more susceptible to the health effects of air pollution; and (iii) because of the combination of greater exposure and susceptibility, these groups are likely to suffer greater relative heal effects from air pollution than their less deprived counterparts. Buzzelli *et al.* (2003) related total suspended particulate (TSP) to socioeconomic status at the census area unit resolution in Hamilton, Canada. The results indicated that environmental injustice persisted but becomes less pronounced over time. In related work, Jerrett *et al.* (2004) examined the modification effect of education and employment sector on the association between air pollution and mortality in Hamilton, finding that areas where the population was less educated and employed in manufacturing occupations presented elevated risk levels. In Los Angeles County, Wilhelm and Ritz (2005) related local variations in carbon monoxide and particulate matter air pollution with adverse birth outcomes. In an environmental justice study in Christchurch, New Zealand, Pearce *et al.* (2006) recently found that more disadvantaged groups were exposed to higher levels of particulate air pollution, although health outcomes were not analysed. In order to understand the nature environmental injustice, one must first have an appreciation for the variability in exposure between different spatial areas where the subjects under study are located in an urban area – this thesis sets out to further understand the nature of intraurban exposures, and thus, has implications for the environmental justice body of knowledge.

1.3.3. Academic papers

This doctoral thesis was completed by preparing a series of four academic papers which are in various advanced stages of the peer-reviewed publication process. Table 1.1 outlines the four papers with their respective authors, targeted journals for publication, and current status at the time of printing of this thesis. The content within each published paper is in effect the same as that found in the chapters of this thesis with the exception Chapter five, which was under review at the time of submission. Some minor changes in the introduction and discussion have been

added to this particular Chapter, but the methods, the data set, and results are identical to the version currently under review. In accordance with the University of Canterbury standards for completion of thesis by papers, the candidate is the first author and primary contributor to all four papers. The candidate was responsible for no less than 85% of the analysis and for no less than 95% of the manuscript writing for any of the papers.

Table 1.1. Thesis chapters, paper titles, authorship, target journals, and current publication status for each of the four journal articles produced for this thesis.

Ch	Paper title (paper designation)	Authors	Journal	Status
2	A review of intraurban variations in particulate air pollution: Implications for epidemiological research (paper A).	Wilson, Kingham, Pearce & Sturman	Atmospheric Environment	2005; **39**, 6444-6462.
3	Intraurban variations of particulate matter air pollution in Christchurch, New Zealand: Implications for epidemiological studies (paper B).	Wilson, Kingham & Sturman	Science of the Total Environment	2006; forthcoming
4	Intraurban-scale dispersion modelling of particulate matter concentrations: Applications for exposure estimates in cohort studies (paper C).	Wilson & Zawar-Reza	Atmospheric Environment	2006; **40**, 1053-1063.
5	The spatial modification effect of environmental and social confounders on intraurban patterns of risk in respiratory hospital admissions (paper D).	Wilson, Sabel, Kingham, Tisch, & Epton	Social Science and Medicine	under review.

1.3.4. Additional contributions by the candidate

It should also be noted that the candidate has undertaken several additional pieces of work beyond the publications included in this thesis. Seven additional peer-reviewed publications were initiated during the PhD programme (Table 1.2). The papers all incorporate aspects of spatial epidemiology or air pollution exposure analysis and are in various stages of the publication/development process. Five of the papers have been accepted for publication.

Table 1.2. Additional peer-reviewed work published or in progress by the candidate associated with the doctoral work.

No.	Title	Authors	Journal	Status
1	Dispersion modelling of particulate matter concentrations at the intraurban scale: epidemiological applications.	Wilson & Zawar-Reza	Proceedings of the 10th Int'l Conf. Harmonisation within Atmospheric Dispersion Modelling	2005; 509-13.
2	Intraurban particulate air pollution and restricted activity days in New Zealand school children.	Wilson, Kingham, Sturman, & Pearce	Epidemiology (reviewed abstract)	2005; **16**, S136-S136.
3	Winter comparison of TEOM, MiniVol and DustTrak PM_{10} monitors in a wood smoke environment.	Kingham, Durand, Harrison, Wilson & Epton	Atmospheric Environment	2006; **40**, 338-347.
4	Spatial analysis of respiratory disease clusters on an urbanised geothermal field.	Durand & Wilson	Environmental Research	2006; forthcoming; online.
5	Ball lightning and fireballs during volcanic air pollution.	Durand & Wilson	Weather	2006; **61**, 40-43.
6	Dry deposition of particulate matter air pollution in an urbanised native bush environment.	Wilson, Zawar-Reza & Cavanaugh	Atmospheric Environment	under review.
7	Error in restricted activity days estimates due to intraurban variability in particulate matter.	Wilson & Kingham	Environmental Monitoring and Assessment (target journal)	in final preparation.

1.3.5. Structure

The structure of the thesis is as follows. Chapter two focuses on laying out the general hypotheses of the thesis (as discussed in section 1.2 above) and provides a thorough review of the literature pertaining to intraurban particulate concentrations. The hypotheses presented in Chapter two are then revisited and verified through empirical field testing in Chapter three. In Chapter four, a model for a new study design (the intraurban cohort study based on an integrated meteorological-emission model) is proposed and tested. Finally, in Chapter five, the results and conclusions derived from Chapters two, three, and four are applied in such a way as to suggest a new way to ascertain the relative effects of various ecological confounders on health outcomes. Chapter six provides a brief summary of the contributions of the thesis. The general structure and flow of the thesis is summarised in Figure 1.1.

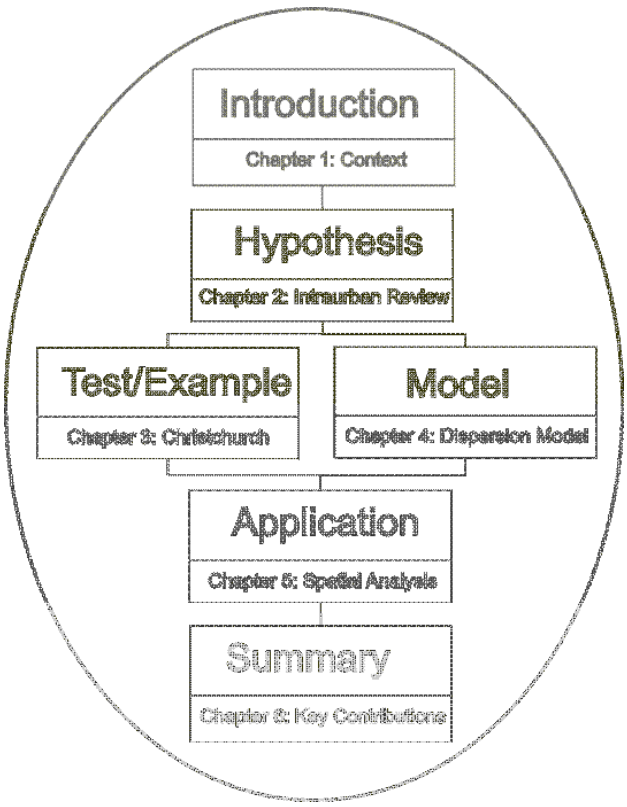


Figure 1.1. Outline of thesis and conceptual linkages.

Chapter 2: A review of intraurban particulate concentrations: implications for epidemiological studies.

2.1. Summary

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 chapter 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.

2.2. 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, Schrenk *et al.* 1949, Ministry of Health 1954). 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 health effects were present even at levels below ambient guidelines for pollution, especially particulate exposure (Pope *et al.* 1992, Dockery & Pope 1994, Brunekreef *et al.* 1995, Mage *et al.* 1999). 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 (Vedal *et al.* 2003, Bell *et al.* 2004). 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., Pope *et al.* 1992, Dockery *et al.* 1993a, Samet *et al.* 2000, Wong *et al.* 2001, Pope *et al.* 2002, 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 & 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, Zhu *et al.* 2002, Ito *et al.* 2004, Pinto *et al.* 2004, Kim *et al.* 2005). 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.* 1995, Ito *et al.* 2005). 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 &

Batterman 2000b, 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 (Kukkonen *et al.* 2001, Brunekreef & Holgate 2002, Sajani *et al.* 2004, Jerrett *et al.* 2005a).

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 (Pope *et al.* 1991, Pope *et al.* 1992, Dockery & Pope 1994, Pope *et al.* 1995a, Moolgavkar & Luebeck 1996, Samet *et al.* 2000). A PubMed search (Mar 2005) containing the keywords "health effects" and "health" with combinations of "particulate air pollution", "PM_{10}" and "$PM_{2.5}$" yielded approximately 1,200 journal articles written in English on the subject since 1991, when Pope (1991) published his 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 (Zeger *et al.* 2000, Brauer *et al.* 2002, Dominici *et al.* 2003, Pinto *et al.* 2004).

The aim of this chapter 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.

2.3. 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 U.S. 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} ($d_a \leq 10\mu m$) and $PM_{10\text{-}2.5}$ ($2.5\mu m \leq d_a \leq 10\mu m$). Smaller fractions are $PM_{2.5}$ ($d_a < 2.5\mu m$), PM_1 ($d_a < 1\mu m$), also called fine particles, and UFP, or ultrafine particles ($d_a < 0.1\mu m$). Total suspended particulate, or TSP, is used less frequently today, and represents particles $d_a < 100\mu m$. 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 $PM_{2.5}$, 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 μm) (Schwartz *et al.* 1996). PM_{10} and $PM_{2.5}$ 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 10μm 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 μg 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 (Peters *et al.* 1997, Englert 2004).

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 1.0μm, 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 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 x 10^{-5} cm, for PM_1 3.5 x 10^{-3}cm, and for PM_{10} 0.23 cm (Willeke & 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 ($PM_{2.5}$ and PM_1) are relatively more uniformly distributed than for coarser fractions (Brimblecombe 1986, Wilson & Suh 1997, Monn 2001). 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 (Chow *et al.* 1994, Ito *et al.* 1995, Adgate *et al.* 2002, Bari *et al.* 2003). 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).

2.4. 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, Thomas *et al.* 1993, Dominici *et al.* 2003). 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 (Bell *et al.* 2004), which we focus on below.

2.4.1. 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 (Kunzli *et al.* 2001, Bell *et al.* 2004). 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 (Schwartz *et al.* 1996, Janssen *et al.* 1998, Schwartz & Levin 1999,

Schwartz 2000, 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 (Selvin 1958, Richardson *et al.* 1987, Piantadosi *et al.* 1988, Greenland & Morgenstern 1989, Nurminen *et al.* 1999, Nurminen & Nurminen 2000, Sheppard 2002, Dominici *et al.* 2003, Wakefield 2003, Berhane *et al.* 2004, Wakefield 2004).

2.4.2. 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 (Greenland 1992, Sheppard & Prentice 1995, Houthuijs *et al.* 2001, Wakefield & Salway 2001, Berhane *et al.* 2004). 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 2.1 outlines epidemiological study designs and associated possibility of errors associated with exposure misclassification for the time-series and cohort study designs.

Table 2.1. 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 & 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.* 1993a); (Pope *et al.* 2002); (Gauderman *et al.* 2002)	Increased

2.5. Review of monitoring-based studies

2.5.1. 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 categorized in accordance with the 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 categorized as 'homogeneous', while those that report greater than 20% variation are categorized 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 the 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).

2.5.2. Intraurban concentration studies

In the next section of this review, we focus on studies that examine intraurban variations in PM. The following criteria guided the 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.

2.5.3. 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 PM_{10}, $PM_{2.5}$ 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 of a high range for PM_{10} ($0.62 < r < 0.96$) 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 & Suh (1997) found high site-to-site correlations ($0.80 < r < 0.96$) for 24-hr $PM_{2.5}$ concentrations at the intraurban scale that were correlated to population density. However, correlations were slightly lower for PM_{10} ($0.79 < r < 0.96$) 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 course 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 μg m^{-3} and monthly concentrations were 13.2 and 21.7 μg 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 D.C. metropolitan area. Correlations were high and significant ($p < 0.05$) 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 Total Suspended Particulate (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 nine-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 μg m^{-3} and 64.6 μg m^{-3} with a high correlation value between sites (r^2=0.94), suggesting regionally homogeneous sources. Researchers in Vancouver measured PM_{10} at eleven 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 one to thirty 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 3.3km of each other found relatively homogeneous PM_{10} mass concentrations ranging from 27.6 to 32.0 μg m^{-3} (Röösli *et al.* 2000). In a related study in the same city, researchers found PM_{10} mass concentrations to be uniformly distributed (+/- 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 micrometers in diameter) levels. However, outdoor levels were on average 9% lower than the fixed site (20.0 versus 21. μg 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 μg m^{-3}, but weekly values were much more variable (37 – 357 μg 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%).

2.5.4. 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 20-64% in the winter, and from 17-69% in the summer. Maximum average PM_{10} ambient concentration differences between types of ambient sites ranged from 26-34% in the winter and from 15-22% 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. Cyrys *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 three week period in El Paso, Texas. Average gravimetrically sampled PM_{10} concentrations between sites varied significantly (61-91 $\mu g\ m^{-3}$), while $PM_{2.5}$ concentrations were more uniform (17-20 $\mu g\ m^{-3}$). 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 U.S. 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 characterise 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:

$$COD_{jk} = \sqrt{\frac{1}{p}\sum_{i=1}^{p}[(x_{ij} - x_{ik})/(x_{ij} + x_{ik})]^2} \qquad (2.1)$$

where x_{ij} and x_{ik} represent the 24-hr average particulate concentration for sampling day i at sampling site 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 ($0.082 <$ max COD < 0.27), whereas cities in other areas of the country, especially in the west, showed higher degrees of spatial variation ($0.20 <$ 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, Missouri. $PM_{2.5}$ 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 COD ≥ 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 to 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 concluded that source-oriented evaluations of PM associated health effects should consider the uncertainty associated with spatial representativeness 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 $\mu g\ m^{-3}$ in Munich, 8 to 16 $\mu g\ m^{-3}$ in Stockholm, and from 14 to 26 $\mu g\ m^{-3}$ 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 17.8-28.8 $\mu g\ m^{-3}$ for particulates less than four micrometers in aerodynamic diameter (PM_4) and from 22.4-34.8 $\mu g\ m^{-3}$ for PM_{10}. Temporally non-concurring average ultrafine particle concentrations between the three sites in the city ranged

in number from 5,690/cm^3 to 19,300/cm^3. These differences were also attributed to variations in traffic densities between locations. Goswami *et al.* (2002) monitored $PM_{2.5}$ at forty 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 total suspended particulate (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 μg m^{-3}, 466 μg m^{-3} and 477 μg 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 one year at five sites within 15 km of the city centre of Brisbane, Australia. Average mass concentrations ranged from 18.9-37.8 μg 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.

2.6. 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.

2.6.1. 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 conclusions 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 2.2). For the purpose of comparison, we have again separated studies into homogeneous and heterogeneous groups based on their conclusions, as outlined in Section 2.5.1.

Table 2.2. Reviewed studies by absolute and relative methodological characteristics.

Conclusion / Study*	Particle d_a[1]	Absolute measure (averaging period) [2]	Relative measure[3]
Uniform (Homogeneous)			
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
Röösli 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)	$r^2 = 0.94$
Non-uniform (Heterogeneous)			
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	$1.86 \times 10^4 – 9.60 \times 10^4$ (d) cm^{-3}	
Hoek et al., 2002	$PM_{2.5}$	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, 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)
Sajani et al., 2004	TSP		$0.49 < r < 0.91$
Suh et al., 1997	$PM_{10-2.5}$		$0.34 < r < 0.48$
Sun et al., 2004	PM_{10}	184 – 292 (d, winter)	
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)

*Uniformity conclusion based on Section 2.5.1 and study name. [1]Particle aerodynamic diameter: particulate matter (PMx) or particle number (PN) count. [2]Absolute measure of concentration difference between sites in μg/m3 (PM) or cm-3 (PN) with averaging period: minutes (min), hours (h), 24-hr days (d), weeks (w), years (anl), or season. [3]Relative measure of intraurban concentration variation: correlation (r), regression (r^2), coefficient of variation (CV), or (COD) coefficient of divergence.

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 PM1) over urban areas, while UFP (da < 0.1μm) 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.

2.6.2. 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 2.2. Seventeen (52%) studies examined absolute concentration differences between sites, seventeen (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 2.1). 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 90^{th} percentiles of absolute concentration differences are appropriate. A study by the U.S. Environmental Protection Agency (2004) is illustrative of the

fact that correlation coefficients and absolute measures of concentration can yield different conclusions about uniformity. Figure 2.1 shows that concentrations at paired monitoring sites in three Metropolitan Statistical Areas (MSAs) were highly correlated, yet exhibited various ranges of concentration heterogeneity. Paired sites in Columbia, South Carolina; Chicago, Illinois; and Detroit, Michigan 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.

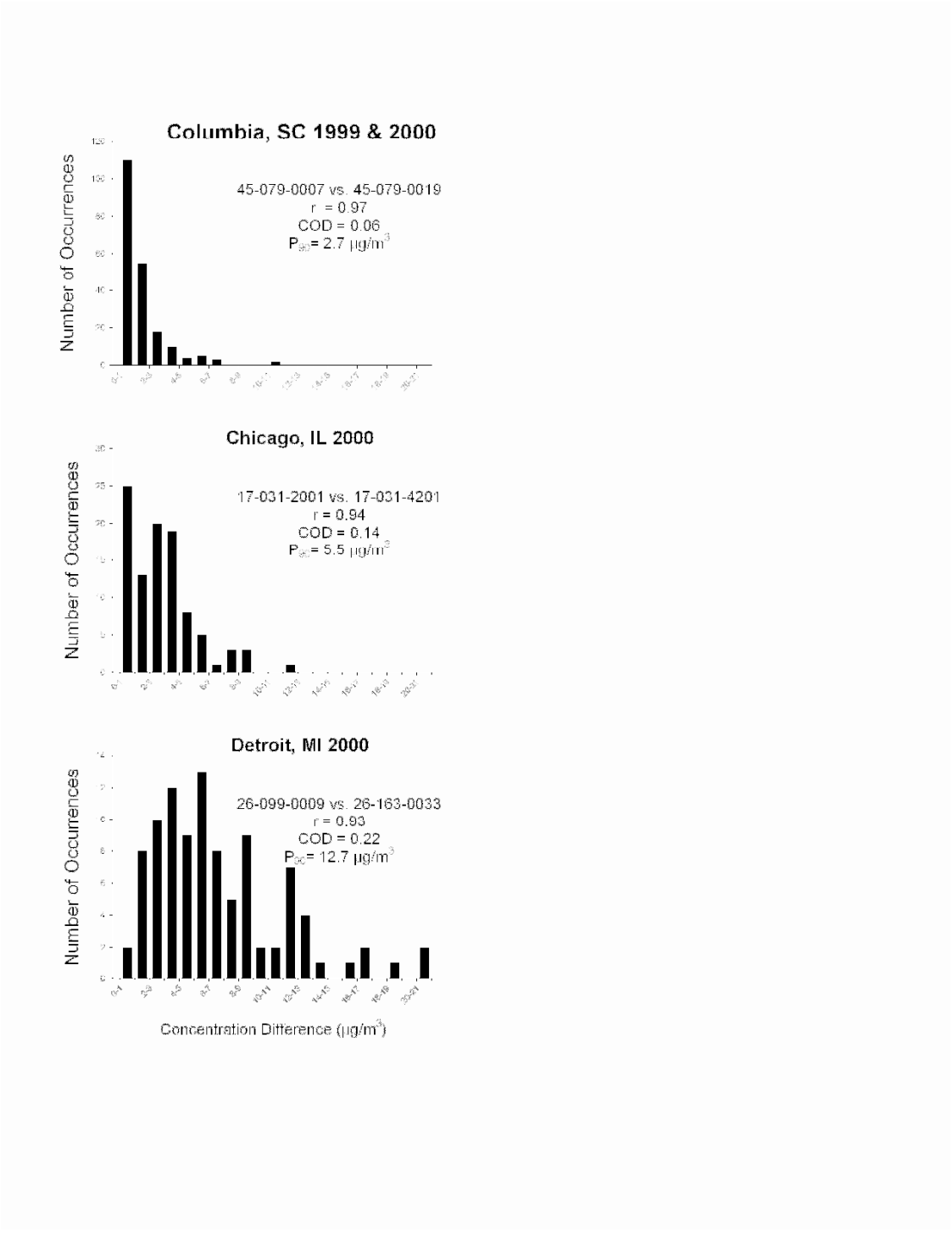


Figure 2.1. Three examples of well correlated site-pairs with coefficients of divergence (COD) and 90th percentiles of daily concentration differences (P90). Note that the y-axis scales are not uniform (Source: US EPA 2004).

2.6.3. 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, the 31 studies are reviewed by the types of sources monitored, the numbers of intraurban monitoring sites, the averaging times, and numbers of concurrent samples between sites (Table 2.3).

Table 2.3. Reviewed studies by quantitative site and sampling characteristics.

Conclusion / Study*	Particle d_a[1]	Site type[2]	Site N[3]	Avg. time[4]	Sample N[5]
Uniform (Homogeneous)					
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
Röösli 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
Non-uniform (Heterogeneous)					
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
Cyrys 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., 2002	$PM_{2.5}$	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, 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

*Uniformity conclusion based on Section 2.5.1 and study name. [1]Particle aerodynamic diameter: particulate matter (PMx) or particle number (PN) count. [2] Monitoring site types: ambient (a), traffic (t), or industrial (i). [3]Number of concurrent intra-community sites. [4]Averaging times are minutes (min), hours (h), 24-hr days (d), or weeks (w). [5]Number of concurrent intra-community samples.

Particulate sources are an important component in determining not only spatial heterogeneity, but also exposure classification and health effects (Wongphatarakul *et al.* 1998, Laden *et al.* 2000, Oglesby *et al.* 2000, Ito *et al.* 2004, Kim *et al.* 2005). 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, while 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 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 utilised 2-30 concurrently operating sites with a median of six sites, while studies finding heterogeneous concentrations utilised from 2-42 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-hrs. 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 with homogeneous findings, 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 twenty four (29%) of the studies finding heterogeneous concentrations had concurrent sample sizes of ≤ 20, while only two studies finding uniform particulate concentrations conducted twenty 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.

2.6.4. Comparison by study area

As a final step in the 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. Twelve studies were selected in four metropolitan areas: Basel, Switzerland; Los Angeles, New York City, and Philadelphia (Table 2.4).

Table 2.4. Comparison of intraurban studies conducted on same particle size fraction by city.

City/ Study	d_a[1]	Measurement (averaging period) [2]	Site type[3]	Site N[4]	Avg. time[5]	Sample N[6]	Conclusion[7]
Basel, Switzerland							
(A) Junker et al., 2000	PM_{10}	22.4 – 34.8 (27h)	a	3	min	17	Heterogeneous
(B) Monn, 1997	PM_{10}	CV = 13%	t	4	d	176	Heterogeneous
(C) Röösli et al., 2000	PM_{10}	27.6 - 32.0 (d)	a,t	4-6	d	52	Uniform
Los Angeles							
(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	$PM_{2.5}$	$0.07 \leq COD \leq 0.48$, $4.3 \leq P_{90} \leq 43.1$; 10.5 – 31.3 (anl)	a	4-5	d	>60	Heterogeneous
(C) Wongphatarakul et al., 1998	$PM_{2.5}$	$0.09 \leq COD \leq 0.572$	a,t,i	5	d	~60	Heterogeneous
New York City							
(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
Philadelphia							
(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); $0.82 < r < 0.96$; $0.08 \leq COD \leq 0.16$	a	5	d	>60	Uniform
(C) Wilson and Suh, 1997	$PM_{2.5}$	$0.80 < r < 0.96$	a,i	6	d	>140	Uniform

[1]Particle aerodynamic diameter: ≤ 10 μm (PM_{10}) or ≤ 2.5 μm ($PM_{2.5}$). [2]Absolute and relative measures of intraurban concentrations in $\mu g/m^3$ with averaging period: minutes (min), hours (h), or 24-hr days (d). [3] Monitoring site types: ambient (a), traffic (t), or industrial (i). [4]Number of concurrent intra-community sites. [5]Averaging times in minutes (min), hours (h), or 24-hr days (d). [6]Number of concurrent intra-community samples. [7]Intraurban particulate concentration conclusion.

In Basel, three separate studies of PM_{10} 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 between sites in study B were similar (20.2 - 24.8 $\mu g\ m^{-3}$), but COD values ranged from 0.14 - 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 sixty 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 $PM_{2.5}$ 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-hr 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 & 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 eight years, concluded that daily $PM_{2.5}$ 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, B and C (Burton *et al.* 1996, Wilson & Suh 1997, respectively, Pinto *et al.* 2004). 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.

2.7. Discussion and conclusion

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 particle 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 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 the review, it is recommended that correlation coefficients be used in conjunction with 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 2.3). 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., Monn *et al.* 1997,

Roorda-Knape *et al.* 1998, Kingham *et al.* 2000, Janssen *et al.* 2001, Fisher *et al.* 2002, Kim *et al.* 2004). 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 (Monn *et al.* 1997, Kim *et al.* 2005). It may also be useful to shorten sample averaging times to ≤ 24-hrs 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 intra-urban framework for assessing or integrating intraurban exposure variations of particulates (e.g., Hoek *et al.* 2002a, Jerrett *et al.* 2005c). The most comprehensive study was performed by Jerrett *et al.* (2005c), who used geographic information systems to interpolate exposures over small areas in Hamilton, Canada. When socioeconomic, 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 is 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 (Wakefield 2004, Ito *et al.* 2005).

It should be noted that this review has a number of limitations. First, the spatial representativeness is examined 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. The focus on outdoor

levels specifically was made 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.* 2006). 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 the review, it is suggested 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 chapter 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." The author does not disagree with Burton et al.'s (1996) initial assertion about the uniformity of PM in Philadelphia, as the review of other studies conducted in Philadelphia also concluded relative uniformity for the study area (Table 2.4). 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, it should be emphasized 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 PM_{10}, 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 ($\leq$ 24-hr). 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.

Chapter 3: Intraurban particulate concentrations in Christchurch, New Zealand: implications for epidemiological studies.

3.1. Summary

Epidemiological studies relating air pollution to health effects often utilise one or few central monitoring sites for estimating wider population exposures to outdoor particulate air pollution. These studies often assume that highly correlated particulate concentrations between intraurban sites equates to a uniform concentration field. Several recent studies have questioned the universal validity of this assumption, noting that in some cities, the uniformity assumption may lead to exposure misclassification in health studies. Few studies have compared central fixed site concentrations to intraurban population background sites using actual monitored data in cities with higher levels of pollution. This chapter examines daily concentration variations in particulate matter less than 10 micrometers in diameter (PM_{10}) at the neighbourhood scale over two winter months in Christchurch, New Zealand, a city with high winter pollution concentrations. Daily concentrations of PM_{10} data were collected for two winter months at ten background monitoring sites within 9.3 km of the central fixed monitoring site typically used for estimating exposure in epidemiological studies. Results indicate that while the correlation between PM_{10} concentrations measured at the central monitoring site and most background sites is strong ($r > 0.76$), absolute daily concentration differences between the central monitoring site and population background sites were substantial (mean 90^{th} percentile absolute difference = 17.6 $\mu g\ m^{-3}$). In Christchurch, a central monitoring site does not therefore appear to accurately depict wider area population exposures to PM_{10}. Local intraurban variations in particulates should be well understood before applying central monitoring site concentrations as proxies for population exposure in epidemiological studies.

3.2. Introduction

Air pollution, especially particulate matter (PM) air pollution, has received considerable attention over the past 15 years, primarily due to health effects associated with personal exposure variations (e.g., Pope *et al.* 1991, Schwartz & Dockery 1992, Dockery & Pope 1994). The prevailing approach for estimating exposure to air pollution at a coarse spatial resolution is the

application of an arithmetic mean concentration value from one or few central monitoring stations to the entire population of the study area (e.g., Pope *et al.* 1992, Dockery *et al.* 1993a, Samet *et al.* 2000, Wong *et al.* 2001, Pope *et al.* 2002, Zanobetti *et al.* 2003). This method assumes that the spatial distributions of certain pollutants, especially particulate concentrations, are distributed homogeneously within large urban areas. Early concentration variability studies in the United States supported this assumption, finding homogeneous distributions of particulates across intraurban (*within*-city) areas (Burton *et al.* 1996, Suh *et al.* 1997, Wilson & Suh 1997). However, several recent studies suggest that in some cities there may be greater spatial variation than previously reported and that the ecological method of particulate exposure assessment may misclassify personal exposures when variability is high (Briggs 2000, Zhu *et al.* 2002, Ito *et al.* 2004, Pinto *et al.* 2004, Kim *et al.* 2005, Wilson *et al.* 2005). Further, exposure misclassifications due to the selection of monitoring sites and sampling frequencies have been shown to significantly alter the magnitude of health outcomes in epidemiolgoical studies (Ito *et al.* 1995). Consequently, the study of air pollution exposures at the intraurban scale has been identified as a priority area for future work (Kukkonen *et al.* 2001, Brunekreef & Holgate 2002, Sajani *et al.* 2004, Jerrett *et al.* 2005a).

This chapter has several aims related to intraurban variations in particulate matter air pollution less than 10 micrometers in diameter (PM_{10}) in a city with high levels of winter air pollution, including: (i) to describe the nature of PM_{10} intraurban concentration homogeneity between all study sites in a dense monitoring network; (ii) to investigate the validity of the intraurban homogeneity assumption in Christchurch, New Zealand by comparing relative and absolute daily concentrations between ten population exposure monitoring sites and a central monitoring site; and (iii) to discuss the wider implications of exposure misclassification for epidemiological studies due to spatial heterogeneity of intraurban particulate concentrations.

3.3 Air pollution and intraurban uniformity

Air pollution in Christchurch has been reported for well over a century (Gray 1889) and has been systematically monitored since the 1950s (Wilkinson 1959). Christchurch is a mid-latitude city with a population of 330,000 located on the Canterbury Plains about 70 km east of the Southern Alps (172ºW, 43ºS) and just north of eroded remains of a late Tertiary volcanic complex known

as Banks Peninsula or the Port Hills. Christchurch's mid-latitude location in the southern hemisphere is in a climate significantly influenced by the interaction between the eastward propagating low and high pressure systems and the Southern Alps massif (Sturman & Tapper 1996). Cold air drainage from the Southern Alps converging with more localised cold air drainage winds originating from the slopes of Banks Peninsula is thought to generate zones of stagnant air, enhancing the strength of temperature inversions on cold winter nights (Kossmann & Sturman 2004). The main source of winter particulate air pollution in Christchurch is domestic heating, with approximately 48% of Christchurch homes burning coal or wood in the main living area as a source of heat on a typical winter's night and/or day (Lamb 2003). Domestic heating sources contribute 11.2 tonnes (t) out of the total 13.6 t (82%) of PM_{10} discharged on a typical winter's day (Wilton 2001b, Scott & Gunatilaka 2004). Ambient 24-hr averages of PM_{10} exceed national ambient air quality guidelines of 50 $\mu g\ m^{-3}$ an average of more than 30 times every winter and may reach up to 400 $\mu g\ m^{-3}$ in 1-hr maximum concentrations (Aberkane *et al.* 2004).

In Christchurch, multi-site monitoring studies of intraurban variations in particulate air pollution are limited. The most recent monitoring studies utilising more than four monitoring stations in Christchurch were published in the early 1980s. Sturman (1982) studied daily smoke concentration data for winter months (May through August) at 12 to 13 sites (non-concurrently) in Christchurch. A strong gradient between the city centre and the suburbs was observed, related to the distribution of emissions. The highest areas of concentration were in the east to northeast of the city centre, where population densities were highest. Relative daily variations were small and most of the spatial variation between sites was found in outlying areas of the urban area. In a follow-on study based on the same pollution data set, local wind systems were found to be a major contributor to spatial concentrations and dispersion patterns of smoke (Sturman 1985). More recently, Kossmann and Sturman (2004) studied meteorological conditions at five air pollution monitoring sites and noted significant variations in air pollution and wind characteristics over relatively small distances related to the complex and dynamic relationship between airflow and air pollution patterns over the city. Concentration variations between intraurban sites were not reported in detail.

Several studies have been conducted internationally on spatial variations in particulates at the intraurban scale, with mixed conclusions about local uniformity (Wilson *et al.* 2005). Burton *et al.* (1996) measured PM_{10}, $PM_{2.5}$ and coarse particles ($PM_{10-2.5}$) at eight sites in Philadelphia, finding high correlations between sites and low concentration variations, concluding 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. Suh *et al.* (1997) measured $PM_{2.5}$, PM_{10}, and coarse particle ($PM_{10-2.5}$) concentrations at six sites across the Washington D.C., 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 $PM_{2.5}$. Nerriere *et al.* (2005) recently measured seasonal $PM_{2.5}$ and PM_{10} concentrations at four metropolitan areas in France. Maximum average PM_{10} ambient concentration differences between types of ambient sites ranged from 26 – 34% in the winter and from 15 – 22% in the summer. $PM_{2.5}$ concentrations, and to a greater degree, 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. Research in Athens, where concentrations are routinely higher than other urban areas across Europe, indicated that spatial variation between sites at the intraurban level was significant. The coefficient of variance was high within the study area (CV = 0.36), and correlation coefficients ranged from 0.57 to 0.84 (Grivas *et al.* 2004). The presence of studies demonstrating clear variability and clear uniformity of intraurban particulate concentrations in various study areas indicates that conclusions about concentration homogeneity are mixed and are not necessarily applicable between study sites. This study in Christchurch focuses on investigating the nature and extent of heterogeneity in an area with high levels of winter air pollution.

3.4. Methods

3.4.1. PM_{10} monitoring network

Monitoring of PM_{10} 24-hr concentrations was conducted for two winter months (July 2003 and June 2004) at one site designated as the 'central monitoring site' (CMS) and at ten sites

designated as ‘population exposure monitoring sites’ in the Christchurch Territorial Local Authority (TLA). The CMS was located at the regional council (Environment Canterbury) ambient monitoring complex on Coles Place in the St. Albans neighbourhood of north-central Christchurch. This site is within 1 km of the ambient monitoring sites used in all local epidemiologic studies conducted over the past 20 years. The ten background population monitoring sites were located on the grounds of primary schools distributed across the city (Figure 3.1). As most of Christchurch is located on the coastal edge of the Canterbury Plains, elevations for most sites were low (elevation above sea level < 26 m) except for one site on the Port Hills (Mount Pleasant), where the elevation was significantly higher (107 masl). Population monitoring sites ranged from 2.3 to 9.3 km from the central monitoring site. Monitoring site locations were selected based on established US Environmental Protection Agency (US EPA) criteria as defined in Chow *et al.* (2002). Samplers were mounted on rooftops, utility poles, flagpoles and one rugby goalpost at distances of more than 5 m from building exhausts and intakes. Monitoring sites were located greater than 20 m from nearby trees and twice the difference in elevation from nearby buildings. Monitors were placed at a height so as to capture a vertical band of concentrations approximating human ambient exposure (2.9-3.4 m above ground). The nearest domestic emission sources were greater than 50 m away (50-250 m) and busy roads were more than 50 m away.

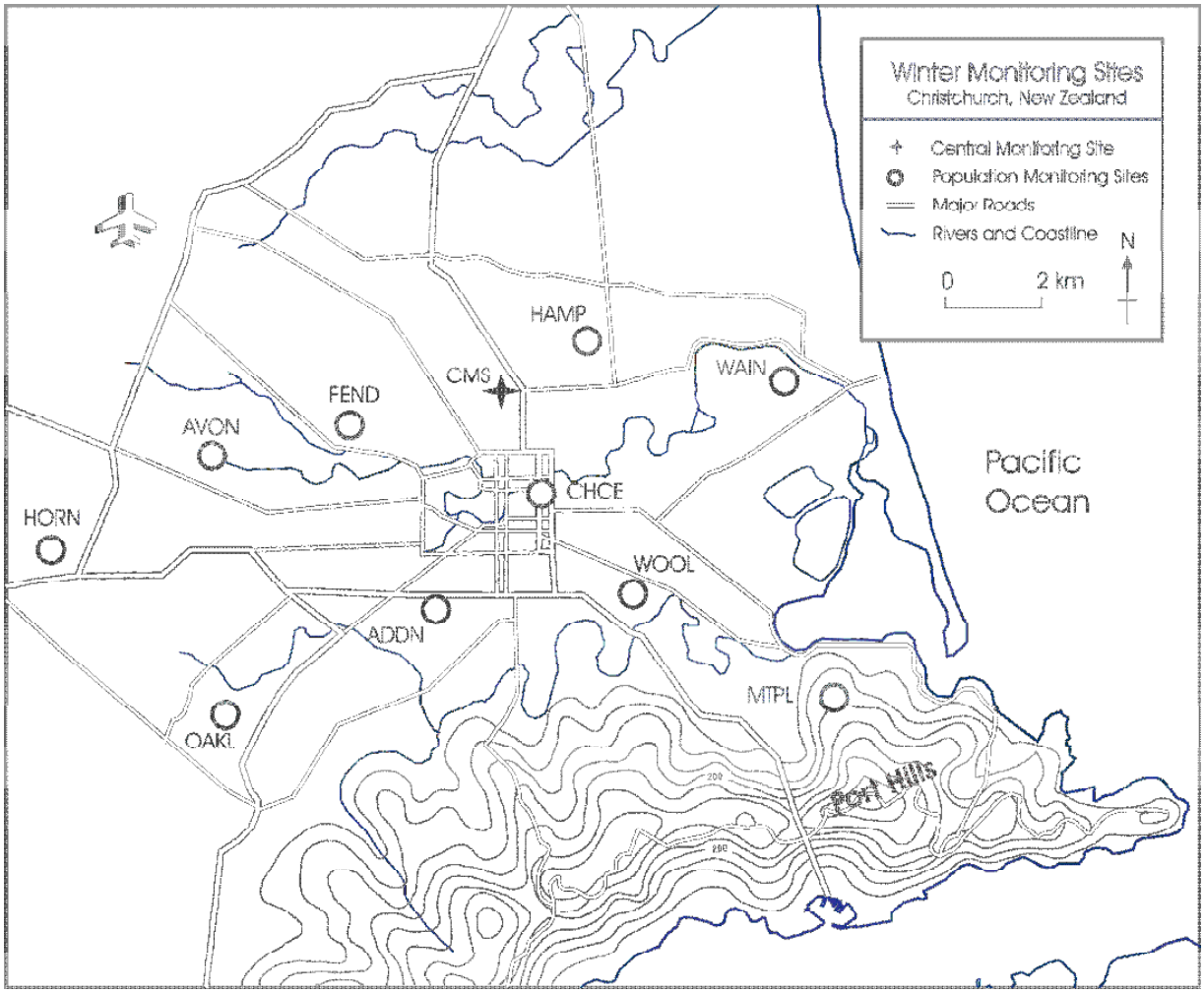


Figure 3.1 Study area and locations of winter monitoring sites for 2003 and 2004.

3.4.2. PM_{10} sampling, analysis and quality control

Airmetrics MiniVol portable samplers (MiniVols) were used to measure PM_{10} at all sites (Airmetrics 2000). MiniVols are not designated as US EPA Federal Reference Method (FRM) samplers, although the MiniVol has been evaluated by the US EPA as part of a preliminary field-testing of prototype FRM samplers and performed well (Yanosky & MacIntosh 2001). MiniVols sample particulate concentrations by drawing a known volume of air through a particle size separator and across a filter over a specified period of time. Pollution particles accumulate on the filter, which is weighed before and after the sampling process. Particles were collected on 47 mm Whatman glass fibre filters which were mounted in plastic filter holders. Flow rates for each of the MiniVols were initially calibrated to 5.5 L/min and were confirmed daily following each filter change. Filters were changed at the same time of day (± 30 minutes) during a time when particulate concentrations are lowest in the local diurnal cycle (0900-1300). Prior to and after sampling, filters were conditioned to a constant temperature (21 ± 3ºC) and relative humidity (40 ± 5%) for at least 24-hr prior to being weighed. Filters were ionized and concentration mass was measured three times on a Sartorius Genius balance with resolution of 0.01 mg (Sartorius AG 2000). After conditioning and weighing, filters were individually stored in a flat Petri dish and covered. Daily concentration averages for each monitoring site were temperature adjusted using co-located HOBO temperature loggers and pressure-adjusted based on data collected at the central monitoring site. Prior to deployment in the field, the MiniVols underwent a calibration and quality assurance testing period for four days in May 2002. All eleven MiniVols were co-located on a rooftop with a Tapered Oscillating Element Microbalance (TEOM). MiniVols were highly correlated with the TEOM (mean r = 0.987) and with each other (mean r = 0.989). Concentration differences were also small between MiniVols in both relative terms (mean coefficient of divergence = 0.05) and absolute terms (mean P_{90} difference in daily concentrations = 5.3 $\mu g\ m^{-3}$).

3.4.3. Data analysis

The statistical analysis of the monitored data was performed using SPSS 11.0 and Microsoft Excel 2003 software (SPSS 2001, Microsoft 2003). Spatial calculations and map layouts were

produced using Arc/MAP 9.0, a Geographic Information System (GIS). Several different measures were utilised to describe intraurban variability between all population monitoring sites, and variability specifically between population monitoring sites and the central monitoring site. The mean, maximum, minimum, and standard deviations of daily absolute concentrations were calculated for each site, as well as the number of days that the ambient concentration exceeded the local ambient air quality guideline of 50 µg m^{-3}. For relative temporal differences between sites, Pearson paired correlations were calculated. Because correlations alone may not sufficiently depict relative concentration differences between intraurban sites, a COD, or coefficient of divergence, was applied to describe relative intraurban uniformity (Wongphatarakul *et al.* 1998, Pinto *et al.* 2004). The COD is defined mathematically as:

$$COD_{jk} = \sqrt{\frac{1}{p}\sum_{i=1}^{p}[(x_{ij} - x_{ik})/(x_{ij} + x_{ik})]^2} \qquad (3.1)$$

where x_{ij} and x_{ik} represent the 24-hr average particulate concentration for sampling day *i* at sampling site *j* and *k*, and where *p* is the number of observations. The COD is important as a relative measure of uniformity as high correlations between intraurban sites demonstrate *temporal* homogeneity (i.e., sites move up and down together), but may not necessarily describe *spatial* concentration uniformity between sites (Pinto *et al.* 2004). A COD of zero means there are no differences between concentrations at paired sites, while a value approaching one indicates maximum differences and absolute heterogeneity. As a means of comparing central site concentrations to population site concentrations in terms of exposure classification, daily absolute concentration differences between the population sites and the central site were calculated, and aggregated the data into 5 µg m^{-3} bands (e.g., 0-5 µg m^{-3}, 6-10 µg m^{-3}, etc.). A linear regression between daily concentrations at each of the population monitoring sites and the central site was matched to the aggregated concentration difference data. The number of days when the CMS over or under estimated the population site was also calculated. In addition, arithmetic means, minimums and maximums of absolute daily differences between the central monitoring site and all population monitoring sites were calculated in order to demonstrate the heterogeneity of daily variation from the central site in greater detail.

3.5 Results

3.5.1. PM_{10} concentrations

24-hr PM_{10} concentrations at eleven sites over 59 mid-winter days in July 2003 and June 2004 were obtained. Table 3.1 provides a summary of winter monitoring results by site. Two full days of data (June 24th and 25th, 2004) were not available due to illness and three sites (Christchurch East, Oaklands, and Avonhead) were missing data during one additional day each during the monitoring period. Monitoring was conducted seven days a week during the two winter months. The arithmetic mean of the ten population monitoring sites during the monitoring period was 43.9 µg m^{-3} with a standard deviation of 26 µg m^{-3}. Maximum values for seven of the population monitoring sites ranged from 132.8 to 171.3 µg m^{-3} while two sites on the western edges of the city, Avonhead and Hornby, had maximum daily concentrations of only 95.4 µg m^{-3} and 93.6 µg m^{-3} respectively. Wind field analysis conducted by Kossmann and Sturman (2004) indicates that the western and south-western regions of the Christchurch TLA receive fresh cold air draining from the Southern Alps and the Port Hills, explaining the lower maximums at these monitoring sites. The Mount Pleasant population site measured substantially lower concentrations than all other sites during the winter, with an arithmetic mean concentration of 16.3 µg m^{-3} and a maximum of 38.9 µg m^{-3}. The low concentrations at the Mount Pleasant site are due to its location on the Port Hills, where cold drainage flows transport most wintertime pollution towards lower elevations over the city (Spronken-Smith *et al.* 2001)**.** The average number of days over the 24-hr ambient guideline of 50 µg m^{-3} for population monitoring sites was ~19 days out of the 58-59 days of total monitoring, while the central monitoring site recorded 27 exceedences. The summary results from the monitoring suggest that the central monitoring site may be over-estimating exposures for the population of Christchurch during winter months.

Table 3.1 Overview of winter monitoring data.

Site Name	Site ID	n	Mean	SD	Min	Max	Days >50 µg m^{-3}
Hammersley Park	HAMP	59	47.8	29.7	8.7	133.0	19
Christchurch East	CHCE	58	44.7	29.4	6.8	148.9	20
Fendalton	FEND	59	43.6	26.3	11	132.8	19
Addington	ADDN	59	49.3	28.9	15	147.5	19
Woolston	WOOL	59	52.6	33.2	14	171.3	23
Wainoni	WAIN	59	48.6	30.7	9.3	154.0	23
Avonhead	AVON	58	40.5	21.8	3.2	95.4	18
Oaklands	OAKL	58	44.1	26.7	11	167.2	18
Mount Pleasant	MTPL	59	16.3	8.0	1.9	38.9	0
Hornby	HORN	59	41.7	18.3	7.3	93.6	19
Population Sites Mean	XPOP	~59	43.9	26.0	8.7	129.7	18.6
Central Monitoring Site	CMS	59	53.6	34.5	7.6	144.5	27

3.5.2. Intraurban uniformity

Table 3.2 summarises the relative and absolute spatial concentration differences between all ten population monitoring sites using pair-wise Pearson correlations, coefficients of divergence (COD), 90th percentile of absolute daily differences (ΔP_{90}). Pearson correlations were significant ($p < 0.01$) and high ($0.757 <$ mean $r < 0.812$) between all sites except for site comparisons with Oaklands (OAKL) and Mt. Pleasant (MTPL). Correlations between OAKL and other sites were significant, but lower than other sites (mean $r = 0.579$), while pair-wise correlations for MTPL were neither significant nor high (mean $r = 0.140$). Relative measures of uniformity between population monitoring sites across the city showed considerable spatial variation, as reflected by the COD. Mean CODs ranged from 0.20 and 0.24 between all background sites except for Mt. Pleasant, which displayed substantially higher relative variation (mean COD = 0.50). The lowest relative variation was between Addington (ADDN) and Woolston (WOOL) sites (COD = 0.13) and the highest variation was found between the MTPL and WOOL sites (COD = 0.54), which are geographically quite close. In order to investigate the absolute magnitude of relative differences between sites, the 90th percentile of absolute daily concentration differences ($\Delta P90$) between all sites was calculated. The mean ΔP_{90} ranged from 27.4 μg m^{-3} to 37.4 μg m^{-3} between significantly correlated sites, while Mount Pleasant registered notably greater concentration differences (mean ΔP_{90} = 66.0 μg m^{-3}). The minimum ΔP_{90} was between the Hornby (HORN) and Avonhead (AVON) sites (19.1 μg m^{-3}) and the max ΔP_{90} was between the MTPL and Wainoni (WAIN) sites (86.0 μg m^{-3}). The significant variability between sites demonstrates that particulate concentrations are heterogeneous throughout the urban airshed in Christchurch, indicating that a central site may not accurately depict daily population exposures.

Table 3.2. Intraurban uniformity statistics for population monitoring site pairs.

	HAMP	CHCE	FEND	ADDN	WOOL	WAIN	AVON	OAKL	MTPL	HORN
HAMP	1	.798*	.826*	.850*	.839*	.917*	.813*	.514*	0.228	.780*
		0.22	0.19	0.16	0.18	0.18	0.22	0.25	0.50	0.20
		28.6	24.7	25.3	23.9	20.9	34.2	40.5	74.0	32.8
		58	59	59	59	59	58	58	59	59
CHCE		1	.878*	.858*	.868*	.858*	.805*	.627*	0.05	.783*
			0.20	0.19	0.21	0.19	0.21	0.21	0.48	0.21
			23.6	23.0	27.2	26.1	25.3	35.6	61.5	28.1
			58	58	58	58	57	57	58	58
FEND			1	.831*	.811*	.811*	.816*	.595*	0.043	.778*
				0.19	0.21	0.22	0.20	0.23	0.49	0.19
				28.2	38.6	30.5	19.9	37.6	60.8	25.7
				59	59	59	58	58	59	59
ADDN				1	.876*	.843*	.794*	.640*	0.174	.805*
					0.13	0.17	0.22	0.19	0.52	0.17
					27.6	24.2	30.0	33.4	64.3	30.4
					59	59	58	58	59	59
WOOL					1	.870*	.697*	.532*	0.118	.722*
						0.16	0.24	0.22	0.54	0.20
						24.5	35.0	39.6	75.5	34.0
						59	58	58	59	59
WAIN						1	.744*	.484*	0.157	.734*
							0.24	0.23	0.52	0.23
							38.4	37.8	78.9	29.5
							58	58	59	59
AVON							1	.668*	0.202	.859*
								0.20	0.46	0.18
								31.3	50.6	19.1
								57	58	58
OAKL								1	0.079	.597*
									0.49	0.20
									55.3	24.3
									58	58
MTPL									1	0.213
										0.47
										53.4
										59
HORN										1

Key to Table
Site ID abbreviation
Pearson r
Coefficient of divergence
90th percentile of daily absolute differences (ΔP_{90}) (μg m^{-3})
Number of paired observations (n)

	HAMP	CHCE	FEND	ADDN	WOOL	WAIN	AVON	OAKL	MTPL	HORN	MEAN
Mean r	0.792**	0.809**	0.793**	0.812**	0.777**	0.783**	0.775**	0.582**	0.140	0.757**	0.764**
Mean COD	0.23	0.24	0.24	0.22	0.23	0.24	0.24	0.22	0.50	0.20	0.25
Mean ΔP_{90}	33.9	31.0	32.2	31.8	36.2	34.5	31.5	35.0	63.8	28.0	35.79
Min n	58	57	58	58	58	58	57	57	58	58	-

* indicates significant at the 0.01 level. **MTPL site not included in arithmetic mean calculation of Pearson correlation. Table format from US EPA (2004).

3.5.3. Central site suitability

In order to test the suitability of the central monitoring site for predicting population exposures across the urban area, Pearson correlations, coefficients of divergence, 90th percentiles of absolute daily concentrations differences, and absolute concentration differentiation profiles for each population monitoring site related to the central monitoring site were calculated (Table 3.3 and Figure 3.2). Overall, results indicate that although concentration correlations between the daily mean of population monitoring sites (XPOP) and the central site are generally high (mean r = 0.907), significant absolute daily differences between the central site and the mean of population sites are present (mean ΔP_{90} = 39.0 µg m^{-3}). The mean COD between all population monitoring sites and the central monitoring site was 0.23, the mean differed from the central site by more than 10 µg m^{-3} on 34 (58%) of the 24-hr periods. At Christchurch East (CHCE), the site most correlated with the central site in terms of daily concentration averages (r = 0.896), differences were still great in absolute terms (mean ΔP_{90} = 28.5 µg m^{-3}) and relative terms (COD = 0.20, percentage of 24-hr periods > 10 µg m^{-3} absolute difference = 38%). Oaklands (OAKL) had a significant but relatively low correlation with the central monitoring site (r = 0.550) and displayed high variability with a mean ΔP_{90} of 53.9 µg m3 and a COD of 0.24, where 47% of 24-hr periods showed > 10 µg m^{-3} absolute difference. Correlations between MTPL and the central monitoring site were not significant and low ($p < 0.01$, r = 0.063) with extremely high variability (COD = 0.53, ΔP_{90} = 86.0 µg m^{-3}, percentage of 24-hr periods > 10 µg m^{-3} absolute difference = 78%). Spatially, there appears to be greater absolute variability from the central site at population sites on the western edge of the city and at the Mt. Pleasant site (Figure 3.3).

Table 3.3. Summary statistics of differences between all population monitoring sites and the central monitoring site (CMS).

Site ID	Dist (km)	n	r	COD	ΔP_{90}	Daily differences from CMS (CMS - population)				
						Abs mean	Min	Max	Days > CMS	Days < CMS
XPOP	5.6	59	0.907*	0.23	39.0	17.4	-14.4	66.2	16	43
HAMP	2.3	59	0.874*	0.16	28.0	12.3	-30.8	67.0	24	35
CHCE	2.3	58	0.896*	0.20	28.5	12.3	-13.0	63.9	13	45
FEND	3.4	59	0.890*	0.20	31.9	13.7	-29.3	54.4	15	44
ADDN	4.3	59	0.806*	0.18	31.5	14.5	-55.6	77.4	27	32
WOOL	5.0	59	0.820*	0.18	32.5	14.3	-61.5	63.2	34	25
WAIN	6.0	59	0.850*	0.19	32.5	14.4	-36.0	62.5	24	35
AVON	6.2	58	0.831*	0.24	39.1	16.3	-22.1	88.8	12	46
OAKL	8.8	58	0.550*	0.24	53.9	19.3	-109.5	99.6	21	37
MTPL	9.3	59	0.063	0.53	86.0	38.3	-19.7	136.2	3	56
HORN	8.8	59	0.760*	0.23	43.1	18.8	-27.4	80.6	18	41
Mean**	5.6	59	0.734*	0.23	40.69	17.4	-40.5	79.4	19	39
Min**	2.3	58	0.063	0.16	28.04	12.3	-109.5	54.4	3	25
Max**	9.3	59	0.896	0.53	86.00	38.3	-13.0	136.2	34	56

*Significant to the 0.01 level. ** Does not reflect population average (XPOP). Abbreviations: Dist, distance between site and central monitoring site; n, number of paired observations; r, Pearson correlation; COD, Coefficient of Divergence; CMS, Central Monitoring Site; SD, Standard Deviation; ΔP_{90}, 90^{th} percentile of absolute differences from the central monitoring site;XPOP, daily average of the population sites concentrations.

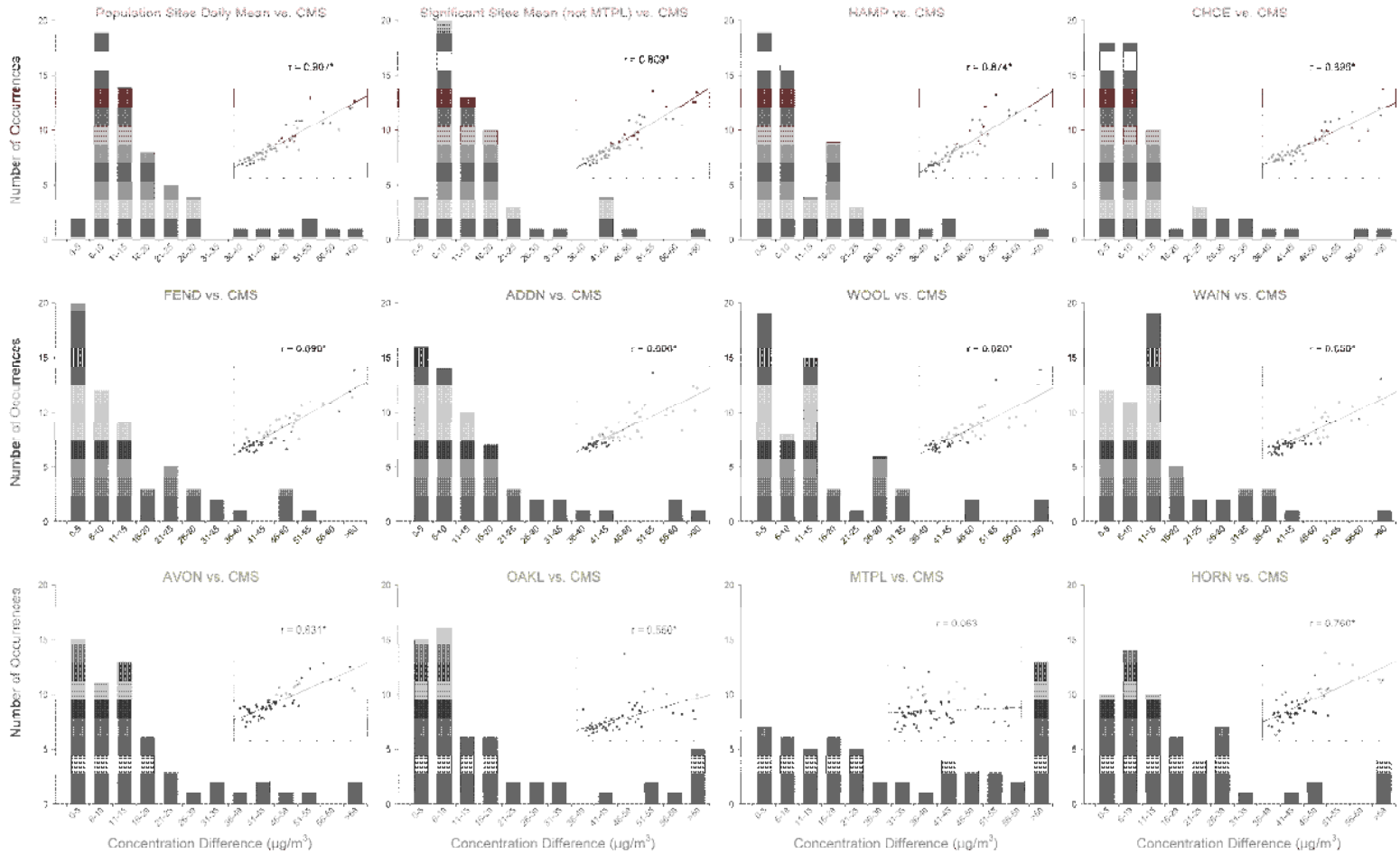


Figure 3.2. Daily absolute concentration difference bars, concentration regression plots* and Pearson coefficients between central monitoring site and population monitoring sites.

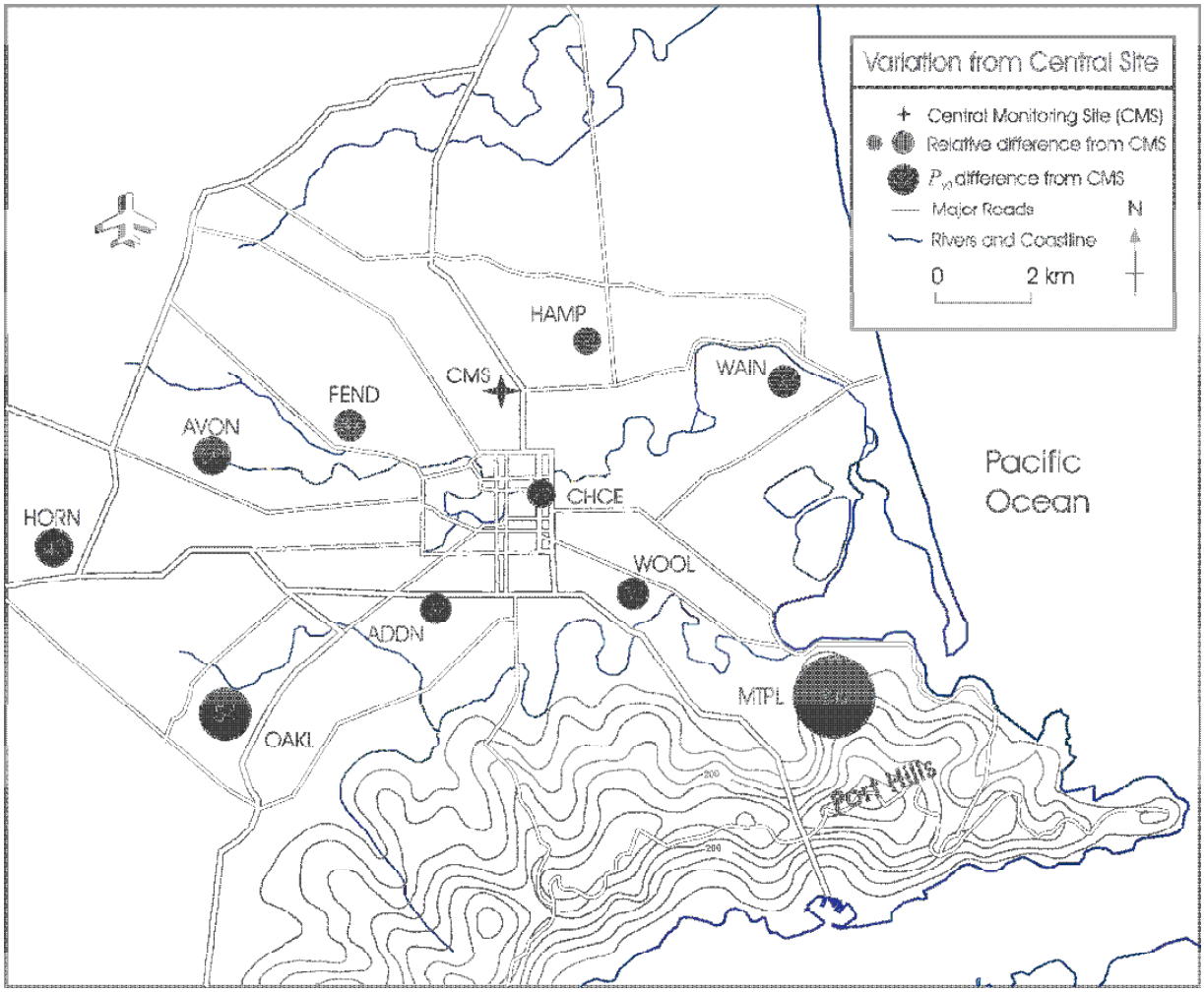


Figure 3.3. The 90th percentiles (P_{90}) of absolute winter daily concentration differences from the Central Monitoring Site (CMS).

In order to test that well correlated sites were not fluctuating up and down from their daily concentration baselines at the same magnitude relative to the central, two population monitoring sites that were similar in terms of several measurements as related to the CMS were selected, then compared their daily divergence from CMS concentrations (Figure 3.4). The Woolston (WOOL) and Wainoni (WAIN) sites were similar in their relationship to the central monitoring site with respective to distance (5.0 km, 6.0 km); correlations (r = 0.820, r = 0.850); CODs (r = 0.18, r = 0.19); and mean 90th percentile differences (ΔP_{90} = 32.5 μg m^{-3}, ΔP_{90} = 32.5 μg m^{-3}). However, they experienced marked differences from the central monitoring site on a daily basis. WAIN exceeded the daily concentration of the CMS on 24 of 59 occasions (41%), while WOOL exceeded the CMS on 34 (58%) occasions and was the only site registering higher than the CMS more than half of the 24-hr hour periods. Absolute differences between the two sites' daily divergence from the CMS ranged from 0.4 - 65.5 μg m^{-3} with a daily mean of 11.5 μg m^{-3}. Daily divergence from the CMS differed between the two sites by more than 10 μg m^{-3} on 23 occasions (39% of the time). These results show that the daily concentrations for population sites need not move up and down in unison, even when population monitoring sites are well correlated to a central site and share similar measures of absolute and relative divergence from the CMS. Our results indicate that the central monitoring site in Christchurch shows significant variation in concentrations from population monitoring sites. Further, use of the central site as a proxy for wider population exposures in Christchurch may lead to significant exposure misclassification in health studies.

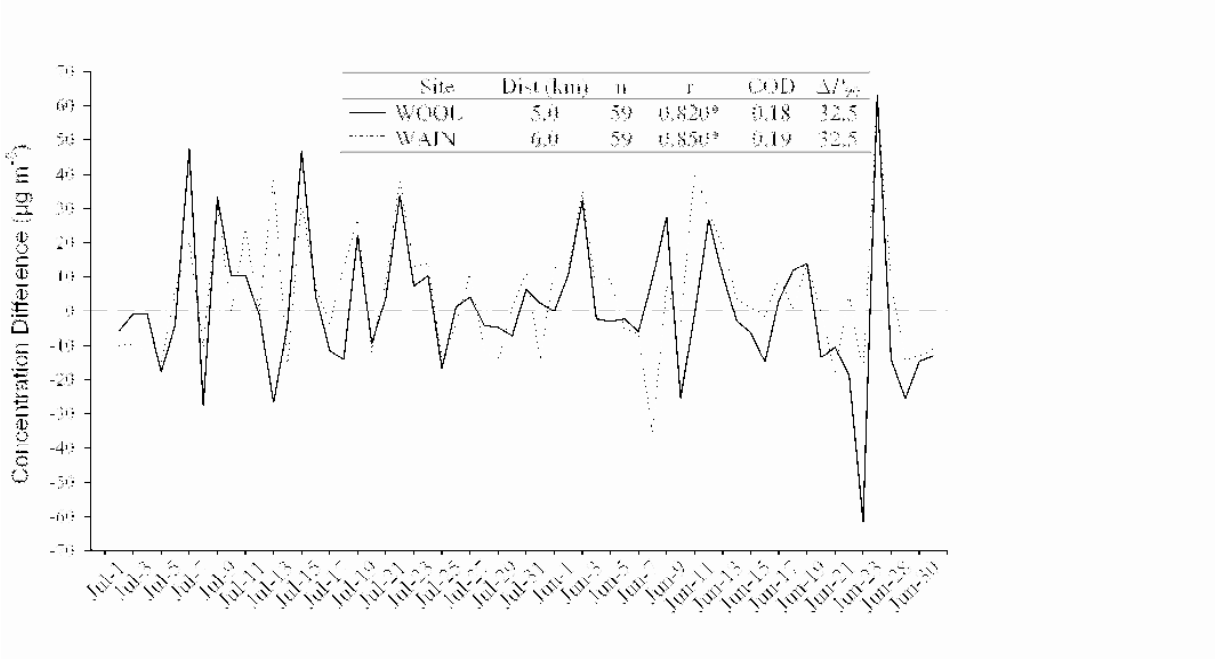


Figure 3.4. Daily concentration differences between two similarly correlated background population sites (Woolston and Wainoni) and the central monitoring site (CMS)*.

3.6. Discussion

This study investigated the spatial variability of winter PM_{10} concentrations at the intraurban level in Christchurch, New Zealand. Our results indicate that daily winter PM_{10} concentrations Christchurch, in a city with high levels of winter particulate air pollution, are not uniform at the intraurban scale. Epidemiologic studies conducted here may have significantly misclassified daily population exposures by using concentration data from the central monitoring site as a proxy for the wider area's exposure. The broader implications of this research are that intraurban particulate matter spatial variability should be considered and investigated prior to making assumptions about wider population exposures based on a central fixed monitoring site. If the nature of intraurban uniformity is not well understood and accounted for, exposure misclassification may occur, possibly affecting the reliability of conclusions drawn in epidemiologic studies, especially of the cohort design-type.

It should be noted that this study has several limitations. First, the ten satellite sites were designated as 'population monitoring sites'. It is difficult to ascertain to what degree these sites are actually monitoring localised population concentrations. The nature of wintertime pollution in Christchurch is such that there is a high density of point sources, namely household chimneys. Monitors were located at sufficient distances from point sources (> 50 m) and from traffic sources (> 50 m), ensuring that the satellite monitoring locations were as representative as possible of the surrounding area. Given that even the closest paired population monitoring sites exhibited relatively large variations (Tables 3.2 and 3.3), the radius of representativeness for spatial uniformity may be quite small. More research of outdoor concentration variations at the neighbourhood scale (< 4 km) would be useful in understanding the nature and extent of PM uniformity. One solution may be the use of integrated meteorological-emission models (Jerrett *et al.* 2005a). Zawar-Reza *et al.* (2005) recently used a mesoscale atmospheric dispersion model that incorporates topography, The Air Pollution Model (TAPM), to predict annual concentrations of PM_{10} in Christchurch with promising results, although predictions for 24-hr averages have yet to be tested.

Our study is also limited by the fact that source apportionment analysis was not performed due to the type of filters used (glass fibre rather than Teflon). Recent research by Kim *et al.* (2005) and

Ito *et al.* (2004) address the contribution of sources to intraurban heterogeneity, which appears to be significant, even in cities where total mass concentrations are relatively uniform. In Christchurch, emission inventories show that a large portion of winter sources are from domestic heating, but sub-species tests are limited in spatial coverage (Wang & Shooter 2001, Wang & Shooter 2002, Scott & Gunatilaka 2004, Wang & Shooter 2004). Future spatial analysis of intraurban variations in source composition is an important next step in determining the causes of intraurban heterogeneity in Christchurch.

Ambient concentrations of particles vary spatially and temporally and effect human health to differing degrees depending on size fraction (Monn 2001, Englert 2004). A general assumption based on stopping velocities, settling times, and field evidence is that spatial distributions for finer particles ($PM_{2.5}$ and PM_1) are relatively more uniformly distributed than for coarser fractions (Brimblecombe 1986, Wilson & Suh 1997, Monn 2001). However, at sizes smaller than fine particles, i.e., Ultra Fine Particles (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). Although PM is widely accepted as a type of pollution with great influence on human health, there is evidence to suggest that finer particles like $PM_{2.5}$ down to nanoparticles size fractions may exert a health burden that exceeds that of larger particle size fractions (Schwartz *et al.* 2002, Englert 2004, Oberdorster *et al.* 2005). The results found here for PM_{10} may depict a greater level of intraurban heterogeneity and lower burden of health effects than if the study had focused on a smaller size fraction, such as $PM_{2.5}$.

Relative and absolute measures of PM_{10} concentration variations between all sites in Christchurch indicate significant daily spatial variability of particulate concentrations at the intraurban scale (Tables 3.2 and 3.3). The intraurban mean COD of 0.25 represents a substantial degree of relative variation between population monitoring sites that is similar to variability found in cities in the western United States (Pinto *et al.* 2004). The sites at Oaklands (OAKL) and Mt. Pleasant (MTPL) were markedly different from the central site and from other population monitoring sites. Although statistically significant, correlations between OAKL and other sites were poor (mean $r = 0.582$), most likely due to the location of the monitoring site. OAKL is located in a zone where cold drainage flows from the Southern Alps foothills and Port

Hills converge, destabilising temperature inversions and ventilating the area (Kossmann & Sturman 2004). MTPL was not significantly correlated with any other site and showed the highest concentration differences from all other sites, due to its elevated position (107 masl) on the hills. The data therefore indicate that ambient PM_{10} exposures in the population subgroup on the Port Hills are severely and consistently over-estimated by the central site on both a daily and average basis in the winter.

Results show that 24-hr PM_{10} concentrations at statistically significant population monitoring sites (i.e., excepting Mt. Pleasant) were well correlated with each other (mean r = 0.734) and with the central site (mean r = 0.764), but observed large variations in absolute concentrations between monitoring sites, (28 μg m^{-3} < mean ΔP_{90} < 35 μg m^{-3}). Relative differences were also high between population monitoring sites (0.20 < COD < 0.24), and between the central site and population sites (mean COD = 0.23), indicating spatial heterogeneity. Further, concentration profiles of daily differences between the central site and the population sites were diverse (Figure 3.2), despite being highly correlated with the central site. It is concluded that correlations alone are not sufficient for describing spatial heterogeneity in Christchurch, as has also been found in studies in the United States (Pinto *et al.* 2004). Coefficients of divergence and 90^{th} percentiles of absolute concentration differences are more representative measures of intraurban particulate uniformity, and should be used in place of correlations when characterising spatial concentration heterogeneity.

The gradient in means of 90^{th} percentile absolute differences from the central monitoring site appears to increase slowly, then rise rapidly in the stations furthest from the CMS, especially to the west (Avonhead, Hornby and Oaklands) and to the southeast (Mt. Pleasant) (Figure 3.3). However, this gradient is probably not related only to distance from the CMS. Correlation coefficients between distance and COD, and distance and mean ΔP_{90} for all sites were low, with r^2 values 0.141, and 0.12, respectively. The steep ΔP_{90} gradient from the central monitoring site at the outer edges of the city was more likely due to monitoring site locations either on the western edges of the city (Avonhead, Hornby and Oaklands), which receive fresh cold air drainage from the Southern Alps on high pollution nights, or on the Port Hills (Mt. Pleasant), where pollution does not accumulate. Oaklands may also receive clean cold air drainage from the

western end of the Port Hills, as suggested by Sturman and Zawar-Reza (Sturman & Zawar-Reza 2002). Sturman (1982) observed a similar gradient pattern of mean concentrations at the city boundary over 20 years ago, which was then attributed to source locations being more dense in the central city. The steep gradient in concentration differences from the CMS along the southern and western edges of the city is more likely due to local-scale meteorology than emissions. Although concentration differences between the central site and the remaining seven sites are lower than for sites located on the hills, absolute and relative concentration differences are still large (20 μg m^{-3} < mean ΔP_{90} < 39 μg m^{-3}, 0.16 < COD < 0.24).

From a cursory standpoint, the average 90th percentile differences from the CMS (Figure 3.3) may indicate uniformity, as the sites located in central Christchurch measured a relatively small range of values (29 μg m^{-3} < mean ΔP_{90} < 32 μg m^{-3}). It should be noted that these represent *averages* over the entire two-month winter period. Longer term averages are known to be less variable, and epidemiological research that study annual or seasonal exposures to particulate matter are probably at less risk of misclassifying exposure at the intraurban level, unless concentration differences *within* urban areas are greater than differences *between* cities, a case which may produce significant errors in health risk estimates (Dominici *et al.* 2003, Wilson *et al.* 2005). While the arithmetic means of 90th percentile differences from the CMS were similar, daily variations are marked (Figure 3.4). The Woolston (WOOL) and Wainoni (WAIN) sites were similar to the central monitoring site in respective terms of distance, correlation, and relative and absolute concentration differences. However, the absolute daily differences from the central monitoring site for the two sites had a wide daily range, differing from each other by 11.5 μg m^{-3} on average. The range of discrepancy in daily particulate concentrations between the central site and population monitoring site articulates the complexity of the intraurban particulate concentration field.

The presence of high PM_{10} concentration variability at the intraurban scale in Christchurch has wider implications for epidemiologic research. Most epidemiological studies conducted in the United States and in Europe have measured particulate population exposures by applying central fixed site values to the wider population (Pope *et al.* 1992, Dockery *et al.* 1993a, Schwartz 1994, Samet *et al.* 2000, Pope *et al.* 2002, Schwartz 2004). In Christchurch, published epidemiologic

studies have used a similar method to international studies, applying 24-hr average concentration data from one central ambient monitoring site to estimate particulate concentrations for the entire urban area. Hales *et al.* (1999) studied PM_{10} in relation to daily mortality in Christchurch and found that a 10 μg m^{-3} increase in daily PM_{10} was associated with a 1% increase in all-cause mortality and a 3% increase in mortality related to respiratory disease. PM_{10} exposure data in this study were from a 'representative, centrally located site' located just north of the city centre. McGowan *et al.* (2002a) studied the associations between daily PM_{10} concentrations and hospital admissions for cardio-respiratory illness. For all age groups there was a 3.37% increase in daily respiratory admissions for approximately each 15 μg m^{-3} rise in daily concentrations of PM_{10} and there was a 1.26% increase in daily cardiac admissions for each 15 μg m^{-3} rise in daily PM_{10}. PM_{10} exposure data in this study was also based on data from one monitoring site north of the city centre. Health effect studies in Christchurch, as well as elsewhere, generally assume that the particulate field is uniform at the intraurban scale. Our research on variability shows the intraurban concentration uniformity assumption to be invalid in Christchurch for daily levels, indicating that exposure estimates for the wider population may be misclassified, confounding health estimates, although risk estimate error from exposure misclassification in these daily time-series studies is probably less severe than for long-term cohort design studies (Zeger *et al.* 2000).

In this study, daily spatial PM_{10} concentrations were shown to be complex and variable in a city with elevated levels of winter air pollution. High correlation between sites was not indicative of relative homogeneity, highlighting the importance of more valid methods for describing uniformity, such as a coefficient of divergence. Variability was substantial, not only between the central monitoring site and the background population sites, but also between all intraurban sites in the study. It was concluded that spatial variations in particulate air pollution in Christchurch are of a level significant enough to warrant caution when using central monitoring sites to estimate wider-population exposures, especially in long-term cohort epidemiological studies. It is recommended that future epidemiological studies in Christchurch and elsewhere incorporate concentration uniformity due diligence into their quality control measures prior to the application of central site concentrations as exposure proxies to wider populations.

Chapter 4: Intraurban-scale dispersion modelling of particulate matter concentrations: Applications for exposure estimates in cohort studies.

4.1. Summary

Epidemiological studies relating air pollution to health effects often estimate personal exposure to particulate matter using values from a central ambient monitoring site as a proxy. However, when there is a significant amount of variation in particulate concentrations across an urban area, the use of central sites may result in exposure misclassification that induces error in long-term cohort epidemiological study designs. When spatially dense monitoring data are not available, advanced dispersion models may offer one solution to the problem of accurately characterising intraurban particulate concentrations across an area. This study presents results from an intraurban assessment of The Air Pollution Model (TAPM) – an integrated meteorological-emission (IME) model. Particles less than 10 micrometers in aerodynamic diameter (PM_{10}) were modelled and compared with a dense intraurban monitoring network in Christchurch, New Zealand, a city with high winter levels of particulate air pollution. Despite the area's high intraurban concentration variability, and meteorological and topographical complexity, the model performed satisfactorily overall, with mean observed and modelled concentrations of 42.9 μg m^{-3} and 43.4 μg m^{-3} respectively, while the mean Index of Agreement (IOA) between individual sites was 0.60 and the mean systematic RMSE was 16.9 μg m^{-3}. Most of the systematic error in the model was due to coarse spatial resolution of the local emission inventory and complex meteorology attributed to localised convergence of drainage flows, especially on the western and southern fringes of the urban area. Given further improvements in site-specific estimates within urban areas, IME models such as TAPM may be a viable alternative to central sites for estimating personal exposure in longer-term (monthly or annual) cohort epidemiological studies.

4.2. Introduction

Epidemiological studies that assess the relationship between air pollution and adverse health effects typically utilise concentrations from one or few central ambient monitoring sites as a proxy for estimating personal air pollution exposure (e.g., Pope *et al.* 1992, Dockery *et al.* 1993a, Samet *et al.* 2000). In the case of particulate matter (PM) air pollution, the common assumption is that concentrations are homogeneous over an urban area, and thus, ambient sites are understood to be a valid substitute for personal exposures. However, recent research has highlighted the fact that this assumption does not always hold true at the intraurban (*within-city*) scale, leading to the potential for error in long-term epidemiological study designs (Dominici *et al.* 2003, Wilson *et al.* 2005). Hence, the development of models that accurately predict long-term air pollution exposures at the intraurban scale has recently been recognised as a priority area of study (Kukkonen *et al.* 2001, Brunekreef & Holgate 2002, Sajani *et al.* 2004, Jerrett *et al.* 2005a).

Interest in intraurban concentration modelling has increased recently due to several factors, including: (i) the growing contributions of traffic emissions within cities; (ii) studies linking exposure variation and resulting health effects within cities; and (iii) advances in Geographic Information Systems (GIS) and spatial statistical techniques (Jerrett *et al.* 2005a). An additional reason for the recent emergence of interest in intraurban concentration variability is the concern for exposure misclassification errors caused by using central sites. There are mixed conclusions in the literature as to the degree of heterogeneity that particulate matter concentrations exhibit within urban areas. Early studies of intraurban concentrations found that finer particles were uniformly distributed (e.g., Burton *et al.* 1996), although more recent studies indicate that levels of uniformity may vary greatly from city to city (Pinto *et al.* 2004, Ito *et al.* 2005). Wilson *et al.* (2005) recently reviewed the intraurban monitoring literature and the possible error implications of unfounded assumptions about intraurban uniformity for long-term epidemiological studies. The review indicated that particulate concentrations across cities did not conform to any concrete set of assumptions about intraurban uniformity, and that the methods and techniques chosen to characterise uniformity could bias conclusions made about homogeneity. In situations where intraurban PM heterogeneity exists, and an adequate number of intraurban monitoring sites (e.g., $n > 4$) is not available, models that effectively simulate exposures based on air pollution

emissions and dispersion may be an appropriate and effective alternative, though they have not been used to date (Jerrett *et al.* 2005a). The aim of this chapter is to evaluate the effectiveness of an integrated meteorological-emission (IME) model, The Air Pollution Model (TAPM), for estimating small-area particulate air pollution exposures within an urban area using winter monitored data from eleven sites.

4.3. Intraurban concentration modelling

Jerrett *et al.* (2005a) recently reviewed intraurban exposure models, designating six classes of models in order of increasing complexity: (i) proximity-based assessments; (ii) statistical interpolation models; (iii) land use regression models; (iv) line dispersion models; (v) integrated meteorological-emission models; and (vi) hybrid models. Hybrid models, which combine personal or regional monitoring with other air pollution exposure methods, were found to be the preferable option. However, hybrid models are dependent on underlying assumptions about the spatial variations in particulate concentrations, which may not be uniform or highly correlated (Jerrett *et al.* 2005a). After hybrid models, integrated meteorological-emission models, which simulate the emission and dispersion of pollutants using coupled meteorological and chemical modules, offer the greatest potential for accurately estimating personal exposures. IME models were found to have a sufficient level of transferability to other study locations, along with complexity and utility in respiratory-related health study applications. Although these integrated models have been shown to be an accurate predictor of annual concentration levels at a single urban point, and have been tested between communities (e.g., Vogel *et al.* 1995, Chen & Dudhia 2001, Pearson & Fitzgerald 2001, Frohn *et al.* 2002, Hurley 2002, Tilmes *et al.* 2002, Hurley *et al.* 2003, Luhar & Hurley 2003, Zawar-Reza *et al.* 2005), they have yet to be independently assessed at multiple and dense intraurban locations for use in epidemiological studies (Jerrett *et al.* 2005a). For more detail on intraurban concentration studies and health studies, see the recently published review by Jerrett *et al.* (2005a).

There are several variations of IME models available to researchers. In the meteorological and dispersion modelling research communities, these models are commonly referred to as *mesoscale* models, referring to their ability to simulate sub-synoptic scale atmospheric processes (Jacobson 2000, Pielke 2002). Mesoscale models not only take into account the effects of the daily weather

cycles on air pollution dispersion, but also incorporate airflow produced by variation in landscape, such as sea-breezes or mountain flows (Whiteman 1990). The most commonly used models are the Fifth Generation Mesoscale Model (MM5: Grell *et al.* 1994, Chen & Dudhia 2001), and the Colorado State University Regional Atmospheric Modelling System (CSU-RAMS: Pielke *et al.* 1992). Models such as these use advanced computational techniques and atmospheric physics to predict the daily variation in wind and turbulence anywhere on the globe at a hierarchy of spatial scales – from the global planetary circulation down to localised flow around a building. IME or mesoscale models like the MM5 and the CSU-RAMS have been used infrequently in applications outside of the modelling community, mostly due to the high level of computing power and expertise needed to set up and run the models (Jerrett *et al.* 2005a). The Commonwealth Scientific and Research Organisation's (CSIRO – www.cmar.csiro.au) new mesoscale model – The Air Pollution Model (TAPM) – has recently become available and has been designed for use by a wider non-specialist community.

TAPM is a three-dimensional incompressible, non-hydrostatic, primitive equation model that uses a terrain-following coordinate system (Hurley *et al.* 2002). The model is a fast, closed-source, PC-based, prognostic and air pollution model with a Graphical User Interface (GUI) allowing for configuration of inputs, the designation of simulation parameters, and the analysis of outputs. As with CSU-RAMS and MM5, high-resolution simulation of meteorology and dispersion is achieved by successively telescoping (nesting) from large geographical domains of 1000 km^2 to finer spatial domains (less than 5 km^2). TAPM requires two basic types of input data: meteorology and emissions. Synoptic meteorology is ingested by the model using data from the Limited Area Prediction System (LAPS) analysis from the Australian Bureau of Meteorology (Puri *et al.* 1998). Sea surface temperatures and surface properties can be configured through the GUI. In addition, the GUI allows for the input of various emission species in gridded, area, and line source configurations. Boundary and surface layer parameterisations and equations are detailed in Hurley (2002). To improve the simulated meteorological fields, a data assimilation option is also incorporated in TAPM, whereby the model is forced towards actual measured wind speed and directional data.

4.4. Methods

4.4.1. Study area

Christchurch is a coastal city on the South Island of New Zealand with a population of 320,000, located on the Canterbury Plains about 70 km east of the Southern Alps (172ºW, 43ºS) and just north of eroded remains of a late Tertiary volcanic complex known as Banks Peninsula or the Port Hills (Figure 4.1). The interactions between meteorology and topography in Christchurch are complex, providing a challenging environment for air pollution models. Christchurch's mid-latitude location in the southern hemisphere is in a climate significantly influenced by the interaction between the eastward propagating high and low pressure systems and the Southern Alps massif (Sturman & Tapper 1996). Cold air drainage from the Southern Alps converging with more localised cold air drainage winds originating from the slopes of Banks Peninsula is thought to generate zones of stagnant air, enhancing the strength of temperature inversions on cold winter nights (Kossmann & Sturman 2004).

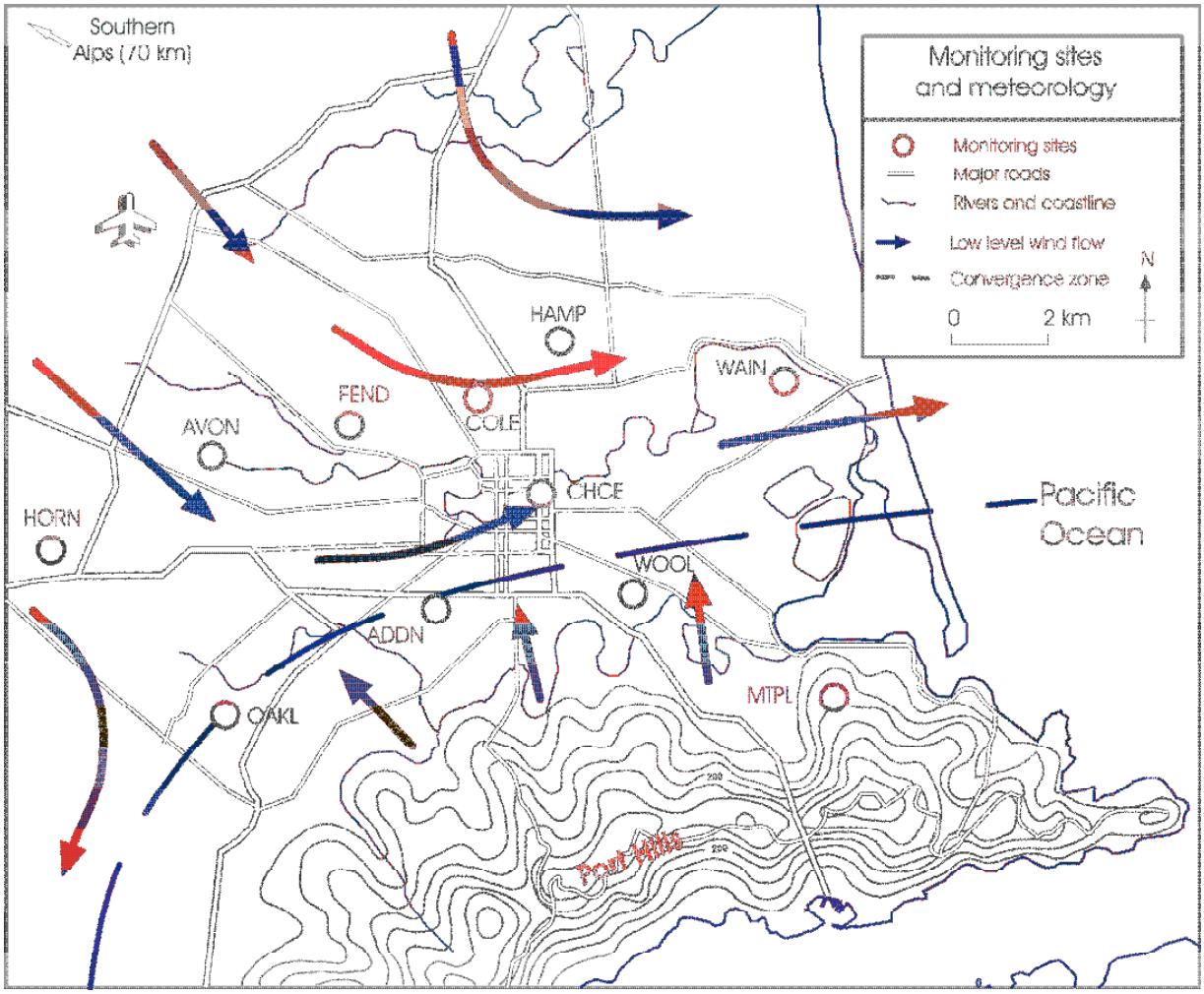


Figure 4.1. Local topography (50 m contour intervals), meteorology and monitoring sites (adapted from Kossmann and Sturman, 2004). Low level wind flows indicated are typical meteorology during high-pollution winter nights.

Air pollution has been a significant and persistent problem in Christchurch since the late 1800s and remains a considerable health risk to the current population (Gray 1889, McGowan *et al.* 2002). Christchurch measures on average 30 exceedances over the 50 μg m^{-3} 24-hr guideline set by the New Zealand Ministry for the Environment (Aberkane *et al.* 2004). Most of Christchurch's PM air pollution in winter is due primarily to the domestic heating emissions from the combustion of solid fuels (Scott & Ganatilaka 2004). Emission inventory data have been collected regularly in Christchurch to monitor trends over time and to determine changes in the relative contribution of sources to emissions (Scott & Ganatilaka 2004). The inventory contains raw emissions data for a 'typical winter's night' in kilograms or tonnes, divided into four six-hour time periods per day. In the 1999 emissions inventory, Christchurch was separated into three different sub-areas: inner Christchurch, suburban Christchurch and outer Christchurch. In work related to this chapter (Wilson *et al.* 2006), daily intraurban concentrations were assessed in Christchurch and found to be highly variable between sites and of similar heterogeneity levels as cities in North America where intraurban variability is highest (Pinto *et al.* 2004). Several local studies have found associations between acute exposure to particulate air pollution and various health effects, including restricted activity days (Wilton 2001a), morbidity (McGowan *et al.* 2002) and mortality (Hales *et al.* 2000).

4.4.2. Monitoring network

A temporary ambient PM_{10} monitoring network was designed and operated over two winter months (July 2003 and June 2004) at 11 sites across the city of Christchurch (Figure 4.1). Selection of the monitoring site locations was based on established US Environmental Protection Agency (US EPA) criteria (Chow *et al.* 2002). The nearest domestic emission sources were greater than 50 m from the monitoring inlets, industrial sources were at distances greater than 75 m and roads with higher traffic densities were more than 50 m away. As most of Christchurch is located on the Canterbury Plains, elevations for sites were mainly low (elevation above sea level < 26 m), except for Mount Pleasant (MTPL), the one site on the northern slope of the Port Hills. The elevation here was significantly higher (107 m above sea level). Airmetrics (Airmetrics 2000) MiniVol portable gravimetric samplers (MiniVols) were placed at a height so as to capture a vertical band of concentrations approximating human ambient exposure (2.9 - 3.4 m). Although

MiniVols are not designated as US EPA Federal Reference Method (FRM) samplers, they performed well when evaluated by the US EPA as part of a preliminary field-testing of prototype FRM samplers (Yanosky & MacIntosh 2001). Particles were separated by size using 10 μm impactors and collected on 47 mm Whatman glass fibre filters which were mounted in plastic filter holders. Flow rates for each of the MiniVols were calibrated to 5.5 lpm and were confirmed daily following each filter change. The sampling duration was 24 hours. Filters were changed at the same time of day (± 30 minutes) during a period when particulate concentrations are lowest in the local diurnal cycle (0900 – 1300 hrs). Filters were conditioned to a constant temperature (21 ± 3ºC) and relative humidity (40 ± 5%) for at least 24 hours prior to being weighed three times on a Sartorius Genius balance with resolution of 0.01 mg (Sartorius AG 2000). Prior to deployment in the field, the MiniVols underwent a calibration and quality assurance testing period over four days in May 2002 when the eleven MiniVols were co-located on a rooftop with a Tapered Oscillating Element Microbalance (TEOM). MiniVols were highly correlated with the TEOM (mean r = 0.987) and with each other (mean r = 0.989). The MiniVols consistently under-predicted TEOM values, which is expected as TEOMs use a heated element that known to burn off some of the mass related to semi-volatile and volatile organic compounds (VOCs), although PM_{10} samples are less affected by differences between methods as most of the semi-volatiles are in the finer fractions (Allen *et al.* 1997). The 90th percentile of absolute concentration differences between MiniVols was 5.3 μg m^{-3}.

4.4.3. TAPM configuration

TAPM was configured using three telescoping grids centred at the Coles Place (COLE) monitoring site with 60 grid nodes in the lateral and longitudinal directions. The distance between the grid nodes was 13.5, 3.5, and 1.5 km. The coarsest resolution grid accounted for the whole of the South Island, while the high-resolution mesh was set to cover Christchurch and most of the Canterbury Plains, allowing for the simulation of local wind systems. The air pollution grid cells were spatially aligned with the meteorological grid. As the emission grid data were only available for the metropolitan area of Christchurch, this grid was significantly smaller in area than the other grids, occupying about 25% of the pollution grid area. To simulate the dispersion of PM_{10}, the air pollution module of TAPM was utilised in tracer mode (with no

chemistry) based on three tracers: one for domestic emissions, one for transport emissions, and one for industrial emissions as defined in the local emission inventory (Scott & Ganatilaka 2004). For a more detailed explanation of the inventory used, see the report by Scott and Ganatilaka (2004) and an earlier study that, whilst not comparing intraurban concentrations, utilised an identical emission inventory (Zawar-Reza *et al.* 2005). Because of the coarse scale of emission inventories, performance of the meteorological component of TAPM was critical to accurate estimations of PM concentrations in small areas. To improve the simulated meteorological field over Christchurch, data assimilation was performed. Wind direction and speed were assimilated into the model from the COLE monitoring site, which is operated by the local environmental council. The zone of influence for the measurements were set for a 5 km radius with a relatively strong forcing value of 0.8 (a value of 1 forces the model to reproduce the monitored data exactly). TAPM simulations for this work were conducted on a PC with a 1.6 GHz processor running version 2.0 of TAPM. Each simulation presented here took five days of runtime to complete.

4.4.4. Data analysis

Daily (24-hr) averages from monitored data were compared with 24-hr averages extracted from the corresponding TAPM grid cells. To assess the agreement between the model and the monitored data, an index of agreement (IOA) was calculated, defined as (4.1):

$$IOA = 1 - \frac{\sum_i (O_i - P_i)^2}{\sum_i (\left|O_i - \overline{O}\right| + \left|P_i + \overline{O}\right|)^2} \qquad (4.1)$$

where the predicted values (P) of the model are compared with the observed values (O) measured by the monitoring network for i samples (Willmott 1981). The IOA is a measure of skill of the model in predicting variations about the observed mean; a value above 0.5 is considered to be good. The error in the model was assessed using Root Mean Square Error (RMSE), as defined by Willmott (1981). There are several types of RMSE, including systematic RMSE ($RMSE_S$), which denotes predictable error inherent in the model and unsystematic RMSE

($RMSE_U$), which indicates error related to factors not intrinsic to the model (Willmott 1981, Willmott 1982). In high-quality models, the $RMSE_S$ should be much smaller than the $RMSE_U$ (Willmott 1982). $RMSE_S$ (4.2) and $RMSE_U$ (4.3) are defined as:

$$RMSE_S = \frac{[\sum_{i=1}^{n} (O_i - fit_i)^2]^{1/2}}{n} \quad (4.2)$$

$$RMSE_U = \frac{[\sum_{i=1}^{n} (P_i - fit_i)^2]^{1/2}}{n} \quad (4.3)$$

where *O* is the observed concentration value at a site, P is the predicted concentration value, and *fit* is the value predicted by the linear fit between the model and the observation. A coefficient of divergence (COD) was utilised to describe relative intraurban uniformity, defined mathematically as (4.4):

$$COD_{jk} = [\frac{1}{p} \sum_{i=1}^{p} [(x_{ij} - x_{ik})/(x_{ij} + x_{ik})]^2]^{1/2} \quad (4.4)$$

where x_{ij} and x_{ik} represent the 24-hr average particulate concentration values at a site for sampling day *i* with observed concentration of *j* and modelled concentration of *k*, and where *p* is the number of observations (Wongphatarakul *et al.* 1998). A COD of 0 means there are no differences between modelled and observed concentrations, while a value approaching 1 indicates a maximum difference. As another means of comparing observed and modelled daily absolute concentration variability, the 90th percentile of daily absolute concentration differences (P_{90}) between the modelled and observed daily concentrations was calculated at each site. In order to analyse the performance of the model spatially over the city, a contour plot was constructed using spatial analyst extension in ESRI ArcMap version 9.0. Concentrations between 1500 m grid points (TAPM output grid) were calculated using a spline interpolation in the spatial

analyst extension with 100 m resolution and five points of interpolation. Excel 2003 (Microsoft 2003) and SPSS version 11.0 (SPSS 2001) were used for the statistical analysis.

4.5. Results

Twenty-four-hour PM_{10} concentrations were obtained for eleven monitoring sites over 59 mid-winter days in July 2003 and June 2004. Two full days of data (June 24th and 25th, 2004) were not monitored, and three sites (Christchurch East (CHCE), Oaklands (OAKL), and Avonhead (AVON)) were missing data during one additional day due to battery malfunction. Table 4.1 displays the summary statistics for monitored and modelled data at the 11 intraurban locations and Figure 4.2 illustrates the mean observed and modelled concentrations for each site. The arithmetic mean of the eleven sites during the monitoring period was 42.9 μg m^{-3} with a standard deviation of 25.3 μg m^{-3}, while the mean of modelled concentrations was 43.4 μg m^{-3} with a standard deviation of 33.0 μg m^{-3}. Mean minimum observed and modelled concentrations were 8.8 μg m^{-3} and 4.5 μg m^{-3} respectively, while mean maximum concentrations were 128.3 μg m^{-3} and 128.2 μg m^{-3} respectively. Maximum observed values for ten of the monitoring sites ranged from 93.6 to 171.3 μg m^{-3}, compared with a modelled range of 85.6 to 205.5 μg m^{-3}, while the MTPL site had substantially lower observed and modelled maximums of 38.9 μg m^{-3} and 35.1 μg m^{-3} respectively. The low concentrations at the MTPL site were due to its location on the Port Hills, where cold drainage flows transport most wintertime pollution towards lower elevations over the city (Spronken-Smith et al., 2001)**.** The number of monitored days over the 24-hr ambient guideline of 50 μg m^{-3} ranged from zero at MTPL for both observed and modelled data, to 27 observed exceedances at COLE and 32 modelled exceedances at COLE and Addington (ADDN). The model underestimated high nights by as many as 15 nights at OAKL and overestimated a maximum of 13 nights at ADDN with an observed mean of 18 nights over the guideline and a mean 20 exceedances for modelled data.

Table 4.1. Summary statistics for winter monitoring and modelled data.

Site name	Site ID	Observed					Modelled			
		n	Mean ± SD*	Min*	Max*	Exceedances[1]	Mean ± SD*	Min*	Max*	Exceedances[1]
Hammersley Park	HAMP	59	47.8 ± 29.7	8.7	133.0	19	35.1 ± 30.0	2.1	114.6	16
Christchurch East	CHCE	58	44.7 ± 29.4	6.8	148.9	20	64.4 ± 55.0	4.7	205.5	28
Fendalton	FEND	59	43.6 ± 26.3	11.0	132.8	19	59.1 ± 42.9	5.2	156.8	29
Addington	ADDN	59	49.3 ± 28.9	14.5	147.5	19	66.8 ± 50.8	5.2	188.0	32
Woolston	WOOL	59	52.6 ± 33.2	13.7	171.3	23	46.5 ± 32.9	4.8	124.8	27
Wainoni	WAIN	59	48.6 ± 30.7	9.3	154.0	23	53.3 ± 39.8	5.8	144.5	28
Avonhead	AVON	58	40.5 ± 21.8	3.2	95.4	18	41.5 ± 30.5	6.7	122.7	20
Oaklands	OAKL	58	44.1 ± 26.7	11.2	167.2	18	20.8 ± 16.7	2.3	85.6	3
Mount Pleasant	MTPL	59	16.3 ± 8.0	1.9	38.9	0	10.9 ± 7.5	1.9	35.1	0
Hornby	HORN	59	41.7 ± 18.3	7.3	93.6	19	35.5 ± 23.9	6.6	104.6	14
Coles Place	COLE	59	53.6 ± 34.5	7.6	144.5	27	65.3 ± 51.9	4.8	189.5	32
Arithmetic mean	MEAN	59	42.9 ± 25.3	8.8	128.3	18	43.4 ± 33.0	4.5	128.2	20

*in $\mu g\ m^{-3}$. [1]Number of 24-hr average periods exceeding the ambient PM_{10} guideline of 50 $\mu g\ m^{-3}$.

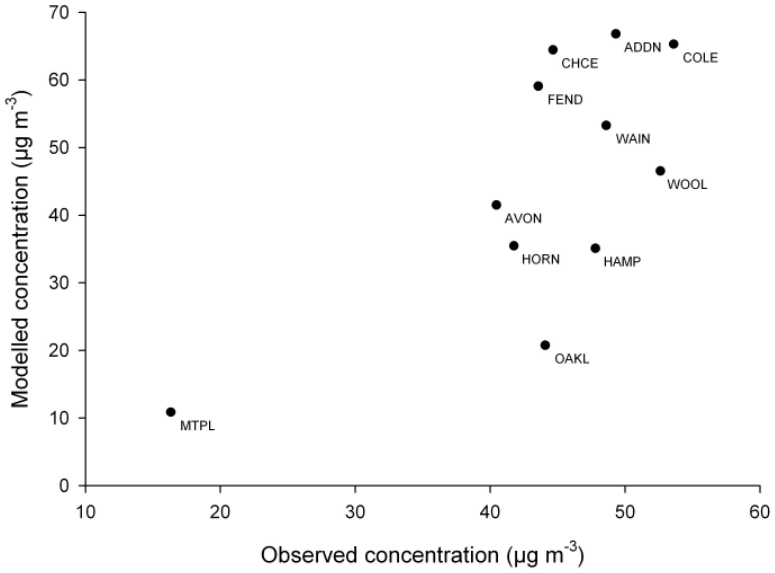


Figure 4.2. Mean observed and mean modelled concentrations at monitoring sites.

Comparative statistics indicate that the model performed well for average long-term values (Table 4.2). Mean IOA (1) was 0.60, with a minimum of 0.40 at MTPL and a maximum of 0.70 at Wainoni (WAIN). At all sites except MTPL, the model scored ≥ 0.53, indicating good agreement. RMSE (2) averaged 34.38 μg m^{-3} for all sites, with the lowest value at MTPL (12.17 μg m^{-3}) and the highest value at Christchurch East (CHCE; RMSE = 50.97 μg m^{-3}). Mean $RMSE_S$ for the model was 16.90 μg m^{-3}, far less than the $RMSE_U$ of 28.89 μg m^{-3}, indicating that the error was largely due to influences outside the model. $RMSE_S$ was greater than $RMSE_U$ at only one site, OAKL, on the western and southern fringe of the Christchurch urban area. The IOAs for wind speed, wind direction, and temperature at COLE were 0.63, 0.75, and 0.87 respectively, indicating that the model sufficiently defines the meteorology over the urban area.

Table 4.2. Summary statistics of model fit.

Site ID	IOA[1]	RMSE[2]	$RMSE_S$[3]	$RMSE_U$[4]	r[5]	COD[6]	P_{90}[7]
HAMP	0.63	34.41	21.25	27.07	0.41*	0.44	62.6
CHCE	0.60	50.97	19.83	46.95	0.52*	0.42	86.3
FEND	0.60	41.26	16.79	37.69	0.42*	0.37	63.7
ADDN	0.65	45.57	17.52	42.06	0.53*	0.37	85.7
WOOL	0.66	35.08	19.41	29.22	0.46*	0.39	59.7
WAIN	0.70	35.90	11.47	34.02	0.50*	0.36	65.0
AVON	0.57	30.60	11.37	28.41	0.47*	0.34	47.0
OAKL	0.61	31.60	28.65	13.32	0.48*	0.49	53.2
MTPL	0.40	12.17	9.64	7.43	0.09	0.42	19.6
HORN	0.53	26.22	13.10	22.71	0.46*	0.33	39.9
COLE	0.65	47.48	15.00	45.05	0.51*	0.40	87.4
MEAN	0.60	34.38	16.90	28.89	0.44	0.39	58.3

[1]Index of Agreement. [2]Root mean squared error (RMSE) in µg m^{-3}. [3] Systematic RMSE in µg m^{-3}.
[4]Unsystematic RMSE in µg m^{-3}. [5]Pearson correlation; *Indicates significant to the 0.95 level.
[6]Coefficient of Divergence. [7]90^{th} percentiles of absolute daily differences between observed and modelled.

Figure 4.3 indicates that from a spatial perspective, there appears to be more agreement towards the central business district than on the outer fringes. Sites on the edges of the city, where emission inventories are not as accurate, yielded lower agreement with the model. The lowest IOAs were for MTPL, HORN, and AVON, all of which are on the hills or the south and western fringes of the city. The highest agreement with the model was found at the WAIN, WOOL, ADDN and COLE sites, which are more centrally located in the urban area. The likely causes of the lower IOAs on the southern and western fringes of the city are the complex wind interactions (Figure 4.1) combined with lower resolution emission inventories on that edge of the urban area.

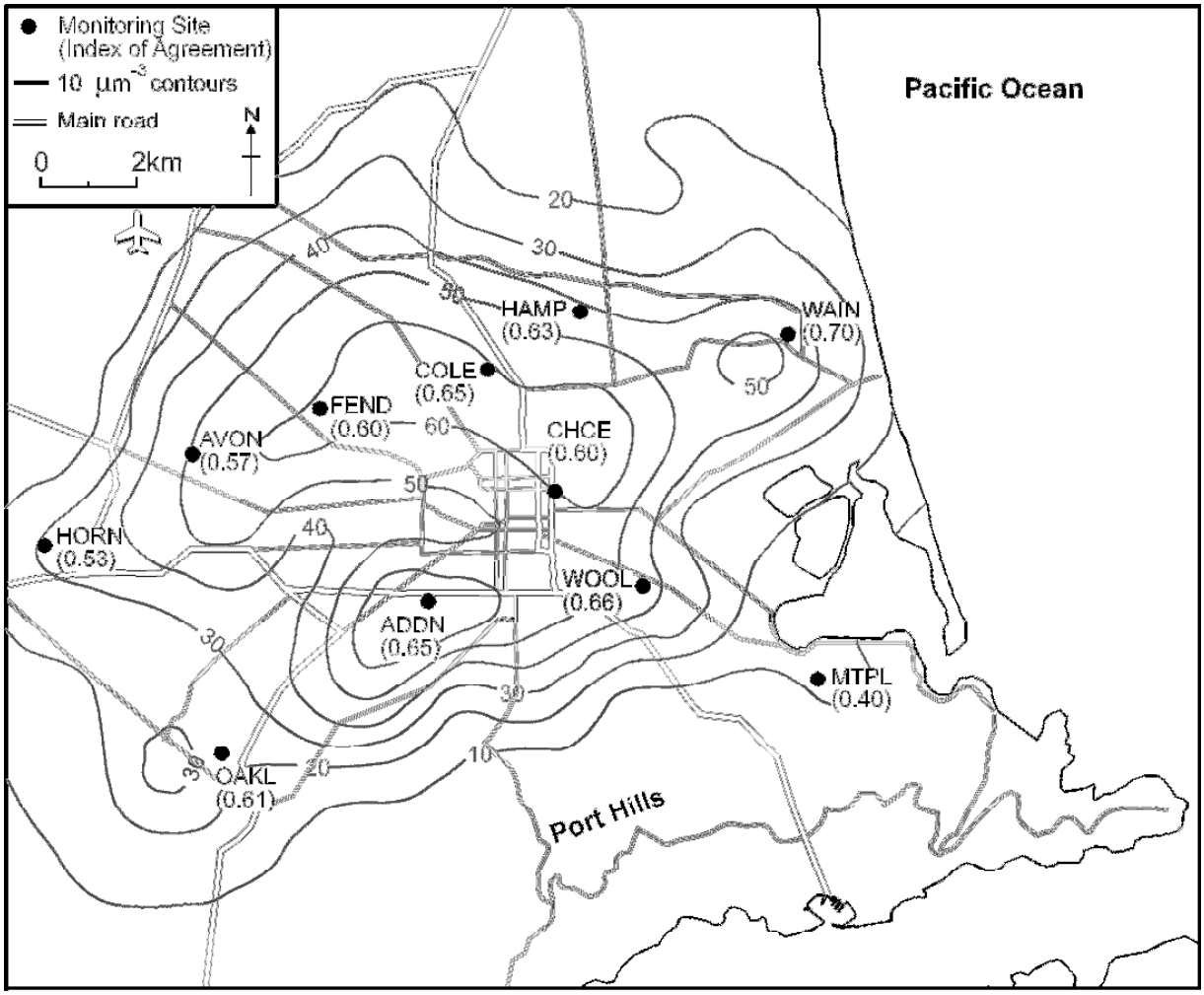


Figure 4.3. Contours (10 µg m^{-3} intervals) of winter average modelled PM_{10} concentrations and Index of Agreement (IOA) at monitoring station locations. An IOA > 0.50 indicates good agreement. The key for site names can be found in Table 4.1.

While average modelled concentrations over the study period were found to be fairly accurate, daily statistical measures of relative and absolute relationships between observed and modelled values indicate that there is significant room for improved performance. The correlations of log-transformed observed versus modelled daily concentrations were mostly significant (except for MTPL) and low (mean r = 0.44). A relatively high COD (3) (mean COD = 0.39) and substantial daily differences between modelled and observed data (mean P_{90} = 58.3 μg m^{-3}), along with the low correlations in daily values indicate that TAPM may not be an effective tool for estimating personal PM exposures in daily time-series studies under the settings described here. However, the more accurate long-term predictions indicate utility for longer-term cohort epidemiological study designs.

4.6. Discussion

The results of this study indicate that TAPM, an integrated meteorological-emission model, predicts medium- to long-term intraurban particulate concentrations with sufficient accuracy, although there is considerable room for improvement. Mean observed and modelled values differed by only 0.5 μg m^{-3} over the study period with a sufficiently high index of agreement (mean IOA = 0.60). Although intraurban particulate concentrations have been established as a priority area of research, there have been no within-community evaluations of integrated meteorological-emission models for use in epidemiological applications conducted to date (Jerrett *et al.* 2005a).

There are several factors that render the accuracy of the modelled concentrations particularly compelling. First, the particulate concentrations in Christchurch have been shown to exhibit considerable intraurban variability. In a recent study, the mean spatial variation between daily intraurban site concentrations was shown to be significantly higher than in most large urban areas in the United States, especially those in the central and eastern regions of the country (Pinto *et al.* 2004, Wilson *et al.* 2006). This variability is a reflection of the nature of the air pollution problem in Christchurch: emissions are from a network of heterogeneously spaced point sources – houses – releasing PM_{10} into an atmospheric inversion layer that suppresses vertical mixing (due to atmospheric stability at night, the size spectrum of turbulent eddies is much smaller than the space between each individual point source). Second, the topography

around Christchurch leads to a complex wind field at night. The nocturnal surface wind field is an interplay between drainage flows from the Port Hills and the Southern Alps and pushes mesoscale models to their limit (Kossmann & Sturman 2004). The poor prediction capability of the model at the Oaklands and Addington sites is likely due to the misplacement of the convergence zone (see Figure 4.1: Kossmann & Sturman 2004). To improve the modelled meteorology, data was assimilated from the only available urban weather station (Coles Place), although TAPM can incorporate many more stations. Additional assimilation sites may increase the accuracy of the meteorological component of the model, enhancing the prediction capability. Finally, given the coarse spatial nature of the emission inventory available in Christchurch, the results of the model were relatively accurate. Many cities in North America and Europe have far greater spatio-temporal resolution in their emission inventories. TAPM may yield closer approximations of actual intraurban conditions when modelling in cities with less intraurban variability, less meteorological-topographical complexity or higher spatio-temporal resolution in their respective emission inventories.

There are several limitations of IME models in general and also of this study specifically. One limitation related to the use of integrated meteorological models such as TAPM is that emissions inventories are required to operate the models. Many urban areas where researchers are interested in modelling small-scale PM variability may not have detailed emissions data, or in some cases, no emissions inventory data at all. The less detail available in the emission inventory, the more likely large unsystematic error will be induced. Second, as highlighted by Jerrett *et al.* (2005a), personnel costs and run-times associated with IME models may be high. The expertise required to configure and run TAPM may not be as high as for other IME models, such as MM5 or RAMS, yet considerable experience and skill are still required. The simulation time may also be an issue. For this two-month analysis of intraurban concentrations, a dedicated computer was used for ten days. Were this computer to run this model to estimate intraurban levels of pollution over a long study period, say, ten years, the computer processing time would be 600 days (~1 year and 8 months). However, this processing time may be shortened by faster CPUs and lower spatial resolution. Of importance to note is that although the model gave a good estimation across sites, there was significant systematic error (mean $RMSE_S$ = 16.9 $\mu g\ m^{-3}$) at individual sites. While this error may be smaller than that produced by using a central site as an

exposure estimate in areas with high spatial heterogeneity, further work should be done to verify the model's accuracy and improve predictions at the intraurban scale. Our study examined only winter months, as guidelines are exceeded in Christchurch typically in the months from May to August. However, as most cohort studies use long-term (e.g., annual) average exposure values, IME models should be evaluated and verified using both winter and summer intraurban monitored data. Lastly, the most significant limitation in the study may be the coarse grid resolution. As noted by Jerrett *et al.* (2005a), a 1 km grid size (this study evaluated a 1.5 km grid) may not provide sufficient spatial resolution for pollutants with high daily spatial variability at the 50 - 100 m scales, such as NO_2, NO_x, SO_2 and CO. Larger size fractions of PM such as PM_{10} and $PM_{10-2.5}$, are also known to be highly variable at smaller spatial scales (Monn 2001).

Models that accurately ascertain intraurban exposures may prove to have significant applications for epidemiological cohort studies. Cohort studies generally rely on the identification of groups with distinctive levels of exposure over long periods of time (Dominici *et al.* 2003). To date, this exposure variation has generally been found between groups by comparing populations between different cities while controlling for variables that may differ between populations, such as age, gender, socio-economic status, smoking and other personal factors (e.g., Dockery *et al.* 1993a, Pope *et al.* 1995b, Pope *et al.* 2002). Some of these inter-community cohort studies have been able to control for components of error due to exposure misclassification by using personal monitors and monitors in homes and comparing these concentrations with those recorded at central ambient sites (Dockery *et al.* 1993a). Although the degree of error between personal and measured pollution may be quantifiable, it is more difficult to assess the health effects of variability in population demographics and particulate species and source composition, which have been shown to be quite heterogeneous even between cities in similar regions (Wongphatarakul *et al.* 1998, Pinto *et al.* 2004, Grahame & Schlesinger 2005, Kim *et al.* 2005). IME models may provide sufficient spatial resolution for conducting long-term cohort studies *within* cities, eliminating some of the possible confounding induced by comparing across diverse airsheds and populations. The use of IME models in these studies is a logical next step in the progress of research, given the results presented here.

In conclusion, an integrated meteorological-emission model, TAPM, predicted intraurban PM_{10} concentration with reasonable accuracy, in an area with a low-resolution emission inventory, complex topography and meteorology. IME models appear to be a promising method for estimating personal exposures in ecological or between-city studies, and show potential for use at the intraurban level. If IME models prove to be successful at modelling intraurban concentrations in future work, they may be a useful tool for studying the health effects of long-term intra-community particulate exposure.

Chapter 5: The spatial relocation effect on intraurban patterns of risk in respiratory hospital admissions due to adjusting for air pollution and deprivation covariates.

5.1. Summary

Epidemiological studies that examine the relationship between exposure and health outcomes often address factors such as deprivation by adjusting for them as covariates. While disease surveillance methods routinely control for covariates such as environmental exposure or measures of deprivation, there has been limited investigative work on the spatial movement of risk at the intraurban scale due to the adjustment. It is important that the nature of any spatial relocation be well understood as a relocation to areas of increased of risk may introduce additional localised factors that influence the exposure-response relationship. This paper examines the patterns of relative risk and clusters of disease by means of spatial analysis techniques applied to an illustrative small-area example from Christchurch, New Zealand. A four-stage test of the effects of covariate adjustment was performed. First, relative risks for respiratory hospitalisations from 1999 to 2004 at the census area unit level were adjusted for age and sex. In three subsequent tests, admissions were adjusted for annual exposure to particulate matter less than 10 micrometers in diameter (PM_{10}), then for a deprivation index, and finally for both PM and deprivation. Spatial patterns of risk, disease clusters and cold and hot spots were generated using a spatial scan statistic and a Getis-Ord Gi* statistic. In all disease groups tested (except the control disease), adjustment for chronic PM_{10} exposure and deprivation modified the position of clusters substantially, as well as notably shifting patterns and hot/cold spots of relative risk across the city. Adjusting for PM_{10} and/or for deprivation shifted clusters in a similar spatial fashion. In Christchurch, the resulting shift relocated the cluster from a purely residential area to a mixed residential/industrial area, possibly introducing new environmental exposures. Researchers should be aware of the potential spatial relocation effects inherent in adjusting for covariates when considering study design and conducting disease surveillance.

5.2. Introduction

An almost ubiquitous method used in ecological or aggregated studies assessing the relationship between contextual and individual determinants of health is the adjustment for determinants that may influence the relationship under study. By correcting for these determinants as covariates, their effects may be, in theory, removed from influencing the relationship under study. While many studies have examined the effects on risk ratios and ecological bias that may occur when controlling for various factors (Schwartz 2000, Katsouyanni *et al.* 2001) the spatial nature of these modification effects at the intraurban scale is not well understood. Thus, the *spatial relocation* of risk may have implications for epidemiological work, as shifts in patterns and locations may change the nature of exposures and environmental risks under study.

Although the analysis of health in space arguably began over one hundred and fifty years ago with the work of John Snow on Cholera's proximity to water pumps in London (Snow 1855), the field has experienced rapid growth recently due to advances in spatial statistical methods and technology. Geographic Information Systems (GIS) now show great potential for environmental-health relationships to be ascertained at increasingly detailed scales (Vine *et al.* 1997, Järup 2004, Nuckols *et al.* 2004). The study of small-area exposures and effects at the intraurban (within-city) scale and below have recently become a priority area of study (Jerrett *et al.* 2005). With the move towards increasingly finer scales of aggregation, it is becoming more crucial to understand the spatial relocation effects of applying methods that have been tested at larger scales (Best *et al.* 2001). Further, as we understand the nature of exposures and pathways of risk at finer scales, it becomes important appreciate the relocation effects of adjusting for covariates, including additional exposures that may be introduced in the process.

The aims of this paper are, firstly, to determine and visually represent spatial trends in relative risks and clusters of hospital admissions for respiratory disease across an urban area while adjusting for an environmental determinant of disease and a measure of deprivation. Next, we aim to quantify the nature of these patterns and trends through spatial statistical techniques and examples from Christchurch, New Zealand. Finally, we comment on the implications of spatial relocation for the analysis of disease patterns and surveillance. In should be noted that the focus

of this paper is methodological – we focus on the patterns of disease and the shifts in patterns, not the specific underlying causes or implications of disease in Christchurch.

5.3. Surveillance and covariate adjustment

This Chapter focuses on spatial characterisation of risk and the shifts in risk associated with adjustment for covariates. One inherently spatio-temporal type of analysis is disease surveillance, which may be either passive or active. Passive surveillance relies on available data and is retrospective in nature, while active surveillance denotes a system in which researchers gather data real-time in the field based on a specific hypothesis or in the event of an environmental contamination (Gordis 2000). While surveillance is often conducted on infectious diseases such as malaria, it is increasingly important for monitoring changes of conditions such as congenital malformations, cancer and asthma (Gordis 2000). Various methods have been applied to addressing aspects of disease (e.g., multi-level modelling, geographically weighted regression) and disease surveillance, although there has been a limited focus on shifts in risk due to environmental and deprivation covariate adjustment.

Scale plays an important role in potential spatial relocation effects. The behaviour of spatial phenomena is often a result of 1st and 2nd order effects (Bailey & Gatrell 1995). First order effects relate to the intensity of global or large scale trends. Second order effects result from the spatial dependence in the process, i.e., the tendency for locations close in space to have similar values, which are local or finer-effects. If the process has a second order effect which alters over the study region, we can say the process exhibits *non-stationarity* or *heterogeneity*. In this paper we are concerned with modelling the second order heterogeneity, or the residuals or deviations from the 1st order trend. Covariates are often adjusted for globally within a dataset, without full consideration of the (local) geographical implications with respect to spatial processes. This is less of an issue if considering the entire study region, but the consequences could be profound if the scale of investigation is the spatial structure of a relationship itself.

Particulate matter (PM) air pollution has been widely studied and is one of the most recognised environmental risk factors for cardio-respiratory health complications, including both mortality (e.g., Dockery *et al.* 1993) and morbidity (e.g., Schwartz 1996). Air pollution may be considered

either a risk factor or a potential covariate, depending on the study design and outcomes under study. In most research, including the studies cited above, particulate matter is an independent variable in the analysis against the dependent health outcome. However, ambient PM might be controlled for as a covariate, for example, in a surveillance study examining temporal changes in rates of lung cancer in proximity to exposures from an industrial point source. PM concentrations have often been studied in relation to its modification effects on the relationship between contextual social factors and health (Jerrett *et al.* 2004). Small-area intraurban (within-city) concentration exposure to particulate matter has recently emerged as a priority area of research in wider epidemiological studies (Monn 2001, Jerrett *et al.* 2004, Jerrett *et al.* 2005).

Social deprivation is a common factor which is controlled for in surveillance studies as a covariate. Deprivation is often calculated by means of an index of weighted factors (Morris 1991). Weighted factors include measures of education, income, home ownership, and access to transport, among others. Although deprivation has been shown to be indirectly and contextually associated with some forms of disease (e.g., respiratory and circulatory illness) it has also been shown to be inversely related to some forms of disease (e.g., breast cancer) and has shown no association with others (Saul & Payne 1999, Finkelstein *et al.* 2005).

5.4. Methods

5.4.1. Study area

Christchurch is a coastal mid-latitude (43ºS) city of 320,000 located on the South Island of New Zealand. Air pollution has been a significant and persistent problem associated with winter temperature inversions in Christchurch since the late 1800s and remains a considerable health risk to the current population (Gray 1889, McGowan *et al.* 2002). Christchurch reports on average 30 exceedances over the 50 $\mu g\ m^{-3}$ 24-hr guideline set by the New Zealand Ministry for the Environment for PM_{10} every winter (Aberkane *et al.* 2004). Approximately 80% of Christchurch's PM air pollution arises from domestic heating emissions from the combustion of solid fuels (Scott & Ganatilaka 2004). Several Christchurch studies have associated daily winter PM_{10} variations with morbidity and mortality, although these have all been aggregated to the

urban scale (Hales *et al.* 2000, McGowan *et al.* 2002) – a scale which has been shown to exhibit heterogeneous concentrations in Christchurch (Wilson *et al.* 2006).

5.4.2. Data

Census population data for 1996 and 2001 were obtained from Statistics New Zealand for each Census Area Unit (CAU) in Christchurch and included age, deprivation and gender. A CAU is the second smallest unit of dissemination of census data in New Zealand, representing approximately 2,300 people. The CAU level was chosen in place of the finer unit (mesh blocks) due to the scaling of our pollution exposure measurement (see below). Linear interpolation was used to calculate the annual population in each CAU between census years and after 2001. Age was re-coded into six categories (< 5, 5 – 14, 15 – 24, 25 – 39, 40 – 64, and 65+) and gender was re-coded into male and female categories to create 12 age-sex groups per CAU.

Deprivation was defined using the 2001 New Zealand Deprivation Index (NZDep), a weighted index calculated by the Ministry of Health and Wellington School of Medicine and Health Sciences as a means of describing deprivation relative to the whole of New Zealand. The deprivation index is determined by nine census variables including employment status, income, access to transport and communication services, home characteristics, and education, and ranges from one (least deprived) to ten (most deprived) (Salmond & Crampton 2002). The NZDep raw scores were aggregated into quartiles at the CAU level based on relative deprivation within the Christchurch study area (Figure 5.1a).

Particulate air pollution data in this study was derived from an air pollution dispersion model, The Air Pollution Model (TAPM). Validation work on TAPM in Christchurch has shown that the model performs well over longer averaging times (Zawar-Reza *et al.* 2005, Wilson *et al.* 2006). In a one-year comparison, TAPM's estimate of 22 μg m^{-3} was slightly higher than the measured annual level of 18 μg m^{-3} while hourly modeled and measured values were similar to the 99^{th} percentile (Zawar-Reza *et al.* 2005). The measured concentrations may have been slightly underestimated due a lack of correction for lost volatiles associated with heated samples in woodsmoke pollution environments – this fact further enhances the confidence in the model's

estimate of long-term exposure (Allen *et al.* 1997). TAPM annual concentration outputs are produced on a grid with 1,500 m resolution. This gridded annual data was input into a GIS, ArcGIS 9.1, and then interpolated using a regular spline with five points of interpolation and a 100 m grid resolution. The zonal statistics tool in spatial analyst was then used to calculate the mean concentration for each CAU. The resulting concentrations by CAU were re-coded into quartiles to be used as covariates in the statistical analysis (Figure 5.1b). It should be noted that annual concentrations between adjacent TAPM grid cell outputs are not highly variable (see Fig. 1 in Pearce *et al.* 2006), so the error associated with down-scaling was most likely negligible, given that the data was re-coded into quartiles before being entered into the analysis rather than using actual concentration estimates for each CAU.

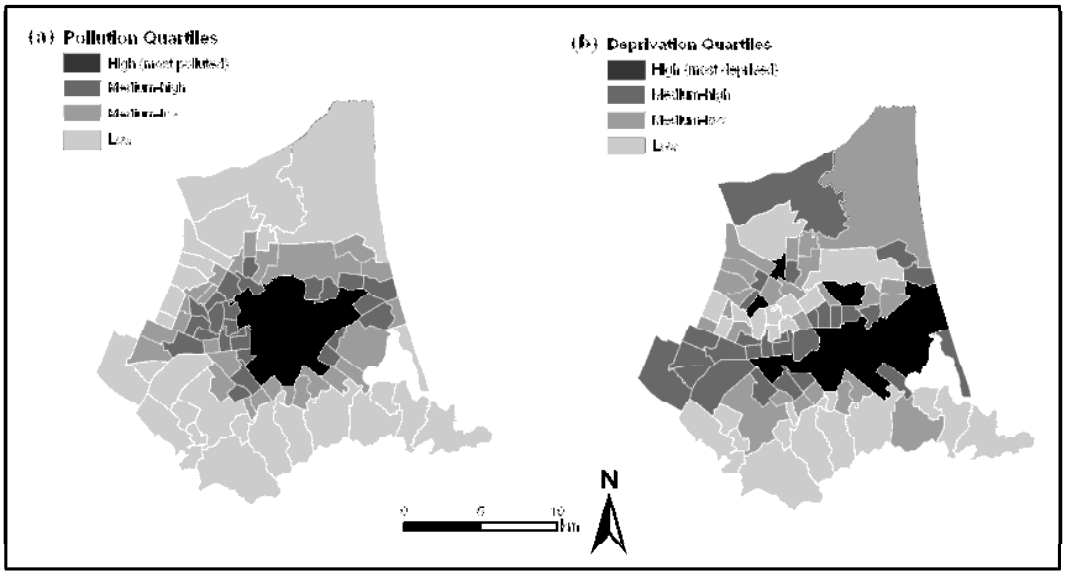


Figure 5.1. Quartiles of (a) annual exposure to particulate matter air pollution and (b) deprivation index scores at the census area unit level.

Hospital admissions data for the years 1999-2004 were obtained from the New Zealand Health Information Service. Primary and secondary diagnoses of International Classification of Disease (ICD–9 or ICD-9 adjusted equivalent) codes for all diseases of the respiratory system (ICD–9 460-519) were included as well as admission data for acute appendicitis (540-542). Acute appendicitis was used as a control in the study, as appendicitis risk is not known to vary substantially as a function of either deprivation or chronic pollution exposure (Burkitt 1971). Four separate disease groupings were investigated within the respiratory set: (i) all respiratory disease (460-519), (ii) acute respiratory illness (460-466), (iv) pneumonia and influenza (480-487), and asthma (493). The final study area for data capture contained 102 CAUs (Figures 1a and 1b).

In total, there were 30,796 total respiratory admissions to hospital for respiratory disease (ICD-9 460-519) over the six years ending in 2004. Due to incomplete domicile/CAU data for individuals, 182 (0.59%) of the hospital admissions were removed from the data set, leaving 30,614 admissions. There were 2008 admissions to hospital for appendicitis over the same period, of which 18 were removed for incomplete domicile coding, leaving 1990 total appendicitis admissions. There is only one major admitting hospital in Christchurch, so capture of initial admissions is not a significant issue with the dataset. Three CAUs on the western boundary of the metropolitan area were omitted from the analysis, as the Templeton Mental Hospital (which also provided initial admissions for respiratory disease) closed there in 2002 and the resulting shift in admissions to the central hospital could not be adequately accounted for. The final study area for data capture contained 102 CAUs (Figures 1a and 1b).

5.4.3. Analysis

Clusters of disease were determined and analysed by means of a spatial scan statistic, calculated using the SaTScan (v. 3.1) software package available from the U.S. National Cancer Institute (Kulldorff 1997). The spatial scan statistic is commonly used in health applications as a method of determining clusters because: (i) it eliminates the problem of pre–selection bias by searching for clusters without specifying their sizes or locations, (ii) under a situation where the null hypothesis is rejected (i.e., when the null hypothesis is that no statistically significant spatial clusters exist when cases are assumed to follow a Poisson distribution in space), the approximate location of the cluster that causes the rejection can still be located, (iii) it is suitable for heterogeneous population density (i.e., the population of

Christchurch), (iv) secondary clusters can also be reported, and (v) the method avoids the problem of multiple testing present in most methods which alter the size of the tested population. The spatial scan statistic has been used to test for disease clustering in a number of recent studies, including: a focused test investigating potential clusters of soft-tissue sarcoma and non-Hodgkin's lymphoma around a solid waste incinerator in France (Viel *et al.* 2000); breast cancer in the United States (Kulldorff *et al.* 1997); amyotrophic lateral sclerosis (ALS) birthplace clustering in Finland (Sabel *et al.* 2003); and reports of lower respiratory disease in a large group practice in the United States (Kleinman *et al.* 2005).

The method used by SaTScan imposes a circular scanning window on the map and lets the centre of the circle move over the study area so that at each position the window includes different sets of neighbouring administrative areas. For each circle centroid, the radius varies continuously from zero to a user-defined maximum. SaTScan evaluates the statistical significance within each population window by examining the likelihood ratio, which is maximized over all the windows to identify the most likely disease clusters (Kulldorff 2002).

SaTScan requires three files for input: a case file, a population file, and a location file. The case file was created from hospital admission records, the population file from Statistics New Zealand census data, and the location file was created using a GIS (ArcGIS 9.1). A Poisson distribution model was selected (the other option, a Bernoulli model, is only available with 0/1 event data such as cases and controls) with a 50% population window scanning for high clusters. Several size windows were tested, but the 50% window achieved the most consistent results – this is also the recommended window for maximum detection (Kulldorff 2002). In addition to cluster data, SaTScan also outputs a file that estimates the relative risk (RR) in each CAU of the health outcome under study, after indirectly adjusting for variables as covariates. These adjusted relative risk estimates were used as the basis for exploratory spatial analysis, which has been shown to be useful in identifying variability in disease risk (Lawson & Kulldorff 1999). It should be noted that SaTScan's method of covariate adjustment process is not a 'step-wise' adjustment, i.e., adjusting for a first covariate, then another. Rather, SaTScan adjusts for the all the variables entered as well as the interaction terms of those variables. As a result, the order of the covariates is inconsequential – we chose to follow standard epidemiological convention in the 'order' of our tests – first controlling for pollution (test B), then for our measure of deprivation (test B). Surfaces for each disease category were created using a regularized spline interpolation in ArcGIS 9.1 with the adjusted

relative risk for each CAU centroid as the z-value, a 0.1 weighting, five points of interpolation, and a 100 m output cell size (ESRI 2004). The 100 m cell size was chosen as it best approximated the surface of risk for display purposes.

As a means of quantitatively comparing patterns of risk, a Getis–Ord Gi* statistic was calculated for each CAU (Getis & Ord 1992). The Hot Spot Analysis tool in ArcGIS 9.1 was used to identify spatial clusters of statistically significant high or low attribute values by indicating whether high values or low values (but not both) tend to cluster in a study area (ESRI 2004). A high value for the Gi*-statistic indicates that high values (i.e., higher than the mean for the study area) are found close to one another. A low value for the Gi*-statistic indicates that values lower than the mean tend to be found together (i.e., a 'cold spot'). The input field for the tool was the respective adjusted RR of each CAU, with a threshold distance of 3 km. Distances from 2 km to 5 km were tested as possible thresholds and 3 km yielded the most consistent results.

Relative risks, clusters, and hot spot analyses were performed for all respiratory disease groupings in four 'test' stages. First, a baseline analysis was conducted which only adjusted for age/sex (test a). Secondly, age/sex and particulate matter air pollution were controlled for (test b) followed by an analysis of patterns when age/sex and deprivation were controlled for (test c). Lastly, age/sex, air pollution and deprivation were adjusted for as multiple covariates (test d). ArcGIS was used to visualise the relative risks, clusters and hot/cold spots for each type of analysis and for every disease code grouping. Spatial relocation vectors (azimuth and distance) were calculate based on the movement from the baseline age/sex cluster (test a) to the centres of most likely clusters in tests b, c and d. In all, 60 analyses (3 spatial analyses x 5 disease groups x 4 covariate tests each) were completed.

5.5. Results

All focus disease groupings under study showed marked shifts in spatial patterns of risk when controlling for additional factors beyond the age/sex adjusted baseline (tests b, c and d). Figures 2 and 3 illustrate the nature of the spatial shifts in patterns of relative risks and clusters across the urban area for pneumonia and influenza and asthma, respectively. Areas of high (black) and lower (white) adjusted relative risks experienced shift upon visual inspection. Reassuringly, there was no observable shift in the patterns of risk for appendicitis control in any of the four tests.

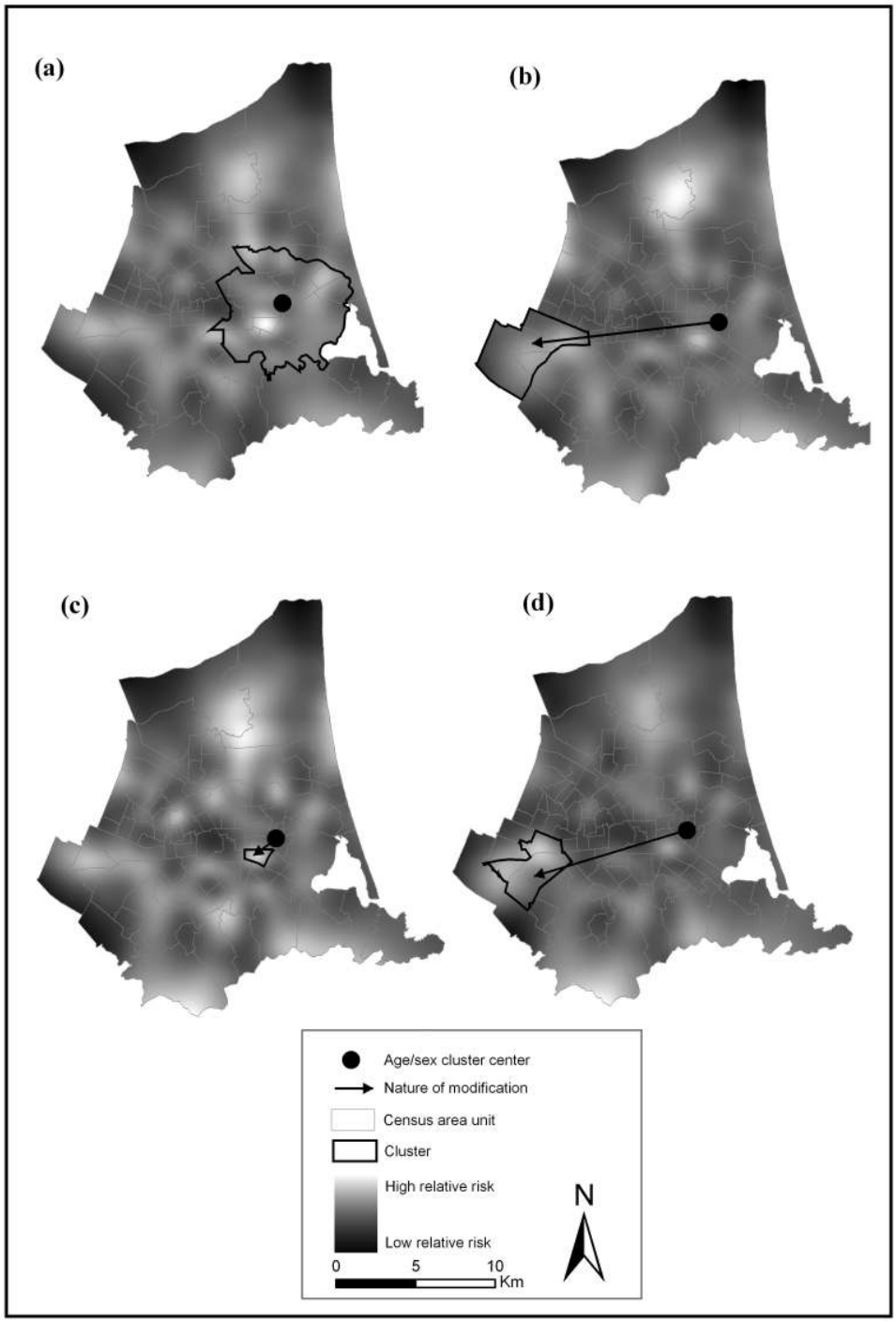


Figure 5.2. Spatial patterns of relative risk and spatial relocation of pneumonia and influenza clusters after adjusting for (a) age and sex; (b) age, sex and deprivation; (c) age, sex and air pollution; and (d) age, sex, air pollution and deprivation. Arrows indicate the distance and direction from the centre of the age/sex cluster to covariate-adjusted clusters with the endpoint of the vector marking the new centre.

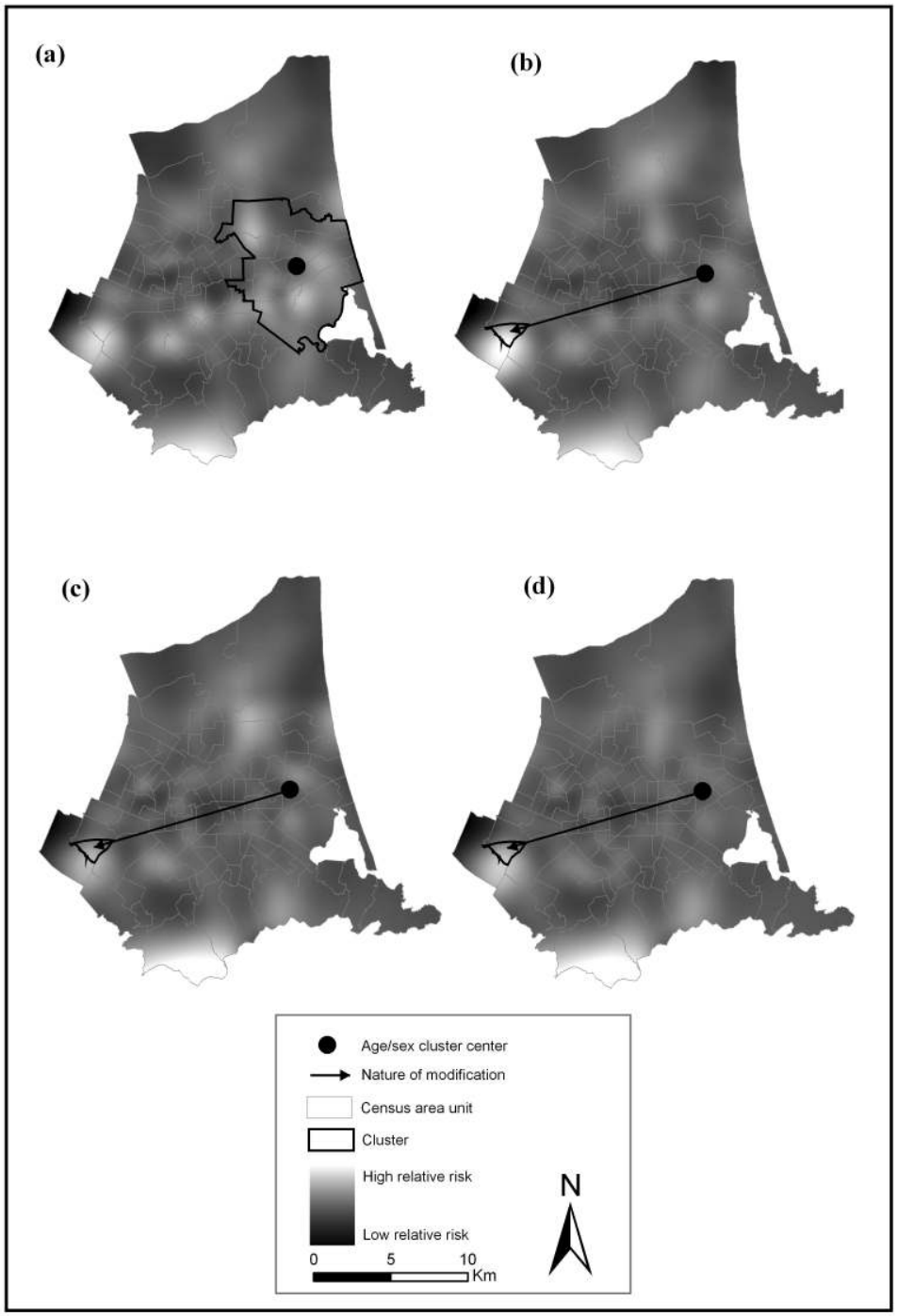


Figure 5.3. Spatial patterns of relative risk and spatial relocation of asthma clusters after adjusting for (a) age and sex, (b) age, sex and air pollution (c) age, sex and deprivation and (d) age, sex, air pollution and deprivation. Arrows indicate the distance and direction from the centre of the age/sex cluster to covariate-adjusted clusters with the endpoint of the vector marking the new centre.

As the spatial shifts in relative risk are apparent yet difficult to interpret, CAUs containing the most likely cluster as determined by the spatial scan statistic and maximum likelihood ratio have also been displayed (Figures 5.2 and 5.3). Table 5.1 details the nature of these shifts as well as the number of CAUs contained in each cluster, centre coordinates, radii, the distance and azimuth (vector) of spatial relocation and the relative risk for each cluster. In the case of every disease code tested (except for the control), there was a significant shift in the position of the clusters and number of CAUs in each cluster when adjusting for deprivation, air pollution, or both. Spatial shifts were from as little as 1.3 km (for age/sex and deprivation adjusted asthma) up to 13.4 km (for any combination of deprivation or air pollution adjustment). Relative risks of clusters quantitatively shifted away from the null (Table 5.1) for all disease codes tested when controlling for factors beyond age/sex. It should be noted that while some relative risks were similar when controlling for different covariates, the positions of clusters could vary greatly. For example, in Figure 5.2c the relative risk of the pneumonia and influenza cluster adjusted for age/sex and deprivation (test c) is 1.92 whereas in Figure 5.2d, the relative risk for the age/sex, air pollution and deprivation (test d) adjusted cluster is 1.99. However, the two clusters are located at significantly different distances from the original cluster (1.3 km vs. 10.6 km) and contain different quantities of CAUs within their cluster boundaries. This example clearly indicates a spatial shift in risk when factors are adjusted for as covariates in this study.

Table 5.1. Summary statistics of cluster spatial relocation by disease groupings after adjusting for age/sex, particulate matter air pollution (PM) and/or deprivation covariates.

Disease and ICD Code(s)	Test[+]	Age/ sex	PM	Dep	N[#]	Centre Coord.* (Lat, Lon)	Radius[1] (km)	Spatial shift[2] (º; m)	Obs[3]	Exp[4]	RR[5]	*p*
Acute Respiratory Illness ICD 460-466	a	x			18	-43.533, 172.682	3.03	-	1456	1159.6	1.26	0.001
	b	x	x		5	-43.540, 172.530	2.30	226; 12,300	476	323.4	1.47	0.001
	c	x		x	3	-43.540, 172.530	1.35	226; 12,300	335	237.2	1.41	0.001
	d	x	x	x	1	-43.549, 172.530	0.00	262; 12,433	172	99.2	1.74	0.001
Pneumonia and Influenza ICD 480-487	a	x			28	-43.530, 172.672	3.76	-	2469	2020.5	1.22	0.001
	b	x	x		5	-43.540, 172.530	2.30	264; 11,553	565	344.1	1.64	0.001
	c	x		x	1	-43.538, 172.661	0.00	225; 1,269	136	71.0	1.92	0.001
	d	x	x	x	3	-43.554, 172.553	2.04	244; 10,646	380	191.4	1.99	0.001
Asthma ICD 493	a	x			27	-43.520, 172.690	4.26	-	1954	150 8	1.30	0.001
	b	x	x		1	-43.549, 172.530	0.00	256; 13,355	168	60.2	2.79	0.001
	c	x		x	1	-43.549, 172.530	0.00	256; 13,355	168	84.6	1.99	0.001
	d	x	x	x	1	-43.549, 172.530	0.00	256; 13,355	168	74.2	2.27	0.001
All Respiratory Admissions ICD 460-519	a	x			28	-43.530, 172.672	3.76	-	10825	8627.1	1.26	0.001
	b	x	x		5	-43.540, 172.530	2.30	264; 11,533	2509	1467.6	1.71	0.001
	c	x		x	2	-43.540, 172.530	1.00	264; 11,533	1335	806.9	1.65	0.001
	d	x	x	x	5	-43.540, 172.530	2.30	264; 11,533	2509	1613.4	1.56	0.001
Appendicitis (control) ICD 540-542	a	x			3	-43.550, 172.508	2.09	-	102	61.5	1.66	0.002
	b	x	x		3	-43.550, 172.508	2.09	360; 0	102	59.3	1.72	0.001
	c	x		x	3	-43.550, 172.508	2.09	360; 0	102	62.4	1.64	0.002
	d	x	x	x	3	-43.550, 172.508	2.09	360; 0	102	57.3	1.78	0.001

[+]factors which were adjusted for in the analysis: (a) age/sex only; (b) age/sex and air pollution; (c) age/sex and deprivation; (d) age/sex, air pollution and deprivation.
[#]number of census area units in the most likely cluster.
*centre coordinates of most likely cluster.
[1]when only one census area unit is found in the cluster, a value of 0.00 km is the returned radius of the cluster.
[2]shift in centre of cluster in azimuth north degrees and metres from baseline age/sex adjusted cluster.
[3]observed number of cases in cluster.
[4]expected number of cases in cluster.
[5]relative risk of the cluster.

A further example of the modifications in spatial risk is illustrated by the Gi* statistical results for pneumonia and influenza (Figure 5.4) and asthma (Figure 5.5). For comparative and reference purposes, the spatial scan statistic's most likely cluster for each test were included in the figures as well. The hot/cold spot analysis also indicated that there were significant spatial modifications in risk when adjusting for air pollution and deprivation. Areas of elevated risk (dark areas) were spatially co-located with the clusters determined by the spatial scan statistic for all disease groupings studied and accordingly shifted in the subsequent tests (b, c, and d). In the baseline examples for both pneumonia and influenza (Figure 5.4a) and asthma (Figure 5.5a), there are hot spots (darker shades) in the eastern, more deprived areas of the city and cold spots (lighter shades) in the less deprived areas to the west/northwest of the city centre. These cold spots appear when pollution alone is controlled for (Figures 5.4b and 5.5b), then disappear when deprivation is controlled for (Figures 5.4c and 5.5c).

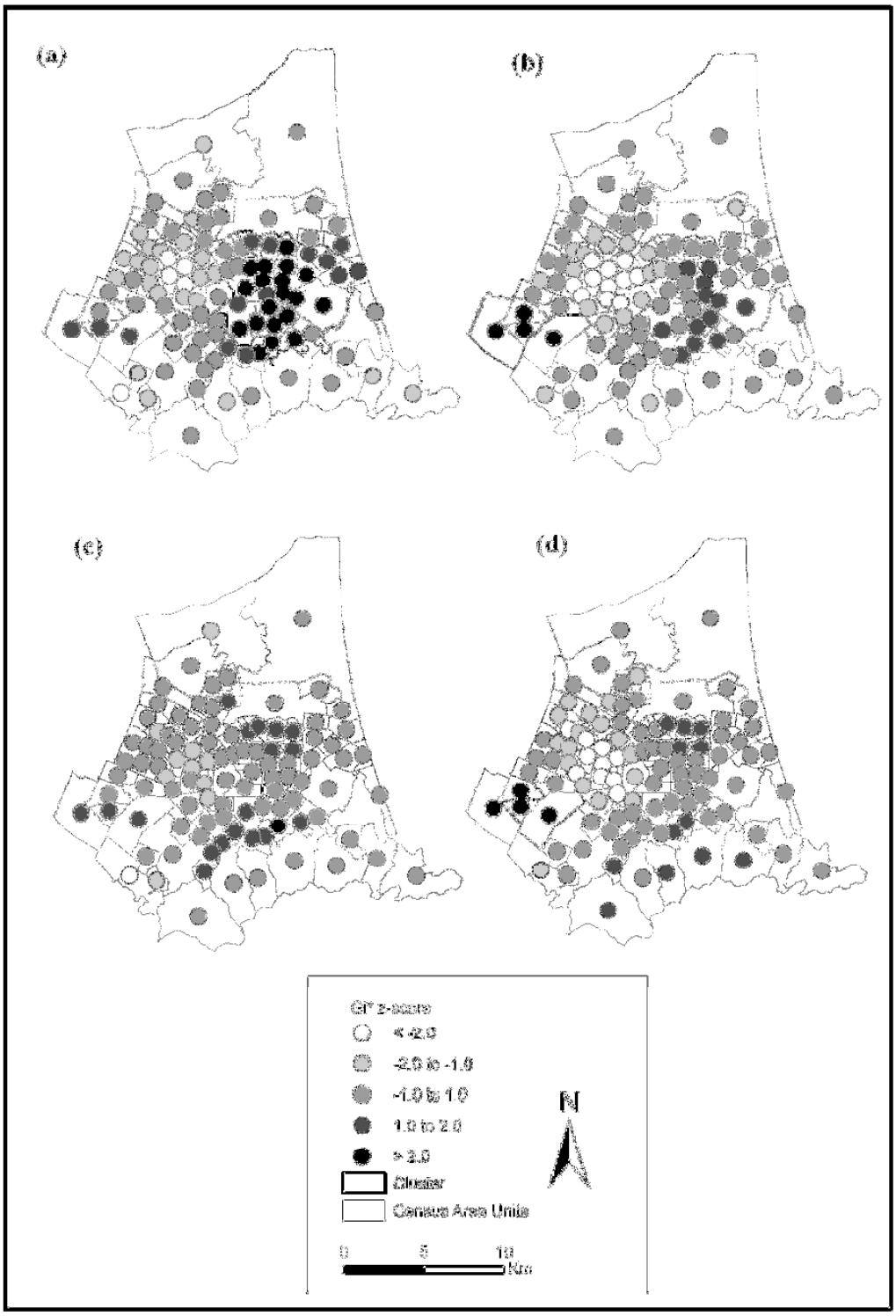


Figure 5.4. HotSpot G-statistic z-scores and SaTScan clusters of pneumonia and influenza hospital admissions after adjusting for (a) age and sex; (b) age, sex and air pollution; (c) age, sex and deprivation; and (d) age, sex, air pollution and deprivation. High z-scores (darker shades) indicate clustering of elevated relative risk ('hot spots') and low z-scores indicate reduced relative risk areas ('cold spots').

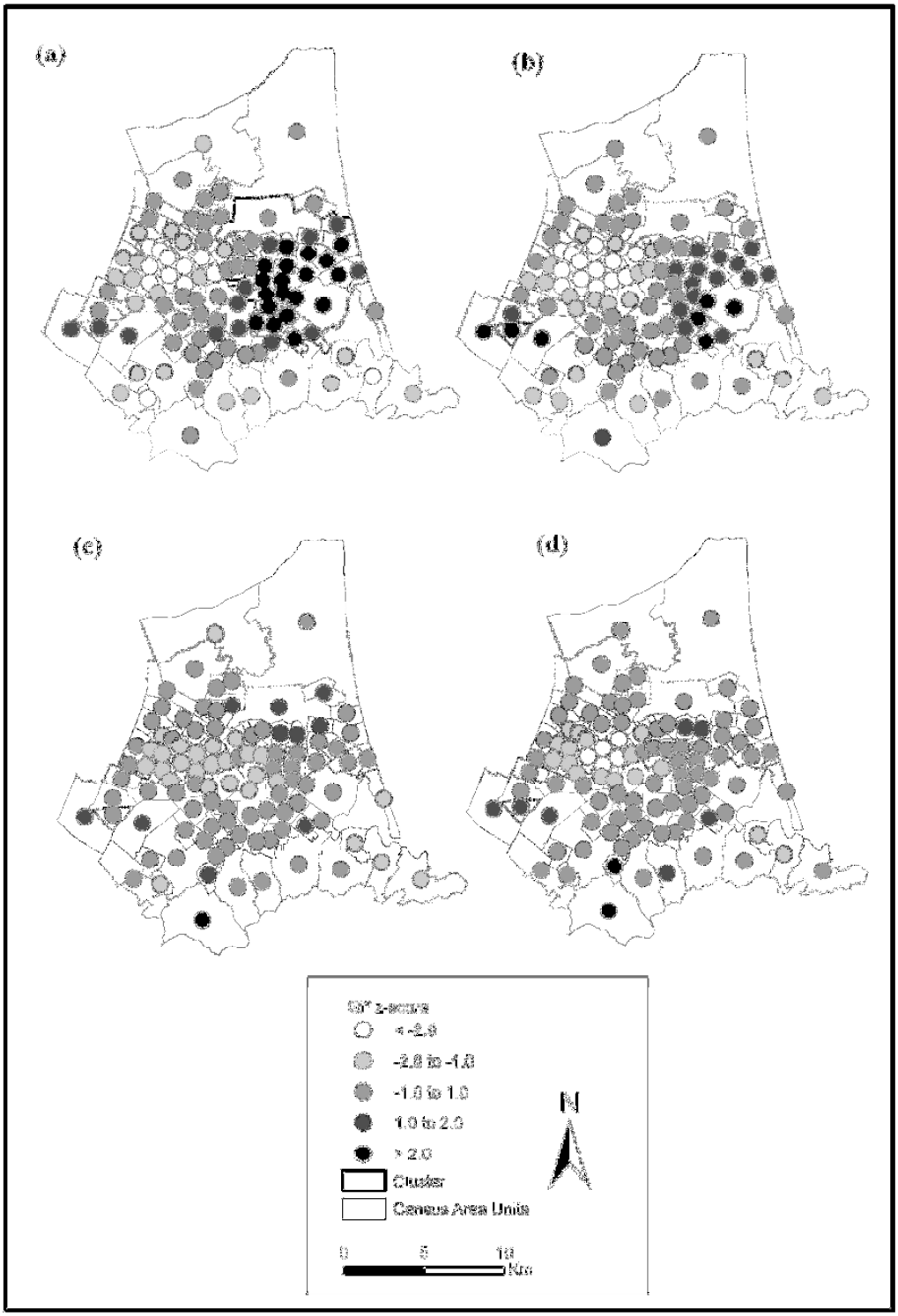


Figure 5.5. HotSpot G-statistic z-scores and SaTScan clusters of asthma hospital admissions after adjusting for (a) age and sex; (b) age, sex and air pollution; (c) age, sex and deprivation; and (d) age, sex, air pollution and deprivation. High z-scores (darker shades) indicate clustering of elevated relative risk ('hot spots') and low z-scores indicate reduced relative risk areas ('cold spots').

These shifts appear to indicate that some of the indirect effects of deprivation on patterns of risk have adequately been removed. However, when deprivation is re-introduced along with air pollution (Figures 4d and 5d), the cold spots remain. The results show that there is a spatial risk modification component to the re-emergence of the cold-spots as well.

As noted in Table 5.1 and Figures 5.2 – 5.5, the shifts when controlling for either deprivation or pollution, or both, were remarkably similar, except for a slight alteration in risk for pneumonia and influenza. In all other disease classes tested, vector adjustment of the centre of air pollution or deprivation clusters were identical and the number of CAUs included in adjusted clusters differed by no more than four. When adjusting for chronic air pollution exposure and deprivation, the shifts were also largely similar. In Christchurch, where air pollution and deprivation have been linked (see Pearce *et al.* in press), these results appear to suggest that adjusting for deprivation and/or pollution may produce similar results.

5.6. Discussion and conclusion

In this paper we find that controlling for social deprivation and chronic PM_{10} air pollution exposure notably alters the spatial patterns of risk and the positions of clusters and hot/cold spots of hospital admissions for respiratory disease (Figures 2 – 5). This work represents the first presentation of the spatial relocation effect due to adjusting for covariates at the intraurban scale that we are aware of in the literature, and may have future applications for epidemiological studies, especially in the area of disease surveillance.

We found that in every disease category, for every combination of disease determinant variables tested, controlling for either or both particulate matter air pollution and deprivation shifted the pattern of risk from the baseline case and shifted the centre and nature of clusters significantly. An additional, although reassuring, outcome of our results indicate that the area and location of the spatial scan statistic closely approximates the Getis-Ord statistic. These results indicate that the two methods produce similar results of elevated clusters of relative risk and lends confidence to the use of both methods in surveillance applications. A more detailed analysis at how results of the two methods compare should be explored.

This work may have several implications for future studies examining air pollution, deprivation and hospital admissions at the intraurban scale. In a spatial epidemiological context, a description of the trends we have observed is instrumental in identifying potential clues as to the cause of the observed spatial structure, which might lead to insight regarding the aetiology of the diseases under investigation. However, if the process of adjusting materially alters the spatial patterns observed at a local scale, then the implications of our findings for other studies of this type could be profound. First, take as an example the case where deprivation–related modifiers of risk for respiratory illness are being investigated at the intraurban scale. When adjusted for age and sex, the elevated risk area may be in a residential area of the city dominated by domestic heating point sources. However, upon adjustment for deprivation, the areas of high risk could shift to a new area of the city, say, a populated area near an industrial park, as was the case in our Christchurch asthma example (Figure 5.6). The new, spatially modified location of the cluster may now introduce new pollutants (e.g., SO_2 or hazardous air pollutants) that were not relevant in the original location, and thus, did not require adjustment. The inverse could also be true, where an area pre-adjustment has high concentrations of a possible pollutant or risk factor which are removed from the exposure equation post-adjustment due to spatial relocation. The spatial relocation of risk may, hence, be an important factor in environmental health studies, and should be well-understood and integrated into the study design process. It may also be useful to map the spatial modification of risk when evaluating the modification effect of various factors on risk outcomes.

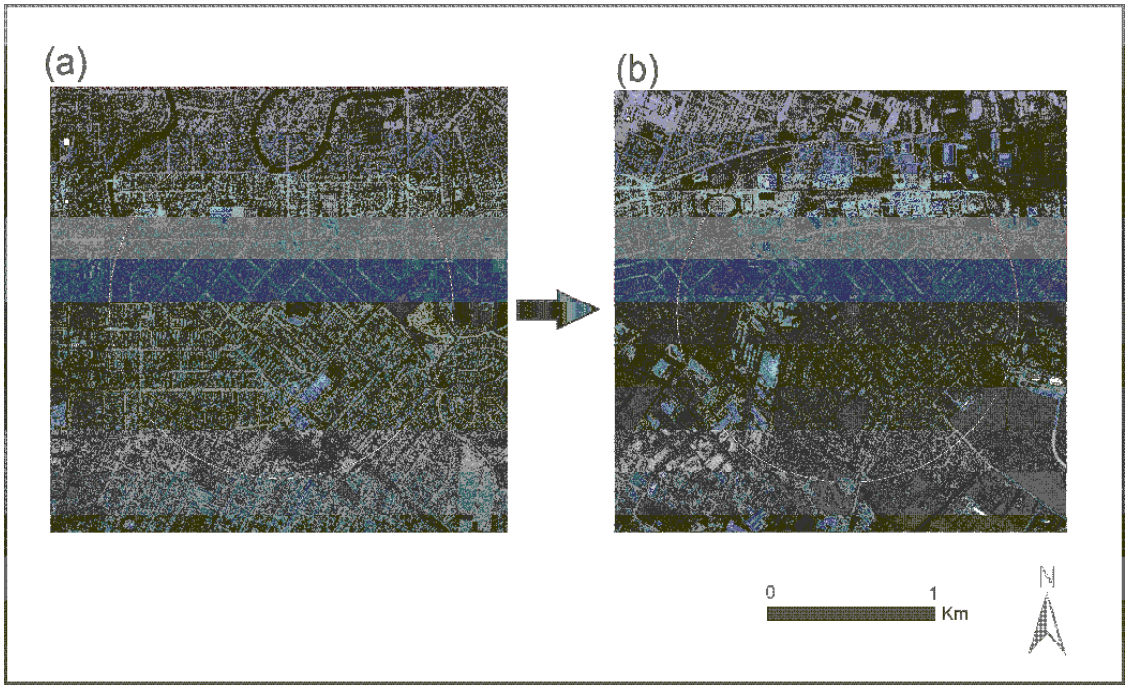


Figure 5.6. Localised landuse characteristics and one kilometre radius around (a) the original age/sex-adjusted asthma cluster centre and (b) the cluster centre adjusted for age/sex, air pollution and/or deprivation. On left, the original location of the cluster is largely residential, while the area of the shifted cluster centre is mixed industrial/residential landuse. The shift in the location of the cluster may introduce additional exposures from industrial sources which were not present in the original location.

It should be noted that this study presents several limitations. First, the nature of our risk pattern analysis is largely descriptive. Although maps of disease risk are an accepted method in the identification of possible areas of risk, especially for diseases such as cancer, risk estimate maps are often open to misinterpretation. We mitigated the descriptive nature of the visual analyses by quantitatively analysing cluster results using a Gi* and spatial scan statistic. Although the maps of spatial shifts in risk may seem trivial at first glance, they can have important practical applications, especially for use in surveillance (Järup 2004). Second, the underlying associations between socioeconomic, environmental, and other factors which we set out to study are inherently complex. O'Neill et al. (2003) have addressed this fact in reference to environmental justice work, and we acknowledge that the spatial relocation effect which we observed may only be due in part to the adjustment for the covariates we adjusted for – their may be other factors at play influencing the relationship of which we are not aware. Third, there are several inherent problems with fine-scale studies. A well understood problem in spatial analysis is the modifiable areal unit problem (MAUP), as defined by Openshaw (1984). The MAUP generally concerns two issues: scale and zone aggregation. Bias in this study may be associated with either issue, but the scale issue has more obvious potential to introduce error into the analyses, i.e., that the effect of socio-economic or air pollution determinants against health may be markedly different depending on the scale of analysis (Jerrett & Finkelstein 2005). Our decision to aggregate at the CAU level was based on findings that there is significant variability in Christchurch for both of the variables tested (Figure 5.1), as has been verified in other intraurban studies locally (Wilson *et al.* 2006, Pearce *et al.* in press). It may be informative to estimate the MAUP-scale effect by testing the spatial relocation at different scales of ecological aggregation using multilevel analysis in tandem with the methods presented here. Our modelled exposure values may also be subject to some misclassification due to the fact that our dispersion model was set at 1,500 m and rasterised to 100 m, even though the exposures were recoded to quartiles before they were entered into SaTScan. Wilson et al. (2006) found PM_{10} concentrations to be heterogeneous at the intraurban level in Christchurch, but none of the monitoring sites used in this assessment were within 1,500 m of each other so the intra-neighbourhood variability of concentrations was not determined. Finally, we used a Poisson estimate of risk, which may not be a valid approach for the estimation of risk when the sample of cases is large, as recent research has shown (Salway & Wakefield 2005). In this research, the focus is on non-rare events (hospital admission), where the

Poisson approximation to the binomial distribution may not be appropriate, especially when characterising the direction of bias. However, as we are more concerned with the spatial direction and distance (vector) of bias or relocation, and not the effect on the dose-response or exposure-health outcome, the point may not be critical in this context.

In this Chapter, patterns of risk and clustering were spatially analysed at a fine scale across an urban area when adjusting for different environmental and deprivation-related determinants of disease. The results indicate that both chronic exposure to particulate matter air pollution and deprivation significantly altered the patterns of risk as well as the position and size of clusters of disease. Further, it was found that the spatial shift in risk was similar, whether controlling for pollution, deprivation or both. It is critical that researchers adjusting for covariates understand the spatial relocation nature of relationships when considering study design and when discussing results of surveillance analyses.

Chapter 6: Outcomes and contributions.

6.1. Summary of the research

This thesis has addressed a base assumption of air pollution epidemiology: that intraurban particulate matter air pollution concentrations are homogeneous within urban areas. Beyond reviewing the literature of monitored intraurban studies, this research has tested the assumption in an area with high air pollution, and has found the assumption to be false. An innovative model for estimating intraurban particulate concentrations has been evaluated, and may be applicable in an emerging study design: the intraurban cohort study. This research has also demonstrated how spatial modification of disease clusters may be used in furthering the understanding of the inter-relationships between social confounders, air pollution, and respiratory morbidity. Collectively, these findings contribute to the wider body of knowledge of exposure assessment of particulate matter and its implications and applications.

6.2. Summary of outcomes and contributions

This thesis has produced several tangible outcomes in the context of the wider exposure assessment and spatial epidemiological literature:

6.2.1. Outcome one – the first systematic review of the intraurban particulate literature, challenging the widely-held assumption that PM concentrations are spatially uniform and clarifying the implications of this assumption for specific epidemiological study designs.

In Chapter two, as well as in the work published by Wilson *et al.* (2005), particulate matter air pollution exposure studies were systematically reviewed for the first time with a focus on monitored-data studies at the intraurban spatial scale. Portions of the literature were found to provide contradictory conclusions as to the homogeneity of intraurban particulate concentrations. In part, this is due to local conditions such as source composition, meteorology, locations of monitoring sites and topography, but it may also be a result of the methods and definitions used

to quantify relative and absolute spatial concentration variations. It was found that special attention should be given to local conditions and methods when using one or few monitoring sites to characterise wider population exposures. Further, it was found that PM 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') and that, 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. The outcome of Chapter two also suggests that 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 (Wilson *et al.* 2005). 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.

6.2.2. Outcome two – an experimental test showed that the homogenous assumption was false in a city with high wintertime particulate matter concentrations.

Chapter three demonstrated that, in the city of Christchurch, New Zealand, daily concentrations of PM_{10} were found to be substantially spatially variable (Wilson *et al.* 2006). High correlation between sites ($r > 0.76$) was not indicative of relative homogeneity, highlighting the importance of appropriate statistical methods (such as a coefficient of divergence) when describing intraurban uniformity. Variability was substantial, not only between the central monitoring site and the background population sites, but also between all intraurban sites in the study. Spatial variability in particulate air pollution in Christchurch was of a level significant enough to warrant caution when using central monitoring sites to estimate wider-population exposures, especially in long-term cohort epidemiological studies. Health and PM studies in cities with similar variability in intraurban PM concentrations should use caution when drawing conclusions about population exposures from a central monitoring site.

6.2.3. *Outcome three – an atmospheric dispersion model was evaluated for the first time at the intraurban level for PM and a new study design was suggested.*

Despite Christchurch's high intraurban concentration variability (Wilson *et al.* 2006), and meteorological and topographical complexity, the integrated meteorological emission (IME) model tested in Chapter four performed satisfactorily (Wilson & Zawar-Reza 2006). Mean observed and modelled concentrations were 42.9 μg m^{-3} and 43.4 μg m^{-3} respectively, while the mean Index of Agreement (IOA) between individual sites was 0.60 and the mean systematic RMSE was 16.9 μg m^{-3}. Most of the systematic error in the model was due to coarse spatial resolution of the local emission inventory and complex meteorology attributed to localised convergence of drainage flows, especially on the western and southern fringes of the urban area. Given further improvements in site-specific estimates within urban areas, IME models such as The Air Pollution Model (TAPM) may be a viable alternative to central monitoring sites for estimating personal exposure in longer-term (monthly or annual) cohort epidemiological studies.

6.2.4. *Outcome four – the spatial modification effect of ecological confounders was analysed with respect to respiratory hospital admissions and PM.*

In Chapter five, the spatial modification of relative risk and clusters of disease by means of spatial analysis techniques was applied to an illustrative small-area example from Christchurch, New Zealand (Wilson *et al.* under review). In all disease groups tested (except the control disease), adjusting for deprivation and chronic PM_{10} exposure was found to modify the position of clusters substantially, as well as notably shifting patterns and hot/cold spots of relative risk across the city. Further, it was found that the spatial shift in risk was similar, whether controlling for deprivation, pollution or both. This research suggests that it is critical to understand the spatial nature of effect modification when considering study design and when discussing results of ecological analyses. Further, this work may be an early step in the application of a 'spatial modification effect' in the study of determinants of exposure and downstream health effects.

6.3. Collective contribution of the thesis and chapter linkages

Collectively, this thesis is more than an anthology of separate chapters and outcomes (as might be presumed from reading Section 6.2) – the work represents a contribution to the literature in a more synergistic way as well. There are several ways which a collective contribution can be addressed, including cross-ways interactions between the chapters, and the relationship between the chapters and the existing literature and theory.

First, there is a baseline linear progression of the chapters, from hypothesis, to a testing of the hypothesis, to modeling, and finally to an example using health outcomes (as seen in Figure 1.1). However, the interactions between the chapters do not actually occur in a linear fashion. Though each of the chapters builds on one another in a retrospective fashion, there is also collective material evidence that amasses as the thesis progresses (Figure 6.1). For example, the hypothesis laid out in Chapter 2, that particulate concentrations may not be homogeneous under some circumstances, becomes a key tenet in all future chapters. Chapter 3 demonstrates the methodological problems associated with the use of correlation coefficients in determining heterogeneity, while applying lessons learned in Chapter 2. Accordingly, Chapter 4 validates a model against the concept of intraurban variability in Christchurch (thus linking to the monitored data in Chapter 3), and suggests a novel study design (the intraurban cohort study based on IME models), taking into account the lessons learned about exposure misclassification error in Chapter 2 (see Table 2.1). Finally, Chapter 5 integrates all of the previous aspects: from methodology (Chapter 2), to ground testing (Chapter 3), to modelling (Chapter 4), to the case of examining risk patterns and the spatial relocation effect of adjusting for both chronic intraurban variability in exposures as well as an index of deprivation. The interactions, therefore, between the chapters and concepts in the thesis, are more inter-related and cumulative rather than stand-alone and linear, as one might surmise from Figure 1.1 at first glance.

Next, the issue of how this work relates to the wider literature should briefly be mentioned. Health geography may be defined as the broader study of health in a spatial and contextual sense, beyond the concerns of disease, risk factors, aetiology and the more traditional concerns of the medical world (Kearns and Moon 2002). While exposure assessment has traditionally been a part of both environmental science and environmental epidemiology, it also has a home within health

geography, as many the primary interaction of concern of exposure assessment is the time-space-dose component between an individual and some environmental contaminant (Monn 2001). As health geographers are largely concerned with the question 'does where you live matter to your health?' (Boyle *et al.* 2004), a certain component of what matters to one's health are the environmental exposures at the intraurban level – or neighbourhood – one lives within. Chapters 2, 3 and 4 largely deal with matters of exposure assessment and measurement error as well as their effects on risk outcomes. In Chapter 5, the focus shifts towards a spatial epidemiological approach. While the 'new' geography of health is likely an evolved version of spatial epidemiology and medical geography (Kearns and Moon 2002), a link remains. In Chapter 5, this thesis builds on the previous four chapters to understand what might be classically defined as spatial epidemiology from a methodological point-of-view: spatial relocations of disease clusters and shifting patterns of disease risk. Finally, though tangential to the overall theme of the thesis, this work does present opportunities for expansion into the field of environmental justice. As stated in the Chapter 1, of primary importance to understanding exposure variability across socioeconomic position gradients is the assessment of true environmental exposures in small areas (O'Neill *et al.* 2003). In Chapter 5, small-areas (census units) are the basis of aggregation – the importance of geographical scale is a key debate in the environmental justice and related literature, as aggregation levels (e.g., national, regional, local) may have implications for the relationship between measures of deprivation (e.g., income inequality) and health (Subramanian and Kawachi 2004). This thesis contributes to the understanding of measurement and modelling of intraurban particulate concentrations at the finer scales, which will support future work in air pollution and environmental justice, helping to clarify the true nature of inequality and health. The chapter interactions with each other as well as with the literature are displayed in Figure 6.1.

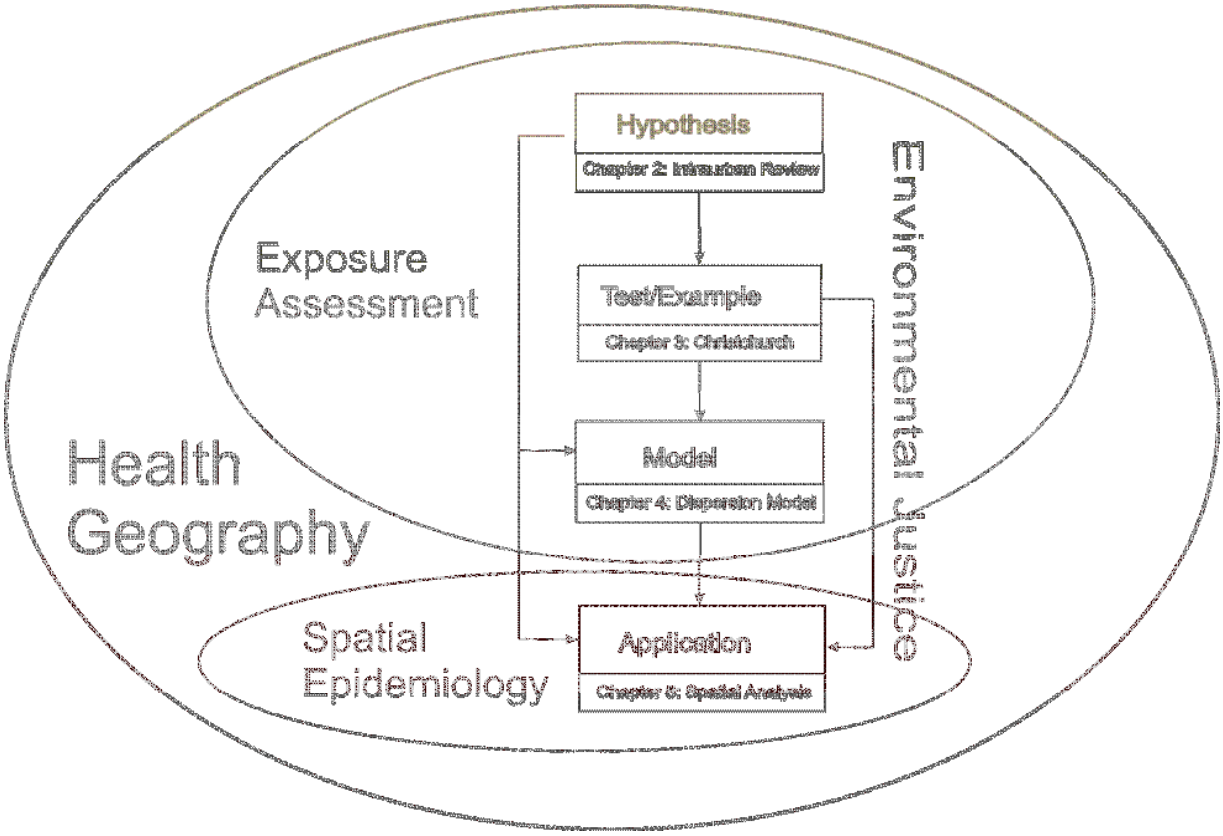


Figure 6.1. Conceptual linkages between chapters and linkages to the theoretical framework

6.4. Limitations

Beyond the limitations outlined in the discussion sections of Chapters 2, 3 and 4, several additional limitations of this work should be noted, along with collective limitations of the thesis. First, the intent of chapter one, and this thesis in general, was to more thoroughly understand the *methods* for determining spatial heterogeneity or particulate air pollution. This work did not set out to understand the *underlying causes*. Some of the causes underlying variability include source composition and density (Goswami *et al.* 2002), land use (Jerrett *et al.* 2005a), population density (Suh *et al.* 1997), localised metrological conditions (Kossman and Sturman 2004), and seasonal trends (DeGaetano and Doherty 2004). In short, predicting variability is a complex and chaotic undertaking and the factors influencing particulate concentration across a city tend to vary substantially from one urban area to the next (Wilson *et al.* 2005). Although the underlying causes may be important, what this thesis stresses is that the appropriate methods be used to describe and measure variability, as it is critical that these measurements and methods be consistent if a true definition of uniformity between cities is to emerge.

Next, the issue of recommendations for measuring uniformity should be addressed. In Section 2.7, the following statement is made:

> "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 (Monn *et al.* 1997, Kim *et al.* 2005). It may also be useful to shorten sample averaging times to $\leq$ 24-hrs as longer averaging times may not capture diurnal patterns that are important in characterising intraurban uniformity."

It should be noted that these recommendations for sample size, number of sites and averaging times are to be taken purely as *examples* of what might constitute a proper combination for determining variability – not hard and fast rules to be applied in every situation. The characterisation of variability is highly dependent on study design, the size of the study area, sampling times, and health outcomes for which the exposures are to be applied against (Wilson *et al.* 2005). One recommendation would be to conduct a short pilot study to ascertain variability before conducting any long-term measurement campaign. The pilot study should contain the

maximum amount of stations over the smallest sampling times, and should be during the season when variability is expected to be greatest.

Finally, a mention of the performance of the TAPM integrated meteorological-emission model should be included. Since the publication of Chapter 4 (Wilson and Zawa-Reza 2006), there have been several observations contributing to our understanding of factors influencing the performance of the model. As a review, the model performed well over long periods (also see Zawar-Reza *et al.* 2006), but daily performance was quite poor (mean RMSE = 34.38 μg m^{-3}). Ironically, the model performed better on days when winds are strong and air pollution levels are low (models are designed to simulate *high* air pollution levels). This is the result of the need to simulate turbulence properly during inversion conditions in Christchurch, a region that also experiences thermally driven flows like the drainage winds at night (Kossman and Sturman 2004). This issue is still unresolved, though it was by-passed in the model design in Chapter 4 by means of data-assimilation. More recent work also indicates that the model performed best during deep anti-cyclonic situations with stagnant winds at 3 km altitude. For any other anti-cyclonic periods the performance was reduced. A manuscript outlining these concerns is in preparation (*pers. comm*. Zawar-Reza, May 2006) and discussions of these findings are currently underway with the group responsible for developing TAPM (CSIRO) – it is anticipated that the problem will be corrected in future versions of the modeling software.

6.5 Further work

This research highlights several opportunities for future work. First, there is an opportunity to further quantify the error induced by the use of central monitoring sites when intraurban variability of PM is substantial. This thesis has established that the use of central monitoring site exposure estimates may induce error, but that variability of that error has yet to be systematically quantified. A useful study would involve evaluation of the effects of variability on health risk estimates based on a meta-analysis approach: that is, one utilising data from several urban areas that have a range of variability. It would also be interesting to address the actual effect on exposure assessment by modelling the exposure distribution across the study area on a daily and/or longer-term basis, while comparing several different estimate methods, namely: (i) the central monitoring site; (ii) the average of the background monitoring sites; (iii) the nearest

monitoring site (e.g., using Thiessen polygons in a GIS); and (iv) an interpolated surface (e.g., Kriged or IDW using a GIS). This comparison might yield a more exact and applicable estimate of the actual magnitude and direction of exposure error due to central site and other methods. Next, at the time of publication of this thesis, there have been no studies to date utilising an air pollution dispersion model to generate exposure estimates in an intraurban cohort study (Jerrett *et al.* 2005a). The results from a recent intraurban cohort study which adopted a GIS approach for estimating respiratory health effects associated with $PM_{2.5}$ exposures by postal code were promising, and give merit to the idea of using APD models as suggested in Chapter four (Jerrett *et al.* 2005b). Lastly, this thesis may provide a foundation for promising applications in the areas of spatial epidemiology and environmental justice; a key component to environmental justice work is ascertaining accurate intraurban exposure estimates across gradients of socioeconomic measures. Early work in this field has been conducted in Christchurch (Pearce *et al.* 2006) and the United Kingdom (Briggs & Fecht 2005), but little work has been performed to date in developing countries. It is the aim of the candidate to develop the work performed in this thesis towards an applied focus on environmental justice concerns in the developing world.

6.6. Final thoughts

In the final analysis, urban air pollution, although menacing, is not a predicament without solution. By its nature, air pollution is not necessary or by any means permanent – lower emissions immediately translate to cleaner air and greater well-being amongst individuals (Clancy *et al.* 2002). With greater knowledge of the nature of urban air pollution, we will gain understanding. This thesis is but a stone in the path of that knowledge and understanding. Walking this path will bring forward truth and, hopefully, the will to clean the urban air we breathe. Then can come the healing.

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