
**The determinants of levels of secondary
particulate pollution and nitrogen
dioxide in urban New Zealand – Part 1**

**NIWA Client Report: AKL2008-053
July 2008**

NIWA Project: PCAC081

The determinants of levels of secondary particulate pollution and nitrogen dioxide in urban New Zealand – Part 1

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Prepared for

FRST

NIWA Client Report: AKL2008-053
July 2008

NIWA Project: PCAC081

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Reviewed by:

Approved for release by:

Executive Summary

“Protecting New Zealand’s Clean Air” is a research programme currently being funded by The Foundation for Research Science and Technology (FRST. Contract number C01X0405). The Processes: Modelling and Chemistry research objective of this Programme aims to develop and apply modelling methods that will be used to quantify air pollution effects and predict the outcomes of mitigation and management options. The objective also aims to determine the specific pathways leading to air pollution in New Zealand, and apply these to emissions management options in order to reduce concentrations of chemically reactive contaminants. It contributes to the programme outcomes by assessing levels of secondary air pollutants (specifically NO₂, ozone and secondary particulate matter) in populated areas of New Zealand.

This is part one of a two-part report looking at the determinants of high levels of some secondary pollutants in urban New Zealand. Taken as a whole, these reports specifically focus on secondary sulphate arising from shipping emissions and nitrogen dioxide, although in doing so it also aims to develop tools for future research into other secondary pollutants and to further our understanding of the general physical and chemical processes which lead to elevated levels of air pollution in New Zealand’s cities.

Part One introduces the existing state of knowledge, discusses some general spatial patterns and temporal trends from monitoring data and identifies some key issues and knowledge gaps. It also introduces and trials some analytical methods which may fill those knowledge gaps. Part Two will report on extension of those initial trials and the combination of various methods. It will then review the major determinants of high levels of secondary sulphate and nitrogen dioxide, and propose a range of mitigation options, as well as indicating how the tools developed can be used to assess these options.

One of the motivations for this research was the tentative finding from a recent PM₁₀ source apportionment study in Auckland that episodically elevated levels of sulphate in the Auckland airshed might be associated with an SO₂ source in the Port of Auckland. In Part One we have shown how advanced meteorological modelling has predicted that one of these episodes (on 19th March 2006) corresponded to a period in which air originating in the vicinity of the Port was advected into the Hauraki Gulf, but recirculated back to land and back to Auckland within 24 hours due to a local land/sea breeze system. We have concluded that such recirculation may not be uncommon in Auckland and that this method is a valuable tool which, in combination with source apportionment (receptor modelling) data may reveal the potential for mitigating elevated levels of sulphate (and other secondary particulates) in coastal airsheds containing ports.

This report reviews monitoring data captured by Auckland Regional Council, Greater Wellington Regional Council, Environment Canterbury and Transit NZ. We find that the highest long-term and short-term peak levels of nitrogen dioxide are found in downtown locations alongside streets with busy traffic in Auckland, especially where the buildings form street canyons. It is at these locations that breaches of the National Environmental Standard or World Health Organisation annual guideline are most likely to occur and initial analysis shows that lower NO_x concentrations are required to trigger an NES breach at the Queen Street II site in Auckland compared to other monitoring sites. A lack of observational data prevents us from determining how probable such breaches are in other cities as the few current sites probably do not represent the worst case. Current trends in NO₂ concentrations in such locations appear to be downwards. After these locations, the highest observed concentrations are alongside motorways in Auckland. Although the NES is far from being breached at any existing motorway monitoring sites there is some evidence of an increasing trend in NO₂ alongside motorways, even where a decreasing trend in total oxides of nitrogen (NO_x) is observed.

The NO₂ concentrations reported by a roadside monitor are highly sensitive to its precise location. ARC have found that monitors either side of a busy motorway report significantly different concentrations. Distance of the monitor to the road and the presence of a street canyon all appear to be very significant modifiers. These influences will be discussed in detail in Part Two.

Our initial research has revealed no evidence to suggest these trends are caused by changes in level of background regional oxidants. Photochemical episodes involving a doubling or more of regional ozone concentrations appear to be exceedingly rare events in Auckland and are not significant in terms of long-term NO₂ trends.

The motorway-site trends may be related to long-term changes in traffic flow, with congestion becoming increasingly common on Auckland's motorways in this decade, and with increases in the proportion of NO_x emitted directly as NO₂ which we show is very significant at roadside sites. This suggests that changes in the traffic fleet mix and technology mix within that fleet may be a significant influence on future roadside NO₂ levels. Part 2 will consider the issue of changes to traffic flow and direct emissions in more detail.

We have concluded that high resolution meteorological modelling and trajectory analysis, semi-empirical chemical source apportionment for NO₂, and empirical photostationary state modelling are three analytical approaches which together can help us to understand the processes underlying the patterns and trends discussed in this report. These approaches, and others, will be developed further in this Programme and will be discussed in detail in Part Two. However, our analysis is limited by the lack of observational data, especially NO₂ monitoring at peak sites outside Auckland, roadside and urban ozone data and regional background nitrogen dioxide.

1. Introduction

1.1 Objectives

This is Part One of a two-part report. It describes research conducted within the FRST Programme “Protecting New Zealand’s Clean Air”. It is concerned with levels of secondary sulphate and nitrogen dioxide of anthropogenic origin in the atmosphere of urban New Zealand. For both pollutants Part One seeks to

- Review data on ambient concentrations in terms of NES, RAQT and WHO guideline compliance,
- Review the available data to scope its ability to describe and explain the determinants of ambient concentrations,
- Trial some analyses that may help to improve our understanding of the processes leading to elevated concentrations,
- Identify analyses which should be pursued in Part Two and beyond to address knowledge gaps regarding spatial variability, relevant physico-chemical processes, future projections and our ability to propose and evaluate mitigation strategies.

Part Two will be delivered to FRST at the end of September 2008. This second report will

- Extend the trial analyses presented in Part One,
- Summarise the current state of knowledge, including projections into the future,
- Identify some key mitigation strategies,
- Identify the remaining knowledge and data gaps that need to be addressed to confidently assess mitigation strategies.

1.2 Scope

Although this research is motivated by the need to understand whether control of primary emissions can be used to meet air quality standards and guidelines for secondary pollutants it does not rely on explicit chemical transport modelling. The limitations in our ability to conduct such modelling and use it to meet our objectives is discussed in chapter 3. In attempting to understand the scope for mitigating secondary PM this report focuses on what can be inferred about formation of secondary sulphates in urban airsheds from meteorological modelling alone. Part two of this report will review whether the future application of chemical transport modelling to this problem is justified and likely to produce credible information. Nitrates will not be directly considered at this point due to the much lower quantity and quality of information about nitrates in the urban air in New Zealand compared to sulphates. The future direction of research into nitrates will be discussed in Part 2. Other secondary particulates, including organic species and halides are not covered in this report.

This report is deliberately biased towards a consideration of peak levels. This is due to a focus on building a knowledge base to help understand the causes, occurrence and nature of exceedences of threshold-based targets (the NES, MfE and WHO guidelines and regional targets).

1.3 Acknowledgements

The monitoring data used for the analysis in this report was kindly provided by Auckland Regional Council (courtesy of Janet Petersen), Environment Canterbury (courtesy of Teresa Aberkane and Vicky Lucas), Greater Wellington Regional Council (courtesy of Tamsin Mitchell) and Transit New Zealand (courtesy of Annette Bolton & Carl Reller). We are grateful for the assistance of Shanju Xie of Auckland Regional Council for providing a useful review of an early draft. Further comments and corrections to the final draft were also provided by Teresa Aberkane, Vicky Lucas and Shanju Xie.

2. Background

2.1 Secondary sulphate from shipping

Regional Councils in New Zealand are charged with introducing policies and regulating emissions so as to achieve compliance with the National Environmental Standards for Air Quality within their designated airsheds. Each Region has the power to introduce Regional Air Quality Targets (RAQT) which in some cases are more stringent than the NES, or cover additional pollutants and/or averaged over different timescales.

In most urban areas in New Zealand the most challenging Standard is thought to be that for PM_{10} , which is to be met by 2013. Council's have developed (or are developing or revising) Straight Line Paths (SLiPs) which describe the estimated PM emission cuts required within an urban airshed to be able to meet the PM_{10} NES fully by 2013. In many airsheds the emission cuts required by the SLiP are very demanding, such that multiple policy options need be considered. In most airsheds primary emissions from residential heating are the dominant source that contributes to NES exceedence. Other sources which are significant in many airsheds, and which are amenable to policy influence, are primary traffic and industrial emissions. There are also several other semi-natural PM emission sources, such as disturbed soil and road dust which are only partially amenable to policy control, plus totally natural sources, such as sea spray (which is a major contributor to PM concentrations in urban New Zealand), volcanic and biological sources.

Another class of PM emissions which are potentially amenable to control are secondary particles arising from anthropogenic emissions of primary precursors. Recent large-scale source apportionment (by receptor modelling) studies conducted by GNS (for Auckland Regional Council) have quantified the amount of 'sulphate' found on PM_{10} and $PM_{2.5}$ filters sampling the air at five sites in Auckland (Davy *et al.*, 2007). However, the average amount of sulphate is not a large fraction of PM_{10} , compared to the fraction attributable to home heating and mobile sources. Furthermore, the receptor modelling cannot apportion the sulphate to natural or anthropogenic sources. If an anthropogenic source can be confirmed, especially a local one, it opens up another potential means of controlling ambient PM_{10} concentrations by reducing emissions of the precursors (in this case most likely to be sulphur dioxide).

The Auckland study provided some evidence (based on triangulated wind directions, see Figure 2.1) that suggested that emissions from activities in the Port of Auckland and the associated shipping lanes might be a locally significant source of secondary

sulphate (Davy *et al.*, 2007). Conventional wisdom would suggest that there is insufficient time for SO₂ to convert to sulphate during air transport over the few kilometres from the source to the receptor. One of the aims of the research which this report introduces is to determine whether atmospheric dispersion modelling may assist in investigating this hypothesis. Thus, the main question will be whether chemical transformation during a longer, indirect route might provide a plausible explanation for sulphate formation leading to its detection in urban Auckland. Specifically, we wish to investigate

1. Can dispersion modelling verify the shipping source?
2. Can it quantify the impact?
3. Can it indicate the potential for mitigation?
4. Can we make inferences about other ports?

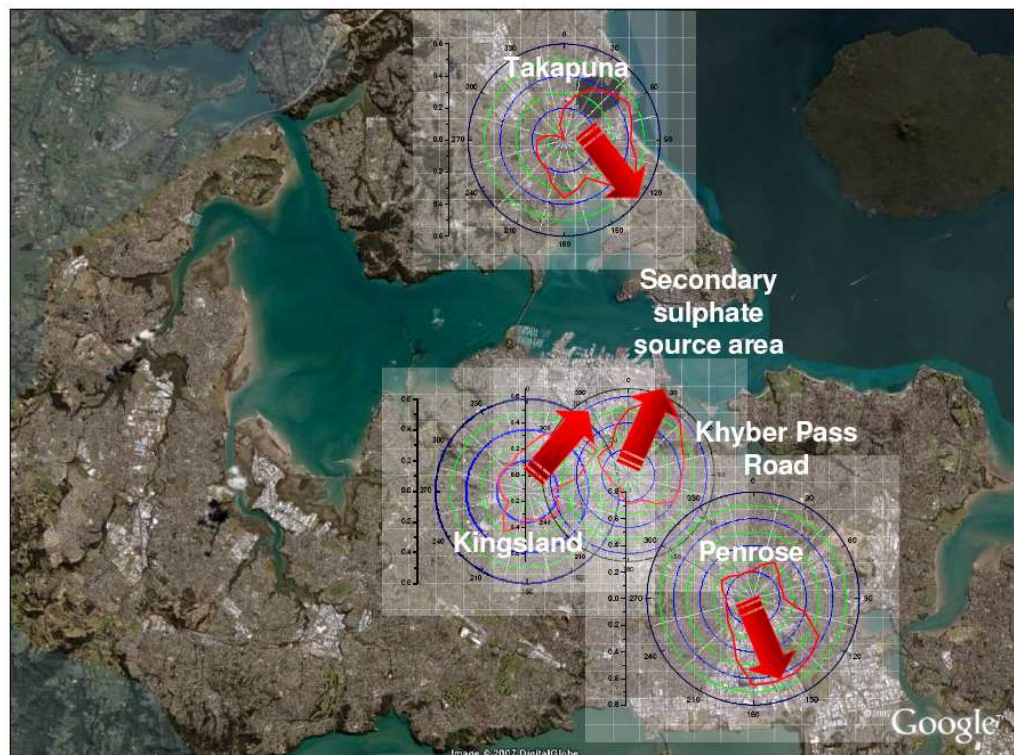


Figure 2.1: Combined secondary sulphate conditional probability function (CPF) analyses indicating source locations. Figure from the Auckland source apportionment report (Davy *et al.*, 2007).

Our aim is to use Auckland as a case-study to determine whether we can develop the tools to investigate the role of local, coastal meteorology on sulphate formation from a shipping source at any New Zealand port-town. In the process, we also expect to gain insights into the implications of coastal meteorology for atmospheric dispersion in New Zealand in general, as well as scope the potential for similar studies of other precursor sources of secondary pollutants.

2.2 The current state of NO₂ compliance

After the NES for PM₁₀, the second most problematic Standard, in terms of the number of people exposed to exceedences, is that for nitrogen dioxide. The NES for NO₂¹ is currently in effect (unlike the PM₁₀ Standard, which must be met by 2013). Regional Councils can (and in many cases have) introduce Regional Targets for NO₂ which may be more stringent than the NES. The MfE ambient air quality guidelines include an additional 24-hour guideline for NO₂ (of 100 µg m⁻³). Monitoring data indicates that the NES for NO₂ is regularly exceeded at two roadside sites in central Auckland. What is not yet established is how representative these sites are for NO₂ i.e.

1. How spatially widespread are the exceedences?
2. How big an area is affected?
3. What are the relative roles of geographical location, local traffic volume (fleet mix and speed), and the precise position of the monitor in determining peak NO₂ and NES compliance?
4. Do exceedences occur outside Auckland or in other locations where NO₂ is not monitored?

The World Health Organisation ambient air quality guidelines include a guideline for annually-averaged NO₂ concentrations. This is not represented in any New Zealand standards, guidelines or targets. Nevertheless, monitoring data has revealed that this guideline is exceeded at a small number of roadside locations in Auckland. The WHO clearly warn that

“If ... nitrogen dioxide is monitored as a marker for the concentrations and risks of the complex combustion-generated pollution mixtures, an annual guideline value lower than 40 µg m⁻³ should be used instead.” (WHO, 2006)

¹ 200 µg m⁻³ as a 1-hour average, with 9 permitted exceedences in a 12-month period

The research that this report introduces seeks to generalise monitoring data to determine the general state of annual NO₂ levels in urban New Zealand, and particularly to understand the roles of location, traffic characteristics and monitor location on these levels.

2.3 Looking to the future – emission trends influencing urban NO₂

Considering the current state of NO₂ standard/guideline/target non-compliance, we aim to scope the efficacy of a range of mitigation options. However, before we can do this we need to review the effects of current trends. The non-linearity and complexity of atmospheric nitrogen chemistry means that emission cuts do not necessarily lead to similar falls in ambient concentrations. It is therefore necessary to verify whether our current understanding of atmospheric chemistry is sufficiently developed and valid to confidently describe the response of the urban New Zealand atmosphere to policy-driven changes. Thus, this strand of the research seeks to

- determine current trends,
- review what monitoring data tells us about local atmospheric chemistry,
- review our ability to model that behaviour,
- scope whether we are yet in a position to confidently develop mitigation options.

The key trends we will be addressing will be

- Recent falls in NO₂ measured at downtown roadside sites in Auckland – will these trends continue?
- Recent steady rises in NO₂ concentrations at two motorway-influenced sites in Auckland, despite falling trends in NO_x concentrations – what is the cause and how general are the trends?
- Rises in the fraction of NO_x emitted as NO₂ reported in Europe – is this, or could this, happen in New Zealand and what would be the consequences?

3. Local secondary PM formation

3.1 What secondary PM is and how it is formed

Secondary particulate matter refers to particles suspended in the atmosphere which have a different physico-chemical composition to those directly emitted from the surface. Conventionally the term is used to describe particles which have arisen through gas to particle conversion or chemical reaction in the atmosphere. Precursor particles may be primary or may themselves be secondary. Intermediates may have very short lifetimes and be effectively impossible to measure.

There are numerous processes and reactions which form secondary particles in the atmosphere. However, for the purpose of this report we are primarily concerned with those that lead to the production of a substantial mass concentration of particulate matter in an urban airshed from gaseous precursors, i.e. those which lead to a net increase in ambient urban PM₁₀ concentrations. A proportion of gaseous precursors will arise from natural emission sources (e.g. VOCs from forestry, SO₂ from volcanoes) and a proportion from anthropogenic sources. Identifying the latter is of value as these sources may be amenable to emission control policies.

The key secondary particulate species of interest to urban air quality management, and some primary precursors, are listed in Table 3.1. Gaseous and semi-volatile organic compounds are oxidised in the atmosphere in multiple stages to form heavier, less volatile and more soluble organic compounds (more likely to exist in the particle phase). Some organic compounds are more potent precursors of secondary organic aerosol (SOA), including monoterpenes and aromatic hydrocarbons such as toluene. The large-scale anthropogenic emission of organic material means that organic species can dominate aerosol chemical and physical properties in urban plumes and on a regional scale, in which SOA generally becomes internally mixed with nitrate and sulphate (Zhang *et al.*, 2007). This complex suite of reactions and the thousands of organic compounds present in the atmosphere means that it is not surprising that our knowledge regarding the quantity and rate of formation of SOA is far from complete.

Table 3.1: The most important secondary particulate species for urban air quality management, and their primary precursors.

Primary precursors	Secondary species
Ammonia	Sulphates
Nitric oxide	Nitrates
Nitrogen dioxide	Organic acids
Nitrous acid	Chlorides
Volatile organic compounds	Fluorides
Sea salt	
Hydrochloric acid	
Hydrofluoric acid	

The rate of conversion of primary gaseous emissions into secondary particles is dependent upon the mutual concentrations of the species which take part in the processes (and the intensity of radiation as a function of wavelength if photochemical processes are involved). Sulphur dioxide emissions usually arise from a point source. Conversion to particulate sulphate usually occurs when the plume mixes with diffuse ammonia or more rapidly when the SO₂ is dissolved in ammonium solution in cloud droplets. The rapidity of this conversion depends upon the rate of dilution of the plume and its trajectory (i.e. its passage through the concentration ‘field’ of ammonia/ammonium). In common scenarios a rate of conversion of 1 % hour⁻¹ is considered typical, or 3 days for a 50 % conversion. Conversion will be accelerated for higher initial concentrations or a longer passage through cloud.

Ammonium sulphates are generally fine mode particles, but sulphates may occur in the coarse mode through two main processes. One is the formation of sodium sulphates through the chloride depletion of sea salt in contact with sulphuric acid. Sulphates (including ammonium, calcium and potassium) may form on the surfaces of coarse mineral dusts, especially where a desert dust plume passes through an industrialised area or interacts with an urban plume.

3.2 The “Conventional Wisdom” of secondary PM

The formation of secondary PM from primary precursors, especially anthropogenic ones, has received limited attention in New Zealand. This is probably associated with the conventional wisdom that nearly all primary emissions will be advected offshore

before enough time has elapsed for any significant physico-chemical transformation to occur. Therefore secondary particles in an urban airshed are not generally considered to be related to primary emissions in that airshed, but more likely emissions from an offshore or highly diffuse and distant source. New Zealand is substantially rural with urban areas representing only 3 % of the land area, yet 17 of the 20 largest urban areas are on the coast. The 5 largest urban areas are all over 300 km from each other (except Hamilton and Auckland, separated by 110 km) so emissions from one are unlikely to strongly affect concentrations in another. Furthermore, the prevailing south-westerly winds present in large parts of the country help strengthen the perception that wind “blows all troubles away”. These prevailing winds do in fact advect air masses towards the north-east, away from New Zealand. However, even if it is true that the predominant wind direction is generally from the south-west, this is not true all the time and there certainly are variations from that “standard” wind direction. South-westerlies are usually associated with stationary high pressure systems over the Tasman Sea. This feature is the most common synoptic pattern in New Zealand in summer but during winter, low pressure systems are more frequent with winds from the north to north-east. Therefore, it is reasonable to think that between two alternating synoptic system (a high and a low pressure systems) there is a considerable amount of low, veering and backing winds such that air mass trajectories are rarely straight, but often looping. This means that there is a mechanism for recirculating and ageing polluted air masses and potentially returning them to their airshed of origin.

3.3 Choice of modelling approach

The complex meteorology of coastal regions such as Auckland can adversely affect the transport and dispersion of air pollutants. Local flows, such as land/sea breeze circulation, limit the ventilation of an air shed by recirculating air pollutants. Numerical modelling may serve as an important tool to assess and predict the transport and dispersion of atmospheric pollutant on local and regional scale (Luhar & Hurley, 2004). Elsewhere in the world considerable research effort has been expended on chemical transport modelling as a means of understanding the spatial and temporal relationships between primary emissions and ambient concentrations of particulate matter or its constituent parts and the process which link them.

Such an approach was considered for our research. However, one must carefully consider how the quality and credibility of such modelling is to be assessed, which is dependent upon the objectives of the study. Accurate modelling is dependent upon accurately capturing a myriad of atmospheric processes (either through direct explicit representation in the model, or through parameterisation or providing assurance that a given process can be neglected without compromising the accuracy of the prediction),

and is equally dependent upon adequate description of the emissions. The amount of detailed emissions information desired is rarely available and this severely limits the applicability of such modelling. A given model simulation may therefore be “accurate” within the limitations of the input data by which it is constrained. Whilst perfectly adequate for research into the fundamental atmospheric processes, these limitations can also be crippling if the objective is accurate forecasts of concentrations of a given substance at a given time and place.

The modelling of sulphate formation from an anthropogenic sulphur dioxide precursor is a relatively simple problem compared to some other chemical transformations in the atmosphere. Yet, it was felt by the research team that even this simple case was too demanding relative to the data available to run the model simulations. Whereas SO₂ emissions could be reasonably estimated (e.g. Luhar & Hurley, 2004) the regional emissions of ammonia, and resulting concentration field could not be specified to any satisfactory degree in order to model the formation of ammonium sulphate (or bisulphate). Local wind flows such as land/sea breezes play an important role in atmospheric aerosol dispersion (e.g. Celia *et al.*, 1999, Norman *et al.*, 2006, Song *et al.*, 2008). Furthermore, the role of cloud condensation nuclei, primary and secondary organics and the particle size distribution of each could not be specified. Previous studies employing explicit chemical transport modelling in simulations of the Christchurch airshed have employed a sensitivity approach to ‘tune’ such input parameters using ground-level PM observations. It was felt that this approach was exceedingly intensive and introduced as many new uncertainties as it removed. As our intention was not to delve into the physico-chemical processes, but to reproduce actual site-specific concentrations then an alternative, indirect approach was considered more suitable.

3.4 Method for modelling the Auckland Region

Concentrating on meteorological modelling alone removes many of the uncertainties and reliance on highly uncertain emission estimates associated with chemical transport modelling. It is well established that meteorology is the major determinant of temporal and spatial variability in urban air pollution. Our approach was to use advanced airshed modelling to investigate the characteristics of local and regional airflow during periods associated with elevated levels of particulate pollution, focussing specifically on episodes of elevated secondary PM. This research coincided with the reporting of results from the Auckland source apportionment study which reported estimated levels of sulphate at 5 sites across the city. As well as being able to exploit that resource, we

chose to model the Auckland airshed due to its high level of anthropogenic emissions and coastal geography (allowing us to test the assumption that “it all blows out to sea” and the role of land/sea breezes in re-circulating polluted airmasses). Other potential benefits of modelling Auckland included the wealth of meteorological and air quality datasets, the opportunity to study the impacts of its complex topography, studying the Port and shipping lanes as a major and localised SO₂ source and the high population potentially affected.

The main questions we wish to address across Parts One and Two are:

- Can airshed modelling assist in increasing confidence in the link between sulphate loadings observed on PM filters in Auckland and a presumed source of shipping emissions in the Port of Auckland and the shipping lanes of the Hauraki Gulf (as discussed in Davy *et al.*, 2007)?
- Can airshed modelling determine whether offshore recirculation provides sufficient source-receptor atmospheric residence time to permit a rate of SO₂–sulphate conversion sufficient to lead to a significant enhancement of PM₁₀ as measured in Auckland?
- Can airshed modelling extrapolate these findings to assist in determining the probability of the shipping source leading to significant enhancements of onshore PM₁₀ in other ports in NZ?

The tool used for all modelling simulations described below was WRF, the Weather and Research Forecasting Model (www.wrf-model.org) modelling system version 3, a next-generation mesoscale numerical weather prediction system designed to serve both operational forecasting and atmospheric research needs. Further details of the model and its configuration for this study are described in Appendix One.

Since our primary concern is to identify the meteorological processes determining whether pollutants are advected entirely to the ocean or whether there is potential for them to be transported back to land, the modelling exercise in this Part One report is comprised of a single case-study and aims to characterise the local air flow associated with a synoptic weather conditions when air pollutant levels tend to be high. Further situations will be analysed in the following Part Two report. Two days (48 hours) in the month of March 2006 were selected to model the local meteorology of the Auckland region. The selection of the modelling period was based on the Auckland’s source apportionment data (Davy *et al.*, 2007) that suggests higher than average sulphate loadings at several monitoring sites during mid to late March 2006. The

synoptic weather conditions (Figure 8.1) suggests that there was a high pressure system over the Auckland region at this time, with low synoptic winds and sunny days. These meteorological conditions are considered favourable for sea/land breezes to occur at this time of the year (Hessell, 1988). Previous studies (e.g. McGill, 1987, Revell, 1984) have shown that in Auckland sea breezes occur from 17 to 40 % of days over the five-month period from November to March.

3.5 Meteorological modelling results for Auckland

According to Auckland's source apportionment data (Davy *et al.*, 2007) higher than average sulphate loadings were observed at several monitoring sites during mid to late March. In this Part One report, we focus on a single example case study aimed at characterising the air flow associated with this period. Further situations will be analysed in the following Part Two report. An example of the output from the modelling is shown in Figure 3.1 below. Additional output (including Figures 8.1 – 8.6) is provided in Appendix 1.

Sea/land breezes are common in Auckland region during summer on warm and sunny days under weak synoptic winds (Bell & Fisher, 1995; Haigh, 1992). In addition to other factors, the strength and depth of the sea/land breezes predominantly depends on the strength of the synoptic pressure gradient. In the third week of March 2006, New Zealand in general and Auckland in particular was at the centre of a high pressure system while a low pressure system was to the S/SW of NZ (Fig 8.1). At the surface level, however, under weak W/SW synoptic winds, land breeze generated at both east and west coast of the Auckland region (Figure 8.4), although the land breeze flow is also modified due to orographic disturbances. Such conditions are typical for coastal areas and the modelling results show an extremely complicated wind regime.

During the mid morning (11:00 LST) under weak synoptic winds, the model predicts that sea breezes were generated at both east and west coast of Auckland (Figure 8.5) and were gradually penetrating inland (Figure 8.5), while in the afternoon (14:00 LST) mature sea breezes developed about Auckland (Figure 8.6). The modelled inland penetration of sea breezes from both east and west of Auckland is up to 50%. Figure 8.6 shows the easterly and westerly sea breeze flow meeting in the middle of the land mass to form a sea breeze convergence zone (SBCZ). The location of the SBCZ depends on the strength and direction of synoptic winds. The synoptic pressure gradient was weak during the simulation period. In the mid morning when the sea breeze was weak at the east coast, the SBCZ was formed close to the east coast.

However, as the day proceeded and the sea breeze strengthened the SBCZ moved westwards to the centre of the land mass (Figure 8.6).

In general Sea/Land breezes are not considered good pollutant ventilators for three reasons:

1. the speed is usually low (less than 7 m s^{-1}),
2. they are closed circulation systems, and
3. they exhibit a diurnal reversal in direction of flow, that is the polluted air aloft drifts seawards, at a short distance from the coast it may be partly caught in the downward current and returned landwards by the sea breeze (Forsdyke, 1997).

Another important consideration that is usually ignored is that sea-breezes can restrict the vertical distribution of pollutants by collapsing the daytime mixing-layer.

The convergence line plays an important role in the horizontal and vertical distribution of pollutants. Convergence zones may inhibit horizontal pollutant dispersion while pollutants that are advected to SBCZ and carried aloft and then to the sea, may come onshore due to recirculation.

Forward trajectory analysis was conducted to understand the path and the time an air parcel takes to reach a particular point from the point of its origin. The air mass trajectories were plotted for two different points of origin a) 48hour trajectory plot originating in the port of Auckland (Figure 3.1), and b) 48 hour trajectory plot originating near the shipping lane north of Rangitoto Island (Figure 8.3). Trajectories from both points started at mid night. They show that small changes in point of origin lead to diverging trajectories. However, after an initial westerly track, several of the trajectories return to land in under 24 hours. Furthermore, several of the trajectories return to central Auckland (Figure 3.1).

The generally circular trajectories illustrated are caused by land and sea breezes superimposed over a weak southerly flow. The nocturnal south-westerly land breeze initiated the advection of the air masses out into the Gulf, but the 11am and 4 pm simulations (Figure 8.4) shows a mature sea breeze over North Shore and the Hibiscus Coast drawing the subject air masses back inland. Between the coasts the model indicates that the sea breeze convergence zone acts to divert trajectories back towards Auckland.

Simulation Testbed
 Fcst: 49 h
 Valid: 13 UTC Sat 18 Mar 06 (01 LST Sun 19 Mar 06)
 Trajectories from hour 48.000 to 96.000
 Terrain height AMSL

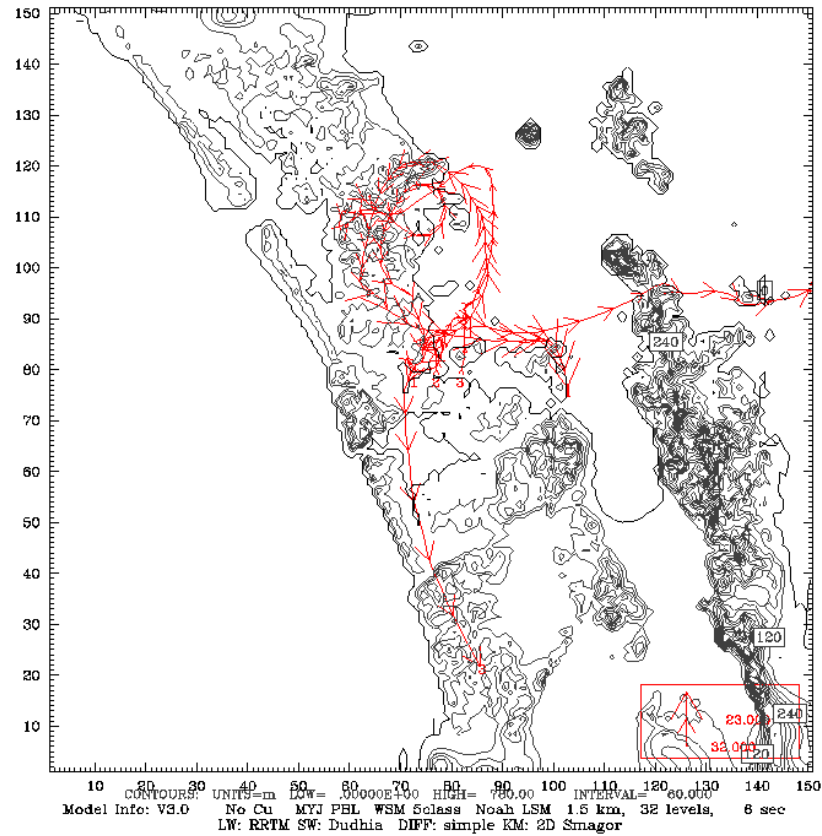


Figure 3.1: 48 hour trajectory Plot of wind field (12 metres above ground) originating at Port of Auckland (00:00 hours 19th Mar 2006 to 00:00 hours 21th Mar 2006).

4. Where and when do urban nitrogen dioxide concentrations peak – monitoring data

4.1 NO₂ Standards, Guidelines and Targets

4.1.1 World Health Organisation guidelines

The World Health Organisation's 2005 Global Update of the Ambient Air Quality Guidelines (WHO, 2006) considered new evidence on the effects of nitrogen dioxide published since the organisation set guidelines of 200 $\mu\text{g m}^{-3}$ as a 1 hour average and an annual mean of 40 $\mu\text{g m}^{-3}$ in 2000. The Update found that no change to these guidelines was justified. However, the Update noted that ...

“Numerous epidemiological studies have used nitrogen dioxide as a marker for the air pollution mixture of combustion-related pollutants, in particular traffic exhaust or indoor combustion sources. In these studies, the observed health effects might also have been associated with other combustion products, such as ultrafine particles, nitric oxide, PM or benzene.”

“If ... nitrogen dioxide is monitored as a marker for the concentrations and risks of the complex combustion-generated pollution mixtures, an annual guideline value lower than 40 $\mu\text{g m}^{-3}$ should be used instead.”

4.1.2 The MfE Ambient Air Quality Guidelines and the National Environmental Standards

The WHO 1-hour guideline (200 $\mu\text{g m}^{-3}$) was adopted as one of the Ambient Air Quality Guidelines for New Zealand by the Ministry for the Environment in 2002 (reduced from 300 $\mu\text{g m}^{-3}$ in the 1994 guidelines). Additionally a 24-hour average guideline of 100 $\mu\text{g m}^{-3}$ was also adopted in 1994. The WHO annual guideline was not adopted in New Zealand.

Only the 1-hour guideline was incorporated into the National Environmental Standards (NES). The NES for nitrogen dioxide came into force from 1st September 2005. It adopts the WHO/MfE value of 200 $\mu\text{g m}^{-3}$ averaged over 1 hour. The Standard permits this concentration to be exceeded 9 times in a 12 month period before a breach has occurred.

4.1.3 Regional Targets

Auckland Regional Council has adopted Regional Air Quality Targets (RAQT). The targets for NO₂, which apply in ‘Urban and Rural areas (i.e. not Industrial areas) are equivalent to 66% of the MfE guideline values, i.e. 132 µg m⁻³ as a 1 hour average and 66 µg m⁻³ as a 24 hour average. The same values have been adopted by Environment Waikato, Otago Regional Council and Environment Canterbury.

4.2 Sources of NO₂ data

4.2.1 Auckland Regional Council

NO_x has been monitored within the Auckland Region since at least 1982. Since 1987 NO_x has been monitored at 16 locations. Long-term data (> 3 years) has been gathered at 8 sites, as summarised in Table 4.1.

Table 4.1: List of ARC monitoring sites that have recorded > 3 years of NO₂ data.

Site	Type ²	From	to
Penrose II	Industrial/motorway/urban	1989	Still operating
Mt Eden II	Urban background	1991	2006
Khyber Pass Road	Roadside/urban	1998	Still operating
Musick Point II	Urban periphery	1999	Still operating
Takapuna	Motorway/urban	2001	Still operating
Henderson	Roadside/suburban	2003	Still operating
Kingsland	Urban background/weak roadside influence	2004	2007
Queen Street II	Street canyon/downtown	2004	Still operating

Note: At the time of writing only provisional data was available for 2007. Therefore the following analysis considers data up to and including 2006.

² NIWA, not ARC interpretation

4.2.2 Greater Wellington Regional Council

NO₂ has been measured continuously in Greater Wellington since 2001 (at Upper Hutt) and in central Wellington (Corner V) since March 2005. By the end of 2007 there were 8 stations measuring NO₂ and NO_x in the region: 3 ‘transport’ sites (Corner V, Ngauranga and Melling). Other sites are at Upper Hutt (Savage Park), Lower Hutt (Birch Lane), Wairarapa College in Masterton, Tawa and Karori Park in the outer western suburbs of Wellington.

4.2.3 Environment Canterbury

NO₂ has been measured continuously in St Albans, Christchurch since 1988. A site was established in St Albans (Packer Street) in May 1988 until late 2002. A similar site was established a kilometer away in August 1998, with an overlap of a few years to enable comparisons of concentrations at both St Albans sites to be made. A new monitoring site, including NO₂, was established in Burnside in 2003. In Timaru some NO₂ monitoring was carried out at Grey Road from July to December 2005. All of these sites are urban background sites, i.e. they are influenced by diffuse traffic sources but not by any single identifiable road. Validated data was only available for this report up to the end of 2005, and we do not, therefore, include recent data from ECan’s roadside site which began operation in 2006.

4.3 NO₂ Standard and Target exceedences

4.3.1 Exceedences in Auckland Region

Table 4.2: Summary of exceedences of the NO₂ NES threshold in Auckland Region (1997 – 2007).

Monitoring site	NES threshold exceedence (i.e. 1 hr NO ₂ > 200 µg m ⁻³)
Khyber Pass Road	Numerous exceedences every year, except 2006 and 2007
Queen Street II	exceedences every year
Penrose	exceedence in all 3 years monitoring was conducted (1987-9)
Penrose II	exceedence in 2001 and 2005 only
Dominion Road I	exceedence in 1994 only (site closed in 1996)

The downward trend in annual mean NO₂ at Queen Street and Khyber Pass Road does not necessarily translate into a downward trend in NES exceedence. Exceedences of the NES threshold of 200 µg m⁻³ of NO₂ have only been observed at 5 ARC monitoring locations, as detailed in Table 4.2. 9 exceedences are permitted in a 12-month period before the NES is considered to have been breached. Since 2004 breaches have only been reported at Khyber Pass Road (2004 and 2005) and Queen Street II (2007 - provisional). Those sites where the NES threshold has not been exceeded have not yet experienced exceedences of the RAQT (132 µg m⁻³) either.

All of the sites where hourly NO₂ concentrations above 200 µg m⁻³ have been observed (as listed in Table 4.2) are sites strongly influenced by busy roads. The Takapuna site is also influenced by the Auckland Northern Motorway. However, the site is 60 m from the motorway and this appears to be sufficient distance to prevent hourly NO₂ from reaching the RAQT value of 132 µg m⁻³. The Henderson (Lincoln Rd I) site is 10 m from Lincoln Road, a major thoroughfare in Henderson. However, the traffic volume on this road (and perhaps also the speed) is sufficiently low to prevent hourly NO₂ exceeding 100 µg m⁻³.

Whether the observed increasing trend at Penrose and Takapuna may lead to future RAQT exceedences, or whether exceedences are already occurring closer to the motorways than these monitors (60 m and 106 m respectively) is one of the key

questions that has motivated this research, and will be addressed in the following sections and Part 2.

As described further below, emissions of NO_x and NO₂ are evolving rapidly. For this reason the following detailed analysis focuses on data from the years 2004 – 2006 inclusive only.

At the start of 2004 there were 6 operational permanent NO_x monitoring sites (Musick Point II, Khyber Pass Road, Penrose II, Mt Eden II, Takapuna and Henderson). By the end of 2006 one had shut down (Mt Eden II) and three new sites had begun operation (Kingsland, Glen Eden and Pukekohe Mobile). Plus, a NO_x monitor had been added to two existing sites (Queen Street II and Pukekohe), so that in total there were 10 operational NO_x monitors on 31st December 2007. Only 4 and 3 months of data had been collected at Pukekohe and Pukekohe Mobile respectively by the end of 2006 and so these sites do not feature as much as the others in this analysis.

Over the period 2004 to 2006 inclusive, the RAQT has been exceeded at Queen Street II 136 times. The median hourly NO_x concentration measured during hours when the RAQT was exceeded was 635 µg m⁻³, although RAQT exceedence has occurred when NO_x was as low as 354 µg m⁻³. The NES was exceeded 6 times with a corresponding median NO_x concentration during exceedences of 796 µg m⁻³. The lowest NO_x concentration during an NES exceedence was 541 µg m⁻³.

Over the period 2004 to 2006 inclusive, the RAQT has been exceeded at Khyber Pass Road 1017 times. The median NO_x concentration measured during hours when the RAQT was exceeded was 906 µg m⁻³, although RAQT exceedence has occurred when NO_x was as low as 370 µg m⁻³. The NES was exceeded 30 times with a corresponding median NO_x concentration of 1195 µg m⁻³. The lowest NO_x concentration during an NES exceedence was 785 µg m⁻³.

These results indicate that although NES and RAQT exceedences have been more common at Khyber Pass Road (where NO_x concentrations are higher – see chapter 5) than at Queen Street II, the NO_x concentration required to trigger an NES or RAQT exceedence for NO₂ is approximately 30 % lower at Queen Street compared to Khyber Pass Road. The possible reasons for this will be discussed further in this report.

4.3.2 Exceedences in Greater Wellington Region

No exceedence of the NES for NO₂ has ever been recorded in Greater Wellington. The equivalent value to the Auckland RAQT (i.e. 132 µg m⁻³) has been exceeded only once at Corner V (central Wellington) in each of the years 2004 – 6 inclusive.

4.3.3 Exceedences in Canterbury Region

Long-term trends in peak (maximum 1 hr concentration) NO₂ in Christchurch are shown in Figure 4.1. Although measurements began in St Albans in 1988, there is limited data regarding the instruments used and the maintenance and calibrations performed before 1994. ECan’s 2005 Monitoring Report states that “Data from this period are reported for comparison to concentrations measured in later years, but care should be taken in drawing conclusions from them.” For this reason data from this period is not presented in Figure 4.1.

Trends in this peak hourly NO₂ are expected to be more prone to random variation. Nevertheless considerable consistency can be seen in the period 1997 – 2003. Although there have been an hour or more over 200 µg m⁻³ in the early to mid 1990s in St Albans, there have been no breaches of the NES observed in Canterbury.

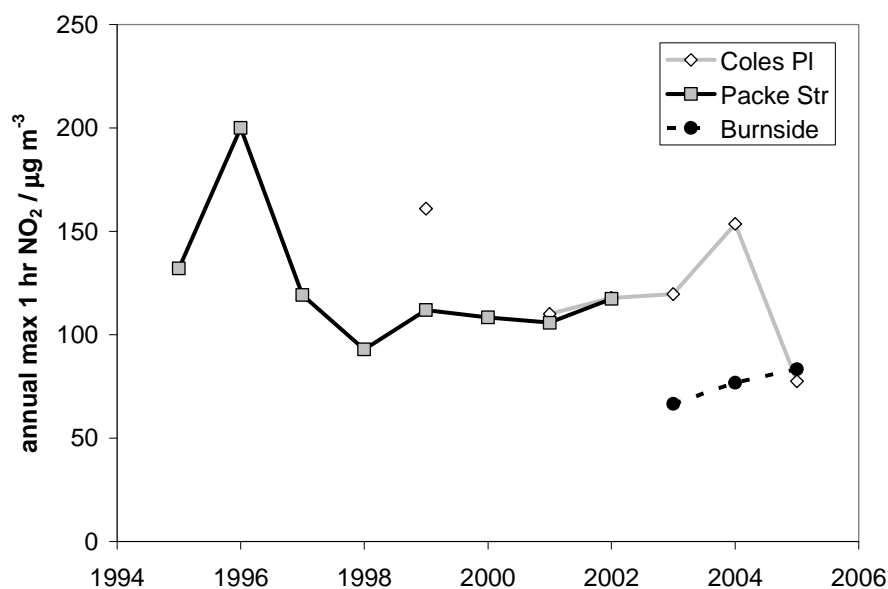


Figure 4.1: Annual maximum 1 hour NO₂ at three monitoring sites in Christchurch (restricted to sites reporting > 50 % data coverage per year).

4.3.4 Relating continuous and long-term NO₂ data

Figure 4.2 plots the maximum observed NO₂ concentration in a given year versus the annual mean NO₂ for that year, for a range of Auckland monitoring sites. This shows that in most cases the data points tend to fall within the bottom-left-hand box, which describes compliance with both the NES and WHO annual guideline. The points on the right hand side of the figure, which exceed both the NES and WHO guideline all relate to Queen Street II and Khyber Pass Road, and show that years in which the NES was exceeded, so was the WHO guideline. The remaining points describe NES exceedence without WHO exceedence. These points originate at the Penrose, Penrose II and Dominion Road sites. As a first approximation, we conclude that a site which is WHO-compliant (i.e. annual mean NO₂ < 40 µg m⁻³) is more likely than not to be NES-compliant.

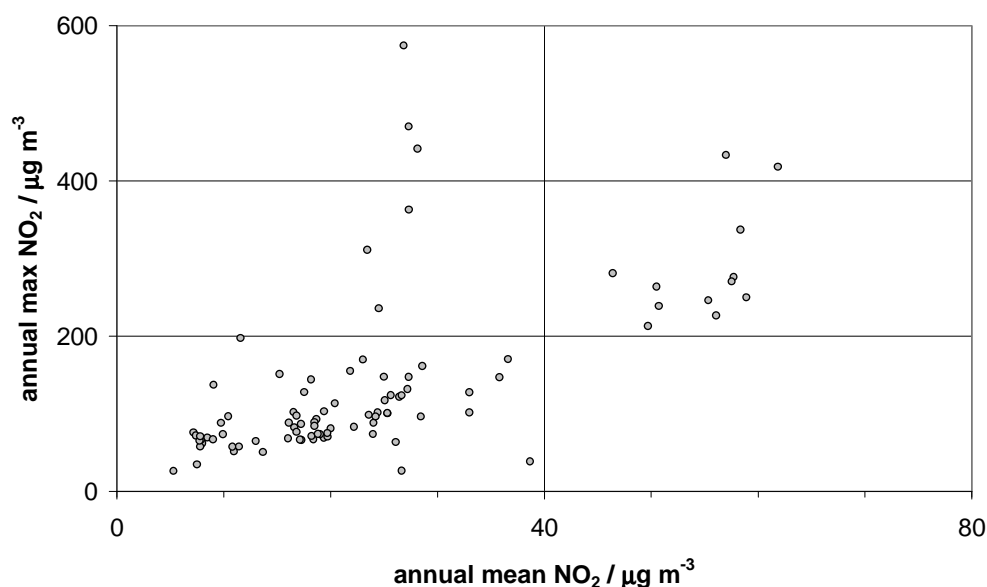


Figure 4.2: Annual maximum NO₂ versus annual mean NO₂ for a wide range of Auckland monitoring sites, 1989 – 2006. The gridlines are in multiples of the NES (y-axis) and WHO annual guideline (y-axis).

4.4 Spatial distribution of urban NO₂

4.4.1 ARC passive sampling

Auckland Regional Council has commissioned several NO₂ diffusion tube surveys since 1994. Full details have recently been published (ARC, 2007). The most spatially comprehensive survey was that of winter 2006.

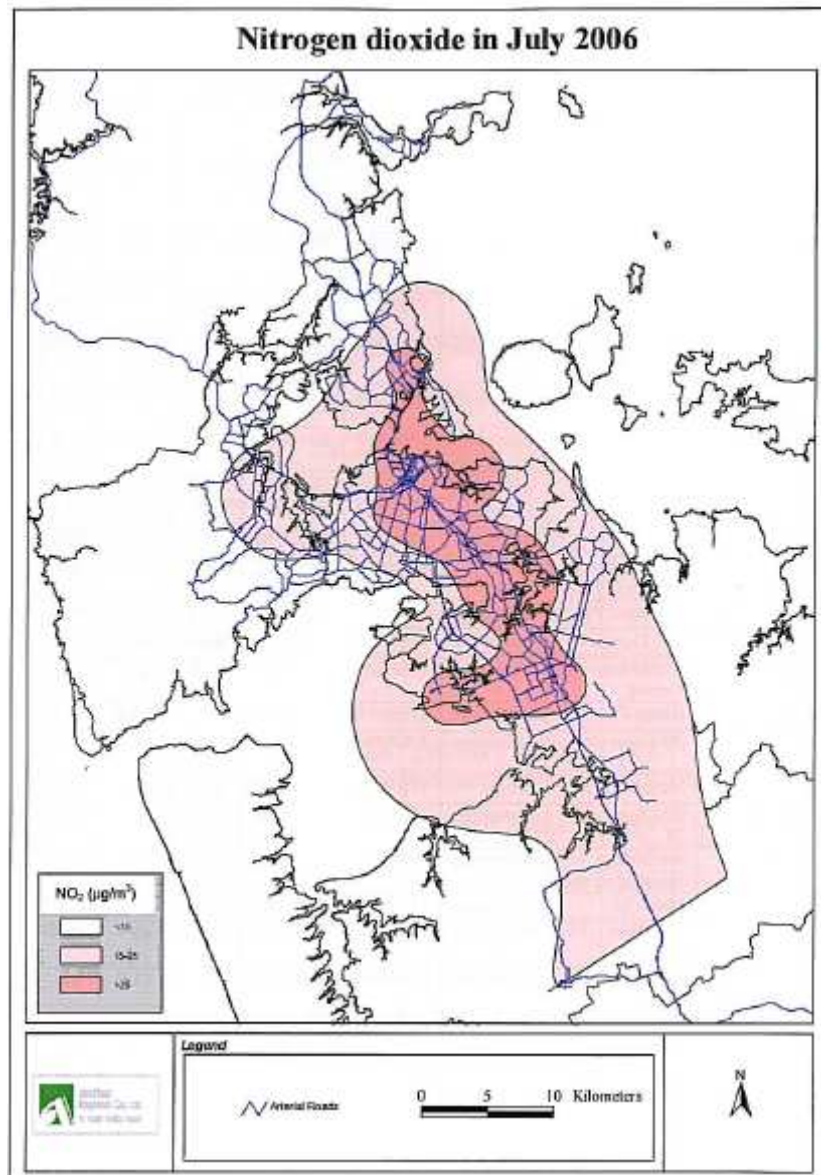


Figure 4.3: Spatial distribution of NO₂ concentrations in July 2006, derived from ARC diffusion tube data (from ARC, 2007).

The survey revealed (Figure 4.3 reproduces one of the figures from that report which is indicative of the main findings) three or four areas of raised NO₂ concentrations, centred on

1. central Auckland,
2. the Mt Wellington/Otahuhu/Otara areas (which are partly industrial but also affected by the Southern Motorway – see below)
3. central Manukau City (also industrial and motorway),
4. Auckland Airport (this peak appears to be weaker than the others).

The winter 2006 survey was conducted over the months July – September. NO₂ concentrations in Auckland have a distinctive and persistent seasonal cycle with concentrations peaking in the winter. For this reason, the mean concentrations recorded over this survey are expected to represent over-estimates of the annual mean. In this survey 8 sites reported mean concentrations (over the three months) above the WHO **annual** mean guideline of 40 µg m⁻³. 7 of these 8 sites were in downtown locations. We estimate that when extrapolated to true annual means only one of the ARC tubes will have exceeded the WHO guideline (Broadway). However, the WHO note that a lower guideline (which it cannot yet specify) should be adopted in traffic-exposure environments, and so the value of 40 should not be used as a ‘safe threshold’.

4.4.2 NO₂ near the Auckland motorways

Auckland has four urban motorways. The Southern and Northern Motorways both form part of State Highway 1. These two motorways and the North-Western Motorway (SH16) converge at the Central Motorway Junction (CMJ) in central Auckland. The South-Western Motorway (SH20) is (currently) not connected to the rest of the system. The Northern and Southern Motorways carry the most traffic, with annual average daily traffic of >100,000 between Tristram Ave and East Tamaki Road (a length of ~ 27 km) and > 150,000 over the central 9 km section. The AADT on the North-Western Motorway is 90 – 110,000 within the Auckland isthmus and < 90,000 in Waitakere City. The AADT on the South-Western Motorway is below 70,000 along most of its length, peaking at 90,000 on the Mangere Harbour Bridge.

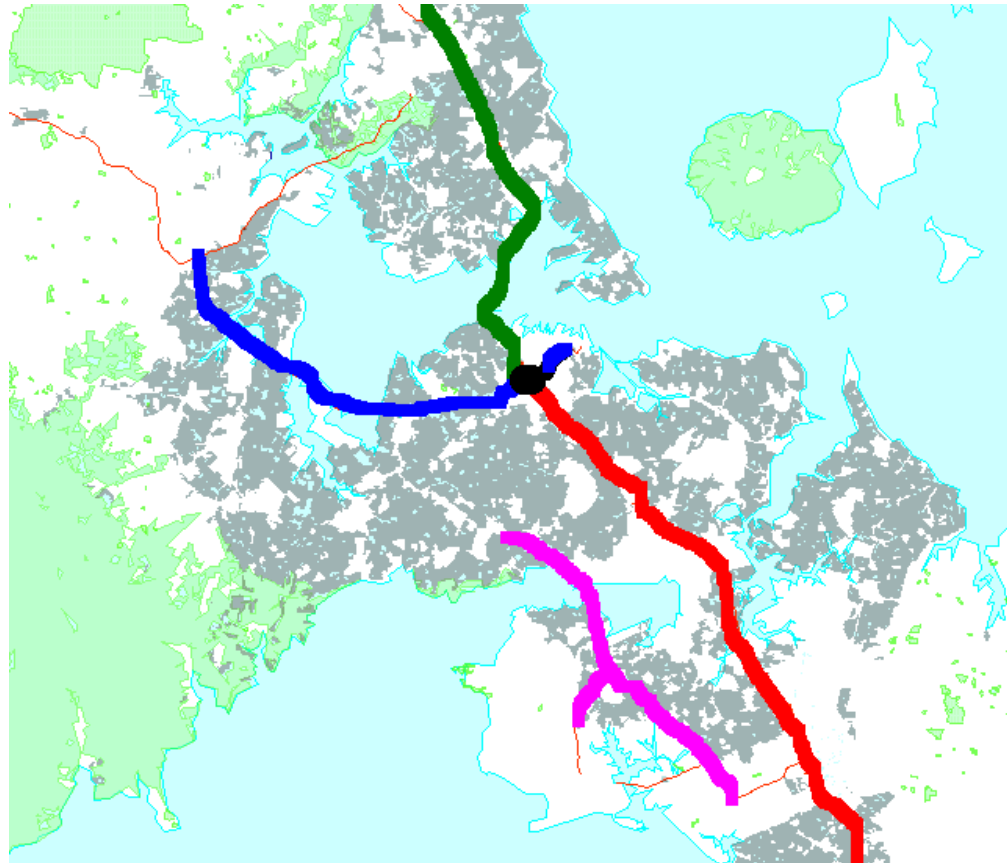


Figure 4.4: The Auckland motorway system. Red = Southern Motorway, Green = Northern Motorway (both part of SH1), Blue = North-Western Motorway (part of SH16), Pink = South-Western Motorway (part of SH20). Black = Central Motorway Junction. Grey = residential areas.

The ARC winter 2006 diffusion tube survey data (Figure 4.3) shows a strong influence of the Southern Motorway on the spatial pattern of NO_2 in Auckland. The influence of the Northern Motorway is not as clear, but the far fewer measurement sites in North Shore might be a significant factor. There is no clear influence of the South-Western or North-Western Motorways.

Transit NZ commissioned a diffusion tube survey of key points on the State Highway network which commenced in January 2007 (Watercare, 2008). 23 tubes were deployed in Auckland, 4 along the Northern Motorway, one at the Central Motorway Junction, 10 on the Southern Motorway, two on the South-Western Motorway and 4 on the North-Western Motorway. Two additional sites were located close to future extensions of the South-Western Motorway. After one year only one site – that at Canada Street, adjacent to the CMJ - reported an exceedence of the WHO annual guideline ($41.9 \mu\text{g m}^{-3}$). Three more sites exceeded $30 \mu\text{g m}^{-3}$, (Gavin Street, Mt Hobson Rd, Niger Street) all of which were on the north or east side of the motorways

close to downtown Auckland. All other SH1 sites reported concentrations between 20 and 30 $\mu\text{g m}^{-3}$.

Auckland wind roses show that the predominant wind direction across most of the region is south-westerly. Based on this observation we classed sites as ‘downwind’ if they were to the north or east of the motorway, and ‘upwind’ if they were to the south or west. The effect of wind direction is clearly suggested in the comparison of two ARC sites: St Peters College and Auckland Grammar School (Figure 4.5). These two schools are located on opposite sides of the most heavily trafficked section of the Auckland motorway system, with ~ 200,000 vehicles per day. St Peters College is on the north-east (i.e. ‘upwind’) side.

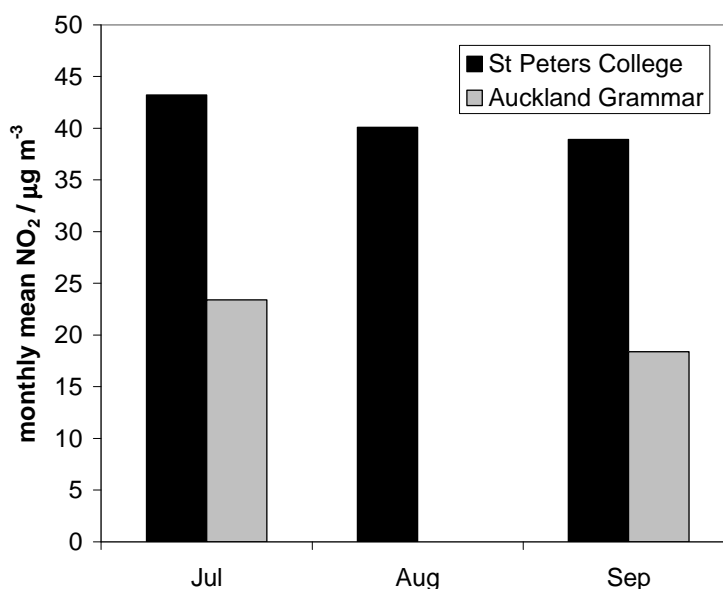


Figure 4.5: Monthly mean NO₂ observed in 2006 (ARC, 2007) at two sites on opposite sides of the busiest section of motorway in New Zealand (SH1 at Newmarket, Auckland). No results were available for August at Auckland Grammar.

When the Transit data are plotted against traffic levels a clear association is revealed for the downwind sites only (Figure 4.6). This figure only includes upwind sites alongside SH1. All of these reported values are above 20 $\mu\text{g m}^{-3}$. Figure 4.7 shows the remaining observations from sites upwind of the South-Western and North-Western Motorways (which are less central than SH1) and non-motorway sites. All of these values are 20 $\mu\text{g m}^{-3}$ or below.

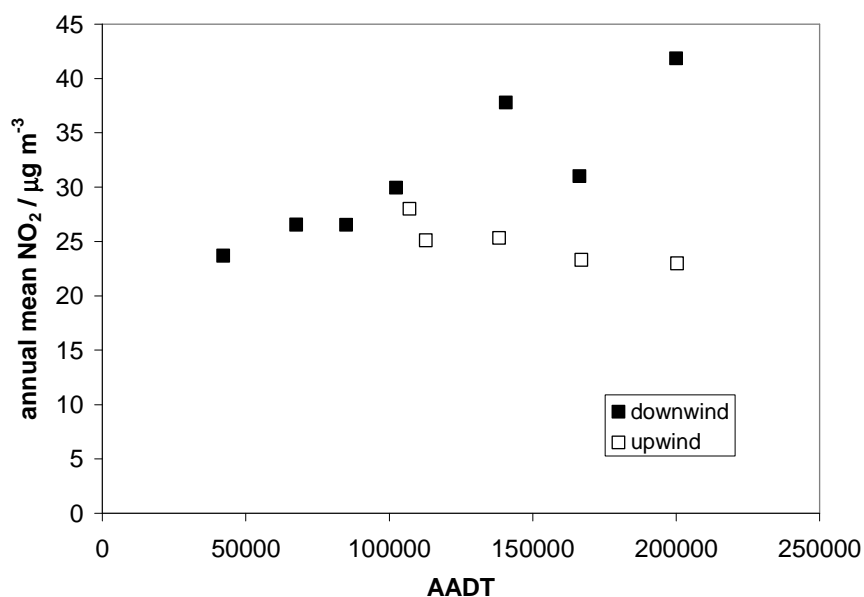


Figure 4.6: Annual mean NO₂ reported by diffusion tubes alongside Auckland motorways, 2007, against annual average daily traffic for the adjacent section (2006). Sites upwind of SH16 and SH20 have been omitted for clarity (they are shown in Figure 4.7).

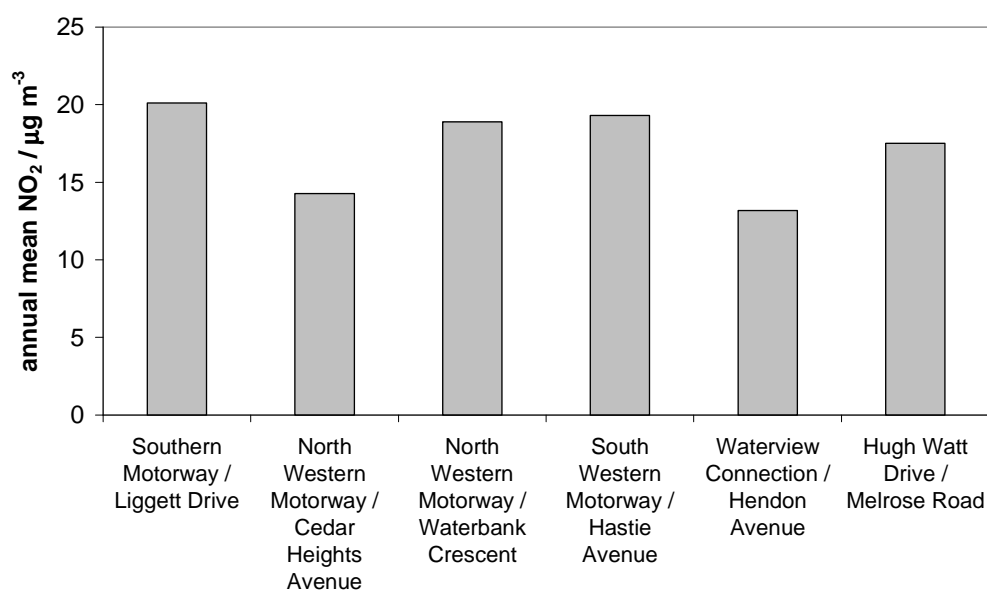


Figure 4.7: Annual mean NO₂ (2007) from Transit sites upwind of motorways, or not adjacent to motorways.

An additional 2 diffusion tubes were added to Transit NZ's network alongside SH1 from September 2007 in the vicinity of the future Victoria Park Tunnel, where AADT

is approx. 138,000. In our classification one site (Gaunt Street) is downwind and one (Hepburn Street) is upwind. In the first four months of operation concentrations (when extrapolated to an estimated annual mean) are lower than might be expected following the pattern illustrated in Figure 4.6. However, firm conclusions cannot be drawn until a year of data are collected (the assumption of upwind and downwind locations may not be valid for a restricted period of time). Also, these sites may be further than average from the motorway (the effect of distance from the road is treated in more detail in section 4.5 and Part 2).

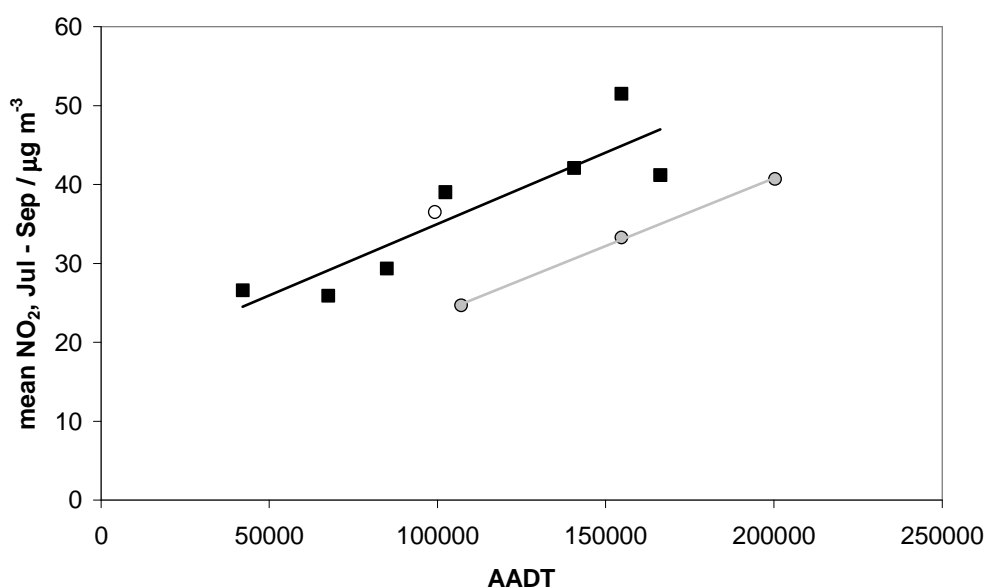


Figure 4.8: Mean NO₂ observed over July to September inclusive at sites ‘downwind’ of Auckland motorways as a function of annual average daily traffic on the adjacent link. Black squares = Transit NZ data (2007). Grey circles = ARC data (2006), open circle = Clemow Drive (see text).

Figure 4.8 shows a comparison of some of the observations made at motorway sites by ARC and Transit NZ. We have restricted this comparison to those months of the year when data is available from both sources (July – September, albeit in different years). It can clearly be seen that the ARC data, although limited to three data points, follows the same relationship with AADT, but with a lower offset. Although this could be due to a systematic error associated with the diffusion tubes, it may also be explained by differences in weather or by different distance of the tubes from the motorway. One site is plotted separately, as an open circle. This is data from an ARC tube located on an off-ramp of the Southern Motorway in Mt Wellington, and is thus much closer to

the traffic source than the other ARC sites. This point fits more closely to the pattern of the Transit data. This will be investigated further in Part 2.

4.5 Role of monitor distance from road

Both the ARC and Transit diffusion tube studies included investigations of the role of distance of the tube from a motorway. ARC conducted surveys at various distances downwind of the Southern Motorway at Otahuhu in 1997/8, Penrose in winter 2006, and across 12 locations both upwind and downwind of the South-Western Motorway at Mangere in winter 2006 (ARC, 2007). At Mangere concentrations downwind were in excess of upwind concentrations by up to $4 - 19 \mu\text{g m}^{-3}$ over the range 500 – 50 m. Results for August and September were more consistent than for July. During these months we have found that the downwind concentrations exceeded the upwind concentrations (average of concentrations at distances of 150, 300 and 500 m upwind) by a factor approximated by $1.8 - x$, where x is the distance downwind in kilometres. Whether or not this relationship is constant, or varies seasonally is dependent upon the degree to which the same processes are controlling both the formation and dilution of NO_2 both upwind and downwind of the road. This may be considered further in Part 2.

The ARC report fitted a logarithmic profile to the reduction in NO_2 downwind of SH1 at Otahuhu in a response to a much steeper spatial gradient close to this busier motorway (ARC, 2007). We have insufficient data to add any further insight to this dataset.

The Transit NZ report (Watercare, 2008) also included a similar study, with 5 tubes (of which 3 were co-located) located at 5, 100 and 130 m from the Southern Motorway at Penrose covering the whole of 2007. Concentrations alongside the motorway were higher than those at 100 m by $0 - 20 \mu\text{g m}^{-3}$, or $10.7 \mu\text{g m}^{-3}$ on average.

5. Trends influencing urban nitrogen dioxide

5.1 NO_x trends

Urban concentrations of NO₂ generally arise due to the oxidation of urban emissions of NO, with additional primary emissions of NO₂. Figure 5.1 below shows the annual mean concentrations of NO_x at the same sites discussed above. Decreasing trends are evident at all sites, as described in Table 5.1.

Table 5.1: linear trend in annual mean NO_x concentrations for long-term ARC monitoring sites to 2006 inclusive, given to nearest integer.

Site	Type	Trend / $\mu\text{g m}^{-3} \text{ year}^{-1}$
Mt Eden	Urban background	-1
Henderson	Roadside	-3
Penrose	Motorway (106 m) / Industrial	-3
Takapuna	Motorway (60 m)	-8
Khyber Pass Road	Roadside	-7
Queen Street	Street canyon	-26

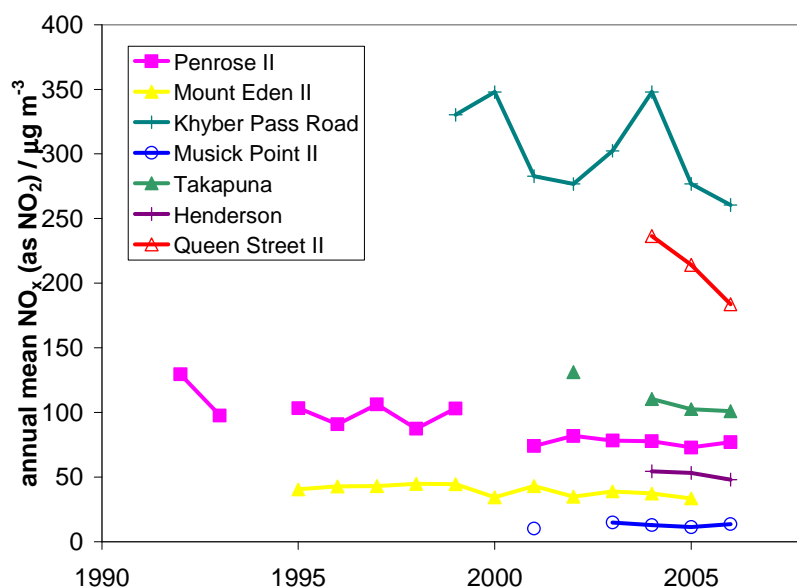


Figure 5.1. Annual mean NO_x at a range of monitoring sites in Auckland Region (restricted to sites reporting > 80 % data coverage per year for at least 2 years).

5.2 NO₂ trends

Figure 5.2 shows the long-term trend in annual mean NO₂ in Auckland (showing only data for years with > 80 % data coverage, and stations with 2 or more years of qualifying data). Queen Street II and Khyber Pass Road clearly report much higher concentrations than elsewhere, although both appear to be experiencing downward trends (which appears to be confirmed by the provisional 2007 data, not shown). A striking trend amongst the other sites is the divergence between Mt Eden (representative of urban background) and Penrose II, 106 m from the Southern Motorway, with a long-term decreasing trend in the former, but an increasing trend at the latter. This increasing trend is weakly confirmed by the other motorway-influenced site, Takapuna.

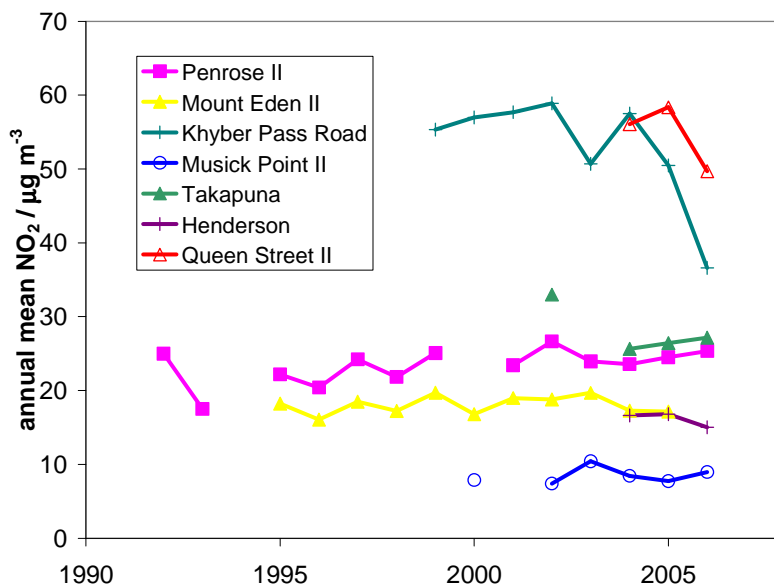


Figure 5.2. Annual mean NO₂ at a range of monitoring sites in Auckland Region (restricted to sites reporting > 80 % data coverage per year for at least 2 years).

This raises questions about NO₂ concentrations near motorways and major roads. The Penrose II data, in particular, show that NO₂ concentrations are consistently rising despite falls in NO_x concentrations. The long-term trend in the relationship between NO_x and NO₂ at Khyber Pass Road is more variable and difficult to interpret and the Queen Street dataset is currently too short to make any confident comments about long-term trends.

Figure 5.3 below shows similar data for Christchurch. There is no consistent trend visible. The St Albans monitors are in a location with similarities to the Mt Eden II location in Auckland, i.e. an urban background location close to the downtown area. Mean concentrations at St Albans are similar to those at Mt Eden II.

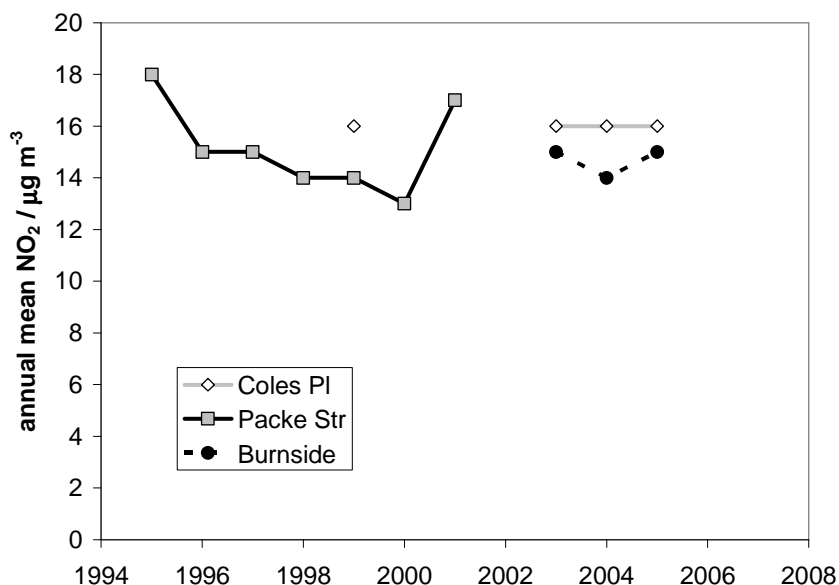


Figure 5.3. Annual mean NO₂ at a range of monitoring sites in Christchurch (restricted to years reporting > 75 % data coverage per year for at least 2 years).

Figure 5.4 displays the trends in annual mean NO₂ in Greater Wellington. Weak downward trends were evident between 2001 and 2005. Trends from 2005 to 2007 are unclear. Concentrations at the non-“transport” sites (including Karori, but excluding Tawa, which is influenced by SH1 – both not shown as <3 years of data available) have been below 12 µg m⁻³ since 2004, and have averaged 8 µg m⁻³ in that time. This is significantly lower than at comparable urban background locations in Christchurch and Auckland (13 – 20 µg m⁻³). With similar transport sources and no expectation of major differences in background oxidant, we tentatively ascribe this lower level of NO₂ in Wellington to the well-established windiness of the city.

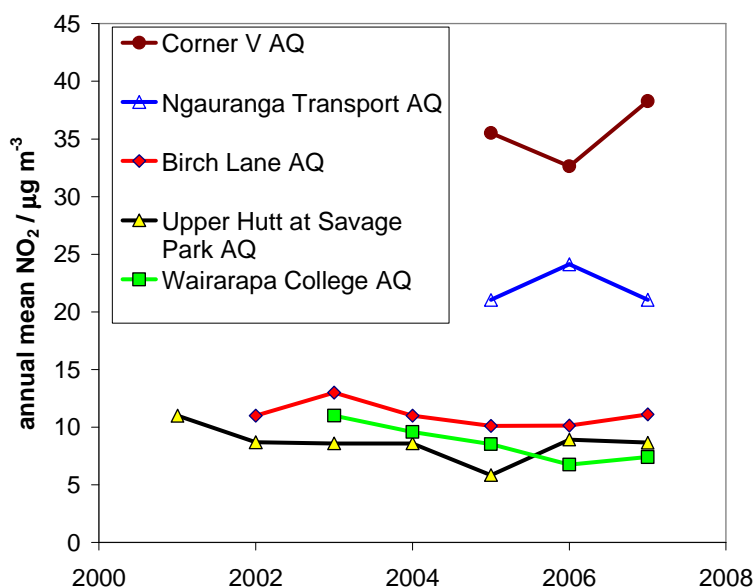


Figure 5.4. Annual mean NO₂ at a range of monitoring sites in Greater Wellington (restricted to years reporting > 75 % data coverage per year for at least 3 years).

The rise in NO₂ concentrations at motorway sites in the face of falling NO_x concentrations can potentially be explained by a number of factors:

1. an increase in the rate of NO oxidation via
 - a. increased levels of background oxidant,
 - b. changes in emissions of VOCs and/or nitrous acid from traffic or other sources.
2. an increase in primary NO₂ emissions from traffic due to
 - a. changes in speed or congestion,
 - b. new engine/exhaust technologies & their penetration into the fleet.

The first factor will be addressed below. The others will be dealt with in Part 2.

5.3 Trends in regional oxidants

5.3.1 Available ozone data

Ozone has been monitored at 7 sites in the Auckland Region since 1995, as listed in Table 5.2. Long-term data is available from Musick Point (although the site was relocated in 1999), Whangaparaoa, Pukekohe and Sky Tower. Their locations are indicated in Figure 5.6 and the long-term trend in annual mean concentrations at each site is illustrated in Figure 5.7. Shorter-term data has been gathered from 3 other sites, also listed in Table 5.2.

Table 5.2: List of ozone monitoring sites in Auckland Region. Long-term sites used in the following analysis are in bold.

Site	Site characteristics	from	to
Mangere Bridge	Urban edge/harbour	Aug 1995	May 1996
Musick Point I	Urban/coastal	Jan 1996	Jan 1999
Pukekohe	rural	Oct 1996	ongoing
Sky Tower	Urban elevated (250 m)	Apr 1998	Mar 2004
Whangaparaoa	Urban edge/coastal	Apr 1998	ongoing
Musick Point II	Urban/coastal	Feb 1999	ongoing
Kingsland	urban	Apr 2004	2008



Figure 5.6: Location of three ARC ozone monitoring sites referred to in the text.

5.3.2 Long-term mean trends

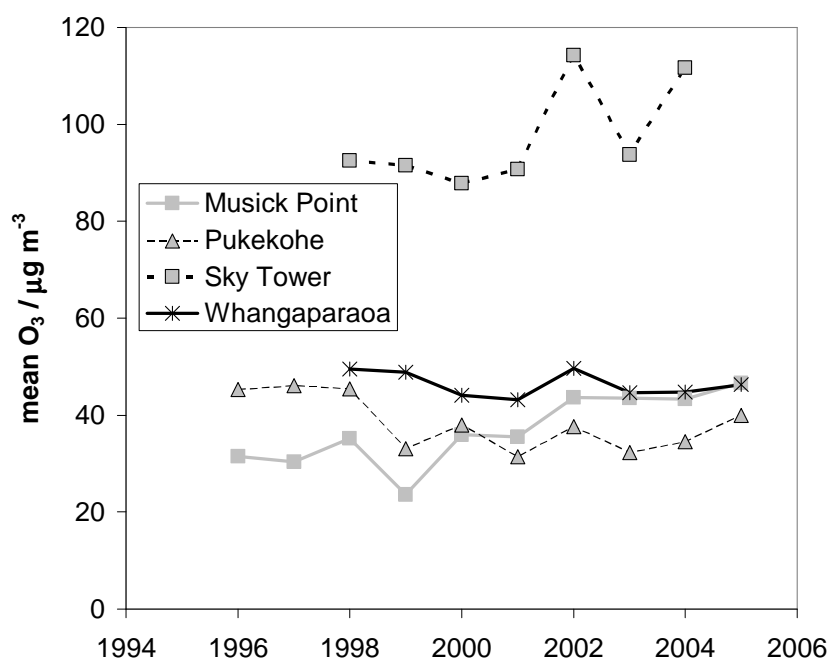


Figure 5.7: Annual mean ozone observed at four Auckland Regional Council monitoring sites.

As can be seen from Figure 5.7 mean observed ozone levels in Auckland region tend to 30 – 50 $\mu\text{g m}^{-3}$ (~15 – 25 ppb). Since 1999, mean ozone has been relatively constant at Whangaparaoa and Pukekohe, but there has been a rising trend at Musick Point. Mean concentrations have been consistently lower at Pukekohe compared to

Whangaparaoa by $10 \mu\text{g m}^{-3}$ (5 ppb) on average. However, this difference is more evident at night, such that the difference between daytime mean concentrations at these locations over the years 2004 – 2006 inclusive is only $5 \mu\text{g m}^{-3}$ ($42 \mu\text{g m}^{-3}$ at Pukekohe and $47 \mu\text{g m}^{-3}$ at Whangaparaoa).

Long-term measurements at Baring Head (9 km SE of the edge of Wellington, 1991 – 2004, NIWA data) reveal that annual mean ozone there is $41 \mu\text{g m}^{-3}$.

Only limited ozone data are available from Christchurch. Three surveys have been conducted, in 1998, 2003 and 2007. In 1998 O_3 was observed at Lincoln (~ 10 km SW of the edge of Christchurch) from 17th January – 27th April and at Kainga (~ 8 km N of the edge of Christchurch and 2 km S of Kaiapoi) from 15th January – 20th April. In 2003 and 2007 repeat measurements were made at Lincoln from 12th December 2002 – 31st March 2003 and 22nd December 2006 – 17th April 2007. The mean O_3 concentrations observed were $35 \mu\text{g m}^{-3}$ in 1998 (at both sites) and $36 \mu\text{g m}^{-3}$ in 2003. These data were all recorded in the summer to assess trends and the seasonal variation cannot be assessed in the absence of data from other seasons. In Auckland ozone is at a minimum in the summer with an average of around 30 to $35 \mu\text{g m}^{-3}$ in 2005.. Consequently we are unable to determine at this point whether typical annual ozone levels in Christchurch are higher, lower or similar to Auckland. Nevertheless, this analysis shows that differences in background ozone between Auckland, Wellington and Christchurch are not large.

5.3.3 Photochemical episodes

European and American literature discusses photochemical pollution episodes in which levels of ground-level ozone can rise significantly to, for example, 3 times the long-term mean (e.g. Jenkin et al., 2002). These are generally related to summer anti-cyclonic conditions and back trajectories that indicate an extensive multi-day air mass passage over VOC and NO_x emission source areas. New Zealand is distant from the extensive source areas required to generate such elevated ozone levels, and such episodes would not be expected.

The ARC data show that peaks consistent with photochemical episodes are exceedingly rare and of a smaller magnitude than observed in Europe. One such event occurred on the 27th October 2002, when ozone concentrations at all four monitors operating at the time (Sky Tower, Pukekohe, Whangaparaoa and Musick Point II) were elevated to almost double the typical values before and after the events for

almost half a day. Smaller events were observed late in 2003. Two events in 1997 were only observed at single monitors and cannot be confirmed as regional events.

5.4 Trends in Auckland motorway traffic

Figure 5.8 shows the long-term trend in traffic flow at three sections on the Auckland Southern Motorway (data from Transit NZ). The Penrose monitoring site is located adjacent to the section between Ellerslie-Panmure Highway and the South-Eastern Highway (the grey line in Figure 5.8). The discontinuities in the dashed line (Mt Wellington – Otahuhu) may be due to changes in traffic counting methods.

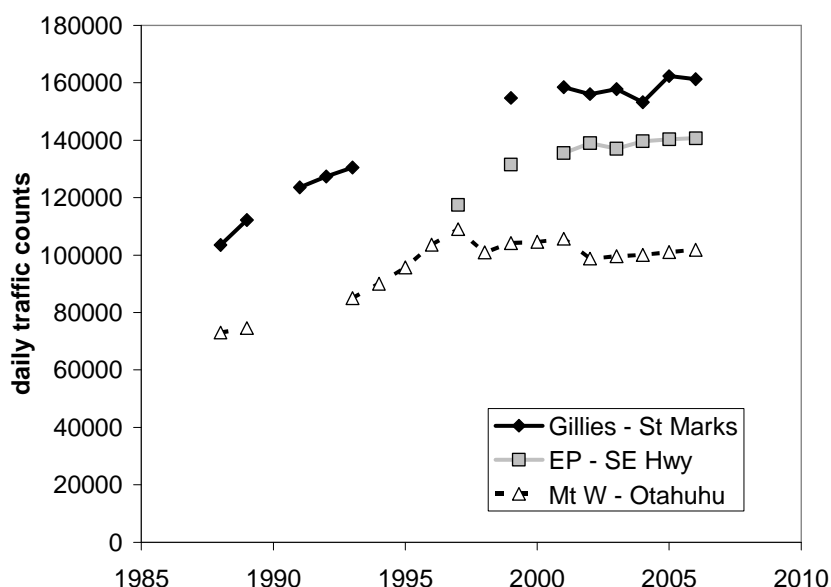


Figure 5.8: Long-term trends in traffic volume at three points on Auckland’s Southern Motorway (Gillies Ave – St Marks Rd, Ellerslie-Panmure Highway – South Eastern Highway and Mt Wellington Highway to Otahuhu). Data courtesy of Transit NZ.

In general this figure shows that the rate of traffic growth experienced on this motorway in the 1990s has slowed considerably in the current decade. Anecdotal evidence indicates that this has been caused by an increase in congestion and associated reduction in peak-time speeds rather than a reduction in the rate of growth of travel demand.

5.5 Trends in primary NO₂ emission

5.5.1 Review of international data on emissions

Conventionally it has been assumed that most NO₂ arising from vehicle emissions forms indirectly via the oxidation of nitric oxide (NO), which forms as a result of the high temperatures of combustion in the engine cylinders. The proportion of NO_x leaving the tailpipe in the form of NO₂ has historically been found to be 5 % or less. Whereas recent developments in engine technology and exhaust after-treatment has led to a reduction in typical NO_x emission factors, recent research has found that the proportion of NO_x emitted as NO₂ has increased in some new vehicles. Thus, cuts in NO_x emission may not necessarily lead to proportional cuts in NO₂ emissions, with consequences for ambient NO₂ concentrations and NES or RAQT compliance.

Almost three decades ago, Hilliard & Wheeler (1979) reported values of the NO₂/NO_x ratio at the tailpipe (which is referred to as $f(\text{NO}_2)$ to distinguish it from the NO₂ / NO_x concentration ratio in the ambient atmosphere) of 2 – 5 % for non-catalyst petrol cars, but more recent data has suggested that more recent vehicles equipped with three-way catalysts, and more recent emission-reduction technology may have much higher ratios. For instance, Soltic & Weilenmann (2003) investigated emissions from Euro II vehicles, observing ratios of 2 – 8 % (with one outlier above 10 %) from cars, rising towards 20 % at cold start and 10 – 40 % from light-duty trucks. More recently, the Dutch and Swiss institutions TNO and EMPA jointly conducted tests on 69 passenger cars spanning pre-Euro to Euro IV (Gense *et al.*, 2006). The values of $f(\text{NO}_2)$ for the petrol vehicles generally increased with more recent emission standards ranging in total from a few % to 20 %. For diesel cars the data ranged from 5 % to 80 %. A large step increase was observed between Euro II and Euro III from ~ 15 – 20 % to ~ 50 %. The highest values (70 – 80 %) were observed for Euro IV diesels with D-Kat (diesel particulate filter with NO_x absorber-catalyst) or catalysed diesel particulate filters.

The net $f(\text{NO}_2)$ ratio from a stream of traffic may be estimated from measurements of NO₂ and NO_x inside a road tunnel, if the concentrations are corrected by removal of external background concentrations. There have been remarkably few examples of this reported in the open literature. Measurements inside a short road tunnel in Birmingham (UK) conducted in 1992 and 1993 (note that the Euro I standard for cars and HGVs only came into force in 1992) indicated a net $f(\text{NO}_2)$ emission ratio of 3 – 8 % with the higher values observed during traffic congestion (Harrison & Shi, 1996).

More recent research has investigated the variability in the $f(\text{NO}_2)$ emission ratio as a function of traffic. Carslaw & Beevers (2005) summarised dynamometer

measurements by Latham *et al.* (2001) to show how the $f(\text{NO}_2)$ emission ratio for London traffic in 2001 varied with vehicle type and speed. The ratio was shown to be a minimum at speeds above 60 km h^{-1} , whilst peaking at low speeds for petrol cars and at $40 - 50 \text{ km h}^{-1}$ for diesel LGVs, buses and HGVs. For the mean London fleet, $f(\text{NO}_2)$ was estimated as $10 - 12 \%$ below 50 km h^{-1} falling to below 5% above 80 km h^{-1} .

Subsequently, UK research has identified a recent and rapid rise in the $f(\text{NO}_2)$ emission ratio, rising from $5 - 7 \%$ in 1997 and reaching 30% by 2006 (Carslaw, 2005, Abbott, 2005, Carslaw *et al.*, 2007). This rise has been attributed principally to the penetration of Euro III light duty diesel vehicles fitted with oxidation catalysts and the fitting of regenerating particle traps to buses. A similarly rapid increase in the $f(\text{NO}_2)$ emission ratio from 14% in 1992 to 23% in 2004 has been observed at a motorway monitoring site in Switzerland (Hueglin *et al.*, 2006) and a rise from 5% in 1995 to 20% in 2005 in monitoring sites in Baden-Wurtemberg, Germany (Kessler *et al.*, 2006).

5.5.2 Penetration of high $f(\text{NO}_2)$ vehicles into the NZ fleet

NIWA is currently in the early stages of developing a National Pollutant Inventory with an initial focus on road transport emissions. One of the activities within this research will be to co-ordinate and enhance different sources of data on vehicle fleet composition. Part 2 of this report will summarise progress made on this issue to date.

5.5.3 What is the average $f(\text{NO}_2)$ on urban roads in New Zealand?

Once well into the oxidant-limited regime, the slope of the $\text{NO}_2 - \text{NO}_x$ curve reduces asymptotically towards a value representing the amount of locally emitted oxidant. Most (if not all) of this oxidant will be in the form of NO_2 at the roadside and will be derived either from direct emission or will be chemically produced by other means (such as free-radical initiated oxidation of VOCs). Other evidence suggests that the latter source is small compared to direct emission, such that the slope of the $\text{NO}_2 - \text{NO}_x$ curve to converge towards the fraction of NO_x emitted locally as a fraction of NO_x , i.e. $f(\text{NO}_2)$. I.e. if oxidant is completely limited so that no NO is converted to NO_2 , and there are no local emission sources of NO_2 , then the slope of the $\text{NO}_2 - \text{NO}_x$ curve will tend to zero. This is shown in Figure 5.9 for four Auckland and two Wellington roadside sites. All of the lines can be seen to be levelling off towards a value somewhere between 0.05 and 0.12 . As $f(\text{NO}_2)$ is a function of the vehicle fleet mix on the adjacent road we may expect the minimum value of the slope to be site-specific. Although far from conclusive, Figure 5.9 suggests that $f(\text{NO}_2)$ is higher at Queen Street and Khyber Pass Road than at the other sites.

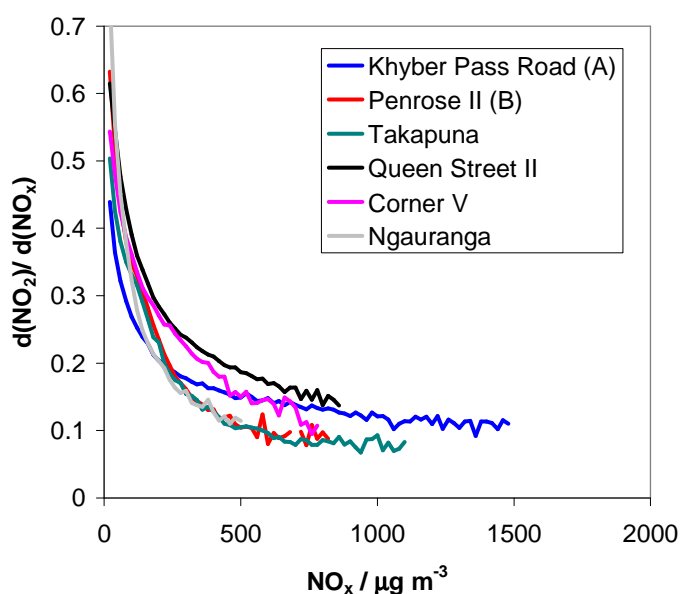


Figure 5.9: Slope of the binned average NO_2/NO_x versus NO_x graph as a function of NO_x for some roadside monitoring data, 2004 – 6.

5.5.4 Significance of primary NO_2 emissions in urban New Zealand

At high NO_x concentrations, as exists near major roadsides during hours of busy traffic, the available ozone will rapidly be depleted by titration of emitted nitric oxide. This depletion prevents the oxidation of further NO to NO_2 until such time that the plume is advected away from the road (and the NO emissions) and mixes with air from aloft which is richer in ozone. Thus, NO_2 production is effectively ‘delayed’ and displaced away from the roadside. With the conversion of NO to NO_2 stalled at the roadside the fraction of NO_x emitted directly as NO_2 acquires much greater importance. To investigate the significance of direct NO_2 emissions we have trialled a method of NO_2 source apportionment in which the NO_2 reported from a roadside monitor is apportioned to three sources:

1. primary NO_2 directly emitted from the tailpipe by traffic on the adjacent road,
2. background NO_2 advected into the urban area from beyond, and
3. secondary NO_2 formed from titration of NO and O_3 in the urban background.

The method will be described in detail in Part 2. Trial results are shown in Figures 5.10 and 5.11. Figure 5.10 shows the diurnal average NO_2 for 2004 and 2005 as measured at Khyber Pass Road in Auckland. The NO_2 is segregated into the three

sources as discussed above. The method is imperfect and has some limitations. The consequence is that this figure (and any data derived from this method) should be treated with caution such that arguments and conclusions should not be built around any individual value. Rather, the value of this method is in the general trends and patterns it reveals.

Figure 5.10 shows that during most of the day, when traffic levels and total NO₂ concentrations are high, primary NO₂ (shown in blue) dominates over the other two sources. This dominance is seen through every month of the year. Non-direct sources dominate only at night. At Queen Street (Figure 5.11) we have modelled a similar dominance of direct NO₂ but with the difference that this dominance even continues through the night. These results can offer clues to the chemical and physical processes influencing NO₂ levels and will be explored further in Part 2. They also clearly indicate that mitigation strategies must consider the much larger ‘leverage’ offered by aiming to control the directly emitted fraction

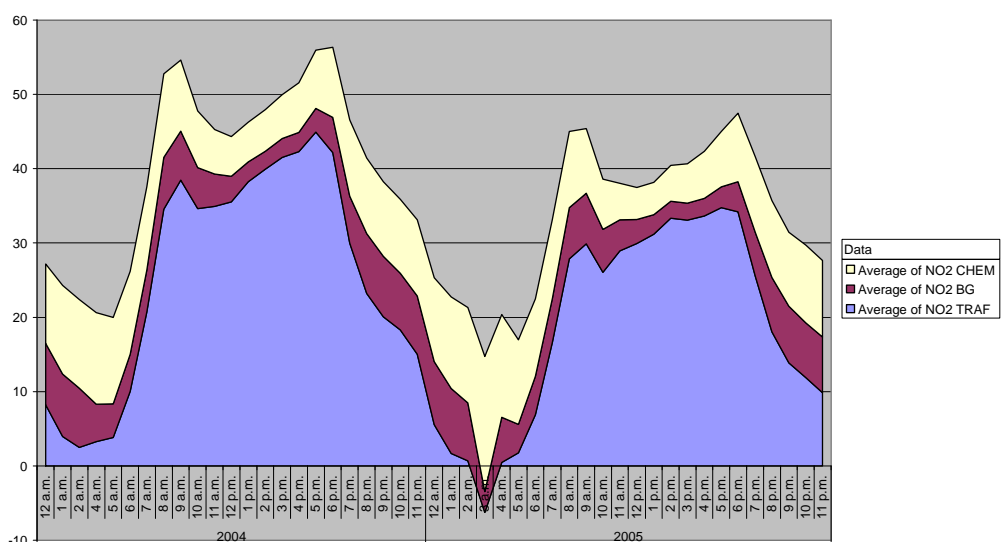


Figure 5.10. Diurnal variation of the averages of the estimated apportionment of NO₂ concentrations at Khyber Pass Road for 2004 and 2005. Blue = NO₂ attributed to direct primary emission, purple = NO₂ attributed to regional background, yellow = NO₂ attributed to conversion from NO in the urban airshed.

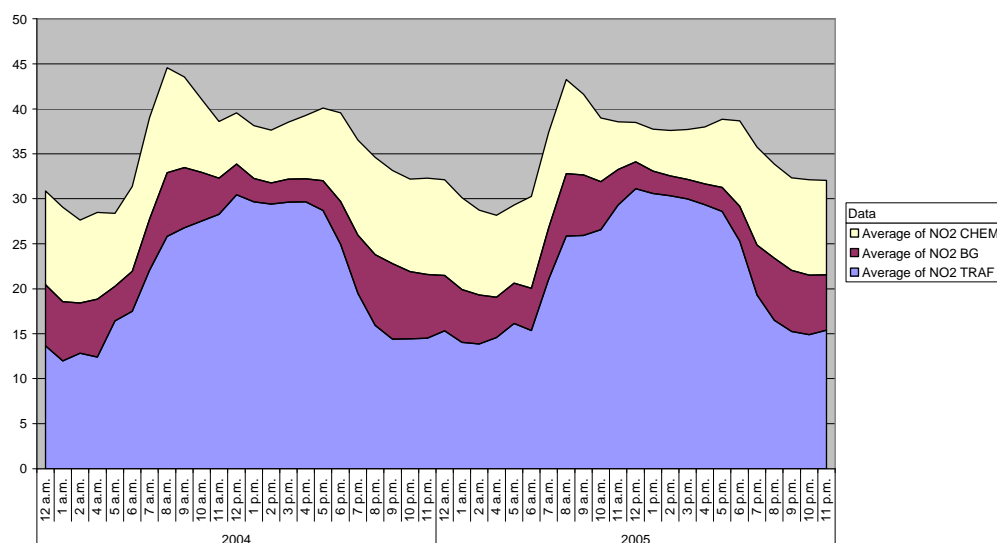


Figure 5.11. Diurnal variation of the averages of the estimated apportionment of NO₂ concentrations at the Queen Street II site for 2004 and 2005. Blue = NO₂ attributed to direct primary emission, purple = NO₂ attributed to regional background, yellow = NO₂ attributed to conversion from NO in the urban airshed.

5.6 How are NO₂ and NO_x concentrations linked at different sites?

5.6.1 Urban nitrogen partitioning in NZ – review of data

Figure 5.12 shows the best fit relationship between NO₂ and NO_x for the four Auckland sites with the highest concentrations, and the Corner V street canyon site in Wellington. This data was derived by finding the mean NO₂ for each corresponding NO_x concentration (in 10 µg m⁻³ bins) and then fitting a cubic expression to the resulting curve. This method leads to a small error at the low concentration end of the graph. However, our primary interest is the general shape of the curve, especially at higher concentrations. Figure 5.12 shows clearly three classes of behaviour represented by:

1. Takapuna and Penrose II,
2. Queen Street II and Corner V,
3. Khyber Pass Road.

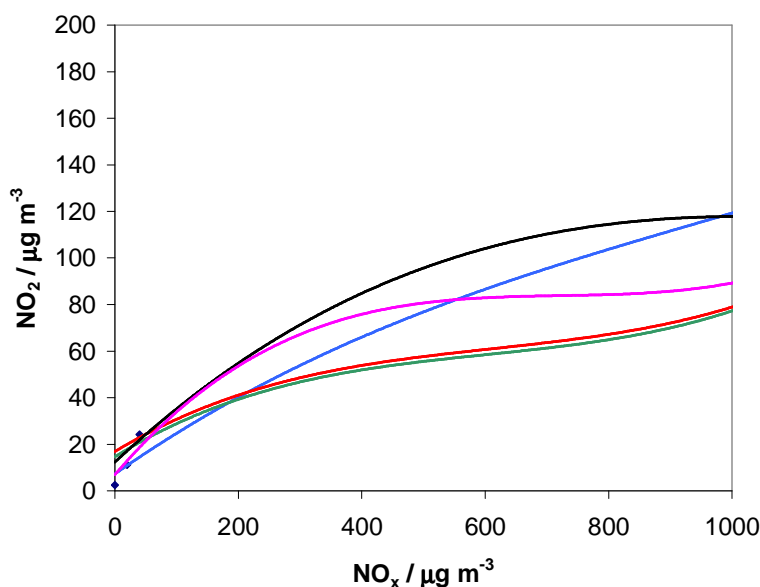


Figure 5.12: Best fit relationships between NO₂ and NO_x for five key roadside monitoring sites, 2004 – 2006 data inclusive. Black = Queen Street II, pink = Corner V (Wellington), blue = Khyber Pass Road, green = Takapuna, red = Penrose II.

The Wellington motorway site (Ngauranga) and other shorter-term motorway sites (Penrose IV) also exhibit very similar behaviour to Takapuna and Penrose II (not shown for clarity). Queen Street II and Corner V are both downtown street canyon sites. They have similar behaviour at low concentrations but they deviate at higher NO_x with higher NO₂ at Queen Street. The reason for this will be investigated further in Part 2. In our review Khyber Pass Road appears to be unique. Thus, we can reclassify our sites as:

1. motorway
2. downtown street canyon
3. Khyber Pass Road

Figure 5.13 shows the same curves for NO_x concentrations below 400 µg m⁻³ (this improves the fit at lower concentrations). This shows that the less road-influenced urban sites (Mt Eden II and Kingsland) also have a similar behaviour, with the semi-background site of Music Point II being distinctly different again, leading us to define two more classes of site:

4. urban background (e.g. Mt Eden, Kingsland),
5. urban periphery (e.g. Musick Point)

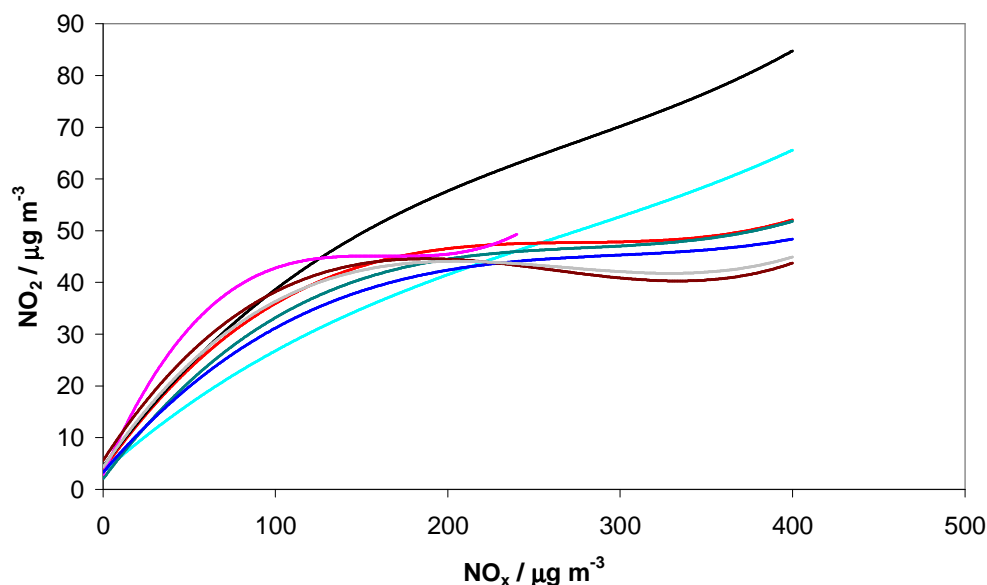


Figure 5.13: Best fit relationships between NO_2 and NO_x at 8 monitoring sites in Auckland Region, 2004 – 2006 inclusive. Black = Queen Street II, light blue = Khyber Pass Road, red = Penrose II, pink = Musick Point II, brown = Mount Eden II, green = Takapuna, grey = Kingsland, dark blue = Henderson.

These plots indicate two regimes of nitrogen chemistry. At the simplest level these regimes may be characterised as *oxidant-available* (or *NO_x -limited*) and *oxidant-limited* regimes. The low concentrations regime represents the ready conversion of emitted NO into NO_2 . The high concentration regime represents the reduced conversion of NO to NO_2 due to the reduced availability of oxidant. The transition between the regimes is unique for each site. For less polluted sites (and less urbanised sites) the transition occurs at lower NO_x concentrations (e.g. 40 – 100 $\mu\text{g m}^{-3}$) and is more well-defined (i.e. represented by a sharper change of slope in the chart, e.g. Musick Point II). For more polluted sites, the transition occurs at higher concentrations and is smoother and therefore harder to quantify.

5.6.2 Street canyon sites

Why is NO_2 higher for a given concentration of NO_x at the street canyon sites, relative to the motorway sites? Below approximately 150 $\mu\text{g m}^{-3}$ of NO_x most sites exhibit similar behaviour in terms of NO_2 concentrations. However, above this level NO_2

concentrations at Queen Street II and Corner V are far higher for a given value of NO_x than at all other sites. This is clearly within the ozone-limited regime, suggesting that within these street canyons the conversion of NO to NO_2 is not limited in the same way, or that there is an extra source of NO_2 . As NO_x rises above $260 \mu\text{g m}^{-3}$ the two sites diverge with NO_2 at Corner V being limited but less so at Queen Street II. The key differences between Queen Street and Corner V are that

- a) Queen Street is a deep canyon whereas Corner V is shallow,
- b) Queen Street has atypical traffic characteristics with a relatively low total volume but high proportion of buses, while Corner V has more typical traffic.

One of the key features of dispersion in a street canyon is the recirculation of air caused by low pressure zones which form in the lee of buildings. Complex, three-dimensional intermittent vortices can form which, compared to more open locations, can retard net dilution and increase the length of time an air parcel remains within the 'street'. These vortices can aid entrainment of ozone from aloft to ground level and aid the mixing of ozone and nitric oxide emissions. The restriction on sunlight penetrating to the bottom of deep canyons may also inhibit the photolysis of NO_2 (which regenerates NO and ozone) leading to total oxidant (i.e. the sum of NO_2 and O_3 , also known collectively as OX) and NO_x being more biased towards NO_2 (Grawe *et al.*, 2007).

The traffic fleet operating in Queen Street is biased towards diesel vehicles which are associated with higher emissions of direct NO_2 . Supporting evidence is supplied by the source apportionment results (see Appendix 2).

We are unable as yet to corroborate which, if any, of these factors explain the observed NO_2 - NO_x behaviour at Corner V and Queen Street. Our understanding would be vastly improved if concurrent and co-located ozone measurements and detailed traffic observations were undertaken at these sites.

5.6.3 Khyber Pass Road

The Khyber Pass Road data exhibits two unusual features:

1. low NO_2 at low NO_x ,
2. very little change in the $\text{NO}_2 / \text{NO}_x$ slope at high NO_x

The low slope at low NO_x is most likely related to the very close proximity of the monitor inlet to the road (approx 2.5 m from the kerb). This ensures that a large proportion of NO_x is in the form of NO due to the very short time from emission to observation relative to the time required to convert the NO to NO_2 . This effect of distance can be observed in the data from ARC's Penrose IV site. This was a mobile monitor that was moved to different distances downwind of Auckland's Southern Motorway in 2004 – 2006. Cubic equations have been fitted to the average NO_2 for 20 $\mu\text{g m}^{-3}$ wide NO_x bins for periods when the monitor was 15, 42 and 62 m from the motorway, as shown in Figure 5.14. The influence of distance on the slope of these curves at low concentrations can clearly be seen.

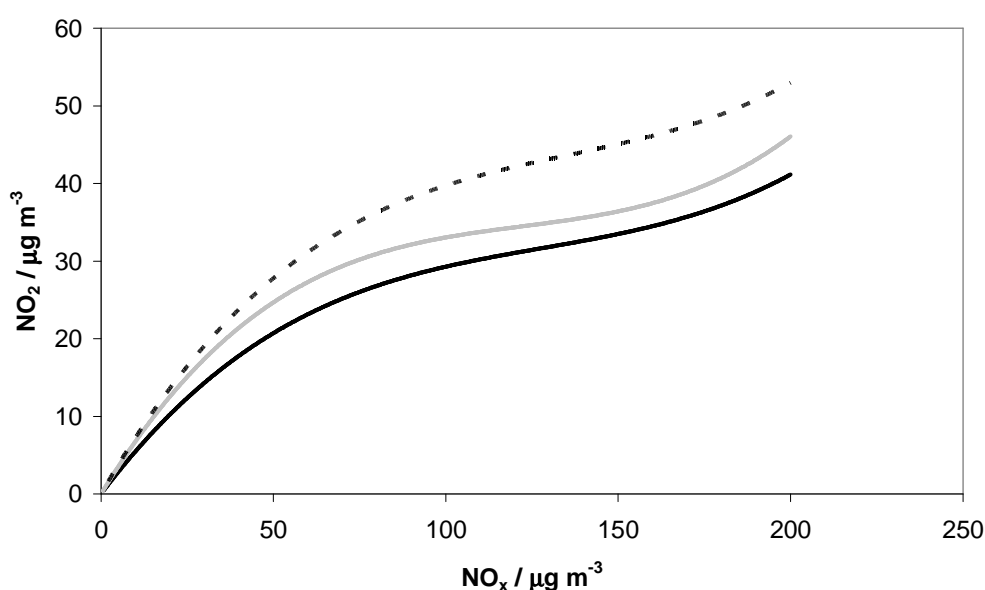


Figure 5.14: Best fit relationships between NO_2 and NO_x at the Penrose IV mobile site (ARC) at 15 m (black), 46 m (grey) and 62 m (dashed) downwind of Auckland's Southern Motorway.

The lack of any significant turning point in the curve as occurs for most other sites, indicative of ozone-limitation, is intriguing. It suggests that at higher NO_x concentrations the ozone depletion which must occur is compensated for by another NO_2 production mechanism. Like Queen Street, Khyber Pass Road is quite heavily used by buses. It is also frequently used by heavy duty trucks. Other than that it has a high traffic volume compared to Queen Street. The monitor is also on a section of the road which has an uphill gradient in the direction adjacent to the inlet and a traffic signal, such that vehicles are aggressively accelerating right in front of the monitor. In Part 2 we will investigate further the potential role of these traffic characteristics in explaining the observed relationships between NO_2 and NO_x at Khyber Pass Road.

5.6.4 Use of co-located ozone data to interpret NO₂ sources

None of the monitoring sites discussed in detail above have co-located ozone measurements. To illustrate the value of roadside ozone data we briefly indicate what further knowledge can be gained by considering the Kingsland site. This monitoring site is on the Auckland isthmus and can generally be considered an urban background site, although there are two moderately busy roads to the north and west. Kingsland is the only urban site at which NO_x and O₃ have been simultaneously measured.

We can use this data to investigate not just how NO_x is partitioned between NO and NO₂, but how total oxidant (OX) is partitioned between NO₂ and O₃ (where OX[ppb] = NO₂ [ppb] + O₃ [ppb]). We have calculated daily averages of NO₂, NO_x, O₃ and OX using daytime data only for the period 2004 – 2006 inclusive. We have applied a linear regression to the relationship between total oxidant and NO_x to get

$$\text{OX [ppb]} = 25.8 + 0.031 \text{ NO}_x[\text{ppb}],$$

i.e. total oxidant consists of a background component, of mean value 25.8 ppb and a component derived from local emissions of 3 ppb for every 100 ppb of NO_x. These two values can be compared to those observed elsewhere. At present we are restricted to international comparisons in the absence of more urban O₃ data in New Zealand. The local OX production rate of 0.03 ppb ppb⁻¹ NO_x is consistent with the lower values observed at urban sites in the UK (Clapp & Jenkin, 2001) and the assumption of a lower f(NO₂) (fraction of NO_x emitted as NO₂) arising from the New Zealand vehicle fleet compared to the more diesel-influenced UK fleet. Compared to the findings in this study, Auckland's (and we may presume all of New Zealand's) lower background oxidant and lower local oxidant production rate mean that higher NO_x concentrations are required to produce any given NO₂ concentration, including the WHO annual guideline, Regional Targets or NES.

Figure 5.15 shows that at the urban, partially road-influenced Kingsland site, the OX crossover point, i.e. the level of NO_x above which NO₂ dominates over ozone is at ~40 ppb of NO_x (i.e. ~ 80 µg m⁻³ of NO_x). This is consistent with a photolysis rate of NO₂ of approximately 0.012 s⁻¹ based on the assumption of a photostationary state (making the simplified assumption that VOC oxidation plays no role in NO₂ production). This is consistent with findings for the only other site for which concurrent NO₂ and O₃ data is available covering high enough concentrations to observe this crossover (Musick Point II, data not shown). There is no NO₂ crossover point, i.e. NO always exceeds NO₂ on average.

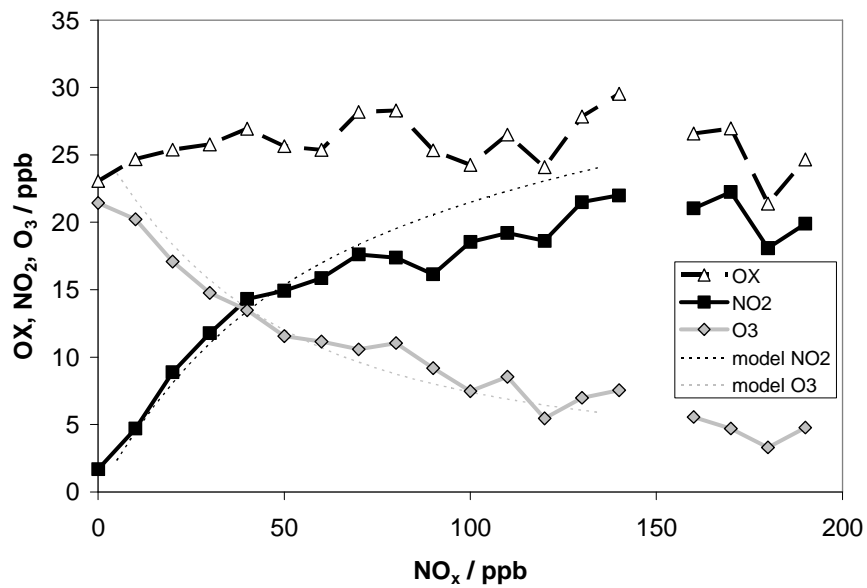


Figure 5.15: Mean concentrations of (in ppb) OX, NO₂ and O₃ for given concentrations of NO_x (in bins of 10 ppb width) at Kingsland, 2004-6 inclusive, and as predicted by a photostationary state model. Units of ppb have been adopted so that numerically O₃ + NO₂ = OX.

6. Conclusions

6.1 Sea breeze recirculation and sulphate formation

Meteorological modelling of airflow in the Auckland airshed during an anti-cyclonic period in March 2006 has identified a recirculating pattern of airflow around the east coast. Trajectory modelling suggests that air originating in the region of the Port of Auckland and the Hauraki Gulf shipping channels was advected out to sea at night but drawn back to the land in the morning. Some of the trajectories returned to Auckland city in ~ 1 day. It seems plausible to consider that gaseous precursor pollutants emitted into this recirculating air mass from a shipping source would be constrained in this recirculation such that upon return to Auckland a significant amount of SO₂ may have been converted to ammonium or sodium sulphate and thus contribute to PM₁₀ concentrations in Auckland. This may offer an explanation of the elevated levels of sulphate observed by receptor modelling in this period.

We conclude that this meteorological modelling approach is worth pursuing. The trajectory analysis is able to identify the links between emissions along shipping lanes and sulphur pollution on land. Further investigation will allow us to quantify the probability of emissions originating in Auckland being returned to Auckland some hours or days later by advection, how this process is determined by meteorological variability, whether there are key conditions that increase that probability, the variability in residence or recirculation time and the potential scale of the impact on PM₁₀. Thus, further modelling will be conducted covering a much wider range of case studies and meteorological conditions. The use of the 2006 Auckland source apportionment data (Davy *et al.*, 2007) for validation will be further investigated, plus the use of limited chemical modelling to explicitly simulate SO₂ to sulphate conversion along identified plume trajectories.

6.2 Peak nitrogen dioxide

This review of data originating principally from Auckland Regional Council and Transit NZ has shown that New Zealand's highest levels of observed nitrogen dioxide, both in the short-term (i.e. 1 hour averages) and in the long-term, are found on streets in downtown Auckland carrying busy traffic. Many of these streets could be described as street canyons due to their being lined with tall buildings, although high concentrations have also been observed at locations with more modest buildings, such as in Newmarket on the fringe of the city centre. Although traffic levels can be high at these locations, they are often significantly lower than on Auckland's motorway and major road network, suggesting that the well-known phenomenon of air recirculation

in street canyons and in the lee of buildings is a significant process causing elevated concentrations.

After downtown Auckland, the highest concentrations are observed alongside motorways. Higher concentrations are associated with busier motorways on the side of the motorway which is predominantly downwind only. Concentrations on the upwind side of the busiest motorways appear to be unrelated to traffic volume and similar to those downwind of less busy motorways. There is still some more progress required in defining the spatial extent of the influence of a motorway on NO₂ concentrations. However, this initial review suggests that concentrations are significantly raised within at least 100 m of a motorway and are raised at 5 m relative to 100 m by up to 100 %.

Auckland appears to possess higher NO₂ concentrations than other New Zealand cities, however our ability to compare cities is limited by the relative paucity of data outside Auckland.

In Part 2 of this report we shall extend this review to include additional sources of data. We also aim to advance the general understanding of atmospheric chemistry in New Zealand such that the roles of traffic volume, monitor location, distance to the road and street canyon situations can be generalised and interpreted.

6.3 Trends influencing future peak NO₂ levels

Generally, trends in ambient concentrations of NO_x are down, especially at roadside sites. Yet trends in NO₂ are not so clear, with many sites reporting static or upward trends in annual mean concentrations. Auckland's two motorway sites, in particular, display upward trends in NO₂ despite downward trends in NO_x.

Our initial research has revealed no evidence to suggest these trends are caused by changes in the level of background regional oxidants. Typical background ozone concentrations upwind of Auckland, Wellington and Christchurch are of the order 40 µg m⁻³. There is no clear inter-annual trend, although this is based on fewer data than are available for NO₂ and NO_x. Photochemical episodes involving a doubling or more of regional ozone concentrations, as discussed extensively in American and European literature, appear to be exceedingly rare events in Auckland and are not significant in terms of long-term NO₂ trends.

The motorway-site trends may be related to long-term changes in traffic flow, with congestion becoming increasingly common in this decade, and with increases in the

proportion of NO_x emitted directly as NO_2 . This latter change is associated with increased fleet penetration of diesel vehicles and/or vehicles fitted with some modern exhaust treatment technologies (especially oxidation catalysts and particulate filters). We have shown that the proportion of NO_2 arising from direct emission is very significant at roadside sites. Part 2 will consider the issue of changes to traffic flow and direct emissions in more detail.

The probability of exceedence of the NES for NO_2 (or any other short-term target) has been investigated as a function of the level of NO_x likely to lead to a given concentration of NO_2 . The threshold NO_x for a given NO_2 is much higher at Auckland's Queen Street II site than for any other we have considered. We have tentatively attributed this to the deep street canyon location of this site. However, we cannot yet rule out that the particular traffic fleet mix (bus-dominated) and subsequent emissions mix being a contributing factor.

The relationship between NO_2 and NO_x at the Khyber Pass Road site is quite different to all other sites. We have tentatively identified the very close proximity of the monitor inlet to the road as a major reason for this. We intend to investigate this further in Part 2.

6.4 Future research and recommendations

The purpose of this Part One report was to summarise the current state of knowledge regarding two key secondary pollutants in urban New Zealand – particulate sulphate and nitrogen dioxide. It was also to trial some tools that may be exploited to develop our understanding, particularly

- High resolution meteorological modelling and trajectory analysis,
- Analysis of monitoring data,
- Semi-empirical chemical source apportionment for NO_2 ,
- Empirical photostationary state modelling.

In Part Two we intend to report on the extended use of these methods and other methods not covered in Part One, and how they can be combined to fill the knowledge gaps and help explain the trends and patterns identified in the data.

Progress in understanding the physical and chemical processes in the air of urban New Zealand is limited by some substantial data gaps. Modelling, whether it is empirical or semi-empirical chemical modelling, dispersion modelling or chemical transport modelling is ultimately dependent upon observational data. Observational data in New Zealand is biased towards NES-compliance monitoring. Our analyses are hindered by the lack of roadside and urban ozone data, the presence of which would greatly increase the accuracy and reduce the uncertainties in the chemical source apportionment technique. Similarly, data on regional background nitrogen dioxide is very limited. Urban/roadside ozone and rural NO₂ are rarely monitored as NES-compliance is not an issue at those particular locations, but atmospheric processes are such that rural NO₂ and urban roadside O₃ play key roles in determining whether urban locations comply with the NES for NO₂.

Furthermore, this report has not covered nitrates or the role of other nitrogenous species, such as HONO or ammonia. This is principally because of the general paucity of data regarding these substances. We note that the report to ARC regarding the 2006 source apportionment study (Davy et al., 2007) recommended monitoring of ammonium nitrate and ammonia upwind of Auckland, and we endorse that recommendation.

7. References

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Appendix 1: WRF Auckland March 2006 case-study

1. Model and configuration

To produce the meteorological fields we have employed the NCAR Weather Research and Forecasting (WRF ARW Version 3) Model. WRF is a next-generation state-of-the-art mesoscale numerical weather prediction system designed to serve both operational forecasting and atmospheric research needs. WRF is based on Eulerian mass solver and intended to be used for a broad spectrum of applications across scales ranging from metres to thousands of kilometres (Developmental Testbed Center, 2006). It is suitable for operational weather forecasting, regional climate prediction, driving air-quality models, data assimilation, parameterized physics research and idealized dynamical studies (Michalakes et al., 2006). The model consists of fully compressible non-hydrostatic equations. The prognostic variables include the three-dimensional wind, perturbation quantities of potential temperature, geopotential, surface pressure, turbulent kinetic energy and scalars (water vapor mixing ratio, cloudwater etc). The model's vertical coordinate is terrain following hydrostatic pressure and the horizontal grid is Arakawa C-grid staggering. A 3rd order Runge–Kutta time integration is used in the model. The model has several options for spatial discretization, diffusion, nesting, lateral boundary conditions and physics parameterization.

For Auckland region, the WRF model was designed with 3 nested grids with horizontal resolution of 4500, 1500 and 500 metres. One-way interactive nesting was used where information is passed from the coarser grid to the finer grid. All three grids have the size of 151 x 151 grid points and all domains contain 32 vertical levels each. The 6-hourly FNL Global Analysis Data of the NCEP Global Tropospheric Analysis with 1 x 1 degree resolution were used. The centre of all three grids was located at Mount Eden in Auckland (36.8778oS, 174.7644oE). WRF single-Moment 5-class scheme was used for cloud and ice processes, Dudhia-scheme was employed for short wave physics and RRTM scheme for long-wave radiation, surface temperature predicted by four layers soil diffusion unified Noah land-surface model while Monin-Obukhov (Janjic Eta) scheme used for surface-layer physics; Mellor-Yamda-Janjic (Eta) TKE scheme was used to calculate to boundary layer physics. The model was integrated for two days (48 hours) from 12:00 LST 19th March to 12:00 LST 21 March 2006. Once the simulation was completed, a meteorological output visualizing program RIP (Read, Interpolate, Plot) version 4.4.

2. Modelling output

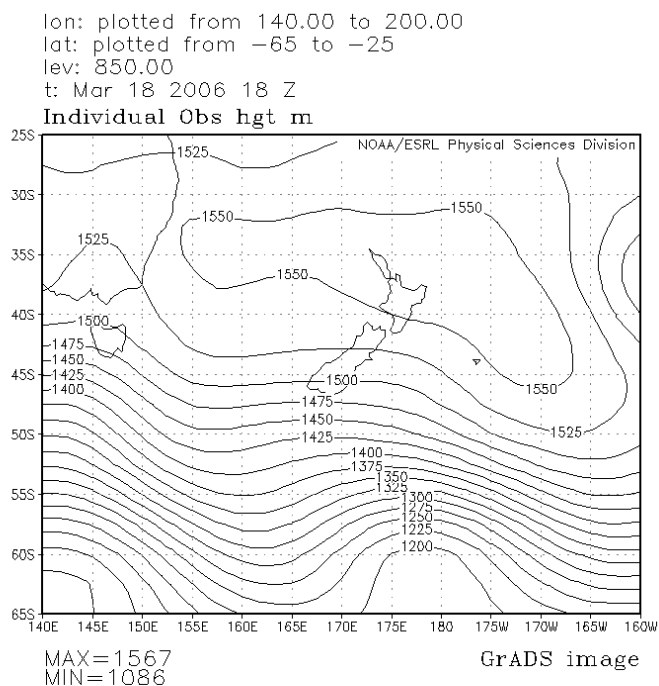


Figure 8.1. Geopotential height at 860 mb at 0600 hours 19th March 2006; NOAA image.

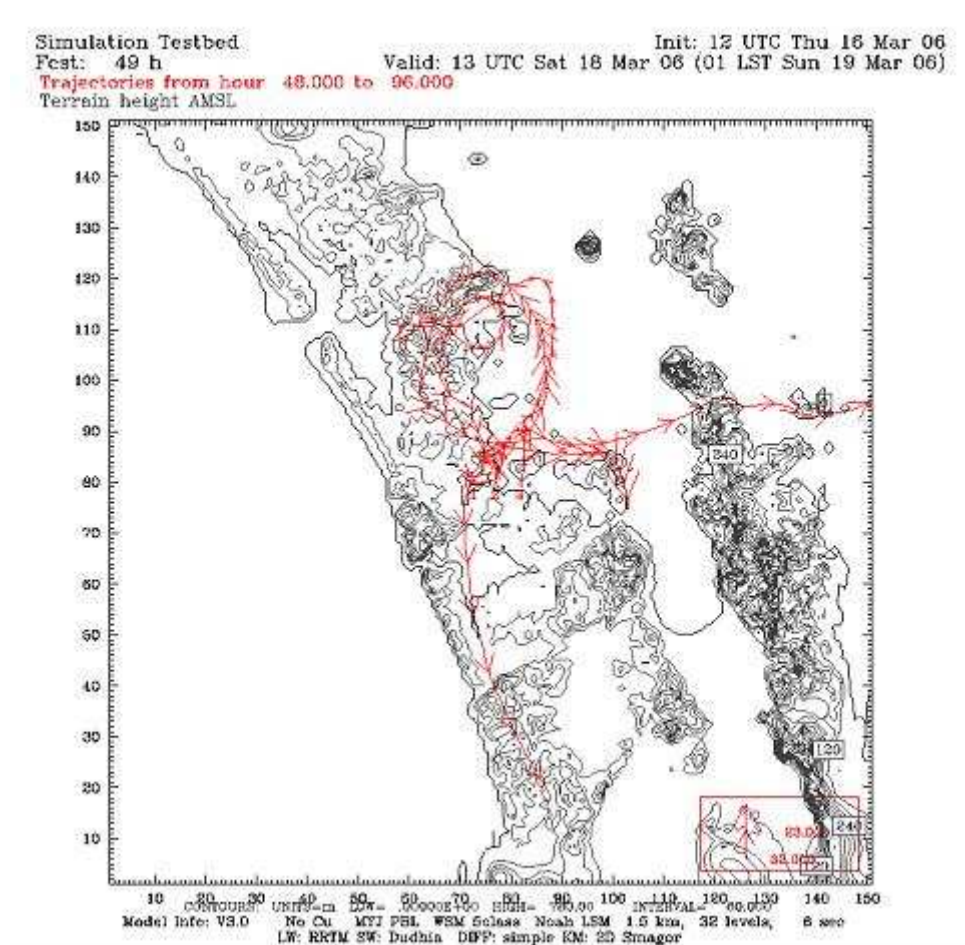


Figure 8.2: 48 hour trajectory Plot of wind field (12 metres above ground) originating at Port of Auckland (00:00 hours 19th Mar 2006 to 00:00 hours 21th Mar 2006).

Simulation Testbed
 Fcst: 48 h Valid: 12 UTC Sat 18 Mar 06 (00 LST Sun 19 Mar 06)
 Trajectories from hour 48.000 to 96.000
 Terrain height AMSL

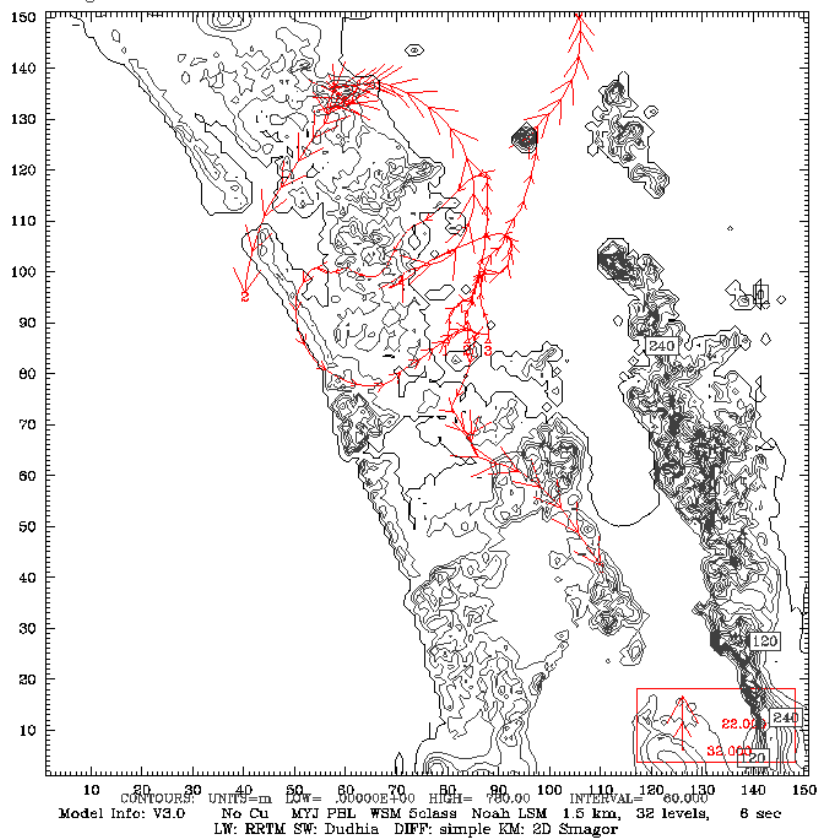


Figure 8.3: 48 hour trajectory Plot of wind field (12 metres above ground) originating north of Rangitoto (00:00 hours 19th Mar 2006 to 00:00 hours 21th Mar 2006).

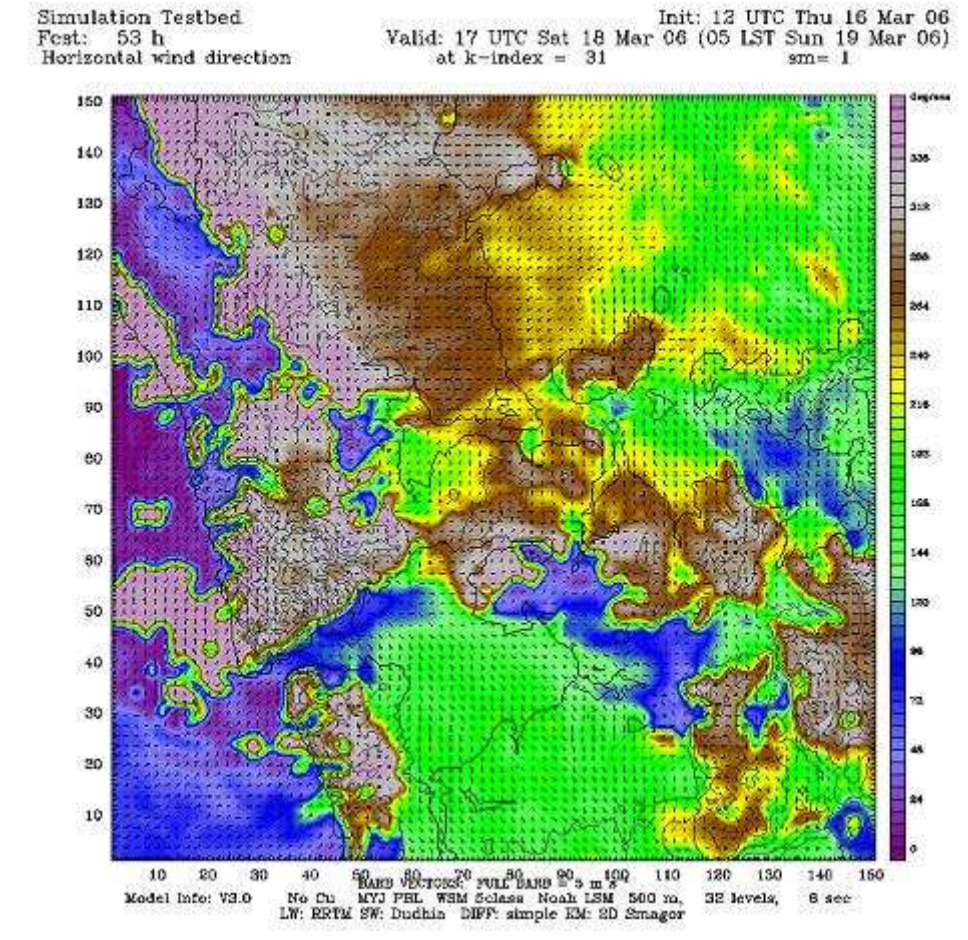


Figure 8.4: Wind vector plot (05:00 hours 19Mar 2006) showing mature land breeze on the east coast of the Auckland region, extending ~20 km into the Hauraki Gulf. Opposing land breezes are also seen on the west coast (blue-purple).

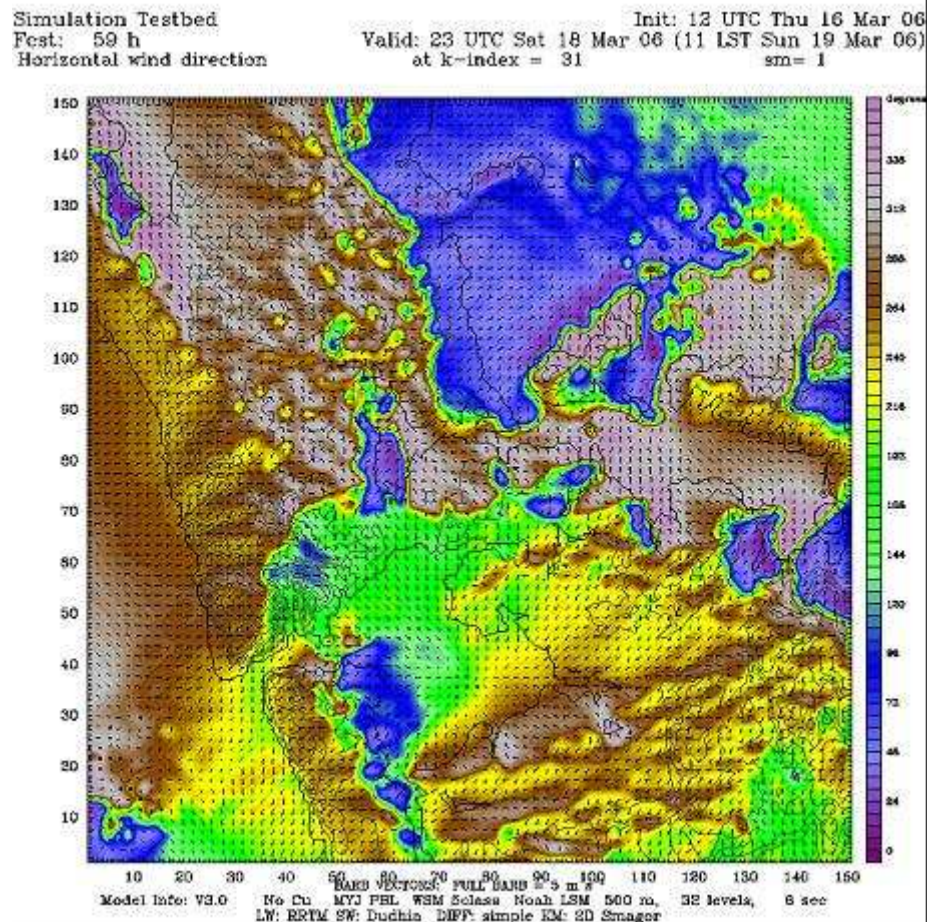


Figure 8.5: Wind vector plot (11:00 hours 19th Mar 2006) showing mature sea breezes on both east and west coast of the Auckland region and further north in the afternoon. To the north of Auckland, the inland penetration of easterly sea breezes on the east-coast is up to 15% of the total width of the land mass. In this region, the easterly sea breezes are forming convergence zone with the opposing westerly sea breezes from the west coast.

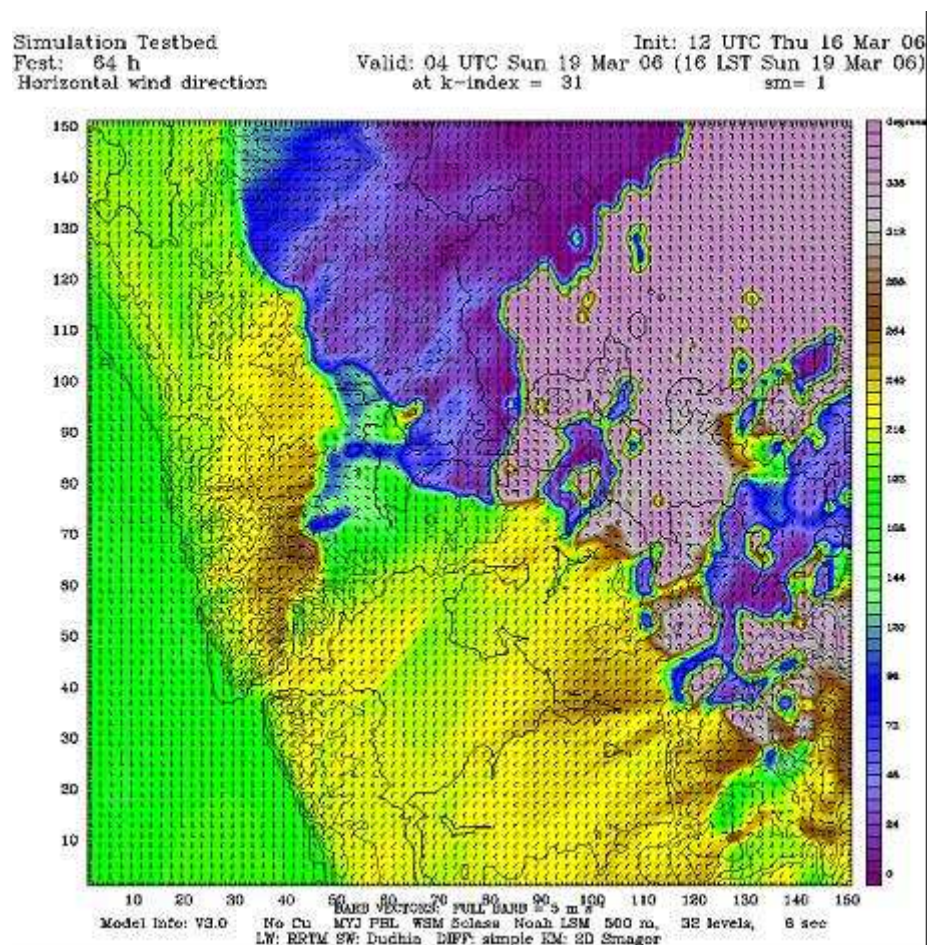


Figure 8.6: Wind vector plot (16:00 hours 19th Mar 2006) showing mature sea breezes on both east and west coast of the Auckland region and further north in the afternoon. To the north of Auckland, the inland penetration of easterly sea breezes on the east-coast is up to 50% of the total width of the land mass. In this region, the easterly sea breezes are forming convergence zone with the opposing westerly sea breezes from the west coast.