Assessment of different approaches to determining personal exposure

Final Report

W.Physick[1], J.Powell[1], M.Cope[1], K.Boast[1], S.Lee[1], W.Lilley[2], R.Gillett[1], G. Edgar[3]

[1] CSIRO Marine and Atmospheric Research
[2] CSIRO Energy Technology
[3] Environment Protection Authority Victoria

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Contact: Khokan Bagchi
Air Quality Section
PO Box 787
Canberra ACT 2601

Enquiries should be addressed to:

Dr. Bill Physick
tel: (03) 9239 4400
fax: (03) 9239 4444
email: bill.physick@csiro.au

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EXECUTIVE SUMMARY

The primary aim of this Clean Air Research Programme (CARP) project is to evaluate methodologies for estimating personal exposure from ambient monitoring data and from simulation data from complex ambient air quality models. We focussed our efforts on nitrogen dioxide (NO_2), but also present measurements and modelling of fine particulate matter ($PM_{2.5}$).

Following a literature search, we developed a conceptual model of personal exposure to NO_2 based on time-weighted sums of exposure in the microenvironments of home, transit and work. In this model, personal exposure in each microenvironment is linked to ambient concentration by indoor-outdoor concentration ratios. Previous studies indicated that gas cooking appliances and house ventilation rates are strong influences on indoor NO_2 concentrations, and thus on indoor-outdoor ratios. Unflued gas heaters are also significant contributors, but there are restrictions on the installation of such heaters operating on Natural Gas in Victoria and consequently their use is not widespread. There were none in any of the homes in which measurements were taken in our study. To allow us to both develop and evaluate our model, we designed a measurement program involving volunteers across Melbourne wearing personal samplers. Participants' diaries were designed to record details of time and activities in each microenvironment, especially those associated with cooking and ventilation. Measurement of house ventilation rates was also conducted at five dwellings.

The field-work for the project entailed the measurement of NO_2 concentrations (cumulative) across Melbourne for 15 - 17 volunteers wearing personal passive samplers in each of four events (each of two days). In addition, $PM_{2.5}$ concentrations were continuously measured over the same periods by four volunteers with portable DustTrak™ monitors (TSI inc.). Both working and non-working participants were included in the study. All participants were non-smokers. The study was done for a total of four separate two-day events, in April 2007, May 2006, May 2007 and June 2006. These times of year were chosen for the stable light-wind conditions to maximize concentrations and the spatial variation in concentrations across the city and suburbs. Participants also wore additional samplers for sub-periods of each 48-hour exposure, at home, at work and in transit between work and home. Outdoor concentrations were also measured in these microenvironments, except for travel on public transport, allowing indoor-outdoor ratios to be calculated.

Nitrogen dioxide

A wide range of NO_2 personal exposures (average concentrations), from 6.1 ppb to 19.8 ppb, was experienced across the different activity profiles of the participants. The highest exposures were measured in the transit microenvironment (mean 46.2 ppb), but the major portions of the total dosage (exposure multiplied by time spent in an environment) were experienced at home and at work. For each of the four events, the highest personal exposure did not exceed the maximum ambient NO_2 concentration measured by the EPA Victoria monitoring network, suggesting that for our people profile (office workers, stay at home people and one outdoor worker) the maximum monitored concentration is a conservative estimate for a city's population exposure. However, for 19% of measurements, the personal

exposure was less than the minimum concentration measured across the monitoring network, indicating that assignation of the city's maximum ambient exposure to everyone would strongly overestimate exposure.

Ratios of indoor to outdoor concentrations at home varied from 0.12 to 1.37 (mean 0.57 ± 0.27), with extreme values attributed to indoor NO_2 sources and to low ventilation rates in new houses. Ratios at workplaces were all less than 1.0 (no indoor sources) and showed much less variability (mean 0.74 ± 0.16). For each home, a mass balance equation was used to calculate indoor NO_2 concentrations, given the outdoor concentration, assuming a steady state and using an assumed deposition rate from previous studies, emission rates according to the time spent cooking with gas and ventilation rates according to house age. This approach gave good agreement with the measured indoor concentrations (correlation 0.78). A simpler approach depending only on whether a gas cooking appliance was installed in the home gave acceptable results, with a correlation of 0.63. Indoor-outdoor ratios were calculated from each approach and evaluated in our personal exposure model.

The in-vehicle to out-vehicle concentration ratio was calculated for 16 vehicles by attaching samplers to the side mirror. For all trips, windows were closed and air-conditioning was off. A mean value of 0.63 (±0.17) was obtained when the vehicle was driven with the external vent open. Readings of 0.37 and 0.07 were measured for two trips with the vent closed.

The Australian Air Quality Forecasting System (AAQFS) and CSIRO's air quality model TAPM-CTM were run for each two-day event, and results compared to the outdoor (ambient) concentrations measured by the samplers at home and at work. The models underestimated NO_2 concentrations, especially during the daytime, but good agreement with sampler ambient data was obtained with a blending procedure in which EPA Victoria monitoring data were incorporated into the TAPM-CTM model predictions.

Our conceptual model of NO_2 exposure involves linking personal exposure to ambient exposure by indoor-outdoor ratios. Methodologies evaluated in this study included three approaches to calculating ambient exposure and three methods of estimating indoor-outdoor ratios. Ultimately it is hoped that our recommended methodology can be used in epidemiological studies where pollutant exposure of many subjects needs to be estimated. To this end, we have introduced two simplifications. Firstly, following our finding that the major exposure components occur in the home and work environments, we omitted the transit environment from our model. In fact, an evaluation of our methodology for in-vehicle exposure showed very weak correlation between ambient and on-road concentrations. Secondly, recognising the practicalities of an epidemiological study, we assumed that all participants are at home between 1800 Eastern Standard Time (EST) and 0800 EST, and at work between 0800 EST and 1800 EST.

Ambient NO_2 exposures for each person for these periods were obtained by two methods: (1) concentrations at the *nearest monitor* in the EPA Victoria ambient monitoring network to home or work were assigned, and (2) concentrations at the home and workplace were assigned from the gridded hourly NO_2 concentrations obtained by *blending* the modelled and EPA Victoria monitored data. Home indoor-outdoor ratios were calculated from two methods for computing indoor NO_2 concentrations developed from diary data. The use of *measured indoor-outdoor concentration ratio* averaged across all homes for each participant was also evaluated. For the workplace, a constant indoor-outdoor ratio was used for all workplaces and was the mean value measured in the study. All methods produced good

agreement with the measured personal exposure values, especially by the criterion that a prediction method is deemed to be valid if the root mean square error (RMSE) is less than the standard deviation of the measurements. Importantly, the standard deviations predicted by these spatial-variation techniques match well the variation seen in the measurements. Evaluation statistics were poor for a commonly-used method whereby each person is assigned the same ambient concentration, taken to be the mean concentration across all monitors in the EPA Victoria network.

For estimation of the personal exposure to NO_2 of a large number of people, it is recommended that best results would be obtained with the I/O ratio calculated from a mass balance method. This requires participants to record daily gas cooking periods and approximate house age, although a simpler but slightly less accurate method dependent only on the existence or not of a gas cooking appliance also produces satisfactory results. The recommended method for calculating the required ambient outdoor concentration is to use the *nearest monitor* approach. However there is very little difference between results from the *nearest monitor* and *blended* approaches and the former is only recommended as it is simpler and researchers may not always have access to an emissions inventory or model for the *blended* approach.

While these findings are promising, they can only be related at this stage to NO_2 and to the existing EPA Victoria monitoring network. The findings are also relevant only to persons who spend the majority of their time at indoor work and/or home, allowing time spent in other microenvironments such as transit to be ignored. Results may be different for those who drive for a living or who spend a significant amount of time near roads. For those situations, further work could be done to relate transit exposures to key variables, including traffic volume and ambient concentration from the nearest monitor. Alternatively, modelling at fine-resolution (e.g. 10 m) in the vicinity of roads of interest, using a specific vehicle emission inventory for each road, could be explored.

Fine particulate matter

For each of the four events, DustTrak monitors logging one-minute $PM_{2.5}$ data were assigned to three 'workers' and to one person who stayed at home. This resulted in data for 15 home, 10 transit (motor vehicle) and 10 work microenvironments, consisting of eight different homes, eight different transit routes and three different workplaces. The highest personal exposure (a two-day mean of 23.2 $\mu g\ m^{-3}$) was just below the advisory Air NEPM standard for $PM_{2.5}$ of 25 $\mu g\ m^{-3}$ (24-hour average). This participant was a truck driver who spent 41% of the 2-day period in the transit microenvironment, where his mean exposure was 30.1 $\mu g\ m^{-3}$. The range across the participants of personal exposure (2-day mean concentration over an event) for each event lay within the range of 2-day mean ambient values measured across the EPA Victoria monitoring network. The highest values measured in the home (25.8 $\mu g\ m^{-3}$) and transit (30.1 $\mu g\ m^{-3}$) microenvironments exceeded the advisory NEPM standard, although the standard was also exceeded in the ambient monitoring network for that event. Comparison of hourly-averaged values to the nearest work and home monitors showed that the ambient concentration was a strong component of the personal exposure of the participants.

The 1-minute averages of $PM_{2.5}$ in each microenvironment showed short-period concentration excursions (two to 15 minutes) reaching values five to 10 times higher than the longer-term average concentration. In the home, these were involved with cooking, a hairdryer and extinguishment of a candle, and in transit were associated with traffic congestion, smoky vehicles and an idling truck in which its exhaust fumes entered the cabin. These findings are relevant in the light of epidemiological and toxicological work showing stronger respiratory health impacts from $PM_{2.5}$ concentrations measured over intervals shorter than the NEPM averaging period of 24 hours.

The indoor concentration trends tracked hourly-averaged ambient concentrations from the nearest monitor, with short-term deviations associated with activities in the home. The mean indoor-outdoor ratio was 0.90 (±0.19), ranging between 1.26 and 0.52, with the dominant source being cooking, and smaller contributions from hair dryers and candles. In the workplaces, with no obvious sources, ratios were all below 1.0 with a mean of 0.58 (±0.15). While no $PM_{2.5}$ readings were taken directly outside the vehicles, concentrations from the nearest monitor were used to obtain ratios of in-vehicle to ambient concentration. These ranged from 1.44 to 0.80 with a mean of 1.07 (±0.19). Only 6% of a 24-hour day was spent in transit by our predominantly office-worker cohort.

The AAQFS and TAPM-CTM models for $PM_{2.5}$ did not perform as well as they did for NO_2, with the mean concentrations under predicted, typically by 50%, and standard deviation only fairly predicted. RMSE for both models was larger than the observed standard deviation for all events, indicating that the models did not predict with any skill. Correlations were 0.53 and 0.61 respectively. The models' worst performance was for event 3, when RMSE values were much higher than the observed standard deviation and there was almost no correlation between model and observations. The major reason for this was the presence of smoke haze on both days of the event, as there is no source in the models for particulate matter from fires. Improvements in model performance are likely to be seen with an updated $PM_{2.5}$ inventory and incorporation of an algorithm to predict secondary organic aerosols.

In a similar manner to the analysis of NO_2, simple methods for estimating personal $PM_{2.5}$ exposure were evaluated using (1) monitored data, and (2) a blended combination of monitored data and TAPM-CTM modelled predictions. The same approach was adopted, assuming that all participants were at their workplace between 0800 LT and 1800 LT and at their home location between 1800 LT and 0800 LT. The indoor-outdoor ratios used to link personal exposure to ambient exposure were the mean measured home and work ratios.

Both the nearest monitor and blended data methods gave acceptable results, though not as good as for NO_2. A simpler approach in which all participants were assumed to be at home for the duration of each event also gave acceptable results. However, it was not possible to conclude whether this or the home plus work approach is superior or whether it is better to use the nearest monitor or the blended data method. This is probably due to the small number of participants and the fact that eight out of 10 workplace data sets were measured at the one location (Aspendale). Even so, the differences between results using the nearest monitor data set and the blended data set were not large, and as for NO_2 suggest that either approach is suitable.

General

In epidemiological studies, the exposure assigned to an urban dweller over a period is often the mean pollutant concentration for that period, averaged over all monitors in the urban monitoring network. Hence, each member of the population receives the same exposure value. For comparison with our spatial variation methodologies, we examined the statistics arising from assigning the mean ambient concentration from the EPA Victoria monitoring network to each participant in all four events, and using the mean measured indoor-outdoor ratios to convert to personal exposure.

For NO_2, the mean, RMSE and correlation were poor and this approach is clearly inferior to the techniques developed in this project. It must also be remembered that there is no exposure variation between participants using this mean concentration approach, whereas the standard deviation predicted by the spatial variation techniques matched well the variation seen in the measurements.

For $PM_{2.5}$, the mean and RMSE values were good, but as for NO_2 there is no exposure variation between participants using this constant concentration approach. However, the relatively low RMSE values for an approach which assigns each home location the same $PM_{2.5}$ concentration implied that there was not a lot of spatial variation between those locations. This does not mean that there was not much variation across Melbourne (for each event there was typically a factor of two between the highest and lowest 2-day mean concentration at the monitors), only that the small number of chosen home and work sites did not capture that variation. This is in contrast to the findings from the NO_2 part of the project, for which there were four times as many measurements from a wider variety of locations, and in which greater confidence can be assigned to the results.

Our results for NO_2 and $PM_{2.5}$ are relevant for estimating the personal exposure of individuals in epidemiological cohort studies or for calculating an average exposure for a population. In a population exposure study, the best results would be obtained by using a representative number of participants for various activity profiles within the population. Exposure results from each profile would be weighted according to the profile sub-population, and summed. Such activity-profile categories could include 1) people who predominantly stay at home, 2) those who go to work indoors, 3) those who work outdoors or spend recreation time outdoors, and 4) those who spend a significant amount of time on or near roads.

We believe that our research findings contribute to estimating exposure within the above activity profiles of staying at home and working indoors (categories one and two). Our time at work period (0800 EST to 1800 EST) could perhaps be reduced for some sub-groups within the working indoors category (e.g. school children), with an outdoors category (3) added for two or three hours. Our encouraging results for estimating home and work outdoor concentrations from the ambient monitoring network suggest that exposure while outdoors, at work or recreation, could be assigned from concentrations at the nearest monitor.

Our work indicates that there is not a strong relation between on-road concentrations and ambient concentrations, thus ruling out application of our methodology for estimating on-road exposure. For this activity profile (4), further research should be done to relate transit exposures to key variables, including traffic volume and ambient concentration at the nearest

monitor. Measurement work on concentration as function of distance from a road, such as that done by EPA Victoria (2006), and the relation between in-vehicle to out-vehicle concentration ratios and in-vehicle comfort settings and cabin volume are also important for developing robust exposure methodologies. Contributions can also be made through modelling concentrations at fine-resolution (e.g. 10 m) in the vicinity of roads of interest, using a specific vehicle emission inventory for each road.

As the meteorological conditions for our field campaigns were similar for all four events and were chosen to maximise concentrations of both pollutants, as well as their spatial variation, it is expected that our methodology would be equally applicable under more dispersive conditions, such as more uniform or stronger winds across the area of interest when concentrations and spatial variation would be smaller. For estimates of annual population exposure, it is necessary to evaluate exposure under the major meteorological conditions and then weight the results according to the annual frequency of each category.

The above discussion is equally as relevant for personal exposure of individuals in an epidemiological cohort study, except that their exposure is estimated every day of the study and so the previous discussion re the weighting of results under different meteorological conditions does not apply.

While our research has identified a simple exposure methodology that could be widely applied, without the need for access to air quality models and with only minimum information from respondents, there are some simplifying assumptions that need support from further research. Strictly speaking, the findings can only be related at this stage to NO_2 and to the existing EPA Victoria monitoring network, although it is expected that the methodology would also be valid for cities with monitoring networks of similar density to that of Melbourne. Our sample size for NO_2 was necessarily limited to a total of 24 volunteers, with between 15 and 17 participating in each of the four events. However our methodologies were valid for each event, as well as for the combined data set involving 59 samples. Repetition of our work, ideally in another city and with a higher number of participants, is highly desirable and would strengthen the findings of this project. More participants would also widen the variety of homes, workplaces and even ages.

Our methodologies were also successful for $PM_{2.5}$, where the relation between indoor and ambient concentration was stronger than for NO_2, but the sample size was only 25% that of the NO_2 data set. Consequently, similar work is needed in this area too.

In achieving the project goal, to evaluate methodologies for estimation of personal exposure, there have been interesting developments in several research areas along the way. These include the approach of blending ambient monitoring data with model predictions to produce hourly estimates of gridded concentration fields; the application of a mass balance approach to estimate indoor NO_2 concentrations; and the measurement of simultaneous in-vehicle and out-vehicle concentrations.

CONTENTS

1. INTRODUCTION

The primary aim of this Clean Air Research Programme (CARP) project was to evaluate methodologies for estimating personal exposure from ambient monitoring data and from simulation data from complex ambient air quality models. In this Report, exposure is defined as the mean concentration of a pollutant over the period under discussion. We focussed our efforts on nitrogen dioxide (NO_2), but also present measurements and modelling of $PM_{2.5}$. NO_2 is known to irritate the throat and the lung, with the principal site of toxicity being the lower respiratory tract. Recent studies indicate that low-level NO_2 exposure may cause increased bronchial reactivity in some asthmatics, decreased lung function in patients with chronic obstructive pulmonary disease, and an increased risk of respiratory symptoms and infections, especially in young children (USEPA).

Australian studies have found associations between exposure to NO_2 and negative health effects at NO_2 levels to which the Australian population is typically exposed. Pilotto et al. (1997) found significant increases in respiratory symptoms in children who lived in houses with gas appliances and/or attended a school that used gas heating. Garrett et al (1998) found that respiratory symptoms were more common in children exposed to a gas stove, even though the mean indoor NO_2 levels were low, with a median of six ppb. They also found a statistical association between the presence of a gas stove and asthma in children. The authors suggested that the association with low concentrations on average may be caused by short term exposure to high levels of NO_2 during the use of the gas stove. Cuik et al (2001) also found higher levels of asthma and respiratory symptoms in preschool children that were exposed to unflued gas heaters and cookers. Pilotto et al (2004) looked at the influence of unflued gas heater emissions on the health of children in Adelaide schools and found that when half the schools had the unflued gas heaters replaced with another form of heating that the rates of respiratory symptoms in children with asthma decreased in those schools without the gas heaters. The authors suggested that the reduction in symptoms was due to exposure to lower levels of NO_2.

Australian epidemiological studies have also found associations between ambient NO_2 levels and hospital admissions for respiratory (particularly childhood asthma) and cardiac conditions, particularly in the elderly (EPAV, 2000).

Probably the greatest uncertainty in an epidemiological study is associated with the estimate of each individual's exposure to the pollutant of interest. In urban air quality studies, the traditional approach is to assume that each person in a city has the same exposure. It is well-recognised that there are two assumptions in this approach that are not strictly true. Firstly, air quality on any day is not uniform across a city, i.e. there are spatial gradients, and secondly, most people do not remain in one location over the study period, be it one day or one year or more. Moreover, much time is spent indoors, where air quality is likely to be different from outdoors. The end result is that an individual's true personal exposure can often be quite different to that determined from the 'uniform ambient air quality/fixed site' approach outlined above. Our study aimed to quantify the magnitude of the variation between individuals with different activities and locations and to investigate the feasibility of using ambient air quality models and indoor mass balance models to reduce the uncertainty in assessments of personal exposure.

Linking personal exposure values to ambient exposure values through indoor-outdoor concentration ratios is one way of deriving an estimation of personal exposure from ambient modelled and monitoring data. The ratios vary between microenvironments, and within a microenvironment are usually dependent on various parameters. It is also necessary to know the proportion of time spent in each microenvironment over a period of a day or a week, and this can be obtained from time-activity studies. In Chapter 2, we summarise findings from a literature review of indoor air quality for NO_2 in different microenvironments and use these to not only develop a conceptual model of our current understanding of personal exposure to NO_2, but also to design a measurement programme to investigate some of the less well-known aspects of NO_2 exposure.

On the measurement side of the project, NO_2 data (cumulative) were gathered across Melbourne by between 15 and 17 volunteers wearing personal passive samplers over four two-day periods and maintaining a diary of their activities over these periods. In addition, $PM_{2.5}$ concentrations were continuously measured over the same periods by four volunteers with portable DustTrak™ monitors (TSI inc.). The field work is described in Chapter 3 and results presented in Chapters 4 and 8. Ventilation rates were measured for five houses and the results are used in Chapter 5 with gas cooking information from participants' time-activity diaries to develop models for predicting indoor NO_2 concentrations, and hence indoor-outdoor ratios for each home. The power of the activity-based methodology combined with personal samplers was illustrated by Olaru et al. (2005) who collected sampler data in seven microenvironments from three participants living in the same house, but with different time-activity profiles. At the end of the 5-month study period, there was a difference of 30% in accumulated personal exposure to NO_2 between the participants.

The Australian Air Quality Forecasting System (AAQFS) and CSIRO's air quality model TAPM-CTM were run for each two-day event, and hourly-gridded NO_2 and $PM_{2.5}$ fields were used with EPA Victoria ambient monitoring data to calculate personal exposure for each trip profile. Predictions of on-road NO_2 concentrations using information from a near-road dispersion model, the Lagrangian Wall Model (LWM), were evaluated against concentrations measured while participants were in transit. The models are described in Chapter 6 and analysis of these results is presented in Chapters 7, 8 and 9.

2. CONCEPTUAL MODEL OF NO_2 PERSONAL EXPOSURE

A conceptual model framework for calculating personal exposure to NO_2 is illustrated in Figure 2.1. Different emission sources contribute to pollution levels in different microenvironments (MEs). Total personal exposure (PE) is estimated by weighting exposures in different microenvironments according to the time spent in each microenvironment. Such an approach is based on easy-to-use time-activity diaries. Algebraically, this is expressed as

$$PE = \sum_{i=1}^{n} C_i t_i \Big/ \sum_{i=1}^{n} t_i \ .$$

The pollutant concentration in each microenvironment is dependent upon emission rates of sources in the microenvironment, the rate at which air is exchanged with the external environment (ventilation rate), and the removal rates of the pollutant from the microenvironment (deposition, decomposition, transformation). Our approach in this project begins at the second row of boxes in Figure 2.1, where we measure concentration (exposure) in each microenvironment.

Indoor concentrations of NO_2 can depend on various characteristics of the microenvironment, and these are reviewed in this chapter. In the measurement aspect of our study, we related these microenvironment characteristics from participants' diaries to our measured indoor-outdoor ratios. These ratios were used with outdoor ambient monitoring and modelled data to estimate indoor exposure in each microenvironment and thus personal exposure according to Figure 2.1. These estimates were then compared with our sampler measurements of personal exposure. We did not calculate internal dose.

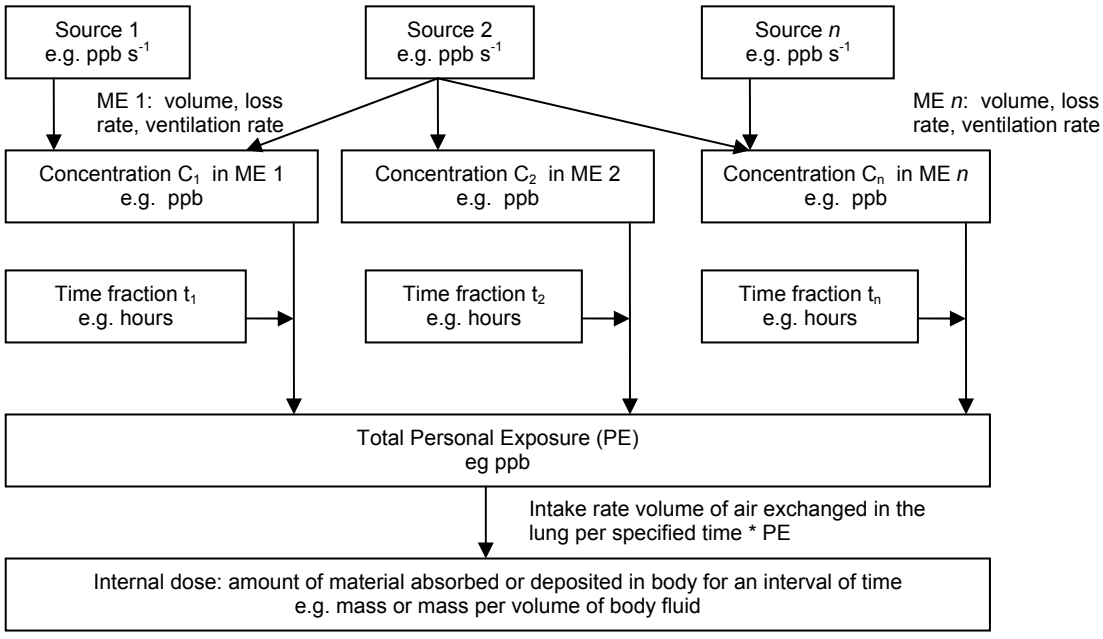

Figure 2.1 The concept of calculating personal exposure using time-activity data and pollutant levels in microenvironments (ME). Adapted from Monn (2001).

According to the Australian Bureau of Statistics (ABS, 1998), Australians over 15 years of age spend their time in various microenvironments according to the percentages listed in Table 2.1. Consequently, while the participants in our field campaign wore a personal sampler at all times, they also wore additional samplers in the microenvironments of home, work and transit (defined here as travelling between home and work). To account for 100% of their time, participants were also issued with a sampler to wear when in none of the above three microenvironments. Accordingly, we review here previous studies of NO_2 in the home, work and transit environments.

Comprehensive reviews of indoor air quality, including NO_2, have been carried out by Brown (1997), DHAC (2000) and FASTS (2002). Based on these reviews and some recent research papers, we present here a brief outline of those aspects of indoor NO_2 that are relevant to our project's microenvironments.

Table 2.1 Time budget for total Australian population for persons 15 years and older.

Environment	Minutes/day in 1992	Percentage of day in 1992	Minutes/day in 1997	Percentage of day in 1997
Home Personal care Domestic activities Child care Voluntary work	775	54	820	57
Work Employment Education	205	14	199	14
Shopping Purchasing goods and services	30	2	29	2
Recreation Social and community interaction Recreation and leisure	299	21	262	18
Transit associated with all environments	70	5	73	5
Outdoor Domestic activities Social and community interaction Recreation and leisure	61	4	54	4

2.1 Exposure to NO_2 in the home microenvironment

2.1.1 Sources and concentrations of NO_2 in homes

Australian and overseas investigations have shown that the major sources of NO_2 in the indoor air of a large number of dwellings and schools is unflued gas heating appliances and cooking appliances. However, cigarette smoking and outdoor air (via ventilation rates) also influence indoor concentrations.

Gas appliances

Indoor gas combustion sources have been identified as the major indoor source of exposure to NO_2 in Australian and overseas homes. Emissions from gas heaters can be very high in NSW, where unflued natural gas space heaters are widely used without restriction and in Western Australia, where 83% of gas heaters in homes are unflued (Farrar et al., 2005). In other states, liquid petroleum gas (LPG) heaters can also be used without a flue or sufficient ventilation. Ferrari et al. (1988) found that NO_2 concentrations in Sydney dwellings exceeded 160 ppb in 50% of cases three hours after lighting unflued gas heaters. Similarly high results were found in NSW school rooms with unflued gas heaters (McPhail and Betts, 1992). Other studies have found that emissions from the pilot light in a gas hot water heater contribute to indoor NO_2 (Lee et al., 2000, Yang et al., 2004). Concentrations measured indoors with and without various gas appliances are listed for a number of studies in Table 2.2. In houses without unflued gas heating, gas cooking has often been identified as the major indoor source of NO_2 (Levy et al., 1998, Monn et al., 1998, Garrett et al., 1999, Lee et al., 2000, Garcia-Algar et al., 2004. The impact of a gas stove on NO_2 concentrations can be seen in the indoor-outdoor concentration (I/O) ratios listed in Table 2.3.

Environmental tobacco smoke

Most emissions of NO_2 from cigarette smoking have been found to be present in aged cigarette smoke, otherwise known as environmental tobacco smoke (ETS). ETS consists of smoke exhaled by the smoker and 'sidestream' smoke, which is emitted from the lit end of a cigarette between puffs (Borgerding and Klus, 2005). In both of these sources, the NO emissions from the cigarette were oxidised within a few minutes to NO_2. Nelson et al. (1998) estimated that 688 µg of NO_2 is emitted per cigarette.

Some studies have found statistical associations between the presence of smokers and elevated indoor NO_2. Levy et al. 1998 found the presence of a smoker with the residence was positively correlated with personal exposure to NO_2, while Algar et al. (2004) found that indoor cigarette smoking was significantly related to indoor NO_2 concentrations. In Australia, Garrett et al. (1998) and Lee et al. (2000) also found significant associations between indoor NO_2 and presence of a smoker. In our study, all homes were smoke free.

Table 2.2 NO_2 concentrations measured indoors in selected studies, illustrating the impact of various sources.

Indoor source(s)	**NO_2 (ppb)**	**Sample period**	**Location**	**Reference**
Gas appliance No gas appliance	17.7 ± 2 8.3 ± 2.5		House, Adelaide	Cuik et al. 2001
None Gas stove Smoker present Multiple sources Unflued gas heater	3.1-3.9 6.4-8.1 5.7-6.7 10.9-14.7 130.7	4-day 4-day 4-day 4-day 4-day	80 houses, Latrobe valley	Garrett et al. 1999
Unflued gas heater	58%>160	1-hour operation	64 houses, Sydney	Ferrari et al. 1988
Unflued gas appliances	190 ± 130	During operation	Around Australia	DEH 2004
Gas stove No gas stove Smoker present No smoker present Gas water heater No gas water heater	13.6 ± 6.2 9.1 ± 4.7 14.9 ± 7.7 9.9 ± 5.0 13.2 ± 5.1 9.8 ± 5.5	2-day 2-day 2-day 2-day 2-day 2-day	87 houses, Brisbane	Lee et al. 2000
None	9.9 (6.7-13.8)	24-hr	Melbourne house, 49 days, winter	Powell 2001
None, summer None, winter	6.9 9.6	7-day 7-day	Melbourne house, 8wks summer, 8 wks winter	Dunne et al 2006
None	8.8 (3.1-17.4)	24-hour	Wallsend, NSW, 51 days, autumn	O'Leary 1999
All houses Peak conc- gas cooker Peak conc. no gas cooker	8.6 (6.8-11.0) 34.0 (25.8-43.6) 13.3 (9.7-18.9)	3-day During operation During operation	Kitchens of 53 non-smoking homes, summer	Franklin et al 2006
Unflued gas heater No unflued gas heater Unflued gas cooker No unflued gas cooker Outdoors	22.6, 23.5, 18.3 13.0, 15.7, 10.1 17.2, 21.1, 13.3 16.7, 16.0, 13.9 9.2	3-day 3-day 3-day 3-day	Living, kitchen, bedroom Homes with/without gas appliances in winter	Farrar et al 2005
Unflued gas heater No unflued gas heater Unflued gas cooker No unflued gas cooker Outdoors	8.2, 8.5, 7.2 8.8, 9.1, 8.2 8.9, 9.6, 8.1 8.1, 8.0, 7.4 7.7	3-day 3-day 3-day 3-day	Living, kitchen, bedroom Homes with/without gas appliances in summer	Farrar et al 2005
No gas appliances Gas appliances	5.6[G] 15.5[G]	7-day	Living rooms of 140 houses in ten of the 17 health regions in NSW	Sheppeard et al 2006
House with gas cooker Gas cooking peak conc.	16 (5-34) 304 (60-800)	7-day During operation	Kitchens of 15 houses, Adelaide, SA	Steer et al 1990
All houses Gas cooker No gas cooker	22.6[G] (5.1-61.9) 18.6 10.5	24-hr	Living rooms of 28 houses, 1 month, Brisbane	Yang et al 2004
Kitchen, gas cooker	22.4±7.4 ppbv),	7-day	Kitchen of 1 house, 22 weeks, Melbourne	Keywood et al 1998

G=Geometric mean

Table 2.3 Indoor-outdoor NO_2 concentration ratios (I/O ratio) in various studies with and without indoor gas stoves.

Indoor source	I/O ratio	Sample period	Location	Reference
Gas stove No gas stove	1.19 0.69		Multi-national study	Levy et al. 1998
Gas stove No gas stove	1.03 0.67	24-hour	87 houses, Brisbane	Lee et al. 2000
No gas stove	0.8	4-day	80 houses, Latrobe valley	Garrett et al.1999
Gas stove No gas stove	0.9 ± 0.3 0.7 ± 0.3	24-hour	28 houses, 30 days, Brisbane	Yang et al. 2004
No gas stove No gas stove	1.03 ± 0.13 0.69 ± 0.10	7-day 7-day	1 home, 8 weeks, Melbourne, summer 1 home, 8 weeks, Melbourne, winter	Dunne et al. 2006
None	0.64 (0.38-.17)	24-hour	1 house, 49 days, Melbourne, winter	Powell 2001
None	0.78 (0.41-2.76)	24-hour	1 house, 51 days, Wallsend, autumn	O'Leary 1999

Contribution from outdoor NO_2

Many studies have found strong associations between indoor and outdoor NO_2. These associations are usually strongest in houses with few indoor sources and high ventilation rates. For example, Yang et al. (2004) found that in both Brisbane (Australia) and Seoul (Korea), there was a significant association between outdoor NO_2 and indoor NO_2. This means that if household ventilation can be estimated, then the contribution to indoor NO_2 from outdoors may be able to be estimated using ambient monitoring network concentrations.

Ventilation is recognised as a significant influence on indoor NO_2 concentrations (Algar et al., 2004). Ventilation affects indoor concentrations by allowing mixing of outdoor air with indoor air. This process can act to dilute indoor concentrations if there are strong indoor sources of NO_2 or it can increase indoor concentrations if outdoor air contains high concentrations of NO_2. Ventilation rate is expressed in air changes per hour (ach or h^{-1}). Different forms of ventilation can be defined as the following:

Infiltration is defined as the air exchange between outdoor air and indoor building air when the building is in its closed up state. Thus the air exchange occurs through cracks, spaces and fixed ventilators in the building shell.

Natural ventilation is defined as air exchange between the building interior and exterior through the same processes as infiltration and additionally through controllable openings such as vents, windows and doors.

The dynamics of infiltration and natural ventilation rely on a pressure differential between inside and outside air caused by external air advection or density differences due to temperature gradients between indoors and outdoors. Thus infiltration and ventilation rates vary according to meteorological conditions outside, temperature differentials between inside and outside and whether windows and doors are open. Natural ventilation is commonly used in single- and double-storey residences in Australia and may include some mechanical ventilation such as extraction fans in the kitchen, bathroom and toilet.

Mechanical ventilation is defined as airflow between outdoors and indoors using active ventilation systems. In Australia this form of ventilation is used (a) when the building design cannot allow sufficient natural ventilation such as high rise apartments; and (b) with evaporative cooling for air conditioning in hot dry climates.

Ventilation rates of buildings (whether domestic or commercial) have varied greatly in recent decades due to a range of factors such as energy conservation practice, changes to building regulations and building practices, and variations in ventilation standards and codes. Limited evidence now indicates that air infiltration rates in some new Australian dwellings are below levels considered overseas as essential for good indoor air quality (Brown, 1997).

Typical infiltration and natural ventilation rates measured in Australian residences are listed in Table 2.4.

Table 2.4. Air exchange rates determined for Australian residences.

Study description	**Ventilation type**	**Air change rate per hour, h^{-1}**	**Type of measurement**	**Author**
Unoccupied houses in Melbourne	Infiltration, no wind Canberra estimate Sydney estimate Hobart estimate Melbourne estimate	0.33 0.44 0.55 0.55 0.57	Pressurization Calculation Calculation Calculation Calculation	Biggs cited in Brown 1997
9 new houses in 1985 Perth	Infiltration	0.05 - 0.41	Tracer gas release	Harrison cited in Brown 1997
30-yr 3-bdr unit, winter Melbourne	Natural ventilation average over 7 weeks	0.5	Modelled	Powell & Ayers 2007
14 houses, Brisbane	Infiltration Natural ventilation	0.61 ± 0.45 3.00 ± 1.23	CO2 depletion CO2 depletion	He et al 2005
20-year house in Melbourne	Infiltration Natural ventilation	0.23 ±0.03 winter 1.4 ± 0.1 summer	CO2 release CO2 release	Dunne et al 2006
20-yr house 5-yr bungalow 30-yr house 40-yr house	No openings (infiltr.) 0.02 m^2 open window 0.02 m^2 open window 0.34 m^2 open window	0.29 0.41 0.55 0.90	CO2 release CO2 release CO2 release CO2 release	Unpublished, CMAR 2008
28 houses, Brisbane	Natural ventilation all House pre-1990 House post-1990	1.44 1.76 1.32	Modelled Modelled Modelled	Yang et al 2004
43 houses,Sydney	Infiltration, winter	0.9 (0.2-2.3)	SF6 tracer release	Ferrari et al 1988
Houses<5yrs old Sydney, winter	Infiltration	0.33	Tracer gas release	Ferrari 1991 cited in Brown 1997
116 houses measured during heater use	During heater use ventilation all Sydney Canberra Victoria 7-house comparison 7-house comparison	1.1 average of all 1.1 (0.12-3.8) 0.91 (0.26-2.8) 1.2 (0.18-3.4) 1.4 (0.60-2.0) 1.3 (0.97-1.8)	CO2 depletion CO2 depletion CO2 depletion CO2 depletion PFC tracer release CO2 depletion	DEH 2004
Kitchens of 15 houses, Adelaide	Natural ventilation during cooker use	1.4 (0.3-4.1)	CO depletion SF6 tracer release	Steer et al 1990
1 House	All openings closed Windows, doors open	0.3-0.6 6	CO depletion SF6 tracer release	Steer et al 1990

PFC = perfluorocarbon
SF6 = sulphur hexafluoride
CO2 = carbon dioxide

2.1.2 Summary

Based on the above discussion, we would expect to see the following home exposure characteristics:

- Higher I/O ratios in homes using gas appliances. The absence of use of an extraction fan or external ventilation when using gas appliances results in higher concentrations indoors.
- A 'tight' home with a low infiltration rate will increase the I/O ratio if the indoor sources are strong.
- In the absence of indoor sources, I/O ratios in homes are likely to be closest to I/O=1 if the homes have high air exchange rates with outdoors (ie many windows and doors are left open).
- It is also expected that homes with low infiltration, no gas appliances and closed doors and windows would register low I/O values.

Study participants were requested to note the above home and activity characteristics in their diaries, and infiltration rates were measured for some of the homes. These were used in an analysis of I/O ratio results in chapter 5.

2.2 Exposure to NO_2 in the work microenvironment

The sources of NO_2 in the workplace are too broad to cover as they can vary across industries. In our study, all participants who spent the day inside worked in mechanically-ventilated offices with no sources of NO_2 – smoking indoors is banned. Thus we would expect indoor concentrations to be related to outdoor concentrations, but lower because of deposition onto indoor surfaces. In our project, NO_2 concentrations were measured simultaneously inside and outside office buildings. However, we could find no studies reported with similar measurements, though there are data for NO_2 concentration inside offices (e.g. Lee et al., 2000).

2.3 Exposure to NO_2 in the transit microenvironment

The transit microenvironment in our project covers any mode of transport, including walking. Comparison studies of pollutant exposure between different modes of transport have been carried out for Sydney by Chertok et al. (2003), for Perth by Farrar et al. (2001), for Hong Kong by Chan et al. (1999) and for London by Adams et al. (2001). Our study was not designed to compare modes (because participants were travelling on different routes), but was designed to compare the personal exposure in the transit microenvironment to that found for the home and work microenvironments.

Sources of NO_2 during transit in correctly-functioning vehicles originate from outdoors. Thus, the biggest influences on the indoor concentrations are the ventilation within the cabin and the concentrations outdoors. The in-cabin concentration is affected by whether windows are open or closed, whether the vent is set to external or recirculation, and the speed of the fan. NO_2 is

removed by deposition to surfaces within the vehicle and it is to be expected that the ratio of in-vehicle to out-vehicle concentrations will be less than 1. The cabin surface area to volume ratio also affects the concentration (Keywood et al., 1998).

The I/O ratio for NO_2 has been measured by Cains et al. (2003) in the M5 tunnel. Passive samplers were placed inside the car and also attached to the outside on the roof. Concentrations were measured for three combinations of in-cabin settings, as shown in Table 2.5. The results showed that in-vehicle concentrations were virtually the same as outside the vehicle (mean concentration 207 ppb) when windows were open, but that this value can be reduced by more than 70% when the windows and external vent are closed (fan on recirculate).

Table 2.5 Ratios of in-vehicle to out-vehicle NO_2 and carbon monoxide (CO) concentrations measured in Sydney's M5 tunnels by Cains et al. (2003).

Cabin Ventilation State	**NO2**	**CO**
Windows up, Fan on recirculate, No A/C	0.30	0.23
Windows up, Fan on recirculate, A/C on	0.25	0.25
Windows down	0.96	0.98

2.4 Our conceptual model

The main aim of this project was to investigate methods of using ambient concentrations (both monitored and modelled) of NO_2 to estimate personal exposure, by linking the two via an indoor-outdoor ratio. We have chosen to follow the approach outlined in Monn (2001) in which total personal exposure is estimated by weighting exposures in different microenvironments according to the time spent in each microenvironment. Following a literature search, we investigated I/O ratios in our project by simultaneously measuring with passive samplers indoor and outdoor concentrations in the home, work and transit microenvironments, as well as introducing an additional 'other' sampler to be worn in any other microenvironments. Participants' activity diaries were designed to reflect previous findings that indoor concentrations in the home are a function of the use of gas stoves and heaters, and that concentrations in vehicles have been shown to depend on ventilation settings. Ventilation rates in homes have also been found to be important and these were measured in a sample of participants' homes.

It is impractical in epidemiological studies to measure the concentration and time spent in every microenvironment for each individual in a cohort. However by considering only those microenvironments in which a considerable amount of time is spent, and estimating the indoor concentrations in those microenvironments according to common characteristics, it is likely that realistic estimates of personal exposure can be obtained.

3. FIELD EXPERIMENTS

NO_2 data (cumulative) were gathered across Melbourne by between 15 and 17 volunteers wearing personal passive samplers for each of four two-day periods and maintaining a diary of their activities over these periods. In addition, $PM_{2.5}$ concentrations were continuously measured over the same periods by 4 volunteers with portable DustTrak™ monitors (TSI inc.). Both working and non-working participants were included in the study. All participants were non-smokers. The study was done for a total of four separate two-day events, in April 2007, May 2006, May 2007 and June 2006. These times of year were chosen for the stable light-wind conditions in order to maximize concentrations and the spatial variation in concentrations across the city and suburbs.

Participants wore a small Ferm-type passive gas sampler (about 2.5 cm diameter), attached to chest clothing, that measures the ambient concentration of NO_2 (Keywood et al., 1998; Beer et al., 2001). DustTrak monitors for $PM_{2.5}$ were carried by three people for each Event. An additional DustTrak was installed in the house of a non-working participant.

The NO_2 samplers were analysed by CSIRO Marine and Atmospheric Research (CMAR) at the end of the 48-hour period and the cumulative NO_2 uptake over that period was obtained. From this value, an average concentration for the period of exposure can be calculated. The term 'passive' is used because the method uses a passive diffusion process rather than an active pumping process to collect the nitrogen dioxide gas. A filter within the sampler is coated with a chemical that reacts with nitrogen dioxide gas and stores the by-product on the filter. As the gas is removed from the air onto the filter, more nitrogen dioxide is drawn into the sampler to replace the nitrogen dioxide lost from the air. This process continues until all the chemical on the filter has reacted or its exposure to nitrogen dioxide ceases.

Sampler measurements were compared to data from the EPA Victoria monitoring network. Hourly-averaged concentrations of NO_2 were collected at the ten locations shown in Figure 3.1. The RMIT station (RMI) was not in operation for the two Events in 2007. $PM_{2.5}$ data were gathered at Alphington and Footscray using continuous TEOM® particulate monitors (Thermo Scientific, USA), while hourly values of a backscatter index, which can be converted to $PM_{2.5}$ concentrations, were measured by nephelometry at all stations except Richmond and Altona North.

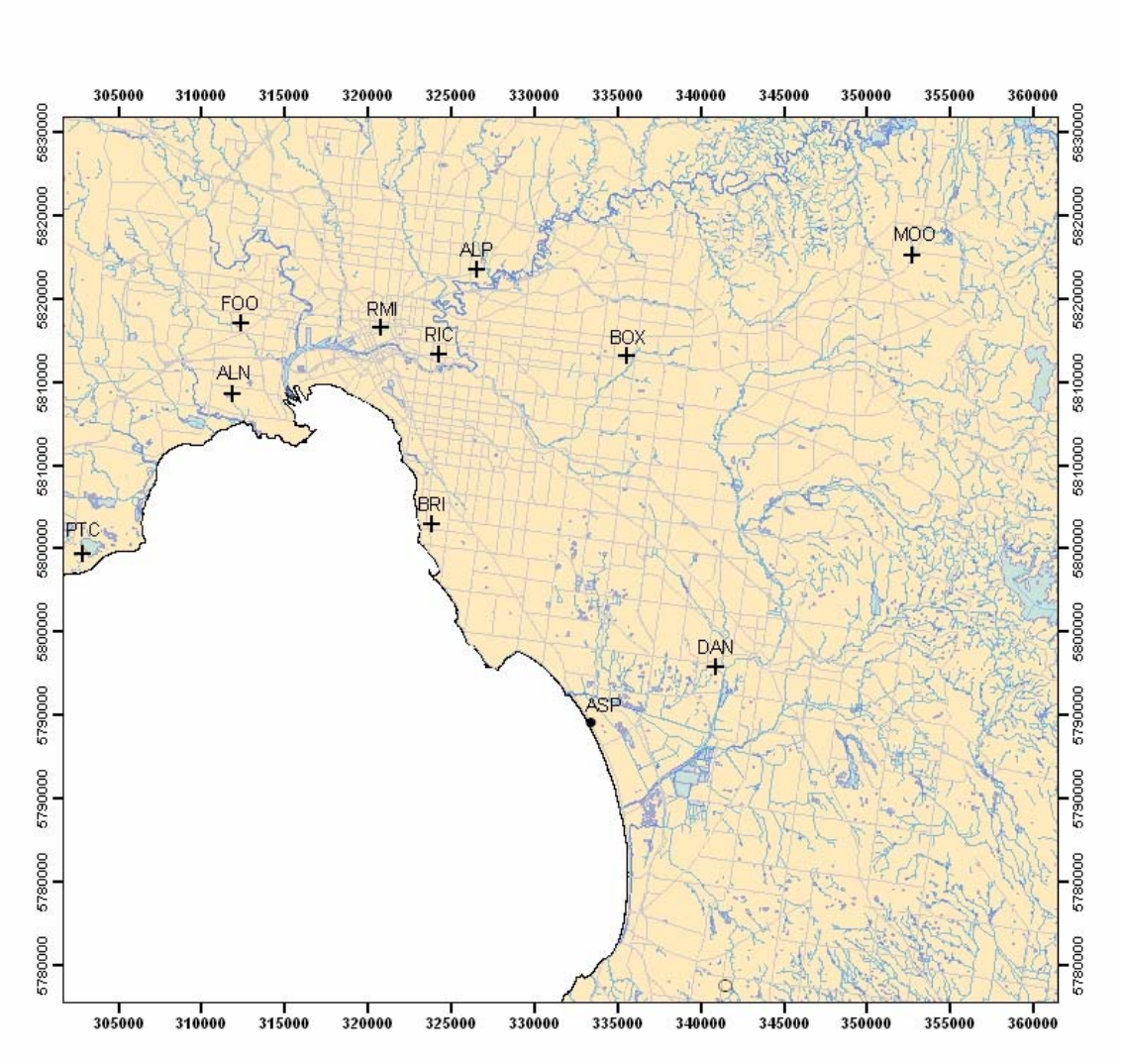

Figure 3.1 Locations of EPA Victoria monitoring stations (+) and the CSIRO Bayside monitoring station (●) at Aspendale (As). Main roads and waterways are also shown.

3.1 Logistics and activities

Events covered a 48-hour period beginning and ending at 2100 hours local time (9 pm). Home and workplace locations across Melbourne were chosen to maximise the variation in exposure arising from location and activity.

Table 3.1 shows the number of different microenvironments in which each pollutant was measured. The number of volunteers wearing samplers was 16, 17, 16 and 15 for Events one to four, consisting of 24 different people. Personal sampler data were found to be invalid for two volunteers, reducing the number of valid samples to 62. Three individuals participated in all four events while 11 participated in three events. Locations of homes and workplaces are shown in Figure 3.2, with participants' home sites grouped (by symbol) according to work location. Five work locations in the central business district (CBD) are grouped under the location denoted as CBD. The transit modes (and trips) included car (30), train (8), tram (4), bus (2), bicycle (6) and pedestrian (2). On 12 occasions, people were based at home over an event.

Participants wore two sets of NO_2 passive samplers at all times, each set containing two samplers to enable precision checking to be done. One set was worn at all times throughout the 48-hour period and the second set depended on which microenvironment (home, work or transit between work and home) was being experienced. When in none of the above environments, volunteers wore a set of samplers labelled 'other' for their second set. 'Other' included such activities as shopping, visiting friends, attending the cinema etc.. When not being worn, a sampler was closed by returning it to a canister fitted with a lid. An additional pair of samplers was placed outside a participant's home and workplace and opened only while the volunteer was in that environment. In this way, ratios of indoor to outdoor concentrations were obtained. For vehicles, a sampler was placed on the mirror-side of the side mirror to enable ratios of in-car to out-car concentrations. Activity diaries were kept by each person, noting times of arrival and exit in the different environments. At home, details of heating and cooking appliances and times, and open doors and windows were noted.

Over the same period, DustTrak $PM_{2.5}$ monitors accompanied three participants at home, at work and in vehicle transit between the two sites, while a further monitor was installed in one home for the full period. Throughout one event, a monitor travelled in the cabin of a delivery truck. Concentrations were logged at minute intervals.

Table 3.1 Measurement statistics over the four Events.

	Personal exposure measurements	Different Home Locations	Different Work Locations	Different Transit Trips
NO_2	62	24	8	25
$PM_{2.5}$	15	8	3	8

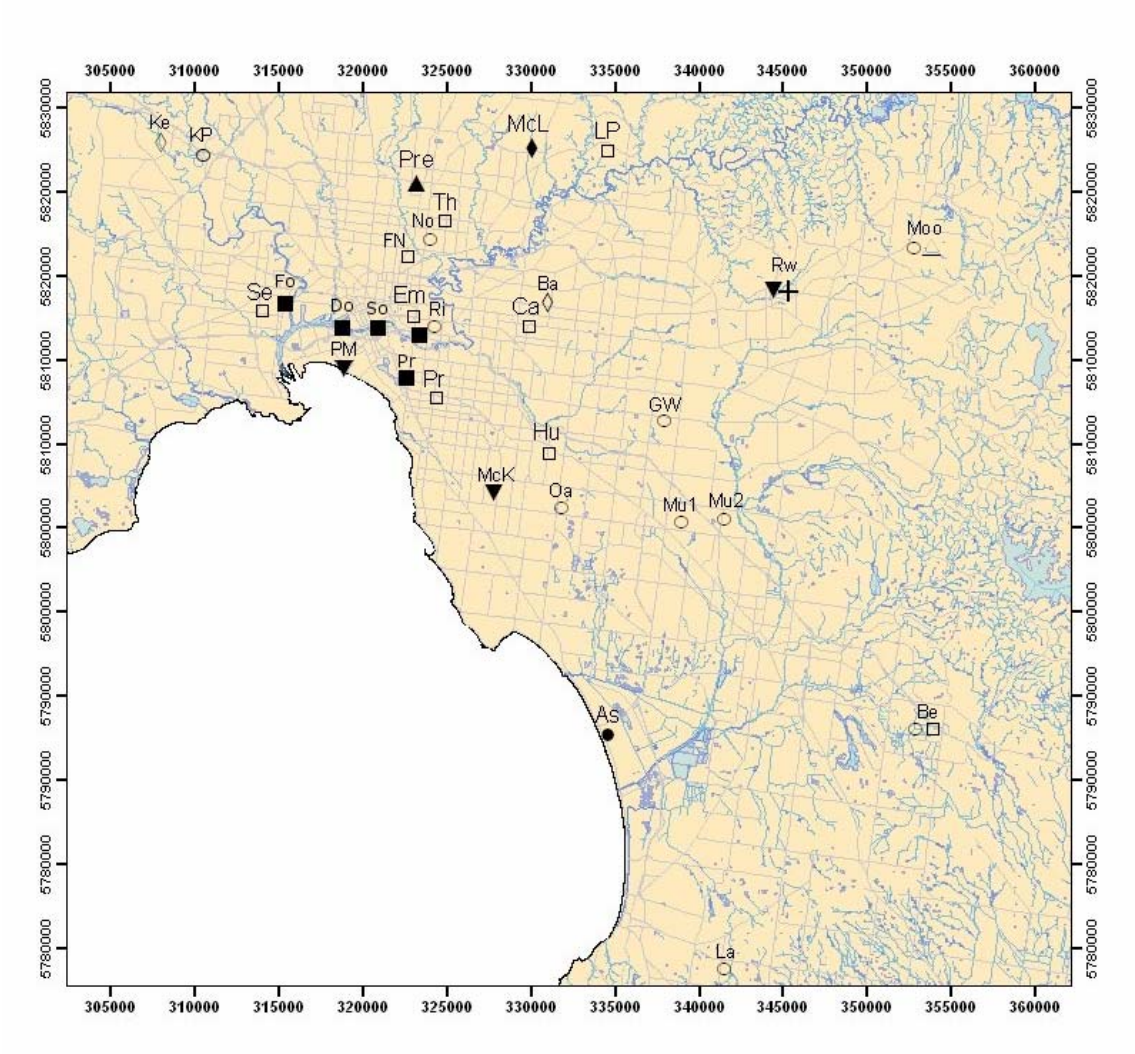

Figure 3.2 Work and home locations around Melbourne, covering all events, of participants wearing NO_2 samplers for the personal sampling project. Main roads and waterways are also shown. Symbols denote the following groupings. ● CMAR Aspendale, ○ Trip to Aspendale, ♦ EPA Victoria McLeod, ◊ Trip to McLeod, ■ CBD locations, □ Trip to CBD, ▲ Preston, Δ Trip to Preston, ▼ Based at home, + Working outdoors.

3.2 Event 1 25-26 May 2006

3.2.1 Synoptic situation

The mean sea-level pressure (MSLP) charts for each day in Figure 3.3 show Melbourne under the influence of a weak ridge between two low pressure systems over South Australia and the Tasman Sea. Winds were light on both days, with the Victorian Regional Office of the Bureau of Meteorology summarising the weather as follows.

25 May 2006. Early mist patches clearing to a cloudy afternoon and evening, mainly low cloud but some high cloud early in afternoon. Light southerly breeze. Maximum temperature 15.2^{o}C, minimum temperature 6.4^{o}C.

26 May 2006. Early fog and mist patches clearing to a hazy day with mainly high cloud and a little isolated low cloud. Winds were light and variable. Maximum temperature 14.9^{o}C, minimum temperature 4.6^{o}C.

Wind speed and direction at a height of 10 m for the two days at Footscray monitoring station are shown in Figure 3.4. Apart from the afternoon and early evening of 25 May, wind speeds were below two m s^{-1} and with a southerly component for most of the time. Wind behaviour at Footscray was typical of all sites across Melbourne, apart from the coastal site of Point Cook where winds were stronger.

NO_2 and $PM_{2.5}$ concentrations at the inner-suburban EPA Victoria monitoring station Footscray are shown in Figure 3.5. NO_2 levels during the daytime were around 20 ppb, a product of titration of nitrogen oxide (NO) by background ozone, levels of which in winter are typically 25 ppb. On the afternoon of 26 May, NO_2 levels rose to 40 ppb. The sharp rise was also observed at all other monitors, except Point Cook, Dandenong and Mooroolbark. $PM_{2.5}$ concentrations, between five and 15 μg m^{-3} for most of the period, also rose sharply on the second afternoon.

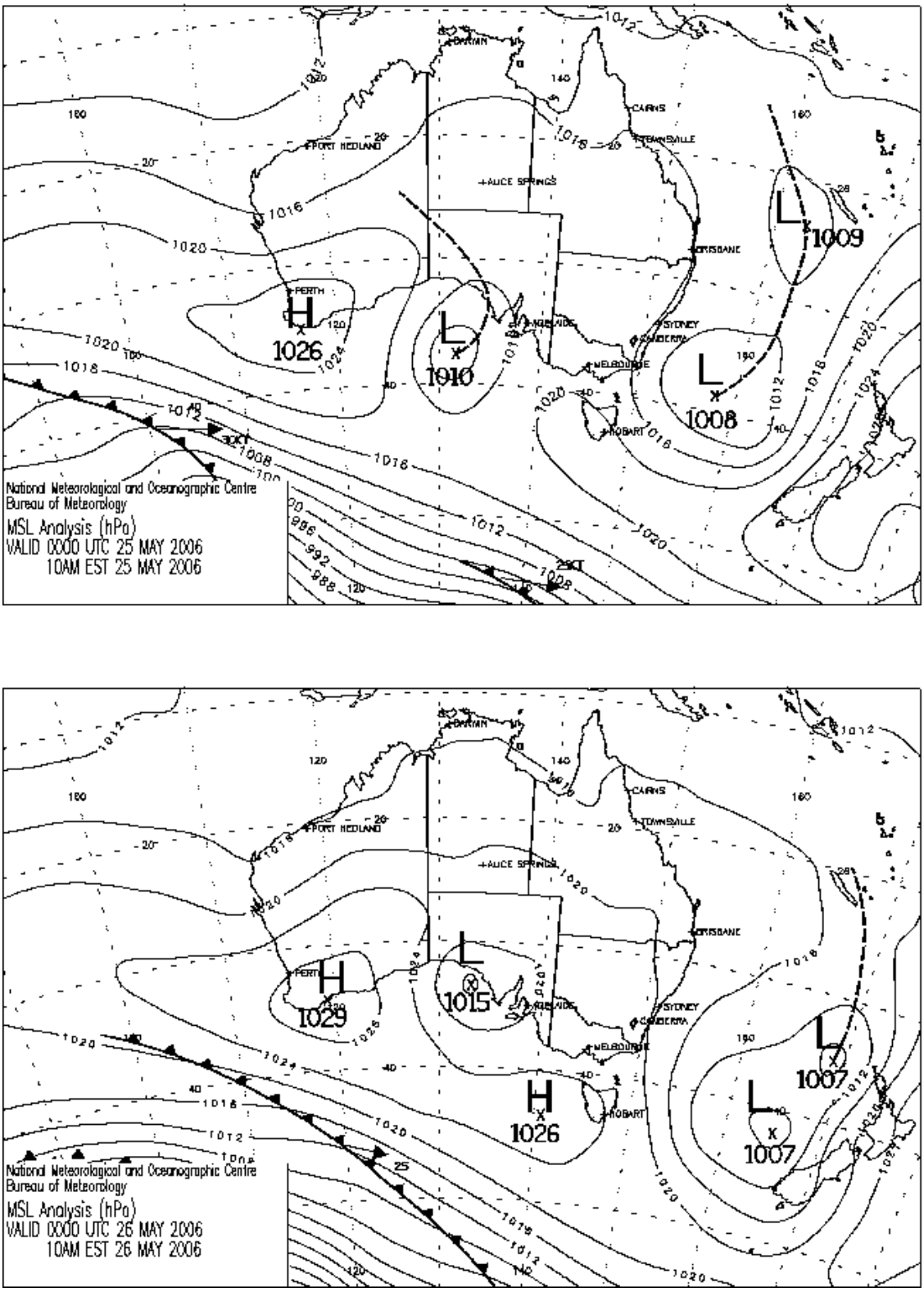

Figure 3.3 Mean Sea-Level Pressure charts at 1000 Eastern Standard Time on 25 May 2006 (top) and 26 May 2006 (bottom).

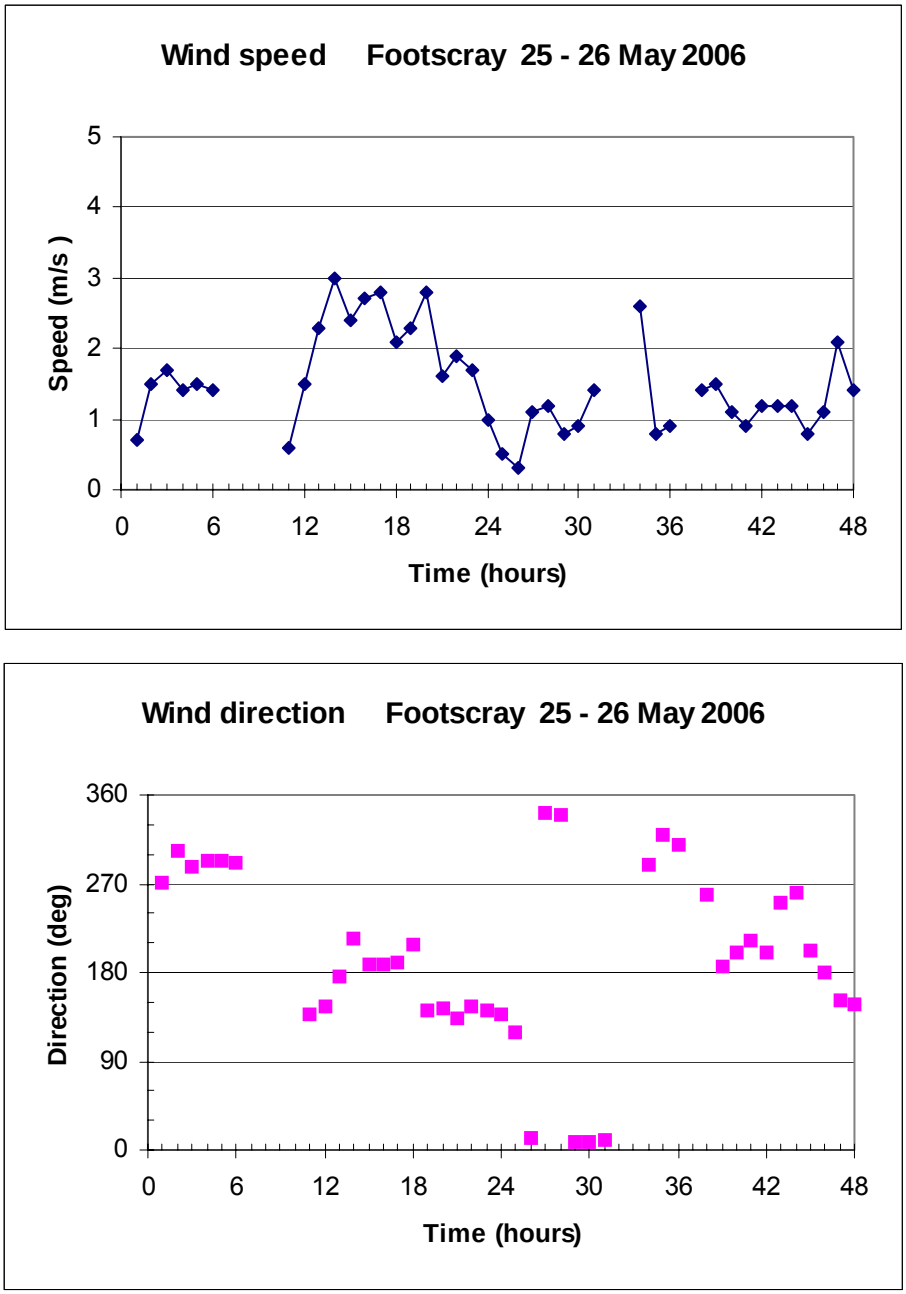

Figure 3.4 Wind speed (top) and direction (bottom) at EPA Victoria's Footscray monitoring station for the period 25-26 May 2006.

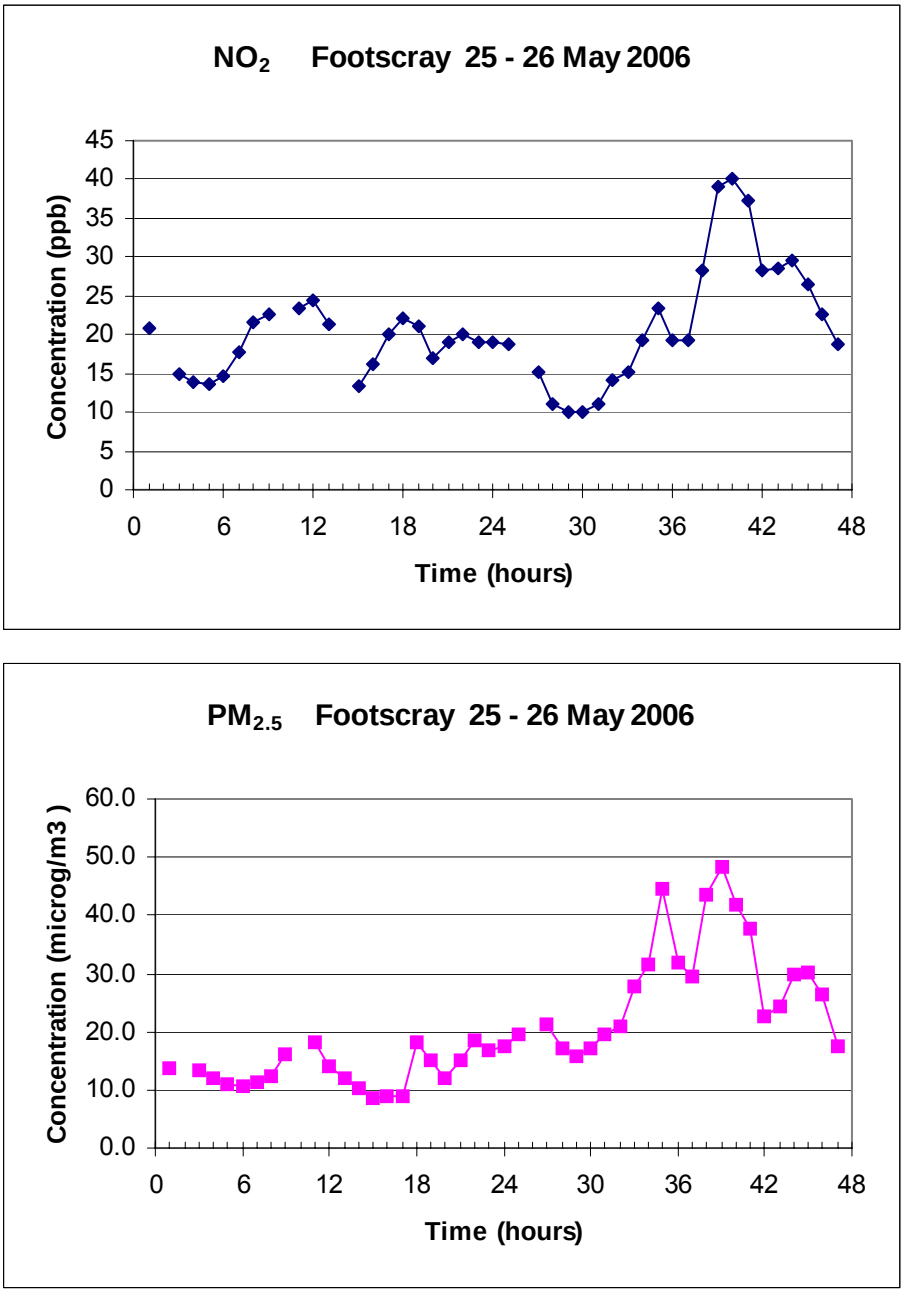

Figure 3.5 Concentrations of NO_2 (ppb) (top) and $PM_{2.5}$ (μg m^{-3}) (bottom) at EPA Victoria's Footscray monitoring station for the period 25-26 May 2006.

3.3 Event 2 7-8 June 2006

3.3.1 Synoptic situation

The mean sea-level pressure (MSLP) charts for each day in Figure 3.6 show Melbourne near the centre of a large high pressure system. Winds were light on both days, with the Victorian Regional Office of the Bureau of Meteorology summarising the weather as follows.

7 June 2006 Early fog patches clearing to a hazy day with isolated low cloud. Moderate northerly winds backing light south to southeast later. Maximum temperature 15.9 ^{o}C, minimum temperature 4.8 ^{o}C.

8 June 2006 Hazy day with high cloud. Moderate northerly winds backing light south to southeasterly in the afternoon. Maximum temperature 14.6 ^{o}C, minimum temperature 3.7^{o}C.

Wind speed and direction at a height of 10 m at Footscray monitoring station are shown in Figure 3.7. At this site, and across the network, winds were less than two m s^{-1} for virtually the entire period. During the night, wind direction was northerly, but changed to south to southeasterly during the daytime.

NO_2 and $PM_{2.5}$ concentrations at the inner-suburban monitoring station Footscray are shown in Figure 3.8. NO_2 levels during the daytime were above 30 ppb on both days, perhaps indicating a little photochemical activity. Levels at Footscray on 7 June were typical of those in the western suburbs, but on 8 June concentrations were more even across the CBD and inner to middle suburbs. Maximum $PM_{2.5}$ concentrations, between 25 and 30 μg m^{-3} were observed on both evenings as the near-surface atmosphere stabilised.

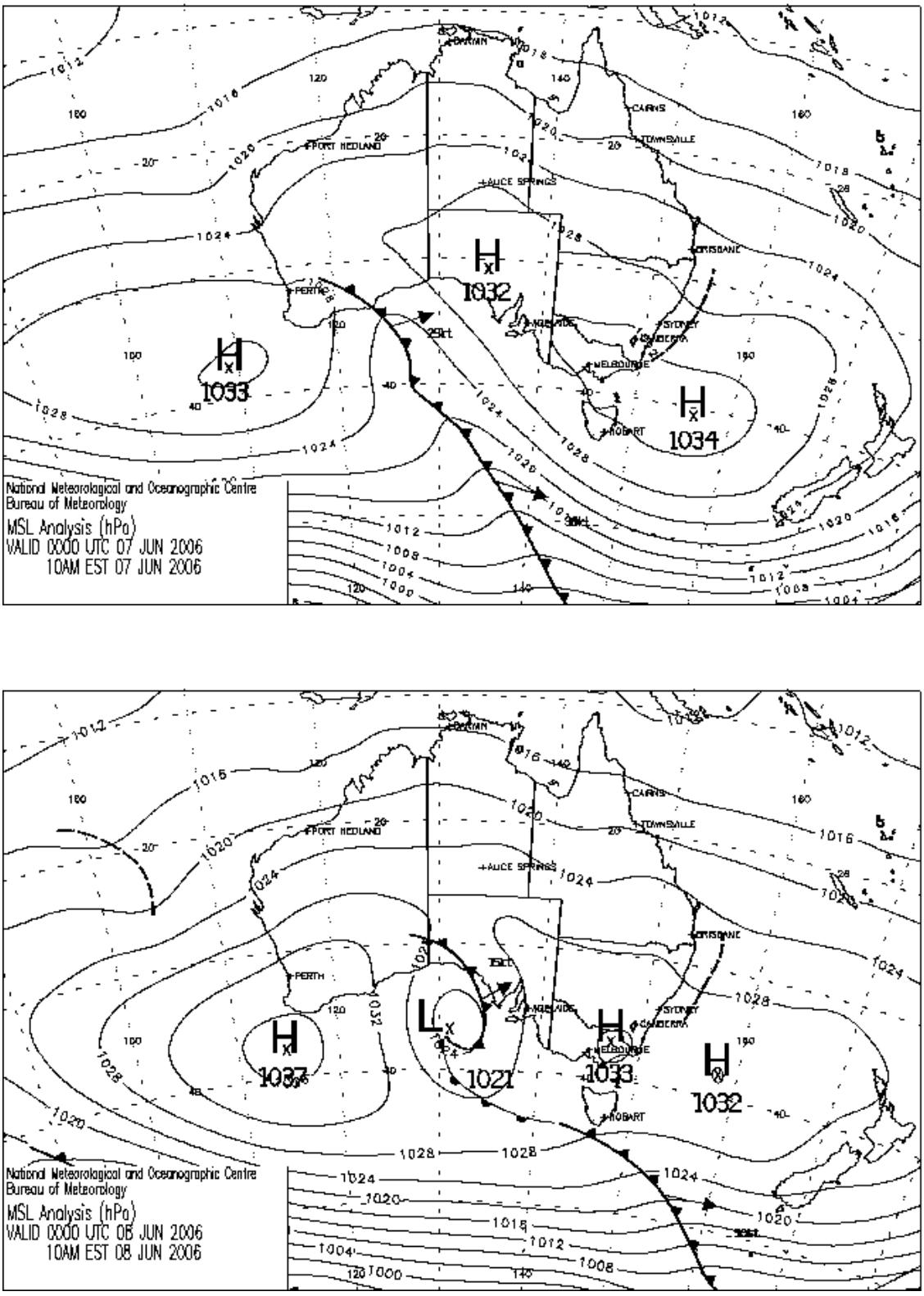

Figure 3.6 Mean Sea-Level Pressure charts at 1000 Eastern Standard Time on 7 June 2006 (top) and 8 June 2006 (bottom).

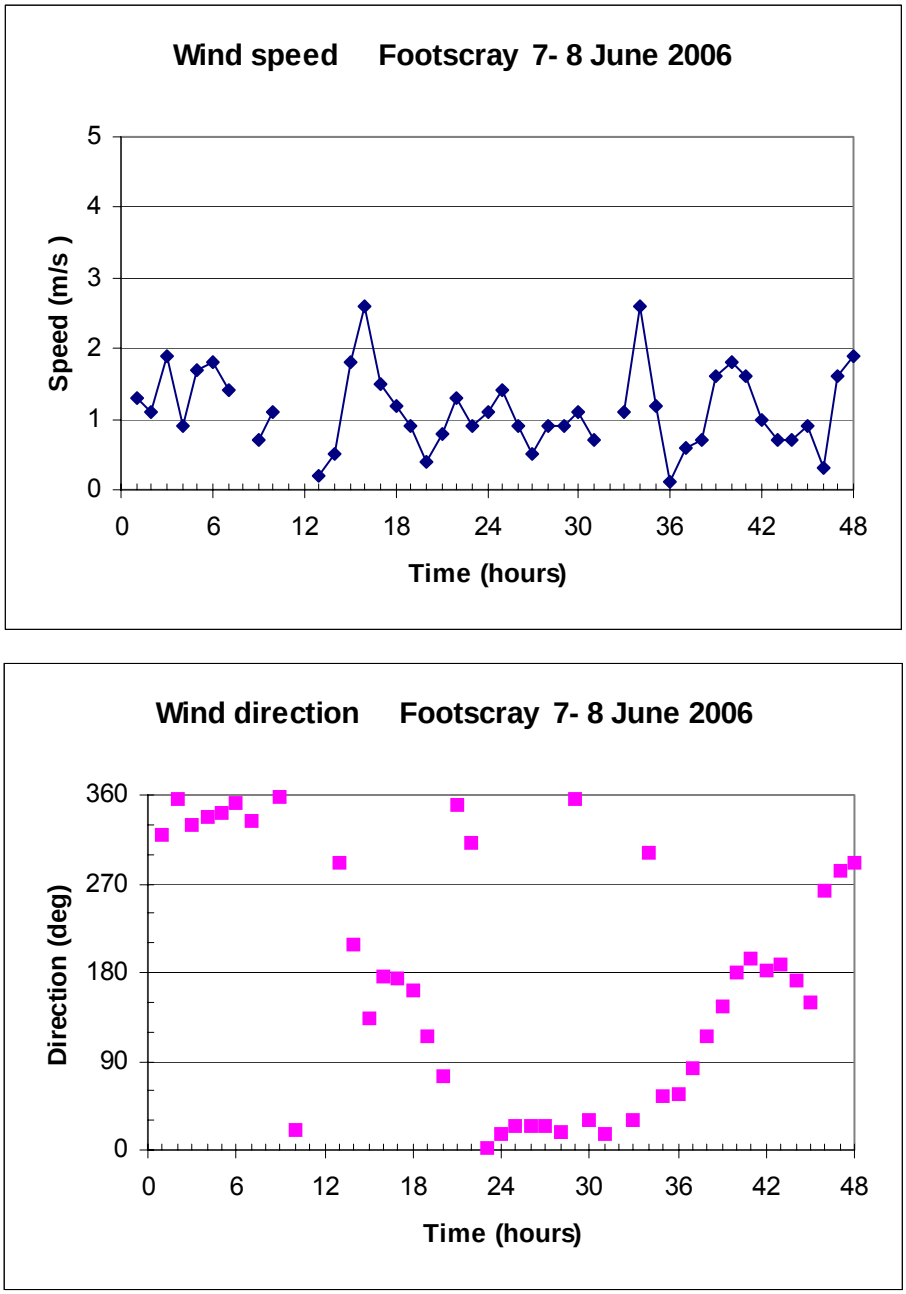

Figure 3.7 Wind speed (top) and direction (bottom) at EPA Victoria's Footscray monitoring station for the period 7-8 June 2006.

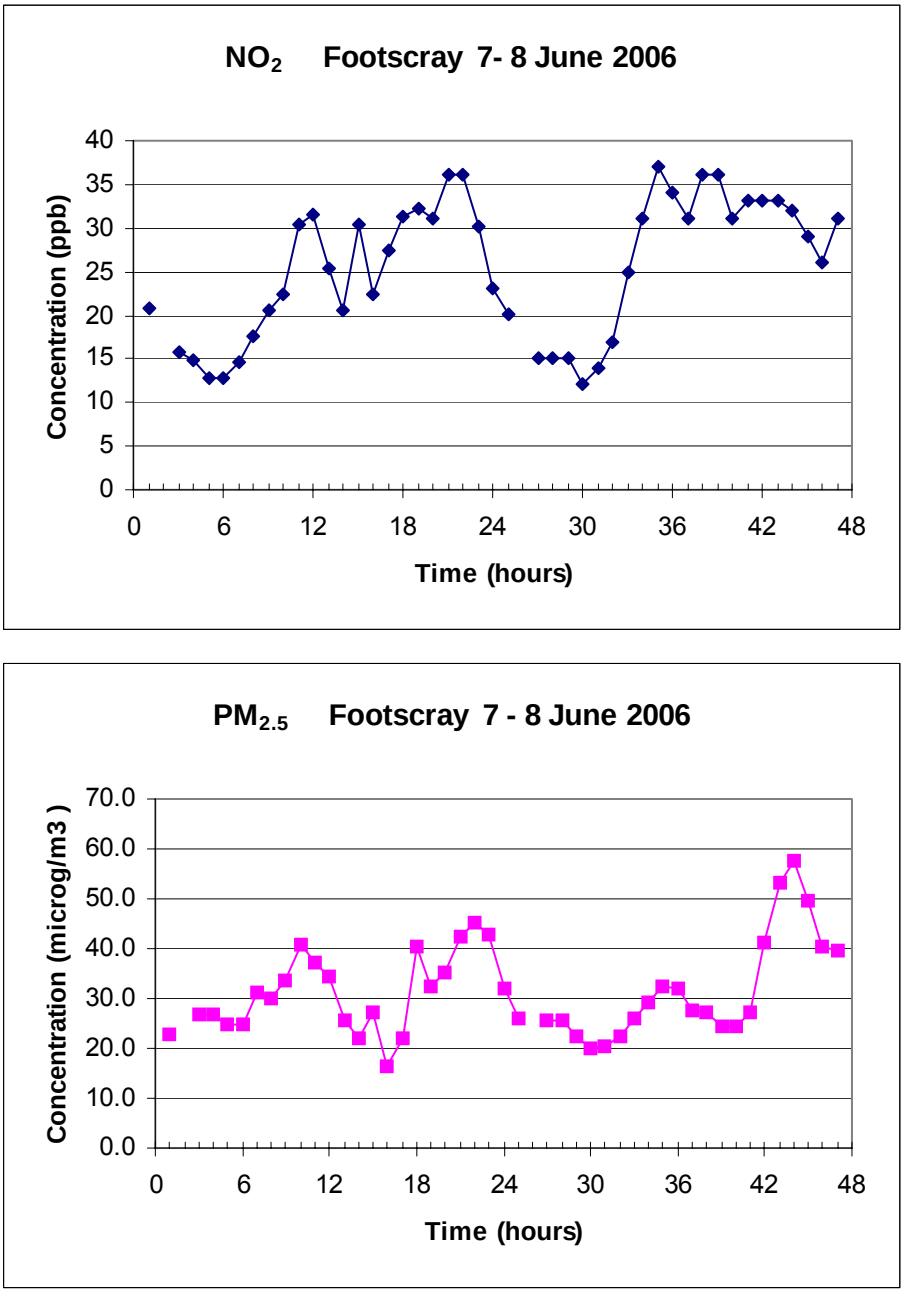

Figure 3.8 Concentrations of NO_2 (ppb) (top) and $PM_{2.5}$ ($\mu g\ m^{-3}$) (bottom) at EPA Victoria's Footscray monitoring station for the period 7-8 June 2006.

3.4 Event 3 12-13 April 2007

3.4.1 Synoptic situation

The mean sea-level pressure (MSLP) charts for each day in Figure 3.9 show that a large high pressure system passed just to the south of Melbourne over the two days. Winds were light in the morning and moderate in the afternoon on both days and a little smoke was present, with the Victorian Regional Office of the Bureau of Meteorology summarising the weather as follows.

Thursday 12 April 2007. Hazy day after some early cloud, smoke haze (pollution) in PM. Visibility reduced to 10km at times. Light northerly wind in the morning turning moderate southerly in the afternoon. Max temp: 21.3ºC. Min temp 10.0ºC.

Friday 13 April 2007 Hazy / smokey (pollution) again with visibility reduced to 6km in afternoon. Light northerly wind in the morning turning moderate southerly in the afternoon. Max temp: 24.6ºC. Min temp 10.4ºC.

Wind speed and direction at a height of 10 m at Footscray monitoring station are shown in Figure 3.10. At this site, and across the network, wind speeds are less than two m s^{-1} at night and early morning, increasing to 2.5 to 3 m s^{-1} in the afternoon. During the night, wind direction at Footscray was northwesterly, but changed to south by noon, before reverting to a northerly component in the early evening. At stations in the eastern suburbs, the very light northerly winds at night and early morning had an easterly component.

NO_2 and $PM_{2.5}$ concentrations at the inner-suburban monitoring station Footscray are shown in Figure 3.11. NO_2 levels during the daytime were around 15-20 ppb on the first day and rose to around 40 ppb at most stations on the second afternoon, coinciding with the winds turning southerly. Smoke levels were at their highest on this afternoon too. On the first day, maximum $PM_{2.5}$ concentrations of around 20 μg m^{-3} were observed at most stations in mid-afternoon. Similarly, maxima were measured mid-afternoon on the second day with concentrations typically reaching 40 μg m^{-3}.

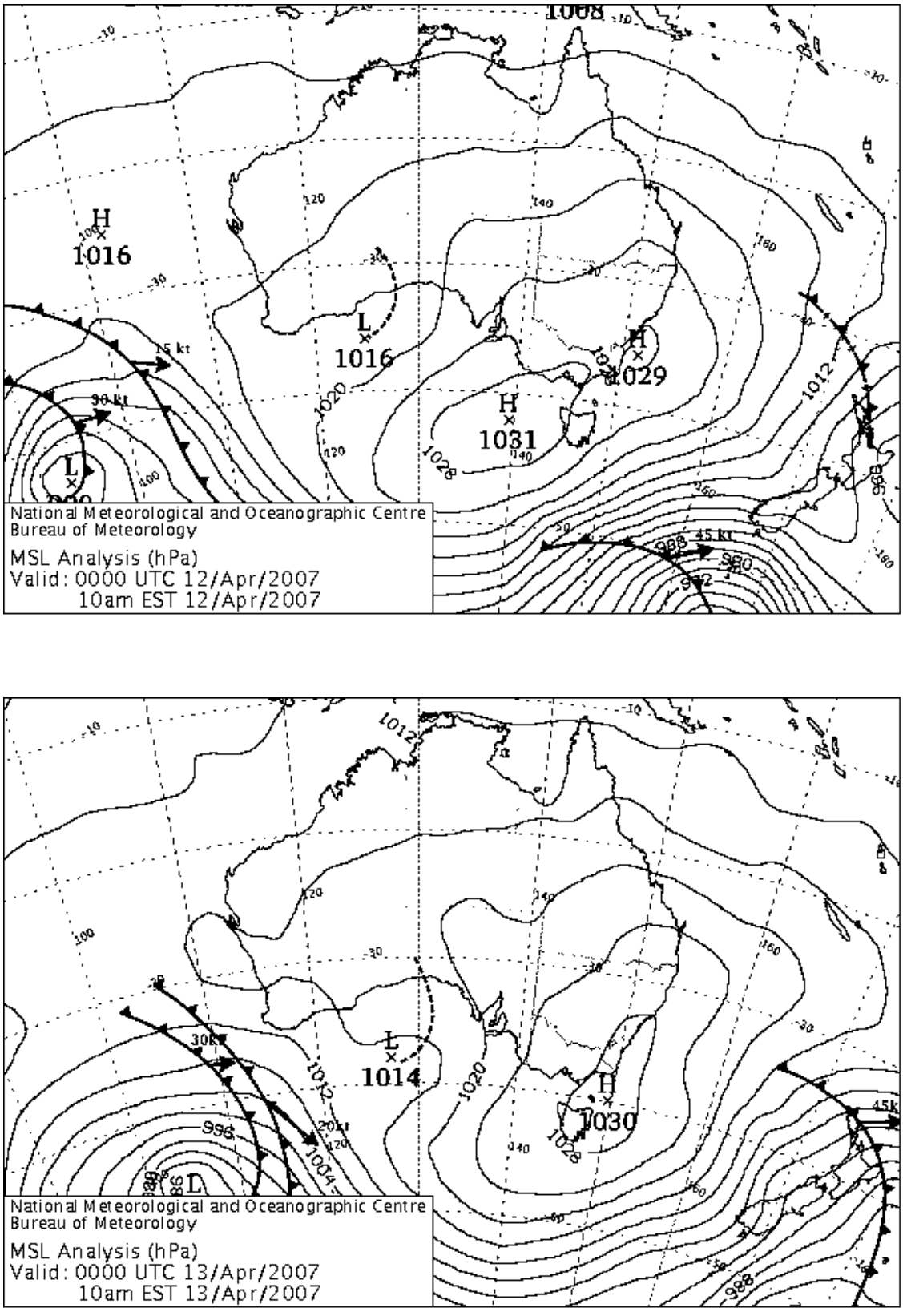

Figure 3.9 Mean Sea-Level Pressure charts at 1000 Eastern Standard Time on 12 April 2007 (top) and 13 April 2007 (bottom).

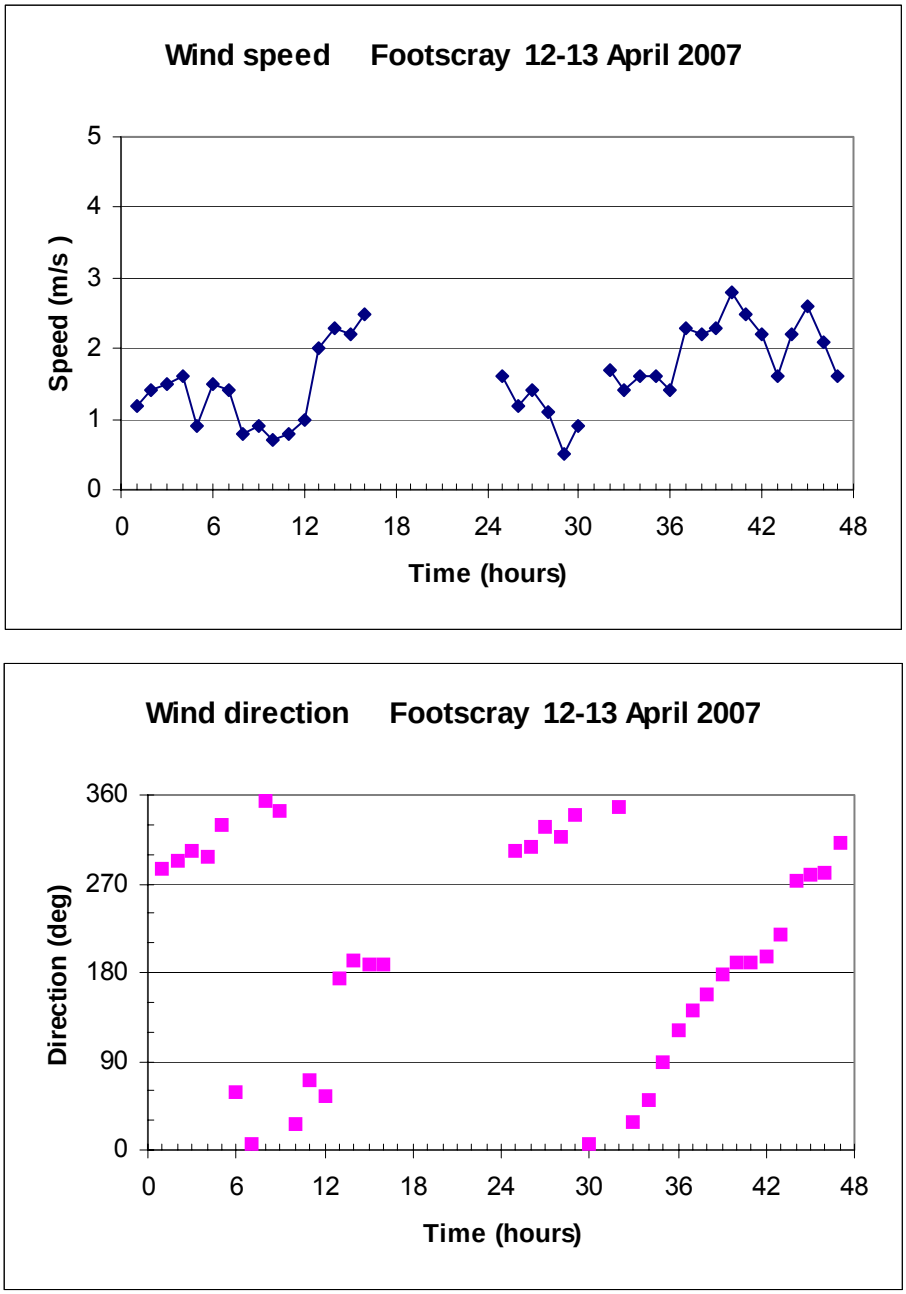

Figure 3.10 Wind speed (top) and direction (bottom) at EPA Victoria's Footscray monitoring station for the period 12-13 April 2007.

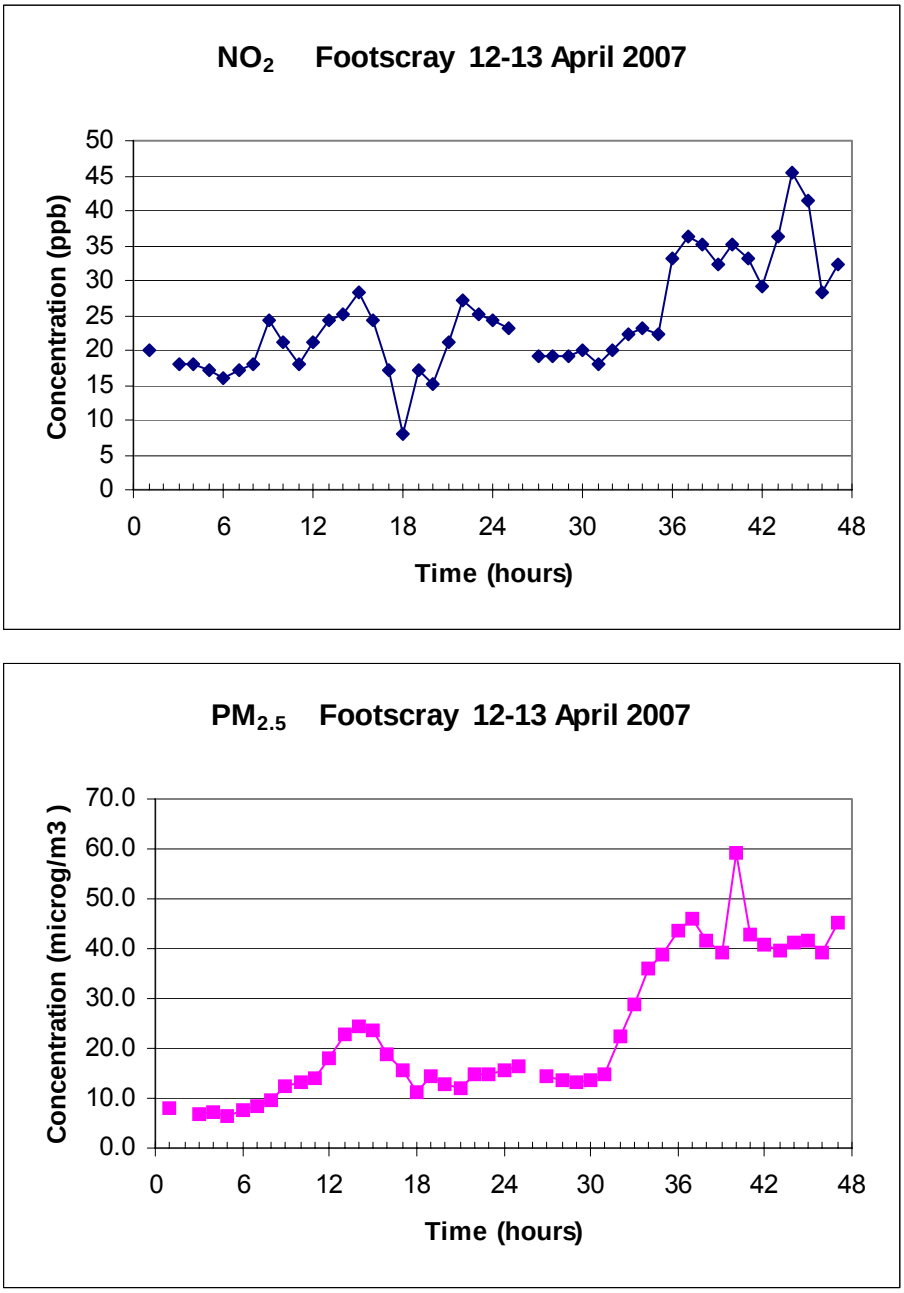

Figure 3.11 Concentrations of NO_2 (ppb) (top) and $PM_{2.5}$ ($\mu g\ m^{-3}$) (bottom) at EPA Victoria's Footscray monitoring station for the period 12-13 April 2007.

3.5 Event 4 10-11 May 2007

3.5.1 Synoptic situation

The mean sea-level pressure (MSLP) charts for each day in Figure 3.12 show Melbourne under the influence of a high pressure system. Winds were light on both days, with the Victorian Regional Office of the Bureau of Meteorology summarising the weather as follows.

Thursday 10 May 2007. Hazy, cloudy day. One or two scattered showers seen around lunch time. Moderate southerly breeze. Max temp: 17.4ºC Min temp 11.9ºC.

Friday 11 May 2007. Another hazy day with cloud dissolving during the morning. Light to moderate southerly wind. Max temp: 19.5ºC Min temp 11.9ºC.

Wind speed and direction at a height of 10 m at Footscray monitoring station are shown in Figure 3.13. At this site, and across the network, winds were less than two m s^{-1} for virtually the entire period. During the night, wind direction was northwesterly on the western side of Port Phillip Bay and north to northeast to the east of the Bay, but changed to south to southwesterly everywhere during the daytime.

NO_2 and $PM_{2.5}$ concentrations at the inner-suburban monitoring station Footscray are shown in Figure 3.14. The morning and evening peak traffic periods are evident in the NO_2 levels, rising to above 30 ppb in the evening on both days. This pattern and magnitude were typical of all stations, with the late morning drop in concentrations coinciding with the onset of the southerly breeze. Maximum $PM_{2.5}$ concentrations were below 20 μg m^{-3} at all stations throughout the observing period, except for the final evening when Alphington rose to above 30 μg m^{-3} and Footscray to near 25 μg m^{-3}.

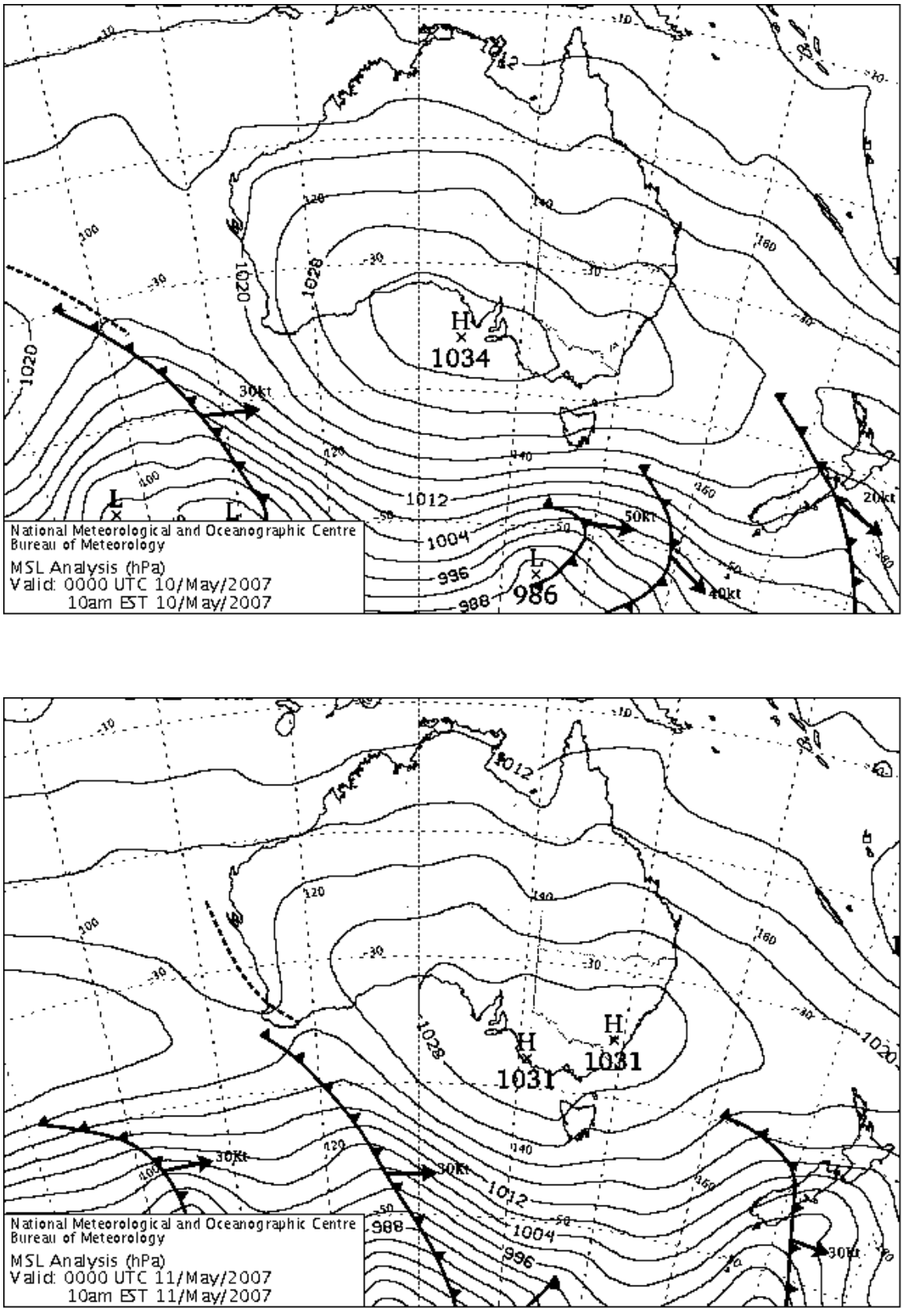

Figure 3.12 Mean Sea-Level Pressure charts at 1000 Eastern Standard Time on 10 May 2007 (top) and 11 May 2007 (bottom).

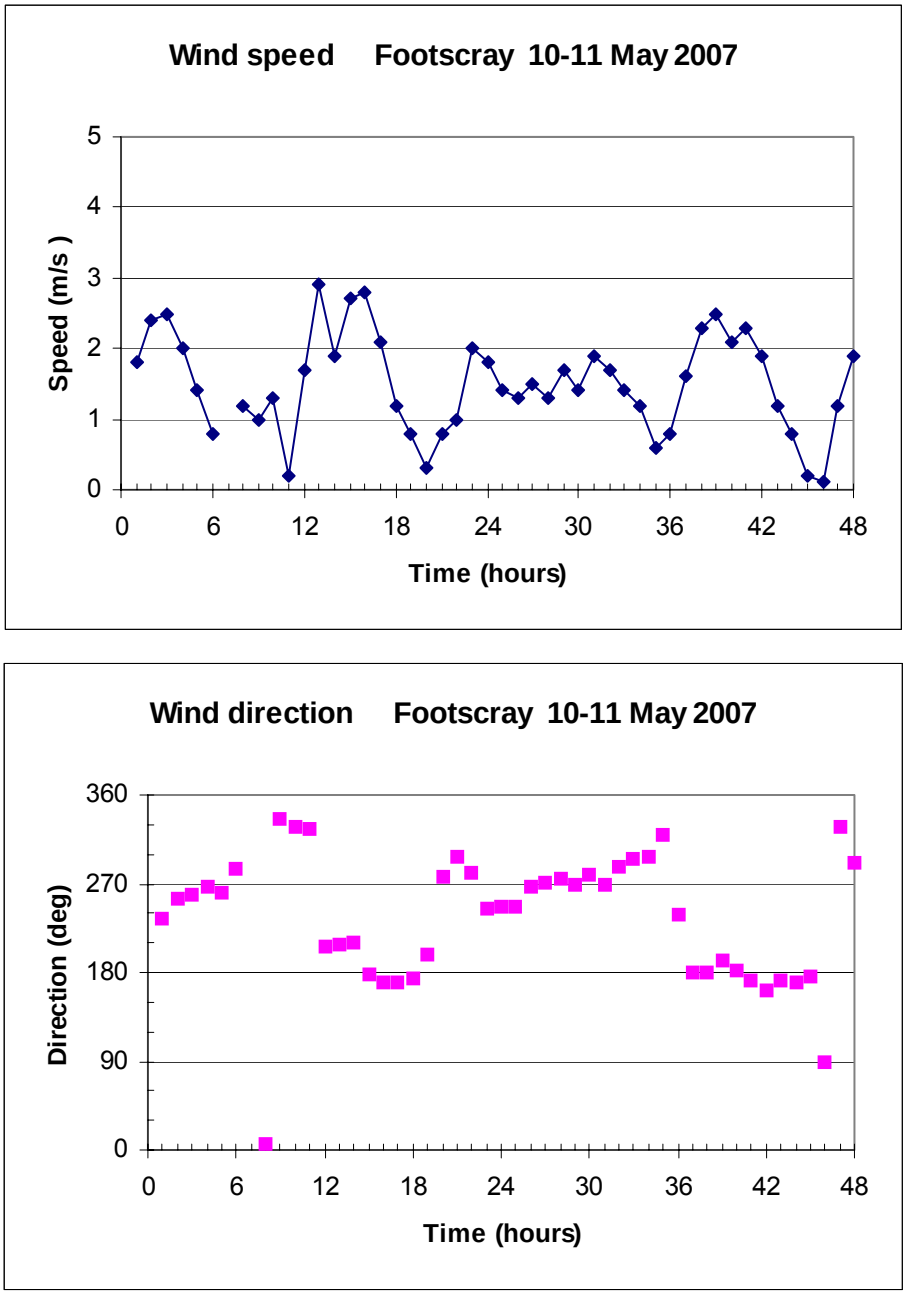

Figure 3.13 Wind speed (top) and direction (bottom) at EPA Victoria's Footscray monitoring station for the period 10-11 May 2007.

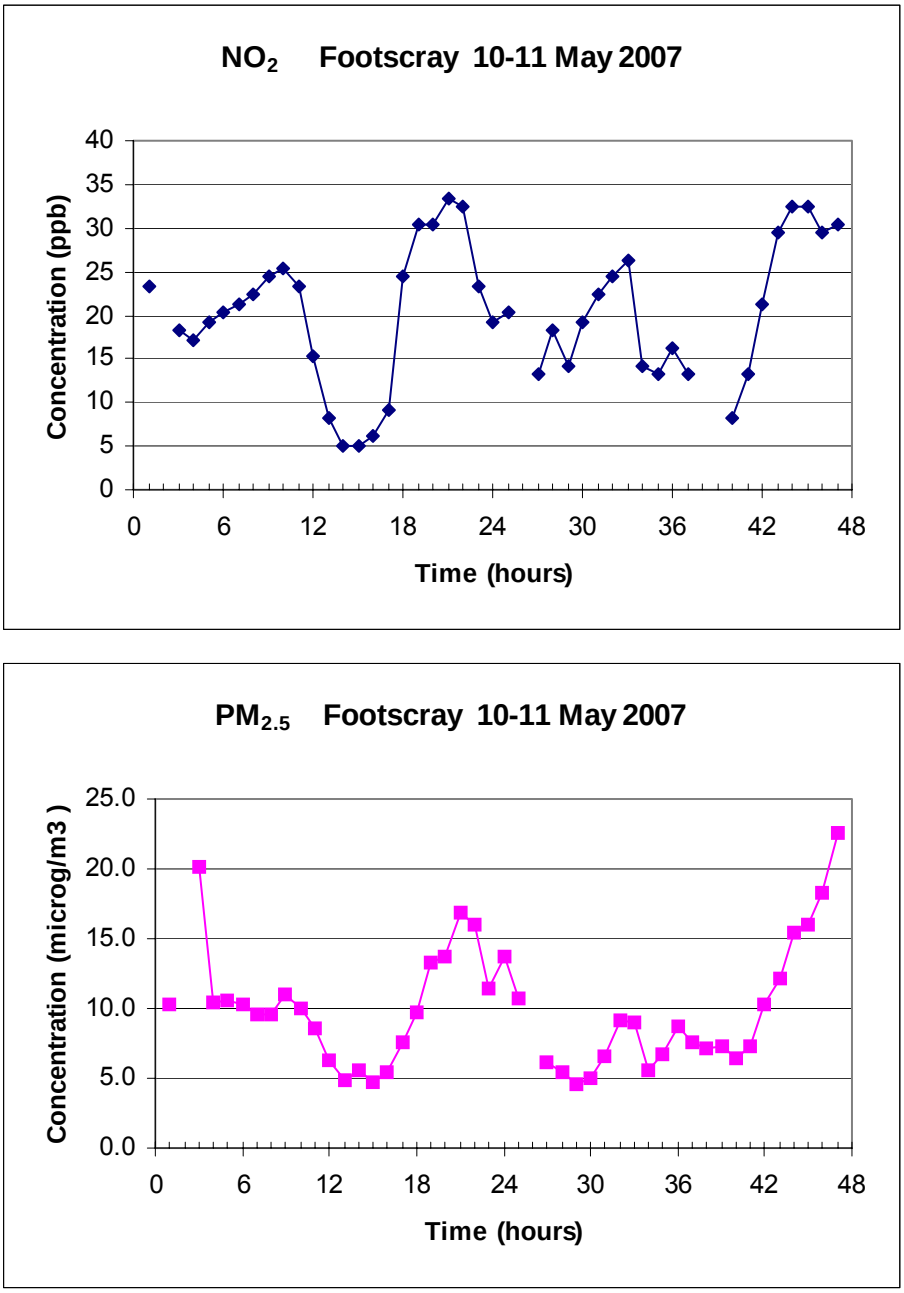

Figure 3.14 Concentrations of NO_2 (ppb) (top) and $PM_{2.5}$ (μg m^{-3}) (bottom) at EPA Victoria's Footscray monitoring station for the period 10-11 May 2007.

4. NO_2 FIELD RESULTS

4.1 Activity profiles

Our cohort profile consisted of 24 different people contributing to 62 valid measurements of personal exposure over the four events. Of the 62 samples, 46 were from office workers, 12 were obtained from stay at home people, three from an outdoors worker, and one from an on-road truck driver. Table 4.1 shows the percentage of time spent in each microenvironment by the average participant for each event. The values for the home (64%) and transit (6%) categories are similar to those from the ABS survey for Australians 15 years and older in Table 2.1 (57% and 5% respectively), but the time spent at work in our study (26%) is almost double that of the corresponding value in the ABS survey (14%). This is a reflection of the makeup of our study cohort, which was skewed towards people of working age. It should also be kept in mind that our two-day events all took place during the week, whereas weekends are more likely to be when people engage in activities which would fit into our 'other' category. The ABS survey microenvironments of shopping, recreation and outdoor (totalling 24%) correspond to other in our study.

Table 4.1 Percentage of time, averaged across all participants, spent in the four microenvironments of our study.

	Event 1	Event 2	Event 3	Event 4	All events
Home	60	63	64	65	64
Work	26	26	27	26	26
Transit	7	8	6	6	6
Other	7	3	3	3	4

4.2 NO_2 personal exposure and dosage

It is probably useful to re-iterate that we define exposure as the mean concentration of a pollutant over the period under discussion. Thus personal exposure is the mean concentration measured by each participant's sampler over a 2-day event, whereas indoor exposure is the mean concentration measured during those periods when the participant is indoors. Outdoor (or ambient) exposure is the mean concentration measured by samplers outdoors. As a check on the performance of the samplers and on the degree to which participants followed the procedures, concentrations from microenvironment samplers were weighted and summed for each participant and compared to the concentration measured by the sampler worn at all times. On average, both sets of samplers were worn for 98.3% of the time (standard deviation of 1.7%),

with a mean difference between the concentration sets of 0.1 ppb (standard deviation of 0.8 ppb). The excellent agreement can be seen in Figure 4.1, in which the concentration pairs are plotted for all measurements.

The wide range of personal exposures measured across the participants can be seen from Figure 4.2, where the maximum and minimum values of each event are plotted. There is not much variation in the extreme values across the four events, consistent with the meteorological conditions being similar for each event (Section 3). The highest personal exposure (19.8 ppb) was experienced by a participant working in the CBD, cycling to work and living in an inner suburb, and the lowest (6.1 ppb) by a participant working at Aspendale, driving to work and living in an outer Melbourne suburb.

Also shown in Figure 4.2 is the range of 2-day mean concentration measured by the 10 monitors in the EPA Victoria monitoring network across Melbourne. It can be seen that exposure to transit and indoor NO_2 sources and sinks did not cause any participant's personal exposure to exceed the maximum ambient concentrations measured across Melbourne for each event. However the lowest participant exposure in each event was consistently lower than the minimum ambient concentration measured by the network.

Figure 4.3 shows the maximum and minimum values of exposure measured in the microenvironments of work, transit, home and other. Not surprisingly, the highest exposures were measured while participants were travelling, most of them on roads. The highest transit value recorded (46.2 ppb) was by a cyclist, and the second-highest (39.0) by a truck driver. On average, values were higher at work than at home, presumably due to higher average ambient concentrations at work (21.3 ppb at work compared to 16.6 ppb at home) and indoor/outdoor ratios at work (see Table 4.3). Some very low values were measured in homes and vehicles, and these are discussed in Section 4.2 on indoor/outdoor ratios. The maximum, minimum, mean and standard deviation statistics are shown in Table 4.2 for each microenvironment.

Table 4.2 Summary of findings re 2-day cumulative exposure from 62 NO_2 passive sampler measurements. 'All' denotes the personal exposure measured by the sampler worn in all environments.

EXPOSURE (ppb)	Maximum	Minimum	Mean	Standard Deviation
Home	21.1	1.9	9.2	4.3
Work	26.8	7.2	15.5	4.9
Transit	46.2	2.2	24.0	9.1
Other	33.2	4.9	18.8	7.7
All	19.8	6.1	12.1	3.1

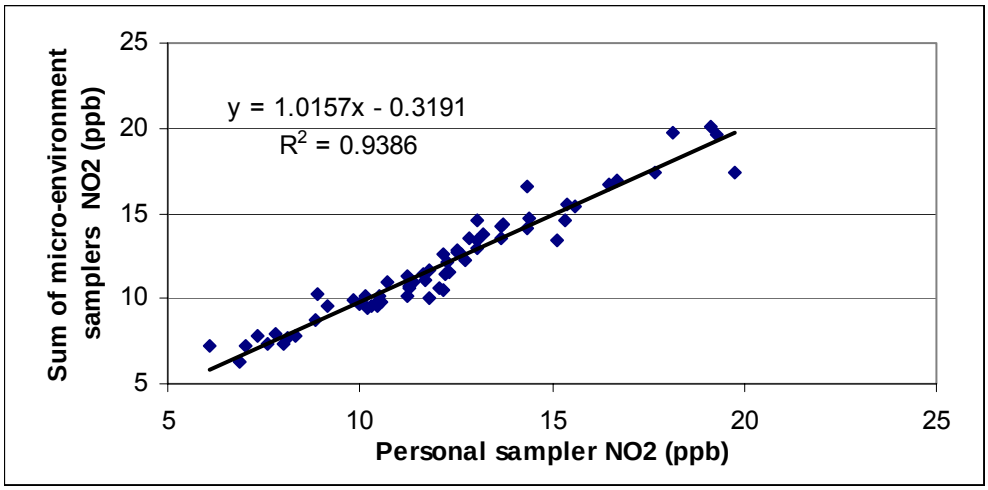

Figure 4.1 For each participant, the weighted sum of NO_2 concentration (ppb) from the samplers worn in each microenvironment versus the concentration from the personal sampler worn at all times.

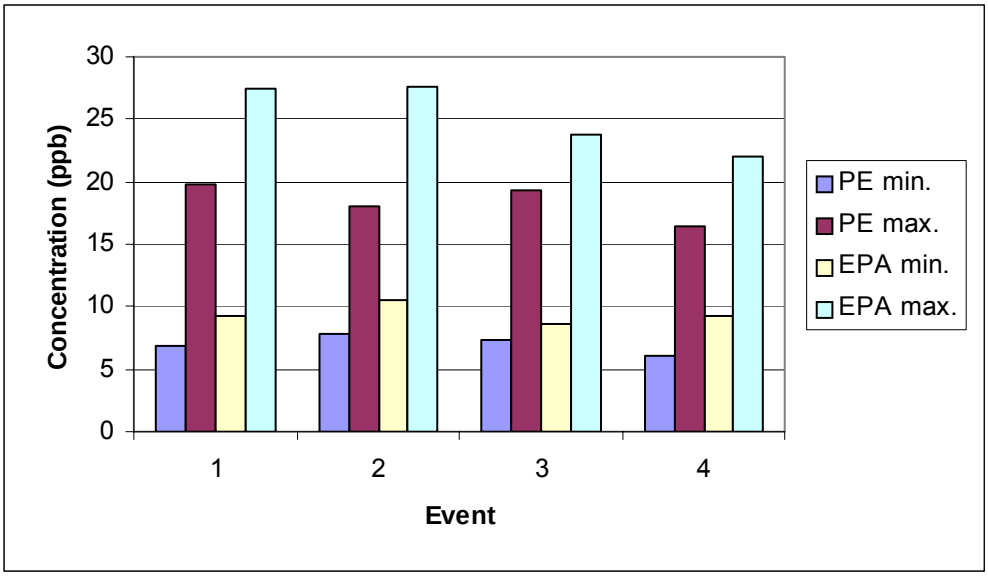

Figure 4.2 Highest and lowest mean NO_2 concentration measured over 48 hours by participants wearing personal passive samplers for each of the four events (PE). Also shown is the range of mean ambient NO_2 concentration recorded across the EPA Victoria monitoring network for each event (EPA).

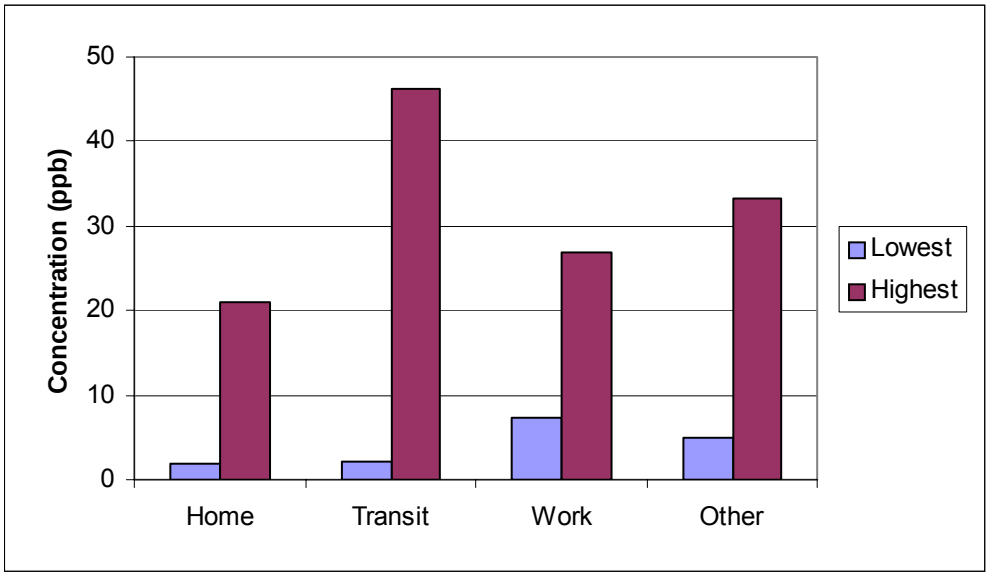

Figure 4.3 Highest and lowest mean NO_2 concentration (across all four events) measured by participants wearing personal passive samplers in the microenvironments of home, transit, work and other.

Further information can be obtained by examining the dosage of participants in each microenvironment. In this study, dosage is defined by the mean concentration experienced in a microenvironment multiplied by the time in hours spent in that environment. The dosage for each environment, expressed as a percentage of the total dosage, is shown in Figure 4.4 where it is seen that people obtained their dominant dosages from the home and work environments, largely due to the amount of time spent in each. In fact 50% of dosage for the average participant was experienced at home. However, there are some cases that do not fit this picture. For example, five participants received considerably higher dosages at work than at home, because of the low concentrations of NO_2 measured in their home environments. This is discussed further in Section 5 in relation to dwelling characteristics. Another received equally high doses while at work and in transit, due both to his transit mode (bicycle) and transit time (5.1 hours over the 48-hour period). Although transit times were considerably less than time spent at home or at work, the higher concentrations experienced in transit can lead at times to dosages that approach those measured in home and work environments. Naturally, dosage for the home environment was greatest for those who are based at home and do not travel to work. For a few participants, the 'other' category was a significant contributor when the evenings were not spent at home.

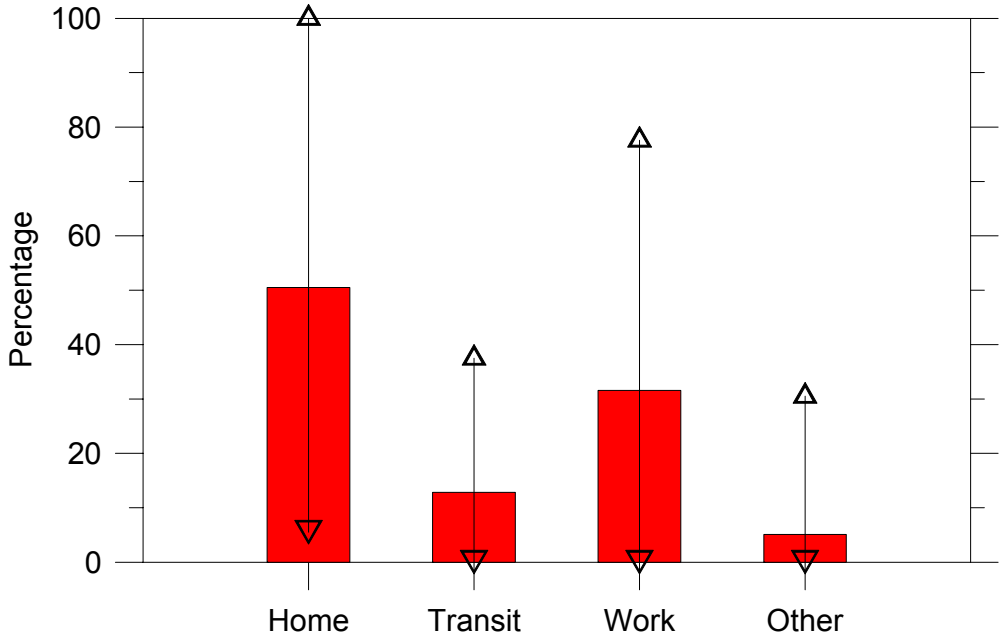

Figure 4.4 The percentage of total dosage (ppb-hrs) experienced in each microenvironment. Arrows indicate range across participants.

4.3 NO_2 indoor–outdoor concentration ratios

Ratios of inside to outside concentration while participants are in each of the three microenvironments are listed in Table 4.3. As expected, there is a wide variation in ratios for the home environment, from more than 1.0 to the quite low value of 0.12. The reasons range from windows and/or doors left open, to indoor sources of NO_2, to poorly- or highly-ventilated homes. Results are analysed according to home characteristics in Section 5. The mean value is 0.57 with a standard deviation of 0.27. At all workplaces, the ratio was less than 1.0 for the first three events, but in Event four five measured ratios (at three different workplaces) were equal to or greater than 1.0. It is difficult to explain this as windows or doors were not left open and the samplers were not analysed in the same batch. The outdoor sampler values at the Aspendale workplace agreed closely with the mean concentration over that period measured by an ambient monitor at Aspendale's on-site Bayside Air Quality Station (BAQS) laboratory. Over all events, the mean value was 0.74 with a standard deviation of 0.16. The variation in the ratio is less than for homes, and is likely due to less variation in the ventilation properties of offices, lack of indoor sources and to the fact that the 50 work measurements were spread over only eight workplaces, compared to the 60 home measurements covering 24 homes.

The mean in-vehicle to out-vehicle concentration ratio from the 16 vehicles (0.63) with the external vent open was not vastly different from the mean home (0.57) and work (0.74) values. For all trips, windows were closed and air-conditioning was off. Three vehicles had climate control turned on and 10 had the heater on, but the ratios for these vehicles were evenly distributed from highest to lowest. The only factor noted in the diaries that seemed to influence the ratio was whether the external vent was open or closed. The two lowest readings (0.07 and 0.37) were during the only trips when the vent was closed, and are consistent with the values (0.30 ± .07) obtained under the same conditions by Cains et al. (2003). When these two vehicles travelled with the external vent open, the readings were 0.76 and 0.61 respectively.

The variation across vehicles in I/O ratios obtained in trips with the vent open may be a function of materials inside the car, volume to surface area, and the ventilation rate, influenced strongly by the fan speed (not recorded).

Table 4.3 Summary of indoor/outdoor ratios calculated from NO_2 passive sampler measurements for 2-day periods. Vehicle transit ratios were measured in vehicles with the external vent open.

IN/OUT RATIOS	Maximum	Minimum	Mean	Standard Deviation	Number of values
Home	1.37	0.12	0.57	0.27	60
Work	1.13	0.47	0.74	0.16	48
Transit (vehicle)	0.96	0.39	0.63	0.17	16

5. A MODEL FOR ESTIMATING INDOOR NO_2 CONCENTRATION

In this chapter, a technique for predicting the indoor concentration of NO_2 is developed to enable the estimation of indoor-outdoor concentration ratios for use in our personal exposure model.

5.1 House characteristics

As discussed in the conceptual model, the concentration of indoor NO_2 in residences is determined by the ventilation rate, the contribution to indoor concentrations from sources indoors, and outdoor sources.

Of the 62 NO_2 measurements, 60 measurements from 22 houses provided sufficient information to enable prediction of indoor NO_2. Information included completed time activity diaries (see Appendix B) and house characteristics including type of cooking fuel, type of heating, age of house, materials, floor area and volume (Table 5.1). Ventilation rates were measured for five selected houses.

Table 5.1 Characteristics of houses used in the study.

House ID	Type	Bed rms	Outdoor material	Age yrs	Area m^2	Vol. m^3	Heater type	Cooking type		No. of events
								Cooktop	Oven	
1	House	3	Brick	3	180	405	gas ducted	gas	electric	4
2	Flat grd floor	2	Concrete	5	67	205	electric	electric	electric	3
3	House	3	Hardie board	5	145	406	woodheater	gas	electric	1
4	House	3	Brick	8	142	355	gas ducted	gas	gas	3
5	House	2	Brick/h-board	12	256	743	gas ducted	gas	electric	4
6	House	7	fibro	15	214	514	gas ducted	gas	electric	2
7	House	3	Brick	18	132	330	gas ducted	gas	gas	1
8	House	4	Brick	25	130	319	gas ducted	electric	electric	3
9	Flat 3rd floor	1	Brick	30	48	115	electric	gas	electric	2
10	Flat 2nd floor	2	Brick	30	72	173	electric	gas	gas	2
11	House	3	Brick	30	94	230	electric	gas	gas	1
12	House	4	Brick	35	180	540	gas ducted	gas	gas	3
13	House	3	Brick veneer	40	88	210	gas ducted	gas	electric	3
14	House	3	Brick veneer	40	97	232	gas ducted	electric	electric	4
15	House	3	Weatherboard	40	100	240	gas ducted	gas	electric	3
16	House	3	Weatherboard	53	80	216	gas space	gas	gas	3
17	House	1	Weatherboard	54	124	337	hydronic	electric	electric	2
18	House	3	Weatherboard	70	110	297	gas wall	gas	gas	3
19	House	3	Weatherboard	75	113	318	gas ducted	gas	electric	4
20	House	5	Brick	79	211	632	gas ducted	gas	electric	3
21	House	3	Brick	80	118	360	gas ducted	electric	electric	3
22	House	2	Weatherboard	100	79	248	gas wall	gas	gas	3

As no participant residences had indoor sources from unflued gas heaters or smokers, the only sources of indoor NO_2 were assumed to be emissions from gas cooking and infiltration of outdoor NO_2 indoors. Thus the prediction of indoor NO_2 was based on developing a model that accounted for gas cooking emissions, ventilation rate and outdoor concentration. The presence of gas water heaters outdoors was not included in the model.

5.2 Indoor NO_2 using a mass balance equation

Prediction of indoor NO_2 was performed using a conservation of mass equation that assumes a steady-state over the sample duration, as described in Dockery and Spengler (1980). The steady-state assumption is valid if changes in indoor concentration over the sample period are small compared to product of sampling duration (48 hours), loss rate and air exchange rate.

$$\overline{C_i} = \frac{Pa}{(a+K)}\overline{C_o} + \frac{1}{V(a+K)}\overline{S}$$

Where:

$\overline{C_i}$ = mean indoor concentration (μg m^{-3})

$\overline{C_o}$ = mean outdoor concentration (μg m^{-3})

P = penetration of outdoor pollutant through building shell, assumed to be 1.0 for NO_2

a = air changes per hour (h^{-1})

K = rate of decay and removal of indoor NO_2 per hour (h^{-1})

$\overline{S}$ = rate of emission from indoor sources in micrograms per hour (μg h^{-1})

V = interior volume of the building

The rate of loss of NO_2 indoors depends on many variables such as surface area to volume ratio, type of surface materials and relative humidity (Spicer et al., 1989; Yamanaka, 1984; Grontoft and Raychaudhuri, 2004). Spicer et al. (1989) reported that a typical rate loss in a residence resulting from the interaction of these variables was 0.80 h^{-1}; we have also used this value in the mass balance equation.

5.2.1 Gas cooking source

A regression of NO_2 indoor/outdoor ratio versus time spent using gas cookers in the absence of ventilation (Figure 5.1) explained 33% of the variance in indoor NO_2 when gas cooking was used. A simple approach to simulating indoor concentrations from gas cooking emissions is to represent emissions in the steady state model as constant throughout the sample period, rather than modelling peak emission/decay events (eg Dimitroulopoulou et al., 2006). The constant emission source input was calculated from the proportion of time a participant used gas cooking whilst at home, multiplied by a typical gas cook top NO_2 emission rate (Relwani et al., 1986).

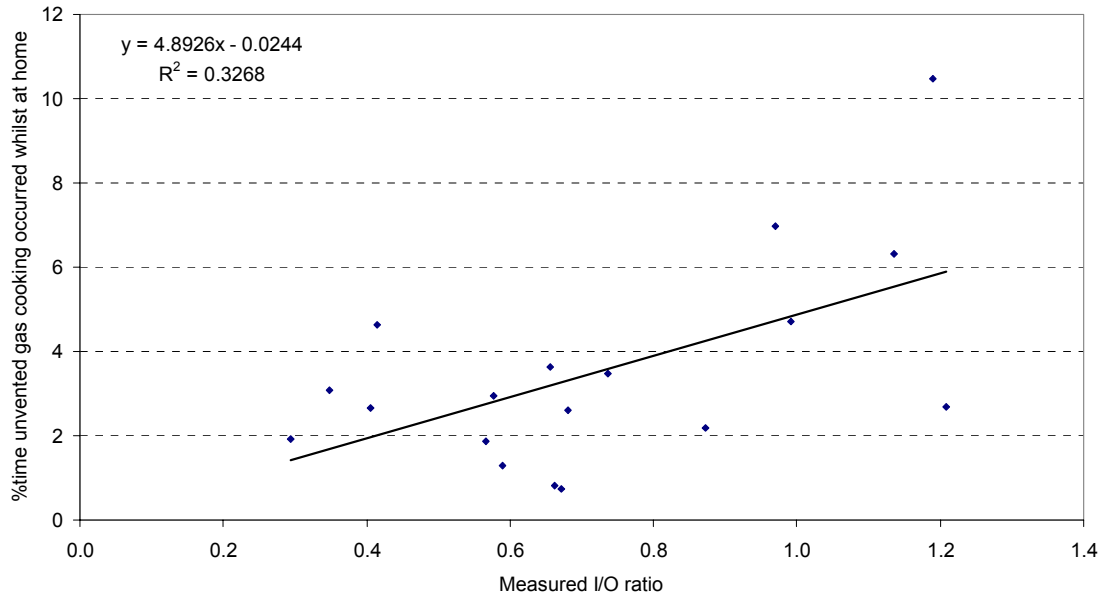

Figure 5.1 A regression of NO_2 indoor/outdoor ratio versus time spent using gas cookers in the absence of ventilation.

5.2.2 Ventilation rates

Based on the participant diaries, most households (20 of 22) did not have their windows open during the measurement periods, thus it was decided that measurement of the houses in a closed-up state was representative of the sampling conditions. To assign a ventilation rate for each house, infiltration rates of five of the 22 houses were measured using the CO_2 release method similar to that described in Dunne et al. (2006). Houses were filled with 5000 ppm of CO_2, and between one and three CO_2 *QTrak* monitors (TSI Inc.) were left in various areas of the house and one CO_2 *QTrak* monitor outside the house. The unoccupied house was closed for two to three hours and the monitors logged the decay at 1-minute intervals. The averaged decay curve of all indoor measurements was used to determine the infiltration rate.

Table 5.2 shows that the main influence on the air exchange rate appears to be house age. There is a relationship between house age and house ventilation due to changes to building materials, construction techniques, building codes and house design over time. Newer houses are designed to be more energy efficient and have lower infiltration rates (minimum ventilation rates). Examples include changes to the 1990 Building Code that removed the requirement for fixed ventilation. Ventilation in newer houses can be manually controlled by the state of openable external windows, doors, fan units, etc. In older houses, shifting of foundations and the building shell can also result in a 'leakier' building, although this does not necessarily apply to all old buildings. Ventilation rates from the Yang et al. (2004) and Brown (1997) papers, shown in Table 2.4, illustrate the difference in ventilation and infiltration rates from newer and older houses. For further discussion, see Brown (1997).

It appears from Table 5.2 that carpet coverage may be a factor contributing to the air exchange rate. Although house furnishings and materials indoors can affect the decay rate of pollutants,

an inert gas (CO_2) was used for the infiltration rate measurements in our experiments. Thus it appears to be just a coincidence and it could be that newer houses may have less carpet than older ones.

Table 5.2 Infiltration rates (air change) measured for the five selected houses.

House ID	Air change h^{-1}	Age years	Floor area m^2	Volume m^3	Outdoor material	Floor materials %				Wall & Ceiling material
						Carpet	wood	tile	lino	
3	0.19	5	145	406	Hardie board	35	50	15	0	Plaster
2	0.30	5	67	205	concrete	40	52	8	0	Plaster
14	0.39	40	97	232	brick veneer	55	37	0	8	Plaster
19	0.36	75	113	318	weatherboard	69	0	11	20	Plaster
13	0.39	40	88	210	brick veneer	76	17	7	0	Plaster

Both the 5-year old renovated ground floor apartment (ID 1) and a 5-year old house (ID 14) had lower air exchange rates than older houses. As discussed in the conceptual model, this is to be expected due to energy efficiency and building material improvements made to residences. A significant change in infiltration rates occurred from implementation of the 1990 building code, which removed requirements for fixed ventilation and enabled ventilation only by controllable openings (ie windows). Thus in the mass balance equation, values for air exchange rates were assigned to the participant houses based on whether they were built before or after 1990.

Values initially used were 0.5 h^{-1} for residences built before 1990 and 0.2 h^{-1} for residences built after 1990. However better results were obtained in the model by increasing each of the values by 0.1 h^{-1}, to allow for slight increases in air exchange rates driven by thermal gradients during heater operation, used by 80% of participants during the study. For a house with windows open throughout the measurement period, the air exchange rate used was doubled; a method used by Dimitroulopoulou et al. (2006).

5.2.3 Outdoor NO_2 concentration

Three data sets were evaluated for obtaining the outdoor concentrations of NO_2 needed in the mass balance equation.

1. The *local* outdoor sampler concentration measured at the participant's house.
2. Ambient concentration at the nearest *monitor* in the EPA Victoria monitoring network.
3. Ambient concentration predicted at the house location by a *blending* of EPA Victoria monitoring data with gridded model predictions from TAPM-CTM – see section 6.3

Parameter values used for all variables in the mass balance equation are listed in Table 5.3.

Table 5.3 Parameter values used in the mass balance equation for calculating indoor NO_2 concentration.

Parameter	Value
Penetration factor, *P*	1
Air exchange rate, *a*	
Built after 1990, no windows open	0.3 h^{-1}
Built after 1990, windows open	0.6 h^{-1}
Built before 1990, no windows open	0.6 h^{-1}
Built before 1990, windows open	1.2 h^{-1}
NO_2 decay rate, *K*	0.8 h^{-1}
NO_2 outdoor average concentration, $\overline{C_o}$	
Measured outdoor NO_2	10.7 - 45.7 µg m^{-3}
Nearest monitor NO_2	19.4 - 51.7 µg m^{-3}
Blended obs/model NO_2	11.6 - 46.8 µg m^{-3}
NO_2 emission rate, $\overline{S}$	
Maximum fuel input, blue flame burning, 9.49 MJ hr^{-1} cook-top	7.74 µg kJ^{-1}
Range of emission rates used according to %time used	0 - 7699 µg h^{-1}
Volume of residence, *V*	115 - 746 m^{-3}

5.3 Results

Predicted indoor concentrations using the three outdoor concentration data sets are plotted against measured indoor concentration in Figure 5.2. It is probably not surprising that the best predictions, in terms of R^2 the percentage of observed variation predicted by the model, are made when the local outdoor measurements are used, but it is encouraging that they are almost matched by those from the blended monitor and model concentrations. Results from using the nearest monitor ambient concentrations are also quite respectable.

The ability of our steady-state mass balance model to be able to predict up to 61% of the variance in indoor concentration compares favourably with the predictive abilities of more complex multiple regression models. Lee et al. (1998) were able to predict 42% of indoor variation in the Boston residential nitrogen dioxide characterisation study by including the presence of gas appliances. Monn et al. (1998) were able to describe 58% of variance in indoor NO_2 using outdoor concentration, gas cooking, smoking and ventilation. Sheppeard et al. (2006) were able to predict 72% of indoor variation in NSW houses using hours of use of unflued gas heating, gas cooking and outdoor level of nitrogen dioxide. The addition of variables for quantity of cigarettes smoked during the week, heater type, region, type of oven and house age increased the prediction of indoor variation to 87%.

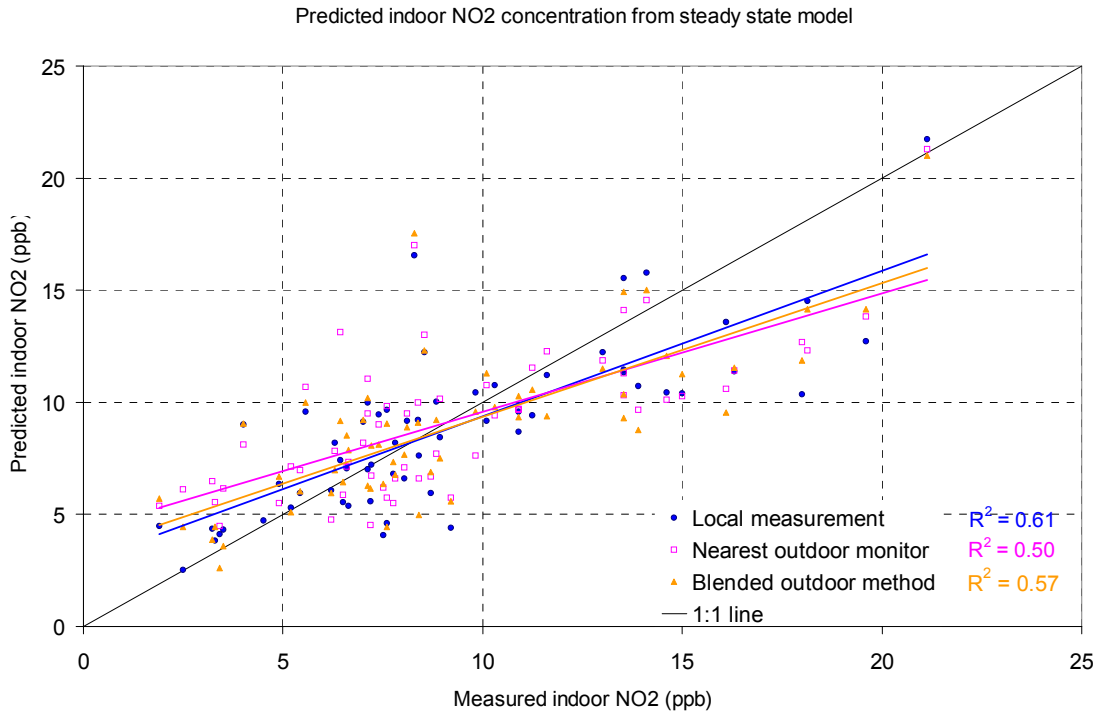

Figure 5.2 Prediction of indoor NO_2 using a steady-state mass balance model. Outdoor concentration is obtained from three different data sets.

A simpler approach to estimating indoor NO_2, using the average indoor-outdoor ratio measured for houses that did or did not use gas cooking, was also evaluated. Between 23% and 39% of variance (across the three data sets) in indoor NO_2 could be explained using this approach (Figure 5.3). However the higher indoor NO_2 levels from people who frequently used gas cooking were underestimated.

All approaches developed for predicting indoor NO_2 levels in this chapter are evaluated in section 7.2.1 for their suitability as part of a predictive personal exposure model.

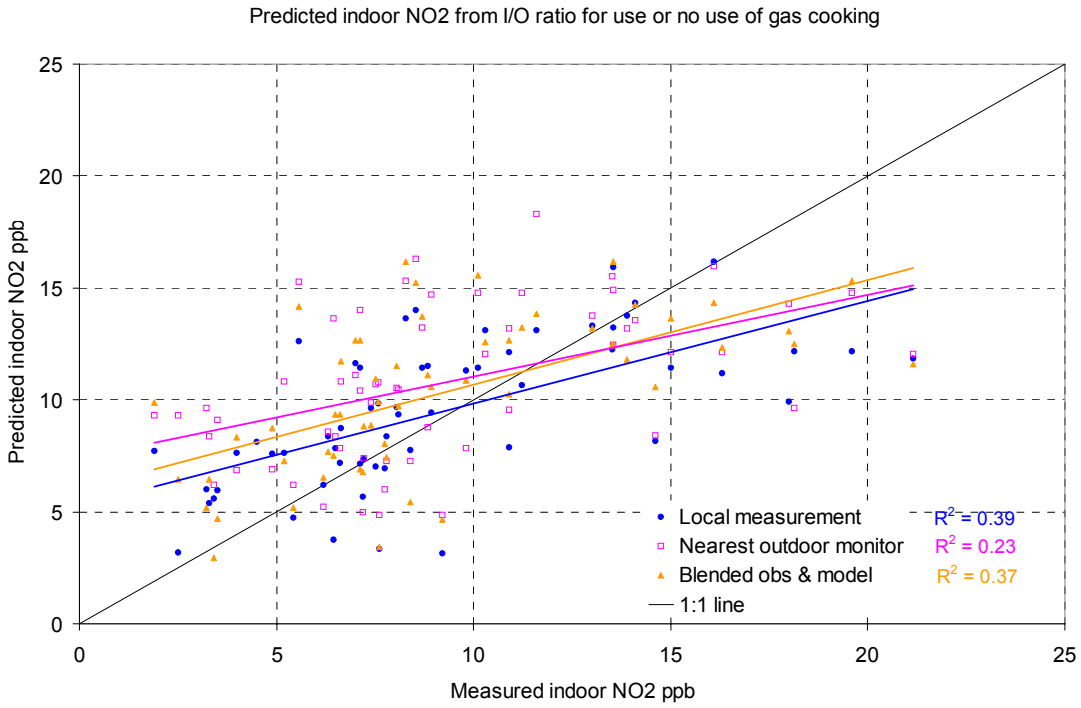

Figure 5.3 Prediction of indoor NO_2 using an average indoor/outdoor ratio dependent on whether or not a gas cooking appliance is installed.

6. AIR QUALITY MODELS

The ambient exposure of each participant was calculated by two air quality models, AAQFS and TAPM-CTM. Personal exposure at home and at work was calculated by scaling the ambient exposure by an indoor/outdoor ratio.

6.1 Australian Air Quality Forecasting System (AAQFS)

AAQFS is an operational forecast model run twice daily by the Bureau of Meteorology (BoM) for Sydney, Melbourne and Adelaide (Cope et al., 2004). Results are sent to the state EPAs in each city, where they contribute to the next day's air quality forecast. The weather forecast model LAPS provides the meteorological fields that drive dispersion of emissions and pollutant concentrations in the chemical transport model CTM. The latter can be run for an arbitrary number of chemically reacting gaseous and aerosol species. Emission inventories for each city are produced by the respective EPAs.

6.2 TAPM-CTM

TAPM-CTM is a similar type of model to AAQFS, except that the meteorological driver is not LAPS, but the meteorological component of The Air Pollution Model TAPM (Hurley et al., 2005). TAPM-CTM is not used as a forecasting model, but is run instead for case studies and annual simulations of the production and dispersion of urban and regional photochemical smog and aerosols. In this hindcast mode, TAPM-CTM makes use of BoM's GASP archived meteorological analysis fields for initial and boundary conditions. The air chemistry model CTM is used in both AAQFS and TAPM-CTM.

Modelling of NO_2 and $PM_{2.5}$ concentrations for the four events was done with version 1.7b of TAPM-CTM (Cope et al. 2004). A comprehensive anthropogenic emissions inventory, based on the EPA Victoria's Port Phillip air emissions inventory (EPAV, 1998), was used as well as a natural (includes emissions from plants and soil) emissions inventory, which is generated 'on-the-fly' in TAPM-CTM during a simulation.

Version 3.2 of TAPM was used for the meteorological modelling. This version includes a multi-level soil temperature/moisture scheme and improved coupling with the overlaying vegetation. For the current study TAPM was configured with three 70 x 70 x 25 nested computational grids with horizontal spacing of nine km, three km and one km. In the vertical, model levels are at 10, 25, 50, 100, 150, 200 m above the ground, with layer spacing gradually increasing up to the 4000 m level, above which a spacing of 1000 m is employed up to the top of the model domain (which extends to 8000 m).

Boundary conditions for TAPM were taken from the Bureau of Meteorology 6–hourly GASP analysis (~75 km spatial resolution). Sea surface temperatures were taken from a 100 km NCAR data set and a 300 m resolution data set was used to generate the topographic grids.

In order to generate the most accurate wind fields for the chemical transport modelling, predictions of near-surface winds in TAPM were nudged to observations from seven EPA Victoria sites, five Bureau of Meteorology weather station sites, and the Aspendale site. This data-assimilation mode of simulation is referred to as 'T-CTM assim' in section 6.4. For comparison, results are also shown from simulations without assimilation of data (T-CTM no assim).

6.3 Blended fields

In Physick et al. (2007), a methodology was presented for computing daily ambient pollutant concentration fields that takes account of spatial variation in air quality. This approach, using elliptical influence functions, involves the optimum blending of observations from a monitoring network with gridded pollution fields predicted by a complex air quality model. Such fields can contribute to epidemiological studies by allowing more spatial and temporal information to be incorporated in the exposure fields, and by enabling cohort studies to take advantage of the information in daily activity diaries.

In this study, we have successfully applied the blending technique to TAPM-CTM hourly-averaged fields, rather than daily-averaged, for the 48 hours of each event. Data from the EPA Victoria monitoring network and from the Aspendale site were incorporated. These fields are used in section 7.2 to estimate the exposure of participants as they move according to their daily activities.

6.4 Evaluation of models and blending

6.4.1 NO_2

Predictions of ambient concentrations from AAQFS, TAPM-CTM and the blended fields cannot be compared to indoor or transit measurements, but a valid comparison can be made with the outdoor concentrations measured at the home (60 values) and workplace (48 values) of each participant. These are shown in Figure 6.1 for NO_2, in which Measured refers to the average of the mean measured concentrations, and AAQFS, T-CTM no assim, T-CTM assim, and Blended refer to predictions from the various models at the same locations and time periods.

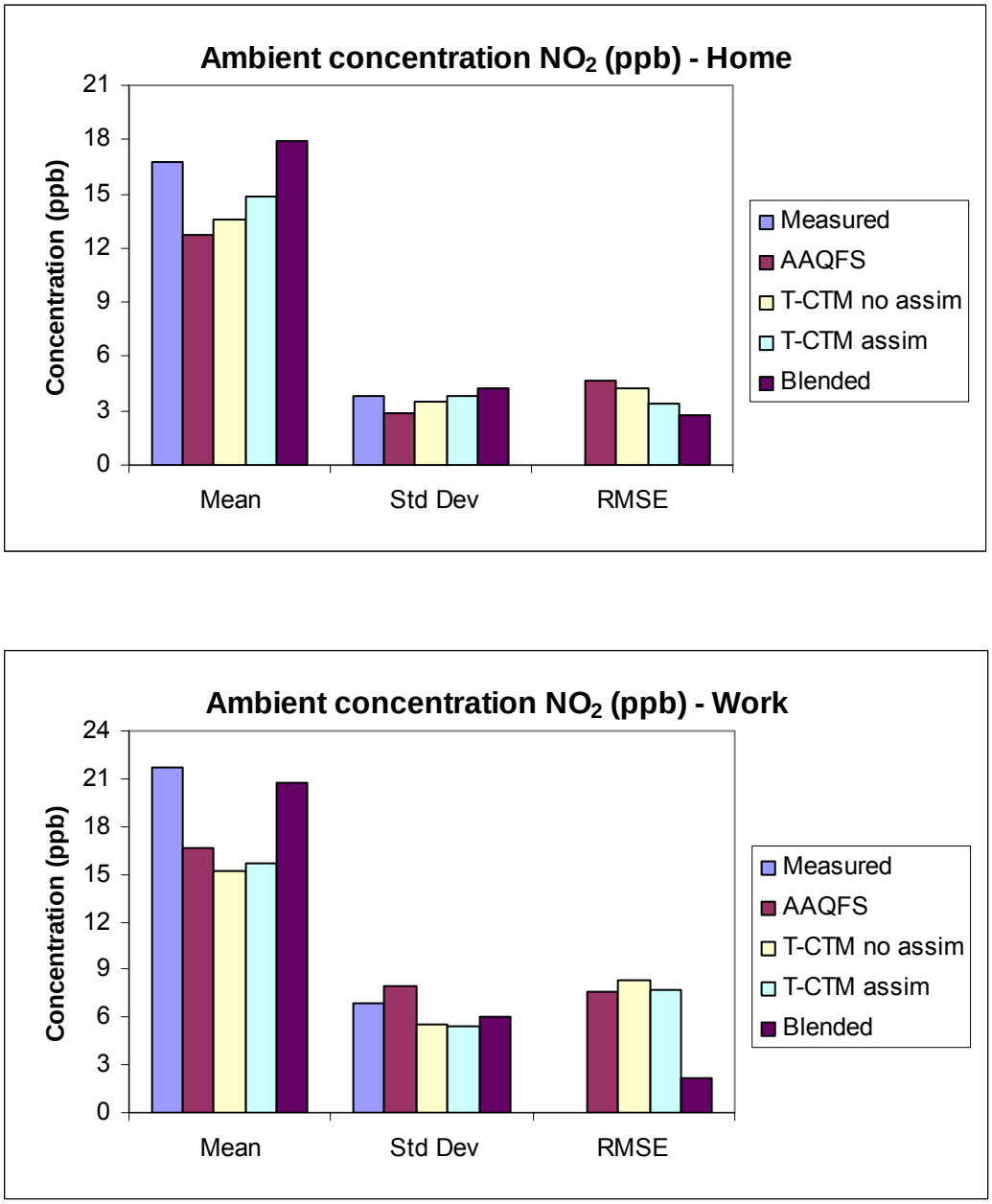

Figure 6.1 Mean, standard deviation and root mean square error (RMSE) of measured, modelled (AAQFS, T-CTM no assim, T-CTM assim) and blended ambient NO_2 concentrations. Values at home (upper) and work (lower) are shown.

Assimilation of wind data has improved the results for both the home and work environments. All three model simulations produced respectable results at the home environments (predominantly night time), with the T-CTM assimilation run underestimating the NO_2 exposure by only 11%. The ratio of RMSE to observed standard deviation is less than 1, indicating predictive skill, only for this model. The models did not perform as well at the work environments (predominantly day time conditions), with the best result being an under prediction of 23% by AAQFS, and no models indicating skill. It should be noted though that the good correlation between measured and modelled correlations, 0.72 for T_CTM assim at night

time and at day time, indicates that the models were reproducing the spatial distribution of exposure well, if not quite the correct magnitude.

There are two main reasons that the mean exposure comparison was worse at the work locations. In all four events at the majority of stations, winds turned from about midday onwards to become off the sea with a southerly component. In the first three events, NO_2 levels rose as the sea breeze reached stations, but in the last event NO_2 levels dropped on the sea breeze arrival. The models predicted concentrations well for the last event, but overall did not predict the increase for the first three events. An example is shown for event one (25-26 May 2006) in Figure 6.2, which shows a comparison of the TAPM-CTM model results (with wind assimilation) against hourly-averaged observations at the EPA Victoria monitoring station at Footscray and at CSIRO's Aspendale station. Firstly, in the afternoon of 26 May, NO_2 levels at most stations across Melbourne (except Dandenong and Mooroolbark) rose to above 30 ppb for three to four hours as a sea breeze moved onshore. The sea breeze was captured well by the model but not the NO_2 increase.

The second reason concerns the models' performance at Aspendale, where 21 of the 50 work observations over the four events were taken. At this station, the models' daytime predictions were worse than at all the other monitors, consistently underestimating NO_2 levels, especially in the morning peak traffic period. Comparison of measured and modelled NO_x concentrations at Aspendale suggests that NO_x emissions may be too low in the inventory for the Aspendale grid square. This may be due to increased traffic volumes since 1996, the year for which the inventory was compiled, or it may be that the impact of the high-volume Nepean Highway (200 m from the monitor) cannot be represented adequately in a model in which both the emissions and predicted concentrations are averages over one square kilometre.

The home and work ambient concentrations estimated from the blended monitor and model fields (denoted Blended in Figure 6.1) agreed very well with the passive sampler concentrations. Average concentration at home was overestimated by 8% and at work was too low by just 5%. Measured and Blended standard deviations were very close, and Blended RMSE was a much smaller value than Measured standard deviation. Both these indicators suggest that the Blended methodology is a valid approach to estimating ambient exposure for Melbourne.

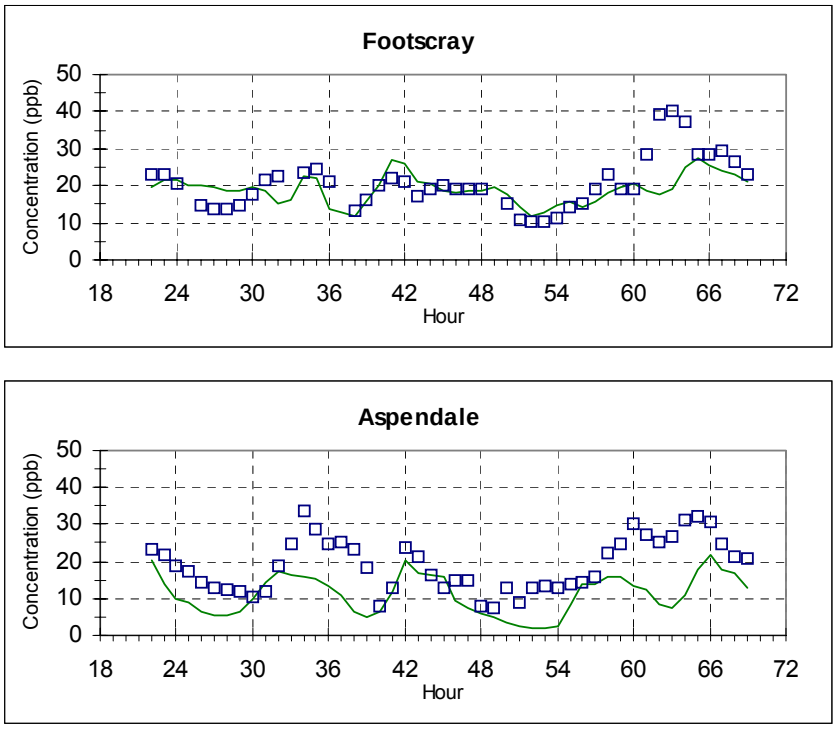

Figure 6.2 Observed (□) and modelled (—) NO_2 concentration (ppb) for the TAPM_CTM (assimilation) simulation for 24-26 May 2006 at Footscray (top) and Aspendale (below).

6.4.2 $PM_{2.5}$

Although predictions of ambient $PM_{2.5}$ concentrations from AAQFS and TAPM-CTM cannot be compared to the indoor and transit measurements, a valid comparison can be made with the outdoor concentrations measured by the EPA Victoria monitoring network plus the Aspendale site. Model performance statistics averaged over all four events are shown in Figure 6.3, in which Monitored refers to the 4-event average of the mean event concentrations measured by the nine EPA Victoria and one Aspendale monitor. AAQFS and T-CTM assim refer to predictions from the AAQFS model and TAPM-CTM model with assimilation of wind data. Neither model performed well, with the mean concentrations being under predicted and standard deviation only fairly predicted. RMSE for both models was larger than the observed standard deviation, indicating that the models were not predicting with skill. Correlations over all four events for the two models were 0.53 and 0.61 respectively.

Statistics for the individual events are listed in Table 6.1, where it can be seen that the model results varied between events. The models' worst performance was for event 3, when RMSE values were much higher than the observed standard deviation and there was almost no correlation between model and observations. The major reason for this was the presence of smoke haze on both days of the event, as there is no source in the models for particulate matter from fires. TAPM-CTM consistently under predicted, with improvements likely to be seen with an updated $PM_{2.5}$ inventory and an incorporation of code to produce secondary organic aerosols. Although AAQFS appeared to predict higher concentrations than TAPM-CTM, this was mostly due to much-higher than observed concentrations being predicted for a couple of hours during

the early-morning and late-afternoon peak traffic hours. This behaviour was reflected in higher values of standard deviation than those predicted by TAPM-CTM.

Table 6.1 AAQFS and TAPM-CTM model performance statistics for each event, evaluated against PM2.5 data from the EPA Victoria monitoring network and the Aspendale site. For each event, there are nine monitors.

Event 1	Monitored	AAQFS	T-CTM assim
Mean	13.5	8.7	6.1
Std Dev	3.0	6.1	3.2
RMSE		6.5	7.6
Correlation		0.64	0.80
Event 2			
Mean	22.2	21.6	11.3
Std Dev	4.7	8.6	3.6
RMSE		8.8	11.8
Correlation		0.11	0.35
Event 3			
Mean	16.3	8.6	5.4
Std Dev	3.8	3.9	2.2
RMSE		9.2	11.6
Correlation		0.05	0.07
Event 4			
Mean	9.5	10.4	5.7
Std Dev	2.5	5.1	3.0
RMSE		4.7	4.9
Correlation		0.31	0.34

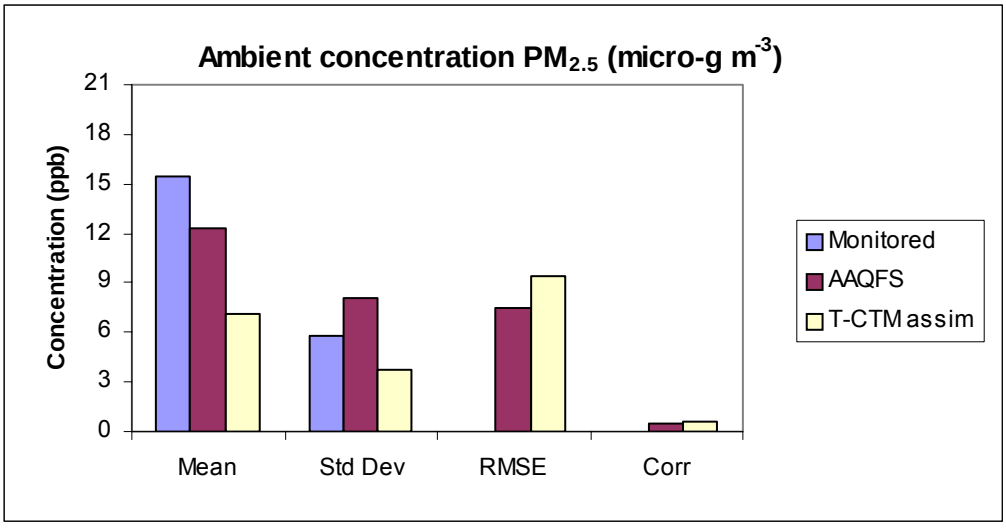

Figure 6.3 Mean, standard deviation, root mean square error (RMSE) and correlation across four events of Aspendale and EPA Victoria-monitored and modelled (AAQFS, T-CTM assim) ambient $PM_{2.5}$ concentrations ($\mu g\ m^{-3}$). Cumulative concentrations over each of the four two-day events at each monitoring site are evaluated. Sample size is 36.

7. ESTIMATES OF NO_2 EXPOSURE

Estimates of exposure to air pollutants over a range of time periods are an essential input to epidemiological studies. Most of these analyses investigate links between *ambient* air quality and health outcomes, because in most cases ambient exposure is the major part of total exposure, and it is easier for regulatory agencies to control sources affecting the population as a whole. In some cases though, it may be important to estimate the actual (or personal) exposure experienced by a population.

Using the diaries kept by each participant, we were able to track their movements 'through' the output fields of each model and calculate the average concentration to which they were exposed over the two-day period. In the same way, average exposures for the time spent in each microenvironment can be calculated and compared to measurements. In this section we present results from an evaluation of simple methods for estimating ambient and personal exposure using (1) monitored data, (2) modelled predictions, and (3) a blended combination of monitored data and modelled predictions.

When analysing large pollutant data sets and activity diaries involving many people over a long time period, it may not feasible to calculate each individual's exposure according to their exact time and space coordinates each day. An approach investigated here is to assume that all participants are at their workplace between 0800 LT and 1800 LT and at their home location between 1800 LT and 0800 LT. Note that time spent in other microenvironments such as transit is ignored in this approach. Our dosage results for the different microenvironments in Figure 4.4 largely support this assumption. An even simpler approach, in which people are assumed to stay inside at home for 24 hours each day, was also investigated. This approach is probably quite representative of more susceptible groups such as the very young, the very old and the ill.

When using only monitored data, we assigned NO_2 concentrations from the nearest EPA Victoria monitor to each participant's home and work locations. Across all participants, the nearest monitor to a home site was at 0.3 km and the two furthest were at 19.1 and 12.9 km. The mean and standard deviation were 6.1 ± 3.9 km. The nearest and furthest work monitors were at 1.0 km and 8.2 km, with a mean and standard deviation of 5.6 ± 2.7 km. A visual comparison can be done by inspecting Figure 3.1 and Figure 3.2.

7.1 Ambient exposure

7.1.1 Using monitoring network data

The average of the mean home outdoor NO_2 concentration measured by samplers while each participant was at home is plotted in Figure 7.1. Also shown is the average of the mean ambient concentration at each participant's nearest EPA Victoria network monitor between 1800 and 0800 EST for the event. The corresponding plot for work outdoor concentration and nearest work monitor concentration between the hours of 0800 and 1800 EST is also shown. Agreement between the means and standard deviations is promising, and RMSE is less than the measured standard deviation for the work locations, and only slightly larger than the standard deviation

for the home comparison. Correlations between the two data sets are 0.49 and 0.77 for home and work respectively.

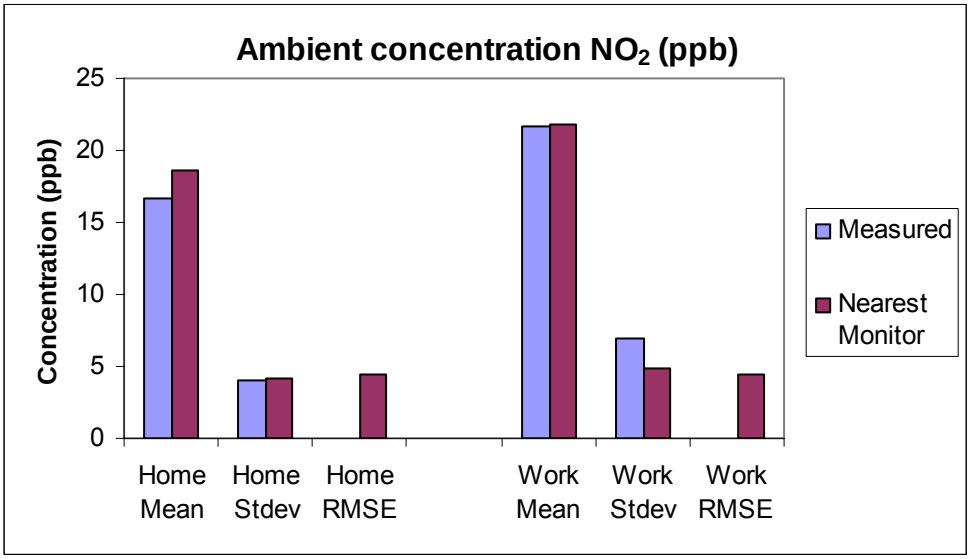

Figure 7.1 Mean outdoor concentration measured when participants were at work and at home, and mean ambient concentration estimated using data from the EPA Victoria monitor nearest to the home and work locations. Standard deviations are shown, as is RMSE between the two data sets.

7.2 Personal exposure

7.2.1 Using a monitoring network data set and a blended data set (monitoring and modelled)

Estimates of personal exposure for each participant were calculated as a time-weighted sum of the mean ambient concentrations (monitored or blended) during the home and work periods, scaled by respective indoor/outdoor (I/O) concentration ratios. Although 62 valid measurements of personal exposure were taken, only 59 could be used for evaluation of the methodologies, due to invalid data at the home or work sites. Ambient concentrations were calculated at the home and work sites (1) from nearest monitor data as done in section 7.1.1, and (2) from blended monitoring and modelled data. In epidemiological studies, the exposure assigned to an urban dweller over a period is often the mean pollutant concentration for that period, averaged over all monitors in the urban monitoring network. Hence each member of the population receives the same exposure value. For comparison with our methodologies here, we also evaluated a third approach to estimating ambient concentration: (3) for each event, assign the mean concentration across all monitors in the network to each participant

The I/O ratios used for each home were computed from the indoor NO_2 concentrations calculated by the two methods developed in section 5.2. A further method, which assigns each home the mean I/O ratio measured across all homes and events, was also evaluated. The three methods can be summarised as:

- *Mass balance*. Applying the steady state mass balance equation, calculate the indoor NO_2 concentration using activity and house characteristics with an outdoor concentration. Then it is straightforward to calculate the I/O ratio.

- *Gas cooking*. One of two indoor-outdoor ratios is assigned to each home according to whether a gas cooking appliance is installed (0.67) or not (0.47). These mean values are obtained from measurements in the current study.

- *Mean measured ratio*. The I/O ratio used is the mean value from all homes measured in this study (0.57).

The same I/O ratio is used for all workplaces and is the mean value measured in this study (0.74).

The mean of all participants' personal exposure values and statistics are listed in Table 7.1 for each of the above I/O estimation methods, under the three approaches to estimating ambient concentrations (nearest monitor, blended and mean monitor concentration). It can be seen that nearly all combinations produced good agreement with the measured values, especially by the criterion that a prediction method is valid if the RMSE is less than the standard deviation of the measurements. However there was a hierarchy of skill amongst the methods. For example, the two methods that assign an I/O ratio according to home characteristics consistently produced better statistical results than that which assigns the same *mean ratio* to everyone. The *mass balance* method was more accurate than the *gas cooking* method, especially for the RMSE and correlation statistics.

As far as the three approaches for calculating the ambient outdoor concentration are concerned, the *blended* approach led to better personal exposures than the *nearest monitor* approach with respect to RMSE and correlation, but was slightly inferior for mean value and standard deviation. However, results from both were more than satisfactory. All statistics were poor for the method in which the personal exposure was calculated by assigning to each participant the *mean monitor concentration* scaled by the mean measured ratio and this approach is clearly inferior to the techniques developed in this project. It must also be remembered that there is no exposure variation between participants using this *mean monitor concentration* method, whereas the standard deviation predicted by the spatial variation techniques matched well the variation seen in the measurements. Note that it is usually the mean *ambient* concentration that is assigned to participants in such studies – in our study this is 18.6 ppb.

For estimation of the personal exposure to NO_2 of a large number of people, it is recommended that best results would be obtained with the I/O ratio calculated from the *mass balance* method. This requires participants to record daily gas cooking periods and approximate house age, although a simpler but slightly less accurate method dependent only on the existence or not of a gas cooking appliance also produced satisfactory results. The recommendation for calculating the required ambient outdoor concentration is to use the *nearest monitor* approach. However there was very little difference between results from the *nearest monitor* and *blended* approaches and the former is only recommended as it is simpler and researchers may not always have access to an emissions inventory or model for the *blended* approach.

A summary of data and measurements needed for the three methodologies using nearest monitor ambient data is presented in

Table 7.2. It can be seen that all three methods are relatively straightforward and do not require field measurements, apart from time-activity diaries. Diary information is minimised too because of our assumption of set periods for participants at home (6 pm to 8 am) and work (8 am to 6 pm).

The statistics from Table 7.1 for the *mass balance* method using ambient concentration data from the *nearest monitor* and *blended* approaches are plotted in Figure 7.2, denoted by H&W (home and work). Also plotted, and denoted by H, are personal exposures similarly estimated, but under the assumption that participants spend the whole measurement period indoors at home. This is a common simplifying assumption in epidemiological studies, except that in those studies the people are assumed to be exposed to outdoor ambient pollution. The results in Figure 7.2 show that the statistics under this assumption were not nearly as good as when the workplace was also taken into account.

Table 7.1 Statistics for various methods used for estimating personal exposure to NO_2 from 59 measurements. Methods are described in the text.

Method	Source of ambient concentration	Correlation	Mean (ppb)	Standard Deviation (ppb)	Root Mean Square Error (ppb)
Measured			**12.1**	**3.2**	
Mass balance	Blended	0.79	11.1	2.8	2.3
Mass balance	Nearest monitor	0.76	11.8	3.0	2.1
Gas cooking	Blended	0.71	12.1	2.8	2.3
Gas cooking	Nearest monitor	0.65	12.5	3.0	2.6
Mean measured ratio	Blended	0.60	12.0	2.3	2.5
Mean measured ratio	Nearest monitor	0.53	12.4	2.5	2.8
Mean measured ratio	Mean of monitors	0.10	10.7	0.0	3.5

Table 7.2 Summary of data needed for three methods for determining personal exposure to NO_2 .

Method	Concentration data	Location data	Home data	I/O ratio (home)	I/O ratio (work)
Mass balance	Nearest network monitor.	Home and work addresses.	Percentage of time using gas cooking. State of windows and doors. House volume and age.	From mass balance equation using home data.	Literature value. Mean for offices.
Gas cooking	Nearest network monitor.	Home and work addresses.	Gas cooking appliance installed?	Literature values according to gas cooking or not.	Literature value. Mean for offices.
Mean ratio	Nearest network monitor.	Home and work addresses.	-	Literature value. Mean for homes.	Literature value. Mean for offices.

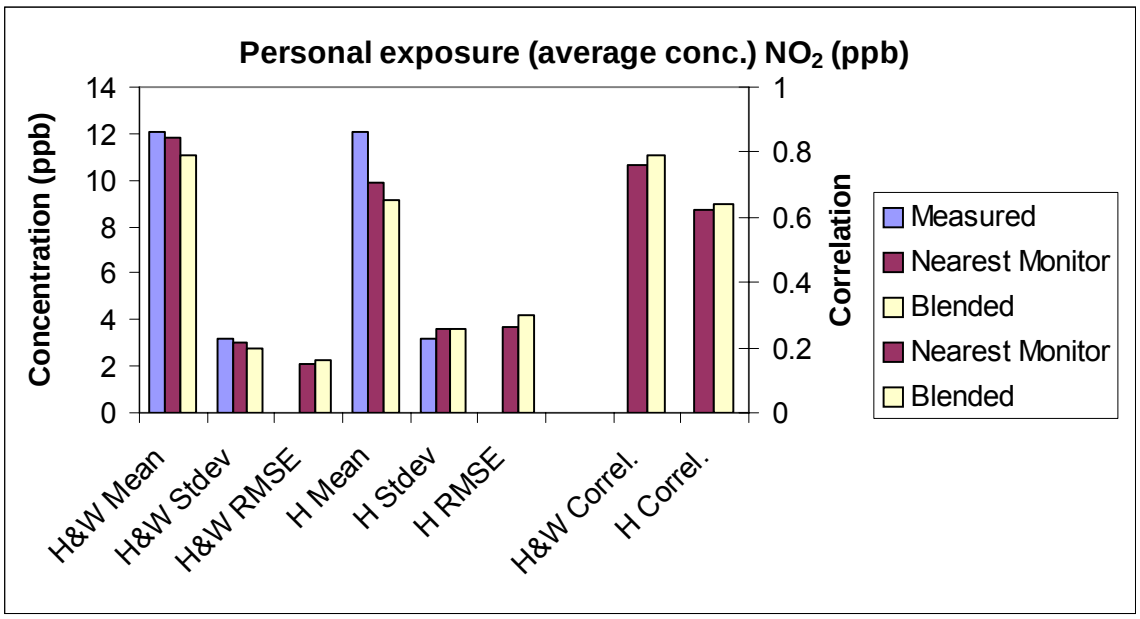

Figure 7.2 Mean, standard deviation and root mean square error (RMSE) across 59 samples of measured, nearest monitor and blended personal exposure (NO_2 concentrations). Values estimated from methodologies using home and work (H&W), and home (H) locations are shown (see text for details).

8. ON-ROAD CONCENTRATION ESTIMATES FOR NO_2

While we have been able to estimate personal exposure with good success by making use of ambient monitoring data, air quality model predictions and activity diaries at home and workplaces without taking account of any transit information, it should be noted that our participants were office workers whose cumulative exposure when travelling was considerably less than that at work and at home. This may not be the situation when estimating personal exposure for people with occupations that entail driving or working near roads. Hence in this chapter we examine whether our methodologies have relevance for estimating exposure in vehicles and at roadsides.

8.1 Inside- and outside-vehicle concentrations

Twenty six measurements of NO_2 concentration, averaged over four journeys per event, were measured inside the vehicles over the four events. Eighteen concentration measurements were also made with samplers attached to the outside of the vehicle. In addition, on five occasions participants measured NO_2 while cycling to and from work.

Table 8.1 shows the mean and other statistics for NO_2 concentration measured outside of vehicles and by cyclists over the four events. Also shown are the values obtained by tracking each participant's route through the concentrations predicted using the blended model and monitor methodology. It is immediately apparent that the predictions significantly underestimated the measured on-road concentrations, but this is to be expected because the former are averaged over a one km^2 square and are unable to resolve the higher on-road concentrations. Naturally the RMSE was high. The estimated standard deviation was less than 50% of the observed value and the correlation was only 0.38, suggesting that there is little predictive skill.

Statistics for the ratio of measured to blended ambient predictions are listed in the final column of Table 8.1. The mean value of 1.75 is the average of values that varied widely, due to the varying meteorology, traffic conditions and volumes across the 23 participant routes. EPA Victoria have conducted a monitoring programme at a number of roadside sites around Melbourne (EPA Victoria, 2006), and results for ratios of near-road to nearest ambient monitor concentrations of NO_2 are listed in Table 8.2. Our on-road value of 1.75 is consistent with the EPA values, considering that the latter were measured at various distances from the roadside and averaged over a number of months.

The data analysis in section 4.3 found that the mean ratio of in-vehicle to out-vehicle NO_2 concentrations in our study was 0.63. Therefore an estimate of in-vehicle concentration can be obtained for each participant by multiplying the out-vehicle blended estimate by the ratio (1.75) in Table 8.1, and then by the ratio 0.63. The resulting statistics for the 26 estimates compared to the measured in-vehicle concentrations are listed in Table 8.3. The mean concentration predicted for inside the vehicle agreed well with the mean measured value, but the other statistics were not good. The predicted standard deviation was only 57% of the observed value and the correlation was poor (0.15). The RMSE was a little higher than the measured standard deviation, indicating too that this approach has little skill as a predictive tool for estimating

transit exposure. This is due to application across all roads of a single value for the on-road to ambient concentrations. Further uncertainty is introduced by assigning a single in-vehicle to out-vehicle concentration ratio, when it is likely that this value is influenced by several factors (see section 4.3 for discussion). The results suggest, not surprisingly, that there is not a strong link between on-road and ambient concentrations and that, apart from direct measurement which is often not possible, further approaches for estimating on-road exposure need to be explored. Modelling at fine-resolution (e.g. 10 m) in the vicinity of roads of interest, using a specific vehicle emission inventory for each road, is one possibility. Roadway models include AUSROADS (EPAV, 2002), CALINE4 (Caltrans) and LWM (Cope et al., 2005).

Table 8.1 Statistics for measured and predicted (blended methodology) NO_2 concentrations on roadways, averaged over 23 values. Measurements were made outside vehicles or on bicycles.

NO_2 (ppb)	Outside Measured	Outside Blended	Ratio: Measured to Blended
Mean	36.0	21.1	1.75
Standard Deviation	9.7	4.2	0.47
Maximum	56.9	27.7	2.59
Minimum	19.3	10.2	0.91
RMSE		17.3	
Correlation		0.38	

Table 8.2 Ratio of near-road to nearest ambient monitor concentrations of NO_2, measured at various roadside locations by EPA Victoria. Also shown is the ratio from our project.

Location	Measurement period (months)	Distance from roadside (m)	Ratio
Francis Street Yarraville	8	5	1.41
Springvale Road Nunawading	6	6	1.54
Westgate Freeway Brooklyn	9	10	1.46
Hoddle Street Collingwood	3	60	1.23
Vehicle trips CARP project	2-4 hours	On-road	1.75

Table 8.3 Statistics for measured and predicted NO_2 concentrations inside vehicles, averaged over 26 estimates and measurements. See text for description of methodology.

NO_2 (ppb)	In-vehicle Measured	In-vehicle Predicted
Mean	21.4	22.8
Standard Deviation	8.0	4.6
Maximum	35.8	30.4
Minimum	6.3	11.2
RMSE		8.5
Correlation		0.15

8.2 Lagrangian Wall Model (LWM)

LWM calculates concentrations in the vicinity of roads with a 10 m grid interval, potentially resulting in a more-realistic estimate of the exposure experienced by people on or near roads. The model solves a similar set of chemistry equations to those in TAPM-CTM (Cope et al., 2005). The two-dimensional wall is moved at the speed of the vertically-averaged wind, allowing considerable speed-up of the solution of the model equations. This allows the model to be operated at very high resolution, O (20 m), making it suitable for modelling near-road air quality impacts. Initial concentrations in the 'wall' (upwind of the road or sources of interest), and boundary conditions at the edges of the wall, are obtained from TAPM-CTM, albeit at a larger scale. Alternatively, the boundary conditions can be merely specified as a typical background concentration. The model moves with the wind across a grid square(s) of a larger-scale model, taking account of emissions from small-scale sources such as roads and point sources.

While we have not actually run LWM for the journeys in our project, we have used information from another Clean Air Research Programme (CARP) project (Cowie et al., 2008) to extend each journey's TAPM_CTM estimate of exposure to include an on-road estimate. Cowie et al. (2008) used LWM to calculate near-road NO_2 concentrations for roads in the Lane Cove area of Sydney for comparison with sampler data obtained in the vicinity of the Lane Cove tunnel before and after its opening. TAPM-CTM was also run in that project and concentrations were calculated for the 16 1-km^2 grid squares covering the area of interest. Over a 12-month period, LWM was run each hour with a 20x20 m grid spacing for each of the 2500 sub-cells in the one km^2 TAPM-CTM cells/grid squares. The mean ratio of the LWM sub-cell concentrations to TAPM-CTM ambient concentrations was calculated for all sub-cells which contained EPANSW road links. These cells contain a mixture of roadways from very large to small, but do not contain residential roadways. The mean ratio over the 16 cells was 1.65, with a minimum value of 1.48 and maximum of 1.91. The standard deviation was 0.12, an encouragingly small value

considering the variation in the mix of road types (and thus traffic volumes) over the many sub-cells.

On-road estimates have been obtained for our project by applying this ratio to each of our participants' exposure calculated from TAPM-CTM. Statistics for these on-road estimates (denoted outside) are listed in Table 8.4. Note that the blended fields rather than the TAPM-CTM fields have been used in the predictions in this Table as they have previously been shown to be more accurate than those from TAPM-CTM. There was good agreement with the mean measured value, and the RMSE was slightly smaller than the observed standard deviation. However there was not as much variation in the predictions as in the observations. It is possible that more variation may be introduced through examining the relationship between the ratio and traffic volume, and applying the appropriate ratio value to participants according to the traffic volume on their route.

Further work would need to be done to evaluate whether this modelling approach is viable for a relatively long-term epidemiology study with many participants.

Table 8.4 Statistics for measured and predicted NO_2 concentrations on roadways, averaged over 23 values. Measurements were made outside vehicles or on bicycles. Outside predictions are calculated by scaling the blended methodology ambient estimate by an LWM-derived ratio of on-road to ambient concentration.

NO_2 (ppb)	Outside Measured	Outside Predicted. Ratio = 1.65
Mean	36.0	34.9
Standard Deviation	9.7	6.9
Maximum	56.9	45.7
Minimum	19.3	16.8
RMSE		9.4
Correlation		0.38

8.3 Summary

In section 7.2, methodologies for estimating personal exposure to NO_2 were based on the application of an indoor-outdoor ratio to an outdoor concentration at a person's workplace or home. Methods were equally successful whether the outdoor concentration was obtained from a blended monitoring and modelled data set or whether it was assigned from the nearest EPA Victoria monitor to work or home. In this chapter, we have evaluated the same general methodology for estimating personal exposure in the transit (road) mode, but have only used the blended approach for estimating the outdoor concentration. Given the very similar statistics

obtained for both approaches for home and workplace exposures, there is no reason to expect that transit exposures could be estimated any better by the nearest monitor approach.

The results show that, for the vehicle transit mode, there is no skill in our general methodology of predicting personal exposure by scaling an ambient concentration by an indoor-outdoor concentration ratio, indicating that if there is any link between ambient and on-road concentrations, it is only a weak one. A mean ratio of on-road to ambient concentrations was calculated from TAPM-CTM and LWM simulations for a mixture of major and minor roads in Sydney's Lane Cove area. When this ratio was applied to our modelled ambient concentrations, statistics for the predictions of roadway concentrations agreed well with those of the measured concentrations. Even more accurate results may be obtained by applying an appropriate ratio to each participants' route, depending on traffic conditions. A suggested alternative approach for calculating on-road concentrations is modelling at fine-resolution (e.g. 20 m) in the vicinity of roads of interest, using a specific vehicle emission inventory for each road.

9. $PM_{2.5}$ FIELD RESULTS AND MODELLING

9.1 $PM_{2.5}$ personal exposure and dosage

For each of the four events, DustTrak monitors logging one-minute $PM_{2.5}$ data were assigned to three 'workers' and to one person who stayed at home. This resulted in data for 15 home, 10 transit (motor vehicle) and 10 work microenvironments, consisting of eight different homes, eight different transit routes and three different workplaces. The range across the participants of personal exposure (2-day mean concentration over an event) for each event is shown in Figure 9.1. The highest personal exposure (23.2 μg m^{-3}) was just below the advisory Air NEPM standard for $PM_{2.5}$ of 25 μg m^{-3} (24-hour average). This participant was a truck driver who spent 41% of the 2-day period in the transit microenvironment.

Also shown is the range of 2-day mean concentration measured by the eight monitors in the EPA Victoria monitoring network across Melbourne. A quick inspection shows that exposure to transit and indoor $PM_{2.5}$ sources and sinks did not cause any participant's personal exposure to lie outside the range of ambient exposure measured across Melbourne for each event. There was also a monotonic increase in the value of each of the four indicators from the lowest event (4) to the highest event (2), indicating that the ambient concentration was a strong component of the personal exposure of the participants. This is especially so as a comparison between events two and four of the four ratios of the extreme concentrations shows that they were very similar (close to 2).

Figure 9.2 shows the maximum and minimum values of exposure measured in the microenvironments of home, transit and work. The highest home concentration slightly exceeded the advisory NEPM standard and occurred during the highest event (2), when the ambient concentration also exceeded the standard (Figure 9.1). However, there were significant contributions from activities within the home (see later). The highest transit value (30.1 μg m^{-3}) was registered by the truck driver. Concentrations measured in the chosen work sites were comfortably below the advisory standard. The maximum, minimum, mean and standard deviation statistics are listed in Table 9.1 for each microenvironment.

Further information can be obtained by examining the dosage of participants in each microenvironment. In this study, dosage is defined by the mean concentration experienced in a microenvironment multiplied by the time in hours spent in that environment. Over the four events, 70% of dosage for the average participant was experienced at home, with 18% at work. The truck driver experienced 55% of his dosage while on the road. These results are skewed by having four of the 15 observations in the home for the duration of the event, whereas across the community the proportion of people who stay home is likely to be less than this figure.

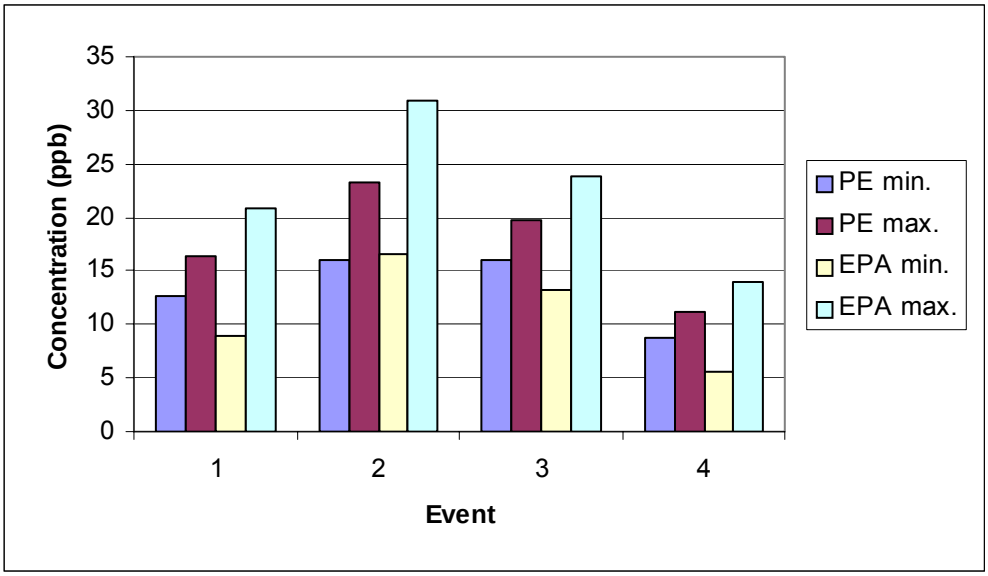

Figure 9.1 Highest and lowest 2-day mean $PM_{2.5}$ concentration measured over 48 hours by participants with DustTrak monitors (PE). Also shown is the range of 2-day mean ambient PM2.5 concentration recorded across the EPA Victoria monitoring network for each event (EPA).

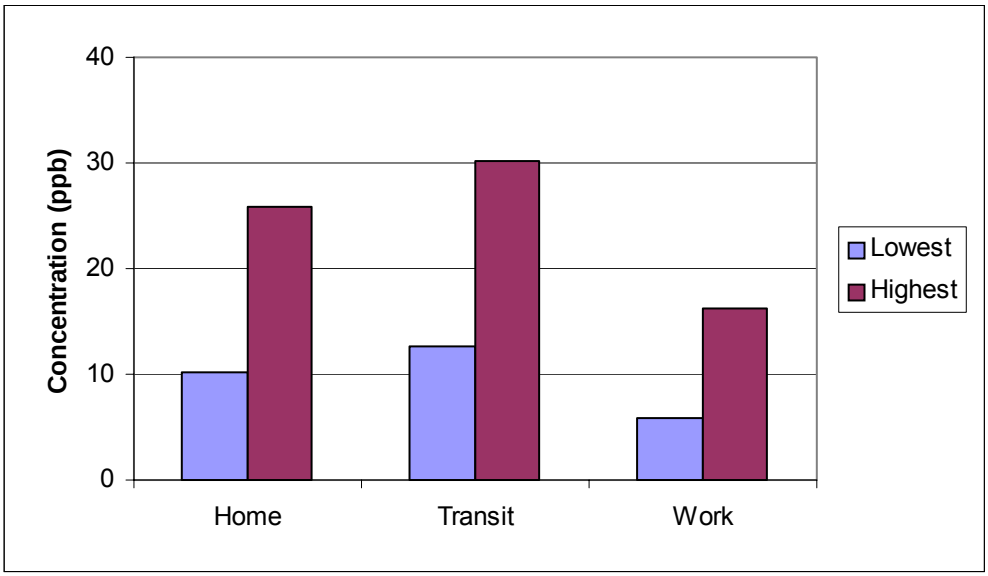

Figure 9.2 Highest and lowest 2-day mean $PM_{2.5}$ concentration (across all four events) measured by participants with DustTrak monitors in the microenvironments of home, transit and work.

Table 9.1 Summary of findings re exposure for $PM_{2.5}$ measurements with DustTrak monitors for 2-day periods. 'All' denotes the personal exposure measured by the monitor across all environments.

EXPOSURE ($\mu g\ m^{-3}$)	Maximum	Minimum	Mean	Standard Deviation	Number of values
Home	25.8	10.1	16.3	4.5	15
Work	16.2	5.8	11.8	3.3	10
Transit	30.1	12.7	23.6	4.9	10
All	23.2	8.7	16.1	4.5	15

Acquisition of 1-minute $PM_{2.5}$ data throughout each event, in contrast to the cumulative NO_2 concentration data, allowed examination of concentrations during situations which are commonly experienced but which occurred for shorter time periods within the 48 hours of each event, and indeed for periods shorter than the averaging periods for the advisory NEPM standards (24 hours and one year). This is especially important in light of findings from the examination by Michaels and Kleinman (2000) of analytical data, and toxicological and epidemiological literature. Their review concluded that for asthmatics, the strength of the PM_{10} association with symptom severity increased as the PM_{10} averaging time varied from 24 h to 8 h to 1 h. In Australia, Simpson et al. (1997) reported that daily mortality in Brisbane was associated with daily maximum 1 h, but not 24 h, average PM_{10} concentrations, while Morgan et al. (1998) reported that admissions of people over 65 into hospitals for chronic obstructive pulmonary disease (COPD) was associated more strongly with daily maximum 1 h PM_{10} concentrations than with 24 h concentrations. The review of Michaels and Kleinman (2000) also examined the findings from experiments involving rats inhaling equal time-weighted average aerosol concentrations, with or without excursions (short periods of high concentration). In these experiments, the area of lung surface developing lesions was elevated in rats breathing the same four-hourly dose of aerosols when the four-hourly average rate of aerosol delivery included at least one short-term ($\geq$ 5 min) burst ($\geq$ 50 %) above the average dose rate.

Figures 9.3 to 9.5 show time series of 1-minute averaged $PM_{2.5}$ concentrations encountered by selected participants during the events of the project. In Figure 9.3, for a participant driving to and working in the CBD in Event 2, the work and home concentrations (windows closed) tracked the outdoor ambient concentration from the nearest monitor fairly closely (see section 7.2 for discussion on nearest monitors), except during cooking periods when the indoor concentrations rose (about 7.30 pm on 7 June and 6 pm on 8 June). The high concentration at the very beginning of the event was due to intense cooking (i.e. burning) 90 minutes earlier. Transit concentrations fluctuated according to speed, congestion and the type of vehicle ahead, as well as travelling through the domain tunnel on each journey.

In the Event one exposure time series shown in Figure 9.4, the work and home indoor concentrations followed the ambient concentration closely, though there was an interesting departure from this trend for the home concentration on the first evening. This occurred when a hair dryer was used for 10 minutes in the same room as the monitor, though not next to it. This may have been a case of the dryer stirring up dust in the room. Concentrations returned to pre-hairdryer values after about 60 minutes. Each transit trip encountered brief high exposure excursions, probably due to congested periods, though a note of 'following a smoky truck' was made at the time of the 87 $\mu g\ m^{-3}$ reading on the final journey.

Interesting transit measurements were also made in Event two by a monitor in the cabin of a delivery truck (Figure 9.5). The high excursions to values between 60 and 100 $\mu g\ m^{-3}$, and which lasted between five and 15 minutes, occurred during delivery stops when the engine was left running; the exhaust outlet on the truck is alongside and just above the cabin. While such excursions could be harmful to health, as noted earlier, it is interesting to note that the mean concentration in the truck over this event was 30.1 $\mu g\ m^{-3}$, which is only slightly above the ambient concentration of 28.2 $\mu g\ m^{-3}$ estimated for the same period from the nearest monitor.

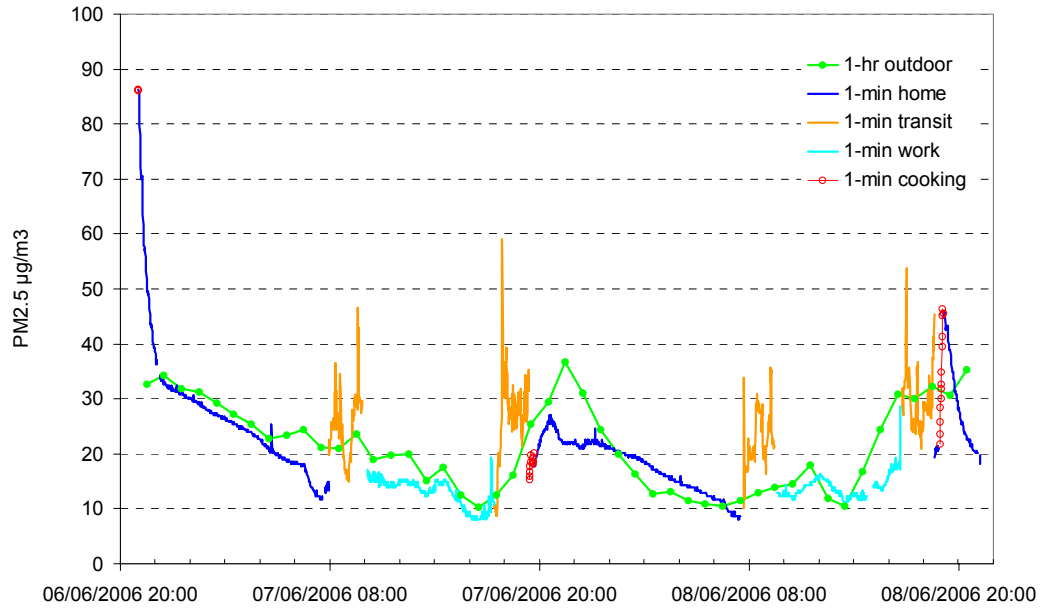

Figure 9.3 One-minute averaged personal exposure (concentrations) of $PM_{2.5}$ for a participant in Event 2. Different microenvironments and activities are denoted by colours according to the legend. Hourly-averaged ambient $PM_{2.5}$ concentrations from the nearest monitor are denoted by the green curve.

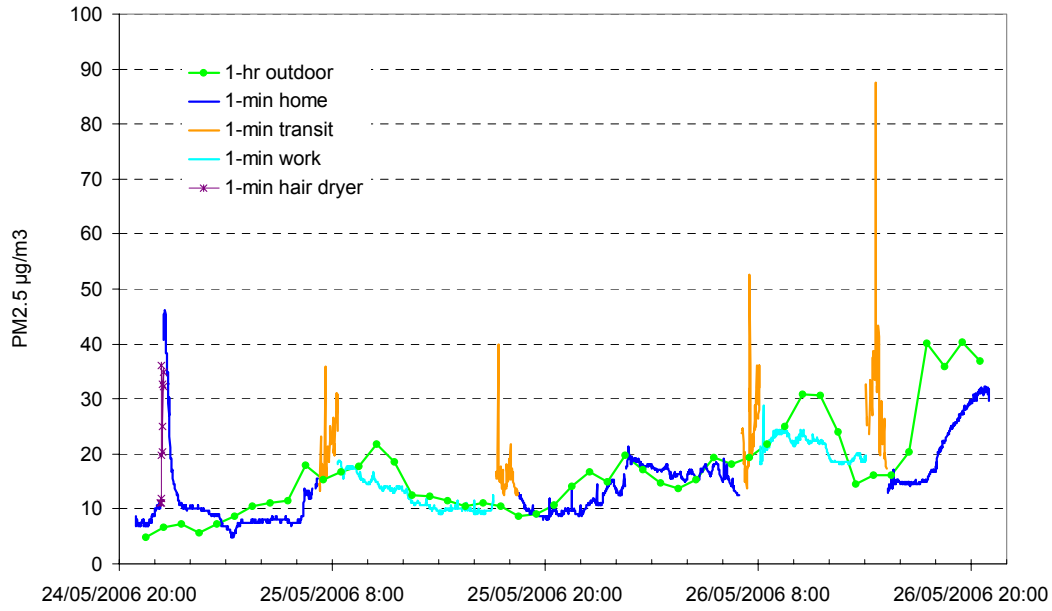

Figure 9.4 One-minute averaged personal exposure (concentrations) of $PM_{2.5}$ for a participant in Event 1. Different microenvironments and activities are denoted by colours according to the legend. Hourly-averaged ambient $PM_{2.5}$ concentrations from the nearest monitor are denoted by the green curve.

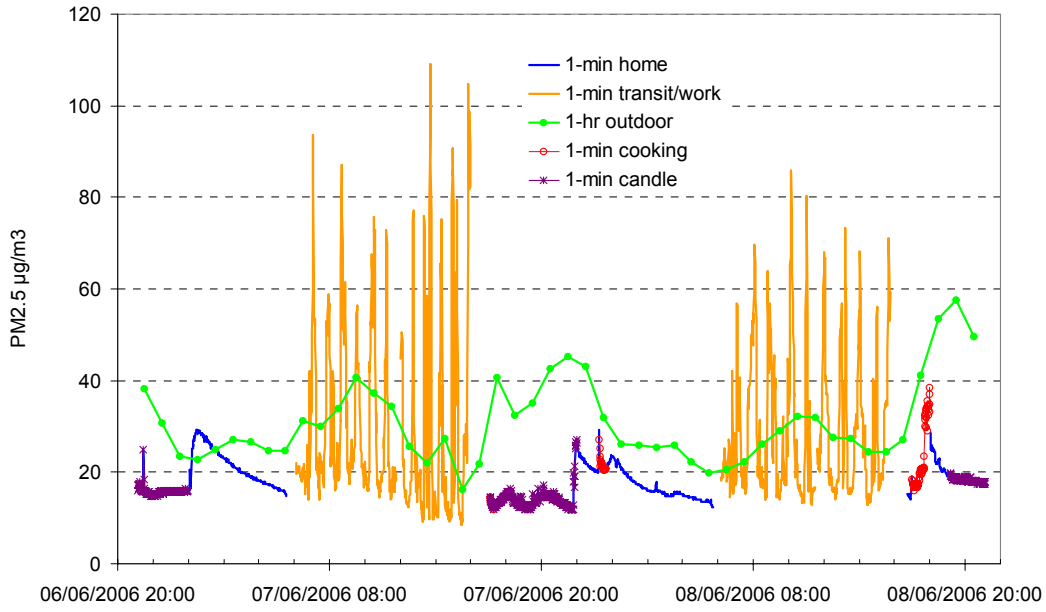

Figure 9.5 One-minute averaged personal exposure (concentrations) of $PM_{2.5}$ for a participant in Event 2. Different microenvironments and activities are denoted by colours according to the legend. Hourly-averaged ambient $PM_{2.5}$ concentrations from the nearest monitor are denoted by the green curve.

Also of interest in Figure 9.5 are the sharp rises in $PM_{2.5}$ concentration just before midnight on 6 June and at 10 pm on 7 June. These were also evident in other events at this home and occurred when the household retired for the night. The activity common to all events at this time was the extinguishing of a candle that has been burning through the evening. The candle was situated about 5 m from the monitor, and the concentrations began to rise about six minutes after the candle had been extinguished. Subsequent $PM_{2.5}$ measurements have been made in the same house, with and without a candle, and confirmed our initial diagnosis. As was the case with peaks arising from cooking or hair drying in Figure 9.3 to Figure 9.5, the $PM_{2.5}$ concentrations took more than one hour to return to pre-activity levels.

9.2 $PM_{2.5}$ indoor–outdoor concentration ratios

Computation of indoor–outdoor concentration ratios for $PM_{2.5}$ is not as straightforward as for NO_2 because no outdoor measurements were made at home or work locations, apart from Aspendale. Ambient concentrations from the nearest EPA Victoria plus Aspendale monitor were examined for their suitability as an estimate for outdoor $PM_{2.5}$. For each event, time series of indoor and nearest monitor hourly-averaged concentrations were examined for similarity in their pattern of behaviour over the two-day period. On most occasions, the correlation was good to very good, in agreement with the finding of Powell and Ayers (2007), from their analysis of 1-minute monitor data inside and outside dwellings, that the indoor concentration closely tracked the outdoor during periods of no indoor activity. However, for some inland home sites in a couple of events in our project, it was found that pattern agreement was poor during part of the event when the nearest EPA Victoria monitor was near the coast (Brighton). On these occasions, substitution of the nearest inland monitor gave quite satisfactory results. Similarly

there were two occasions when a coastal home site showed good agreement with the Brighton monitor, even though it was not the nearest monitor. We believe that these anomalies were due to meteorological factors (the sea breeze penetrating only a short distance inland) and they have been amended in the results that follow.

All inside to outside ratios were calculated using mean concentrations over the two days of each event. The ratios while participants were in each of the three microenvironments are listed in Table 9.2. As expected, there was a wide variation in ratios for the home environment, from 1.26 to 0.52. The mean value was 0.90 with a standard deviation of 0.19. The values greater than or near 1.0 can be traced to high concentrations during periods of activity such as cooking or hair drying (see examples in section 7.1), while one home simply had windows open throughout each event. At all workplaces, the ratio was less than 1.0, with a mean value of 0.58 and standard deviation of 0.15. The workplace value should be treated with caution as eight out of the 10 data values were obtained at Aspendale, with readings varying between 0.41 and 0.64; the other two work places registered ratios of 0.87 and 0.81. Also, the outdoor readings for Aspendale were actually taken at the site, whereas the other two work locations used outdoor values from monitors two and eight km away.

The same 'nearest monitor' procedure was used to calculate inside–outside ratios for the transit mode. It should be noted that the ratios obtained in this way for $PM_{2.5}$ represent an in-vehicle to ambient concentration ratio, whereas the ratios for NO_2 in section 4.3 denote an in-vehicle to roadway concentration ratio. Accordingly the mean in-vehicle to ambient concentration ratio (1.07) from the 10 vehicles with the external vent open in Table 9.2 was higher than for NO_2 (0.63). As discussed for NO_2, windows were closed for all trips and air-conditioning was off. Five vehicles had the heater on, and the four highest ratio readings were registered within this group, but the number of values is too small to suggest an association. As found for NO_2, the vehicle with the vent closed (not included in the statistics) registered the lowest ratio (0.09). The highest value (1.44) received a strong contribution from following a particularly polluting truck for a few minutes, while other sources included tunnels, buses and fire smoke. Interestingly, the ratio in the truck cabin that experienced the regular peaks (Figure 9.5) was only 1.07.

Table 9.2 Summary of indoor/outdoor ratios of $PM_{2.5}$ concentrations from DustTrak monitors averaged over 2-day periods.

IN/OUT RATIOS	Maximum	Minimum	Mean	Standard Deviation	Number of values
Home	1.26	0.52	0.90	0.19	15
Work	0.87	0.41	0.58	0.15	10
Transit	1.44	0.80	1.07	0.19	10

9.3 Modelling personal exposure to $PM_{2.5}$

In a similar manner to the analysis of NO_2 in chapter 7, we evaluated simple methods for estimating personal $PM_{2.5}$ exposure using (1) monitored data, and (2) a blended combination of monitored data and modelled predictions. As in section 7.1, we adopted the simple approach of assuming that all participants were at their workplace between 0800 LT and 1800 LT and at their home location between 1800 LT and 0800 LT. Note that time spent in other

microenvironments such as transit is ignored in this approach. An even simpler assumption, that people stay inside at home for 24 hours each day, is also investigated.

When using only monitored data, we assigned $PM_{2.5}$ concentrations from the nearest EPA Victoria monitor to each participant's home and work locations, even though it was discussed in section 0 that at times the nearest monitor was not the most representative for a particular location. This approach was followed to keep it simple. Across all participants, the nearest monitor to a home site was at 0.9 km and the furthest was at 12.9 km. The mean and standard deviation were 6.5 ± 3.8 km. The nearest and furthest work monitors were at 2.2 km and 8.2 km, with a mean and standard deviation of 7.5 ± 1.9 km.

Estimates of personal exposure for each participant were calculated as a weighted sum of the mean concentrations (monitored or blended) during the home and work periods, scaled by the respective mean indoor-outdoor concentration ratios. The I/O ratios used were the measured home (0.90) and work (0.58) ratios from section 0. The average of all participants' personal exposure values and statistics are plotted in Figure 9.6 and denoted by H&W. Estimates were calculated (1) from nearest monitor data (denoted by Nearest Monitor), and (2) from blended data at the home and work sites (denoted by Blended). Both methods gave acceptable results, though not as good as for NO_2. Observed variation was reproduced well and RMSE was less than observed standard deviation for both approaches.

Also plotted in Figure 9.6, and denoted by H, are personal exposures estimated under the assumption that participants were indoors at the home location over the whole measurement period. For the Blended methodology, these results were better than those obtained with the H&W approach, but the Nearest Monitor results showed a fairly high variation and RMSE. It cannot be concluded whether the H&W or H approach is superior or whether it is better to use Nearest Monitor or Blended Data, because of the small number of measurements and the fact that eight out of 10 workplace data sets were measured at the one location (Aspendale). Even so, the differences between results using the Nearest Monitor data set and the Blended data set were not large, and as for NO_2 suggest that either approach is suitable.

For comparison with our spatial variation methodologies, we have examined the statistics arising from assigning the mean network concentrations to each participant in all four events. The mean and RMSE are plotted in Figure 9.7 (standard deviation is zero) for a personal exposure estimate (Averaged Monitors), obtained by multiplying the ambient values by the mean home indoor/outdoor ratio (0.90) measured in our project. Also shown are the H&W statistics from Figure 9.6. It can be seen that assigning a constant personal exposure value to everyone was a good estimate for the mean participants' exposure. The RMSE values were also good, better than for the Nearest Monitor and Blended methodologies. It must be kept in mind though that there is no exposure variation between participants using the constant concentration approach, whereas the standard deviation predicted by the two spatial variation techniques matched well the variation seen in the measurements. However, the relatively low RMSE values for an approach which assigns each home location the same $PM_{2.5}$ concentration implies that there was not a lot of spatial variation between those locations. This does not mean that there was not much variation across Melbourne (as shown in Figure 9.1), but that the small number of chosen home and work sites did not capture that variation. This is in contrast to the findings from the NO_2 part of the project, for which there were four times as many measurements from a wider variety of locations.

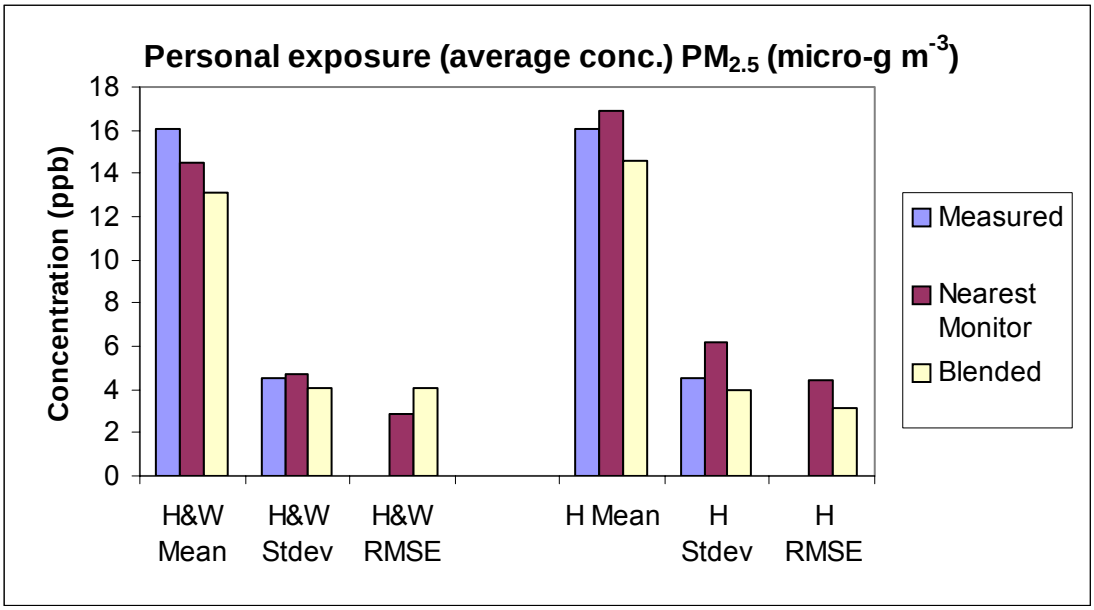

Figure 9.6 Mean, standard deviation and root mean square error (RMSE) across 15 values of measured, nearest monitor and blended personal exposure ($PM_{2.5}$ concentrations). Values estimated from methodologies using home and work (H&W), and home (H) locations are shown (see text for details).

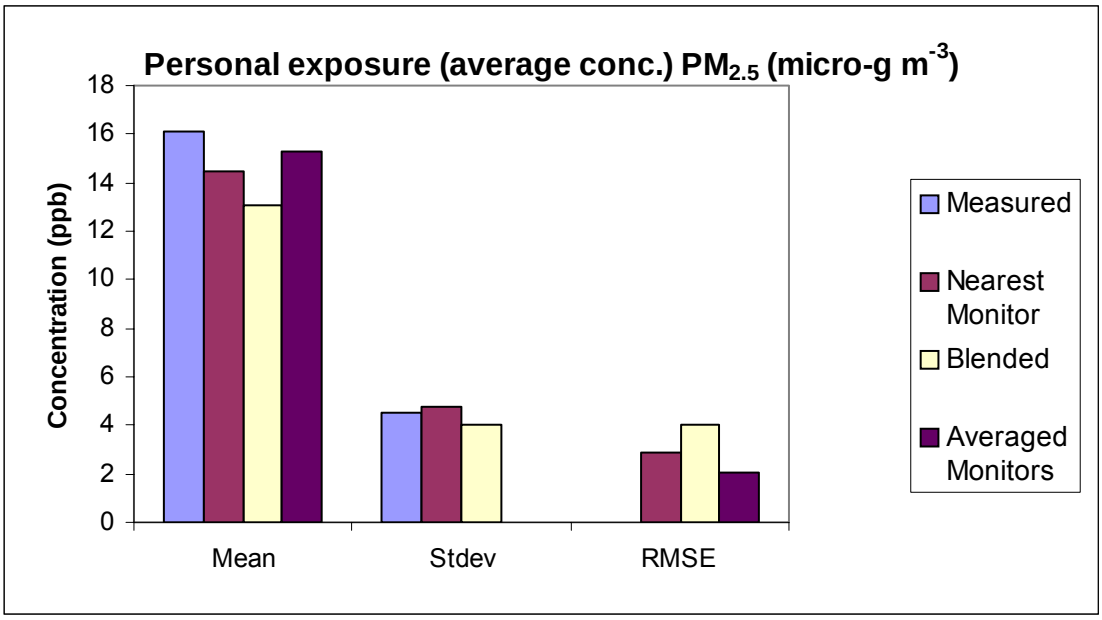

Figure 9.7 Mean, standard deviation and root mean square error (RMSE) across 15 values of measured, nearest monitor and blended personal exposure ($PM_{2.5}$ concentrations). Values estimated from methodologies using home and work (H&W), and from the average of all monitors are shown (see text for details).

10. DISCUSSION AND SUMMARY

10.1 Nitrogen dioxide

A wide range of NO_2 personal exposures (average concentrations), from 6.1 ppb to 19.8 ppb, was experienced across the different activity profiles associated with the 62 measurements. The highest exposures were measured in the transit microenvironment (mean 46.8 ppb), but the major portions of the total dosage (exposure multiplied by time spent in an environment) were experienced at home and at work. For each of the four events, the highest personal exposure did not exceed the maximum ambient NO_2 concentration measured by the EPA Victoria monitoring network, suggesting that for our people profile (office workers, stay at home people and one outdoor worker) the maximum monitored concentration is a conservative estimate for a city's population exposure. However for 19% of measurements, the personal exposure was less than the minimum concentration measured across the monitoring network, indicating that assignation of the city's maximum ambient exposure to everyone would strongly overestimate exposure.

Ratios of indoor to outdoor concentrations at home varied from 0.12 to 1.37 (mean 0.57 ± 0.27), with extreme values attributed to indoor NO_2 sources and to low infiltration rates in new houses. Ratios at workplaces were all less than 1.0 (no indoor sources) and showed much less variability (mean 0.74 ± 0.16). For each home, a mass balance equation was used to calculate indoor NO_2 concentrations, given the outdoor concentration, assuming a steady state and using an assumed deposition rate from previous studies, emission rates according to the time spent cooking with gas and ventilation rates according to house age. This approach gave good agreement with the measured indoor concentrations (correlation 0.78) and could be used in epidemiological studies where only data on cooking time need be recorded in diaries. A simpler approach depending only on whether a gas cooking appliance was installed in the home gave acceptable results with a correlation of 0.63.

The in-vehicle to out-vehicle concentration ratio was calculated for 16 vehicles by attaching samplers to the side mirror. For all trips, windows were closed and air-conditioning was off. A mean value of 0.63 (±0.17) was obtained when the vehicle was driven with the external vent open. However, readings of 0.37 and 0.07 were measured for two trips with the vent closed.

The Australian Air Quality Forecasting System (AAQFS) and CSIRO's air quality model TAPM-CTM were run for each two-day event, and results compared to the outdoor (ambient) concentrations measured by the samplers at home and at work. The models underestimated NO_2 concentrations, especially during the daytime, but good agreement with sampler ambient data was obtained with a blending procedure in which EPA Victoria monitoring data were incorporated into the TAPM-CTM model predictions.

One aim in this study has been to evaluate methodologies for calculating personal exposure. It was hoped to reduce the uncertainty involved in the popular approach whereby a single exposure value is assigned to all persons in a spatially-varying exposure field. To reduce complexity in the methodologies, we assumed that all participants were at home between 1800 EST and 0800 EST, and at work between 0800 EST and 1800 EST. This assumption was supported by our finding that on average only 13% of a participant's dosage was experienced in

the transit micro-environment. Ambient NO_2 exposures for each person for these periods were obtained by two methods: (1) concentrations at the nearest EPA Victoria monitor to home or work were assigned, and (2) concentrations at the home and workplace were assigned from the gridded hourly NO_2 concentrations obtained by blending the modelled and monitored data.

Personal exposure was calculated from these ambient concentrations by using indoor-outdoor ratios at home and work. Home I/O ratios were calculated from the two methods for computing indoor NO_2 concentrations developed from diary data in chapter 5. For each participant, the mean measured ratio across all homes was also evaluated. A constant I/O ratio was used for all workplaces and was the mean value measured in this study. All methods produced good agreement with the measured values, especially by the criterion that a prediction method shows skill if the RMSE is less than the standard deviation of the measurements. Importantly, the standard deviations predicted by these spatial-variation techniques matched well the variation seen in the measurements. Evaluation statistics were poor for a commonly-used method whereby each person is assigned the same ambient concentration, taken to be the mean concentration across all monitors in the EPA Victoria network.

For estimation of the personal exposure to NO_2 of a large number of people, it is recommended that best results would be obtained with the I/O ratio calculated from a mass balance method. This requires participants to record daily gas cooking periods and approximate house age, although a simpler but slightly less accurate method dependent only on the existence or not of a gas cooking appliance also produced satisfactory results. The recommendation for calculating the required ambient outdoor concentration is to use the *nearest monitor* approach. However there was very little difference between results from the *nearest monitor* and *blended* approaches and the former is only recommended as it is simpler and researchers may not always have access to an emissions inventory or model for the *blended* approach.

While these findings are promising, they can only be related at this stage to NO_2 and to the existing EPA Victoria monitoring network. They are also relevant only to persons who spend the majority of their time at indoor work and/or home, allowing time spent in other microenvironments such as transit to be ignored. Results may be different for those who drive for a living or who spend a significant amount of time near roads. With regard to the transit mode, we applied our methodology to estimating in-vehicle exposure, but it did not show any skill. This is primarily because there is little correlation between ambient and on-road concentrations. Our findings may also be of use for the setting of ambient air quality NEPMs.

10.2 Fine particulate matter

The 48-hour averaged values of personal exposure to $PM_{2.5}$ for a smaller group of 15 measurements lay within the range of ambient values measured across the EPA Victoria monitoring network. The highest values measured in the home (25.8 $\mu g\ m^{-3}$) and transit (30.1 $\mu g\ m^{-3}$) microenvironments exceeded the advisory NEPM standard for 24-hour average $PM_{2.5}$ (25 $\mu g\ m^{-3}$), although the standard was also exceeded in the monitoring network for that event. Comparison of hourly-averaged values to the nearest work and home monitors showed that the ambient concentration was a strong component of the personal exposure of the participants.

The 1-minute averages of $PM_{2.5}$ in each microenvironment showed short-period concentration excursions (two to 15 minutes) reaching values five to 10 times higher than the longer-term

average concentration. In the home, these were involved with cooking, a hairdryer and the extinguishing of a candle, and in transit were associated with traffic congestion, smoky vehicles and an idling truck in which exhaust fumes entered its cabin. These findings are relevant in the light of epidemiological and toxicological work showing stronger respiratory health impacts from $PM_{2.5}$ concentrations measured over intervals shorter than the NEPM averaging period of 24 hours.

The indoor concentration trends tracked hourly-averaged ambient concentrations from the nearest monitor, with short-term deviations associated with activities in the home. The mean indoor-outdoor ratio was 0.90 (±0.19), ranging between 1.26 and 0.52, with the dominant source being cooking, and smaller contributions from hair dryers and candles. In the workplaces, with no obvious sources, ratios were all below 1.0 with a mean of 0.58 (±0.15). While no $PM_{2.5}$ readings were taken directly outside the vehicles, concentrations from the nearest monitor were used to obtain ratios of in-vehicle to ambient concentration. These ranged from 1.44 to 0.80 with a mean of 1.07 (±0.19). Only 6% of a 24-hour day was spent in transit by our predominantly office-worker cohort, although this percentage rose for a truck driver who was on the roads for 41% of his time, where he was exposed to a mean $PM_{2.5}$ concentration of 30.1 $\mu g\ m^{-3}$.

The AAQFS and TAPM-CTM models did not perform as well as for NO_2, with the mean concentrations under predicted, typically by 50%, and standard deviation only fairly predicted. RMSE for both models was larger than the observed standard deviation for all events, indicating that the models did not predict with any skill. Correlations were 0.53 and 0.61 respectively. The models' worst performance was for event 3, when RMSE values were much higher than the observed standard deviation and there was almost no correlation between model and observations. The major reason for this was the presence of smoke haze on both days of the event, as there is no source in the models for particulate matter from fires. Improvements in model performance are likely to be seen with an updated $PM_{2.5}$ inventory and an incorporation of code to predict secondary organic aerosols.

In a similar manner to the analysis of NO_2, simple methods for estimating personal $PM_{2.5}$ exposure were evaluated using (1) monitored data, and (2) a blended combination of monitored data and TAPM-CTM modelled predictions. The same approach was adopted, assuming that all participants were at their workplace between 0800 LT and 1800 LT and at their home location between 1800 LT and 0800 LT. The I/O ratios used to link personal exposure to ambient exposure were the mean measured home (0.90) and work (0.58) ratios.

Both the nearest monitor and blended data methods gave acceptable results, though not as good as for NO_2. A simpler approach in which all participants were assumed to be at home for the duration of each event also gave acceptable results. However, it was not possible to conclude whether this or the home plus work approach is superior or whether it is better to use the nearest monitor or the blended data method, probably because of the small number of measurements and the fact that eight out of 10 workplace data sets were measured at the one location (Aspendale). Even so, the differences between results using the nearest monitor data set and the blended data set were not large, and as for NO_2 suggest that either approach is suitable.

For comparison with our spatial variation methodologies, we have examined the statistics arising from assigning the mean concentration from the EPA Victoria monitoring network to each participant in all four events. While the mean and RMSE values were good, it must be kept

in mind that there is no exposure variation between participants using this constant concentration approach, whereas the standard deviation predicted by the two spatial variation techniques matched well the variation seen in the measurements. However, the relatively low RMSE values for an approach which assigns each home location the same $PM_{2.5}$ concentration implies that there was not a lot of spatial variation between those locations. This does not mean that there was not much variation across Melbourne, only that the small number of chosen home and work sites did not capture that variation. This is in contrast to the findings from the NO_2 part of the project, for which there were four times as many measurements from a wider variety of locations, and in which greater confidence can be assigned to the results.

10.3 Bias and variability

The 59 NO_2 personal exposure measurements for which model estimates were calculated are not totally independent. Table 3.1 shows that these were made in 24 different homes, eight different work locations and 25 different transit routes. 24 people took part, with three following the same routine, i.e. the same house, workplace and route in all four events and six doing likewise in three events. It is possible that the model did extra well for these nine routines and that their presence in the data set is introducing bias. They constitute 30 of the 59 measurements, near enough to 50%. However, examination of the 10 most accurate predictions from the *mass balance* and *nearest monitor* methodology revealed that only 40% belong to the set of measurements from these nine repeated routines. In fact nine of the top 10 are measurements made from independent routines, indicating no bias towards any individual(s). Of the 10 worst predictions, 50% are from the nine repeated routines.

As well as the constituency of each cohort (between 15 and 17 people) differing across the four events, the spatial distribution of NO_2 varied between events. This can be seen from Table 10.1 which lists the ranking of each EPA Victoria monitoring station according to its mean NO_2 concentration averaged across the two days of an event. No two ranking sets are remotely alike, indicating different patterns of NO_2 across Melbourne for each event. For example, event two concentrations in the west of Melbourne were higher than those in the east, whereas the opposite situation existed in event 4. Maximum and minimum concentrations and the standard deviation are also shown to illustrate the variation across the network.

Table 10.2 shows the evaluation statistics for our methodology to estimate personal exposure (section 7.2), broken down into the four events. The statistics for the combined data set are also listed. The methodology is the *mass balance* method using ambient concentration data from the *nearest monitor*. The statistics clearly show that the correlation is good, the variability in the predictions matches the observed variability well (column three values are very close to 1), and the magnitude of the error is comfortably less than the observed variation in exposure (column four values less than 1), all of which indicate that the methodology is predicting with skill. This is the situation not only for the combined data set (as shown in section 7.2), but also for each of the four events, which consist of different mixes of individuals/microenvironments and different air quality patterns. While acknowledging that the methodology needs to be applied to larger cohorts and in different cities for further validation, we believe that it is a promising approach that is able to capture the variability in personal exposure across an urban area.

Table 10.1 Ranking of stations in the EPA Victoria monitoring network according to the mean 2-day NO_2 concentration over each 2-day event. Statistics across the stations are also shown.

	Event 1	Event 2	Event 3	Event 4
Alphington	2	4	5	3
Altona North	4	1	7	7
Box Hill	6	8	4	2
Brighton	5	5	6	8
Dandenong	7	7	3	3
Footscray	3	2	1	4
Mooroolbark	8	9	8	6
Point Cook	9	3	9	9
Richmond	1	6	2	1
Maximum NO_2 (ppb)	23.7	27.6	23.7	22.1
Minimum NO_2 (ppb)	9.3	10.6	8.6	9.3
Std. Devn. NO_2 (ppb)	4.8	4.9	4.8	4.6

Table 10.2 Evaluation statistics for the personal exposure estimation methodology referred to as the mass balance method using ambient concentration data from the nearest monitor. Stdev denotes standard deviation and RMSE denotes root mean square error.

	Correlation	Stdev (model) / Stdev (obs)	RMSE (model) / Stdev (obs)
Event 1	0.83	0.91	0.56
Event 2	0.75	0.94	0.69
Event 3	0.79	1.15	0.72
Event 4	0.66	0.84	0.77
All events	0.76	0.93	0.67

10.4 General

Our results for NO_2 and $PM_{2.5}$ are relevant for estimating the personal exposure of individuals in epidemiological cohort studies or for calculating an average exposure for a population. In a population exposure study, the best results would be obtained by using a representative number

of participants for various activity profiles within the population. Exposure results from each profile would be weighted according to the profile sub-population, and summed. Such activity-profile categories could include 1) people who predominantly stay at home, 2) those who go to work indoors, 3) those who work outdoors or spend recreation time outdoors, and 4) those who spend a significant amount of time on or near roads.

We believe that our research findings contribute to estimating exposure within the above activity profiles of staying at home and working indoors (categories one and 2). Our time at work period (0800 EST to 1800 EST) could perhaps be reduced for some sub-groups within the working indoors category (e.g. school children), with an outdoors category (3) added for two or three hours. Our encouraging results for estimating home and work outdoor concentrations from the ambient monitoring network suggest that exposure while outdoors, at work or recreation, could be assigned from concentrations at the nearest monitor.

Our work indicated that there is not a strong relation between on-road concentrations and ambient concentrations, thus ruling out application of our methodology for estimating on-road exposure. For this activity profile (4), further research should be done to relate transit exposures to key variables, including traffic volume and ambient concentration at the nearest monitor. Measurement work on concentration as function of distance from a road, such as that done by EPA Victoria (2006), and the relation between in-vehicle to out-vehicle concentration ratios and in-vehicle comfort settings and cabin volume are also important for developing straightforward exposure methodologies. Contributions can also be made through modelling concentrations at fine-resolution (e.g. 20 m) in the vicinity of roads of interest, using a specific vehicle emission inventory for each road.

As the meteorological conditions for our field campaigns were similar for all four events and were chosen to maximise concentrations of both pollutants, as well as their spatial variation, it is expected that our methodology would be equally applicable under more dispersive conditions, such as more uniform or stronger winds across the area of interest when concentrations and spatial variation would be smaller. For estimates of annual population exposure, it is necessary to evaluate exposure under the major meteorological conditions and then weight the results according to the annual frequency of each category.

The above discussion is equally as relevant for personal exposure of individuals in an epidemiological cohort study, except that their exposure is estimated every day of the study and so the previous discussion re the weighting of results under different meteorological conditions does not apply.

While our research has identified a simple exposure methodology that could be widely applied, without the need for access to air quality models and with only minimum information from respondents, there are some simplifying assumptions that need support from further research. Strictly speaking, the findings can only be related at this stage to NO_2 and to the existing EPA Victoria monitoring network, although it is expected that the methodology would also be valid for cities with monitoring networks of similar density to that of Melbourne. Our sample size for NO_2 was necessarily limited to a total of 24 volunteers, with between 15 and 17 participating in each of the four events. However our methodologies showed skill for each event, as well as for the combined data set involving 59 samples. Repetition of our work, ideally in another city and with a higher number of participants, is highly desirable and would strengthen the findings of

this project. More participants would also widen the variety of homes, workplaces and even ages.

Our methodologies were also successful for $PM_{2.5}$, where the relation between indoor and ambient concentration was stronger than for NO_2, but the sample size was only 25% that of the NO_2 data set. Consequently, more work along the same lines is needed in this area too.

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APPENDIX A - REFERENCES

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APPENDIX B – TIME-ACTIVITY DIARIES

"Household" Exposure Diary

Storage vial numbers for indoor household samplers
Storage vial numbers for outdoor household samplers
Household address, suburb
Position of outdoor samplers
Comments or problems

"Household" Exposure Diary

Household entry date and time	_ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm
Date and time indoor samplers opened and worn	_ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm
Date and time outdoor samplers opened and installed	_ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm

Please complete the exposure and ventilation table

Exposure / Ventilation state	Location in house	Time On	Time Off
Gas heating: wall, space, ducted Other heating / cooling Cooking gas oven / cook top Cooking electric oven / cook top Kitchen extraction fan / hood Bathroom / other extraction fan Window / door open to outside	 Who cooked? Who cooked?		
Gas heating: wall, space, ducted Other heating / cooling Cooking gas oven / cook top Cooking electric oven / cook top Kitchen extraction fan / hood Bathroom / other extraction fan Window / door open to outside	 Who cooked? Who cooked?		
Gas heating: wall, space, ducted Other heating / cooling Cooking gas oven / cook top Cooking electric oven / cook top Kitchen extraction fan / hood Bathroom / other extraction fan Window / door open to outside	 Who cooked? Who cooked?		

Please estimate the duration of each type of activity during this exposure period

Sleeping / lying down	_ _ _ _ _ _ _ _ _ _ _ _ _ _
Sitting (reading, watching TV, eating, etc)	_ _ _ _ _ _ _ _ _ _ _ _ _ _
Personal care (showering, getting dressed, etc)	_ _ _ _ _ _ _ _ _ _ _ _ _ _
Housework (ironing, vacuuming, dusting, cleaning, washing)	_ _ _ _ _ _ _ _ _ _ _ _ _ _
Cooking (food preparation, cooking)	_ _ _ _ _ _ _ _ _ _ _ _ _ _
Other (please describe)	_ _ _ _ _ _ _ _ _ _ _ _ _ _
Outdoor at home (please describe)	_ _ _ _ _ _ _ _ _ _ _ _ _ _

Household exit date and time	_ am/pm
Date and time indoor samplers closed	_ am/pm
Date and time outdoor samplers closed	_ am/pm

"Workplace" Exposure Diary

Storage vial numbers for indoor workplace samplers
Storage vial numbers for outdoor workplace samplers
Workplace street address, suburb
Workplace entry date and time _ _ _ _ _ _ _ _ _ _ _ _ _am/pm Time indoor workplace samplers opened and worn _ _ _ _ _ _ _ _ _ _ _ _ _am/pm Time outdoor workplace samplers opened and installed _ _ _ _ _ _ _ _ _ _ _ _ _am/pm Location of outdoor samplers at workplace

Please complete the exposure and ventilation table

Exposure / Ventilation state	Description and location at work	Time Start (am/pm)	Time End (am/pm)
Near operating vehicles / machinery Near cigarette smoke Near operating gas appliances Window / door open to outside			

Please estimate the duration of each type of activity during this exposure period

Active indoor (walking, moving, cleaning, etc) _ Sedentary indoor (sitting at desk, standing still etc) _ Other indoor (please describe) _ Active outdoor (walking, moving, gardening, etc) _ Sedentary outdoor (sitting, standing still, etc) _ Other outdoor (please describe) _
Workplace exit date and time _ am/pm Time indoor workplace samplers closed _ am/pm Time outdoor workplace samplers closed _ am/pm

Comments or problems

"Transit" Exposure Diary
To be used when travelling to and from the workplace

Storage vial numbers for transit samplers that are worn
Storage vial numbers for samplers placed on car exterior Location of samplers on car exterior
Comments or problems

"Transit" Exposure Diary

Start destination street address, suburb			
End destination street address, suburb			
Date and time travel started _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm Date and time transit samplers opened and worn _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm Date and time car exterior samplers opened _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm			
Fill out the table for all types of transport used to travel from the start to the end destination. Include details of petrol stops (car trips) or waiting times if on public transport.			
Mode of transit (car, bus, bike, train, tram, walk)	Duration	Bus, tram, train route or route number	Roads travelled on route
For car transport, please circle the ventilation and temperature settings used during the trip			
Car windows open / closed	Heating on / off	Climate control on / off	
Fan circulation internal / external	Air conditioning on / off		
Date and time travel ended _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm Date and time transit samplers closed _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm Date and time car exterior samplers closed _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm			

"Other Location" Exposure Diary

"Other Location" samplers to be used when in locations other than home and work. This includes the travel to and from the location.

Storage vial numbers for "other location" samplers			
Description of location and activity (i.e.. indoor shopping centre, movie theatre, football, doctor's office, other people's houses, etc)			
"Other Location" street address, suburb			
"Other Location" entry date and time _ _ _ _ _ _ _ _ _ _ _ _ _am/pm Time "Other Location" samplers opened and worn _ _ _ _ _ _ _ _ _ _ _ _ _am/pm			
Please complete the exposure and ventilation table			
Exposure / Ventilation state	Description of location	Time Start (am/pm)	Time End (am/pm)
In transit (please describe) Near operating vehicles / machinery Near cigarette smoke Near operating gas appliances Window / door open to outside			
Please estimate the duration of each type of activity during this exposure period			
Active indoor (walking, moving, shopping, etc) _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ Sedentary indoor (sitting in cinema, standing in line etc) _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ Other indoor (please describe) _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ Active outdoor (walking, moving, gardening, sport, etc) _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ Sedentary outdoor (sitting, standing still, etc) _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ Other outdoor (please describe) _ _ _ _ _ _ _ _ _ _ _ _ _ _ _			
"Other location" exit date and time _ am/pm Time "other location" samplers closed _ am/pm			
Comments or problems			

"Household" Exposure Diary – DustTrak $PM_{2.5}$

Household entry date and time _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm
Date and time DustTrak monitor opened _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm

Activities	Room/s in house	Time On	Time Off
Cooking activities (please circle or describe)			
Stove top – gas / electricity			
Oven – gas / electricity			
Were you heating / frying food / grilling?			
Toaster / microwave			
Other?			
Heating (please circle or describe)			
Gas - ducted / wall			
Electric fan / radiator / Reverse-cycle (split)			
Wood			
Other?			
Cooling (please circle or describe)			
Evaporative / reverse-cycle split system			
Other?			
Cleaning activities (please circle or describe)			
Sweeping / Vacuuming / Dusting			
Other?			
Smoke activities (please circle or describe)			
Burning candle / incense			
Other?			
Ventilation (please circle or describe)			
Extraction fan kitchen / bathroom / other			
Window(s) open to outdoors			
Door(s) open to outdoors			

Please estimate the duration of each type of activity during this exposure period	
Sleeping / lying down	_ _ _ _ _ _ _ _ _ _ _ _ _
Sitting (reading, watching TV, eating, etc)	_ _ _ _ _ _ _ _ _ _ _ _ _
Personal care (showering, getting dressed, etc)	_ _ _ _ _ _ _ _ _ _ _ _ _
Housework (ironing, vacuuming, dusting, cleaning, washing)	_ _ _ _ _ _ _ _ _ _ _ _ _
Cooking (food preparation, cooking)	_ _ _ _ _ _ _ _ _ _ _ _ _
Other (please describe)	_ _ _ _ _ _ _ _ _ _ _ _ _

Household exit date and time	_ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm
Date and time DustTrak monitor closed	_ _ _ _ _ _ _ _ _ _ _ _ _ _ _ _ am/pm