# Ultrafine Particle Exposure Inside Automobiles

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# Table of Contents

# Abstract

There is an accumulating body of work suggesting that a large proportion of many people's daily exposure to ultrafine (<100nm) particles (UFPs) is incurred during the relatively short period of time spent travelling in an automobile (vehicle). Based on toxicological and epidemiological studies, UFPs are increasingly viewed as a significant risk to public health. However, despite the substantial contribution to total exposure that vehicle travel may constitute, the factors controlling the magnitude of in-vehicle UFP exposures have not been well-documented to-date. The effects of travel through road tunnels (a location where pollutant emissions are often elevated due to their constrained geometry) on daily UFP exposures have not been investigated, despite the increasing number and length of tunnels worldwide. This thesis takes a synergistic and measurement-oriented approach to elucidate the key factors underpinning in-vehicle UFP exposure during tunnel travel that is also relevant to all roadway environments.

To facilitate accurate measurements of the often extreme UFP concentrations encountered on roadways, a TSI 3007 condensation particle counter (CPC) was fitted with a purpose-fabricated dilution system. This system was deployed in five common passenger vehicles encompassing an age range of 18 years, and was used to alternately measure in-cabin and on-road UFP concentrations in the twin-bore 4km M5 East road tunnel in Sydney, Australia. Three hundred and one successful tunnel trips were performed under a wide range of traffic conditions, and the data collected revealed a strong association between heavy diesel vehicle (HDV) volume and median on-road UFP concentrations in the eastbound bore of the tunnel. By comparison, the strength of this association was quite poor in the westbound bore of the tunnel; although in both cases, HDV traffic volume was a significantly better determinant of median on-road UFP concentrations than passenger (i.e. gasoline-powered) vehicle volume.

To investigate the relationship between on-road and in-cabin concentrations, the data collected during tunnel travel under four distinct

ventilation settings were combined with tracer gas measurements that quantified the rate at which the cabins were ventilated with outdoor (i.e. on-road) air. These measurements were obtained following an extensive and systematic field-based data collection campaign. The results indicated a very strong positive linear relationship between outdoor air flow rate and ingress of on-road UFPs into the cabins of all test vehicles under all ventilation settings. This highlighted the role of a vehicle's air-tightness in protecting its occupants from UFP exposure, especially under an air recirculation setting, as the vehicles found to be the least air-tight under such a setting allowed the greatest proportion of on-road UFPs into the cabin, and vice-versa. Newer vehicles fitted with standard pollen/dust filters had the lowest median in-cabin/on-road UFP ratios.

Based on all data collected, a simple mathematical model was employed to predict the average in-cabin UFP exposure incurred during a trip through the M5 East tunnel. The predicted exposures agreed reasonably well with those measured, and the results highlighted both the substantial utility such models could provide and the need for future studies to examine this topic in greater depth. Simulations underpinned by field data combined with the results of other studies estimated that between 1.1 and 22% of an M5 East tunnel user's daily UFP exposure could occur as result of <0.25h of daily tunnel travel, with the above range indicating the best (newer, filter-fitted vehicle) and worst-case (older vehicle, no filter) scenarios, respectively.

This thesis contributes substantial new data and analytical insights regarding the nascent yet significant issue of in-vehicle UFP exposure, in addition to providing suggestions for future work. It is hoped that the results presented here will allow roadway and tunnel managers and public health authorities, regardless of their location, to more effectively manage and mitigate vehicle occupant UFP exposure.

# Declaration

None of the work described in this thesis has been submitted in support of an application for another degree or diploma at Macquarie University or any other institution. The work presented is my own, except where due reference has been given.

Luke D. Knibbs

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"Ah, there's nothing more exciting than science. You get all the fun of sitting still, being quiet, writing down numbers, paying attention...Science has it all."

Principal Seymour Skinner, *The Simpsons.*

# Section I
# Introduction

# Chapter 1

# Introduction and Background

The study of air quality inside automobile (vehicle) cabins has existed for over 40 years. In addition to his pioneering and enduring work related to characterising photochemical smog and ozone, Professor Arie Haagen-Smit was among the first researchers to recognise the significance of vehicle occupant exposure to pollutants whilst driving (Flachsbart, 2007). In 1966 he reported a suite of carbon monoxide (CO) measurements performed on routes in Los Angeles during peak traffic periods (Haagen-Smit, 1966). Later that year, a comparable, albeit more comprehensive, study was published (Brice and Roesler, 1966) based on in-cabin CO and hydrocarbon measurements performed in several US cities. Both of these early forays into the characterisation of air pollutants affecting drivers and passengers yielded results that would be corroborated by scores of related works over the following decades; specifically, that the concentrations of pollutants measured inside vehicles or on roadways were greater than ambient concentrations. One of the first systematic investigations of the magnitude of this effect was performed in terms of Boston residents' CO exposure by Cortese and Spengler (1976), who reported that commuters using automobiles experienced higher exposures than those who used other transport modes. Cortese and Spengler (1976) also reported the concentrations of CO that commuters were exposed to were approximately double those measured by fixed-site monitors (Flachsbart, 2007). Other early in-vehicle pollutant studies include those of Petersen and Sabersky (1975), Mayron and Winterhalter (1976) and Chaney (1978).

The fact that researchers of the calibre of Haagen-Smit and others were attracted to this area of study was a testament to its importance, by virtue of the worldwide number of automobiles and commuters occupying them. This is of equal if not greater relevance over 40 years later, and despite in-vehicle air quality research now being a mature science, many questions still remain to be answered in order to fully achieve the goal that early investigators aimed for; namely, protection of vehicle occupants from harmful pollutants.

This thesis does not attempt the monolithic task of developing an integrative solution to the issue of in-vehicle air pollution in terms of all air pollutants and vehicles. Rather, it aims to deliver a timely and tightly focussed investigation of specific criteria, many of which are relevant to other facets of in-vehicle air quality. The specifics of these criteria and project aims are discussed in detail after the following overview of topics most relevant to the present study.

## 1.1 Aspects of In-Vehicle Environmental Quality

The recognition of the importance of the environment inside vehicles and the need to control it for the improvement of human comfort has existed for many years. Primitive heating and cooling systems for vehicles included hot bricks, fuel burners and blocks of ice, which soon gave way to heat exchangers and evaporative coolers, which in turn were superseded by the antecedent of the modern mechanical air-conditioning system, first introduced in 1939 (Daly, 2006). Since that time, the penetration of air-conditioning into automobile markets worldwide has increased, and now approaches 100% in many regions (Daly, 2006).

There are myriad factors affecting the overall quality of the in-vehicle environment (Gameiro da Silva, 2002; Gameiro da Silva et al., 2006); including ventilation rates, thermal conditions, noise, vibration and the presence of pollutants, allergens and odours. In this respect, vehicle cabins are similar to other built environments. However, due to their mobility and the often highly variable on-road conditions encountered during even a short trip, vehicles represent a unique environment. The total environmental quality of a vehicle cabin seems to be greater than the sum of its individual parts, and it therefore necessitates a synergistic approach to its evaluation and prediction (Gameiro da Silva, 2006). Nevertheless, the overwhelming majority of in-vehicle studies have addressed a single variable of interest.

### Overview of Pollution Inside Vehicles

The introduction of automobiles powered by internal combustion engines brought with it the problem of occupant exposure to the emissions of other proximate vehicles. As automobile numbers increased, so too did the magnitude of this problem, prompting initial efforts to ventilate vehicle cabins; an undertaking that met with varying degrees of success (Daly, 2006). Currently, various components can be included in a standard automotive heating ventilation and air conditioning (HVAC) system in order to better protect occupants from combustion-derived pollutants and other undesirable products. These include pollen filters, high efficiency particulate air (HEPA) filters, germicidal lamps, activated carbon filters, photo-catalytic oxidation-based filters and air quality sensors that can automatically trigger a change in ventilation based on pollutant concentrations (Daly, 2006). Despite the availability and diversity of these accoutrements, they are not necessarily fitted to all new automobiles, with the possible exception of standard dust/pollen filters (Pui et al., 2008). Many older vehicles are unlikely to have any such device.

Fuel combustion-related pollutants typically encountered in the vehicle cabin environment include CO, oxides of nitrogen ($NO_x$), a suite of volatile organic compounds (VOCs) and particulate matter (PM) of varying size (Weisel, 2001), all of which can have short or long-term negative effects on human health. In addition to these pollutants, allergenic pollens (Muilenberg et al., 1991; Hugg et al., 2007) and fungi (Simmons et al., 1997) have been measured inside vehicles. The source of pollutants present in vehicles under most conditions is generally other vehicles, particularly those immediately preceding a given vehicle (Clifford et al., 1997; Fruin et al., 2008). However, in some instances a vehicle can self-pollute. Typical causes of this include exhaust leaks that intrude into the cabin (Ziskind et al., 1981) and malfunctioning fuel distribution systems (Lawryk et al., 1995). Another likely self-pollution mechanism is turbulent entrainment of tail-pipe emissions and their subsequent re-entry via leakage pathways and open windows (Behrentz et al., 2004).

Some incidental in-cabin pollutant sources include VOC emissions from materials in new cars (Brown and Cheng, 2000; Zhang et al., 2008), smoking (Ott et al., 1992; Park et al., 1998; Offermann et al., 2002; Matt et al., 2008; Ott et al., 2008), occupant-produced emissions (Björkqvist et al., 1997), windshield washing fluid (Becalski and Bartlett, 2006), colonisation of HVAC system components by fungi (Simmons et al., 1997), polybrominated diphenyl ethers released from various vehicle components (Mandalakis et al., 2008), air-bag deployment and transport of polluting materials such as dry-cleaned clothes (Park et al., 1998).

### Exposure and Duration of Vehicle Occupancy

For an in-vehicle pollutant exposure to occur, the envelope of an occupant's body must come into contact with a pollutant. This is a different concept to that of dose, which occurs when a pollutant crosses the envelope (Ott, 1985). The duration for which this contact occurs is another key dimension of exposure (Zartarian et al., 2007). Although the concepts of pollutant exposure and dose are closely related, they are fields of study unto themselves, and this thesis deals with assessment of exposure.

The time-activity pattern of a person defines how much time they spend engaged in a range of activities, generally over the course of 24h. An understanding of time-activity patterns representative of various groups of people is integral to the estimation and mitigation of their pollutant exposures (Ryan and Lambert, 1991). One of the largest studies of time-activity patterns was reported by Klepeis et al. (2001). The study, known as the National Human Activity Pattern Survey (NHAPS), surveyed in excess of 9000 United States residents. From this, it was calculated that approximately 6% (~1.5h) of a US resident's daily time is spent in a vehicle, a figure comparable with results reported for the working European population (Weisel, 2005). Based on a large survey of approximately 24,000 people between 2004 and 2007, it was estimated that residents of Sydney, Australia spend 1.3h per day in-transit on average during weekdays, and approximately 70% of those surveyed used private vehicles as their mode of transport, either as a driver or passenger (New South Wales Transport Data Centre, 2008). This result was therefore generally consistent with those reported by the aforementioned American and European studies.

### Studies of In-Vehicle Pollution

There have been scores of in-vehicle pollutant studies published since the 1960's. A complete survey and analysis of the literature is not attempted here. However, one striking factor to emerge when reviewing works on this topic is that in-vehicle studies of particle concentration are seemingly less common compared to studies addressing other pollutants. A similar observation was made by Weisel (2001) in his review of transportation air quality. The reasons for this are not clear, although the availability of instrumentation that is both portable and capable of high temporal resolution with reasonable accuracy (both desirable characteristics for in-vehicle monitoring) is possibly the main cause of the general lack of in-vehicle particulate studies, as such equipment has become only become more widely available in relatively times. The fact that the overwhelming majority of studies have been performed in the last 10 years tends to support this hypothesis.

Particles form part of the airborne cocktail known as an aerosol, which comprises solid or liquid particles suspended in a gaseous envelope (Hinds, 1999). Particles are typically grouped into three broad categories; with $PM_{10}$ and $PM_{2.5}$ respectively describing all particles characterised by an aerodynamic diameter less than 10 and 2.5μm, respectively, and ultrafine particles (UFPs) defined as those of diameter <100nm. A detailed description of particle characteristics is given in section 1.2.

Numerous studies have reported in-vehicle measurements of particles (Morandi et al., 1988; Ptak and Fallon, 1994; Kingham et al., 1998; Rodes et al., 1998; Alm et al., 1999; Gee and Raper, 1999; Praml and Schierl, 2000; Zagury et al., 2000; Adams et al., 2001; Rank et al., 2001; Abraham et al., 2002; Chan et al., 2002a; Chan et al., 2002b; Dennekamp et al., 2002; Levy et al., 2002; Riediker et al., 2003, 2004; South Eastern Sydney Public Health Unit and New South Wales Department of Health, 2003; Gulliver and Briggs, 2004; Gómez-Perales et al., 2004; Greaves, 2006; Kaur et al., 2006; Diapouli et al., 2007; Hammond et al., 2007; Yokoyama et al., 2007; Zhu et al., 2007; Adar et al., 2008; Briggs et al., 2008; Lee et al., 2008; McNabola et al., 2008; Pui et al., 2008; Qi et al., 2008; Rim et al., 2008; Weichenthal et al., 2008; Tsai et al., 2008; Asmi et al., 2009; Boogaard et al., 2009; Huang and Hsu, 2009; Longley et al., 2009). From reviewing these works, several points of interest were noted, namely;

*a)* Some studies focussed exclusively on public transport modes, and of those that investigated passenger vehicles, relatively few of these performed measurements of the UFP size range;
*b)* where the influence of ventilation settings on in-cabin particulates was assessed, it was typically done in a qualitative manner;
*c)* few studies assessed multiple vehicles and the role of vehicle type and age;
*d)* only a modicum of work had related on-road particle concentrations with those inside vehicle cabins, and;
*e)* the effect of roadway environments like underground tunnels was not routinely assessed, despite the likelihood that this location is associated with elevated concentrations of particulates by virtue of its enclosed nature.

Studies focussed on UFP characterisation inside passenger vehicles are a relatively recent feature in the literature. Where UFPs were measured, only a handful of studies (Zhu et al., 2007; Pui et al., 2008; Qi et al., 2008) attempted a systematic investigation of UFP pollution inside passenger vehicles, which, as stated previously, are the most popular transport mode in Sydney. In the first such study, Zhu et al. (2007) concurrently measured UFP concentrations inside and outside three passenger vehicles fitted with HVAC filters under three ventilation settings while travelling on Los Angeles freeways. Particle size distributions were also measured. Zhu et al. (2007) reported that maximum occupant protection from on-road UFPs occurred when an air recirculation setting was used, and that the manufacturer-installed HVAC filters afforded 50% protection from 7-40nm particles, and 20-30% protection from particles between 40 and approximately 200nm. Newer vehicles were found to permit less UFP ingress than older vehicles, although the range of vehicle ages assessed by Zhu et al. (2007) was quite small (2000 model – 2005 model). Based on their measurements, Zhu et al. (2007) estimated that 10-50% of daily UFP exposure could occur in-vehicle, depending on the proximity of workplace and home environments to significant UFP sources. Subsequent estimates reported by Fruin et al. (2008) placed the in-vehicle UFP exposure contribution to total exposure range for Los Angeles residents from 33-45%. Given these findings, it would seem beneficial to be able to employ modelling techniques to predict in-cabin UFP concentrations so that exposures could be better understood and managed. Only one such study (Xu and Zhu, 2009), published very recently, has been identified. Using data collected by Zhu et al. (2007) and Gong et al. (2009), Xu and Zhu (2009) used a mass-balance model to predict on-road UFP ingress into vehicle cabins and elucidate the relative importance of vehicular factors governing this. They found that mechanical (HVAC) air flow, filtration efficiency particle penetration, deposition and vehicle speed all had varying roles to play, depending on in-vehicle ventilation conditions.

Both Pui et al. (2008) and Qi et al. (2008) performed a suite of UFP measurements inside the same two newer vehicles (2003 model and 2007 model). Simultaneous UFP measurements outside the vehicle cabin (i.e. on-road) were also performed in some cases. Both vehicles were filter-equipped. Pui et al. (2008) reported a 46% total UFP removal efficiency based on on-road testing under a non-recirculate ventilation setting inside the 2003 model vehicle. Laboratory wind tunnel tests of HVAC filters highlighted minimum particle filtration efficiencies of 17.4% and 22.9% for ~350nm particles (above the upper size limit of the UFP fraction) under high and medium air flow rates, respectively (Qi et al., 2008). Qi et al. (2008) reported on-road HVAC and filter combined

UFP removal efficiencies of 55.0% and 65.6% for their respective high and medium fan settings (non-recirculate). The removal efficiencies measured in terms of particle number concentration or the surface area of particles depositing in the human lung were comparable. The reduction achieved with the filter removed, based on measurements using the latter metric, ranged between 7.9% and 39.4% for non-winter and winter conditions, respectively, for the medium fan speed condition. The equivalent range for a high fan speed was 10.3% to 26.4%. Qi et al. (2008) attributed the high winter reductions to the gradient effects associated with a temperature difference of >25°C between the outdoor environment and the vehicle's heating system and cabin. When an air recirculation setting was introduced, particle concentrations inside a vehicle capable of filtering recirculated air rapidly (~3 minutes) reached typical background levels, due to a particle removal efficiency of 45.5% (in terms of particle number concentration) and frequent air recirculation (Pui et al., 2008). Where no filtering of recirculated air was present (removal efficiency = 27.2%), or the filter was removed from the HVAC system (removal efficiency = 19.1%), the time taken for such a reduction was 10-13 minutes, with similar results reported by Qi et al. (2008) in terms of surface area of particles depositing in the lung. A model was developed by Pui et al. (2008) that described the reduction through time of in-vehicle particle concentration under a recirculation setting. A continuous and significant reduction in UFPs was observed, while large particles were also removed, albeit with somewhat reduced efficiency (Pui et al., 2008).

In addition to the knowledge gaps noted previously, the synthesis of primary findings from this small pool of relevant studies indicates that;

*a)* a disproportionately large contribution to daily UFP exposure (up to 50%) can be incurred during 1.5h of vehicle occupancy;

*b)* the ability of typical cabin air filters to reduce this exposure is moderate, and;

*c)* ventilation settings and vehicle age are both important determinants of in-cabin UFP levels.

None of the above in-vehicle UFP studies examined the effect of road tunnel travel on in-cabin UFP exposure, which could potentially be significant (Gouriou et al., 2004). Based on the knowledge gaps identified, the relevance of UFPs, road tunnels and vehicle ventilation parameters are described in the following sections, prior to an integration of the information presented in this chapter into the aims of this thesis.

## 1.2 Ultrafine Particles

Aerosols exhibit complex and often highly dynamic behaviour. The constituent particles suspended within them are produced by a multitude of processes, comprise a substantial variety of chemical compositions, range through many orders of magnitude in size and can affect everything from global weather and climate phenomena to the production of microchips. They can negatively impact upon the health of humans and animals, the aesthetic amenity of places and the appearance and quality of valuable cultural artefacts (amongst other facets), and they can do so across a variety of indoor and outdoor environments. Furthermore, by virtue of their diverse parent-processes, airborne particles are ubiquitous in the environment. It is therefore not surprising that particles of various types have been the focus of myriad theoretical and empirical research efforts across diverse disciplines. In any discussion of particles, an understanding of their general characteristics is important. Hence, the following section is devoted to outlining key aspects of UFPs.

### Sources

UFP concentrations typically reflect the contribution of anthropogenic processes to a pre-existing background concentration (Morawska et al., 2008). Background concentrations are generally ascribed to natural processes, such that in environments free from the immediate influence of anthropogenic activities, UFPs are present and their concentrations readily measured. Some natural sources of particles of various sizes include the interaction between wind and oceans (e.g. sea spray), oceanic phytoplankton emissions, VOC emissions from various types of plants, geological processes, seasonal pollen releases, forest fires, wind-entrained dust and natural decomposition processes (Koutrakis and Sioutas, 1996; Eastwood, 2008). Bioaerosols containing bacteria, viruses, spores, pollens and debris can fall into something of a grey area with respect to classification of their source, as the mechanisms by which bioaerosols are released are generally initiated by humans (Eastwood, 2008). The particles produced by the aforementioned sources vary appreciably in size, as shown in figure 1-1. Despite the manifold natural sources of UFPs, vehicular fossil fuel combustion has repeatedly been shown to constitute the dominant source of UFPs in urban areas (Morawska et al., 2008). Other anthropogenic particle sources include industrial processes (e.g. smelting), biomass combustion (e.g. wood-fired heaters or stoves), non-vehicular fossil fuel combustion (e.g. power plants), agricultural processes (e.g. spraying of pesticides), disintegration of materials (e.g. the wearing of a vehicle's tyres, brake pads and clutch), cooking and incineration (Spengler and Wilson, 1996; Eastwood, 2008). In general terms, anthropogenic particles are smaller than those produced naturally (see figure 1-1).

An important distinction is between primary and secondary particles. The primary variety are emitted from their source as particles, whilst secondary particles are formed following homogenous nucleation of gases (Koutrakis and Sioutas, 1996; Jacobson, 2002). This occurs when a gas, or gases, nucleate(s) in the absence of a pre-existing surface (Jacobson, 2002). Primary and secondary particles can be attributed to many natural and anthropogenic sources. Ozone can react with certain VOCs to form secondary aerosols in both indoor and outdoor environments, and this is an oft-studied pathway of particle formation. Given the dominance of vehicle emissions as an urban UFP source, it is of most relevance to describe their characteristics.

The particles produced by vehicular gasoline and diesel combustion differ with respect to their size and prevalence in terms of number concentration, with gasoline vehicles (often light-duty) typically emitting the bulk of particle number counts in the 20-60nm range, whilst the equivalent range for diesel vehicles (often heavy-duty) is 20-130nm (Morawska et al., 2008). Conversely, the overwhelming majority of particle mass emitted by vehicles is constituted by particles between 100-1000nm (Eastwood, 2008). The particles produced by vehicles are largely attributed to the combustion of fuel and lubricant, and, to a much lesser extent, material disintegration (Eastwood, 2008). Particle formation and dynamic processes within the engine, exhaust system and with dilution post-release are highly complex, and constitute a field of study in their own right. As such, a detailed investigation of these is not entered into here.

Heavy-duty diesel-powered vehicles (which rely on compression for fuel ignition) make a disproportionately large contribution to UFP concentrations in many environments, although light-duty gasoline-powered vehicles (which typically rely on spark ignition) can make significant contributions under certain circumstances; during rapid acceleration, for example (Fruin et al., 2008; Morawska et al., 2008). Other fuel types, such as Liquefied Petroleum Gas (LPG) have been shown during dynamometer tests to afford reduced particle emissions in terms of both mass and number (Ristovski et al., 2005), although this is not necessarily applicable to all LPG vehicles, and the number of vehicles powered by such fuels is currently likely to account for only a small proportion of the overall vehicle stock in many countries. For example, the 2006 Australian Motor Vehicle Census (Australian Bureau of Statistics, 2006) reported that LPG and dual fuel (i.e. capable of operating on more than one fuel) vehicles constituted 0.9-4.7% of the total vehicle fleet in Australian states. It should also be noted that some vehicles powered by alternative fuels, especially Compressed Natural Gas (CNG), may emit greater numbers of volatile nanoparticles under certain operating conditions compared to traditional fuels (Jayaratne et al., 2008).

The sulphur content in diesel fuel has been examined in detail recently, as the sulphate particles and sulphuric acid produced by diesel combustion are deleterious not only with respect to environmental pollution, but also in terms of corrosion of engine components and impairment of catalytic converters (Eastwood, 2008; Morawska et al., 2008). The results of several studies conducted in different countries have not been unequivocal regarding their findings on the influence of sulphur content on UFP emissions; however, diesel fuels with lower sulphur content often led to reduced nanoparticle (<50nm) emissions (Morawska et al., 2008). Ultra low sulphur content (generally <10ppm) diesel fuel is currently being introduced in many locations, whilst in others it has already been available for some time.

**Size and Dynamics**

The size of particles is the key factor controlling their behaviour (Hinds, 1999). Particle size is often transient, as particles of certain sizes undergo rapid changes in response to environmental conditions (e.g. air temperature) and the presence of other particles. Typical distributions of particle number and mass reflect this, as figure 1-2 shows. Nucleation mode (<100nm, terminology often used interchangeably with UFP) particles constitute the vast (~90%) majority of particle number emitted by vehicles (Kittelson, 1998, Morawska et al., 2008), as shown in figure 1-3. Particles within this mode impact upon each other and coalesce into larger particles, whilst certain gases condense onto these particles, causing them to grow by heterogeneous nucleation (Jacobson, 2002). The result of such processes is that particle numbers are reduced, while overall mass is relatively unchanged. Following these processes, particles exist in the accumulation mode (100 to ~1000nm), where the majority of vehicle-emitted particle mass lies (Eastwood, 2008). The combined nucleation and accumulation modes constitute what is often referred to as fine PM (Hinds, 1999); a term that often refers to $PM_{2.5}$. Although the mass of UFPs or nucleation mode particles is small, their combined surface area is significant, as figures 1-2 and 1-3 show.

The production of the smallest nucleation mode particles from vehicular combustion is typically a secondary process, and occurs when hot, volatile gases are exhausted from a vehicle and rapidly cooled by, and mixed with, ambient air (Kittelson, 1998; Morawska et al., 2008). However, a variety of mechanisms can substantially dampen the formation of nucleation mode particles, such as the presence of a large accumulation mode capable of scavenging volatile material (Morawska et al., 2008). Some accumulation mode particles are emitted directly as solid carbonaceous agglomerates, which then provide a surface for adsorption of nucleation mode particles (Kittelson, 1998). The characteristics and condition of a vehicle itself can also exert some influence on nucleation mode particle

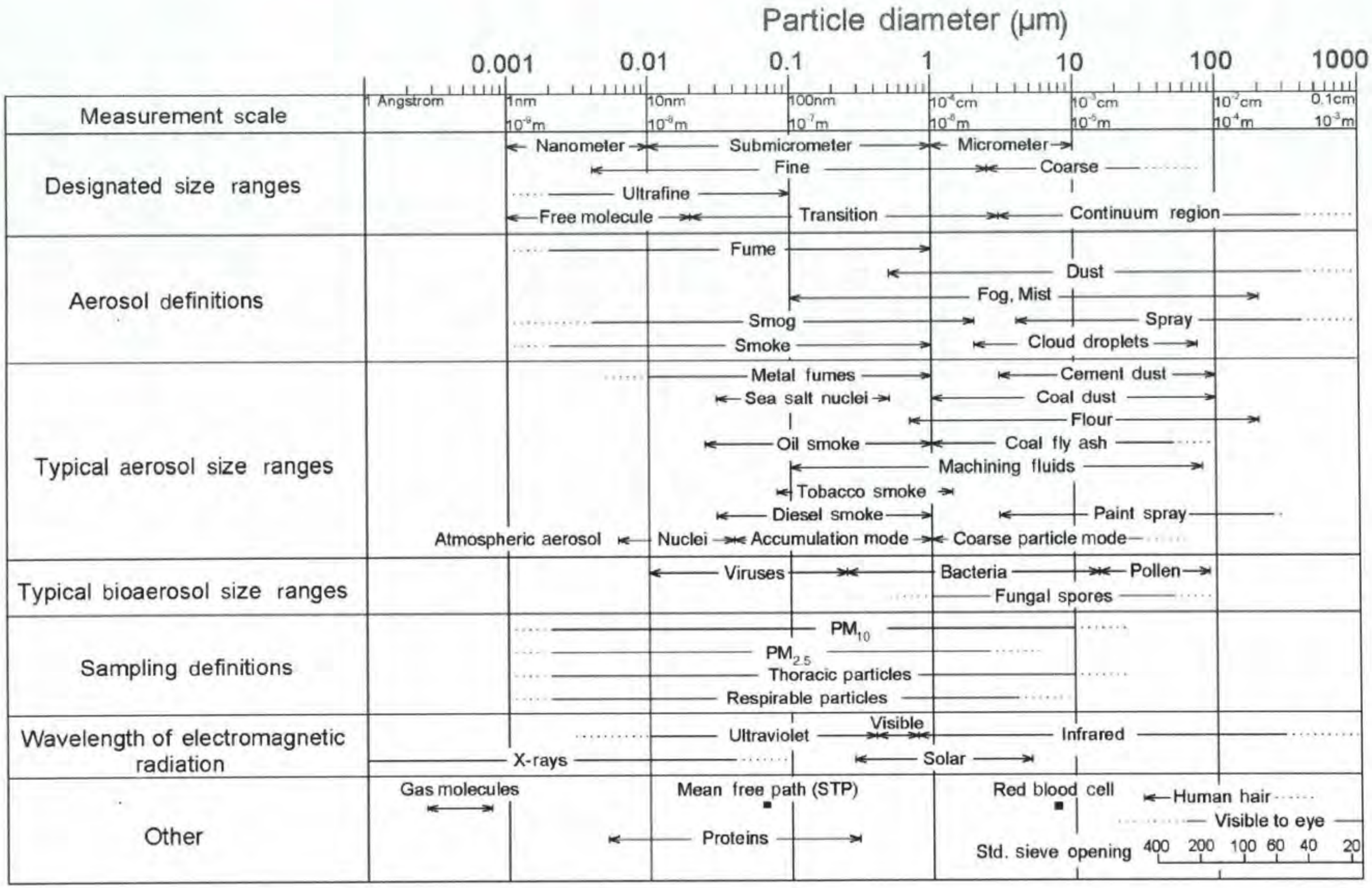



**Figure 1-1.** Definitions of various particle size measurement descriptors and typical size range of some natural and anthropogenic particles. Other natural phenomena are shown for comparison. Source: Hinds (1999).

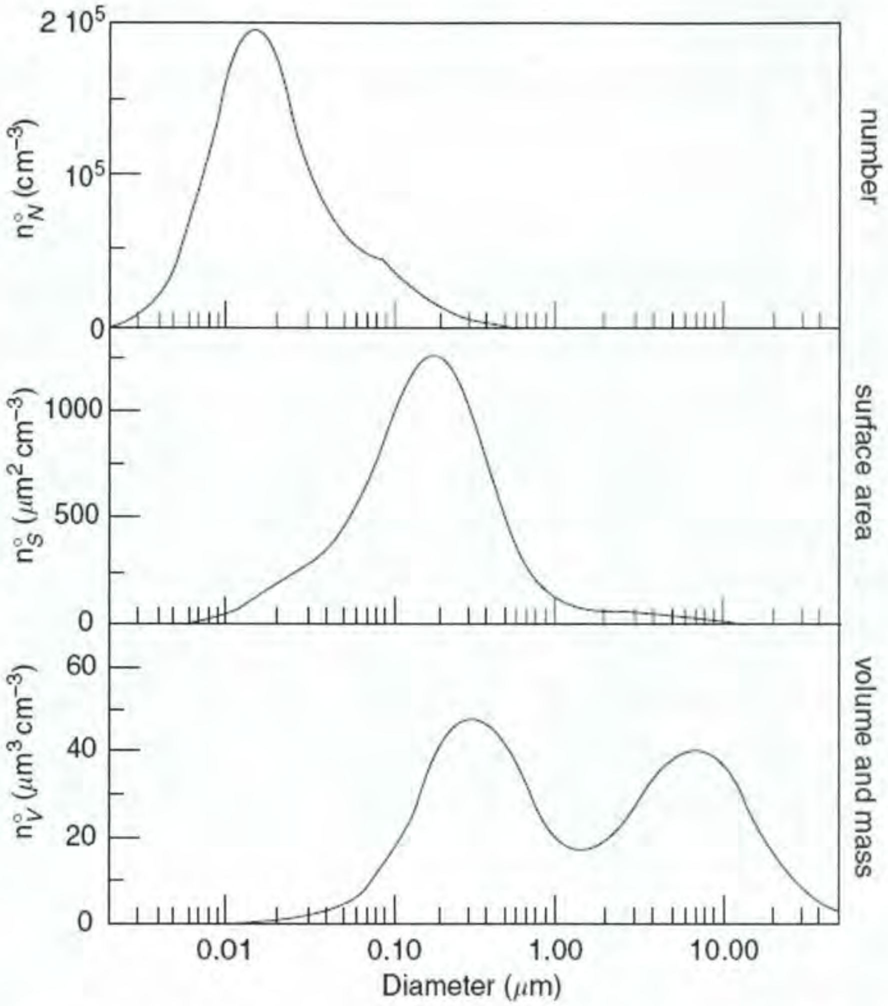



**Figure 1-2.** Size distibution of particles in a typical urban aerosol in terms of number, surface area, mass and volume. Source: Gundel and Sextro (2004), from Seinfeld and Pandis (1998).

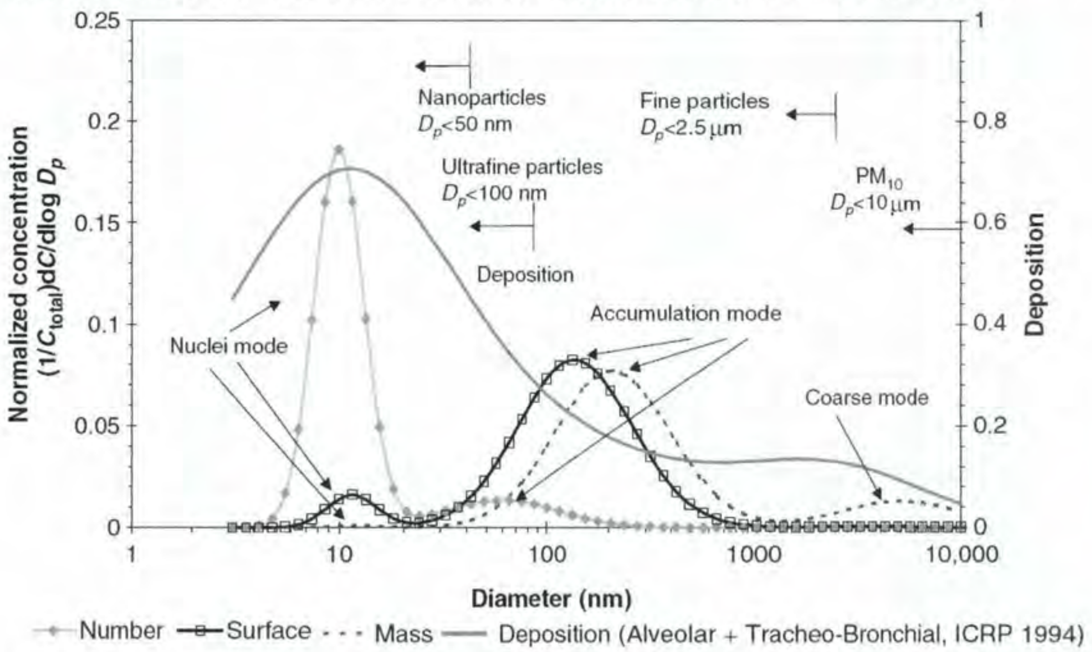



**Figure 1-3.** Typical particle size distribution in a diesel exhaust aerosol in terms of number, surface area, mass and alveolar/tracheo-bronchial deposition. Source: Kittelson et al. (2004c). $D_p$ = particle diameter.

formation. Thus, although UFPs are a dominant constituent of urban air in terms of their number, the conditions necessary for their formation are specific and the emission of UFPs from vehicles of different types can be highly variable. These characteristics of UFPs make the representativeness of real-world conditions sometimes difficult to achieve in laboratory studies, and underscores the value of field-based investigations. Whilst UFPs typically undergo relatively rapid transformation into larger particles, they are also emitted *en masse* with high frequency, making assessment in areas proximate to their source (such as vehicle cabins) an important component of field-based research. The relatively high efficiency with which UFPs deposit in the human respiratory tract, as shown in figure 1-3, is another key aspect that makes exposure assessment important. This is discussed further in a subsequent section.

In any discussion of particle characteristics, especially size, it is useful to note that particle morphology is often irregular and inconsistent, excluding liquid particles, which maintain their sphericity (Hinds, 1999; Eastwood, 2008). Thus, any mention of particulate diameter is generally founded on some concept of equivalent diameter. Metrics most commonly encountered are those of aerodynamic equivalent diameter and Stokes' diameter, with the former more frequently reported than the latter (Hinds, 1999). Aerodynamic equivalent diameter is calculated for a given particle as the diameter of a spherical particle with a density equal to that of water (1g $cm^{-3}$) that would settle with the same velocity as that of the particle (Hinds, 1999). Stokes' diameter is the diameter of a sphere that has the same settling velocity and density of the particle (Hinds, 1999). The concept of aerodynamic equivalent diameter is represented in figure 1-4.

Another key issue related to particle size is the number of particles of a given size within an aerosol sample. This is depicted by the particle size distribution, which describes the number (or mass, or surface area) of particles of various sizes, as shown in figures 1-2 and 1-3. In their raw form, particle size distributions from most sources characteristically exhibit an elongated right tail, and thus appear approximately normally distributed after being logarithmically-transformed (Hinds, 1999). Several methods exist for measuring size distribution

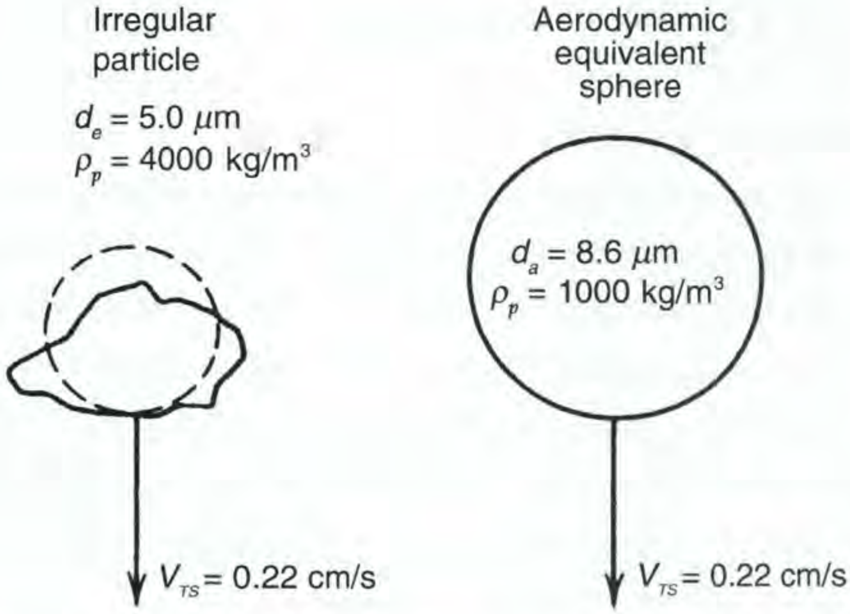



**Figure 1-4.** The aerodynamic equivalent sphere of an irregular particle. Source: Hinds (2004). $d_x$, $\rho_p$ and $V_{TS}$ refer to particle diameter, density and terminal settling velocity, respectively.

in the field or laboratory, and the most common of these are briefly described in an ensuing section.

Numerous natural removal mechanisms exist for particles. Nucleation mode particles have a typically short lifespan post-emission or formation, as they rapidly coalesce and form accumulation mode particles (Hinds, 1999). However, whilst in the nucleation mode, particles may removed by rainout; whereby the particle forms affords the formation of a raindrop and is thus removed from the atmosphere (Hinds, 1999). Brownian (random) motion (i.e. diffusion) and subsequent deposition onto surfaces can also act to remove nucleation mode particles (Hinds, 1999; Eastwood, 2008). Accumulation mode particles have a relatively long atmospheric lifetime, as their small size hinders sedimentation whilst being too large to be affected substantially by the effects of diffusion, as figure 1-5 shows. There is minimal mass exchange between the accumulation mode and the larger coarse (>1000 to 3000nm) mode (Hinds, 1999). When accumulation mode particles are removed from the atmosphere, rainout, washout (direct impaction of precipitation onto a particle) and deposition onto available surfaces (i.e. at ground level) are often responsible (Hinds, 1999; Jacobson, 2002). Given the similarity between the size of accumulation mode particles and the wavelength of visible light (see figure 1-1), they are typically implicated in the effects of air pollution on visibility (Hinds, 1999; Jacobson, 2002). Coarse particles are atypically and inconsistently generated by vehicular combustion (Eastwood, 2008), and are prone to relatively brief atmospheric residence times by virtue of their mass (Hinds, 1999). As such, they are of significantly reduced interest within the context of studies focussed on vehicle emissions and human exposure.

Other mechanisms can act to remove particles from the air. Temperature gradients (thermophoresis), electrostatic attraction, interception with objects and inertial impaction can all lead to particle deposition, although their effects are size dependent (Eastwood, 2008). Such processes can be exploited where deliberate capture of particles is desirable, and many particle control systems installed in locations ranging from vehicle HVAC systems to large power plants rely upon these removal mechanisms. Once particles are deposited onto a surface, they may be resuspended. However, resuspension of deposited particles

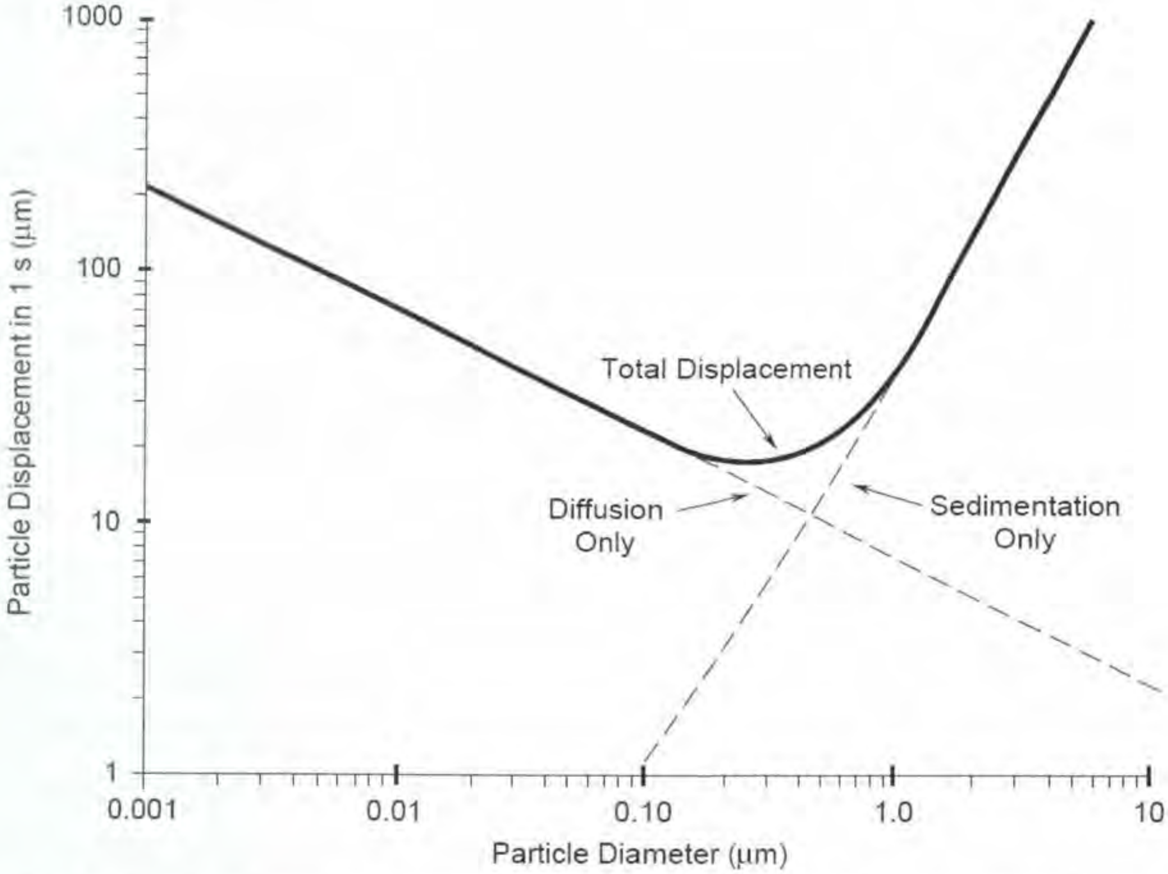



**Figure 1-5.** Particle displacement (assuming density = 1g cm$^{-3}$) in still air attributable to the combined and individual effects of diffusion and sedimentation. Source: Phalen (2008).

is difficult due to adhesive forces (Eastwood, 2008). When resuspension does occur, it mostly affects larger particles, and field studies conducted in indoor environments have shown submicrometer (<1000nm) particles are typically not resuspended as a consequence of common indoor conditions (Morawska and Salthammer, 2003b).

### Chemical Composition

Assessment of the chemical composition of vehicle-generated UFPs has received relatively little research attention to-date (Cohen, 2004; Morawska et al., 2008), and is accordingly given a very brief treatment here. This does not, however, imply that this field of research is of limited relevance. Indeed, the chemical composition of UFPs is significant in determining UFP behaviour within vehicle engines and post-release, and also in terms of human health effects in addition to the many other domains where the influence of UFPs is felt.

UFPs are often produced as a secondary aerosol following homogenous nucleation of $SO_2$, $NH_3$ and $NO_x$ into $SO_4^{2-}$, $NH_4^+$ and $NO_3^-$(Koutrakis and Sioutas, 1996; Morawska et al., 2008). Elemental (EC) and organic (OC) carbon are substantial constituents of vehicular particulates. Both forms are generally present in the same particle, as OC comprises numerous VOCs that coat, in addition to the ions mentioned above, a solid EC core (Eastwood, 2008). A multiplicity of metals (Ca, Fe, Zn and Cu, to name a few) are also present in vehicle combustion-generated UFPs, although their contribution to total UFP mass may be relatively small compared to the aforementioned constituents (Eastwood, 2008; Morawska et al., 2008).

### Measurement

Airborne particles can be measured using many different approaches. Although Coulier is regarded as the first person to have assembled a means of detecting condensation nuclei in his 1875 experiments, it is John Aitken, who reported measurements performed using a portable apparatus for counting atmospheric dust in the 1880's and 1890's (McMurry, 2000; Spurny, 2005), whose name was immortalised in the fields of meteorology and aerosol science. Aitken nuclei mode is a term sometimes used interchangeably with UFPs (Koutrakis and Sioutas, 1996), and usually describes particles that provide a surface for water condensation. Over 100 years since the work of Aitken, there is now an extensive suite of readily available, commercially produced instrumentation for measuring particles. However, for many of those devices that are capable of UFP measurement, the principles underlying their operation are not substantively different from those of Aiken's early devices.

Given their size and relatively insignificant mass, UFPs demand a specific approach to their measurement, usually in terms of number concentration (typically, particles per cubic centimetre), although other properties can be measured (e.g. surface area). The overwhelming majority of UFP measurement studies utilise a condensation particle counter (CPC) either as a stand-alone instrument, or coupled with a device capable of separating particles in an incoming aerosol based on their size using electrostatic classification (Morawska et al., 2008). Many different CPCs are commercially available, although their operating principles are generally similar.

As UFPs are below the detectable limit of traditional light-scattering methods of particle measurement, an integral component of CPC operation is the growth of UFPs to detectable sizes by condensation of a supersaturated working fluid (such as n-butanol, isopropanol or water) onto the UFPs (Cheng, 2005). Following this process, the particles are of a size that is detectable, usually by light-scattering. Once an aerosol has been drawn into a CPC (typically via a pump operating at a set flow rate), it is saturated with the vapour of the working fluid before being cooled, such that condensation of the working fluid and particle growth occurs, and the particles are subsequently detected (Cheng, 2005). A typical CPC schematic illustrating these processes is shown in figure 1-6. Using the methods described above, some CPCs are capable of detecting particles as small as 3nm (McMurry, 2000; Sem, 2002; Cheng, 2005). Three methods have traditionally been employed in CPCs to achieve supersaturation of the working fluid, namely; adiabatic expansion, conductive cooling and mixing of hot and cold air (Cheng, 2005). Many of the currently available CPCs, such as the TSI 3007 depicted in figure 1-6, utilise conductive cooling.

If a CPC is used as a stand-alone instrument, the measurements recorded will typically represent the number concentration of particles within the CPCs detectable size range (Morawska et al., 2008). In certain instances, it is useful to obtain measurements of the particle size distribution. If this is the case, sampled aerosols are first classified electrostatically such that only particles of a certain size become mobile prior to counting by a CPC. Using this method, a particle size

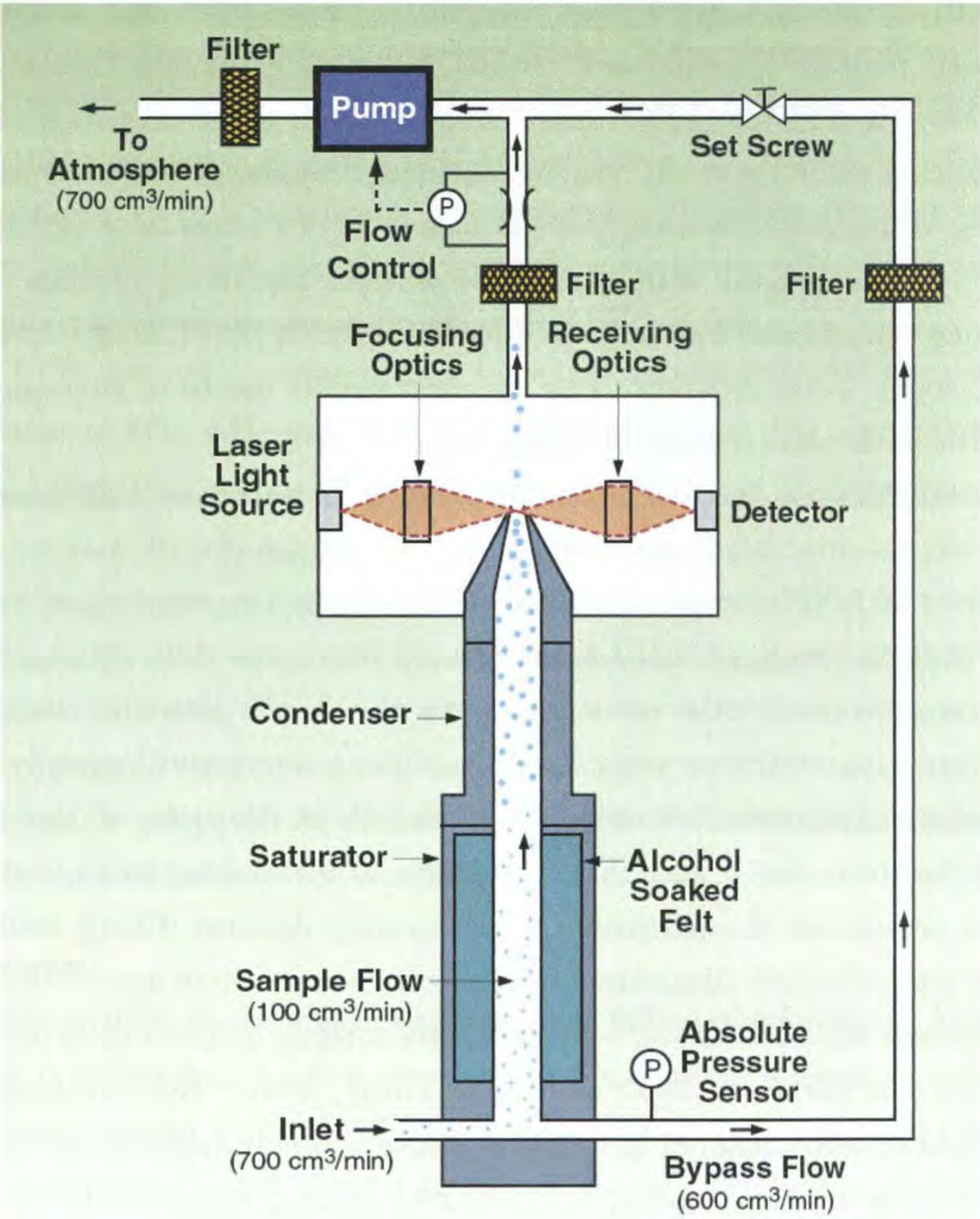



**Figure 1-6.** Schematic of a TSI 3007 CPC. Source: TSI (2004).

distribution measurement is soon acquired. The most oft-encountered system that exploits the well-characterised electrical mobility of differently sized particles is the scanning mobility particle sizer (SMPS) (Morawska et al., 2008). Once a particle size distribution is measured, the total particle number concentration across the size range of the distribution can be calculated. Other means of measuring particle number concentration and size distribution are available, although these are referred to less frequently in the literature than SMPSs and CPCs and are not discussed here (see Baron and Willeke, 2005). Also, there are myriad issues associated with sampling UFPs that can affect the veracity of the collected data. A description and discussion of these is not entered into here (for a comprehensive treatment see Hinds, 1999; Baron and Willeke, 2005), although due reference to, and brief discussion of, the most relevant of these appear in later chapters.

#### Health Effects

The potential effects of poor air quality on human health are graphically highlighted when one considers that a typical adult male human at rest will inhale 10.8 $m^3$ (12.96 kg at room temperature) of air per day; a figure that is one order of magnitude greater than the combined amount of food and water ingested per day in terms of mass (Phalen, 2008). Inhaled air is thus an important vehicle in delivering pollutant doses to the human body (Phalen, 2008).

In contrast to the relatively well characterised physical behaviour of UFPs, their specific effects on the health of humans are not well understood. Numerous studies have established statistically the relationship between concentrations of fine particle pollution and mortality. Of these, arguably the best known is the Harvard Six Cities Study, which began in 1974 (see Dockery et al., 1993). After following a cohort of over 8000 adults from six cities across the United States for 14-16 years (depending on city), and adjusting for confounding factors (e.g. smoking), the authors found strong a relationship between fine (i.e. $PM_{2.5}$) and sulphate particle mass and mortality, with particle pollution associated with deaths from lung cancer and cardiopulmonary disease (Dockery et al., 1993). Studies conducted in other locations have reported similar findings. For example, within the context of Sydney, Australia, Morgan et al. (1998a) largely confirmed the results of Dockery et al. (1993), with respect to the relationship between particulate pollution and mortality, albeit using a different study design and focusing on a reduced time period (1989-1993). Morgan et al. (1998b) found associations in Sydney between particulate pollution and hospital admissions of mainly elderly persons for heart disease and chronic obstructive pulmonary disease (COPD) in the period 1990-1994.

The specific role of UFPs in determining the results of many epidemiological studies is not clear, as the results were based on measurements of particle mass, rather than a parameter more sensitive to the presence of UFPs such as particle number concentration (Morawska et al., 2004). A limited number of studies specifically monitored UFP concentration as part of epidemiological investigations and generally reported inverse relationships between UFP number concentrations in urban air and respiratory function indicators in asthmatic adults and children (Wichmann and Peters, 2000). The effects of UFPs on mortality were observed to be delayed relative to, and possibly induce health effects independent of, those caused by fine particles (Wichmann and Peters, 2000).

The exact mechanisms with which particulates cause morbidity or mortality are not elucidated by epidemiologic studies (Delfino et al., 2005; Pope III and Dockery, 2006). To overcome this, many *in-vitro* and *in-vivo* laboratory and clinical assessments of particulate, especially UFP, effects on human and animal (as human surrogates) health endpoints have been performed. Numerous undesirable health effects have been reported, often as a result of inflammatory responses caused by reactive oxygen species produced following deposition of inhaled particles (Eastwood, 2008). The result of inflammation may not always manifest clinically significant symptoms in mostly healthy people (Gong Jr. et al., 2008); however, those with existing health complaints (COPD, asthma, chronic bronchitis, history of heart problems) most often suffer acute deleterious health effects. The effects of chronic UFP exposure are not well documented, although atherosclerosis (thickening of the arterial walls), heart disease, and potential for carcinogenesis due to significant number of known or suspected carcinogens in vehicle (especially diesel) emissions seem likely (Sioutas et al., 2005; Eastwood, 2008).

The main properties of UFPs that make them damaging to human health are related to (Wichmann and Peters, 2000);

*a)* the high efficiency with which they deposit, via diffusion, in the respiratory system (see figures 1-3, 1-7 and 1-8) and their ability to reach the deepest sections of the lung (i.e. alveoli; see figure 1-7) (Oberdörster et al., 2004; Cohen, 2004);

*b)* their subsequently low removal rates in the alveolar region due to the absence of mucocilary removal processes (capture of particles by a mucus layer and subsequent clearance via the undulations of microscopic, hair-like cilia) and reliance on phagocytosis (a removal process mediated by a type of white blood cell called a phagocyte) for particle clearance (Eastwood, 2008);

*c)* their significant combined surface area (see figures 1-2 and 1-3), which provides an ideal vehicle for efficient transport of toxic air pollutants (Delfino et al., 2005; Sioutas et al., 2005), and;

*d)* their ability, by virtue of their size, to effectively translocate to regions of the body other than where they were deposited (e.g. by evading the blood-brain barrier, passing into interstitial spaces or the bloodstream) (Oberdörster et al., 2004; Cohen, 2004; Pope III and Dockery, 2006).

The largest systematic review of the health effects of UFPs to-date was performed by Morawska et al. (2004). In their review of the literature, Morawska et al. (2004) separated previous work into three distinct categories; namely, epidemiological, toxicological and clinical studies. In terms of epidemiological investigations, Morawska et al. (2004) concluded that the existing data was too limited in with respect to the number of studies performed, number of subjects and location in which studies were conducted, such that authoritative conclusions could not be reached. Health effects induced by UFP exposure could be similar to

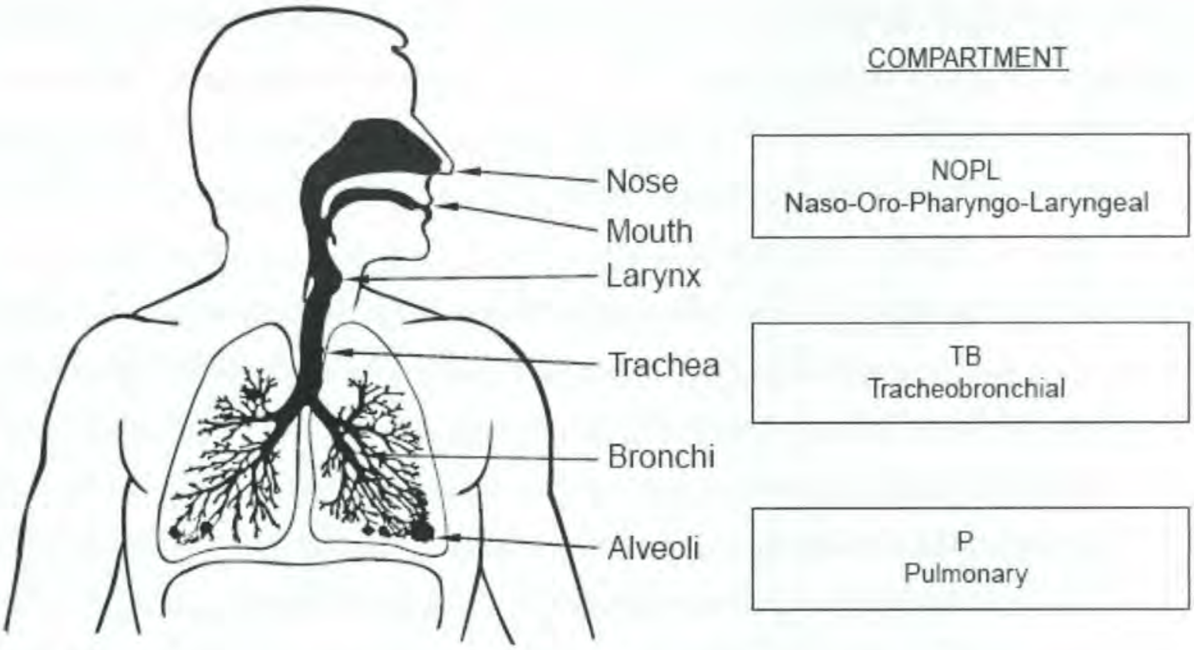



**Figure 1-7.** Major compartments of the human respiratory tract. Source: Phalen (2008).

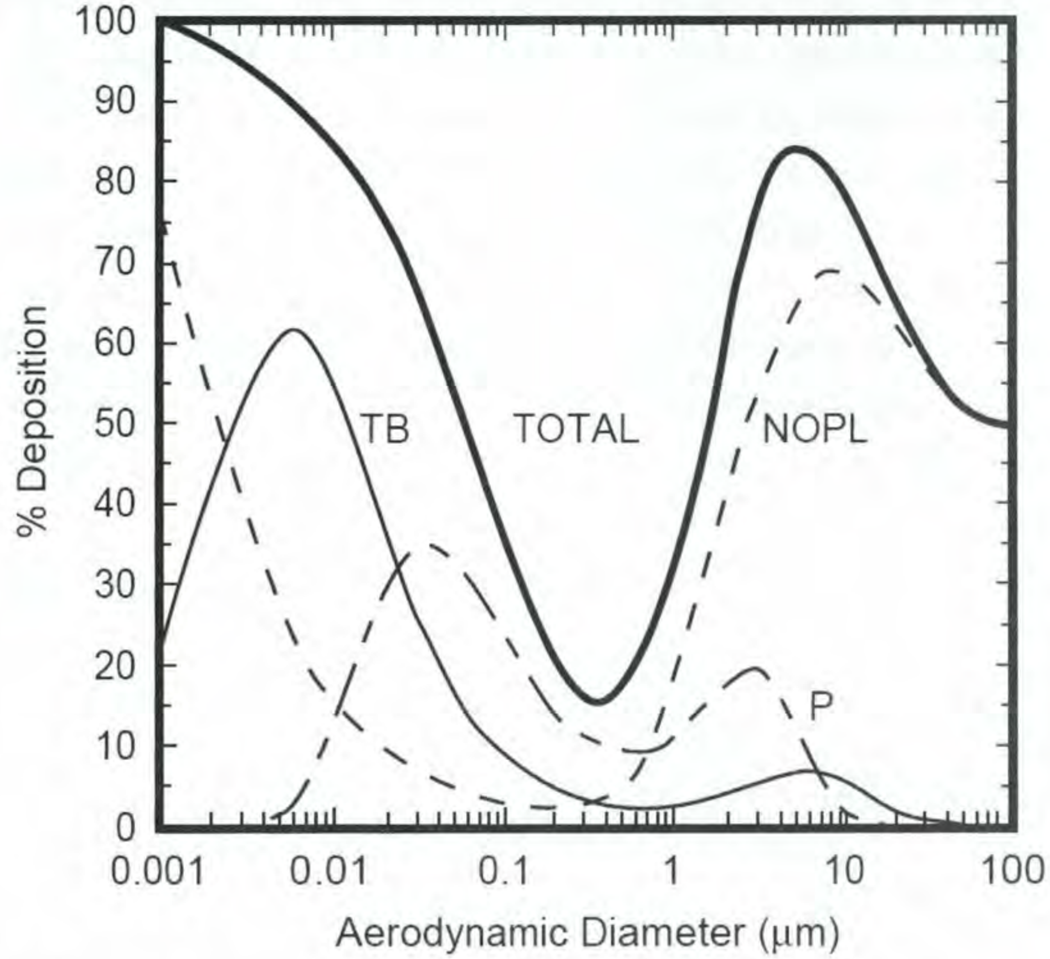



**Figure 1-8.** Deposition efficiency of particles of varying aerodynamic size in primary regions of the human respiratory system during normal breathing. See figure 1-7 for definitions of abbreviations. Source: Phalen (2008).

those caused by fine particle exposure; albeit with the former typically resulting in an effect of greater magnitude than the latter (Morawska et al., 2004; Sioutas et al., 2005). However, it has been noted that UFP number and fine particle mass, and their attendant health effects, are often poorly correlated; despite their shared provenance in vehicular emissions (in urban areas) and propensity to affect similar health end points, they often appear statistically independent (Ibald-Mulli et al., 2002; Morawska et al., 2004; Sioutas et al., 2005).

Toxicological studies of UFP heath effects reviewed by Morawska et al. (2004) indicated that, in general, inhalation of UFPs and associated adsorbed materials produces a cytokine (type of molecule involved in signalling and initiating immune responses) mediated inflammatory response. Some hypothesised pathways via which UFPs may induce negative acute and chronic health effects are shown in figure 1-9. The surface area of UFPs appears to be integral factor in precipitating such responses, as multiple toxic air pollutants absorbed onto the significant combined surface area of UFPs can be efficiently transported to the respiratory system (Sioutas et al., 2005). Given the negligible mass of UFPs, this parameter is thus an ineffective indicator of their potential health effects (Morawska et al., 2004). It should be noted, however, that considerable conjecture persists within the toxicological research community regarding the choice of particle number or surface area as the most appropriate UFP dose metric (Oberdörster et al., 2007; Stoeger et al., 2007; Wittmaack, 2007a,b).

The findings of clinical and controlled exposure studies reviewed by Morawska et al. (2004) typically supported the general toxicological theories noted above, with respect to the chain of events following UFP exposure and preceding an adverse health outcome. It was noted by Morawska et al. (2004) that a number of possible factors can induce bias, particularly in controlled exposure studies, and that future work would ideally address these and other potential short-comings in study design.

Due to the vagaries sometimes inherent in the dynamics and measurement of UFPs, coupled with the absence of a specific dose-response relationship between health outcomes and UFP concentrations, UFPs are not the subject of

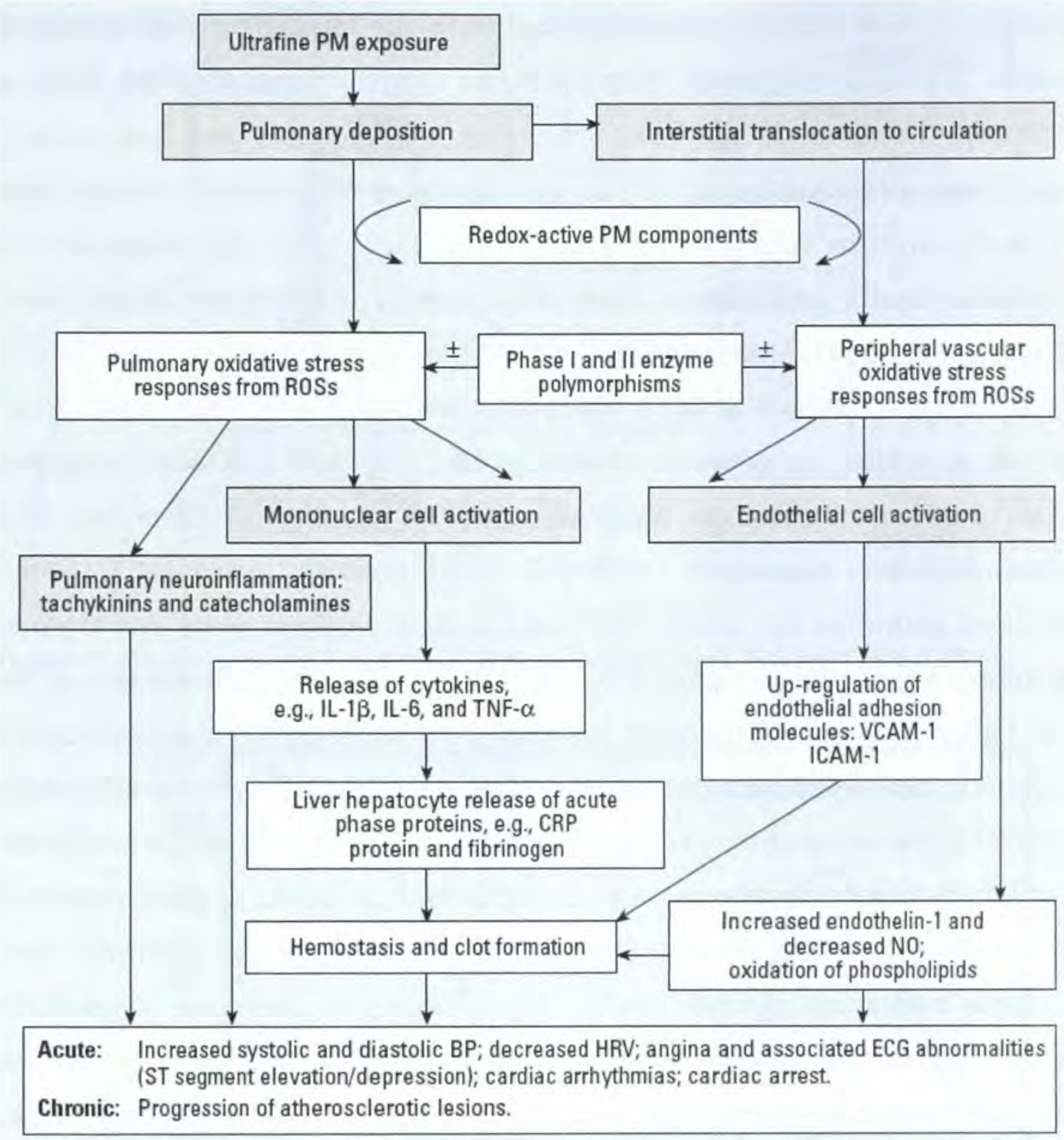



**Figure 1-9.** Suspected pathways via which UFPs could cause acute and chronic health effects in humans. Source: Delfino et al. (2005).

ambient air quality standards, as is the case with larger particle size fractions. However, it is now generally accepted that UFPs are implicated in undesirable human health outcomes, and standards are being promulgated in Europe regarding the emission of particles from vehicles on the basis of number concentration (Morawska et al., 2008). Given the volume of studies focussed on the health effects of UFPs, coupled with the rapid progress made in recent years (Morawska et al., 2004), it would appear to be a matter of 'when', rather than 'if,' the specifics of the health effects caused by UFPs are elucidated by the toxicological and medical research communities. The current lack of such knowledge does not, however, preclude the implementation of UFP particle characterisation and exposure studies, as these serve the highly important purpose of defining how and to what extent people are exposed to UFPs, and compliment studies performed from a health effects standpoint.

## 1.3 Road Tunnels and Ultrafine Particles

Road tunnels are an infrastructure element often required in many cities. This requirement most often stems from a need to alleviate traffic congestion. The benefit provided by the construction of a road tunnel in meeting this need should be reconciled with the local effects associated with the tunnel; especially in terms of air quality, noise and aesthetic implications. Easily overlooked in such considerations are the potentially substantial effects of the above factors on the users of the tunnel. During tunnel travel under free flowing and congested traffic conditions, the psychological impact of such factors on tunnel users, coupled with the enclosed nature and often underground location of tunnels, should not be underestimated (Arias et al., 2008; NHMRC, 2008; Götestam and Svebak, 2009).

The morphology of tunnels acts to confine and concentrate vehicular emissions, as natural dispersion mechanisms (i.e. wind) are not present in most cases. Ventilation must thus be achieved by mechanical means, although short tunnels (<300m) may be sufficiently ventilated by the piston effect associated with the vehicle movement along the tunnel, assuming unidirectional traffic flow (El-Fadel and Hashisho, 2001; NHMRC, 2008). Although most tunnels are short, as shown in figure 1-10, and occupancy times for users should be minimal, many factors can cause an increase in trip time. The nature of tunnel users' pollutant exposure, especially to UFPs, during travel is generally poorly understood (Kuykendall et al., 2009), despite tunnels constituting a location where peak UFP exposures can occur (Gouriou et al., 2004; Morawska et al., 2008). The acute exposures incurred during tunnel travel may precipitate acute health responses and contribute to the chronic effects of UFP exposure shown in figure 1-9.

In the past two decades, Australia has been a frequent user of road tunnel technology, although not to the same extent as Japan, for example. Table 1-1 shows the approximate cumulative length of urban road tunnels opened in various countries and/or cities in the period 1989-2007. There are also approximately 20km of tunnels, comprising four separate tunnels, currently at the proposal or construction stage in Brisbane, Australia (NHMRC, 2008). With the substantial current and future use of road tunnels both in Australia and overseas, the issue of in-tunnel air quality and its effects on the well-being of tunnel users demands attention. To achieve this, initial investigations should define the determinants of air quality and characterise the relationship between these and the pollutants of interest. Through such an approach, subsequent work can focus on the interaction between these parameters and the numerous health end points affected by them.

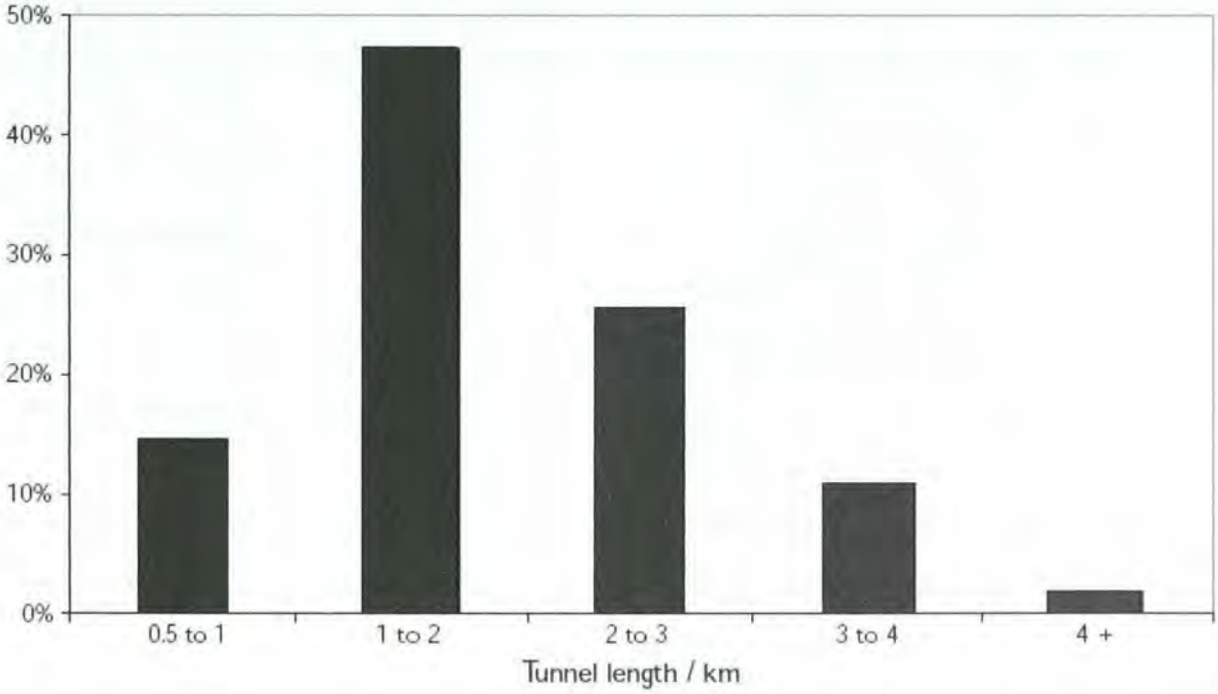



**Figure 1-10.** Distribution of tunnel length for 55 tunnels around the world (qualification criteria: length >0.5km). Source: NHMRC (2008).

### Roadway Environment and In-Vehicle UFP Concentration

The on-road UFP concentration is an integral element requiring consideration in any assessment of in-cabin air quality. Given the high degree of spatial and temporal variability often present in UFP concentrations, data representative of

**Table 1-1.** Approximate total length of urban road tunnels opened in various countries/cities in the period 1989-2007. Source: NHMRC (2008).

| Country, region or city | Tunnel length (km) |
|---|---|
| Dublin | 4.5 |
| Hong Kong | 17 |
| Japan: | |
| Chubu | 55 |
| Kanto | 13 |
| Kyushu-Okinawa | 12 |
| Kinki | 14 |
| Shikoku | 06 |
| Tohoku | 10 |
| Lyon | 06 |
| Melbourne | 05 |
| Sydney | 14 |
| Oslo | 09 |

UFP concentration on roadways must be collected on, or as close to, this environment as possible, as UFP concentrations decay to background levels within ~300m of major roadways (Hitchins et al., 2000; Zhu et al., 2002a,b). Accordingly, several recent studies have employed a mobile laboratory, typically a large van equipped with a suite of instrumentation, which is able to collect data while driving on a roadway. Such an approach is undoubtedly the ideal means to collect data representative of typical on-road UFP concentrations. Studies utilising this technique are described briefly in chapter 3.

Characterising the UFP concentrations on roadways and the factors that determine them are useful in assessing a road user's exposure. Of more use, however, is data relating on-road concentrations to those inside vehicles. As noted previously, very few studies have done this. These studies (Zhu et al., 2007; Pui et al., 2008; Qi et al., 2008) found that in-vehicle UFP levels in late-model vehicles were often substantially lower than those measured on-road, depending on the ventilation setting used. This underscores the importance of relating on-road UFP concentrations to in-vehicle levels, as on-road data are unlikely to be representative of in-vehicle conditions unless the windows of a vehicle are open. Data regarding the effect of tunnel travel on UFP exposure in-vehicles are extremely scant, and data representative of in-cabin UFP exposures incurred by occupants of older vehicles were not identified in the literature.

A number of studies reporting measurements of UFP concentration inside road tunnels have appeared in the literature during past decade (e.g. Kirchstetter et al., 1999; Abu-Allaban et al., 2002; Gidhagen et al., 2003; Gouriou et al., 2004; Jamriska et al., 2004; Geller et al., 2005; Larsson et al., 2007; Lechowicz et al., 2008). Of these, the overwhelming majority were based on measurements performed at a static point or points within a tunnel. While such an approach to measurement is well-suited to the subsequent calculation of vehicle fleet UFP emission factors (e.g. number of particles emitted per km travelled), its representativeness of on-road conditions during tunnel travel has not been comprehensively evaluated in a range of tunnels (see Gouriou et al., 2004), and therefore cannot be extrapolated with confidence.

As stated previously, the extent to which road tunnel travel contributes to daily UFP exposure has not been characterised. Knowledge regarding how the roadway environments used by vehicle occupants affect their daily exposure to UFPs would seem highly beneficial from both a public health and infrastructure planning standpoint. However, it is pertinent to note that individual tunnels are typically idiosyncratic in terms of the concentrations of pollutants within them and associated effects on human health (NHMRC, 2008). Where assessment of multiple tunnels in an area is not practical, it seems logical to focus research efforts towards the tunnel thought to be most affected by poor air quality and/or the tunnel used by the largest number of commuters.

### The M5 East Road Tunnel

The M5 East road tunnel is located in Sydney, Australia. It opened in December, 2001, and is part of the M5 Motorway. The M5 motorway is a constituent of the Sydney Orbital Network of roads. The tunnel lies in an East-West direction and consists of two unidirectional bores, each comprised of two lanes. The tunnel is approximately 4km long, which makes it one of the longest urban road tunnels in the world and Australia's longest operational road tunnel, although it will soon by overtaken in this respect by tunnels under construction in the Australian city of Brisbane (NHMRC, 2008). The tunnel is longitudinally ventilated via 131 jet fans. Fresh air is delivered from an external intake, and tunnel air exhausted through a stack located approximately 1km away. At the exit of each tunnel bore, air is drawn via cross-shafts and mixed with fresh air, before being delivered to the entry of the other bore. A schematic of the tunnel's ventilation system is shown illustrating these processes is shown in figure 1-11, while figure 1-12 shows the local context of the tunnel and its external ventilation components. The roadway gradient reaches a maximum of 1:12 at the tunnel's eastern end, as figure 1-13 shows. A tunnel air filtration plant is currently under construction, with completion estimated to occur in late 2009. The plant will extract air from the westbound bore of the tunnel prior to delivering treated air downstream into the same bore, after which it will delivered to the entry portal of the eastbound bore, as shown in figure 1-14. The tunnel is used by approximately 93,000 vehicles per day. About 7% of these are heavy diesel vehicles (HDVs). There is no toll payable for vehicles using the tunnel.

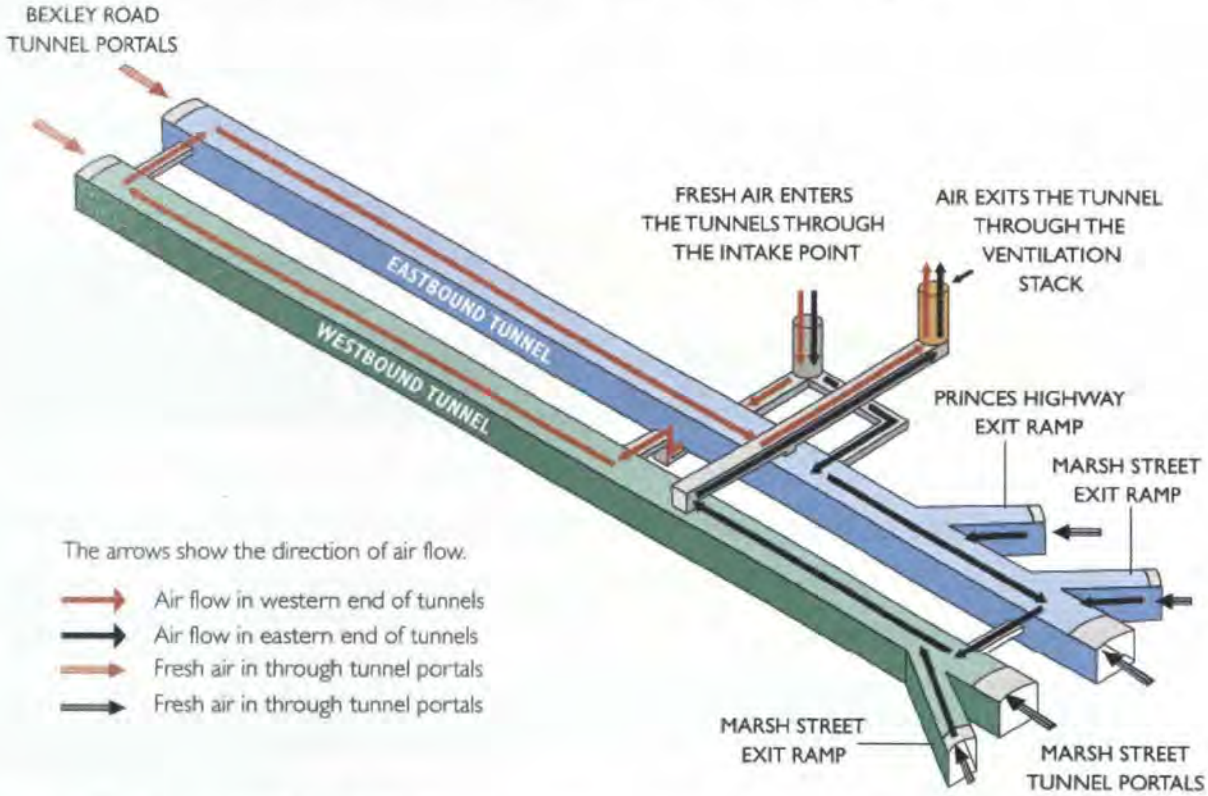



**Figure 1-11.** Schematic of M5 East tunnel ventilation system. Source: NSW RTA (2008a).

The M5 East tunnel presents an ideal case for the study of in-tunnel and in-vehicle UFP determinants and interactions, as it is longest and most heavily trafficked operational road tunnel in Australia (NHMRC, 2008). Also, UFP


**Figure 1-12.** Local context of M5 East tunnel and its associated ventilation infrastructure. Source: NSW RTA (2008a).

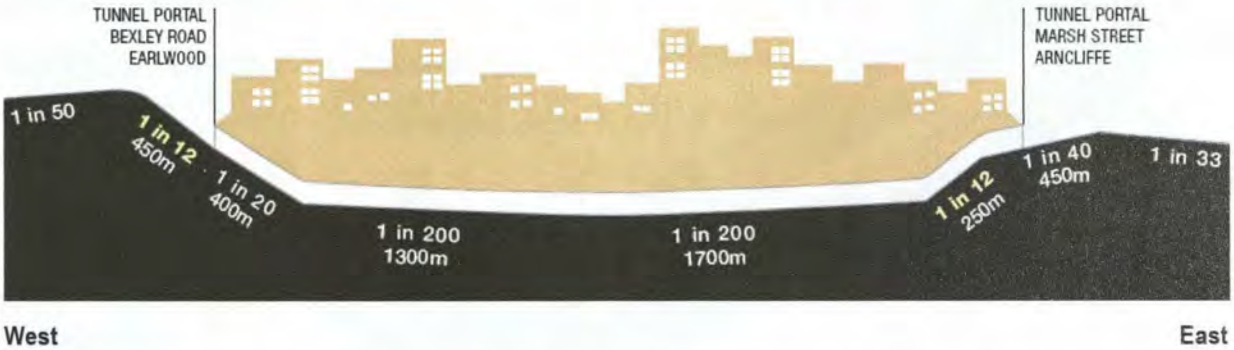



**Figure 1-13.** Roadway grades within the M5 East tunnel. Source: NSW RTA (2008a).

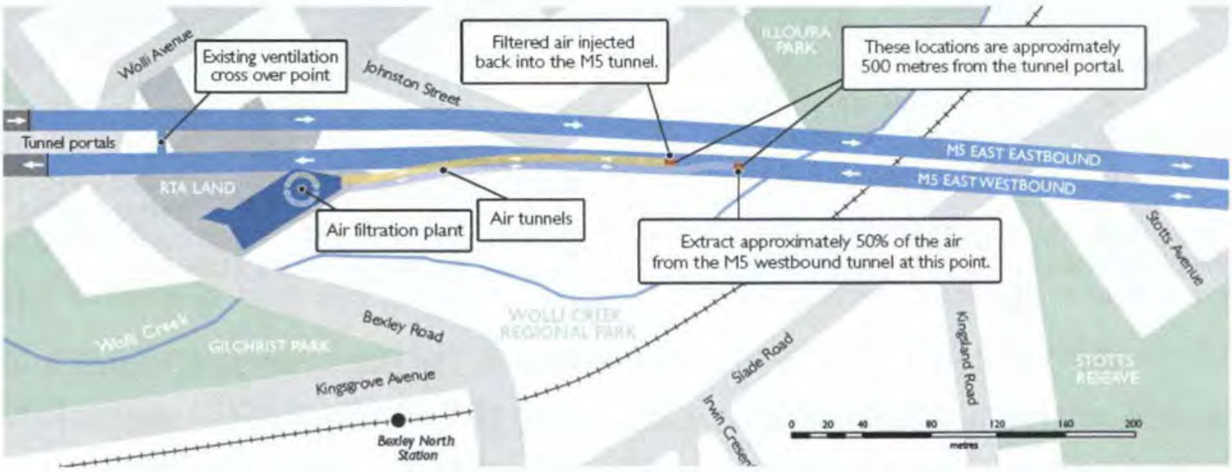



**Figure 1-14.** M5 East tunnel filtration system under construction. Source: NSW RTA (2008a).

concentrations have not been characterised or otherwise investigated in this location. The only published work addressing particulate pollution in the tunnel was focussed on in-vehicle, mass-based measurements of $PM_{2.5}$ (South Eastern Sydney Public Health Unit and the New South Wales Department of Health, 2003). This study reported statistically significant differences between tunnel trip average in-cabin $PM_{2.5}$ mass measured with the vehicle's windows open and that measured with the windows closed (South Eastern Sydney Public Health Unit and the New South Wales Department of Health, 2003). Also, there have been anecdotal and unpublished reports describing negative health effects experienced by tunnel users, particularly following an extended delay (NHMRC, 2008). The issue of air quality both within the tunnel and in the areas proximate to the ventilation exhaust stack has been contentious in the years following its opening, and has received significant mainstream media coverage in Sydney. Such factors, coupled with the aforementioned tunnel characteristics, further highlight the suitability of the M5 East tunnel as the focus of an extensive, integrated assessment of UFPs inside both the tunnel and vehicles using it.

## 1.4 Vehicle Cabin Ventilation

The rate at which a vehicle cabin is ventilated with outdoor (i.e. on-road) air is a key parameter determining the levels of UFPs (and many other pollutants) inside the cabin, whether they originate there or outdoors. Another dominant factor affecting the movement of outdoor pollutants into a vehicle cabin is the efficiency with which they penetrate the vehicle's HVAC system (including filters) and air leakage pathways. Outdoor air ventilation rates are typically expressed in terms of air changes per hour (ACH), whereby a space within which one ACH occurred would experience a volume of air equal to the volume of the space entering over the course of one hour. This is represented conceptually in figure 1-15. If the volume of a space is known, then one can simply multiply ACH by this to obtain a volume flow rate; likewise, if a volume flow rate is known for a space and divided by its volume, then the resulting value is of ACH.

**Previous Work**

ACH have traditionally been assessed in both mechanically and naturally ventilated buildings as a means of assessing the role of ventilation and performance of the building in relation to the concentration of pollutants, acceptability of the thermal environment and perception of odours, amongst other factors (Awbi, 2003). By comparison, ventilation rates inside vehicles have received only modest and inconsistent attention and assessment, which belies their importance as a determinant of in-cabin air quality.

The earliest measurements of ACH inside vehicles were reported in 1975 (Petersen and Sabersky, 1975). In the 34 years since, there have been only a relatively small number of published studies reporting vehicle cabin ventilation rate data (Engelmann et al., 1992; Ott et al., 1992; Fletcher and Saunders, 1994; Ott et al., 1994; Kvisgaard, 1995; Conceição et al., 1997; Park et al., 1998; Rodes

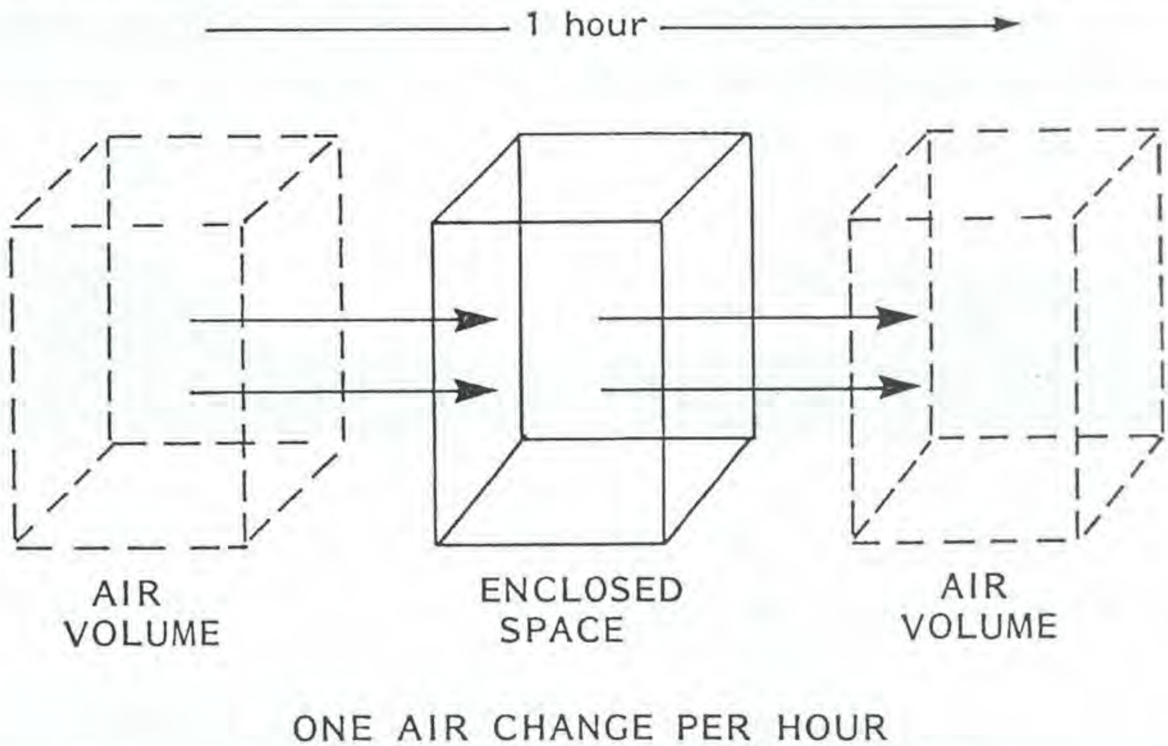



**Figure 1-15.** Conceptual representation of one ACH. Source: Charlesworth (1988).

et al., 1998; Kvisgaard and Pejtersen, 1999; Offermann et al., 2002; Batterman et al., 2006; Nakagawa et al., 2007; Chen and Deng, 2008; Ott et al., 2008; Rim et al., 2008; Zhang et al., 2008). The overwhelming majority of these were performed within the past 15 years, perhaps indicating an increasing awareness of the importance of this parameter in this period. Despite this, very few studies involved the systematic assessment of ventilation rates in terms of replicate measurements incorporating the effects of vehicle type, age, speed and ventilation setting. Measurements of ventilation rate have often been incidental or ancillary to the measurement of a pollutant of interest. While this is acceptable within the context of the aims of the various studies, it has, however, resulted in a paucity of available data on vehicle cabin ventilation, with little knowledge of repeatability or representativeness. Also, some studies dealt exclusively with public transport vehicles (i.e. buses), rather than the private passenger vehicles used by the bulk of the commuting population in cities like Sydney.

Despite the importance of ventilation in controlling pollutant levels, quantitative relationships between ventilation rates and pollutant levels in, and/or ingress into, passenger vehicles are lacking. In one of the earlier studies dealing with measurement of in-cabin ACH, Engelmann et al. (1992) noted the need for future research to focus on measurements of ACH in moving vehicles and the assessment of the mechanisms underpinning the protection offered by the vehicle cabin against particulate and gaseous pollutants. In the 17 years intervening their study and the present, only a modicum of work has been performed to address the research needs outlined by Engelmann et al. (1992). Relevant studies are introduced briefly in chapter 4.

#### Measurement

The measurement of ventilation rate most often involves the deployment of a tracer gas within a space, the concentration of which is then monitored through time and reflects the amount of air unmarked with tracer entering the space during this period. Numerous tracer gases can be employed for this purpose, and should be characterised by very low or absent background concentration, be non-toxic and non-combustible, not react with the environment into which they are released, be readily measurable by instrumentation and not affect the process they are being used to study (Charlesworth, 1988; Sherman, 1990; Awbi, 2003; ASTM, 2006). Commonly used tracers include nitrous oxide ($N_2O$), carbon dioxide ($CO_2$), sulphur heaxafluoride ($SF_6$), freons, perfluorocarbons, ethane ($C_2H_6$) and methane ($CH_4$) (Charlesworth, 1988, Awbi, 2003). Not all of these gases meet the guidelines outlined above, but practicality often dictates that some compromises need be made in regard to tracer gas selection (Sherman, 1990). Accordingly, measures should be implemented to account for the inadequacies of a given gas (i.e. strictly controlling the concentration of potentially explosive gases, such as $C_2H_6$ and $CH_4$, during measurements [Awbi, 2003] or accounting for background concentrations of a gas prior to measurement [Ott, 2007]). In recent years, environmental concerns have resulted in some common tracers, like $SF_6$, being banned in some countries due their substantial global warming potential. In countries where use of such gases is still permitted, the choice of an investigator to use such a gas should include an appreciation of the relevant environmental and ethical issues, whilst noting that in practice, the use of gas for ventilation measurement and atmospheric release of a quantity of tracer (and associated global warming potential) will typically be extraordinarily minute when compared to emissions from industrial and other sources (Kvisgaard and Pejtersen, 1999).

One or more of three possible techniques are generally used in tracer gas analyses of ventilation. These techniques, including concentration-decay, constant emission (or constant injection) and constant-concentration, are all based on a continuity equation, shown here as equation 1-1 (from Charlesworth, 1988), and each has its own advantages and disadvantages.

$$V\frac{dC}{dt}=Q(C_{ext}-C_{(t)})+F \qquad \text{(Equation 1-1)}$$

Where:

$V$ = effective volume of an enclosure ($m^3$)
$Q$ = air flow rate through an enclosure ($m^3\ s^{-1}$)
$C_{ext}$ = concentration of tracer in external air
$C_{(t)}$ = concentration of tracer in internal air at time t
$F$ = production rate of tracer from all internal sources
$t$ = time

The concentration-decay method is the oldest and simplest technique for measuring ventilation rate (Charlesworth, 1988). Using this method, tracer is released into a space until a uniform concentration is achieved. Following this, the release of tracer is ceased, and the decay of its concentration through time is monitored. If an appropriate tracer with zero or negligible concentration in external air is used and no internal source of tracer is present and active, equation 1-1 becomes:

$$V\frac{dC}{dt} = -QC_{(t)} \qquad \text{(Equation 1-2)}$$

Where a constant flow rate (Q) is present, rearranging and integrating equation 1-2 finds:

$$\int_{C_{(0)}}^{C_{(t)}} \frac{dC}{C_{(t)}} = -\frac{Q}{V}\int_{t=0}^{t=t} dt \qquad \text{(Equation 1-3)}$$

Where:
$C_{(0)}$ = tracer concentration at time t=0

Thus:

$$\ln C_{(t)} - \ln C_{(0)} = -\frac{Q}{V}t \qquad \text{(Equation 1-4)}$$

Where:
Q/V = air change rate per unit time (denoted henceforth as N)

And:

$$C_{(t)} = C_{(0)}e^{-\frac{Q}{V}t} \qquad \text{(Equation 1.5)}$$

Given a constant flow rate (Q), an exponential decay of tracer gas will be observed throughout the time period of the measurement and the natural logarithm of tracer concentration plotted against time will be a straight line, the gradient of which is the number of air changes per unit time. This is illustrated in figure 1-16.

The constant emission method relies upon, as its name implies, emission of tracer gas into a space at a constant rate. This can be achieved by using a flow controller or an instrument designed to deliver a precise dose of tracer. If this method is employed in the absence of an external tracer source, the continuity equation (equation 1-1) reduces to (Charlesworth, 1988):

$$V\frac{dC}{dt} = -QC_{(t)} + F \qquad \text{(Equation 1-6)}$$

Solving for $C_{(t)}$ gives:

$$C_{(t)} = \frac{F}{Q} + \left(C_{(0)} - \frac{F}{Q}\right)e^{-\frac{Q}{V}t} \qquad \text{(Equation 1-7)}$$

Assuming constant air flow within the space and the absence of tracer prior to the measurement, equation 1-7 becomes:

$$C_{(t)} = \frac{F}{Q}\left(1 - e^{-Nt}\right) \qquad \text{(Equation 1-8)}$$

Given a constant air change rate (N), a stable concentration of tracer will be reached (as indicated in the section of equation 1-8 in parentheses). Once this occurs, the air flow rate (Q) into the space is given by:

$$Q = \frac{F}{C_{(t)}} \qquad \text{(Equation 1-9)}$$

The two methods outlined above are amenable to measurements performed inside vehicles, with the former suitable when the number of ACH is relatively low and the decay curve can be accurately measured by the instrumentation used (Kvisgaard, 1995), and the latter suitable for higher outdoor air flow conditions (e.g. Kvisgaard and Pejtersen, 1999). The constant-concentration technique is not used in this thesis, and thus, is not described here. Further information on all ventilation measurement techniques can be found in Charlesworth (1988), Sherman (1990), Awbi (2003) and ASTM (2006).

Vehicles present a unique and challenging environment in which to perform ventilation measurements, as the air change rates are often very high when compared to buildings, and the moving nature of the measurement environ-

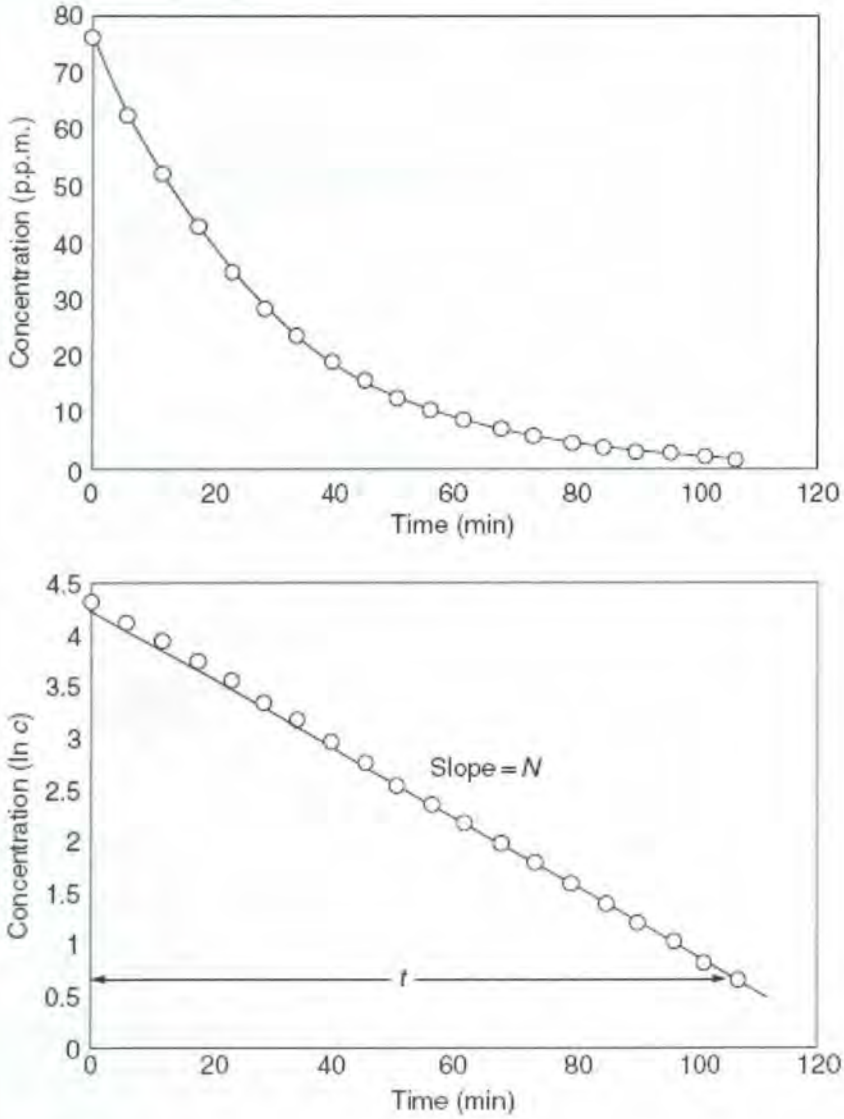



**Figure 1-16**. Decay of tracer gas through time shown in terms of unmodified tracer concentration (top) and the natural logarithm of tracer concentration (bottom). Source: Awbi (2003).

ment can cause instrument errors; through vibration, for example (Ott et al., 2008). Fortunately, the time required to perform ventilation measurements in vehicles is reduced compared to other built environments and automotive HVAC systems are typically quite simple relative to those used in buildings, and rely on mechanical fans and natural infiltration driven by pressure gradients (see figure 1-17) to provide flow to the cabin. A typical modern automotive HVAC system is represented schematically in figure 1-18.

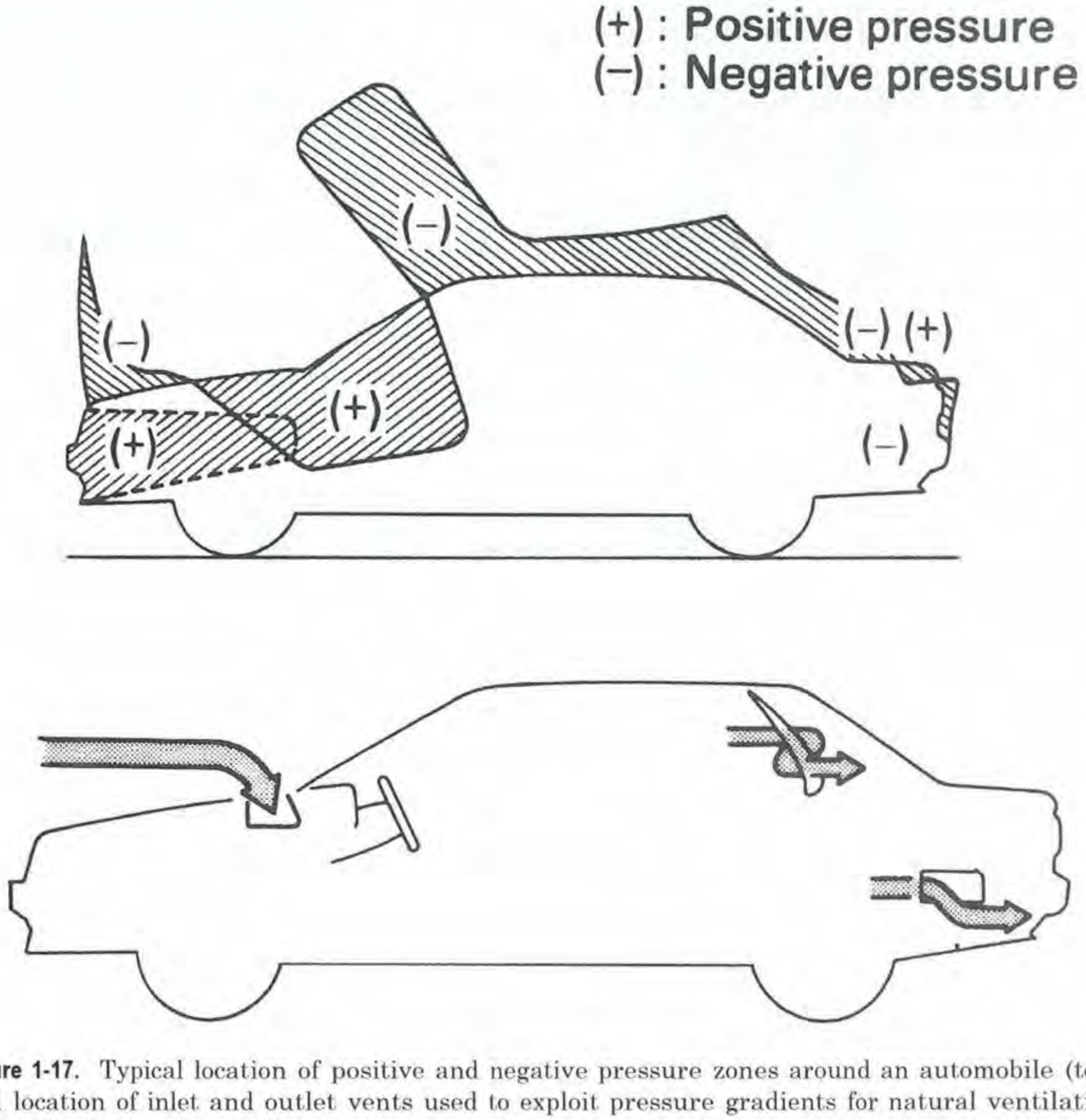
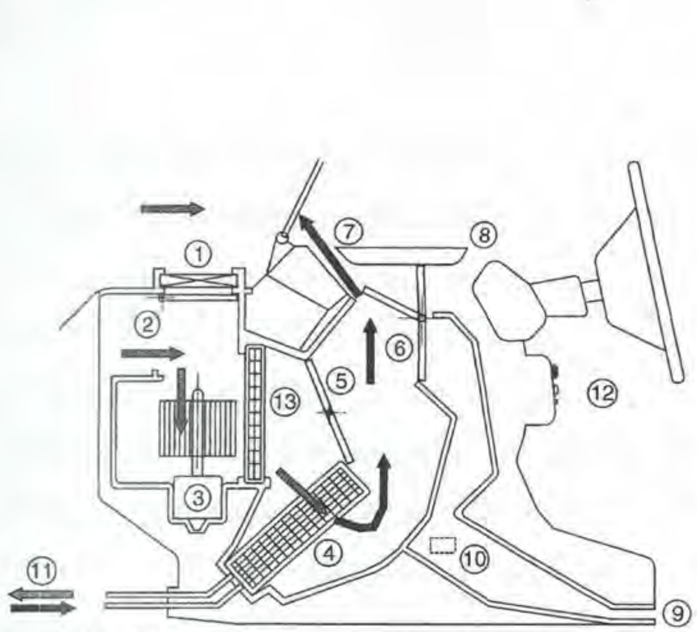



**Figure 1-17.** Typical location of positive and negative pressure zones around an automobile (top) and location of inlet and outlet vents used to exploit pressure gradients for natural ventilation (bottom). Source: Daly (2006).



1. Air filtration
2. Air recirculation door
3. Blower motor and centrifugal fan
4. Heat exchanger
5. Temperature blend door
6. Air distribution door
7. Panel vent (not adjustable)
8. Face/head vent (adjustable)
9. Panel vent – rear passengers' foot well
10. Ducting to passenger foot well
11. Flow of coolant
12. Control panel
13. Evaporator (models with A/C)

**Figure 1-18.** Schematic representation of a modern automotive HVAC system. Source: Daly (2006).

## 1.5 Aims

The preceding sections have outlined the key areas characterised by a paucity of data and knowledge within the context of the vehicle cabin environment. Broadly, these are; the quantification of cabin ventilation rates and analysis of their effect on in-vehicle UFP concentrations, assessment of road tunnel travel on vehicle occupant UFP exposure and examination of the relationship between UFP concentrations encountered on-road and those inside vehicle cabins. Within these, there are various sub-issues that demand systematic enquiry; for example the influence of vehicle age and type on cabin ventilation and infiltration rates. Based on these gaps in knowledge, the specific objectives of this thesis can be expounded. It should be noted that as an umbrella aim encompassing all those described below, this project will seek to place all results within the context established by relevant studies reported in the literature.

### Thesis Objectives

**A)** The primary aim of this project is quantification and evaluation of UFP exposures inside vehicles. The focus of this aim is further refined to address the issue of these exposures during road tunnel travel. While the recognition of the potentially significant role of indoor, rather than outdoor, air pollution in precipitating adverse human health outcomes is not new (Spengler and Sexton, 1983), in general, there is very little information regarding UFPs inside vehicles, and this has been specifically identified as a location requiring assessment (Sioutas et al., 2005). Improved fundamental understanding regarding the nature of in-vehicle UFP exposure carries with it corollary benefits; most importantly, the ability to mitigate these exposures to protect those most susceptible to the undesirable health effects thought to be associated with UFP exposure. As was highlighted previously, the accurate measurement of UFPs requires a judicious choice of instrumentation, and this project aims to incorporate an acute awareness of the value of data veracity, and to ensure appropriate means are in place to ensure collected data are of high quality.

**B)** Investigation of the determinants of on-road UFP concentrations inside the M5 East tunnel. A comprehensive understanding of in-vehicle UFPs necessitates characterisation of the sources responsible for their release into the tunnel environment. Indeed, the importance of knowledge of outdoor air quality issues as a key adjunct to understanding air quality within built environments was identified over 120 years ago (Carnelley et al., 1887). However, time has not diminished the importance of an examination of the synergy between outdoor and indoor air, or as is the case in this study, on-road and in-vehicle air. Within this study, the focus on the M5 East tunnel reflects, as was mentioned previously, its current status as Australia's longest and most heavily-trafficked road tunnel. Notwithstanding this, the investigation of UFP determinants is a topic of external applicability to other roadways, whether tunnelled or open air.

**C)** Systematic investigation of ventilation rates inside passenger vehicles representative of those commonly driven on Australian roads. As noted in an earlier section of this chapter, comprehensive studies involving the assessment and quantification of vehicle cabin ventilation rates are lacking in the literature. The potential benefits of such studies are manifold in terms of improving the understanding of human exposure to pollutants inside vehicles, and are not solely confined to UFPs. The focus on the Australian vehicle fleet is deliberate; however, given the global nature of the motor vehicle market, vehicles commonly encountered on Australian roads are highly likely to be representative of those driven in many other countries. In any assessment of vehicle cabin ventilation, the issue of the shelf-life of results (i.e. for how long the results are likely to be representative of a given vehicle fleet) is worthy of consideration, and this study aims to include older vehicles still commonly driven, in addition to those that will continue to be representative of various vehicle fleets for several years.

**D)** Quantification and assessment of the relationship between on-road and in-vehicle UFP concentrations. An additional aim is the quantification of the role played by cabin ventilation rate in the aforementioned relationship. The identification of relationships between on-road and in-cabin UFPs is relevant not only to the tunnel environment, but to all roadway environments. Tunnels do, however, represent an ideal location in which to perform a field study underpinning the assessments outlined above, as traffic volumes and UFP concentrations are likely to encompass a suitably wide range.

Associated with the objective mentioned above is an associated sub-objective, namely; assessment of the ability of a basic model to predict average in-cabin UFP exposures during travel in the M5 East tunnel. Based on integration of all experimental data proposed above, modelling would provide an end-user (e.g. roadways management authority or tunnel operator) with the facility to predict UFP average exposures for vehicle occupants, and accordingly implement appropriate mitigative measures when required. Modelling would also allow vehicle manufacturers to appreciate the effects of ventilation on intrusion of on-road UFPs into a vehicle cabin. The realisation of practical and usable outcomes from this project is thus of paramount importance, and the examples outlined here are by no means exhaustive.

#### Thesis by Publication Model

This thesis is presented in accordance with Macquarie University's guidelines for a research thesis by publication. As such, all of the chapters in section II have been published in, or submitted to, a peer-reviewed journal during the course of candidature. Where multiple authors contributed to a paper, the specific contribution of the candidate is clearly defined.

Chapter 2 deals with objective A); specifically in relation to assuring the representativeness and veracity of UFP measurements. Chapters 3 and 4 address objectives B) and C), respectively. Based on integration of data presented in the aforementioned chapters, chapter 5 addresses objectives A) and D). The connection between chapters in realising the objectives outlined above, in addition to a final commentary, is the focus of chapter 6.

It should be noted that the chapters in section II exhibit minor variations in terms of formatting and referencing style. This reflects the fact that all of these chapters are faithful reproductions of the various papers as published or submitted that have been typeset similarly here for consistency.

# Section II
# Publications

# Chapter 2

# A Simple and Inexpensive Dilution System for the TSI 3007 Condensation Particle Counter

This chapter describes the development of a basic dilution system capable of substantially increasing the upper particle number concentration measurement threshold of the TSI 3007 CPC. The recognition of the need for such a system stemmed from a pilot study performed in the M5 East tunnel, where it was observed that the CPC was not capable, in its unmodified state, of measuring the high particle concentrations encountered in this location. It was therefore decided that to ensure collected data was as accurate and representative as possible, it would be necessary to dilute air samples prior to measurement by the CPC. To this end, the measures described in this chapter were undertaken.

#### Contribution

The initial pilot study that established the need for a dilution system was performed by me, under the general guidance of Richard de Dear. The nature of the results obtained was discussed with Lidia Morawska, and she raised the possibility of using dilution to ensure data accuracy. Some initial ideas for simple dilution systems were also discussed with her. The specific design of the dilution system described here was performed by Peter Coote and me. The experimental work to calibrate and characterise the response of the CPC with dilution system attached was performed by me during a visit to the International Laboratory for Air Quality and Health at Queensland University of Technology. Data processing and analyses were performed by me. The results were discussed with both Lidia Morawska and Richard de Dear. The draft manuscript was written by me, and I produced the figure presented in this chapter. Following comments and advice from Lidia Morawska and Richard de Dear, the manuscript was submitted in November, 2006 as a technical note to *Atmospheric Environment*. Reviewer comments were received in January, 2007. All post-review amendments to the manuscript and responses to reviewer comments were performed by me with input from Lidia Morawska and Richard de Dear. The revised manuscript was accepted for publication in *Atmospheric Environment* in March, 2007, and published in July, 2007.

# A Simple and Inexpensive Dilution System for the TSI 3007 Condensation Particle Counter

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## Abstract

The aim of this study was to develop a dilution system which would permit the TSI 3007 Condensation Particle Counter (CPC) to operate within its maximum detectable concentration threshold, even when sampling extremely high submicron particle concentrations. The intention of this was to provide a better alternative to coincidence correction factors, which have several limitations; the most significant of which being that they are only applicable to a comparatively low concentration and also that the components of the unit are exposed to concentrations beyond their operating specifications. To achieve the aim, a bifurcation based system was developed and tested repeatedly at concentrations of unleaded petrol combustion particles up to ~$8.5\times10^{6}$ p cm$^{-3}$. The benchmark particle concentration was measured by a TSI 3022A CPC. The results of the tests showed that the nominal dilution ratio based on flow partitioning was applicable up to ~$3.5\times10^{5}$ p cm$^{-3}$, after which particle losses to a capillary tube primarily caused a large increase in apparent dilution. These losses were consistent throughout all tests and allowed the unit to remain below the maximum detection threshold, even under the extreme challenge concentrations encountered. This work represents a useful extension of the operating range of the TSI 3007, without significantly compromising either the quality of data collected or the internal components of the unit.

Keywords: TSI 3007, Condensation Particle Counter, Dilution, Submicron, Particle Measurement

## Introduction

The TSI model 3007 Condensation Particle Counter (CPC) is a hand-held device for measuring the concentration of submicron particulate matter in the air. The unit uses Isopropyl alcohol as an operating fluid, and functions using a regulated continuous flow of ~700 $cm^3\ min^{-1}$. The manufacturer quotes the particle size measurement range of the device as 0.01μm to >1.0μm, with a maximum concentration detection limit of $10^5$ p $cm^{-3}$, and a 50% size detection threshold of 0.01μm (TSI Inc., 2006a). The unit is lightweight and can be powered by AA batteries, which makes it extremely portable relative to many other CPCs. However, this portability means that the unit may easily be taken into a variety of heavily polluted environments where its maximum detectable concentration is exceeded. Once this occurs the output of the unit is unreliable, as the particle concentration is under-estimated due to coincidence error; i.e. more than one particle passes the single particle counting optics at any given time (Hämeri et al., 2002). Once the maximum detectable concentration is exceeded, retrospective coincidence correction can be applied to better estimate the true particle concentration. No on-board coincidence correction is present, unlike some larger CPCs.

Literature searching shows the TSI 3007 to be ubiquitous in many recent air quality projects conducted across challenging and diverse environments, e.g. (Cleary, 2004; Hall et al., 2004; Williamson et al., 2004; Avogbe et al., 2005; Matson, 2005; Vinzents et al., 2005; Westerdahl et al., 2005; Peters et al., 2006; Thomassen et al., 2006; Vlahos et al., 2006). However, despite many of these projects being conducted in locations where the maximum detection limit was exceeded, not all studies addressed this measurement issue. Where steps have been taken, it is often in the form of coincidence correction, although one study made use of diffusion screens to raise the minimum size detection threshold in order to keep sample concentrations below $10^5$ p $cm^{-3}$ (Williamson et al., 2004). Although the data obtained are more reliable following coincidence correction, the components of the unit can be exposed to extremely high particle concentrations, potentially shortening the operational life of consumables such as alcohol wicks and filters, and also soiling other components exposed to the sampled air. Also, the correction factor can only be accurately applied for real concentrations up to ~$4\times10^5$ p cm$^{-3}$ (Hämeri et al., 2002), although less confident corrections can be made up to approximately double this concentration (Westerdahl et al., 2005).

Peters et al. (2006) implemented a dilution system for the 3007 similar to that described in this article, using a filter and small orifice (0.4mm in diameter). Their system filtered all air entering the 3007 with a HEPA cartridge capable of 99.97% capture efficiency for particles ≥ 0.3μm (Whatman Inc., 2006). However, they did not assess the performance of their system at concentrations above $10^5$ p cm$^{-3}$, and therefore assumed a constant dilution ratio at all concentrations. It was not stated whether the challenge aerosol used to determine their system's dilution ratio possessed a similar particle size distribution to the air sampled in their study. Their dilution ratio may have been overestimated if their research samples were taken from air having an increased proportion of ultrafine particles compared to that used during testing of the dilution system, and vice-versa.

Cleary (2004) fabricated a bifurcation-based dilution system for the 3007 using a small tube as a laminar flow device to restrict the flow in the sample line. The flow rate (and therefore dilution ratio) of the sample and filtered bypass line was determined by pressure drop measurement (T. Cleary, personal communication, 2005). The dilution ratio achieved was approximately 20:1. Again, the response of the system at high challenge concentrations was not assessed, and also the test aerosol, ambient room air, was different to the measured aerosols produced by food combustion. The filter type used to clean the bypass flow is unknown.

A project which required a large number of air samples be taken from a heavily-trafficked road tunnel highlighted the 3007's detectable concentration range limitations. A pilot study conducted in September 2004 showed that the maximum threshold was exceeded almost immediately upon entering the tunnel. Rather than expose the unit to high concentrations for prolonged periods, it was decided to develop a simple dilution system. The main considerations for this system were that it was inexpensive, effective at very high concentrations, capable of providing a dilution ratio of approximately 20:1, required little maintenance and was consistent in terms of flow partitioning. It is clear that dilution systems of a similar type have been used previously, however validation is often limited. Increased testing and refinement of simple dilution systems hold promise for improving data quality.

## Methods

As the proposed method of dilution was to bifurcate the instrument flow into bypass and sample lines before combining the two, the flow rate of the TSI 3007 was determined using a bubble flow meter (Gilian Gilibrator 2, Sensidyne Inc., Clearwater, Florida). Repeated testing showed the mean flow rate to be 760.7 $cm^3$ $min^{-1}$, with a standard deviation of 2.1 $cm^3$ $min^{-1}$. A cylindrical glass capillary (outside diameter = 6mm, length = 39mm, and inside diameter = 0.381mm) was placed in a section of Tygon® R-3603 tubing. A bypass line was then attached, and the 3007 was connected to both by the use of a conductive Y-type flow splitter. A new HEPA filter (TSI part no. 1030314) was fitted to the bypass line. This filter is of the same type used to zero-check the instrument before each operation. Monitoring of the flows in both lines showed the capillary reduced the mean sample flow to 36.6 $cm^3$ $min^{-1}$ (standard deviation = 0.3 $cm^3$ $min^{-1}$) while maintaining a mean bypass flow of 723.5 $cm^3$ $min^{-1}$ (standard deviation = 2.2 $cm^3$ $min^{-1}$). Thus, the bypass to sample dilution ratio was 19.8:1. Despite the low flow rate in the sample line, the particle size range being sampled is not thought to be subject to isokinetic sampling issues, due to low particle inertia.

The TSI 3007 with dilution system described above was set up adjacent to a 1$m^3$ smooth-walled test chamber, alongside a TSI 3022A, which is capable of detecting particles down to 0.007μm (50% detection threshold) at concentrations up to $9.99\times10^6$ p $cm^{-3}$, and uses Butanol as an operating fluid (TSI Inc., 2006b). The test chamber was sealed apart from a small air inlet port of approximately 35mm diameter. A short length of tubing was connected to an outlet barb on the chamber and the sample flow from the chamber to both CPCs was split via a conductive splitter approximately 10cm downstream. Both instruments sampled immediately after this bifurcation. Due to the very low sample flow rate of the 3007, a short length of tubing was used to reduce sample residence time to approximately 1.03 s. The 3022A was set to low-flow mode, which was assessed using a bubble flow meter and resulted in a mean of 297.8 $cm^3$ $min^{-1}$ (standard deviation = 1.5 $cm^3$ $min^{-1}$). Before sampling, both units were zero count checked using a HEPA filter, and their time stamps synchronised.

The exhaust port of a 4-stroke unleaded petrol generator (Honda model EU20i) was connected to the inlet port of the test chamber. After ignition, exhaust was allowed to enter the chamber for ~5 s. The instruments then concurrently sampled the chamber air at 1 s intervals as the particle concentration decayed over time. Typically, the CPCs ran for approximately 3 h, at which point the 3007 wick required re-saturation with Isopropyl. Three tests of this nature were conducted.

Data from both instruments were converted to 1 min averages to cancel slight differences in response time. Data collected immediately after petrol smoke plume injection when the maximum detectable concentration of both units had been exceeded were removed. The remaining data were collected under well-mixed conditions in the test chamber.

## Results and Discussion

Fig. 1 shows the side-by-side measurements from both CPCs recorded over three repeat tests. The plot contains 322 one minute average data points. There is a relatively small amount of data collected at concentrations above $2\times10^6$ p $cm^{-3}$, with a maximum concentration of ~$8.5\times10^6$ p $cm^{-3}$. This reflects a relatively rapid decay in particle concentration, despite the low ventilation rate of the chamber which was effectively equal to the total sample flow of the CPCs (334.4 $cm^3$ $min^{-1}$ or 0.02 Air Changes per Hour). However, the small amount of air entering the chamber was ambient lab air characterised by very low particle concentrations (measured range of ~$1.5\times10^3$ p $cm^{-3}$ to ~$3\times10^3$ p $cm^{-3}$ before the test) that would facilitate dilution. In addition to this, surface deposition and coagulation affect particle reduction in the test chamber (Jamriska and

Morawska, 2003). The theoretical response of the 3007 system assuming a constant 19.8:1 dilution ratio is also shown in Fig. 1. For particle concentrations up to ~$4\times10^5$ p $cm^{-3}$, the response of the dilution system is linear. Above this concentration, the response rapidly increases non-linearly. This is very likely due to additional losses at the glass capillary tube in the system. As three tests were conducted, these losses appear to be consistent. There are numerous other factors which may have influenced the non-linear response of the instrument, such as particle volatility and other loss mechanisms in the dilution system. However, given the nature of the study, the theory and dynamics underlying these have not been investigated. It is useful to note that Westerdahl et al. (2005) described the non-linear relationship between concurrent 3007 and 3022A measurements at high particle concentrations. The additional losses observed in this study allow the 3007 to be used within its operating range at much greater particle concentrations than would be possible given a constant 19.8:1 dilution. The difference in minimum detectable size between the two CPCs used is acknowledged as possibly resulting in a slightly overestimated dilution performance, particularly for data collected shortly after smoke plume injection. However, as no particle size distribution information is available, this cannot be confirmed. It should therefore be noted that due caution should be exercised if correcting 3007 readings above $4\times10^4$ p $cm^{-3}$ with the dilution system attached. Even at this level, the system affords a repeatable measurement range increase over the standard threshold of greater than one order of magnitude.

The response of the system described is specific to the challenge pollutant used, standard unleaded petrol smoke produced by 4-stroke spark combustion, which is the focus of many particle characterisation and monitoring projects. This also matched the predominant aerosol source in the location where the 3007 will ultimately be used, namely underground road tunnels. Diesel emissions, also present in the tunnel environment, possess a comparable particle size distribution (Ristovski et al., 1998; Jamriska and Morawska, 2001). Finally, extrapolation of any correction factor for one unit to another of the same model should be undertaken with great caution. Vinzents et al. (2005) found a constant difference of ~9% in counting efficiency between two TSI 3007 CPCs. Wherever possible, unit specific correction factors, be they for coincidence or dilution, should be determined experimentally. Also, it should be considered whether the ambient temperature and humidity in the validation and study environments varies significantly. Suggestions for further development of this work include assessment of the effects of dilution on aerosol samples containing volatile and non-volatile species, which was beyond the scope of this study. Also, the response of the system when sampling particles of varying sizes and sources could be investigated using a Scanning Mobility Particle Sizer (SMPS).

## Conclusions

This study utilised a simple dilution system in order to overcome the limitation of the TSI 3007 CPC in terms of its maximum detectable particle concentration. Sample dilution offers a useful alternative to coincidence correction, which is only viable for particle concentrations up to ~$8\times10^{5}$ p $cm^{-3}$. Another advantage is the greatly reduced exposure of internal components and consumables of the CPC to extreme particle concentrations.

The technique described in this article allowed the unit to operate effectively at particle concentrations up to ~$8.5\times10^{6}$ p $cm^{-3}$. This represents an increase of almost two orders of magnitude over the standard detection threshold, and also the highest concentration likely to be encountered in all but a few very specific sampling environments. When testing any dilution system, use of an appropriate test aerosol is essential to ensure the best possible data quality and reliability. While it is recognised that the methods in this study have some some minor limitations related to the slight difference in size measurement range between the benchmark instrument and test instrument, they afford one of the more reliable and inexpensive solutions available to users who wish to get the most functionality from their CPC, whilst still retaining a good level of data quality. Maintenance requirements are low; cleaning of the capillary tube with clean dry air after each sampling exercise and periodic replacement of the HEPA filter and tubing. The time taken to assemble the system and test flow rates was

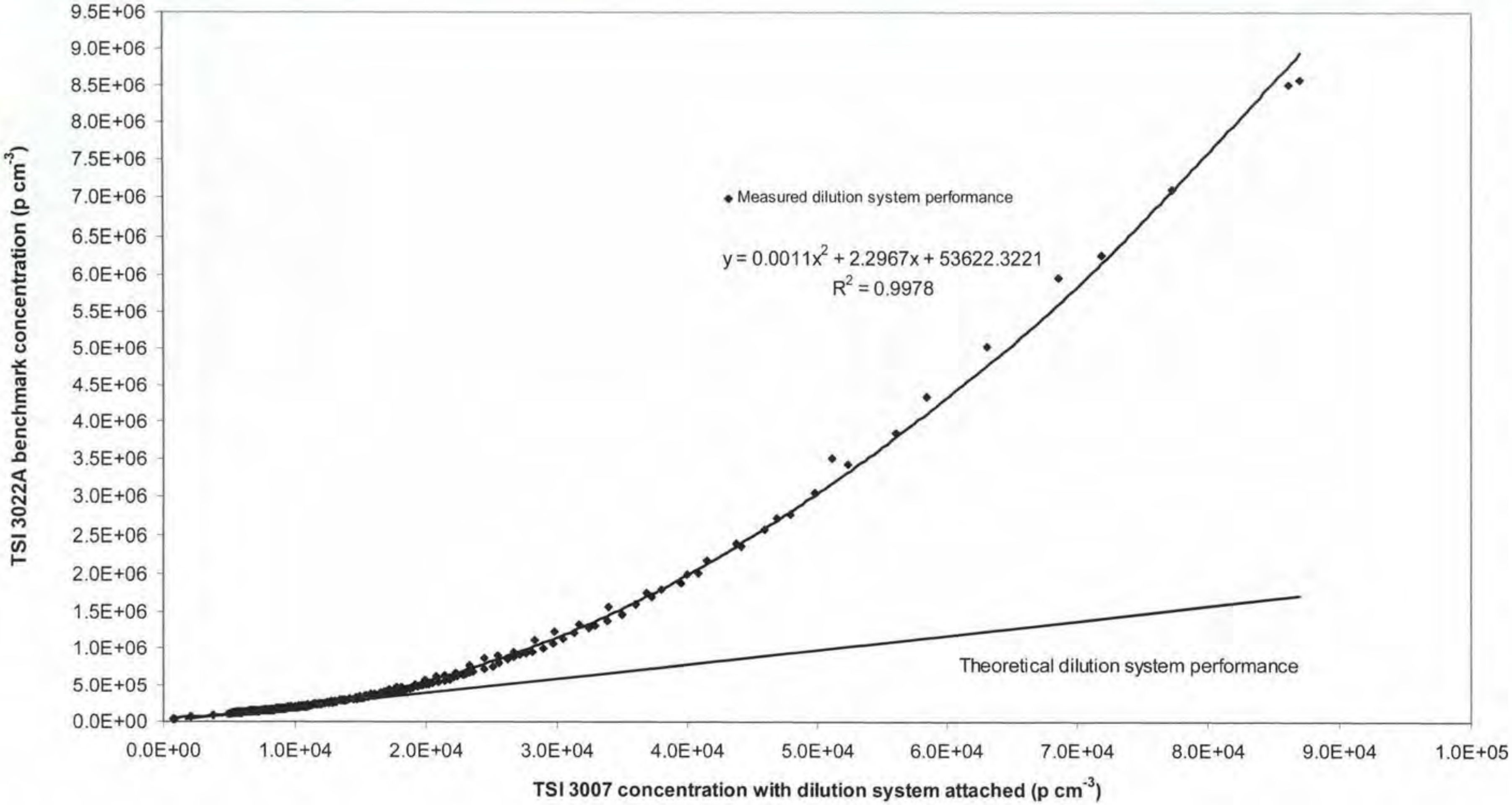


**Fig. 1.** Theoretical and measured performance of the dilution system.

90 minutes, with three replicate chamber validation tests taking 10 hours. The total cost of the modification described was approximately US$100.

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# Chapter 3

# On-road Ultrafine Particle Concentration in the M5 East Road Tunnel, Sydney, Australia

This chapter details the experimental and analytical aspects involved in characterising on-road UFP concentrations in the M5 East road tunnel, in addition to an investigation of their determinants. The TSI 3007 CPC, replete with modifications described in the previous chapter, was deployed on board a mobile sampling platform (moving vehicle). Over three hundred trips through the M5 East tunnel were completed, with the bulk of these occurring in mid-2006. The results presented in this chapter encapsulate the first, as far as I am aware, study of UFPs in the M5 East road tunnel, and underpin the investigation of in-vehicle UFP exposures presented in chapter 5.

## Contribution

The sampling approach and protocols were developed by me in consultation with Richard de Dear and Lidia Morawska. I executed the sampling campaign and was responsible for all data collection in the field and subsequent processing. The analyses described in this paper were performed by me, following consultation with Richard de Dear, Lidia Morawska and Kerrie Mengersen, all of whom also assisted with the interpretation of results. I was responsible for writing the draft manuscript and producing all figures and tables. These were then reviewed and commented on by the three co-authors. The manuscript was submitted to *Atmospheric Environment* in November, 2008. Reviewer comments were received in March, 2009, and with input from all co-authors, I wrote the response to these comments and amended the manuscript. The revised manuscript was accepted for publication in April, 2009, and published in July, 2009.

# On-road ultrafine particle concentration in the M5 East road tunnel, Sydney, Australia

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## Abstract

The human health effects following exposure to ultrafine (<100nm) particles (UFPs) produced by fuel combustion, while not completely understood, are generally regarded as detrimental. Road tunnels have emerged as locations where maximum exposure to these particles may occur for the vehicle occupants using them. This study aimed to quantify and investigate the determinants of UFP concentrations in the 4km twin-bore (eastbound and westbound) M5 East tunnel in Sydney, Australia. Sampling was undertaken using a condensation particle counter (CPC) mounted in a vehicle traversing both tunnel bores at various times of day from May through July, 2006. Supplementary measurements were conducted in February, 2008. Over three hundred transects of the tunnel were performed, and these were distributed evenly between the bores. Additional comparative measurements were conducted on a mixed route comprising major roads and shorter tunnels, all within Sydney. Individual trip average UFP concentrations in the M5 East tunnel bores ranged from $5.53 \times 10^4$ p cm$^{-3}$ to $5.95 \times 10^6$ p cm$^{-3}$. Data were sorted by hour of capture, and hourly median trip average (HMA) UFP concentrations ranged from $7.81 \times 10^4$ p cm$^{-3}$ to $1.73 \times 10^6$ p cm$^{-3}$. Hourly median UFP concentrations measured on the mixed route were between $3.71 \times 10^4$ p cm$^{-3}$ and $1.55 \times 10^5$ p cm$^{-3}$. Hourly heavy diesel vehicle (HDV) traffic volume was a very good determinant of UFP concentration in the eastbound tunnel bore ($R^2 = 0.87$), but much less so in the westbound bore ($R^2 = 0.26$). In both bores, the volume of passenger vehicles (i.e. unleaded gasoline-powered vehicles) was a significantly poorer determinant of particle concentration. When compared with similar studies reported previously, the measurements described here were among the highest recorded concentrations, which further highlights the contribution road tunnels may make to the overall UFP exposure of vehicle occupants.

*Keywords*: On-road, tunnel, ultrafine particles, diesel, measurement

## Introduction

The range and implications of human health effects following exposure to vehicular combustion-generated ultrafine (<100 nm) particles (UFPs) and nanoparticles (<50 nm) remain to be fully elucidated, although they are typically regarded as being deleterious. Whilst UFPs are the dominant constituent in terms of total particle number in many locations, and exposures of varying magnitude are ubiquitous, the roadway and tunnel environments present a location where maximum exposures to UFPs may occur (see Gouriou et al., 2004; Westerdahl et al., 2005; Zhu et al., 2007; Fruin et al., 2008; Morawska et al., 2008). Tunnels are an increasingly necessary infrastructure component in many cities, and tunnel advance in Australia for civil purposes was predicted to be 20 km $y^{-1}$ from 2006 onwards (Day and Robertson, 2004). The health significance of road tunnel exposures has been described by Svartengren et al. (2000) and Larsson et al. (2007), who reported undesirable respiratory effects following road tunnel air exposure in asthmatic and healthy test subjects, respectively. Additionally, Mills et al. (2007) described negative cardiovascular implications that could provide a mechanism for acute myocardial infarction in subjects with existing heart complaints following exposure to UFP concentrations typical of those encountered in tunnels.

Geller et al. (2005) reported elemental and organic carbon as the major constituent species of UFPs emitted in tunnels featuring mixed gasoline and diesel traffic. The same study reported that approximately 80% of particle number was less than 40nm in diameter, with a peak at 15-20nm. The size distribution of particles measured at the exit of a tunnel with diesel bus traffic only was reported by Jamriska et al. (2004) to peak at 20-40nm, with a minor secondary peak of accumulation mode particles at 100nm.

A number of studies focussed on vehicle-based quantification of on-road UFP concentration, amongst other pollutants, have appeared in the literature in recent years (Gouriou et al., 2004; Kittelson et al., 2004a,b; Pirjola et al., 2004; Weijers et al., 2004; Westerdahl et al., 2005; Zhu et al., 2007), as equipment suitable for this challenging measurement environment has become more readily available. This has led to the development of some advanced mobile laboratories aimed at comprehensive assessment of on-road pollution. Some well-documented examples have been described by Bukowiecki et al. (2002), Kittelson et al. (2004a), Pirjola et al. (2004) and Westerdahl et al. (2005). The mobile laboratory approach has been recently extended to incorporate an exposure enclosure inside a van (Zhu et al., 2008), which affords the ability to conduct on-road investigations of human subject health responses to concentrations of UFPs and other pollutants representative of those encountered by many vehicle occupants.

Given the emerging significance of road tunnel UFP exposure, this study aimed to quantify on-road particle concentration in the M5 East road tunnel located in Sydney, Australia, and relate these measurements to traffic volume and fleet composition. The tunnel in question had been studied previously with a focus on in-vehicle concentrations of gaseous pollutants and $PM_{2.5}$ and in-tunnel concentrations of gaseous pollutants (South Eastern Sydney Public Health Unit and NSW Department of Health, 2003). The primary goal of this work was to improve knowledge regarding the role of vehicle fleet volume and composition in determining on-road UFP pollution in the M5 East tunnel, in order to better understand potential exposures of its users, and fortify the prior work described above. We sought to develop a sampling methodology to accomplish this, and supplement the existing data on vehicle-based UFP sampling in tunnels, which is somewhat limited in comparison to vehicle-based measurements on above-ground roadways and fixed-site measurement studies conducted in tunnels (such as Kirchstetter et al., 1999; Abu-Allaban et al., 2002; Jamriska et al., 2004; Kristensson et al., 2004; Geller et al., 2005, Imhof et al., 2006 and Lechowicz et al., 2008). Vehicle-based on-road sampling has the ability to collect data that represent UFP concentrations challenging the protection mechanisms (air tightness, filtration capability, penetration characteristics) afforded by vehicles to their occupants. This is not as easily accomplished during static sampling, which is more suited to development of emission factors. Finally, we aimed to place the measurements into context via comparison with measurements reported in analogous studies, in addition to those reported for a range of other environments, as well measurements performed on a local (Sydney) mixed roads route.

## Methods

### Sampling Location

To fulfil the primary and additional aims of this study, two measurement roadway environments were selected; the M5 East tunnel in Sydney, Australia and a mixed route that commenced close to the tunnel site. The M5 East road tunnel has been in operation since December 2001, and consists of two unidirectional bores, each comprising two lanes. Maximum permitted vehicle speed during normal conditions is 80 km $h^{-1}$. The tunnel is used by approximately 93 000 vehicles per day, about 7% of which are heavy diesel vehicles (HDVs). The tunnel is 4 km long, and reaches a maximum gradient of 1:12 at its eastern end, proximate to the eastbound bore exit and westbound bore entry (NSW RTA, 2008a). Longitudinal ventilation is provided by 131 jet fans (NSW RTA, 2008a). Air is extracted approximately 1/3 of the way along the westbound bore and 2/3 of the way along the eastbound bore, and exhausted through a nearby stack. Fresh air is delivered into each bore slightly downstream of the extraction point. At the exit portal of each bore, air is drawn via cross-shafts and diluted, before being delivered to the entry portal of the other bore (Child and Associates, 2004; NSW RTA, 2008a). Vehicles using the tunnel are not subject to a toll.

Although the construction of a tunnel air filtration plant is currently underway, no such system was present during our measurements conducted from May to July, 2006. Supplemental measurements were performed in February, 2008. During the main measurement campaign, 119 jet fans were present in the tunnel, whilst during the supplemental campaign, an additional 12 fans had been installed (NSW RTA, 2008a).

Additional measurements were conducted on a mixed route, the majority of which consisted of Southern Cross Drive, which was characterised by an average annual daily traffic volume of approximately 128 000 in 2005 (NSW RTA, 2008b). A combination of major roads, toll roads and shorter tunnels were present on the route, including the 2.3 km twin bore Sydney Harbour Tunnel, which carried 86 800 vehicles per day in 2005 (NSW RTA, 2008c).

**Measurements**

A TSI 3007 condensation particle counter (CPC) mounted in a research vehicle was used to measure particle concentration. The manufacturer-stated particle size measurement range of the unit is 10nm (50% detection threshold) to >1000nm, with ± 20% accuracy and a response time of <9 s for 95% response (TSI, 2004). The CPC was zero count checked prior to each use, and its sampling interval set to 1 s. Pilot tests conducted in the tunnel showed the maximum detectable concentration of the unit, $1.0 \times 10^5$ particles per $cm^{-3}$ (p $cm^{-3}$), was often exceeded shortly after tunnel entry. A simple dilution system was developed which allowed the unit to function at concentrations of combustion-derived particles up to $8.5 \times 10^6$ p $cm^{-3}$ (described in detail in Knibbs et al., 2007). The CPC was placed on a raised stand designed to reduce vibration and the tilt effects associated with traversing a roadway incline or decline, and mounted on the passenger seat of a research vehicle. All research vehicles were powered by unleaded petrol and were in very good mechanical condition. None exhibited any signs of exhaust leakage or other sources of self-pollution that could potentially bias the measurements.

To minimise the length of tubing required and associated sample residence time, air samples were taken at the junction of the windscreen base and the rear edge of the hood, which was about 1 to 1.5m above road height depending on the research vehicle used. Samples were transported to the CPC via Tygon® R-3603 tubing. Tubing length from the sample point to the CPC inlet ranged from 0.75 to 1.1m for the research vehicles. Tubing was passed through a small gap in the front passenger side window, which was then sealed. No attempt was made to establish isokinetic sampling conditions despite the moving sampling platform, due to the small size of particles being sampled (Morawska and Salthammer, 2003). An automated Y-type pinch valve fitted with conductive tubing was included in the sampling train, to permit alternate measurement of outdoor and in-vehicle concentrations. However, the work described here is focussed on the outdoor measurements only. As such, the measurements presented are based on transient snapshot measurements of particle concentration inside the tunnel. The valve operation interval was 20 or 25 s, depending on the length of tubing required. The final 10 s of data in each sample block were used for analyses, to account for sample clearance time which ranged from 9-13 s due to the flow partitioning required to achieve sufficient dilution. An overall correction factor for particle loss to the entire sampling train (tubing, connectors, pinch valve and dilution system) was determined experimentally in a test chamber, using 4 stroke spark combustion of standard unleaded gasoline from a warm engine as the pollutant source. A TSI 3022A CPC with a maximum concentration detection limit of $10^7$ p $cm^{-3}$ and minimum size detection threshold of 7nm was used as the reference instrument during these tests, and the maximum concentration detected by the TSI 3007 CPC was approximately $8.9 \times 10^6$ p $cm^{-3}$. Although the specifications of the TSI 3007 CPC define it primarily as an instrument for the measurement of submicrometer particles (<1000nm), due to fuel combustion representing the major pollution source in the study location, the use of UFPs as a descriptor for the measurements described here seems warranted.

Temperature and relative humidity outside and inside of the vehicles was measured by a set of calibrated dry (temp) and aspirated wet (RH) thermistors, or in later tests, Vaisala HMP45A probes, all of which were sited in such that they were not exposed to direct sunlight. Comments made by the investigator during data collection were recorded to enable retrospective production of field notes. Measurements were performed at varying times on from 02:00 to 00:00 h to include a range of traffic conditions. Measurements were conducted primarily on weekdays, although a small number were taken on weekends. Sampling trips were distributed evenly between the two tunnel bores, and 306 tunnel trips were completed in total. Forty one of these trips were completed during the supplemental measurement campaign, with the remainder performed during the main campaign. Each sampling exercise typically comprised 5 transects of each bore. Additional measurements conducted on the mixed route were generally performed after the conclusion of tunnel sampling. Sampling exercises were not conducted during rain.

Traffic data were obtained from the New South Wales Roads and Traffic Authority. Due to the unavailability of traffic data coinciding with the main sampling period, traffic data from 2007 comprising hourly volume measurements collected over the year were used as a surrogate. Annual average daily traffic volume at the main study location exhibits minimal variation between years (New South Wales Roads and Traffic Authority, 2008; personal communication). Hourly traffic averages were computed for each tunnel bore based on 2007 volume data. As volume data were not available for all vehicle classes, heavy vehicle volume (which included a range of vehicles from two axle light rigid trucks to 7+ axle trucks) was determined by subtracting passenger vehicle volume from overall vehicle volume. As the resultant measure was not tantamount to diesel-powered vehicles, the data were modified based on the 2006 Australian Motor Vehicle Census (ABS, 2006), which reported that 73.9% of non-freight carrying trucks, 76.7% of buses, 84.3% of light rigid trucks, 89.3% of heavy rigid trucks and 97.7% of articulated trucks registered in Australia were diesel-powered. Accordingly, hourly heavy vehicle counts were multiplied by the average of these values (84.4%) to generate an estimated hourly heavy diesel vehicle (HDV) count. No changes were made to passenger vehicle counts, as approximately 95% of the Australian passenger vehicle fleet operated on unleaded or supplemented unleaded (lead replacement) fuel in 2006 (ABS, 2006).

### Analyses

Individual trip average particle concentration measurements were split into 24 subsets, depending on hour of tunnel entry. Data collected during both the primary and supplementary measurement campaigns were combined. Temperature differences between the two campaigns for a given hour of day were relatively small (see Section 3.1). Data collected on the mixed road and tunnel route were processed similarly, although trip averages were not calculated. Rather, the data captured on the mixed route were grouped according to the hour during which they were collected and used to produce descriptive statistics. As the in-tunnel data were skewed, the hourly median trip average (HMA) UFP concentration was selected as the variable of choice. Simple linear regression was applied to assess the relationship between the hourly volume of passenger and HDVs and HMA particle concentration in each bore. Regression was also used to examine the influence of average hourly vehicle speed on HMA particle concentration in each bore. Following regression analyses, the distributions of residuals were assessed for approximate normality and homogeneity of variance. Student's t-tests assuming unequal variance were used throughout the analyses, appealing to the Central Limit Theorem for robustness of this procedure. In all cases, the 5% level was used as an indicator of statistical significance. Each pair of regression slopes were assessed for significant differences. Data for each of the four scenarios (eastbound HDV vs. westbound HDV, eastbound passenger vehicles vs. westbound passenger vehicles, eastbound HDV vs. eastbound passenger vehicles, westbound HDV vs. westbound passenger vehicles) were then grouped to assess if one combined slope would better estimate HMA particle concentration compared to separate slopes. To facilitate valid comparisons, HMA particle concentrations for each bore and hourly median measures from the mixed roads route were log-transformed, resulting in close to linear normal scores plots. F-tests for variance were then employed prior to the application of an appropriate student's t-test.

## Results and Discussion

### General

Of the 306 trips through the tunnel, successful data capture was achieved for 301 (98.4%) of these. This encompassed 1204 km and 21.25 h of tunnel travel. Trips were distributed evenly between the eastbound ($n$ = 150) and westbound ($n$ = 151) bores in terms of number, however, 8.5 h was spent in the eastbound bore and 12.75 h was spent in the westbound bore. The longest trip time recorded during sampling was 26m 23s, which occurred in the westbound bore during heavy traffic following a vehicle breakdown. The average trip duration when travelling through the eastbound bore was 3m 22s (average speed = 71 km $h^{-1}$), while the equivalent for westbound travel was 5m 7s (average speed = 47 km $h^{-1}$). Data collection on the mixed roads route comprised approximately 5.5 hours of travel.

Measurements indicated that the temperature inside the tunnel during the main sampling campaign (Australian winter) ranged from approximately

13°C, during late night and early morning measurements, to 22°C during daytime measurements. Relative humidity varied between 35 and 82%. The supplemental measurement exercises were performed in the Australian summer, when in-tunnel temperatures ranged from 19 to 29°C, with a humidity range similar to that recorded during the main sampling period. Outside temperature on the mixed road route was generally 2-4°C lower than the in-tunnel temperature for a given season. In-vehicle temperatures were generally not substantially different from outdoor temperature. Dilution air was therefore supplied at temperatures similar to that of sample air, and the effect of thermophoretic particle loss or condensation in the sampling line as it entered the vehicle cabin is unlikely to have influenced the results. Eighty seven one second data points exceeded the upper limit of the empirical particle loss correction factor (approximately $8.9 \times 10^6$ p $cm^{-3}$). These data points were not discarded, although their value was set to the system's measurement limit.

#### UFP Concentration in the Tunnel Bores

Figs. 1 and 2 provide box plots based on trip average particle concentration for the eastbound and westbound tunnel bores, respectively. The peak trip average recorded in the eastbound bore of $5.05 \times 10^6$ p $cm^{-3}$ (s.d. = $3.77 \times 10^6$ p $cm^{-3}$) occurred between 15:00 and 16:00, while the minimum trip average of $5.53 \times 10^4$ p $cm^{-3}$ (s.d. = $2.10 \times 10^4$ p $cm^{-3}$) was measured between 22:00 and 23:00. The respective trip average maximum and minimum measures in the westbound bore were $5.95 \times 10^6$ p $cm^{-3}$ (s.d. = $2.71 \times 10^6$ p $cm^{-3}$) between 13:00 and 14:00, and $6.59 \times 10^4$ p $cm^{-3}$ (s.d = $1.43 \times 10^4$ p $cm^{-3}$) between 21:00 and 22:00. Evidently, the range of trip average measures in both bores comprised two orders of magnitude. HMA particle concentration values in the eastbound bore ranged from $9.04 \times 10^4$ p $cm^{-3}$ between 02:00 and 03:00, to $9.06 \times 10^5$ p $cm^{-3}$ between 08:00 and 09:00. In the westbound bore HMA concentration ranged from $7.81 \times 10^4$ p $cm^{-3}$ between 02:00 and 03:00, to $1.73 \times 10^6$ p $cm^{-3}$ between 09:00 and 10:00. Average HMA concentration in the eastbound bore was $3.43 \times 10^5$ p $cm^{-3}$, whilst the corresponding figure in the westbound bore was $3.98 \times 10^5$ p $cm^{-3}$.

Results of the various statistical tests applied to the data are summarised in table 1. In three of four cases, the positive gradient of the linear regression model fitted to the relationship between hourly vehicle volume and HMA particle concentration was significantly different from zero. The exception to this was the relationship between passenger vehicle volume and HMA particle concentration in the westbound bore, indicating that passenger vehicles were not a significant determinant of UFP particle concentration in this bore. HDV volume in the westbound bore exhibited a significantly different influence on HMA particle concentration compared to passenger vehicle volume ($p = 0.0238$). However, although significant ($p = 0.0256$), the strength of HDV influence of HMA particle concentration was not as substantial as might be expected ($R^2 = 0.26$). Conversely, HDV volume in the eastbound bore was a highly significant ($p < 0.0001$) determinant of HMA particle concentration ($R^2 = 0.87$), although no significant difference existed between bores in the influence of HDVs ($p = 0.7919$). Similar to the westbound bore, passenger vehicles in the eastbound bore also exhibited a significantly weaker influence on HMA particle concentration compared to HDVs ($p < 0.0001$). Comparison across both bores indicated no significant difference in the influence of passenger vehicle volume on HMA particle concentration ($p = 0.9312$). No significant difference existed between bores in the log-transformed HMA particle concentration ($p = 0.7620$). As shown in table 1, combining data from both bores for a given vehicle type, and combining both vehicle types for a given bore both resulted in reduced predictive ability compared to a single predictor in most cases, and accordingly justified the use of the latter.

Whilst the influence of a given vehicle class on HMA concentration was not significantly different between the two bores, the influence of HDVs was significantly greater than that of passenger vehicles. Despite the relatively low number of HDVs, they represent a substantial source of UFPs. This is in accord with the findings of several previous tunnel studies (Kirchstetter et al., 1999; Abu-Allaban et al., 2002; Geller et al., 2005), and also a street canyon study conducted by Jones and Harrison (2006). Fruin et al. (2008) reported a strong association ($R^2 = 0.84$) between diesel truck count and on-road UFP concentration

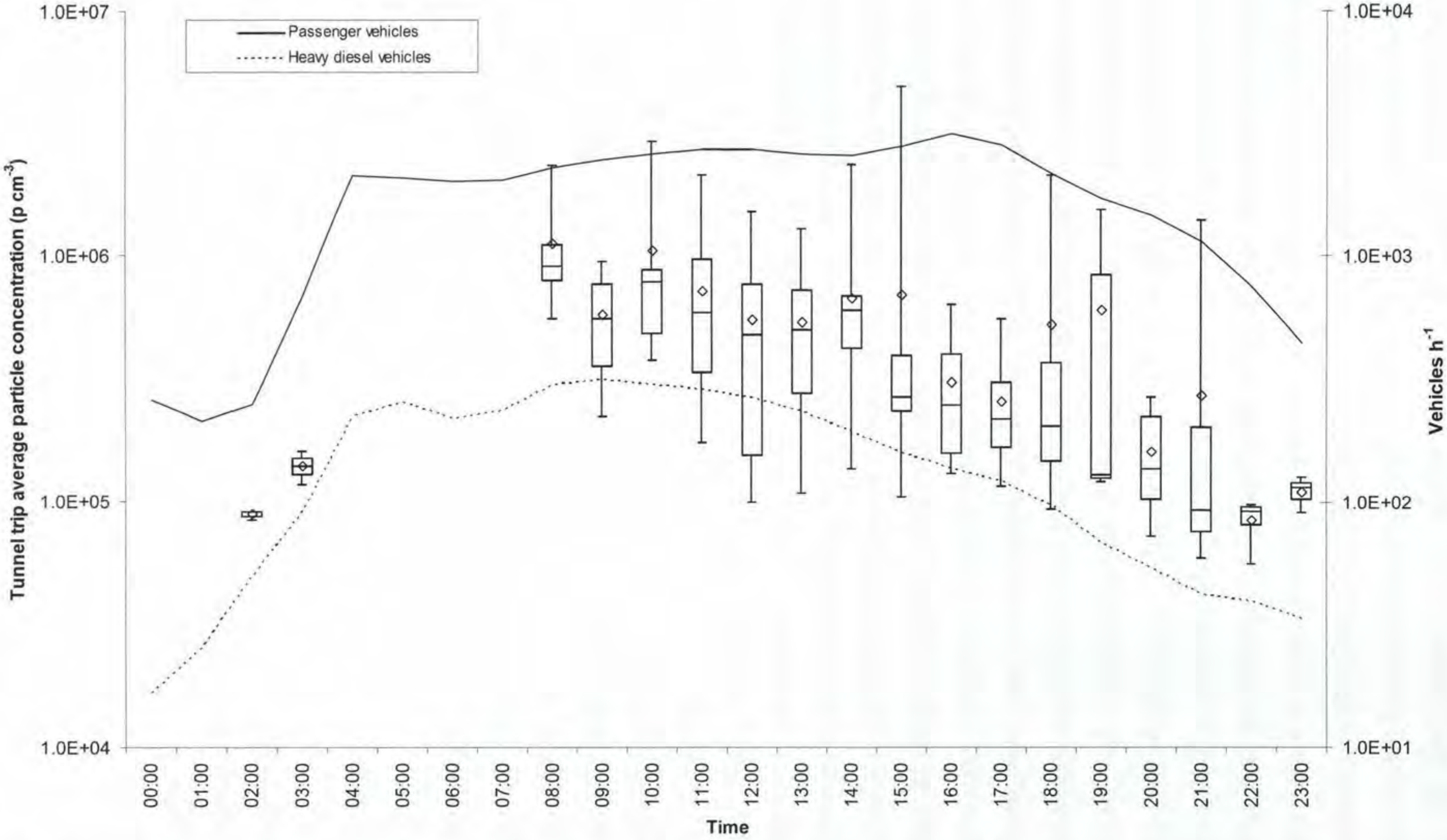


Fig.

**Fig. 1**. Trip average on-road UFP concentration by hour of tunnel entry and hourly passenger and heavy diesel traffic volume in the eastbound bore, based on 150 individual trips. Box-and-whisker plots indicate median (thick horizontal line), average (diamond), first and third quartile (bottom and top edge of box, respectively), minimum and maximum (lower and upper extent of whiskers, respectively) trip average UFP concentration.

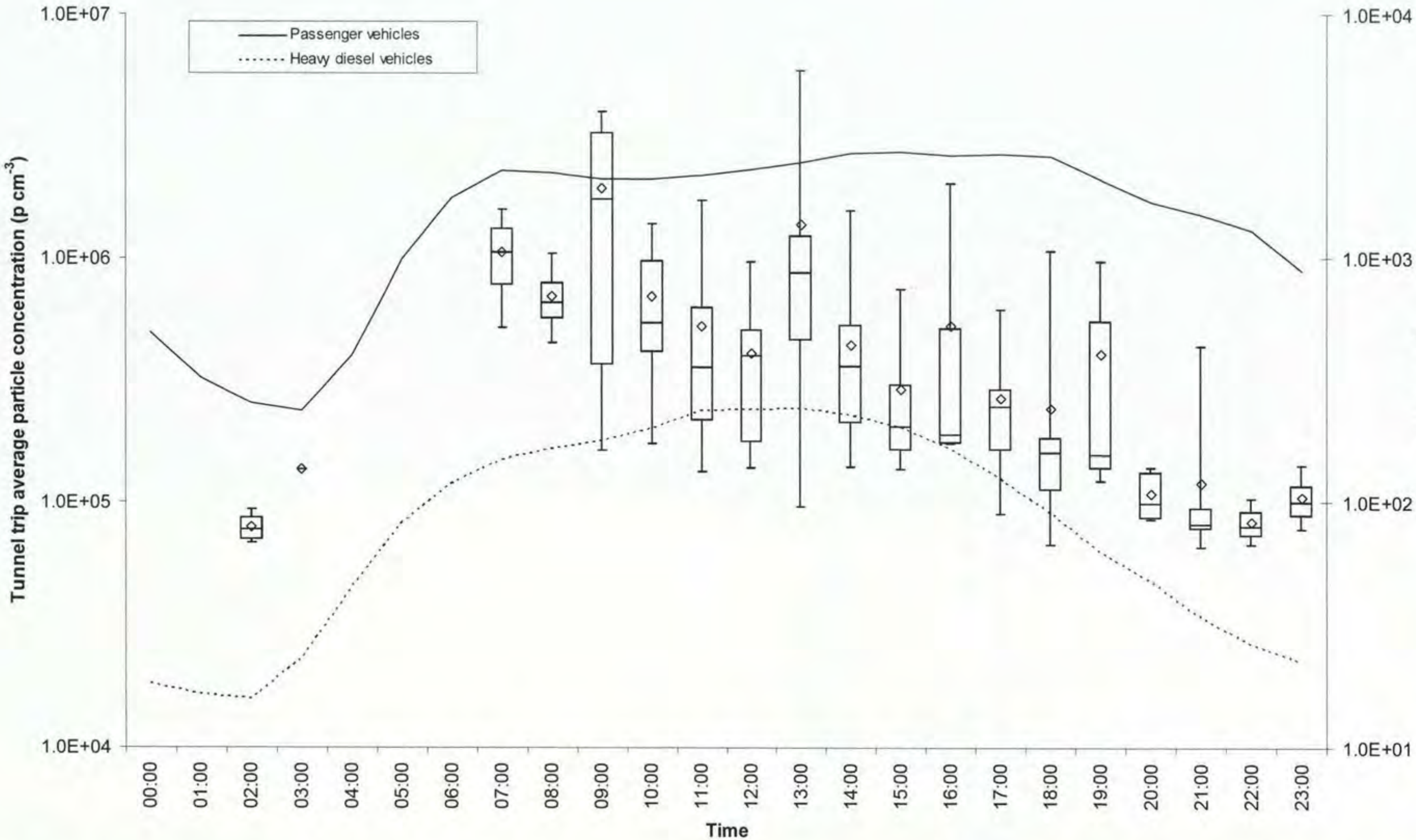


**Fig. 2**. Trip average on-road UFP concentration by hour of tunnel entry and hourly passenger and heavy diesel traffic volume in the westbound bore, based on 151 individual trips. Box-and-whisker plots indicate median (thick horizontal line), average (diamond), first and third quartile (bottom and top edge of box, respectively), minimum and maximum (lower and upper extent of whiskers, respectively) trip average UFP concentration.

**Table 1** – Results summary of statistical procedures. [a] Based on HMA particle concentration. [b] Based on log-transformed data

| Analysis[a] | Slope | SE (slope) | *n* | $R^2$ | t-value | p-value |
|---|---|---|---|---|---|---|
| WB HDV traffic vs. particle conc. | 2548.7 | 1042.0 | 19 | 0.26 | - | 0.0256 |
| WB passenger vehicle traffic vs. particle conc. | 175.7 | 128.2 | 19 | 0.10 | - | 0.1883 |
| EB HDV traffic vs. particle conc. | 2267.7 | 220.6 | 18 | 0.87 | - | 0.0000 |
| EB passenger vehicle traffic vs. particle conc. | 163.7 | 52.9 | 18 | 0.37 | - | 0.0069 |
| WB HDV and passenger traffic vs. particle conc. | 53.2 | 65.9 | 38 | 0.02 | - | 0.4246 |
| EB HDV and passenger traffic vs. particle conc. | 65.1 | 36.2 | 36 | 0.09 | - | 0.0807 |
| WB and EB HDV traffic vs. particle conc. | 2292.3 | 484.2 | 37 | 0.39 | - | 0.0000 |
| WB and EB passenger traffic vs. particle conc. | 166.6 | 63.2 | 37 | 0.17 | - | 0.0124 |
| WB vs. EB HDV influence | - | - | - | - | 0.26 | 0.7919 |
| WB vs. EB passenger vehicle influence | - | - | - | - | 0.09 | 0.9312 |
| WB HDV vs. WB passenger vehicle influence | - | - | - | - | 2.26 | 0.0238 |
| EB HDV vs. EB passenger vehicle influence | - | - | - | - | 9.27 | 0.0000 |
| WB particle conc. vs. EB particle conc. [b] | - | - | - | - | 0.31 | 0.7620 |
| WB particle conc. vs. Mixed route particle conc. [b] | - | - | - | - | -3.80 | 0.0022 |
| EB particle conc. vs. Mixed route particle conc. [b] | - | - | - | - | -4.72 | 0.0002 |
| WB speed vs. particle conc. | 4572.4 | 6406.4 | 19 | 0.03 | - | 0.4851 |
| EB speed vs. particle conc. | -38562.9 | 7966.8 | 18 | 0.59 | - | 0.0002 |
| WB vs. EB speed influence | - | - | - | - | 4.22 | 0.0000 |

on Los Angeles freeways. Our results for a similar analysis of the eastbound bore ($R^2$ = 0.87) are in close agreement with the aforementioned study. Fruin et al. (2008) also noted the generally poor ability of overall traffic volume ($r$ = 0.13), which is largely comprised of gasoline-powered vehicles, to predict on-road UFP concentration. The relative weakness of passenger vehicles as a determinant of in-tunnel HMA UFP concentration in this study ($R^2$ = 0.37 and 0.10 for eastbound and westbound bores, respectively) generally support this finding. Based on measurements at 2 roadside locations in Basel, Switzerland, Junker et al. (2000) noted significant correlations between HDV number and UFP concentration of $r$ = 0.86 and $r$ = 0.67, but did not observe such correlation between light duty vehicle (LDV) number and UFP concentration ($r$ = 0.59 and $r$ = 0.43). Wang et al. (2008) reported correlations ($R^2$ = 0.38 and 0.63) between HDV count and UFP concentration for a busy road intersection in Corpus Christi, Texas. Wang et al. (2008) also recorded comparatively poor associations between total traffic count and UFP concentration ($R^2$ = 0.01 and 0.19) at their study site. Despite differences in measurement location, experimental equipment and/or approach between our study and those described above, the general findings of all studies are in concert with respect to the importance of HDV volume, relative to LDV volume or total traffic volume, as a determinant of on or near-road UFP concentration.

Average hourly vehicle speed through the eastbound bore was a moderate predictor of HMA particle concentration ($R^2$ = 0.57), although particle concentration decreased with increasing vehicle speed. In the westbound bore, average vehicle speed was an insignificant predictor of median average particle concentration ($R^2$ = 0.03). There was a highly significant difference in the predictive strength of vehicle speed between bores ($p < 0.0001$) Kittelson et al. (2004a) reported increased particle concentrations and a reduction in midpoint particle diameter with increasing vehicle speed, whilst also noting the applicability of this relationship to spark ignition vehicles more so than diesel vehicles. Geller et al. (2005) reported a positive correlation ($R^2$ = 0.53 prior to normalisation of vehicle speed; $R^2$ = 0.69 following normalisation) between vehicle speed and particle concentration in a bore of the Caldecott Tunnel in

Berkeley, California. Morawska et al. (2005) found particle number emission factors increased with vehicle speed, as did Kristensson et al. (2004), based on a tunnel study in Stockholm, Sweden. In the above studies, the sampling platform was either static (Kristensson et al. 2004; Geller et al., 2005; Morawska et al, 2005) or direct sampling of exhaust plumes was deliberately avoided (Kittelson et al., 2004a). Neither was the case in this study, and given the combination of a mobile sampling platform and frequent proximity to HDV exhaust plumes in the enclosed tunnel during periods of heavy traffic, the negative relationship we observed in the eastbound bore seems plausible. The effect of higher speed vehicle travel on tunnel ventilation rates and subsequent reductions in UFP concentration has been raised in the literature, although it is not supported by experimental data (Gidhagen et al., 2003; Geller et al., 2005). It is worth noting that due to the factors described above, our measurement approach was not suited to investigation of the influence of vehicle speed on UFP emission factors, nor was it a goal of this study.

We made no attempt to avoid sampling the exhaust plume of vehicles in front of our mobile sampling platform, however, apart from one exercise performed outside of the M5 East tunnel described in a subsequent section, we did not deliberately chase vehicles thought to be high emitters. Individual vehicles can produce high on-road UFP concentrations (see next section), particularly during acceleration from a standing state (Fruin et al., 2008). However, the enclosed characteristic of the tunnel and the associated ability to contain vehicle emissions in a small space, coupled with the number of replicate trips made, largely mitigate the effects of any trips where a single vehicle preceding the measurement platform led to UFP concentration excursions substantially above the combined plume of all preceding vehicles in the tunnel.

The strength of HDV traffic volume as a determinant of UFP concentrations was relatively poor in the westbound bore. Possible, albeit speculative, explanations for this include the fact that the westbound bore feeds traffic away from a major shipping port, so freight trucks entering this bore may be less heavily laden than their eastbound counterparts. However, the reverse could also be true depending on the proportion of HDVs loading or unloading their cargo at the port. Ventilation and air movement parameters may be of increased significance in this bore. The relative position at which air is extracted and injected in each bore could also potentially be of importance. The presence of a sustained (400m) uphill road grade of 1:20 (NSW RTA, 2008a) coincident with the westbound bore exit, and associated heavy vehicle emissions in this vicinity, may exert a substantial influence that is not apparent in the present results which are based on average hourly traffic volume and vehicle based on-road UFP sampling (i.e. individual or multiple HDVs ascending towards the westbound exit may have made a time-disproportionate contribution to measured trip average UFP concentration). A systematic difference in HDV fleet mechanical condition (Jayaratne et al., 2007), age or presence or type of exhaust cleaning device (Jones and Harrison, 2006) between bores seems unlikely, although these factors could exert some individual or combined influence on the disparity in HDV influence observed between bores. As no significant difference in HMA UFP particle concentration existed between the two bores, future research would ideally further investigate the cause of this somewhat counter-intuitive finding. Furthermore, as a filtration plant that will extract tunnel air approximately 500m from the westbound bore exit portal prior to treatment and re-introduction slightly downstream is currently under construction (NSW RTA, 2008a), a follow-up study to assess the efficacy of this device at reduction of on-road UFP concentration would be useful in examining the benefit(s) of such engineering remediation.

### UFP Concentration on the Mixed Route

Fig. 3 presents the results of sampling on the mixed roads and tunnel route. The maximum instantaneous particle concentration recorded was $7.27 \times 10^6$ p cm$^{-3}$ between 12:00 and 13:00, whilst the minimum value of $2.66 \times 10^4$ p cm$^{-3}$ was measured between 23:00 and 00:00. Hourly median particle concentration ranged from $3.71 \times 10^4$ p cm$^{-3}$ between 23:00 and 00:00, to $1.55 \times 10^5$ p cm$^{-3}$ between 13:00 and 14:00. The average hourly median concentration on the mixed route was $7.30 \times 10^4$ p cm$^{-3}$. Despite the presence of numerous smaller tunnels on the mixed route and a generally increased traffic volume compared to the M5 East tunnel, the average hourly median concentration was one order of magnitude lower than the average HMA values recorded in the M5 East tunnel bores. Fewer HDVs were evident on this route, although we did not have traffic fleet composition data to confirm these observations. Highly significant differences were present in the log-transformed hourly median particle concentration measured on the mixed roads route and the HMA concentrations in both tunnel bores ($p < 0.0001$ in both cases). This may be due to the aforementioned, in addition to wind speed and increased exhaust dispersion afforded on above ground roads, amongst other potential factors.

During a mixed roads route sampling exercise on 31/06/2006, a serendipitous opportunity arose to chase an HDV that was visibly emitting a substantial exhaust plume. The vehicle in question was a privately-operated older bus that was ostensibly in poor mechanical condition. Shortly after exiting the eastbound tunnel, the test vehicle was positioned in the trailing plume approximately 20m behind the bus, with no other vehicles present in the intervening space. Fig. 4 shows 10 s average particle concentration measurements prior to, during and after this procedure. Perhaps of most interest is the 5 min period from 18:18 to 18:23 during which the upper concentration detection limit of the dilution-equipped CPC was exceeded, corresponding to an estimated UFP concentration of $>8.9 \times 10^6$ p cm$^{-3}$. It is worthwhile to note that these levels occurred when the vehicle was driving through a shorter (approximately 500m) tunnel and on open roadways during relatively free flowing traffic conditions. The measured particle concentrations occurred despite the dilution of exhaust afforded by the distance between the bus and sampling vehicle (Morawska et al., 2007). From Fig. 4 it is apparent that two orders of magnitude in variation of on-road particle concentration can occur during a relatively short trip. The tendency of on-road UFP concentration to increase rapidly and substantially in the immediate vicinity of a diesel exhaust plume has been similarly described by Gouriou et al. (2004), Pirjola et al. (2004), Weijers et al. (2004) and Westerdahl et al. (2005). The presence of a significant nucleation mode in the exhaust plume seems likely (Morawska et al., 2008), although particle size distribution data required to confirm this postulation were

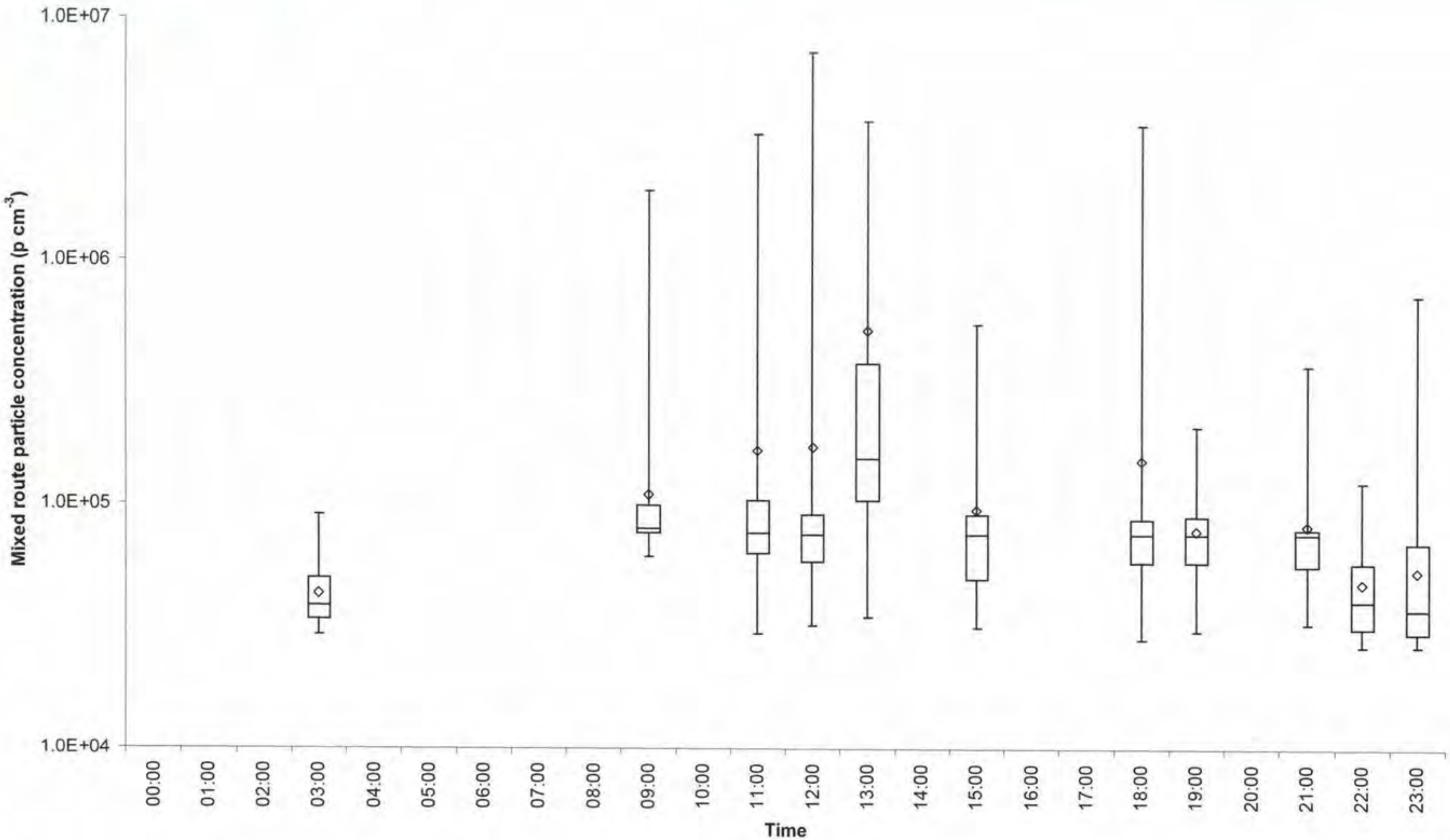


**Fig. 3.** On-road UFP concentration measured on the mixed roads route by hour. Box-and-whisker plots indicate median (thick horizontal line), average (diamond), first and third quartile (bottom and top edge of box, respectively), minimum and maximum (lower and upper extent of whiskers, respectively) UFP concentration.

not available. The data described above were excluded from the overall mixed route particle concentration measurements depicted in Fig. 3.

### Comparison With Other Environments

Morawska et al. (2008) described the results of meta-analyses applied to 71 studies of UFP characterisation conducted in a wide range of environments, as reported in the literature. Excluding their assessment of on-road and tunnel studies, which are the subject of a separate section below, the values reported by Morawska et al. (2008) ranged from $2.9 \times 10^3$ p cm$^{-3}$ to $3.9 \times 10^4$ p cm$^{-3}$ and $2.6 \times 10^3$ p cm$^{-3}$ to $4.8 \times 10^4$ p cm$^{-3}$, for median and average levels, respectively. Minimum median concentrations corresponded to studies of UFP concentration in rural locations, whilst maximum median levels were measured in street canyon environments. The minimum average particle concentrations were recorded in clean background environments, such as boreal forests. Maximum average concentrations were recorded in roadside locations. The minimum and maximum HMA UFP concentrations we measured were both one order of magnitude greater in the eastbound bore than the equivalent measurements reported by Morawska et al. (2008). A similar trend existed for data collected in the westbound bore, although the maximum HMA concentration there ($1.73 \times 10^6$ p cm$^{-3}$) was two orders of magnitude greater than the equivalent value described by Morawska et al. (2008). The hourly median UFP concentrations recorded on our mixed route (see Fig. 3) were generally of the same order as the median reported by Morawska et al. (2008) for street canyon and roadside locations.

The relatively high UFP concentrations we recorded are not unexpected given the proximity of diesel and gasoline emission sources when sampling on-road. Coupled with the enclosed nature of the tunnel and the subsequent limitations on exhaust plume dispersion imposed by this, tunnels clearly represent locations where peak daily UFP exposures are likely to occur for the vehicle occupants using them. The degree to which a person is exposed to levels commensurate with those measured on-road is dependent on the rate and penetration efficiency at which particles enter the vehicle cabin that they occupy. Particle deposition within a vehicle cabin can also be of importance (Xu and Zhu,

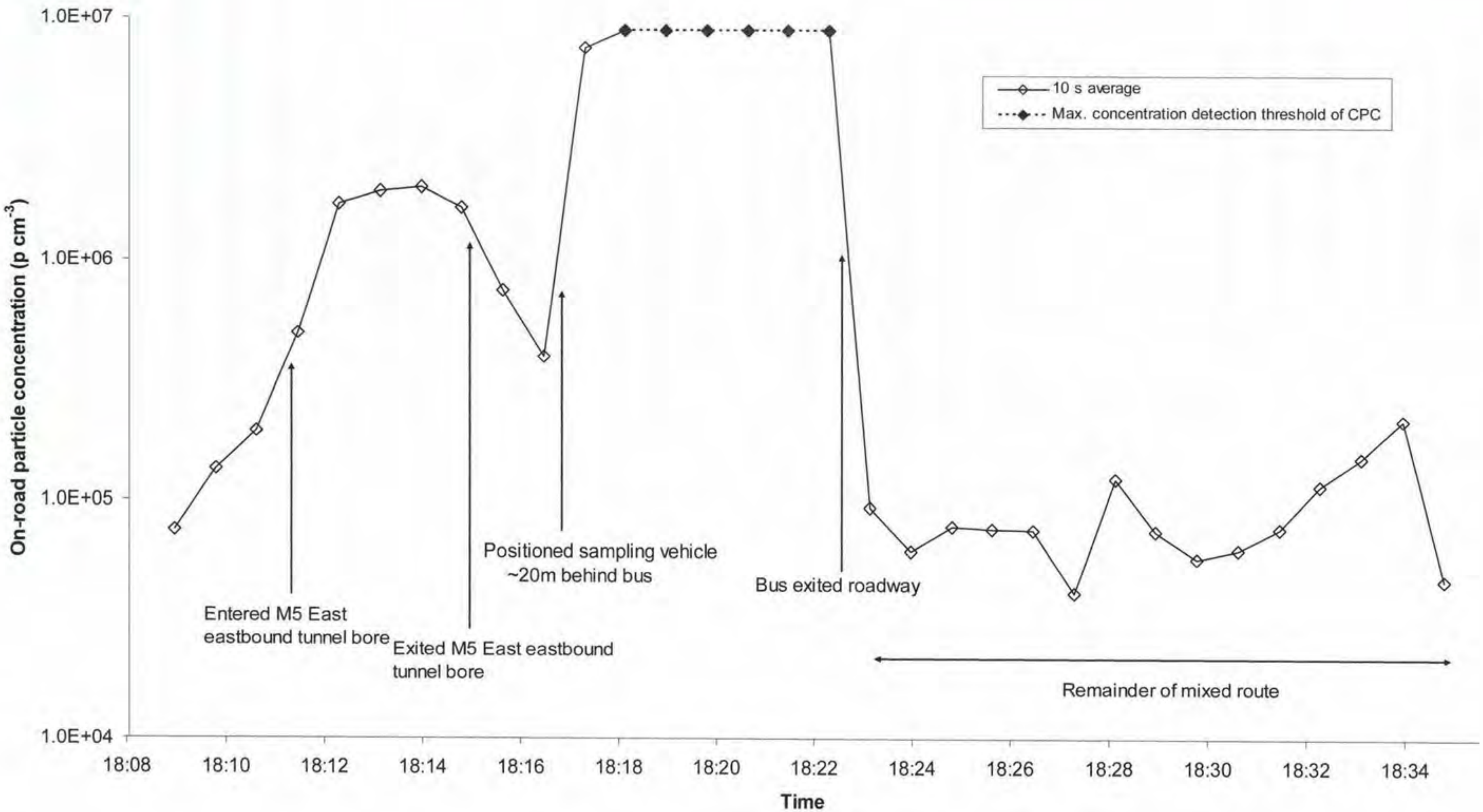


**Fig. 4.** Time-series of 10 s average on-road particle concentration recorded on 31/6/2006.

2009). Newer vehicles fitted with filtration devices have been shown to offer reasonable protection against in-cabin UFP exposure, depending on the ventilation mode used (Zhu et al, 2007; Pui et al., 2008; Qi et al., 2008). However, occupants of older, less air-tight vehicles (Knibbs et al., in press), vehicles with open windows (Ott et al., 2008), open convertible vehicles, motorcycles or other transportation mode characterised by high air change rate and/or lack of filtration may be afforded relatively small reductions in UFP concentration entering their breathing zone.

### Comparison With Other On-road and Tunnel Studies

Table 2 presents a summary of related previous work. A similar comparison was presented by Westerdahl et al. (2005), and given the relatively rapid progression in reported on-road and tunnel-based studies, it seems useful to provide an updated version. It is worth noting that the studies referred to in table 2 are not intended to represent a comprehensive summary of all on-road or tunnel based work, but rather a selection of those most relevant to the present study. Variability existed between these studies in terms of experimental equipment, particle size range of interest, study environment, whether a given on-road study aimed to chase vehicles, and the reported particle concentration statistic. Notwithstanding this, the peak in-tunnel concentrations measured in this study for a given statistic (average, median, maximum) were up to two orders of magnitude greater than those reported in other work. However, it should be considered that our values reflect tunnel transects that generally took 3.5 to 5 min to complete, which is a short averaging period relative to some other studies. As table 2 highlights, compared to other work performed in tunnels from either a static or vehicle-based platform, our results are toward the upper limit of concentrations reported in the literature. Concentrations recorded on the mixed route were more comparable with those reported in other studies, and more specifically, exhibited good agreement with results presented by Weijers et al. (2004) and Westerdahl et al. (2005), both of which employed similar equipment and focussed on an analogous particle size range to the present study.

## Conclusions

(1) Based on 301 individual vehicle-based transects, hourly median average (HMA) ultrafine particle (UFP) concentration in the 4km M5 East road tunnel ranged from $9.04 \times 10^4$ p cm$^{-3}$ to $9.06 \times 10^5$ p cm$^{-3}$ in the eastbound bore, and from $7.81 \times 10^4$ p cm$^{-3}$ to $1.73 \times 10^6$ p cm$^{-3}$ in the westbound bore. Maximum trip average UFP concentrations exceeded $5.0 \times 10^6$ p cm$^{-3}$ in both tunnel bores. Minimum trip average concentrations were two orders of magnitude lower.

(2) Hourly median concentrations recorded on a mixed route comprising major roadways and shorter tunnels ranged from $3.71 \times 10^4$ p cm$^{-3}$ and $1.55 \times 10^5$ p cm$^{-3}$.

(3) Hourly heavy diesel vehicle (HDV) volume was a highly significant determinant ($p < 0.0001$) of HMA UFP concentration in the eastbound tunnel bore ($R^2 = 0.87$), whilst a significant ($p = 0.0256$), yet only poor-fair determinant in the westbound bore ($R^2 = 0.26$). The predictive ability of passenger vehicle volume was significantly different to that of HDV volume in both bores, and was a fair ($R^2 = 0.37$) and poor ($R^2 = 0.10$) determinant in the eastbound and westbound bores, respectively. No significant difference ($p = 0.7620$) in HMA UFP concentration existed between the two bores.

(4) The HMA UFP concentrations measured in-tunnel were one to two orders of magnitude higher than equivalent measurements reported in the literature for a variety of other environments. These results, coupled with those of recent above-ground roadway studies which estimate that up to approximately 50% of Los Angeles commuters daily ultrafine exposure occurs inside vehicles (Zhu et al., 2007; Fruin et al., 2008), further highlight the significance road tunnel transit may have on in-vehicle and overall UFP exposure. As road tunnel numbers and length are likely to increase in many countries, the above issue may be a persistent one, and it would seem beneficial to mitigate vehicle occupant exposures to a range of pollutants encountered in tunnels.

(5) Given the relatively spartan sampling equipment and approach, this study was quite successful from an operational standpoint, although an increased suite of measurement instrumentation would be useful in any ensuing work.

**Table 2.** Summary of related previous work. [a] Maximum value of several reported [b] Non-tunnel route [c] In-tunnel route [d] Maximum average reported [e] Short term average [f] Average of median [g] Maximum median [h] Median of average [i] Instantaneous or very short term peak.

| Study | Location | Measurement platform | Particle size range measured | Measurement equipment | Roadway environment | Diesel % of fleet | Avg. conc. (p $cm^{-3}$) | Med. conc. (p $cm^{-3}$) | Max. conc. (p $cm^{-3}$) |
|---|---|---|---|---|---|---|---|---|---|
| This study | Sydney, Australia | Vehicle-based | 10 to >1000nm | TSI 3007 CPC | 4km tunnel and mixed route (incl. tunnels) | ~7%[c] | $6.0 \times 10^{6}$[ace] | $1.7 \times 10^{6}$[ach] $1.6 \times 10^{5}$[ab] | >$8.9 \times 10^{6}$[bci] |
| Kirchstetter et al. (1999) | Berkeley, USA | Static | >10nm | TSI 3760 CNC | 1.1km tunnel | 4.8%[a] | $4.0 \times 10^{5}$ | - | - |
| Abu-Allaban et al. (2002) | Pennsylvania, USA | Static | 10-400nm | TSI SMPS | 1.6km tunnel | 86.5%[a] | $1.9 \times 10^{5}$[e] | - | - |
| Bukowiecki et al. (2002) | Zürich, Switzerland | Vehicle-based | >3nm (CPC), 7-310nm (SMPS) | TSI 3025 UCPC, TSI SMPS | Non-tunnel routes | - | $7.8 \times 10^{4}$[a] | - | $4.0 \times 10^{5}$[i] |
| Gidhagen et al. (2003) | Stockholm, Sweden | Static | 3-900nm | DMPS | 1.5km tunnel | 5% | $7.6 \times 10^{5}$ | - | >$1.3 \times 10^{6}$[d] |
| Gouriou et al. (2004) | Rouen, France | Vehicle-based & static | 30-10 000nm | Dekati ELPI | 1.6km tunnel and non-tunnel routes | - | $9.5 \times 10^{4}$[b] $5.1 \times 10^{5}$[ace] | $3.5 \times 10^{4}$[b] | $1.5 \times 10^{6}$[i] |
| Jamriska et al. (2004) | Brisbane, Australia | Static | 17-700nm | TSI SMPS | 0.5km tunnel | 100% | - | - | ~$1.3 \times 10^{5}$[e] |
| Kittleson et al. (2004a) | Minnesota, USA | Vehicle-based | 3-1000nm (CPC), 8-300nm (SMPS) | TSI 3025A UCPC, TSI SMPS | Non-tunnel routes | ~8 % | $4.0 \times 10^{5}$ | $9.1 \times 10^{4}$ | $1.0 \times 10^{7}$[i] |
| Kittleson et al. (2004b) | New York, USA | Vehicle-based | 3-1000nm (CPC) | TSI 3025A UCPC, TSI SMPS | Non-tunnel routes | - | $5.6 \times 10^{5}$[a] | - | - |
| Weijers et al. (2004) | Amsterdam/Nijmegen, Netherlands | Vehicle-based | >7nm | TSI 3022 CPC | Mixed routes (incl. ~2km tunnel) | - | $1.6 \times 10^{5}$[ab] | - | ~$1.8 \times 10^{6}$[ci] |
| Pirjola et al. (2004) | Helsinki, Finland | Vehicle-based | >3nm (CPC), 3-50nm (SMPS), 7-10 000nm (ELPI) | TSI 3025 UCPC, TSI SMPS, Dekati ELPI | Non-tunnel routes | - | - | - | $1.3 \times 10^{6}$[i] |
| Geller et al. (2005) | Berkeley, USA | Static | 7-270nm | TSI SMPS | 1.1km tunnel | 4.9%[a] | $7.8 \times 10^{5}$[a] | - | ~$1.0 \times 10^{6}$[e] |
| Westerdahl et al. (2005) | Los Angeles, USA | Vehicle-based | 10 to >1000nm (3007), 7-1000nm (3022A), 5-600nm (SMPS) | TSI 3007 CPC, TSI 3022A CPC, TSI SMPS | Non-tunnel routes | 14%[a] | - | $1.9 \times 10^{5}$[af] | $8.0 \times 10^{5}$[i] |
| Larsson et al. (2007) | Stockholm, Sweden | Static | 20 to 1000nm (CPC), 14-100nm (SMPS) | TSI P-Trak CPC, TSI SMPS | 1.5km tunnel | ~10% | - | $1.1 \times 10^{5}$[a] | - |
| Zhu et al. (2007) | Los Angeles, USA | Vehicle-based | >5nm (CPC), 7.9-217nm (SMPS) | TSI 3785 WCPC, TSI SMPS | Non-tunnel routes | ~25%[a] | $2.6 \times 10^{5}$[a] | - | ~$5.8 \times 10^{5}$[i] |
| Lechowicz et al. (2008) | Brisbane, Australia | Static | >7nm | TSI 3022 CPC | 0.5km tunnel | ~66% | - | $4.1 \times 10^{4}$[f] | $5.4 \times 10^{4}$[g] |
| Rim et al. (2008) | Austin, USA | Vehicle-based | 20nm to 1000nm | TSI P-Trak CPC | Non-tunnel routes | - | $2.5 \times 10^{4}$[a] | $1.2 \times 10^{4}$[a] | ~$1.6 \times 10^{5}$[i] |

Limitations of the present study included the absence of particle size distribution information and lack of traffic data coincident with, and of a temporal resolution analogous to, the measurement periods. Nonetheless, the results were generally consistent with those reported in the limited number of comparable studies. Further work would ideally focus on the cause of the disparity in determinant factors of UFP concentration between the two tunnel bores. Particle size distribution (including possible impacts of ambient temperature) and chemical composition could both be assessed. Development of emission factors and examination of the mechanical condition and age of a representative sample of HDVs may also be of use. Given that two significant events are scheduled to occur in the near future, namely, the reduction of sulphur content in Australian diesel fuel to 10ppm from 2009 and the installation of a tunnel air filtration plant for the M5 East, assessment of their individual or synergistic effects on in-tunnel UFP concentration would be a prudent pathway for any subsequent study.

(6) In road tunnels such as the M5 East control of particle emissions at the source, such as the installation of oxidation catalysts and particle traps in diesel vehicles (Morawska et al., 2008), seems a logical and potentially more effective approach to reduce emissions and subsequent on-road UFP concentrations compared to reliance on tunnel air filtration devices. Developing particle number emissions standards in Australia, similar to those being promulgated in Europe, could prove to be efficacious at reducing UFP concentrations on above ground and tunnel roadways.

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# Chapter 4

# Field Study of Air Change and Flow Rate in Six Automobiles

This chapter describes a comprehensive and systematic field investigation of cabin ventilation rates inside six passenger vehicles representative of those driven on Australian roads. The tracer gas techniques described in section 1.4 were employed throughout the measurement campaign. Over 200 successful ventilation measurements were performed, making this the largest (in terms of number of measurements) identified study to-date of vehicle cabin ventilation rates. The results presented in this chapter are an integral component of the analyses described in chapter 5.

## Contribution

The measurement approach described in this chapter was first formulated by me in consultation with Richard de Dear. Steven Atkinson and I conducted an initial field investigation of the feasibility of this approach, which led to some refinements in the methods used during the sampling campaigns described in this chapter. The final sampling protocols were developed by me. All data collection was performed by me. Analyses were performed by me in consultation with Richard de Dear and Steven Atkinson. Both co-authors assisted with data interpretation. The draft manuscript was written by me, and I was responsible for creating all figures and tables. Some changes to this manuscript were suggested by Richard de Dear and Steven Atkinson, and were included in the final draft. The manuscript was submitted to *Indoor Air* in September, 2008. Reviewer comments were received in December, 2008. I was responsible for addressing all reviewer comments and making amendments to the manuscript, following consultation with the two co-authors. The revised manuscript was accepted for publication in December, 2008 and was published in the August, 2009 issue of *Indoor Air*.

# Field study of air change and flow rate in six automobiles

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## Abstract

For many people, a relatively large proportion of daily exposure to a multitude of pollutants may occur inside an automobile. A key determinant of exposure is the amount of outdoor air entering the cabin (i.e. air change or flow rate). We have quantified this parameter in six passenger vehicles ranging in age from 18 years to <1 year, at three vehicle speeds and under four different ventilation settings. Average infiltration into the cabin with all operable air entry pathways closed was between 1 and 33.1 air changes per hour (ACH) at a vehicle speed of 60 km/h, and between 2.6 and 47.3 ACH at 110 km/h, with these results representing the most (2005 Volkswagen Golf) and least air-tight (1989 Mazda 121) vehicles, respectively. Average infiltration into stationary vehicles parked outdoors varied between ~0 and 1.4 ACH and was moderately related to wind speed. Measurements were also performed under an air recirculation setting with low fan speed, while airflow rate measurements were conducted under two non-recirculate ventilation settings with low and high fan speeds. The windows were closed in all cases, and over 200 measurements were performed. The results can be applied to estimate pollutant exposure inside vehicles.

*Keywords*: Air change rate; Ventilation; Automobile; Vehicle; Exposure.

## Practical Implications

There is increasing recognition of the often disproportionately large contribution of in-vehicle pollutant exposures to overall measures. This has highlighted the need for accurate and representative quantification of determinant factors to facilitate exposure estimation and mitigation. The ventilation rate in a vehicle cabin is a key parameter affecting the transfer of pollutants from outdoors to the cabin interior, and vice-versa. New data regarding this variable are presented here, and the results indicate substantial variability in outdoor air infiltration into vehicles of differing age. The efficacy of simple measures to reduce outdoor air infiltration into 'leaky' vehicles to increase occupant protection would be a worthwhile avenue of further research.

## Introduction

Compared to other built environments, the automobile cabin has seemingly received less attention in terms of the quantification of key parameters affecting its air quality, particularly air changes per hour (ACH) occurring there (Gameiro da Silva, 2002), which is a key determinant of the exposure of occupants to pollutants. Klepeis et al. (2001) reported that 5.5% of a typical day is spent inside a vehicle, based on their survey of about 9400 United States residents. Given the generally short duration of most commuter trips in automobiles, there has perhaps been a tendency to direct many fundamental studies of indoor environmental quality towards the home, work or school environments. Nevertheless, many published in-vehicle studies have identified elevated levels of many unleaded and diesel fuel related pollutants, such as volatile organic compounds (Duffy and Nelson, 1997), carbon monoxide (Chan et al., 1991; Ott et al., 1994), elemental carbon (Adams et al., 2002), $PM_{2.5}$ (Rodes et al., 1998), and ultrafine particles (Zhu et al., 2007), compared to other environments and/or background levels. Recent work has estimated that 1.5 h of vehicle occupancy on a combination of Los Angeles arterial and freeway roads during a day (i.e. typical of a return commute to a workplace) can account for 33–45% of daily exposure to ultrafine particulate matter (Fruin et al., 2008), which supports similar estimates made by Zhu et al. (2007). For persons whose occupation necessitates longer periods be spent inside a vehicle (e.g. police officers, couriers, taxi, and truck drivers), the relative contribution of in-vehicle exposures to overall measures is likely to be greater, particularly if driving in heavily trafficked or congested areas. Exposure to traffic-related pollutants can have deleterious health implications, especially for more susceptible occupants, such as people older than 60 years or those who have existing health issues (Peters et al., 2004). However, other groups may also be affected, and the undesirable short-term health effects of in-vehicle $PM_{2.5}$ exposures on a sample of police officers have been described by Riediker et al. (2004). Particulate exposure inside vehicle cabins may also result in reduced driver vigilance (Wyon et al., 1995). Under low air change rate conditions, in-cabin pollutant sources, such as cigarette smoking (Ott et al., 2008), and material emissions from new vehicles (Zhang et al., 2008) could make potentially significant contributions to vehicle occupant exposure.

The amount of outdoor air entering an automobile can be expressed in terms of ACH or a volume flow rate. Measurement of air change rates inside passenger cars (both stationary and moving), buses and trucks appear sporadically in the literature, with the earliest identified study conducted by Petersen and Sabersky (1975), who measured between 18 and ~39 ACH in a stock Chevrolet at speeds between 0 and ~97 km/h, respectively. Ott et al. (2008) have recently summarized previous work in this area, and also supplemented the limited existing data with measurements of ACH performed in four vehicles under open and closed window conditions. In addition to the work reviewed by Ott et al. (2008), Guillemin et al. (1992) estimated means between 11.62 and 14.51 ACH inside truck cabins. Kvisgaard (1995) described methods for measuring local-mean-age-of-air inside a moving vehicle at up to 100 ACH. Conceição et al. (1997) measured up to 16 ACH inside an intercity bus travelling at 80 km/h, as a means of experimentally assessing the efficacy of a contaminant removal duct. Kvisgaard and Pejtersen (1999) detailed the measurement of outdoor airflow rate in a Honda Civic hatchback under a wide range of heating ventilation and air conditioning (HVAC) system settings and vehicle speeds, using constant injection of sulphur hexafluoride ($SF_6$). Batterman et al. (2006) evaluated a perfluorocarbon tracer ventilation measurement method inside a 2000 model Subaru Legacy with windows closed, recirculation off and a low fan setting, and reported an average of 92 ACH at an average vehicle speed of 105 km/h. Using $SF_6$ decay, Rim et al. (2008) measured between 2.6 and 4.55 ACH inside school buses. Zhang et al. (2008) measured between <0.1 and 0.63 ACH in a variety of small passenger cars parked in an underground garage. As part of a larger study, we required specific measurements of ACH and flow rate in a range of common vehicles representative of those driven on Australian roads, when both stationary and travelling at speed, and under a range of ventilation settings with the windows closed.

## Methods

### Selection of Vehicles and Research Design

There were 11.2 million passenger vehicles registered in Australia in 2006, with an average age of 9.8 years, and 21% of which were manufactured before 1991 (Australian Bureau of Statistics, 2006). We sought a small group of test vehicles to represent those being driven on Australian roads. Our focus was directed toward passenger vehicles, as these represent 78% of the Australian fleet (Australian Bureau of Statistics, 2006). In addition, we wished to include vehicles having very basic HVAC systems (i.e. no air conditioning and no filtration) and those having more advanced systems (air conditioning and filtration). The final sample of vehicles is listed in Table 1. As the vehicles are referred to henceforth by the name they are marketed under in Australia, some common international variants are provided. Two similar vehicles of differing age (2000 model Subaru Liberty and 2007 model Subaru Outback) were included to examine the effect of approximately 7 years of service life on air infiltration into the cabin. Odometer readings prior to the commencement of testing are provided in Table 1. Five of the vehicles were privately owned, while the 2005 Toyota HiLux was a university-owned vehicle. Three of the vehicles were near-new, but all were well-maintained and subject to the regular maintenances recommended by the manufacturer, as indicated by their service log books.

Four ventilation conditions were assessed. In even the most basic automotive HVAC system, the number of possible combinations of fan speed, vent position and air intake position (recirculate or fresh) is large. As such, the ventilation conditions shown in Table 2 were selected to encompass a range of settings and best match the required outcomes of this study. The abbreviated names for each ventilation condition assessed reflected the air intake position (i.e. E = external and R = recirculate) and the fan speed (i.e. 1 = first fan speed position and 3 = third fan speed position, and so on). The INF condition was named as such to reflect that air entering under this setting would do so via infiltration. The inclusion of the baseline infiltration condition was intended to represent the best measure afforded to occupants of a vehicle for mitigation of outdoor air intrusion. Given that Engelmann et al. (1992) reported highly variable differences between their test vehicles in the deliberate provision of outdoor make-up air under an air recirculation condition, we also wished to determine if this was the case in our test group. An additional recirculate ventilation condition, referred to as R4 in Table 2, was assessed in the 2005 Toyota HiLux. Measurements were conducted at nominal vehicle speeds of 0, 60 and 110 km/h. Based on a pilot study in Houston, Texas, Long et al. (2002) reported that 20% of passenger vehicles observed had one or more window (window, sunroof or convertible top) open during rain-free days characterized by an outdoor temperature range of 27.2–35°C. We did not include any open window scenarios in our selection of ventilation conditions, and instead focused our work towards the likely more predominant (in Sydney and most Australian cities, at least) closed window settings. However, automobile air change data collected under an open window setting are available elsewhere (Ott et al., 2008; Park et al., 1998; Rodes et al., 1998). A window opened by even a small amount can substantially increase the air change rate in a vehicle cabin under a given ventilation condition compared to the same condition with windows closed (Ott et al., 2008).

### Experimental Equipment and Methods

Suitable roadways were located where the test vehicles could be driven in a safe and uninterrupted manner at the desired speed for the necessary duration. Tests were conducted at times when traffic volume was low. Due to the high airflows expected under the fresh air ventilation conditions (E1 and E3), and the unsuitability of the measurement equipment for the concentration–decay method at high ACH (Gameiro da Silva, 2002; Kvisgaard, 1995), the constant injection technique using $SF_6$ as a tracer was used for these measurements. For the two remaining ventilation conditions (R1 and INF), the $SF_6$ concentration–decay approach was employed. A detailed handling of the application and theory underlying these techniques can be found in ASTM (2006). An Innova type 1412 photoacoustic field gas monitor and Innova type 1303 multipoint sampler and doser were used. All sampling and dosing tubes used were of the manufacturer recommended composition. The dosing nozzle was flushed and calibrated

**Table 1.** Test group characteristics.

| Automobile | Also marketed as | Model year | Chassis type | Odometer (km) | HVAC filter | Operable vents | AC | Est. volume ($m^3$) |
|---|---|---|---|---|---|---|---|---|
| Mazda 121 | Ford Festiva, Kia Pride | 1989 | Small Hatchback | 156 778 | No | No | No | 3.32 |
| Mitsubishi Magna | Mitsubishi Diamante | 1998 | Large Sedan (Saloon) | 129 241 | No | Yes | Yes | 3.72 |
| Subaru Liberty | Subaru Legacy | 2000 | Mid-Size Station Wagon (Estate Car) | 94 385 | Yes | Yes | Yes | 4.65 |
| Toyota HiLux | Toyota Tacoma | 2005 | Utility (Pick-Up)[a] | 7861 | No | Yes | Yes | 3.33 |
| Volkswagen Golf | Volkswagen Rabbit | 2005 | Large Hatchback | 7120 | Yes | Yes | Yes | 3.88 |
| Subaru Outback | Subaru Legacy Outback | 2007 | Mid-Size Station Wagon (Estate Car) | 7688 | Yes | Yes | Yes | 4.43 |

[a] This vehicle featured a 5 seat cabin ('crew cab'), separated from the cargo tray by the rear firewall.

**Table 2.** Ventilation conditions assessed.

| Condition | Fan | Fan speed | Air intake | Vent state | Vent direction | AC | Temperature | Windows |
|---|---|---|---|---|---|---|---|---|
| E1 | On | Lowest | Fresh | All open | Cabin | On | Coolest | All closed |
| E3 | On | Second-highest[a] | Fresh | All open | Cabin | On | Coolest | All closed |
| R1 | On | Lowest | Recirculate | All open | Cabin | On | Coolest | All closed |
| INF | Off | n/a | Recirculate | All closed | Cabin | Off | n/a | All closed |
| R4[b] | On | Highest | Recirculate | All open | Cabin | On | Coolest | All closed |

[a] In the 2007 Subaru Outback, the fourth setting of six possible was used. In all other vehicles the third setting of four possible was used. [b] This ventilation condition was assessed in the 2005 Toyota HiLux only.

periodically according to specifications. The equipment had been calibrated by the manufacturer prior to use. Innova type 7620 software (LumaSense, Ballerup, Denmark) was used to set up and control all measurements. Some initial measurements were conducted using a Brüel & Kjær type 1302 photoacoustic gas monitor and Brüel & Kjær type 1303 multipoint sampler and doser (Brüel & Kjær, Naerum, Denmark). An empirical correction factor was determined to allow inter-comparability between results obtained with the Brüel & Kjær system and those measured with the Innova system. Foam padding was used to protect the equipment and dampen vibration, which can affect the gas monitor at certain frequencies (LumaSense, 2008a; Ott et al., 2008). The waste air from the ventilation measurement equipment was voided outside of the vehicle. The flow rate of the equipment was minimal and not likely to significantly affect air change rate, even in a stationary vehicle. Following each measurement, the vehicle cabin was flushed with outdoor air for 5–10 min to obviate any effects of background $SF_6$ on the subsequent measurement.

#### Measurements in Moving Automobiles

Sulphur hexafluoride was dosed into the HVAC system inlet, located at the junction of the front windscreen and bonnet (hood). During constant injection measurements, air samples were taken from the passenger side footwell vent, the passenger side centre dashboard vent and the driver's side right dashboard vent. Tests of this nature took 15–30 min. Under the R1 and INF ventilation settings, tracer was dosed into the cabin under the E1 condition with the vehicle stationary and engine running. A single sampling point in the centre of the cabin between the front seats was used. Ott et al. (2008) used sampling points placed in the front and rear of test vehicles and reported that ACH measurements for the two locations were very similar, thus our use of a single sampling point was justified. Once the tracer concentration had stabilised (generally after about 15 min), dosing ceased and the required ventilation setting was selected. The tracer concentration at this point was typically 10–15 ppm. The vehicle was then driven to the required speed for a duration similar to that of the constant injection tests. Vehicle speed was recorded by a global positioning system (GPS). During all tests conducted in a moving vehicle the cabin was occupied by one person of approximately 0.08 $m^3$.

During measurement exercises in moving vehicles, every effort was made to ensure that measurements were distributed evenly between travel directions on the test roadway to avoid the possibility of bias due to the predominant winds (Fletcher and Saunders, 1994). Winds observed during all tests were light, and measurements were performed during periods characterized by stable weather conditions.

### Measurements in Stationary Automobiles

All stationary measurements were conducted with the engine of the test vehicle running. Measurements of E1, E3 and R1 in stationary vehicles were performed in a covered parking garage, while stationary measurements of INF were conducted on an open outdoor car park. During these measurements, wind direction and speed outside of the vehicle were measured at a height of 2 m by a RM Young model 05103 wind monitor (R.M. Young, Traverse City, MI, USA). Air temperature outside and inside of the vehicles was measured using Vaisala HMP45A probes (Vaisala, Helsinki, Finland). The interior volume of the test vehicles was estimated by measuring the number of ACH occurring in a stationary vehicle at a determined flow rate (Fletcher and Saunders, 1994). Three volume estimates were performed in each vehicle and the average of these calculated. During all stationary measurements the vehicle cabin was unoccupied.

### Ancillary Measurements and Repeatability

For every combination of vehicle, speed and ventilation setting, three replicate measurements were conducted. Although most were successful, occasional equipment malfunction or factors which affected the ability to maintain vehicle speed, such as road works or unexpected traffic, resulted in some data being discarded. Excluding interior volume estimates, 212 successful ventilation measurements were performed.

According to the manufacturer, the repeatability of Innova 1412 measurements is ±1% under standard conditions, and the influence of temperature and pressure is negligible (LumaSense, 2008a). The dosing calculation accuracy of the Innova 1303 is stated as ±2% (LumaSense, 2008b). Audio recordings of comments made by the investigator were performed during measurements, and assisted in creating accurate test logs and determining the suitability of data for subsequent analyses. Measurements were conducted in January, May and June, 2006, and September and December, 2007.

### Analysis Methods

Airflow rates measured using the constant injection method were output by the control software and then imported into a spreadsheet program. The time when the vehicle reached the required test speed, and the duration for which this speed was maintained were determined from the GPS and the driver□s audio logs. Basic descriptive statistics were calculated for airflow measurements corresponding to this period. For the majority of tests, these were based on 12–16 airflow measurements, collected over approximately 18–30 min. The mean gas concentration measured across the three sample points during tests was recorded. The deviation of the mean of each sample point from this figure was also calculated. Kvisgaard and Pejtersen (1999) suggest that if this deviation is equal to or <3% for each point, then the uncertainty in the result is ±5% or less.

For all measurements using the concentration–decay technique, the natural logarithm of $SF_6$ concentration was plotted against elapsed time in hours. After removing data points recorded during dosing, linear regression was applied to the remaining points. The number of ACH was recorded as the gradient of the regression line (ASTM, 2006). The cycle time of the equipment during these tests was faster than during constant injection measurements, due the use of one sampling point. An average of 30 data points collected over approximately 15 min were used to determine ACH. Tests conducted under higher ACH conditions took less time, while many of those conducted in stationary vehicles lasted for 1 h or more. Any measurements that failed to result in an exponential decay were not used in further analyses. Due to the relatively small number of samples for the purposes of statistical analysis, only basic descriptive statistics were calculated.

## Results and Discussion

### Vehicle Interior Volume Estimates

Table 1 gives the results of vehicle volume estimates. The estimates for the two hatchback vehicles (1989 Mazda 121 and 2005 Volkswagen Golf) at 3.32 and 3.88 $m^3$, respectively, were relatively large compared to the other test vehicles, and also the limited number of vehicle interior volume measurements reported in the literature (Fletcher and Saunders, 1994; Ott et al., 2008; Park et al., 1998). However, it should be considered that in a typical hatchback vehicle, the boot (trunk) is separated from the cabin by a removable partition. In our test hatchbacks, these partitions had to be removed to accommodate the $SF_6$ cylinder.

The relatively low estimated volume of the 2005 Toyota HiLux at 3.33 $m^3$ is due to the layout of a crew cab type utility vehicle consisting of a cabin physically separated from the cargo tray. The 4.65 $m^3$ estimated in the 2000 Subaru Liberty station wagon was 5% higher than equivalent measurement of 4.43 $m^3$ in the analogous 2007 Subaru Outback. The estimated volume of the 1998 Mitsubishi Magna sedan was 3.72 $m^3$, which is the same as the volume estimated by Ott et al. (1992) in a 1986 model Mazda 626 sedan using a tape measure. Standard deviation of volume estimates based on three replicate measurements in each vehicle ranged from 0.02 to 0.17 $m^3$, for the 2005 Volkswagen Golf and 2000 Subaru Outback, respectively, although the majority were below 0.05 $m^3$.

Based on the limited data reported, gas decay methods of assessing vehicle volume have been shown to result in estimates 4–17% higher than those obtained by manual measurement in the same vehicle (Fletcher and Saunders, 1994; Ott et al., 2008). The exclusion of hollow sections in concealed or difficult to reach parts of the interior during manual measurement is the likely cause of this (Ott et al., 2008). Our tracer gas estimate of volume in the 2000 Subaru Liberty was 13.5% higher than that estimated by Batterman et al. (2006) in the same year and model vehicle, although the estimation method employed by the authors was not stated.

### Constant Injection Measurements: E1 and E3 Ventilation Conditions in Stationary and Moving Automobiles

Descriptive statistics and linear regression models for all ventilation conditions and vehicles are provided in Table 3. The results of airflow measurements conducted under the E1 and E3 ventilation modes are presented in Figures 1 and 2, respectively. Ninety-six per cent of constant injection airflow measurements conformed to the guidelines suggested by Kvisgaard and Pejtersen (1999) for achieving better than ±5% uncertainty. Those measurements which did not meet the guideline did so by a small margin only.

When stationary, the mean measured flows under the E1 condition varied between 96 and 155 $m^3/h$. The equivalent range under the E3 mode was 225–300 $m^3/h$. In both cases, the lowest flow rate was measured in the 1989 Mazda 121 and the highest in the 2005 Volkswagen Golf. The variation in flow rates in stationary vehicles is due entirely to differences in blower fan air delivery between vehicles, as the vehicles were tested in a garage protected from wind.

At the nominal test speed of 60 km/h, the range of mean airflow values recorded under the E1 setting across the test group ranged from 147 to 245 $m^3/h$, with the former measure made in the 2007 Subaru Outback and the latter in the 1998 Mitsubishi Magna. Under the E3 setting, the lowest mean airflow of 271 $m^3/h$ was recorded in the 2005 Toyota HiLux, while the maximum mean value of 343 $m^3/h$ was measured in the 1998 Mitsubishi Magna.

Mean measurements at 110 km/h under the E1 condition ranged from 177 to 281 $m^3/h$. These measures were recorded in the 2000 Subaru Liberty and 1989 Mazda 121, respectively. Under the E3 setting the lowest mean flow of 292 $m^3/h$ was measured in the 2005 Toyota HiLux, while the highest mean flow of 399 $m^3/h$ was recorded in the 1998 Mitsubishi Magna. Under both ventilation conditions, the increase in airflow compared to other speeds was highly variable between vehicles and can be seen in Figures 1 and 2 and Table 3.

Kvisgaard and Pejtersen (1999) measured airflows in a Honda Civic hatchback under a ventilation setting equivalent to our E1 condition of ~140, ~160 and ~190 $m^3/h$, at 0, 60 and 110 km/h, respectively. They also conducted measurements in the same vehicle using a setting equivalent to our E3, which

**Table 3** – Linear regression models and descriptive statistics for all test vehicles.

| Automobile | | E1 ($m^3/h$) | E3 ($m^3/h$) | R1 (ACH) | INF (ACH)[a] |
|---|---|---|---|---|---|
| 1989 Mazda 121 | Regression, ($R^2$) | y=1.78x + 93.83, (0.99) | y=1.16x + 222.50, (0.99) | y=0.45x + 3.90, (0.92) | y=0.32x + 14.12, (0.76) |
| | 0km $h^{-1}$: mean, *n*, s.d. | 96.1, 3, 0.8 | 224.9, 3, 2.1 | 0.2, 2, n/a | 1.4, 2, n/a |
| | 60 km $h^{-1}$: mean, *n*, s.d. | 189.7, 2, n/a | 284.4, 3, 3.0 | 35.6, 3, 5.2 | 33.1, 3, 6.1 |
| | 110 km $h^{-1}$: mean, *n*, s.d. | 281.3, 2, n/a | 346.4, 3, 2.7 | 47.1, 3, 3.6 | 47.3, 3, 3.7 |
| 1998 Mitsubishi Magna | Regression, ($R^2$) | y=1.16x + 156.14, (0.92) | y=1.27x + 266.20, (0.99) | y=0.21x + 1.05, (0.87) | y=0.14x - 0.52, (0.92) |
| | 0km $h^{-1}$: mean, *n*, s.d. | 148.8, 3, 3.2 | 264.4, 3, 0.4 | 3.3, 3, 0.5 | 0.1, 1, n/a |
| | 60 km $h^{-1}$: mean, *n*, s.d. | 245.2, 2, n/a | 343.5, 2, n/a | 8.2, 3, 0.6 | 7.8, 3, 0.2 |
| | 110 km $h^{-1}$: mean, *n*, s.d. | 266.9, 2, n/a | 398.8, 2, n/a | 26.2, 3, 0.3 | 14.6, 3, 1.9 |
| 2000 Subaru Liberty | Regression, ($R^2$) | y=0.41x + 130.61, (0.92) | y=0.40x + 259.32, (0.91) | y=0.06x + 0.41, (0.99) | y=0.05x + 0.42, (0.98) |
| | 0km $h^{-1}$: mean, *n*, s.d. | 132.3, 3, 1.2 | 257.7, 3, 6.6 | 0.5, 3, 0.02 | 0.3, 3, 0.1 |
| | 60 km $h^{-1}$: mean, *n*, s.d. | 151.7, 3, 1.4 | 287.0, 3, 5.0 | 4.1, 3, 0.1 | 3.6, 3, 0.2 |
| | 110 km $h^{-1}$: mean, *n*, s.d. | 176.9, 3, 9.0 | 298.6, 2, n/a | 7.3, 3, 0.5 | 6.0, 3, 0.3 |
| 2005 Toyota HiLux[b] | Regression, ($R^2$) | y=0.78x + 126.37, (0.90) | y=0.29x + 259.02, (0.93) | y=0.06x + 0.90, (0.97) | y=0.04x + 1.82, (0.99) |
| | 0km $h^{-1}$: mean, *n*, s.d. | 131.7, 3, 1.2 | 260.6, 3, 1.2 | 0.7, 3, 0.1 | 1.2, 3, 0.4 |
| | 60 km $h^{-1}$: mean, *n*, s.d. | 154.8, 2, n/a | 271.4, 2, n/a | 4.8, 3, 0.1 | 4.2, 3, 0.1 |
| | 110 km $h^{-1}$: mean, *n*, s.d. | 217.0, 3, 12.0 | 292.5, 3, 4.1 | 6.7, 3, 0.6 | 6.0, 2, n/a |
| 2005 Volkswagen Golf | Regression, ($R^2$) | y=0.63x + 149.99, (0.87) | y=0.30x + 298.05, (0.91) | y=0.03x + 0.05, (0.99) | y=0.03x - 0.95, (0.96) |
| | 0km $h^{-1}$: mean, *n*, s.d. | 155.1, 3, 0.7 | 300.0, 3, 0.3 | 0.1, 3, 0.01 | 0, 3, 0 |
| | 60 km $h^{-1}$: mean, *n*, s.d. | 174.2, 3, 8.1 | 311.8, 3, 3.6 | 1.3, 3, 0.1 | 1.0, 2, n/a |
| | 110 km $h^{-1}$: mean, *n*, s.d. | 224.4, 2, n/a | 333.1, 2, n/a | 2.7, 3, 0.1 | 2.6, 3, 0.2 |
| 2007 Subaru Outback | Regression, ($R^2$) | y=0.50x + 127.14, (0.91) | y=0.10x + 286.49, (0.90) | y=0.08x + 0.02, (0.96) | y=0.31x - 0.04, (0.98) |
| | 0km $h^{-1}$: mean, *n*, s.d. | 131.0, 3, 1.9 | 286.4, 2, n/a | 0.3, 3, 0.05 | 0.3, 3, 0.1 |
| | 60 km $h^{-1}$: mean, *n*, s.d. | 147.0, 3, 0.7 | 292.3, 3, 1.3 | 4.0, 3, 0.3 | 17.4, 3, 1.3 |
| | 110 km $h^{-1}$: mean, *n*, s.d. | 183.9, 3, 4.1 | 296.3, 1, n/a | 8.8, 3, 0.8 | 32.0, 3, 0.9 |

Variable y in regression models refers to ACH or $m^3/h$, depending on ventilation mode. Variable x refers to vehicle speed in km $h^{-1}$. [a] Linear regression equations for a given vehicle under the INF condition were based on measures made at 60 and 110 km/h. Mean wind speeds measured during infiltration tests in stationary vehicles were 9.83, 1.37, 1.09, 5.77, 3.06 and 2.49 km/h for the 1989 Mazda 121, 1998 Mitsubishi Magna, 2000 Subaru Liberty, 2005 Toyota HiLux, 2005 Volkswagen Golf and 2007 Subaru Outback, respectively. [b] The INF ventilation condition was assessed twice in this vehicle. The values given above were recorded in January 2006. Subsequent testing in September 2007 resulted in means of 4.4 ($n$ = 3, s.d. = 0.3) and 6.1 ($n$ = 3, s.d. = 0.5) ACH at 60 and 110 km/h, respectively. The equation derived from linear regression applied to these data was: $y = 0.03x + 2.35$ ($R^2 = 0.89$). An additional ventilation mode, referred to as R4 in table 2, was measured in the 2005 Toyota HiLux. Measured mean values under this setting were 1.7 ($n$ = 4, s.d. = 0.03), 5.5 ($n$ = 3, s.d. = 0.1) and 7.7 ($n$ = 3, s.d. = 0.5) ACH at 0, 60 and 110 km/h, respectively. Linear regression applied to data collected under the R4 setting gave the following equation: $y = 0.06x + 1.82$ ($R^2 = 0.99$).

No page 10
Page 13 repeated.

this condition were not correlated with vehicle speed, due to a vehicle dependent factor. Therefore, the variation between their results and ours is not surprising.

Our data generally confirm a linear increase in airflow with increasing vehicle speed, and the results of linear regression models fitted to data collected in all vehicles under both E1 and E3 condition are given in Table 3. The model gradients are steeper for the two oldest vehicles, and decrease significantly for the newer vehicles, which can be seen in Figures 1 and 2. For example, under the E3 condition there is only a minor increase in mean flow of 10 $m^3/h$ for a speed increase of over 100 km/h in the 2007 Subaru Outback, the newest of the tested vehicles. The model gradients decreased under the E3 condition compared to the E1 condition in all but one case (1998 Mitsubishi Magna). The scope of this work was not intended to cover the mechanics underlying automobile HVAC systems; however, the interested reader is directed to the model for passive ventilation in a moving vehicle with closed windows and fan and recirculation off developed by Fletcher and Saunders (1994) and recently validated by Ott et al. (2008). When viewed in conjunction with our data, the aforementioned studies highlight the influence of an operating fan, and possibly other factors (such as inlet and outlet vent position, filter presence, and size of HVAC ducting) on reduction of natural ventilation flow that would, in some of our test vehicles, otherwise occur at speed under a fresh air intake setting.

### Concentration-decay Measurements: Infiltration in Stationary Automobiles

Figure 3 and Table 3 show the results of concentration-decay measurements performed under the INF ventilation mode with the test vehicles stationary, plotted against mean wind speed during the tracer decay period. During these tests, average wind direction measurements indicated that winds originated from the rear or front the vehicle, based on the definitions of Fletcher and Saunders (1994).

Three replicate tests made in the 2005 Volkswagen Golf of approximately one hour duration each during mean wind speeds of 2.6–3.8 km/h failed to result in a measurable decay in tracer gas concentration. Therefore, the air change rate for these conditions has been plotted as 0 per hour. Measurements made in the

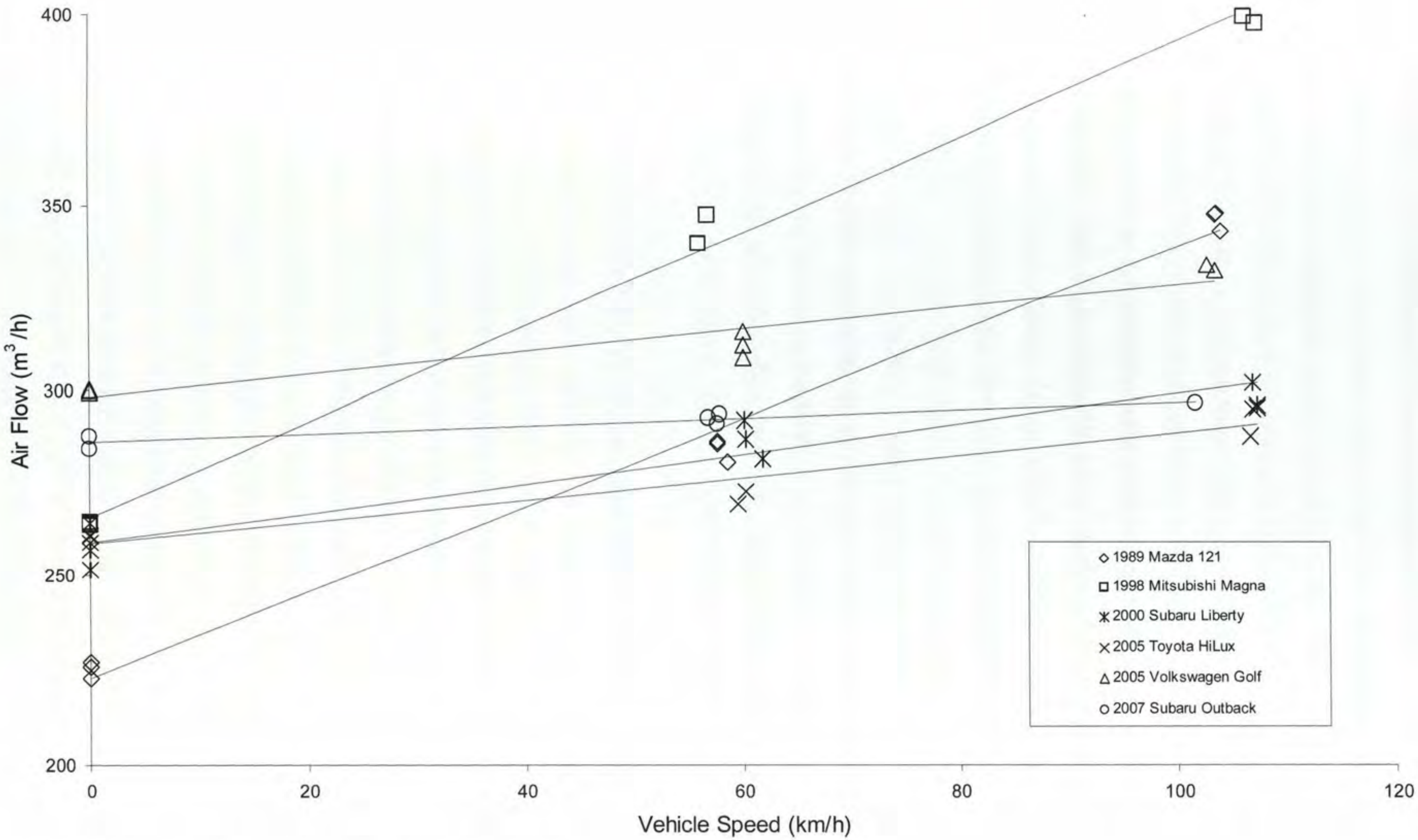


**Figure 2.** Vehicle speed vs. air flow rate under the E3 ventilation condition

resulted in airflows of ~260, ~300 and ~320 $m^3$/h at the vehicle speeds mentioned above. All of their reported measurements lie within the ranges we recorded under the same speed and ventilation settings.

Rodes et al. (1998) reported air change rates measured in three vehicles under a medium fan setting with recirculation switched off and closed windows. A stationary measurement was performed only in a 1997 Ford Explorer, which resulted in value of 20.7 ACH. At ~56 km/h in the same vehicle, the measured value was 35.7 ACH. Measurements were conducted in three vehicles at □89 km/h, which resulted in a range of 56–98 ACH. Although we measured outdoor airflow in differing units from Rodes et al. (1998), based on our estimates of vehicle volume, measurements made under our E1 condition compare moderately well with theirs.

Park et al. (1998) measured between 36.2 and 47.5 ACH in a stationary 1994 model Hyundai Sonata sedan with windows closed and recirculation switched off. Based on the interior volume reported by the authors, this is equivalent to a volume flow rate range of 103–135 $m^3$/h. These values lie within the range we recorded for our test vehicle group under the E1 setting when stationary.

As stated previously, Batterman et al. (2006) applied perfluorocarbon tracer techniques to measure air change rate in a 2000 model Subaru Legacy/Liberty station wagon with recirculation off, a low fan setting and windows closed at an average vehicle speed of 105 km/h. Despite the similarities between test conditions, the mean value of 177 $m^3$/h measured in our 2000 model Subaru Liberty under the E1 setting at an almost identical average speed is considerably lower than their reported mean of 92 ACH, which corresponds to a flow rate of 377 $m^3$/h using the estimated interior volume of 4.1 m3 provided by Batterman et al. (2006). However, the primary motivation for their study was to evaluate a measurement methodology, and furthermore, they stated that their results should not be taken as representative.

Values of ~30 ACH (~72 $m^3$/h) reported by Ott et al. (2008) in a 2005 Ford Taurus under a setting comparable to E1 at a range of speeds were significantly lower than those we measured. However, they noted the results obtained under this condition were not correlated with vehicle speed, due to a vehicle dependent factor. Therefore, the variation between their results and ours is not surprising.

Our data generally confirm a linear increase in airflow with increasing vehicle speed, and the results of linear regression models fitted to data collected in all vehicles under both E1 and E3 condition are given in Table 3. The model gradients are steeper for the two oldest vehicles, and decrease significantly for the newer vehicles, which can be seen in Figures 1 and 2. For example, under the E3 condition there is only a minor increase in mean flow of 10 $m^3/h$ for a speed increase of over 100 km/h in the 2007 Subaru Outback, the newest of the tested vehicles. The model gradients decreased under the E3 condition compared to the E1 condition in all but one case (1998 Mitsubishi Magna). The scope of this work was not intended to cover the mechanics underlying automobile HVAC systems; however, the interested reader is directed to the model for passive ventilation in a moving vehicle with closed windows and fan and recirculation off developed by Fletcher and Saunders (1994) and recently validated by Ott et al. (2008). When viewed in conjunction with our data, the aforementioned studies highlight the influence of an operating fan, and possibly other factors (such as inlet and outlet vent position, filter presence, and size of HVAC ducting) on reduction of natural ventilation flow that would, in some of our test vehicles, otherwise occur at speed under a fresh air intake setting.

#### Concentration-decay Measurements: Infiltration in Stationary Automobiles

Figure 3 and Table 3 show the results of concentration-decay measurements performed under the INF ventilation mode with the test vehicles stationary, plotted against mean wind speed during the tracer decay period. During these tests, average wind direction measurements indicated that winds originated from the rear or front the vehicle, based on the definitions of Fletcher and Saunders (1994).

Three replicate tests made in the 2005 Volkswagen Golf of approximately one hour duration each during mean wind speeds of 2.6–3.8 km/h failed to result in a measurable decay in tracer gas concentration. Therefore, the air change rate for these conditions has been plotted as 0 per hour. Measurements made in the remaining vehicles ranged from 0.14 to 1.8 ACH, which is generally in accord with other reported values (Fletcher and Saunders, 1994; Ott et al., 2008; Park et al., 1998; Zhang et al., 2008). For comparison, data reported by Park et al. (1998) in three unoccupied stationary vehicles under similar ventilation conditions and subject to winds that originated from the front of their test vehicles are also included in Figure 3. Although Park et al. (1998) found no statistically significant relationship between wind speed and infiltration in stationary vehicles, their data combined with ours reveals a moderate tendency of the air change rate to increase with wind speed in vehicles for which a sufficient range of wind speeds occurred during measurements (see also Fletcher and Saunders, 1994). Increasing age of vehicles and the associated weathering of door seals and other leakage pathways is likely to result in higher infiltration rates (Park et al., 1998). However, it should be considered that wind speeds during our measurements in most newer vehicles were consistently low. We did not observe any thermal effects on infiltration despite temperature differences of up to 11°C between the vehicle cabin and outdoor environments. Likewise, the presence of winds originating from the front or rear of the vehicles during our measurements did not result in different infiltration rates. These two findings support those of Fletcher and Saunders (1994), and in the case of the former, Park et al. (1998) also.

#### Concentration-decay Measurements: Infiltration in Moving Automobiles

The results of measurements performed under the INF condition in moving vehicles are shown in Figure 4 and Table 3. Data collected under the INF condition while moving have been plotted separately from those collected with the vehicle stationary, as Fletcher and Saunders (1994) noted that a vehicle driven at a particular speed resulted in a higher number of ACH than would occur in a stationary vehicle subject to an equivalent wind speed. Results obtained increased linearly and ostensibly fell into four distinct categories. At 60 km/h, mean infiltration into the 2005 Volkswagen Golf was 1.0 ACH, while at 110 km/h the equivalent measure was 2.6 ACH, making it the most air-tight vehicle in the test group. Mean values measured in the 2000 Subaru Liberty were

3.6 and 6.0 ACH, at 60 and 110 km/h, respectively. Infiltration into the 2005 Toyota HiLux while moving was re-assessed 20 months after the initial test, during which time the odometer reading increased by 16,226 km. During the first set of measurements in January 2006, mean air changes at 60 and 110 km/h were 4.2 and 6.0 per hour, respectively. Measurements performed in September 2007 under the same conditions resulted in means of 4.4 and 6.1 ACH, indicating no significant changes in the air tightness of the vehicle occurred as a result of the relatively modest amount of usage it received between the two tests. Values reported by Ott et al. (2008) under a comparable ventilation setting in a 2005 Ford Taurus and 2005 Toyota Corolla have been plotted on Figure 4, and it can be seen that our measurements in the 2000 Subaru Liberty and 2005 Toyota HiLux exhibited good agreement with these. Mean infiltration into the 1998 Mitsubishi Magna at 60 km/h was 7.8 ACH, and 14.6 ACH at 110 km/h. Equivalent measurements performed in the 1989 Mazda 121 resulted in mean values of 33.1 and 47.3 ACH, which should be treated cautiously, as the ability of the gas monitoring equipment to adequately measure such high air changes is limited due to its time constant (Kvisgaard, 1995). In addition, the contribution of tracer gas from semi-enclosed sections inside the cabin where the age of air is increased may also become significant under such conditions (G. Clausen, 2008; personal communication). Nonetheless, if these factors affected our measurements, then the 'true' number of air changes in this vehicle would be higher than we recorded. Air infiltration into the 1989 Mazda 121 was one order of magnitude greater than that into the 2005 Volkswagen Golf at the speeds tested.

Results under the INF condition for the 2007 Subaru Outback are not included in Figure 4, as the HVAC system of this vehicle automatically reverted to fresh air intake when the fan was turned off. However, the measured mean values of 17.4 and 32.0 ACH at 60 and 110 km/h, as shown in Table 3, agree reasonably with those presented by Fletcher and Saunders (1994) and Ott et al. (2008) for passive ventilation with both recirculation and the fan switched off and the windows closed.

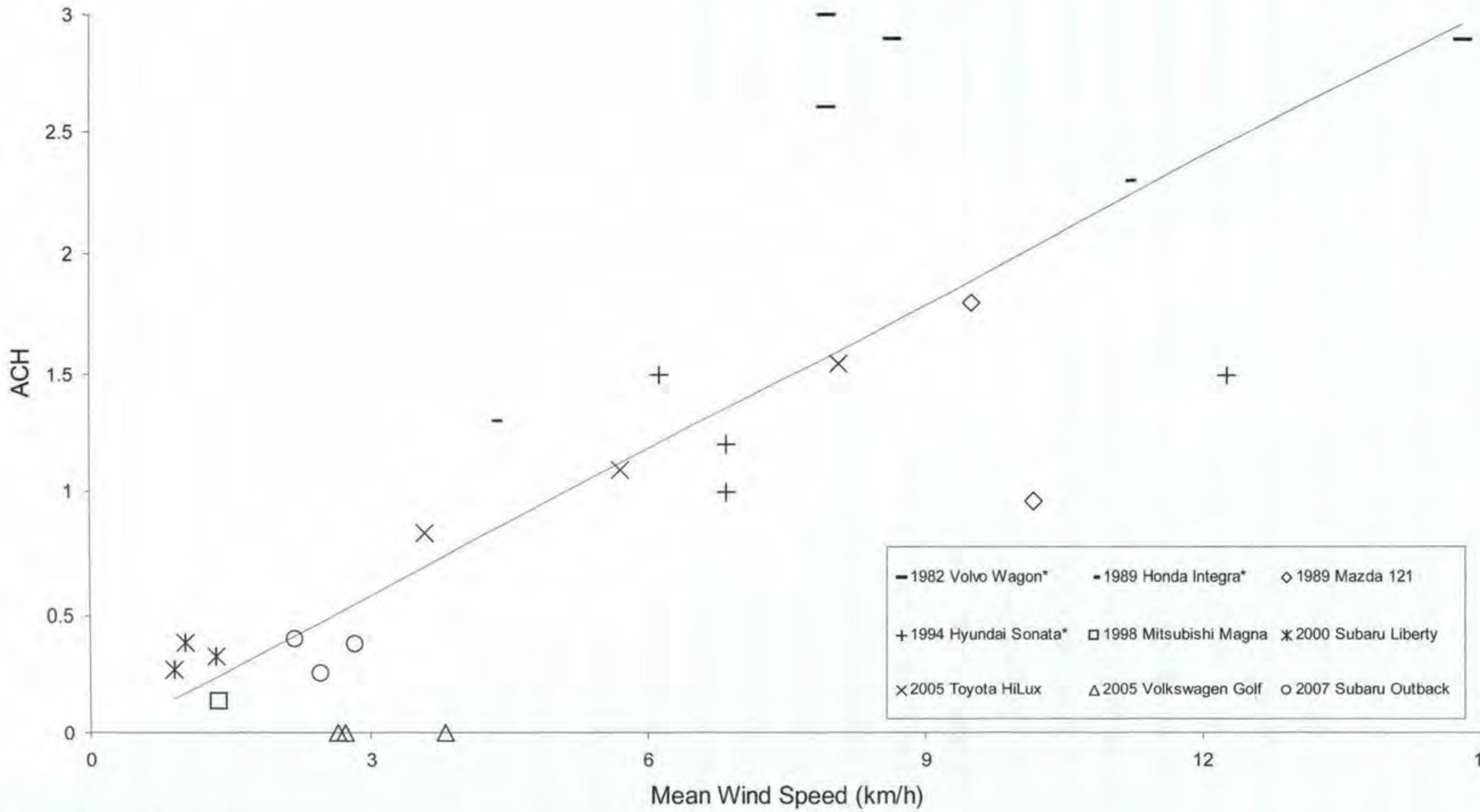


**Figure 3.** Mean wind speed vs. ACH in stationary vehicles under the INF ventilation condition. *Denotes results reported by Park et al. (1998). Linear regression applied to the data reported by Park et al. (1998), combined with our data, returned an equation of: $y = 0.20x - 0.03$ ($R^2 = 0.64$). Variable y in regression model refers to ACH, variable x refers to wind speed in km $h^{-1}$. Note that measurements reported for the 2007 Subaru Outback were obtained after its HVAC system automatically reverted to fresh air intake when the fan was switched off.

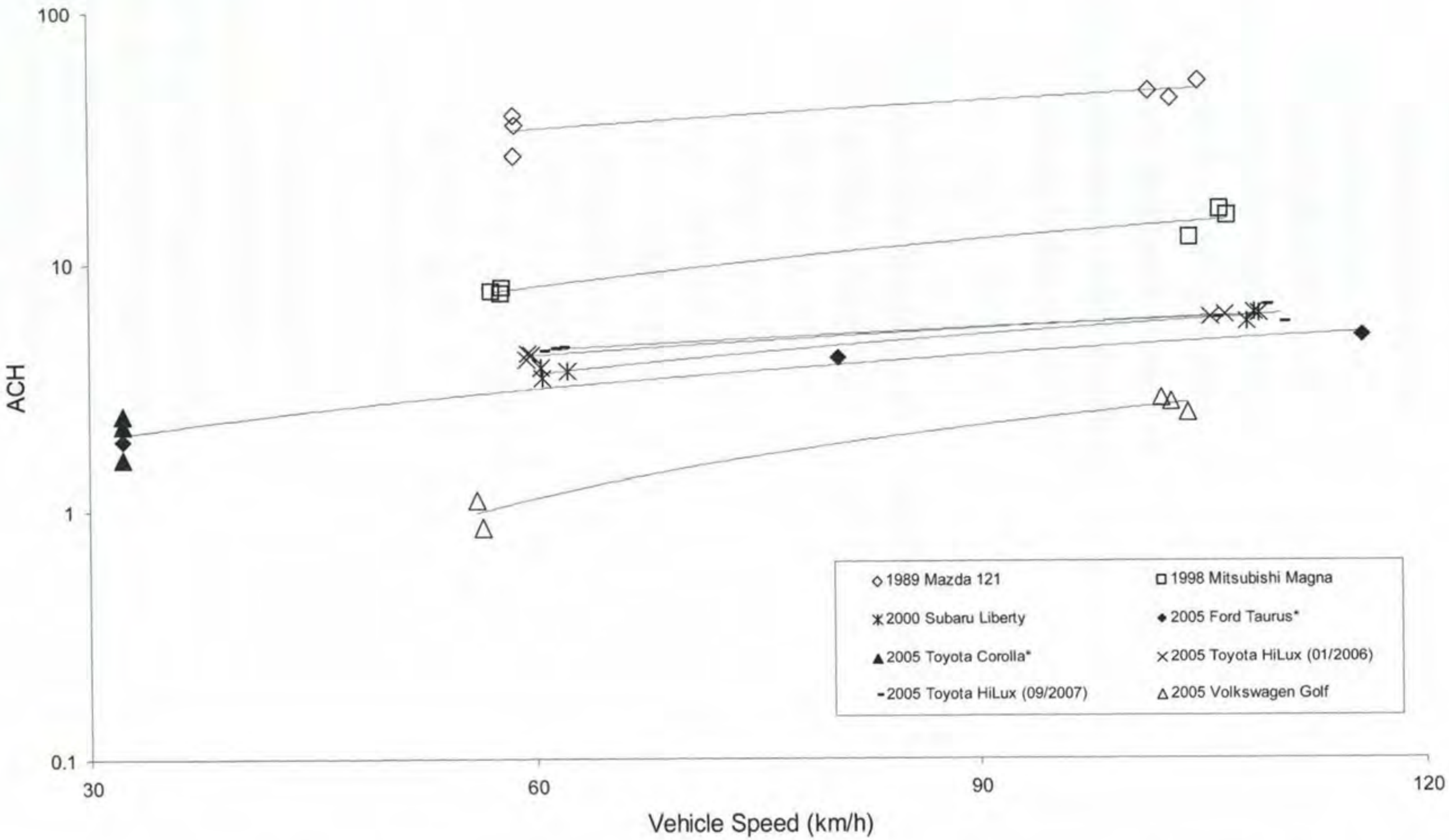


**Figure 4.** Vehicle speed vs. ACH in moving vehicles under the INF ventilation condition. *Denotes results reported by Ott et al. (2008). Linear regression applied to the data reported by Ott et al. (2008) for a 2005 Ford Taurus returned an equation of: $y = 0.04x + 0.81$ ($R^2 = 0.98$). Variable y in regression model refers to ACH, variable x refers to vehicle speed in km/h.

The likely primary pathway of air infiltration is the recirculation damper, which can deteriorate over time and present a less effective barrier to incoming air. Due to the test vehicles being privately owned, we were not able to dismantle their HVAC systems and physically examine the state of the damper. Window and door seals are the other likely sources of infiltration, and can also be prone to reduced effectiveness with age and wear. The factory-fitted air vents inside the cabin of the 1989 Mazda 121 were not operable and permanently set in the open position, which reduces resistance to air infiltrating through the HVAC system. There are a multitude of air infiltration pathways in automobiles. Ziskind et al. (1981) examined these in buses, taxis and police vehicles, and a similar study based on common passenger vehicles currently in use would be a worthwhile avenue of future research.

**Concentration-decay Measurements: Recirculate Setting in Stationary and Moving Vehicles**

Figure 5 and Table 3 show the measured air changes in the vehicle group under a recirculation setting. With the vehicles stationary, the measured air changes were generally <1 per hour, except in the 1998 Mitsubishi Magna where a mean of 3.3 ACH occurred. Due to differing measurement locations, it is difficult to compare results obtained under the INF and R1 settings when the vehicles were stationary. While moving, the air change rate in most vehicles was very similar to the values recorded under the INF setting, as Table 3 shows. This result indicates that little, if any, outdoor air is deliberately introduced into the cabin by the operating HVAC system. However, air change at 110 km/h under the R1 mode in the 1998 Mitsubishi Magna was almost double that measured under the INF condition. It is thought that this is due to a vehicle-dependent factor that allows additional air to enter under this setting. This finding may also explain why a significantly higher air change rates were measured under the R1 setting than under the INF setting when this vehicle was stationary, despite measurements of the former being conducted in a covered parking garage and those of the latter being performed outdoors. Mean air changes measured under the R4 setting in the 2005 Toyota HiLux were approximately 1 per hour higher at all speeds than mean values recorded under the R1 setting. Increasing fan speed and the associated change in negative pressurization in the vicinity of the recirculation damper thus appeared to moderately accentuate infiltration.

Our measurements under the R1 setting compare well with the limited number collected under similar conditions as reported in the literature. Data from Rodes et al. (1998) and Ott et al. (2008) have been plotted on Figure 5. It is interesting to note that the results of Rodes et al. (1998) for a 1991 Chevrolet Caprice and 1997 Ford Explorer and Ford Taurus are comparable to our measurements made in the 1989 Mazda 121 and 1998 Mitsubishi Magna, respectively. As the work of Rodes et al. (1998) was conducted almost 10 years before our measurements, this introduces the likelihood of vehicle air-tightness being a reflection of manufacturing and design techniques, rather than a direct indicator of damper and seal degradation. However, a combination of both factors is likely. In addition, some manufacturers may allow a certain amount outdoor air intrusion to prevent excessive $CO_2$ build-up.

The inability to implement a representative INF mode in the 2007 Subaru Outback made it difficult to compare the effect of its shortened service life on infiltration compared to the 2000 Subaru Liberty. However, using the measures recorded in the 2007 Subaru Outback under the R1 condition as a surrogate for infiltration, it can be seen from Table 3 that the mean values recorded are comparable to those made in the 2000 Liberty under the INF condition. It is not possible to make a conclusive statement based on the data collected, but it would appear that no significant decrease in air-tightness has occurred in the 2000 Subaru Liberty during seven years of service prior to this study.

Pui et al. (2008) reported that significant reductions in nanoparticle concentration inside vehicles were achieved through the use of recirculation in two vehicles fitted with filters. The vehicles tested were both recent models (2003 Saab 93 and 2007 Toyota Camry). Similar findings were previously reported by Zhu et al. (2007). Becalski and Bartlett (2006) noted that the use of a recirculate setting in a 2002 model Honda Civic sedan substantially lowered the concentration of in-cabin methanol (introduced through the application of windshield cleaning fluid) compared to non-recirculate settings. Our data support these findings, but also indicate that outdoor air intrusion under a recirculation setting varies considerably between vehicles of differing age, and conceivably so too does the protection of the occupants from both internal and external pollutant sources.

The recirculate condition is perhaps the most relevant ventilation mode, as it represents, in most cases, increased protection from outdoor air compared to the E1 and E3 modes, while offering the occupant better thermal comfort options compared to the INF mode. Car manufacturers and traffic regulatory authorities often recommend drivers use this setting when travelling through polluted or dusty environments. Recirculation can also substantially reduce the power required to cool or heat air compared to fresh air intake of ambient air, which consequently results in improved fuel efficiency and reduced emissions (Farrington and Rugh, 2000). Therefore, mitigation of high outdoor air infiltration into vehicle cabins is relevant not only to occupant protection, but also to overall vehicle fleet emissions. In the case of an internal pollutant source, discretionary use of fresh air intake ventilation conditions or open windows is advisable.

## Conclusions

- Under fresh air intake ventilation modes and closed windows, outdoor airflow increased with increasing vehicle speed, and generally varied by 50–100 $m^3/h$ between test vehicles at a given speed.
- The amount of increase in airflow with speed was greater in older vehicles. The causes have not been investigated in this study, but changes in HVAC system design in newer vehicles are thought to be a significant influence.
- Under fresh air intake conditions and closed windows, the operation of the HVAC fan is likely to have reduced the influence of pressure gradient-driven passive ventilation into some vehicles.

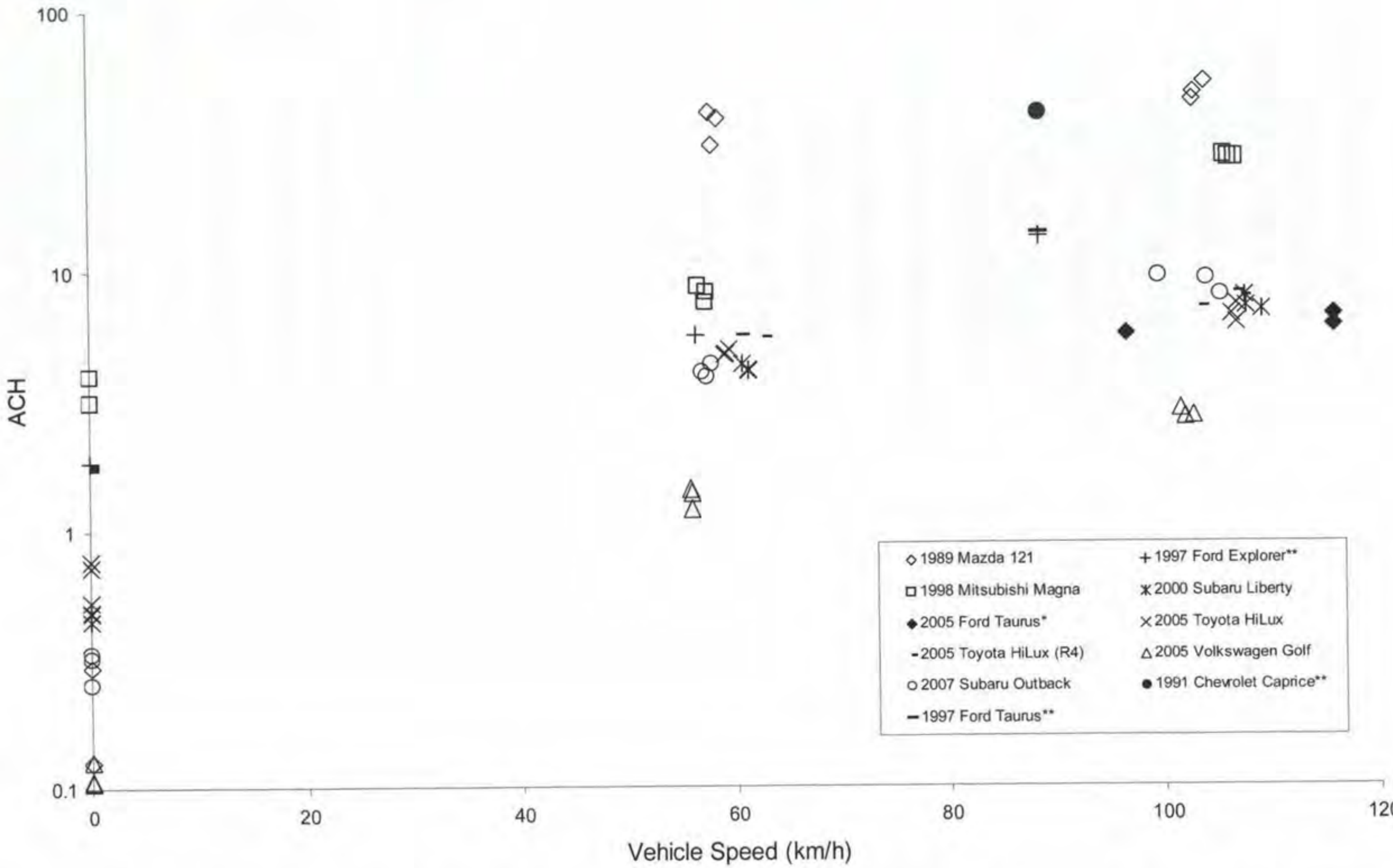


**Figure 5.** Vehicle speed vs. ACH under the R1 ventilation condition. *Denotes results reported by Ott et al. (2008). **Denotes results reported by Rodes et al. (1998). Linear regression applied to the data reported by Rodes et al. (1998) for a 1997 Ford Explorer returned an equation of: $y = 0.12x + 0.94$ ($R^2 = 0.88$). Variable y in regression model refers to ACH, variable x refers to vehicle speed in km/h.

- Infiltration of outdoor air with all operable leakage pathways closed under all test conditions was one order of magnitude greater in the least air tight vehicle (1989 Mazda 121) compared to the most air tight (2005 Volkswagen Golf).
- There is considerably less variation between vehicles in passive ventilation with the fresh air intake mode selected (Fletcher and Saunders, 1994; Ott et al., 2008) compared to air infiltrating with recirculate selected, as the influence of vehicle-dependent factors related to the recirculation damper, window and door seal air tightness increase under the latter setting.
- Based on limited data, we found no apparent evidence of a significant reduction in air-tightness in both the 2000 Subaru Liberty and 2005 Toyota HiLux over periods of 7 years and 20 months, respectively.
- There was little evidence to suggest any additional outdoor air is deliberately brought into the most test vehicles by the HVAC system under a recirculation setting, other than that which infiltrates naturally. However, this may not be the case for the 1998 Mitsubishi Magna, nor for all vehicles generally.
- The substantial variability in outdoor airflow and change rate between vehicles highlights the importance of accurate quantification of these parameters as a means of better estimating and understanding variations in exposure of occupants of different vehicles to a range of pollutants.
- The results of this study generally agree well with the limited number of previously published studies for similar closed window cases. We have provided substantial new data to supplement existing results, in addition to filling gaps in the knowledge of automobile cabin airflow and change rates. In conjunction with recent contributions by Ott et al. (2008), we feel the available data now better represent the vehicle fleet in many countries.
- The results presented here can be applied to estimation of pollutant concentrations in automobile cabins with closed windows, from sources both internal and external.

- Further work could include detailed assessment of the determinants of air leakage and examination of its specific pathways in automobiles. The efficacy of remedial measures to reduce leakage in less air tight vehicles, while preventing excessive occupant generated $CO_2$ accumulation, could also be assessed.

## Acknowledgements

Luke Knibbs gratefully acknowledges Macquarie University for providing a MQRES scholarship and postgraduate research fund grant that partially funded this research. We would like to thank Associate Professor Mark Luther of Deakin University for the loan of equipment. Professor Lidia Morawska, of the International Laboratory for Air Quality and Health at Queensland University of Technology, is thanked for her continued interest in this project. We greatly appreciated the useful discussions provided by Associate Professor Geo Clausen of the International Centre for Indoor Environment and Energy, Technical University of Denmark. Many thanks to Jenny Spagnolo of Macquarie University for assistance with data logger programming. During the preparation of this paper, Luke Knibbs was a guest PhD student at the International Centre for Indoor Environment and Energy, Technical University of Denmark. He thanks all the staff and students of this facility for making his visit enjoyable and productive. We thank the two anonymous reviewers for their useful comments.

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# Chapter 5

# Effect of Ventilation Rate on Ultrafine Particle Concentration Inside Automobiles

This chapter describes the investigation and quantification of the influence of ventilation rate (presented in chapter 4) on UFP intrusion into five test vehicles (the 2000 Subaru Liberty was not assessed). Also investigated is the relationship between on-road UFP concentrations in the M5 East tunnel and in-cabin UFP concentrations for the group of test vehicles. In this respect, this chapter is the integration of data presented in chapters 3 and 4, in addition to some new data presented for the first time. In-cabin UFP exposures during tunnel travel are quantified in terms of different ventilation settings and vehicles A simple mathematical model is employed to predict the average UFP exposure of vehicle occupants during tunnel travel. The contribution of morning and evening peak hour travel through the M5 East tunnel to total daily UFP exposure is estimated for tunnel users and related to vehicle type and ventilation setting. The results presented here are the first estimates of the effect tunnel travel on daily UFP exposure, as far as I am aware. In addition, this is only the second study to-date that quantifies the role of ventilation rates in determining UFP exposures during travel in passenger vehicles and models in-cabin concentrations, and the first to assess older vehicles and vehicles without HVAC filters, which are still common in many locations.

## Contribution

The sampling approach and protocols were developed by me in consultation with Richard de Dear and Lidia Morawska. I executed the sampling campaign and was responsible for all data collection in the field and subsequent processing. The analyses described in this paper were undertaken by me, following consultation with Richard de Dear and Lidia Morawska. Both co-authors assisted with data interpretation. I was responsible for writing the draft manuscript and producing all figures and tables. The co-authors then commented on the draft, and appropriate amendments were made. The manuscript was submitted to *Environmental Science & Technology* in June, 2009, and is currently (October, 2009) under review.

# Effect of ventilation rate on ultrafine particle concentration inside automobiles

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## Abstract

We alternately measured on-road and in-vehicle ultrafine (<100nm) particle (UFP) concentration using 5 passenger vehicles comprising an age range of 18 years. A range of ventilation settings were assessed during 301 trips through a 4km road tunnel in Sydney, Australia. We quantified ventilation rates under these settings using tracer gas techniques. Considerable variation in median in-cabin/on-road (I/O) UFP ratios was observed (0.10 to >1.0) as a consequence of ventilation setting, filter presence and vehicle type/age and air-tightness. Based on data spanning all test vehicles and ventilation settings, a strong linear relationship ($R^2 = 0.82$) existed between measured outdoor air flow and median I/O UFP ratio. UFP concentrations recorded in-vehicle during tunnel travel were significantly higher than those reported by comparable studies performed on open roadways. A simple mathematical model afforded the ability to predict tunnel trip average in-cabin UFP concentrations with reasonable accuracy. We estimate that for the vehicles and ventilation conditions assessed in our study location, 1.1% to 22% of road tunnel user's daily UFP exposure could be contributed by <0.25h of tunnel travel. This article presents extensive new data to compliment the limited amount currently available.

## Introduction

The exposure of humans to ultrafine (<100nm) particles (UFPs) is increasingly thought to be implicated, via a range of mechanisms, in undesirable acute and chronic health effects (1). The comparatively large surface area presented for a small mass of UFPs (2) carries with it the potential for adsorption of toxic components (1). The deposition efficiency of UFPs in the human respiratory tract is substantial (3), and laboratory studies of animals are suggestive of UFP translocation across the blood-brain barrier and into the central nervous system (4). The balance of evidence to-date suggests UFPs have a role in adverse human health outcomes. Accordingly, the quantification and assessment of UFP concentrations and determinants in a wide range of environments is integral, in order to better understand human exposures, guide future health-based studies and afford retrospective assessment of those conducted previously.

Fuel combustion is the primary source of UFPs in most locations (5), and due to its proximity to this source, the automobile cabin represents a microenvironment where peak UFP exposures are likely to occur. It has been estimated that up to 50% of daily UFP exposure for Los Angeles residents can occur inside automobiles during commuting activities that comprise only approximately 6% of daily time (6, 7). The increasing cognisance of this nascent issue is evident by the number of recent studies addressing its various aspects (6-18). Despite the recent advances and efforts described above, only a modicum of work to-date has addressed the role of cabin ventilation rates in the relationship between on-road UFP concentrations and those inside automobile cabins. As such, to supplement and bolster the existing data, the primary aims of this study were to; 1) assess the relationship between on-road and in-cabin UFP concentrations under a range of ventilation settings in a varied fleet of passenger vehicles; 2) quantify ventilation rates under the settings used; 3) assess their influence on in-cabin UFP levels and; 4) integrate the two key measured parameters into a simple predictive tool. We directed our study towards the road tunnel environment, as this location has been identified as one where peak UFP exposures may occur for vehicle occupants (see 21).

## Experimental Methods

### Vehicles and Ventilation Conditions

Based on the 2006 Australian Motor Vehicle Census (19), a group of five passenger vehicles generally representative of the overall stock was selected. The vehicles encompassed an age range of 18 years and are described in Table 1. Two of the vehicles (2005 VW Golf and 2007 Subaru Outback) were equipped with their original factory-fitted Heating, Ventilation and Air Conditioning (HVAC) system pollen filters. The manufacturers of these two vehicles advised that recirculated air was not filtered, which is a common configuration in many passenger vehicles (11). None of the vehicles were affected by exhaust system or firewall leaks. Four ventilation conditions were assessed (see 20),: (A) - recirculation off, fan at lowest setting, air conditioning on; (B) - same as (A), except fan at second-highest setting; C) - same as (A), except recirculation on; (D) - same as (C), except fan off and all internal air vents closed, where possible.

### UFP Measurements

We selected the 4km twin-bore M5 East road tunnel in Sydney, Australia as the primary study location. Approximately 93,000 vehicles per day use the tunnel, of which ~7% are heavy diesel vehicles. A more detailed description of the tunnel's characteristics is given in Knibbs et al. (21). Particle concentration measurements were performed using a TSI 3007 Condensation Particle Counter (CPC) that sampled every second. Sampling points were sited immediately upstream (i.e. outside of the vehicle next to the HVAC intake) and downstream (i.e. proximate to the in-cabin air vents) of the HVAC system of each vehicle. A valve fitted with conductive tubing was controlled by a datalogger to enable alternate (every 20 or 25s) measurements from each sample point, of which the final 10s were retained for analyses, to account for sample clearance. It was necessary to equip the CPC with a dilution system (22) that facilitated operation within its specifications when challenged with very high UFP concentrations. The presence of sub-isokinetic sampling is unlikely to have influenced the results substantially, given the typical size distribution of particles measured on-road (6, 12). Particle loss to the sampling system was determined in laboratory tests and used to correct data collected in the field (21). Information regarding the CPC is provided in supporting information. Trips through both bores of the study tunnel were performed in each sampling vehicle, spanning a range of times. Sampling was performed during rain-free periods from May through July, 2006, with supplemental measurements conducted in February, 2008. Three hundred and one successful tunnel trips were completed of 306 attempted. Additional data collection was performed on above ground roadways (21). Throughout the measurement campaign, the cabin of the test vehicle was occupied by one person.

**Table 1**. Characteristics of study vehicles. Modified from Knibbs et al. (20).

| Automobile | Mazda 121 | Mitsubishi Magna | Toyota HiLux | Volkswagen Golf | Subaru Outback |
|---|---|---|---|---|---|
| Model year | 1989 | 1998 | 2005 | 2005 | 2007 |
| Chassis type | Small Hatchback | Large Sedan | Utility | Large Hatchback | Mid-Size Station Wagon |
| Odometer (km) | 159,253 | 137,884 | 11,252 | 17,160 | 10,907 |
| HVAC filter | No | No | No | Yes | Yes |
| Operable vents | No | Yes | Yes | Yes | Yes |
| AC | No | Yes | Yes | Yes | Yes |
| Est. surface area ($m^2$) | 16.08 | 19.41 | 18.52 | 23.50 | - |
| Est. volume ($m^3$) | 3.32 | 3.72 | 3.33 | 3.88 | 4.43 |
| Est. S/V ratio ($m^{-1}$) | 4.84 | 5.22 | 5.56 | 6.06 | - |

#### Ventilation and Additional Measurements

Sulphur hexafluoride ($SF_6$) tracer gas techniques were used to quantify outdoor air ventilation rate ($m^3\ h^{-1}$ or air changes per h) in each study vehicle under all ventilation conditions at 0, 60 and 110 km $h^{-1}$. Vehicle interior volume was also estimated using tracer gas methods to permit conversion between the two units of measurement used in ventilation tests, which depended on the condition being assessed. A comprehensive treatment of all ventilation measurements is given in Knibbs et al. (20). Interior surface area was estimated for all vehicles (excluding the 2007 Subaru Outback) using manual measurement. Temperature and relative humidity inside and outside of the study vehicles was recorded throughout the UFP sampling campaign.

#### Analyses

UFP ingress into each vehicle for a given ventilation condition was determined by pairing individual 10s average outside and in-vehicle UFP measurements collected in the bores of the tunnel, and, in some cases, on a mixed route comprising above ground major arterial roads and tunnels (21) . The overall median inside/outside (I/O) UFP ratios were obtained from these. It should be stated that an I/O ratio is not necessarily equivalent to a particle penetration factor (23). Given the automobile environment investigated in this study and our experimental approach, the following descriptions define more precisely the I/O measurements we performed for each ventilation setting investigated: under conditions (A) and (B) we measured the penetration of UFPs through the HVAC system pathways and filter (where fitted); under condition (C) we measured the combined effect of the penetration of UFPs contained in air infiltrating through HVAC system pathways and filter (where fitted), combined with the effect of mixing with HVAC return air; under condition (D) we measured penetration of UFPs contained in infiltrating air. It was assumed that air infiltrating under condition (D) did so via the HVAC system, specifically, the recirculation damper (20), and was therefore subject to the effects of the HVAC ducting, components and filters.

Analyses of ventilation measurements are described in Knibbs et al. (20). A simple mathematical model assuming uniform mixing (24) was employed to predict in-cabin UFP levels. For practicality, the model was employed to predict tunnel trip average in-cabin UFP concentration, based on the corresponding value outside of the vehicle (i.e. in-tunnel). The value output by the model was C(t), which we took to be representative of the average in-cabin concentration during a tunnel trip, taking into account the large variability often present in on-road UFP concentrations (see 21). Particle processes (deposition, coagulation etc.) were not included in the model. Of the 301 successful tunnel trips, 30 validation cases were selected using a random number generator and excluded from calculations of particle ingress to ensure the veracity of model output. The model was parameterised with data described here in addition to that presented by Knibbs et al. (20, 21).

The general solution to the model was as follows (24):

$$C(t) = \frac{C_{O/A}[Q_{O/A} \cdot (1-\varepsilon_{S/A}) + Q_{INF}] + G}{Q_{O/A} + Q_{EXF} + Q_{R/A} \cdot \varepsilon_{S/A}}$$

$$\times[1 - \exp(-\frac{Q_{O/A} + Q_{EXF} + Q_{R/A} \cdot \varepsilon_{S/A}}{V} t)]$$

$$+ C_0 \cdot \exp(-\frac{Q_{O/A} + Q_{EXF} + Q_{R/A} \cdot \varepsilon_{S/A}}{V} t)$$

Where C(t) is particle concentration at time (t) (p $cm^{-3}$); $C_{O/A}$ is particle concentration in outdoor air (p $cm^{-3}$); $Q_{O/A}$, $Q_{INF}$, $Q_{EXF}$ and $Q_{R/A}$ are the flow rates of outdoor, infiltration, exfiltration and return air, respectively ($m^3$ $s^{-1}$); $\varepsilon_{S/A}$ is the supply air filtration efficiency of an air-handling system (-); G is the generation rate of particles due to the occupants (p $s^{-1}$); V is the volume of the space ($m^3$) and t is time (s). Many of these parameters were not required for our predictions. A description of the assumptions underlying each parameter we used is provided in supporting information. It was assumed that no internal UFP sources were active (i.e. parameter G was set to 0).

## Results and Discussion

Tunnel trips took from 3 to 26.4 minutes to complete, depending on traffic conditions. An average of 17 tunnel trips was completed per ventilation setting in each vehicle. Interior surface area to volume estimates for all vehicles except the 2007 Subaru Outback are provided in Table 1. In the 1989 Mazda 121 and 2005 VW Golf, the partition between the cabin and trunk was removed during volume and surface area estimates, therefore the estimates of these parameters presented in Table 1 include the trunk. These estimates were not required for our model, however, a brief discussion of our results is provided in supporting information. On-road UFP concentrations during the sampling campaign, in addition to in-vehicle and on-road temperature and relative humidity measurements, are described in Knibbs et al. (21).

### I/O UFP Ratios

Figure 1 presents box-and-whisker plots of I/O UFP ratios for each ventilation condition and vehicle, based on the paired 10s readings. Also presented are the flow rates of outdoor air for each ventilation condition at the average vehicle speed maintained during measurements. UFP ingress under setting (D) was not assessed in the 1998 Mitsubishi Magna or 2005 Toyota HiLux. There was a substantial reduction in the median proportion of on-road UFPs entering the cabin of the test vehicles under conditions (C) and (D) when compared to conditions (A) and (B). The presence of median I/O ratios exceeding 1.0 under condition (B) reflected the nature of the sampling methodology and the high variability in on-road UFP concentration over time periods less than the 20-25s valve switching interval used when sampling. Nevertheless, the relative relationship between ventilation modes and vehicles in terms of UFP penetration should be largely preserved. The two lowest median I/O ratios (0.72 and 0.80) under condition (A) were recorded in the 2005 VW Golf and 2007 Subaru Outback, respectively, both of which were fitted with filters. Similarly, under condition (B), these vehicles featured the lowest median I/O ratios (0.83 in the VW Golf, 0.92 in the Subaru Outback). The increase in UFP ingress under condition (B) is attributable to the observed increase in outdoor air flow, and thus filter face velocity, which results in a decreased UFP filtration efficiency (12, 25). Under conditions (A) and (B), the highest median I/O values were observed in the unfiltered and non air-conditioned 1989 Mazda 121 (1.01 and 1.27, respectively), although the use of air conditioning has previously been reported as having no apparent effects on UFP ingress into vehicles (6). Median UFP I/O ratios recorded in the 1998 Mitsubishi Magna and 2005 Toyota HiLux were 0.92 and 0.97, respectively, for ventilation setting (A), and 1.21 and 1.13, respectively, under setting (B).

Yokoyama et al. (26) reported an I/O UFP ratio under a fresh air intake setting of 1.09, for their otherwise unspecified, compact (i.e. hatchback) test vehicle. This is generally in accord with the results presented here. Zhu et al. (6) measured average on-road I/O UFP ratios between approximately 0.3 and 0.7 under a fresh air intake and low-medium fan speed condition in three filter-fitted vehicles up to 5 years old at the time of their study. Pui et al. (11) reported an average on-road UFP penetration efficiency of 54% in a 2003 model Saab 93 fitted with HVAC filtration, under fresh air intake setting and medium fan speed. Qi et al. (12) measured average on-road UFP particle penetration in the same model vehicle of 45% and 34.4% with fresh air intake on for high and medium fan speeds, respectively. Qi et al. (12) also found that particle penetration into their vehicle's cabin measured using number concentration (p $cm^{-3}$) and surface area of particles depositing in the human lung ($\mu m^2$ $cm^{-3}$) resulted in similar values. The same study assessed the protection offered by the test vehicle's HVAC system alone (i.e. with the filter removed) to on-road UFP penetration. When in-cabin and outdoor temperatures did not differ significantly, they found average penetration of 92.1% and 89.7%, for their respective medium and high fan speed conditions. The studies of Zhu et al. (6), Pui et al. (11) and Qi et al. (12) all employed simultaneous measurement of UFPs inside and outside of their test vehicles, and this is likely explanation of the lower UFP penetration values they recorded compared to the present study.

Under ventilation condition (C), substantial reductions in UFP intrusion were observed in all vehicles. The 2005 VW Golf and the 2005 Toyota HiLux, permitted the lowest proportion of outdoor UFPs into the cabin. The respective

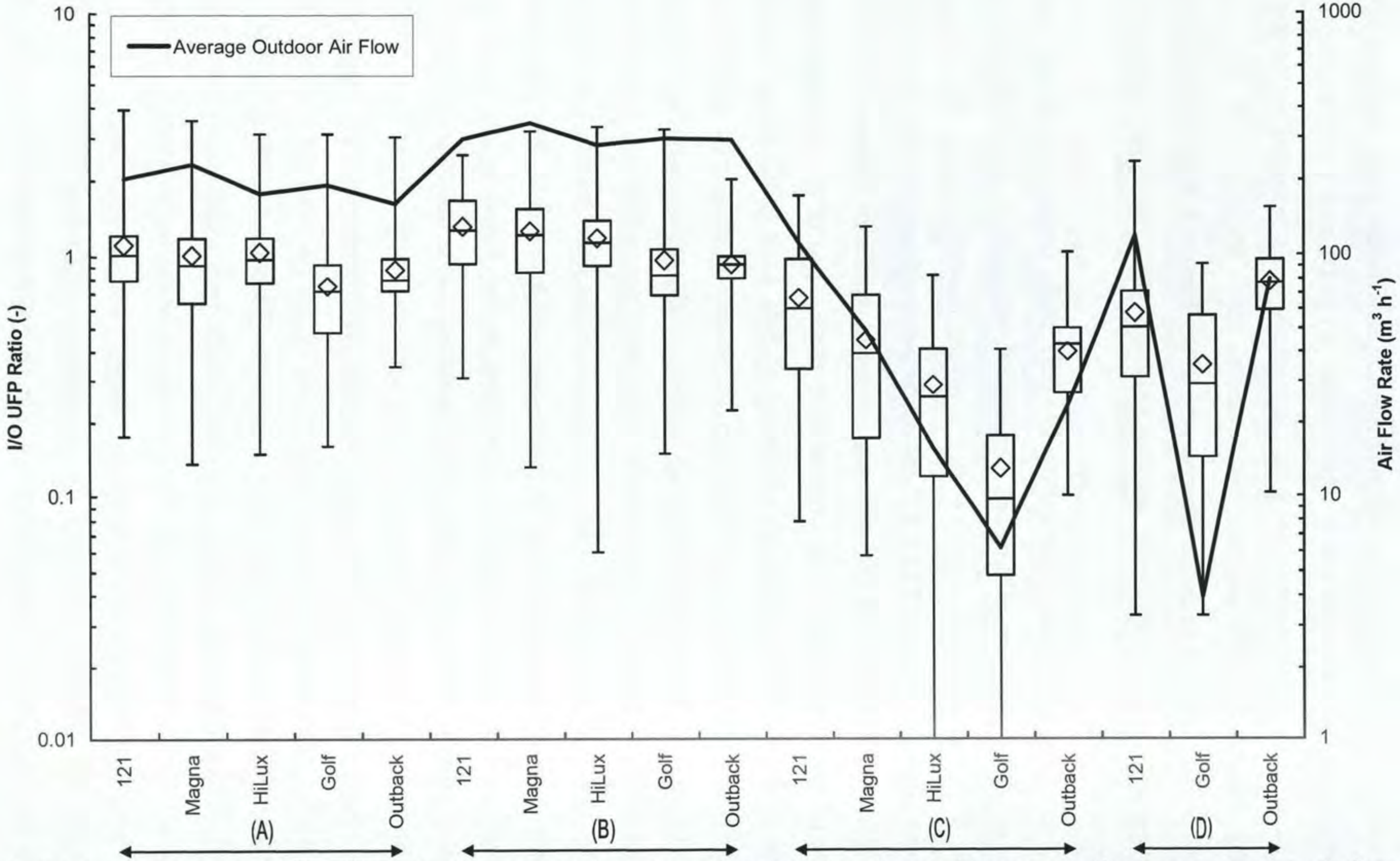


**Figure 1**. Box-and-whisker plot of I/O UFP ratios for each vehicle and ventilation setting, shown with corresponding average outdoor air flow rate. Thick horizontal line, diamond, lower and upper edges of box, lower and upper extent of whiskers denote median, average, 25th and 75th percentile, minimum and maximum, respectively.

median I/O ratios for these vehicles were 0.10 and 0.26. A median I/O value of 0.39 was recorded in the 1998 Mitsubishi Magna. Despite the presence of a filter in the 2007 Subaru Outback, its median I/O ratio was 0.43. Many measurements performed in the Subaru Outback were collected in the mid-late evening when average in-tunnel UFP concentrations were generally lower (median = $9.38 \times 10^4$ p $cm^{-3}$) than those present during measurements performed in other vehicles (median = $2.58 \times 10^5$ p $cm^{-3}$). This is likely to have resulted in the background in-cabin concentration becoming more significant under setting (C), and hence inflating the I/O ratio. As Figure 1 highlights, I/O ratios generally decreased with decreasing infiltration under condition (C). The highest median I/O ratio of 0.61 was measured in the least air-tight vehicle (1989 Mazda 121).

Zhu et al. (6) measured on-road average I/O UFP ratios under a recirculate setting between approximately 0.10 and 0.60 in their test fleet of three post-2000 model filtered vehicles. Our results generally agree with theirs, despite the presence of substantially older, vehicles lacking HVAC filters in our test group. However, following the conclusion of the sampling campaign, the air intake of the Mazda 121 was observed to contain a substantial amount of leaf detritus as a consequence of being parked outdoors, which may have afforded a minor reduction in particle penetration for a subset of the particle size range investigated. It should be noted that the two newest vehicles assessed by Zhu et al. (6) resulted in average I/O values of <0.2, which is largely in accord with our measurements in the 2005 VW Golf and 2007 Subaru Outback. Yokoyama et al. (26) measured a UFP I/O ratio of 0.44 when recirculating air in their test vehicle. Pui et al. (11) estimated that the UFP reduction when recirculating air in a 2007 Toyota Camry attributable to non-filter processes was 19.1%. Thus, for most vehicles, given a situation where recirculation had been in effect for several minutes, it is likely that return air would be characterised by a relatively low UFP concentration and subsequently act as a diluent on infiltrating air. Qi et al. (12) reported significant reductions in the surface area of UFPs depositing in the lung measured inside vehicles after operating the ventilation system in recirculation mode. The time taken for this reduction to occur was less in the vehicle where return air was filtered, which was also reported by Pui et al (11) with respect to particle number concentration. Our measurements under setting (C) are in good agreement with the limited number of comparable studies. This reflects the reduced influence that the absence of simultaneous in-vehicle and outdoor UFP measurements in our study had under this ventilation setting.

Under condition (D), median I/O UFP ratios varied significantly across the three vehicles assessed. For the 2005 VW Golf and 2007 Subaru Outback, the respective median I/O values of 0.29 and 0.77 were substantially higher than those measured under setting (C) for each vehicle. This is possibly a reflection of the absence of effects associated with the aforementioned mixing of infiltrating air with return air. This is supported by the observation of a relatively minor change in median I/O ratio measured under setting (D) (0.51) compared to that measured under (C) (0.61) in the 1989 Mazda 121, where infiltrating air is likely to constitute a significant proportion of supply air under setting (C) at the typical vehicle speeds maintained in-tunnel.

### Relationship Between Ventilation Rate and I/O UFP Ratio

Figure 2 shows the result of linear regression applied to average outdoor air flow and median I/O UFP ratio measured for all vehicles and ventilation settings. There is a highly significant ($p < 0.0001$) linear relationship between these variables. The ventilation rate of the cabin with outdoor air under all ventilation conditions in the vehicles assessed is a very good ($R^2 = 0.82$) determinant of median I/O UFP ratio. For our test vehicles, outdoor air ventilation rate was evidently a dominant process governing UFP intrusion into their cabins. Xu and Zhu (16), based on field measurements (6) and subsequent modelling, reported a positive correlation between vehicle HVAC system mechanical air flow rate and in-cabin I/O ratio of 20-80nm particles (although the gradient of the relationship varied with particle size), at air flow rates above 108 $m^3\ h^{-1}$ under a ventilation setting similar to our (A) and (B) conditions. Our data for these conditions, as Figure 2 shows, generally support this finding, although our air flow rate data collected under the (A) and (B) settings reflected varying degrees of infiltration through the HVAC system of the test vehicles as a consequence of vehicle speed, in addition to mechanical air flow delivered by the HVAC fan (20). Under a recirculate condition, Xu and Zhu (16) found I/O ratios of 20-80nm particles decreased with an increasing mechanical air flow rate, however, they found a positive correlation between vehicle speed (and thus infiltration rate) and in-cabin I/O ratio for the same particle size range. The outdoor air flow data depicted in Figure 2 for the (C) and (D) settings were a measure of infiltration, rather than mechanical air flow, and as such, also largely support the conclusions of Xu and Zhu (16) with respect to the significance of ventilation parameters as determinants of in-vehicle UFP concentration. The relative roles of other (non-ventilation) parameters are investigated in the work of Xu and Zhu (16).

**Model Performance**

Figure 3 shows the results of applying the model to predict in-cabin average UFP concentration during a tunnel trip. The 30 randomly selected trips used for model validation encompassed all test vehicles and ventilation modes. Given the simplicity of the model, the results depicted in Figure 3 indicate a reasonably good agreement between measured and predicted average in-cabin UFP concentrations over 2 orders of magnitude. A 1:1 line has been plotted on Figure 3 and the results obtained appear generally proximate to it. For measured in-cabin UFP concentrations below approximately $2.50 \times 10^5$ p cm$^{-3}$, there is a roughly equivalent number of data points falling either side of the 1:1 line, suggesting no obvious systematic over or under-prediction in this range for the scenarios investigated. Above this concentration, the model typically under-predicted UFP levels, although performance remained quite sound. It was not an aim of this study to investigate the factors underlying the performance of the model. The results presented here largely confirm its applicability to the vehicle cabin microenvironment. It is worth noting that a more comprehensive (in terms of input parameters) approach to modelling in-vehicle UFPs was adopted by Xu and Zhu (16), with good results. Given a roadway or tunnel where average on-road UFP concentration can be reliably characterised as a function of heavy diesel vehicle volume (e.g. 7, 21) or other key variable, the application of the model described here, or that of Xu and Zhu (16), parameterised with relevant

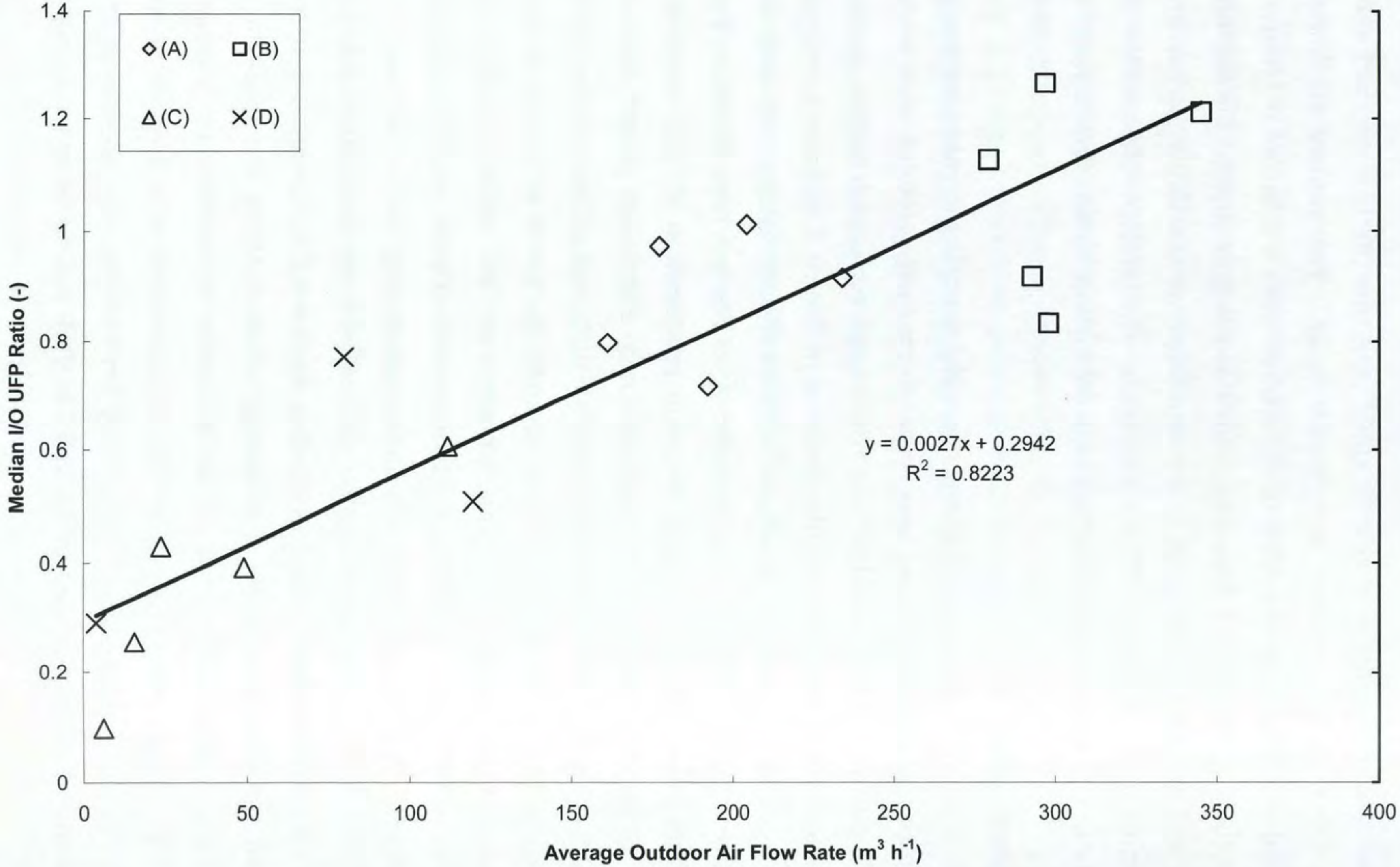


**Figure 2.** Average outdoor air flow rate vs. median I/O UFP ratio, incorporating all vehicles and ventilation settings.

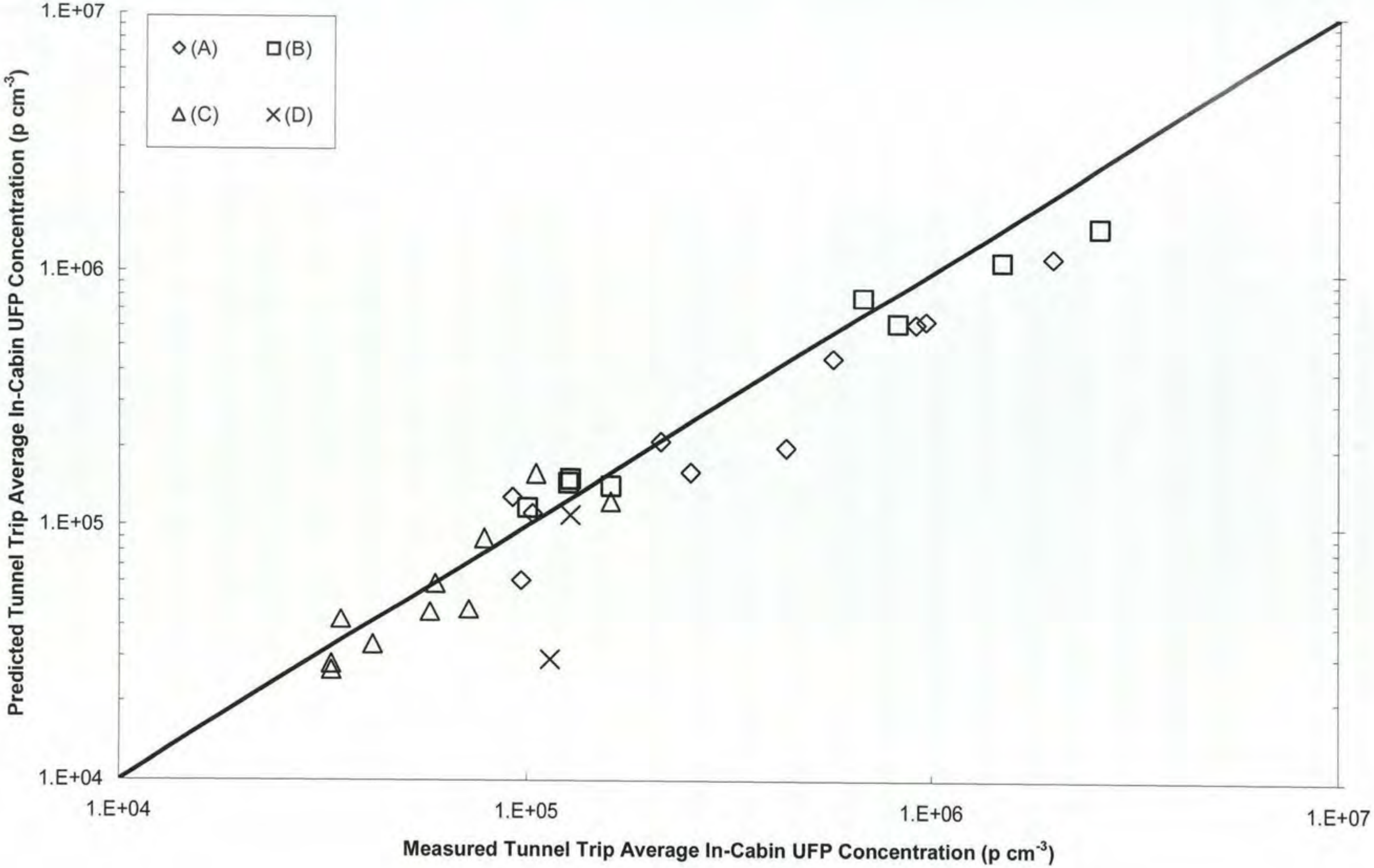


**Figure 3.** Measured and model-predicted in-cabin tunnel trip average UFP concentrations for 30 randomly selected validation cases.

data (6, 12, 17, 20, 27) could provide an effective means to better understand and mitigate in-vehicle UFP exposures.

### In-vehicle UFP Concentration Measured During Tunnel Travel

Table 2 presents summary statistics describing UFP concentrations measured inside each vehicle when travelling through the M5 East road tunnel. It is difficult to directly compare the data presented in Table 2, as a considerable range of in-tunnel conditions prevailed during the measurements (21). The table is therefore presented as an indicative guide. The UFP concentrations measured inside our group of study vehicles were typically substantially higher than those reported by several other in-cabin UFP studies (9, 10, 13, 14, 18).

For comparison, Zhu et al. (6) recorded average in-vehicle UFP concentrations in their group of three relatively new (at the time of their study) filter-equipped vehicles ranging from $8.5 \times 10^4$ p $cm^{-3}$ to $2.39 \times 10^5$ p $cm^{-3}$ and $1.50 \times 10^5$ p $cm^{-3}$ to $2.56 \times 10^5$ p $cm^{-3}$ on Los Angeles freeways characterised by a respective diesel vehicle usages of 5% (total traffic flow 13,860 vehicles $h^{-1}$) and ~25% (total traffic flow 12,180 vehicles $h^{-1}$ ). These concentrations reflect data averaged over three different ventilation settings (2 non-recirculate and 1 recirculate). Although not shown in Table 2, the comparable range (i.e. excluding setting (D) measured inside our vehicles was from $1.41\times 10^5$ p $cm^{-3}$ (2007 Subaru Outback) to $5.08 \times 10^5$ p $cm^{-3}$ (1989 Mazda 121). These measurements, from our tunnel study featuring ~7% heavy diesel vehicles and a comparatively low traffic flow rate, were roughly double the equivalent concentrations recorded by Zhu et al. (6) for their 5% diesel study roadway (405 Freeway). This increase is unremarkable given it largely reflects both the difference in study location, and the greater range of vehicle ages (and reduced air-tightness/lack of filter presence) assessed in our study. Had our study utilised the same CPC as that employed by Zhu et al. (6), which had a lower particle size detection limit (5nm) than our CPC (10nm), it is likely the magnitude of the increase noted above would have been greater. Figures 4a and 4b show on-road and in-cabin UFP concentration for all tunnel trips performed in the 2005 VW Golf under settings (C) and (A), respectively. The dampening of in-vehicle UFP levels (6), relative to

**Table 2.** Summary of in-vehicle UFP concentrations measured for each vehicle and ventilation setting during tunnel travel.

| Automobile | Ventilation Condition | Mean (p cm$^{-3}$) | S.D. (p cm$^{-3}$) | Median (p cm$^{-3}$) | Min. (p cm$^{-3}$) | Max. (p cm$^{-3}$) |
|---|---|---|---|---|---|---|
| 1989 Mazda 121 | (A) | 7.02E+05 | 1.17E+06 | 3.51E+05 | 7.63E+04 | 7.45E+06 |
| | (B) | 6.60E+05 | 1.21E+06 | 3.30E+05 | 8.28E+04 | 8.01E+06 |
| | (C) | 1.63E+05 | 1.40E+05 | 1.06E+05 | 8.10E+04 | 8.49E+05 |
| | (D) | 1.15E+05 | 5.64E+04 | 8.72E+04 | 8.10E+04 | 3.40E+05 |
| 1998 Mitsubishi Magna | (A) | 4.83E+05 | 5.33E+05 | 3.61E+05 | 6.94E+04 | 3.44E+06 |
| | (B) | 7.90E+05 | 1.08E+06 | 4.32E+05 | 8.24E+04 | 6.09E+06 |
| | (C) | 1.02E+05 | 7.88E+04 | 8.17E+04 | 5.53E+04 | 6.12E+05 |
| 2005 Toyota HiLux | (A) | 4.69E+05 | 7.68E+05 | 1.36E+05 | 4.68E+04 | 3.82E+06 |
| | (B) | 3.60E+05 | 5.09E+05 | 1.49E+05 | 6.84E+04 | 2.91E+06 |
| | (C) | 5.28E+04 | 1.37E+04 | 5.02E+04 | 3.30E+04 | 9.31E+04 |
| 2005 Volkswagen Golf | (A) | 2.82E+05 | 2.52E+05 | 1.78E+05 | 3.15E+04 | 1.02E+06 |
| | (B) | 6.11E+05 | 8.02E+05 | 1.69E+05 | 8.10E+04 | 3.88E+06 |
| | (C) | 3.38E+04 | 5.20E+03 | 3.26E+04 | 2.72E+04 | 5.47E+04 |
| | (D) | 4.36E+04 | 7.73E+03 | 4.69E+04 | 3.12E+04 | 5.52E+04 |
| 2007 Subaru Outback | (A) | 1.27E+05 | 1.05E+05 | 9.13E+04 | 4.23E+04 | 5.32E+05 |
| | (B) | 2.57E+05 | 3.75E+05 | 9.99E+04 | 6.33E+04 | 1.71E+06 |
| | (C) | 4.02E+04 | 7.33E+03 | 3.79E+04 | 3.19E+04 | 6.34E+04 |
| | (D) | 6.37E+04 | 1.42E+04 | 6.12E+04 | 3.05E+04 | 8.66E+04 |

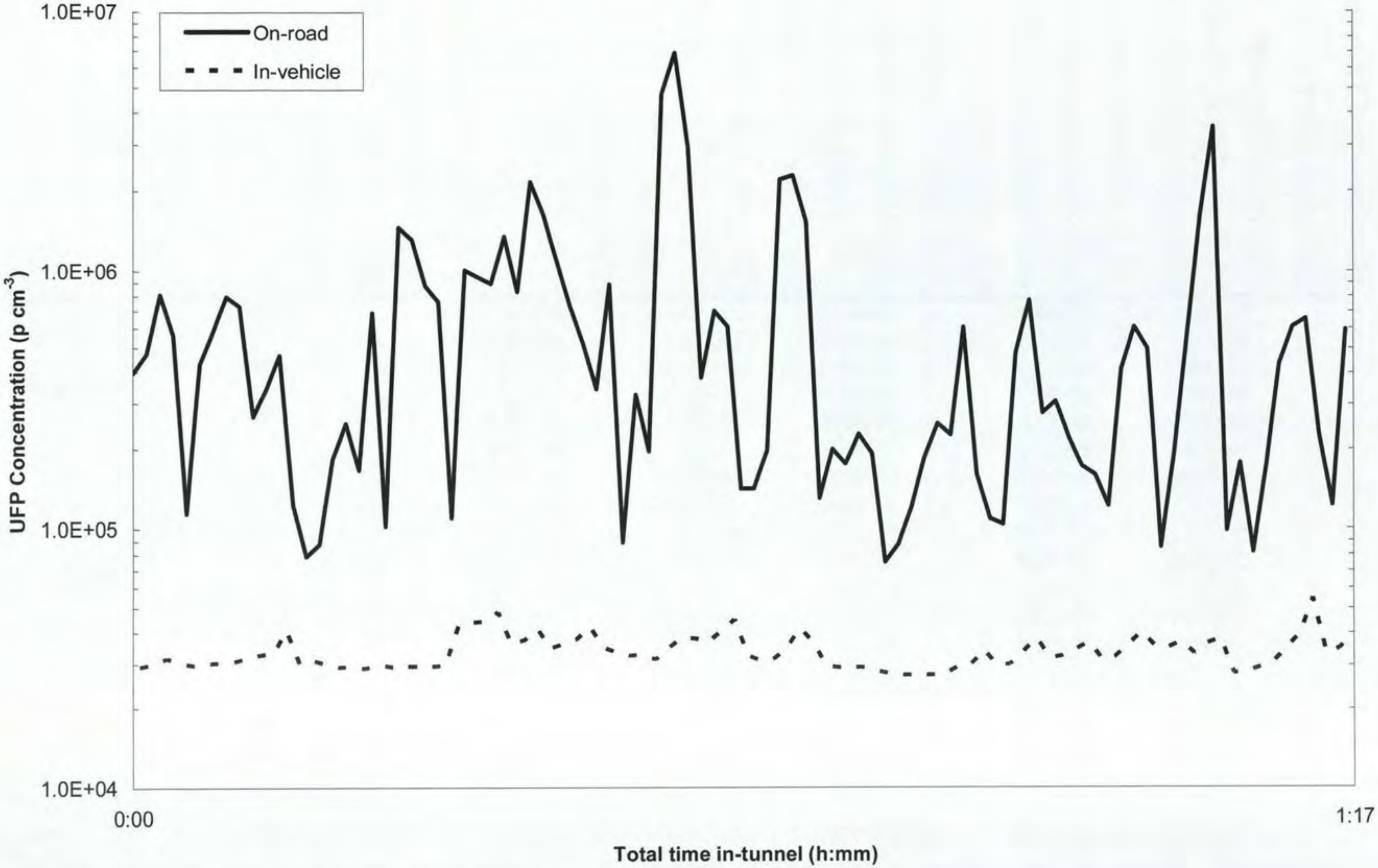


**Figure 4a**. In-vehicle and on-road UFP concentration for all tunnel travel in the 2005 VW Golf for setting (C) (recirculation).

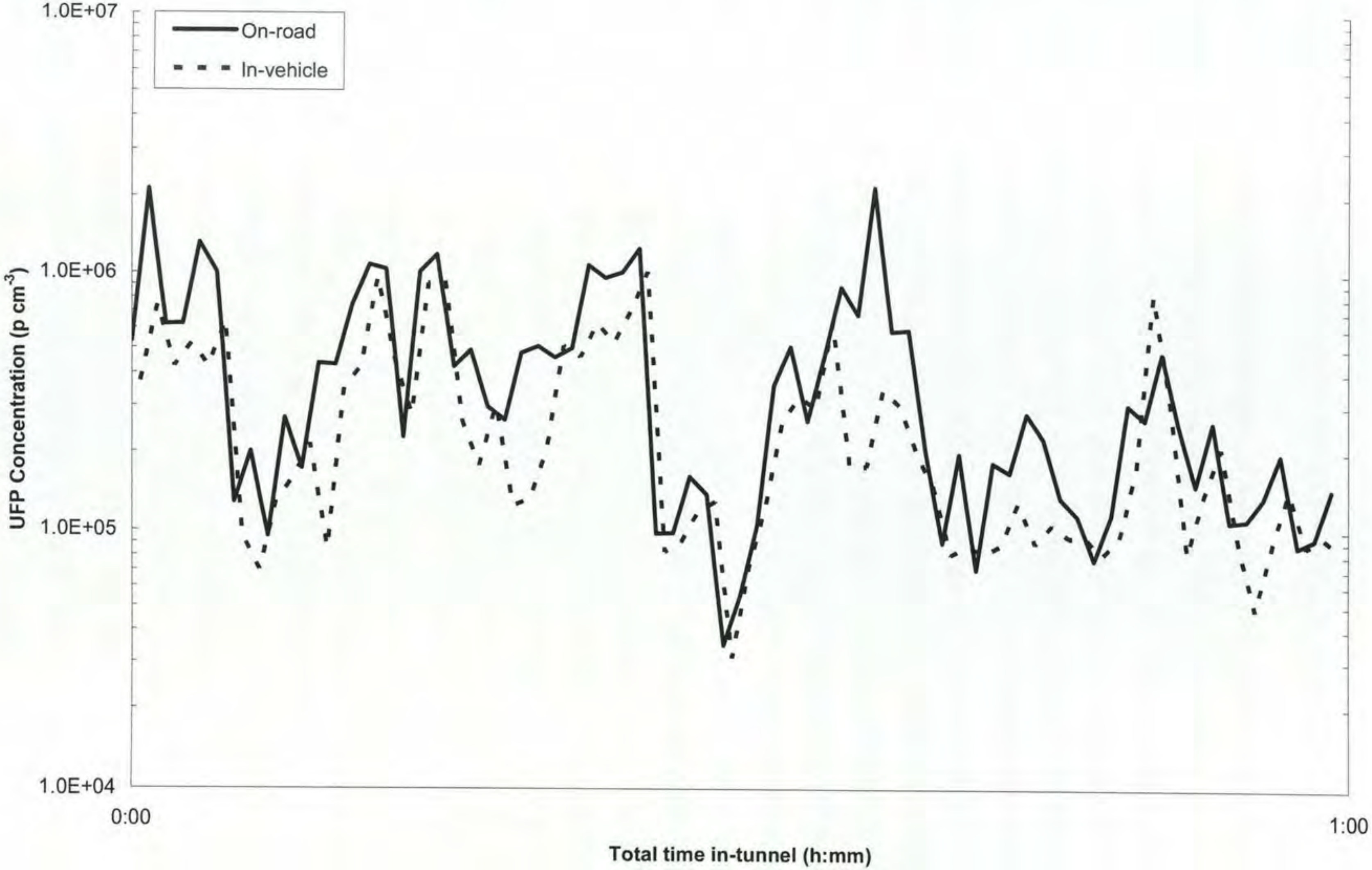


**Figure 4b**. In-vehicle and on-road UFP concentration for all tunnel travel in the 2005 VW Golf for setting (A) (fresh air intake).

those measured on-road, afforded by the use of recirculation is apparent in Figure 4a. Conversely, the extent of this effect was greatly reduced under setting (A), as highlighted by Figure 4b.

**Potential Contribution of In-vehicle UFP Exposures Incurred During Tunnel Travel to Overall Daily Exposure**

To estimate the influence of road and tunnel travel on Sydney tunnel user's UFP exposure, we employed our model to predict average in-cabin UFP concentrations inside the 1989 Mazda 121 and 2005 VW Golf during a eastbound morning (8-9 am) and westbound evening (5-6 pm) trip through the M5 East tunnel under both the (B) and (C) ventilation settings. The I/O values we measured were used for simulations involving the (C) setting, whilst for those addressing the (B) setting, we used values reported by Qi et al. (12) for comparable conditions (i.e. high fan speed, variable filter presence, non-winter conditions). The UFP concentration in outdoor air (i.e. parameter $C_{O/A}$) was taken as the hourly median trip average in-tunnel UFP concentration reported by Knibbs et al. (21) for the two respective time periods modelled. The results were combined with the averages presented by Fruin et al. (7) in their exposure log. In order to accommodate tunnel travel into the daily activity log, it was altered such that 0.25 h was spent on freeway and tunnel travel combined, during both morning and evening commutes. Fruin et al. (7) designated the total time spent in-vehicle was 1.5h $day^{-1}$, which is very similar to that of many Sydney commuters (28). Trip time for the morning and evening tunnel trips was calculated as the average time taken for all transects during these periods throughout the data collection campaign (3m 47s and 10m 10s, respectively). The measurements reported by Fruin et al. (7) were of on-road UFP concentration, rather than in-cabin UFP concentration. Accordingly, the freeway on-road UFP concentrations reported by Fruin et al. (7) were substituted with the median of our measurements recorded on a Sydney mixed road route (21) and, in the absence of accurate vehicle speed data that would enable mathematical modelling, multiplied by the relevant I/O ratios to obtain an in-cabin exposure estimate. Initial in-cabin UFP concentration for the modelling cases at time = 0 was assumed to be equal to the output of the above process. As we did not have arterial roads UFP concentration data, those reported by Fruin et al. (7) were used, after being processed in the same manner as the freeway data, such that they were converted to in-cabin estimates. All other data reported by Fruin et al. (7) were left unchanged.

Under setting (B), tunnel travel inside the 2005 VW Golf and 1989 Mazda 121 respectively constituted 15.3% and 22.0% of estimated total daily UFP exposure, while the equivalent measures under setting (C) were 1.1% and 10.7%. The estimated daily in-vehicle contributions to total UFP exposure for 1.5 h of travel in Sydney in the 2005 VW Golf under the (C) and (B) settings were 4.0% and 25.7%, respectively, whilst the corresponding figures for the 1989 Mazda 121 were 24.9% and 39.2%. These are average estimates for non-smoking, non-occupationally exposed persons only, and should not be taken as authoritative or extrapolatable. However, the estimates do indicate; 1) the potentially large contribution of <0.25 h of daily M5 East tunnel travel to overall UFP exposure of Sydney commuters using this tunnel; 2) that substantial reductions in UFP exposure incurred whilst travelling in various vehicles can be achieved by using recirculation, and; 3) that this reduction can vary markedly and depends strongly upon the air-tightness of the vehicle.

This study has shown that UFP ingress into passenger vehicles is highly variable and dependent on ventilation setting (i.e. rate at which the cabin is ventilated with outdoor air), air-tightness and filter presence. The ability of a simple mathematical model to predict average in-cabin UFP exposure during travel through a road tunnel for a wide range of vehicles and ventilation settings was reasonably good, and offers an alternative, based on minimal input parameters, to more complex models. Despite the paucity of data on UFP ingress into vehicle cabins, the results of the present study generally exhibited good agreement with comparable works, with due consideration to the inherent differences in experimental and analytical methods. Further investigation of in-vehicle UFP exposures and assessment of total daily UFP exposure, particularly for persons who due to their occupation spend a large proportion of their day inside vehicles, in addition to further studies of the health effects associated with in-vehicle exposures, would seem beneficial from a public health perspective.

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## Supporting Information

Effect of ventilation rate on ultrafine particle concentration inside automobiles

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### Instrument Details

A TSI 3007 Condensation Particle Counter (CPC) was used to measure ultrafine particle (UFP) concentration. The CPC was capable of measuring particles from 10nm (50% detection threshold) to >1000nm at concentrations up to $1 \times 10^5$ p $cm^{-3}$, with an accuracy of ± 20%, and 95% response time of <9 seconds (1). Although given its detectable size range the CPC was not a true UFP counter, given the study environment and its associated pollutant source, UFPs are known from the literature to constitute the dominant size range (2). A simple dilution system (3) was implemented, and the maximum detectable concentration increased to $\sim 8.5 \times 10^6$ p $cm^{-3}$.

To achieve sufficient dilution, the sample line was partitioned to possess a low flow rate (see 3), and given the moving nature of the sampling platform, sub-isokinetic sampling was present. However, due the particle source in the study location (diesel and gasoline fuel combustion) and the associated size of particles, this was not a significant limitation. The sampling interval of the CPC was set to one second, and prior to measurements, the unit was zero count checked using the manufacturer-supplied HEPA filter. The dilution system and sampling hardware were periodically treated with clean dry air to prevent fouling. Samples were transported using Tygon® R-3603 tubing, with the sample point to CPC inlet distance varying between 0.75 to 1.1m (vehicle dependent) for the outside sampling line, and 0.72 to 0.84m (vehicle dependent) for the inside sampling line.

Total particle loss in the sampling system was assessed in the laboratory using a test chamber injected with unleaded petrol exhaust produced by 4-stroke spark ignition. A TSI 3022A CPC was used as the benchmark instrument in these tests. Following the calculation of correction factors, the TSI 3007 CPC was capable of measurement of particle concentrations up to approximately 8.9 ×

$10^6$ p $cm^{-3}$. The particle loss in the sampling system attributable to factors other than dilution was minor when compared to that caused as a result of dilution.

#### In-vehicle S/V Ratios

S/V ratio estimates ranged from 4.84 (1989 Mazda 121) to 6.06 $m^{-1}$ (2005 VW Golf). Manual measurement of vehicle interior surface area is a challenging task, and our results should be interpreted with this in mind. Notwithstanding this, vehicle interior S/V value presented by Jakobi and Fabian (4) of 5.0 fell within the range measured in this study. Similarly, our results fell within the range of in-vehicle S/V ratios reported by Gong et al. (5). The S/V value for a vehicle is higher than that in many other built environments, such as houses (6), and also typically include a range of surface materials, each with varying roughness. The parameter most affected by this is particle deposition, which has been recently investigated by Gong et al. (5) who reported in-cabin UFP deposition rates to be up to 20 times higher than those recorded in other indoor environments. Deposition can be an important factor affecting of in-vehicle UFP concentration under certain ventilation conditions (5, 7).

#### Model Parameters

It was assumed that $Q_{INF}$ constitutes part of the measured value of $Q_{O/A}$ under ventilation modes (A) and (B), and the entirety of the measured value of $Q_{O/A}$ under modes (C) and (D). Hence, $Q_{INF}$ was not explicitly used. For modes (A) and (B), we calculated the difference between measured air flow at the relevant vehicle speed for each validation case and that measured with the vehicle stationary and used this as the value of $Q_{EXF}$. For modes (C) and (D), $Q_{EXF}$ was set to the measured value of $Q_{O/A}$ under mode (D) (i.e. leakage air). For mode (C), $Q_{R/A}$ was not used for modelling as our measured values of UFP ingress under this setting included the effects of return air.

For each randomly selected trip, the average speed through the tunnel bore was used to calculate outdoor air flow from the linear relationship observed between vehicle speed and ventilation rate observed in each vehicle under all ventilation settings. The in-cabin UFP concentration measured immediately prior to tunnel entry was taken to represent $C_0$. The trip average in-tunnel (i.e.

on-road) concentration was entered as $C_{O/A}$. The median I/O ratio from the paired 10s sets of observations for each vehicle and ventilation mode was used for $\varepsilon_{S/A}$.

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# Section III
# Discussion and Conclusions

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# Chapter 6

# Summative Discussion and Conclusions

This thesis has taken a synergistic approach to assessing in-vehicle UFP pollution. A quantitative focus has prevailed throughout this work, underpinned by an acute awareness of the methodological considerations inherent in such an investigation. The key determinants of in-vehicle UFP exposure (i.e. on-road UFPs, vehicle cabin ventilation, UFP ingress into vehicle cabins) have been measured and brought together in an integrative and cohesive conclusion. A deliberate focus on the M5 East road tunnel has resulted in a substantial database and better understanding of UFP concentrations in this location, which previously didn't exist. Despite the local focus of the primary study location, many of the results presented here are applicable to a range of roadway environments. The following sections summarise the findings of this work as they relate to the initial objectives, discuss the limitations of the present study and suggest avenues of investigation worthy of consideration in future research endeavours.

## 6.1 Realisation of Project Objectives

### A): Quantification and Evaluation of UFP Exposure Inside Vehicles

The first objective of this project was the measurement of UFP exposure inside vehicles and evaluation of the nature of these exposures, with a specific focus on the exposures incurred during travel through the M5 East road tunnel in Sydney. The initial motivation to pursue this pathway of research was driven by identification of knowledge gaps in the literature. The objective described above was realised through the various measurement and analytical efforts described in section II.

Chapter 2 detailed the methods implemented to ensure the quality of UFP measurements. Specifically, a simple dilution system that would allow the TSI 3007 CPC to function when challenged with the often very high particle concentrations encountered in-tunnel was assembled and tested repeatedly. The system afforded an increase in the maximum detectable particle concentration of almost two orders of magnitude. Although not noted in chapter 2, I can confirm that the CPC with attached dilution apparatus performed admirably during the measurement campaign, and occasions where data quality was affected by instrument limitations or error were extremely rare. Whilst not the primary focus of this project, the assessment of the reproducibility and validity of the measurements obtained with the TSI 3007 CPC were cornerstones upon which the quality of the field sampling campaign described in chapters 3 and 5 rested. As such, chapter 2 met one aspect of objective A), with respect to the assurance of the veracity of UFP measurements.

The quantification and assessment of in-cabin UFP exposures during tunnel travel was dealt with in chapter 5. It was found that the range of average in-cabin UFP exposures incurred during M5 East tunnel travel in the five vehicle study group were approximately double those reported for a freeway with a similar traffic composition but greater volume (Zhu et al., 2007). The most obvious query arising from this finding was related to the fact that the time spent in the M5 East tunnel each day would likely be substantially lower than that spent on other roadway types. This was duly investigated in the final segment of chapter 5, where it was estimated that <0.25h (a figure selected on the basis of observed tunnel trip times during field sampling) of total daily tunnel occupancy in the M5 East could account for 1.1 to 22.0% of total daily UFP exposure, depending on the vehicle travelled in and ventilation setting used. The short duration of tunnel travel also constituted a substantial proportion of the total UFP exposure incurred during 1.5h of daily vehicle travel.

It is being increasingly reported and accepted that time spent inside vehicles can be a disproportionate contributor to total UFP exposure. However, as chapter 5 notes, during tunnel travel, a vehicle occupant may experience the highest UFP exposures present in an already high exposure environment (i.e. roadways), and the influence of vehicle characteristics and ventilation rates are significant determinants of such exposures. Prior to this study, the nature of in-cabin UFP exposures experienced by tunnel users were poorly defined, as this area of research has lagged behind other aspects of on-road particulate exposure (Kuykendall et al., 2009). However, this study has elucidated the issue, whilst identifying the need for further work in this area. As was noted in section 1.4, the characteristics of a tunnel may often preclude the extrapolation of air quality investigations performed in it to another tunnel. As such, this study, with its deliberate focus on the M5 East tunnel, has further highlighted the need for UFP exposure assessments in other tunnels worldwide, especially those with known or suspected air quality problems.

Chapters 2 and 5 described the successful achievement of objective A) of this thesis. The results obtained, with particular reference to chapter 5, are of direct relevance to the domain of pubic health practice. Many people are likely to use tunnels without compunction and consideration of the potential health effects ascribed to UFP (or other pollutant) exposure following tunnel travel. As such, tunnel operators and public health authorities should ensure appropriate measures are implemented to minimise and mitigate the exposure of tunnel users to potentially deleterious pollutants. This is especially true for persons most susceptible to the effects of such pollutants, and tunnels where the duration of travel is large due to either the tunnel's length or frequent traffic congestion. This study has provided the initial steps forward in terms of managing in-vehicle UFP exposures during tunnel travel, and some pragmatic avenues of investigation for future studies are briefly introduced in section 6.3.

**B): Investigation of the Determinants of On-road UFP Concentrations Inside the M5 East Tunnel**

As was noted previously, a wholesale understanding of in-cabin UFP exposures demands knowledge of on-road concentrations and their causes, as it is through this knowledge that effective measures can be implemented to reduce on-road UFP levels, such that in-cabin concentrations will follow. The importance of field measurements on roadway environments should not be underestimated in terms of its ability to collect data representative of conditions typical of those challenging a vehicle's ability to protect its occupants from UFPs.

The second objective of this project was quantification of on-road UFP concentrations and their determinants in the M5 East tunnel. Chapter 3 described the efforts undertaken to address this objective. Following an extensive sampling campaign involving over 300 trips through the tunnel bores, it was found that trip average on-road UFP concentrations in the M5 East encompassed two orders of magnitude ($5.53 \times 10^4$ p $cm^{-3}$ to $5.95 \times 10^6$ p $cm^{-3}$). The minimum and maximum medians of these trip averages, sorted by the hour of tunnel entry, were each one order of magnitude greater than equivalent measurements than those recorded in other environments, based on the analysis by Morawska et al. (2008) of results from 71 worldwide studies of UFP concentrations. The in-tunnel UFP concentrations were also substantially elevated, again, by an order of magnitude, compared to those measured on local (i.e. Sydney) above ground major roadways. Relative to other on-road and/or in-tunnel studies, the measurements we recorded were typically elevated by a significant margin.

The investigation into the determinants of average in-tunnel UFP concentrations was hampered slightly by the limitations of traffic volume data (a factor that was beyond the control of this study) but nevertheless yielded results broadly consistent with those reported by studies performed overseas. It was found that, in both tunnel bores, heavy diesel vehicle traffic volume was significantly better correlated with hourly median trip average UFP concentrations than passenger (i.e. unleaded gasoline-powered) vehicle traffic. This finding supported those reported by similar studies performed on other types of roadway. However, the strength of the impact of HDV volume on UFP concentration was much greater in the eastbound bore than in the westbound bore. Some speculative explanations for this were proffered in chapter 3 *vis-á-vis* potential differences in tunnel ventilation system parameters/layout, heavy vehicle cargo loading and roadway gradient between the two bores. As this was the first investigation into UFPs in the M5 East tunnel, and not the sole focus of this thesis, the roles of all possible UFP determinants were not investigated. However, this study did identify patent and relevant pathways for future work, which are summarised in section 6.3.

It would be an exaggeration to state that the work described in chapter 3 has provided a definitive understanding of all causes underlying UFP pollution in the M5 East tunnel. With its deliberate focus on assessing and parsing the key elements of UFP pollution, this study has, however, provided the first measurements of UFP concentration in the tunnel, based on an extensive sampling campaign. The findings identified the role of traffic volume on in-tunnel UFP concentrations, placed them into the context of international literature and established a baseline from which future studies may extend the knowledge of UFPs in the M5 East tunnel. Given the good agreement between the results presented here and those reported in the literature, the applicability of the results, especially in a qualitative sense (e.g. relative role of HDVs and passenger vehicles in determining on-road UFP concentrations), to other roadway environments is thought to be reasonable, with due consideration of the sometimes idiosyncratic nature of tunnels noted in section 1.3.

#### C): Systematic Investigation of Ventilation Rates Inside Passenger Vehicles

Given the importance of vehicle cabin ventilation rate in terms of the ingress of UFPs, or indeed many other pollutants, the general lack of comprehensive and systematic studies of this parameter identified a key knowledge gap requiring assessment. Accordingly, to address this issue and in the process fulfil several of the various objectives of this study, the largest known study of vehicle cabin ventilation and air-tightness to-date was performed.

Chapter 4 detailed the measurement and analyses involved in the investigation of vehicle cabin ventilation rates. A group of vehicles generally representative of those driven on Australian road was selected based on the 2006 Australian Motor Vehicle Census (ABS, 2006). A systematic approach, incorporating 4 ventilation settings and three vehicle speed conditions, was employed, and replicate concentration-decay and constant injection ventilation measurements using $SF_6$ as a tracer gas were implemented.

The most significant finding from the work described in chapter 4 was that when driven at speed, the oldest vehicle in the study group (1989 Mazda 121) permitted air exchange rates one order of magnitude higher than those measured in one of the newest vehicles assessed (2005 VW Golf) under an air recirculation or completely closed cabin (i.e. all user operable air infiltration pathways closed) setting. Also, there was very little to suggest in our test group that the air entering the cabins of the test vehicles under a recirculation setting did so due to the operating HVAC system (i.e. air entered due to natural infiltration, rather than mechanical means). In general, the inter-vehicle variability in ventilation rate when operating under a given speed and ventilation setting was substantial, which as was noted in chapter 4, underscores the importance of accurately quantifying this parameter in a range of vehicles. While it would not be appropriate to suggest that the data presented in this thesis are universally applicable to passenger automobiles, the inclusion in the test group of vehicles encompassing a substantial range of ages and types has resulted in an equally substantial range of ventilation rate measurements, which, it would be reasonable to assume, were representative of similar vehicles not assessed in the present study.

The findings presented in chapter 4 have profound implications for occupants of older vehicles (e.g. similar to the 1989 Mazda 121), in terms of their in-cabin pollution exposure. As explained in chapter 5, the exposure of vehicle occupants to UFPs during tunnel travel was strongly related to ventilation rates, with the presence or lack of an HVAC filter also an important parameter.

Despite the scarcity of analogous data, where suitable data sets were identified, the results of the ventilation rate assessment in chapter 4 were often in concert with other studies. Based on this, and the efforts taken in selecting the test vehicle group, employing suitable measurement techniques and assuring instrument reliability, the veracity and external validity of the data is felt to be of a high standard, with due reference to the caveats noted above. It is hoped that the data presented in chapter 4 will be used by other researchers, and will afford improved accuracy in the assessment of in-vehicle exposure to a multiplicity of pollutants. Based on all information presented above, it can be stated that objective C) of this project was met categorically.

**D): Quantification and Assessment of the Relationship Between On-road and In-vehicle UFP Concentrations**

The measurement of the relationship between on-road and in-vehicle UFP concentrations in the study vehicles under the various ventilation settings was described in chapter 5. The need for such measurements stemmed not only from the comparative lack of data in this area when compared to other built environments, but also the proposed modelling efforts detailed in objective D), which necessitated the measurements in order to derive appropriate input parameters. In an integrative assessment of in-vehicle UFPs, once quantification and assessment of determinants of roadway concentrations have been undertaken, the next logical step is to examine and characterise the relationship between on-road and in-cabin UFP concentrations. This was explored in chapter 5, and the role of the ventilation rates discussed in chapter 4 was highlighted.

In chapter 5, the results of alternate on-road and in-cabin UFP sampling in the test vehicle group during the 301 trips through the M5 East tunnel (and, in some cases, on a mixed roads route) during sampling were described. The 2000 Subaru Liberty was not included in these measurements. The ingress of UFPs into the cabin of the test vehicles under each ventilation setting was determined from these measurements, and expressed in terms of the median I/O UFP ratio. Lowest protection to UFP exposure for a given vehicle was recorded under ventilation setting (B) (also referred to as E3 in chapter 4). This was ascribed to the higher ventilation rates observed under this setting, as the total time available for diffusion for an incoming UFP-laden air parcel, whether to a filter (Qi et al., 2008) or HVAC components, is reduced under such conditions. Highest UFP protection was measured, for the overwhelming majority of vehicles, under ventilation setting (C) (also referred to as R1 in chapter 4).

Under this setting, the main mechanism that acted to suppress UFP intrusion was the relatively low ventilation rate observed under this setting for a given vehicle. Also, the mixing of incoming on-road air with recycled cabin air is thought to have resulted in lower in-cabin UFP levels under this setting. However, the inter-vehicle variability in UFP ingress was substantial under this setting compared to others, which largely reflected the same trend in ventilation rates. In general, older vehicles allowed a greater proportion of on-road UFPs to enter the vehicle cabin, and the lower ventilation rates and, in some cases, presence of pollen filters in newer vehicles afforded better occupant protection against UFP intrusion.

The role of ventilation rates in governing the penetration of on-road UFPs into the cabins of the test group was described in chapter 5. A strong linear relationship between outdoor air ventilation rate and I/O UFP ratio was observed, encompassing all vehicles and ventilation settings assessed. The mechanistic relationship between the ventilation rate of the cabins of our test vehicles and the proportion of on-road UFPs able to reach the cabin thus appeared to be a dominant factor determining in-cabin UFP exposure. Although collected primarily in a road tunnel, the data described in chapter 5 should be applicable to most roadway environments. The work presented in chapter 5 not only contributes to, and extends knowledge of, the mechanisms underpinning in-cabin UFP exposure, but also meets objective D) of this thesis. It is prudent to note, however, that while the ability of a filter when combined with a relatively high level of air-tightness to minimise in-vehicle UFP concentration is of substantial benefit to the occupants, it is only one of myriad parameters affecting their well-being and comfort. For example, Rudell et al. (1999) found that the ability of an automotive cabin filter to reduce particle number count when challenged with dilute diesel exhaust did not in itself indicate an ability to reduce undesirable subjective symptoms (such as headache, eye irritation and others).

The final primary objective of this project was the integration of all measured data into a simple model capable of predicting the trip average UFP exposure encountered by vehicle occupants during a trip through the M5 East tunnel. Chapter 5 detailed the efforts involved in implementing a basic particle number-balance model for this purpose. It was intended that the utility provided by such a model could conceivably be applied by an end-user (i.e. roads or tunnel management authority) to estimate average vehicle occupant exposures to UFPs.

The model chosen was a standard particle mass (or in this case, number) balance model, previously implemented successfully by Jamriska et al. (2000) to predict indoor UFP concentrations in an office building. To ensure the integrity of the predictions made by the model, 30 tunnel trips completed during the field sampling campaign, encompassing all vehicles and ventilation settings, were selected using a random number generator and excluded from any calculations of UFP ingress. These validation data, representing 10% of all tunnel trips, were therefore completely 'new' when used in the model. The model was parameterised with relevant data collected during the sampling campaigns described in chapters 3, 4 and 5. When the model-predicted trip average UFP concentrations were compared with those measured, the agreement was reasonably good, particularly at UFP concentrations below $2.50 \times 10^5$ p $cm^{-3}$. It was never an intention of this study to conduct comprehensive modelling and analyses of model output; indeed, this is a field of investigation unto itself. Nonetheless, the performance of the simple model was of a suitable standard for use in a semi-quantitative fashion by a road or tunnel management authority. In this way, when model output indicated it was likely that in-cabin UFP exposures were likely to be above a threshold (e.g. above a certain percentile of previously predicted average exposures), then appropriate mitigative measures could be implemented (e.g. recommending susceptible persons use an alternate route). As noted in chapter 5, if the on-road UFP concentration for a given roadway can be predicted reasonably reliably as a function of vehicle volume or other parameter, then the model could be used to produce a suite of predictions, from worst to best case UFP exposure based on vehicle type and ventilation setting. An improvement on this approach would involve the continuous monitoring of UFP concentration, in a road tunnel for example. However, this not likely to be feasible in many locations due to cost and the current lack of air quality standards for UFPs that would necessitate this type of monitoring.

It should be noted that the modelling described in this thesis, while successful in meeting the original objective, was one of several objectives investigated. Additionally, there have only been two reported studies of in-cabin UFP modelling including chapter 5 of the present study; the other being the work of Xu and Zhu (2009). Accordingly, there is considerable scope for future studies to further investigate and improve the hitherto under-researched area of in-vehicle UFP modelling.

## 6.2 Limitations

The various limitations within this thesis have largely been noted in the appropriate sections of chapters 2-5; however, they are also summarised here. In any sustained research effort, especially one that is founded on field sampling campaigns as was the case with this project, limitations are typically going to arise. Given that the sampling location throughout this study was the cabins of vehicles moving at speeds up to 110 km/h, it was initially anticipated that this would give rise to operational issues with experimental equipment. In practice, however, such problems were minimal and rarely affected data collection campaigns. All experimental equipment was powered by two 120 ampere hour deep cycle absorbed glass mat batteries, which were fed through a pure sine wave inverter to produce the necessary 240 volt power. A rapid charger was used to recharge the batteries at the conclusion of each sampling session. The mobile power system performed faultlessly throughout the sampling campaigns, and lack of power was never a limiting factor.

The ventilation measurements described in chapter 4 were performed under strict protocols using calibrated equipment and well-defined methodology. As noted previously, the data gathered is thus felt to be of high quality. Nevertheless, it is appropriate to note that the data is representative of specific vehicle types fitted with basic HVAC systems. While very effort was taken to include a substantial range of vehicles, due judiciousness should be exercised if extrapolating the results presented here to other passenger automobiles.

The bulk of limitations present in this study are related to the UFP measurements. Firstly, the TSI 3007 CPC used throughout this study, whilst an admirable performer in a difficult measurement environment, is a relatively simple CPC capable of obtaining basic data. As stated in chapters 3 and 5, its specifications do not define it as a true UFP counter; however, given the size distribution of particles encountered on-road, UFP is a justifiable descriptor for the measurements performed in this study. The 3007, by virtue of its size and portability, was an appropriate option for the measurement environment. The simple dilution system described in chapter 2 was implemented to overcome the limitations of the 3007 with respect to its maximum detectable concentration. Repeated testing of this system showed it to be consistent in terms of particle concentration reduction due to dilution and other processes when challenged with a four-stroke unleaded gasoline combustion aerosol. No efforts were made to ascribe the behaviour of the system to dynamic aerosol processes, as this would have constituted a study in its own right and was not an objective of this thesis. Once it was determined that the system performed satisfactorily, it was deployed in the field. The trade-off resulting from the development of an effective dilution system that was reliant on a moderate diluent to sample air ratio (i.e. in terms of volume flow rate) was the associated increase in sample residence time in the sampling train. This too was characterised in the laboratory prior to the field sampling campaign, and was found to result in comparatively minimal UFP loss when compared to the effects of the dilution system. Nonetheless, an overall (rather than dilution-specific) correction factor was calculated and applied to all data collected (see appendix). Due to the relatively long sample residence time, use of a single CPC and short duration of most tunnel trips, it was necessary to switch the sampling location (i.e. in-vehicle or on-road) every 20 or 25 seconds, depending on vehicle being tested and the sample tubing length used. Of each 20-25 second data block, the first 10-15 seconds were occupied by clearance of the previous sample and arrival of the new sample and were excluded from the analyses. Obviously, this level of data redundancy is not ideal, but given the nature of the sampling equipment, dilution system, study location and the objectives of the study, it afforded a useful compromise and worked well in practice. Also, the generally good agreement between the results of this study and others described in chapters 3 and 5 is seemingly testament to the absence of any substantial sampling deficiencies in this study.

The lack of particle size distribution data is an obvious limitation of this study. However, given the short duration of most tunnel trips, coupled with the time required by most portable instruments capable of scanning the particle number size distribution, measurements of particle size distribution using the mobile sampling platform approach employed in this study were not practicable. Also, on-road particle size distributions for various vehicle fleet compositions have been reasonably well-defined in recent years, and there is little to indicate that the distributions present in the sampling locations used in this study would differ significantly from these. Similarly, no particulate chemical speciation analyses were performed as part of this study, but given the absence of significant UFP sources besides vehicular combustion in the sampling environment, the results of chemical analyses of UFPs (see section 1.2) produced by mixed vehicle fleets should be largely representative of those in the M5 East tunnel. Nonetheless, these UFP parameters could in investigated in future studies of the tunnel.

During the main UFP sampling campaign in the tunnel (mid-2006), the system responsible for recording the hourly volume and type of vehicles using the M5 East tunnel was offline. This became apparent only after the conclusion of the sampling campaign and was beyond the control of this study. Accordingly, the average daily traffic statistics for 2007 were used as a surrogate for this data. The authority responsible for recording this information advised that this data would be highly representative of conditions during the sampling campaign, as traffic volume and fleet composition in the tunnel is extremely consistent.

The absence of simultaneous in-vehicle and on-road UFP measurements are likely to have resulted in the inflation of I/O UFP ratios under high ventilation rate conditions (i.e. conditions (A) and (B) or E1 and E3). This was driven by the tendency of on-road UFP concentrations to undergo significant fluctuations over short time scales. The use of two identical inter-calibrated CPCs would have overcome this, although such an approach was not possible in this study. The primary limitation imposed by this is that the I/O UFP ratios measured under the two aforementioned ventilation settings were overestimated. This did not, however, affect the in-cabin UFP measurements or the modelling efforts described in chapter 5. The gradient of the linear relationship observed between outdoor air ventilation rate and I/O UFP ratio (shown as figure 2 in chapter 5) may have been slightly reduced had the two CPC approach been employed. However, given such a scenario, the goodness-of-fit of the regression line applied to these data is very likely to have been similar to that obtained with the data described in chapter 5. Hence, the general characteristics of the relationship between the two variables, especially in a qualitative sense (e.g. the relative position of each ventilation setting on the graph), were largely unaffected by the sampling approach utilised.

## 6.3 Recommendations for Future Studies

While the study of the exposure of vehicle occupants to some combustion-derived pollutants is not a particularly new field of endeavour, the measurement and analysis of in-cabin UFP exposure is. It is felt that this thesis makes a substantial contribution to this nascent area of research, both in terms of fundamental data and deeper analytical insights. Nevertheless, there are numerous issues that remain to be addressed.

Given the potentially large contributions of time spent in a vehicle to a person's daily UFP exposure identified by other studies (Zhu et al., 2007; Fruin et al., 2008) and applied to the tunnel environment by this study, a priority area of research is the comprehensive assessment of exposure to UFPs incurred by professional drivers, couriers, taxi drivers, police officers and the like. The work of Zhu et al. (2007) and Fruin et al. (2008) suggested that up to 50% of daily UFP exposure can be ascribed to commuting activities comprising 1.5h; therefore, both the in-vehicle contribution and overall UFP exposure for a person who spends 8 or more hours in a vehicle cabin environment may be of substantial magnitude, and thus demand appropriate investigation. Given the results of this study, further studies of the effects of tunnel travel on daily UFP exposure, including assessment of the role of tunnel characteristics, would be a logical and useful research pathway.

Associated with the above recommendation is the investigation of the effects of exposure to on-road UFPs on human health end points. This is a very new area of research, with many attendant logistical, instrumental and ethical issues. Elder et al. (2004) performed on-road UFP exposure studies and health end point monitoring using rats as the exposure subjects. Recently, Zhu et al. (2008) described the development of a mobile UFP (and other pollutant) exposure system designed for human subject health studies on roadways. The results of studies performed with this and similar systems will be of great interest in terms of better elucidating the relationship between in-vehicle UFP exposures and subsequent health effects.

The on-road assessment of the UFP removal efficiency of advanced filtration devices to reduce UFP concentrations entering vehicle cabins would be of use as one means of mitigating occupant UFP exposure. Current cabin air filter standards only specify performance for particles sizes above 300nm, and it has been suggested that this needs to be redressed to include UFPs (Qi et al., 2008). While standard cabin air filters offer moderate protection from UFPs (Pui et al., 2008; Qi et al., 2008), it would seem beneficial to both develop and assess the real-world ability of alternative filtration types to capture on-road UFPs prior to their ingress into vehicle cabins.

While the above suggestion is directed towards new vehicles, occupants of older vehicles lacking any filtration device may benefit from investigation of inexpensive and simple means to reduce excessive on-road air infiltration. 'Leaky' buildings have, for some time, been retrofitted with various products in order to reduce energy consumption. While the benefits of such retrofitting in buildings may be questionable in terms of the potential indoor air quality impacts, there is no reason why some less air-tight, older passenger vehicles couldn't be similarly outfitted. For example, the 1989 Mazda 121 examined in this study would be a prime candidate for such a procedure. Obviously, if these measures were implemented, due caution should be exercised not to over-tighten a vehicle and potentially exacerbate the effects of occupant's $CO_2$ emissions or in-cabin pollutant sources. It would also seem beneficial to perform an extensive study of air leakage pathways in vehicles in order to improve understanding of pollutant intrusion pathways, and so that the appropriate locations to direct retrofitting activities could be identified. The magnitude of UFP exposure reduction effects attributable to the retrofitting of older vehicles with filtration systems (and/or improving their air tightness), where possible, would also be a useful area of investigation.

As referred to earlier in this chapter, the development and refinement of models capable of predicting in-cabin UFP concentration would be of significant utility in terms of reducing occupant exposures. Future studies should focus on further improving the accuracy of such predictions and assessing the performance of modelling in a practical application (a tunnel would be an ideal location), as the latter was not possible in the present study.

With specific reference to the M5 East tunnel, as noted above, further studies of UFP pollution and characterisation of UFP size distributions and chemical composition would be a useful complement to this study. Assessment of the efficacy of the tunnel air filtration plant (currently under construction) to reduce on-road UFP concentrations would be a key adjunct to the work presented here. Similarly, the effects of the reduction of sulphur content (from the start of 2009) in Australian diesel fuel on UFP concentrations in the M5 East tunnel could also be assessed via comparison with the results contained in chapter 3 of this thesis.

As a final comment, it is important to highlight that the work described here has been deliberately focussed on assessing and analysing one of the 'symptoms' caused by vehicular emissions of UFPs. Possibilities for improving the quality of in-cabin air and reducing UFP intrusion have been suggested here. However, as the adage states, "prevention is better than cure". In this respect, it is of much greater benefit to reduce the emission of UFPs at their source, thereby directly treating the cause, than to implement measures to address the myriad resultant symptoms; in this case, in-vehicle UFP exposure. The fact that this thesis and many related research works addressing such symptoms needed be undertaken is testament to the magnitude and pervasiveness of UFP pollution.

Thankfully, particle number (as opposed to mass) emission guidelines are now being promulgated in some parts of the world. The implementation and future development of such measures hold promise for directly treating the predominant cause of UFP pollution; to the benefit of both humans and the environment.

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# Appendices

**Figure A-1 (top).** 1989 Mazda 121. **Figure A-2 (bottom):** 1998 Mitsubishi Magna.

**Figure A-3 (top)**. 2000 Subaru Liberty. **Figure A-4 (bottom)**. 2005 Toyota HiLux.

**Figure A-5**. 2005 Volkswagen Golf.

**Figure A-6**. 2007 Subaru Outback.


**Figure A-7.** Experimental equipment and power supply set up in the boot of the 1998 Mitsubishi Magna prior to an early (Jan 2006) ventilation measurement. The Brüel & Kjær 1303 sampler/doser (rear left) and 1302 photacoustic gas monitor (rear right) can be seen, along with the two 120 Ah absorbed glass mat deep cycle batteries and pure sine-wave inverter used to produce the required 240 volt power. The $SF_6$ cylinder cannot be seen in this figure; in order to transport it in a safe (i.e. upright) manner, it was secured in the rear of passenger compartment. Note that foam padding was used to protect the instruments, but was removed for clarity prior to taking photos.

**Figure A-8.** The same experimental instruments described in figure A-7 can be seen here in the boot of the 2005 Volkswagen Golf, in addition to the $SF_6$ cylinder (prior to being secured). The batteries and power inverter were fitted after this photo was taken.

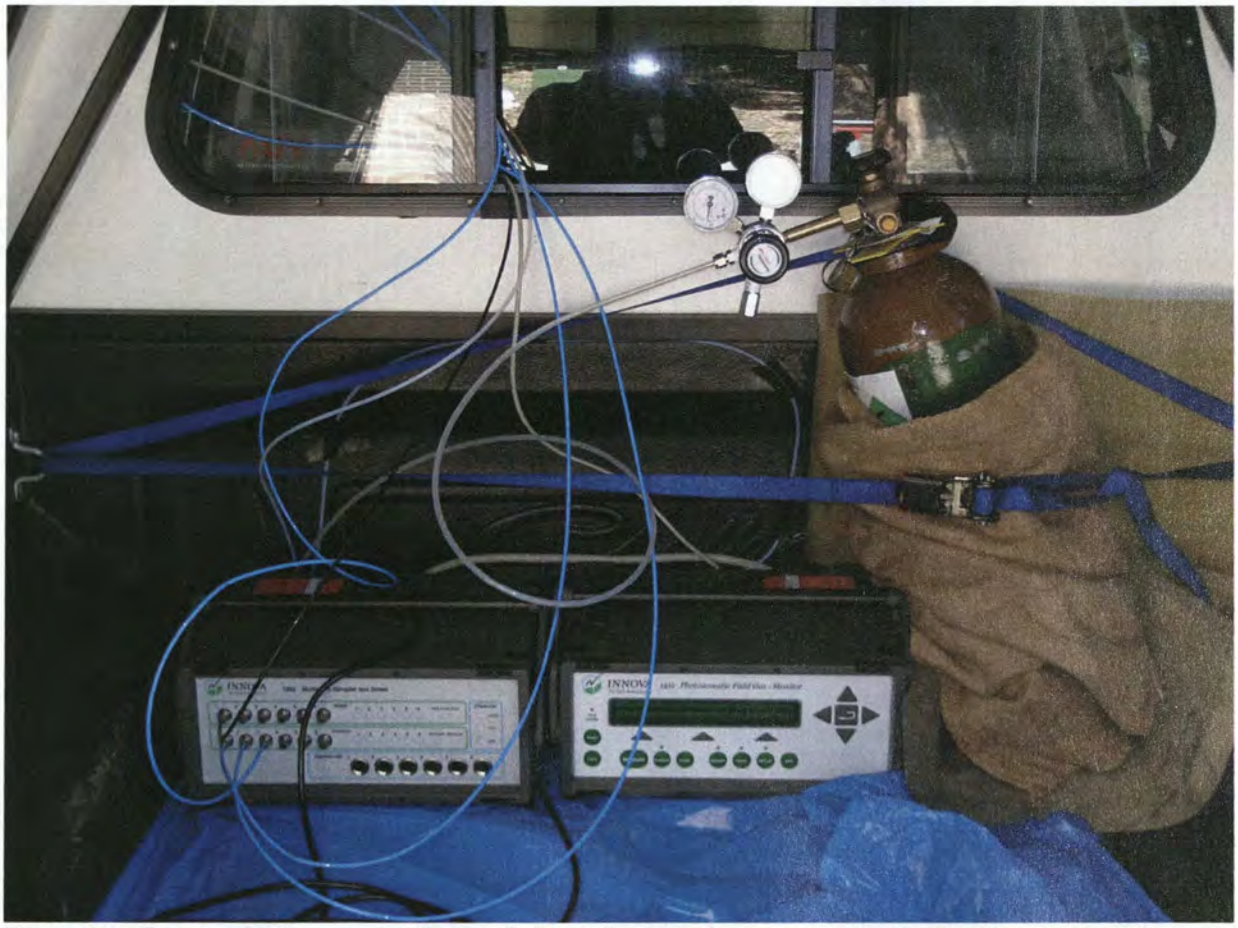

**Figure A-9**. Innova 1303 sampler/doser (left) and 1412 photoacoustic gas monitor (centre) next to the $SF_6$ cylinder in the cargo tray of the 2005 Toyota HiLux.

**Figure A-10**. Sampling points for a constant tracer gas emission measurement inside the 2005 Toyota HiLux.

**Figure A-11**. Same as figure A-10, except inside the 2007 Subaru Outback.

**Figure A-12**. Open car park used for stationary infiltration measurements (2005 Toyota Hilux in the distance).

**Figure A-13**. 2005 Toyota HiLux prior to a stationary infiltration measurement; RM Young 05103 wind speed and direction monitor in the foreground.

**Figure A-14**. Same as figure A-13, except test vehicle is the 1989 Mazda 121.


Figure A-15. Four stroke petrol generator, test chamber, TSI 3022A CPC (top left), TSI 3007 CPC (top centre) and laptop computer set up prior to laboratory test of dilution system performance.

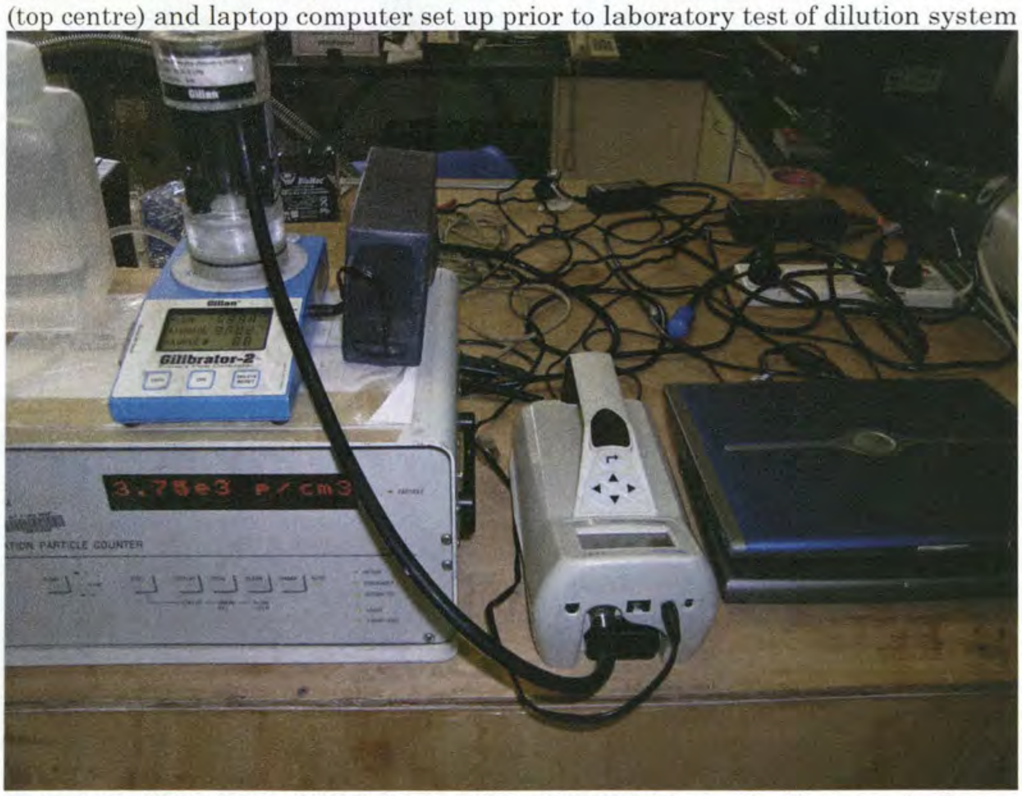


Figure A-16. TSI 3022A CPC (left) and TSI 3007 CPC (centre). The sample flow rate of the 3007 is being measured.


**Figure A-17**. TSI 3007 CPC (affixed to cardboard by Velcro) with dilution system, sampling point switching valve and small fan seen from through the front windscreen of the 2007 Subaru Outback.


**Figure A-18**. As above, except the vehicle is the 1989 Mazda 121. The external (i.e. on-road) sampling line can be seen extending from a small hole made through a thick tape seal placed over the front passenger side window

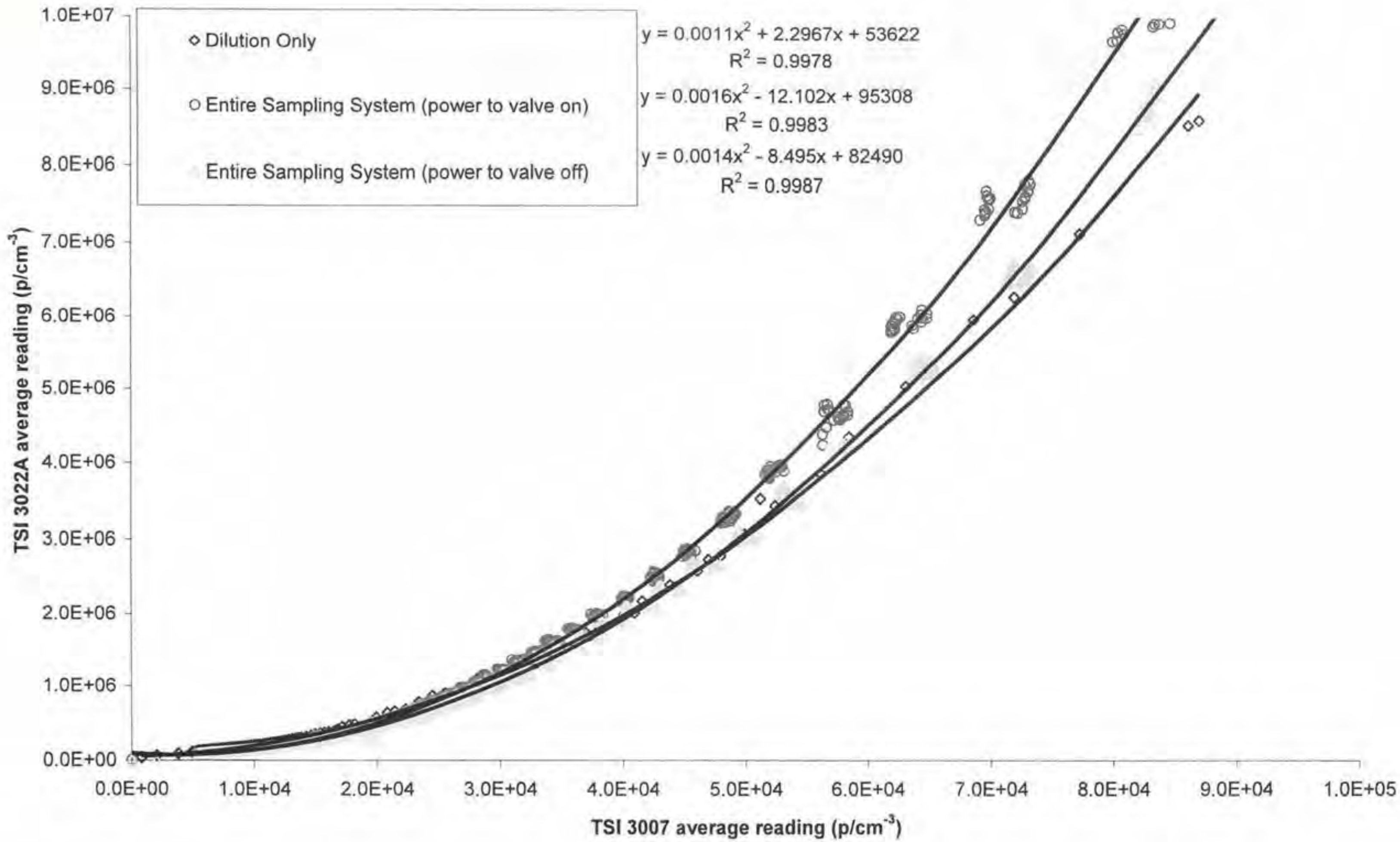


**Figure A-20**. Particle loss attributable to dilution system only (see chapter 2), and also to the entire sampling train (including dilution system), depending on valve power state.

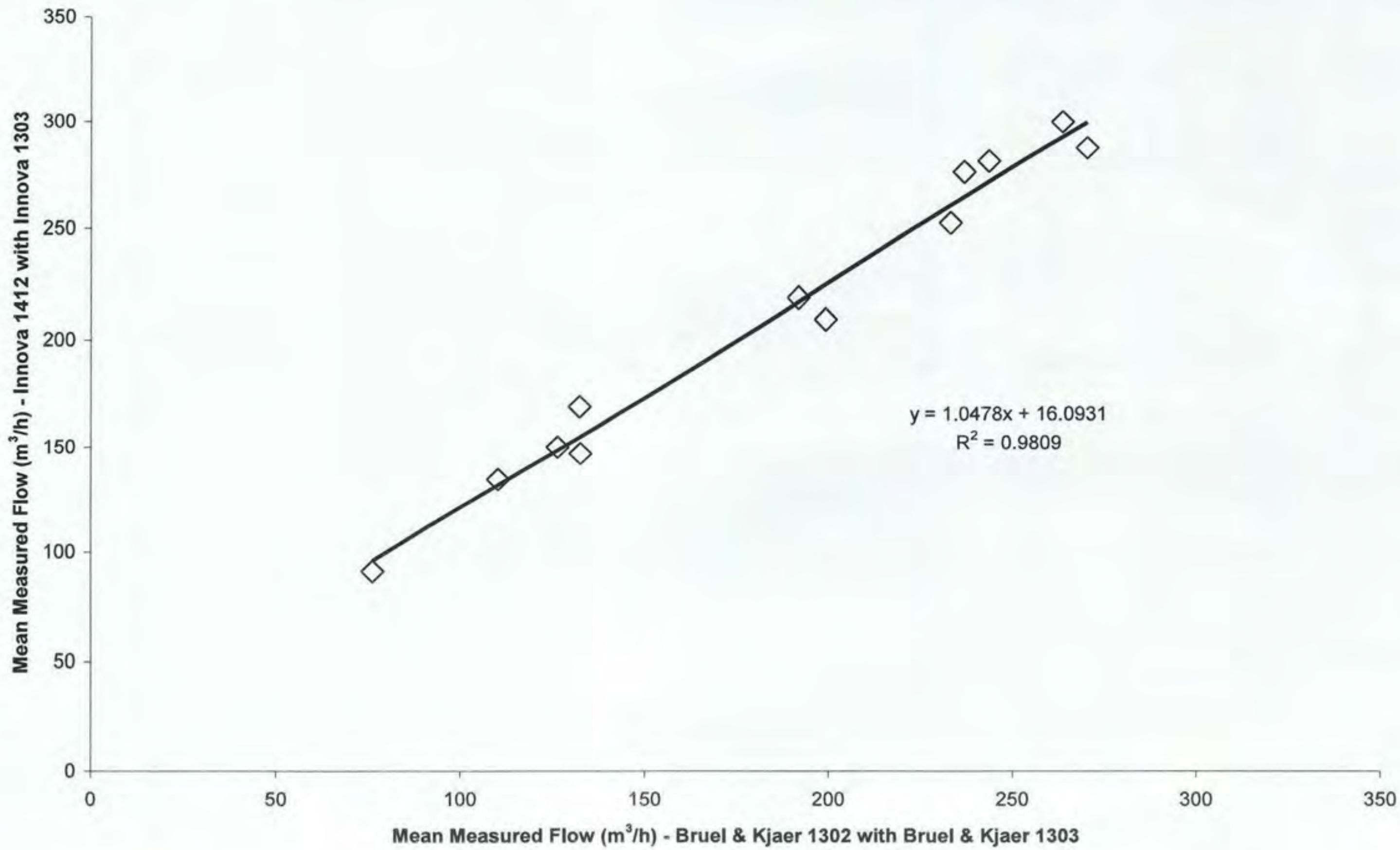


**Figure A-19**. Inter-comparison of air flow rate measurements performed with Brüel & Kjær and Innova equipment. B&K measurements within the range of the data above were standardised to those performed with the Innova system, which had been manufacturer-calibrated prior to the field campaign.