
A Review of Regional and Natural Background PM₁₀ in New Zealand

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Contents

Executive Summary	iv
1. Introduction	1
1.1 Scope	1
1.2 Context	1
1.3 Acknowledgements	3
2. What is background?	4
2.1 An overview of definitions	4
2.2 Natural versus anthropogenic	5
2.3 Location-based definitions	5
2.4 Visualising background	7
2.5 Urban background	8
2.6 Definition of background in source apportionment	9
2.7 Agreement on definitions	10
3. Regional background in NZ: summary	11
3.1 Regional background – definition, sources and variability	12
3.2 Sea salt	14
3.3 Soil and dust	16
3.4 Sulphate	17
3.5 Contributions of profiles to background PM ₁₀ and NES exceedences	18
4. Other background PM ₁₀ data sources	20
4.1 Summary estimates	20
4.2 Monitoring data regression	21
4.3 Background monitoring sites	24
4.4 Filtering urban-edge monitoring data	25
4.5 Discussion – comparison of background PM ₁₀ estimates	27
5. Conclusions	30
5.1 General conclusions	30
5.2 Recommendations for future monitoring	31
5.3 Recommendations for future research	32
6. Technical Annex – Review of sources and transport of regional background PM ₁₀	33
6.1 Sea spray	33
6.1.1 Source variability	33
6.1.2 Inland transport & deposition	33

6.1.3	Consequences of sea spray emission and dispersion for background PM ₁₀	34
6.1.4	NZ data and studies	35
6.2	Mineral dust	37
6.2.1	Source areas	37
6.2.2	Trans-Tasman transport	38
6.2.3	New Zealand mineral dust	39
6.3	Other dusts	39
6.3.1	Anthropogenic resuspension of dusts	39
6.3.2	Biogenic dusts	40
6.3.3	Volcanic dust	41
6.4	Intra-national transport	42
6.5	Non-mineral long-range transport	42
6.5.1	Sources of sulphate	42
6.5.2	Source areas of long-range transport to New Zealand	43
6.5.3	Remote and marine background	44
6.6	What are the outstanding questions?	45
6.6.1	The source of the sulphate	45
6.6.2	How to deal with episodic emissions/transport	48
6.6.3	Nitrate, bound water and secondary organic aerosol (SOA)	48
6.6.4	Sea breeze and local recirculation	49
7.	Appendix – Notes from the Background PM Workshops held at NIWA in May 2007	51
8.	References	55

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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 Monitoring and Network Design research objective of this Programme aims to improve PM₁₀ monitoring networks and measurement systems in New Zealand. The outputs from the research will help resource managers and resource users meet the requirements of the NES as effectively as possible.

This report has been prepared subsequent to another FRST report (Wilton *et al.*, 2007), which presents results of PM source apportionment studies recently carried out in NZ.

A recurring question in Air Quality management is how to estimate background or baseline concentrations now and in the future to assess the effects of abatement strategies or new projects.

As part of the research programme we are carrying out monitoring and source apportionment studies of PM₁₀ in collaboration with various partners in order to determine what proportions are coming from which sources. The results of these studies can be used to refine emissions inventories. We are also carrying out modelling studies for various purposes such as interpolation of monitoring data and forecasting of pollutant concentrations for AQ management and resource consent. All these activities require an estimation of the background concentrations to close the gap between monitoring and modelling. For AQ management there is also a need to estimate background as a starting point for straight-line paths to NES compliance.

However, "Background" means different things to different people depending on the context in which it is being used: Urban, rural, remote, natural or anthropogenic? And do we mean background or baseline?

For resource management in NZ, background PM₁₀ is generally used in one of two contexts, model/attainment strategies and consent processing. This Report focuses primarily on the former while providing the foundation for a subsequent report focussing on the latter.

Regional background PM₁₀ may be defined as any PM₁₀ that is natural or is sourced from beyond the regional boundary. Receptor modelling (RM) is particularly useful for quantifying this. If time and/or resources are not available to do RM, a data mining approach could be considered. Techniques for estimating background values for these purposes include simple indicative values included in the MfE good practice guides, regional background monitoring and filtering of monitor data and regression of co-located monitor data.

Source apportionment studies using receptor modelling indicate that in summer, non-anthropogenic sources can constitute up to 80% of the measured PM₁₀ although on high pollution days in winter the value tends to be much lower, e.g. 10-20% in Masterton (Davy., 2007), 15% in Kowhai or 8-10% in Hastings (Wilton *et al.*, 2007).

Other estimates of background such as regression give values for non-combustion sources, which may still be anthropogenic. An example is given of 8 - 10% in Masterton (Xie *et al.*, 2006).

Estimates of long-term average regional background PM₁₀ have been compared from three methods across 5 locations. All but one analysis suggested a value in the range 8 – 12 µg m⁻³, with a higher range (at Pukekohe) of 10 – 14 µg m⁻³ potentially linked to higher wind-driven resuspension in south-westerlies.

Coastal sites can have very high background concentrations due to high sea salt loadings. Marine background measurements of the order of 15 µg m⁻³ have been made at Christchurch and Kaikoura with a strong spatial gradient reported in Christchurch (Clark *et al.*, 2007).

The following recommendations have been made:

Recommendation 1

Report writers (applicants, consultants, researchers, and council staff) should provide definitions of 'Background' or 'Baseline' within their report. This should indicate whether it refers to sources, concentrations or locations. It should indicate what it DOES and DOES NOT include. For example: *"In this report, Background is assumed to mean any natural sources such as sea salt, pollen and biogenic particulates and any wind blown dust (including that arising from human activity) but does not include any other anthropogenic emissions."*

Recommendation 2

Air quality assessment of sites within 5 km of the coast should consider the impact of sea spray. Background PM₁₀ is likely to have a directional dependence due to the influence of wind speed-dependent sea spray in onshore winds. In non-urban areas monitoring data may be filtered between onshore and offshore winds, or by wind direction generally, to identify this variation. Onshore winds may not necessarily be taken as the worst-case, if offshore winds carry high contribution from an inland source.

Recommendation 3

PM₁₀ monitoring sites within 200 m of the coast cannot be considered representative of locations further inland or whole airsheds, nor are they particularly stable due to rapidly changing locally elevated surf-generated sea spray. Such sites should be avoided for the purpose of NES compliance monitoring unless the local contribution of sea salt is explicitly to be included.

Recommendation 4

As a first estimate of long-term average regional background PM₁₀ in New Zealand we find that a value of 10 µg m⁻³ is reasonable at a non-coastal site.

Recommendation 5

Further PM₁₀ monitoring at regional background sites (such as Pongakawa) is highly recommended.

Recommendation 6

Further co-located monitoring of CO with PM₁₀ monitoring is recommended. The use of regression with NO_x or measures of combustion-related particles should be researched.

1. Introduction

1.1 Scope

This report is aimed at air resource users. The key objectives of this report are to

- provide a conceptual framework for background PM, its definitions and uses,
- review the current state of knowledge regarding the sources of background PM in New Zealand,
- provide an overview of previous estimates of background concentrations of PM in New Zealand and compare to estimates from the recent source apportionment studies, and
- present some methods for estimating background in different circumstances.

1.2 Context

“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 Monitoring and Inventories research objective of this Programme aims to improve PM₁₀ monitoring networks and measurement systems in New Zealand. The outputs from the research will help resource managers and resource users meet the requirements of the NES as effectively as possible.

This report has been prepared to support contracted output 3.4.5, which specifically requires;

“ A report or workshop which,

- Defines “background” air pollution and considers vehicle, industry, domestic and natural sources
- Presents an analysis of air quality and source apportionment monitoring data that aims to determine the contribution of background air pollution to urban air quality.
- Provides a method by which background air quality can be estimated for the major urban areas in New Zealand”

The workshop was conducted in Auckland and repeated in Christchurch in May 2007. An overview of the workshops, including lists of attendees, is included in Appendix 1.

It follows an earlier report (Wilton *et al.*, 2007), which presented results of PM source apportionment studies recently carried out in NZ. An early version of this report was submitted to FRST and NESRAG in June 2007. Following reviews by members of NESRAG substantial changes were made. A decision was made late in 2007 to withhold the release of a final version until after the completion and publication of a large source apportionment study (based on analysis of ~ 1400 PM₁₀ and PM_{2.5} filters) commissioned by Auckland Regional Council. This final version has incorporated findings from this study, which was published in May 2008 (Davy *et al.*, 2007).

A recurring question in air quality management is how to estimate background or baseline concentrations now and in the future to assess the effects of abatement strategies or new projects.

Regional Councils are charged with achieving compliance with the PM₁₀ AQNES by 2013. Reducing concentrations inevitably means reducing emissions. However, not all emissions to the atmosphere can be managed, and some emissions occur in one airshed or Region, but impact concentrations in another. **The relative impacts of emission cuts can only be predicted if the contribution to concentrations of other emissions, including natural, non-inventory and non-local, can be quantified and understood.** As the New Zealand AQNES for PM₁₀ specifies an averaging time of 24 hours, and the WHO annual guideline has not been adopted in the Standards, an understanding of the temporal variability of the background is crucial. The key question is **how much do background sources contribute to concentrations on those days when the NES is exceeded?**

This Report focuses on the current state of knowledge regarding background PM₁₀ and some existing and proven means of estimating it. It will inform a follow-up report providing more specific advice on how to determine baselines for industrial Resource Consents to be completed by October 2008. It will also be used as one of the foundations of the next FRST air quality programme: 'Healthy Urban Atmospheres', which will begin in October 2008.

However, "Background" means different things to different people depending on the context in which it is being used: Urban, rural, remote, natural or anthropogenic? And do we mean background or baseline?

1.3 Acknowledgements

We would like to thank Paul Franklin (Ministry for the Environment) and Louise Wickham (Ministry for the Environment) for their helpful reviews of the 2007 draft version, and especially Paul Baynham (Northland Regional Council) for reviewing several versions and providing key advice on the layout of the report. The monitoring data included and analysed in this Report is provided courtesy of Auckland Regional Council, Environment Canterbury and Environment Bay of Plenty.

2. What is background?

- Different uses of the term ‘background’ are discussed.
- For air resource users the most common useful definition is emission sources (and resulting contributions to concentrations) beyond the direct control of a local authority.
- A single definition for background across all contexts is not appropriate.
- Background should be clearly defined in each study/project/report.

2.1 An overview of definitions

The word ‘background’ is frequently used in air quality management – perhaps too frequently, for it has come to mean many different things to different people, and different things to the same person. Therefore any discussion of ‘background’ in the sense of atmospheric contaminants cannot begin until the multiple uses, meanings and nuances of the word are disentangled.

Background can refer to an emission source, an ambient concentration (or contribution to it) or a location. It may or may not include anthropogenic emissions. In general, readers need to be aware that no consensus exists on a single general definition that can be used in all contexts, and it is quite possible that no consensus could ever be reached. Within a specific context however, mutual agreement on definitions is achievable. Thus, careful definition of background is a key requirement in any report or assessment that uses the term. In this report we will present an overview of the many different concepts of background and examine the circumstances where it is most likely to be encountered by air quality resource users in New Zealand.

When defined in terms of emission sources, background can mean ‘natural’ as opposed to anthropogenic (discussed further below). More commonly it can refer to sources not in the emission inventory. This approach is most likely to be taken by Regional Councils in the context of NES compliance. This definition may be taken to mean sources outside of our control (e.g. sea spray), but that is not necessarily the case as in certain Regions non-inventory sources could include shipping or small-scale cooking operations, which *are*, in principle, amenable to control.

Another common use of the word background is to describe emissions from outside the area of concern, i.e. emissions that are not 'local'. For example, if we are managing an airshed, emissions from outside of our airshed are often described as background. If we are considering the whole of New Zealand, then background may refer to emissions external to our country, whether they are continental or oceanic.

Furthermore, background may be defined as '*not foreground*', in other words, emissions or concentrations from any source that pre-exist the source, activity or receptor that we are interested in. This approach is more likely to be taken in the case of Resource Consent or specific environmental impact assessments. This concept is similar to 'baseline' – the differences will be explored further below.

2.2 Natural versus anthropogenic

One definition of background is to align the concept with 'natural' as opposed to anthropogenic emissions, i.e. those emissions that would occur in the absence of human activities. However, this is not as clear-cut as it may at first seem. Mineral dusts can form a significant component of PM₁₀. Whereas much of this dust is resuspended by the wind, animals are also responsible for its resuspension, and for the disturbance of soil crusts, which exposes soil to wind erosion. Although 'soil' may be thought of as a natural source, it is resuspended by agricultural activities, mining, quarrying, track-out on vehicle tyres and resuspension of road dust by vehicles - all human activities.

To correctly quantify the impact of human activities on the atmosphere it is necessary to know what the state of the atmosphere would be without the impact of human sources. However, the measurement of the state of the background (i.e. natural) atmosphere is not an easy task. The main problem is to find a location not affected by human activities. This is further complicated by the fact that after several thousands of years of human activities there is almost no place on the planet that can be viewed as truly background. For instance, measurements in the Arctic show that the particulate matter observed there has a noticeable impact from human activities (Stohl *et al.*, 2006). Furthermore, measurements of greenhouse gases (GHG) in supposedly pristine locations show variations that are *very likely* to be the result of human activities worldwide (IPCC, 2007).

2.3 Location-based definitions

Background may refer to a location where monitoring may be conducted, or the composition of the air sampled there. Sub-definitions include:

Urban background. Considering that one of the main objectives of air quality measurement in urban areas is to know the population exposure to potentially dangerous concentrations, it is useful to have measurements that are representative of a large urban area (Harrison *et al.*, 1999). Therefore, even though it is not a proper background, the concept of urban background is widely used to describe the mean concentration of pollutants in urban areas. Furthermore, the concept of urban background has been overused so that there are several different concepts that fall within the definition of urban background:

- Industrial background. Location where industrial emissions are dominant.
- Residential background. Location far from industrial sources and mainly dedicated to residential activities.
- Suburban. Location far from commercial centres and industrial sources but dominated by residential activities.
- Urban centre. Location deep in the city but distanced from roads, where people spend significant time during the day.

Rural background. This is generally defined as referring to typical concentrations in rural areas well removed from urban centres and not subject to impacts from point source discharges. Contributions to rural background include pollen, wind blown dust fertiliser application and occasionally smoke from rural burnoffs. A definition of “rural background” may be relevant when considering resource consent applications to discharge to air in predominantly rural locations.

Continental background. This is the land based equivalent to the marine background. It is defined as the concentration over large extensions of land far from the coast and human activities. However, the different types of climates in continental areas and the different vegetation or soil composition means that background measurements are dependent on the location of the measurement (Laakso *et al.*, 2003).

Marine background. It is understood as the composition of the atmosphere over large extensions of salt water (oceans) not disturbed by landmasses or ship tracks in the vicinity. This is one of the most difficult background concentrations to measure because it requires the location of a sensor in open seas in a platform as small as possible. Alternatively small islands or exposed coastal sites can be used for such measurements (McGovern *et al* 1994).

Polar background. Supposedly the simplest of the background definitions, the difficulty of setting a measurement site that does not disturb the background makes these measurements logistically difficult.

2.4 Visualising background

Measurements at a given point can be conceptualised as representing the sum of contributions from different sources in different locations. Figure 1 below illustrates this emission-source based concept. Inventory sources will make up different proportions of each box.

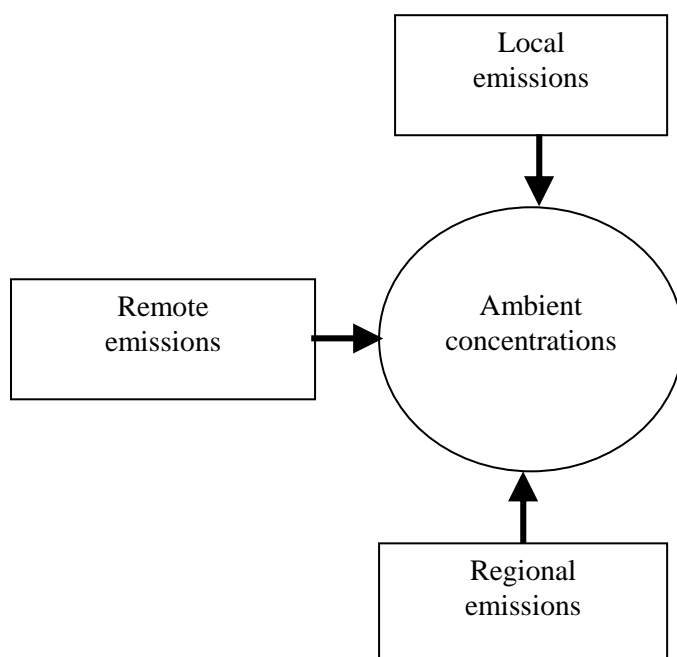


Figure 1. The different scales of source emissions contributing to measured ambient concentrations

This can also be visualised as a hierarchy of layers (Figure 2) with concentration represented by height and location represented horizontally. If we define background spatially then it applies to either just the remote emissions (if our zone of interest is the country), or remote plus regional emissions (if we are considering a region or airshed only). If we define background by emission source (i.e. natural sources, or non-

inventory sources) then each of our boxes partially contributes to background, although the proportion of background differs.

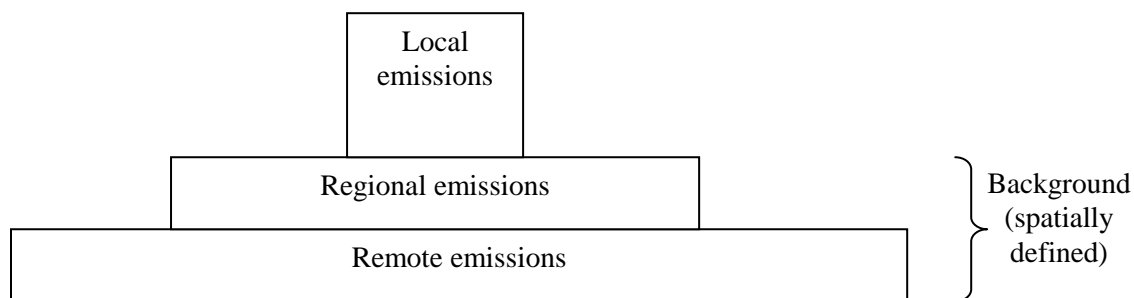


Figure 2. A hierarchy of background source regions

2.5 Urban background

In urban areas the picture becomes slightly more complicated and it is here, particularly, that definitions tend to diverge. Concentrations at an urban roadside consist of the layers illustrated in Figure 3.

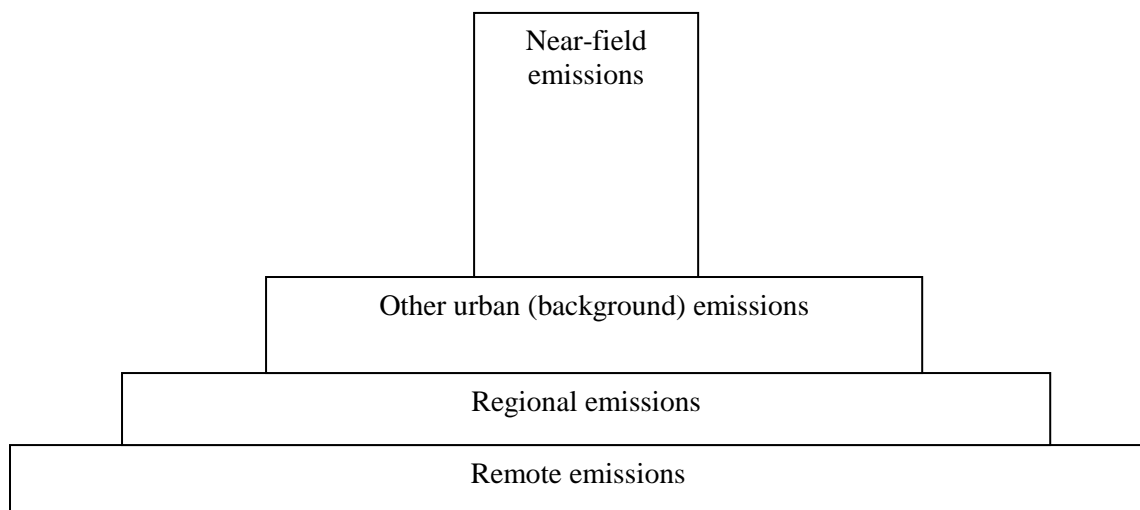


Figure 3. A hierarchy of contributions of source regions to urban concentrations. Some confusion may arise when describing concentrations, as the term ‘urban background’ is sometimes applied to the third layer alone, but also to the sum of the lower three layers.

When defined in terms of emissions, ‘urban background’ describes the third layer in Figure 3 i.e. urban emissions other than those near-field sources causing an immediate impact at any given site.

In terms of locations, ‘urban background’ any urban location far enough from any locally dominating near-field source such that the contribution of that source (e.g. a road, stack, etc) cannot be distinguished due to the mixing of its emissions with other urban emissions.

In terms of concentrations, ‘urban background’ can mean one of two things:

- either the concentration at an urban background location (i.e. the sum of the lower three layers in Figure 3), or
- the contribution to concentration due to urban background emissions only (i.e. the third layer only in Figure 3).

Both definitions are in common use.

2.6 Definition of background in source apportionment

The new Hastings and Auckland source apportionment studies reported in 2007 and discussed below deconstruct urban PM_{10} and $PM_{2.5}$ samples into 5 sources (two sources were described differently in the 5-site Auckland study as noted below in parentheses):

- Motor vehicles
- Domestic heating (biomass burning)
- Sulphate
- Sea spray (marine aerosol)
- Soil

Sea spray and soil are classed as ‘background’. This satisfies the source-based definition in that they do not appear in emission inventories. Sea spray is formed in the absence of any human activity, but we have noted above how human activity contributes significantly to soil resuspension. Whether these components are ‘background’ in the geographical sense (i.e. not locally emitted) is less clear.

The origin of sulphate is even less clear. Natural sources include oceanic dimethyl sulphide and volcanic activity. This component fits most definitions of background. However, estimates of the precursors of most anthropogenic sulphate sources (i.e. SO₂ sources) appear in emissions inventories (combustion, especially industrially or for power generation, but also domestic), so is this background? From the spatial point of view the sulphate may have trans-oceanic origins making it part of the background, but intra-national and even intra-airshed sources cannot be ruled out at this stage, so that some of the sulphate in a given airshed may be local while the majority is likely to be regional. Thus much sulphate may be background from the point of view of a Regional Council that cannot control its emission, but it is anthropogenic and thus amenable to control in its airshed of origin.

2.7 Agreement on definitions

Two focus groups were held to help assess the current state of understanding and priorities for the future of the subject of background concentrations of PM₁₀ in New Zealand. It was appreciated by all that the definition of background varied with the use to which it was being put and although it would be useful for everyone to agree on a standard definition, or set of definitions, the word is so entrenched in each context that it would be hard to change. Suggestions of modifiers such as “Natural background” or “Urban background” were made but they were thought unlikely to change things. Therefore it is probably simpler for it to be defined by the user depending on the context.

Recommendation 1

Report writers (applicants, consultants, researchers, and council staff) should provide definitions of ‘Background’ or ‘Baseline’ within their report. This should indicate whether it refers to sources, concentrations or locations. It should indicate what it DOES and DOES NOT include. For example: *“In this report, Background is assumed to mean any natural sources such as sea salt, pollen and biogenic particulates and any wind blown dust (including that arising from human activity) but does not include any other anthropogenic emissions.”*

3. Regional background in NZ: summary

- The current state of knowledge regarding the sources of background PM₁₀ in New Zealand is briefly reviewed. A more thorough discussion of the science is present in the Technical Annex.
- Sources are considered roughly in order of decreasing significance.
- Sea salt is found to be highly significant, especially considering NZ's high coastal population. Concentrations are strongly elevated within a few hundred metres of the coast, but the elevation is sensitive to wind speed and humidity and changes rapidly. Monitoring data from sites within 1 km of the coast should be interpreted with care.
- As a first approximation sea salt concentrations halve between 1 km and 50 km from the coast, but this is also highly dependent on variations in wind speed across that distance.
- A recent study of the transport of fine desert dust from Australia suggested it contributed $\sim 5 \mu\text{g m}^{-3}$ to PM₁₀ in New Zealand. This transport peaks in autumn, whereas transport through spring and summer tends to become more sporadic leading to potentially larger but much more episodic concentrations.
- Numerous natural, agricultural and other anthropogenic activities (such as quarrying, mining and construction) crush soils and minerals into particles below 10 μm , and also resuspend them into the atmosphere. Effects are highly variable and mostly localised, and thus very difficult to predict. Vehicle 'trackout' can transport this material away from the source area.
- Long-range transport of aged anthropogenic emissions (sulphates and nitrates) principally from Australia occurs sporadically. The east coast of South Island is relatively protected by precipitation scavenging over the Southern Alps.
- Natural marine and remote sources make a small contribution to PM₁₀ ($\sim 0.5 \mu\text{g m}^{-3}$). The natural marine source dominates and varies seasonally, peaking in summer.
- Information is incomplete and many contributions remain unquantified. Some sources are better understood than others.

3.1 Regional background – definition, sources and variability

This section of the report reviews the current state of knowledge regarding regional background PM₁₀ in New Zealand. It considers two types of sources: anthropogenic sources that are transported into an airshed from beyond its boundaries, including those derived from gaseous precursors, and natural sources both imported and local. It does not include local anthropogenic emissions (whether included in inventories or not).

This section summarises the findings of a review which is found in the Technical Annex. The key sources of information are a literature review and the source apportionment (receptor modelling) studies conducted in Hastings and Auckland. The Hastings results and initial results from the Kingsland (*aka* Kowhai) site in Auckland were reported in Wilton *et al.* (2007). The Kingsland data, however, forms part of a larger 5-site Auckland study commissioned and funded by Auckland Regional Council and reported by Davy *et al.* (2007). In both Auckland and Hastings, five sources of PM₁₀ were identified: domestic home heating, sea spray, motor vehicles, sulphate and soil. Sea spray and soil were described as ‘background’ although it was acknowledged that sulphate from natural sources could contribute to background.

The Technical Annex reviews our current state of understanding of regional background PM₁₀ in New Zealand, disaggregated into the main compositional components. The relative significance of these components varies between locations. In general, however, Table 1 lists the principal persistent components of NZ’s regional background PM₁₀ with some estimates of their contribution. In terms of New Zealand as a whole the regional background is probably dominated by long-range transport of anthropogenic fine particles emitted both in New Zealand and in Australia, and fine dust from Australia. However, most of the population lives near the coast, and in coastal locations it appears that sea spray is likely to be the dominant component.

The review of the various contributions to regional background above have identified that some components display more spatial and temporal variability than others. Some such components may be of lower significance in the long-term but can lead to high concentrations on a localised and/or episodic basis (e.g. desert or volcanic dusts). As a general rule coarse particles are more variable than fine particles. This calls into question the validity of any long-term average concentration, and the spatial representativeness of any measured, modelled or estimated concentration. Table 2 lists sources of short-term episodic peaks in the regional background.

Several background components have seasonal cycles. Sea spray and local soil resuspension is driven by wind and the impacted footprint will depend on wind

direction, both of which are subject to seasonal climatic variation such that the season of local peaks will depend upon the local climate. We have reviewed how desert dust emission and long-range transport trajectories vary seasonally, as does sulphate emission arising from phytoplankton blooming.

Table 1: typical source contributions to long-term average PM₁₀ concentrations in New Zealand.

Component	Typical long-term average concentrations / $\mu\text{g m}^{-3}$	Data Source
Sea salt	0.5 – 1.0 (> 20 km inland) 1 - 15 (near coast)	Kaikoura monitoring, Clark <i>et al.</i> (2007), Wilton <i>et al.</i> (2007), Davy <i>et al.</i> (2007), Cole <i>et al.</i> , 2003
Soils and fine mineral dust	1 – 2 ~ 5 (indirect estimate based on radioactivity measurements)	Wilton <i>et al.</i> (2007), Davy <i>et al.</i> (2007), Marx <i>et al.</i> , (2005)
Biogenic secondary organic aerosol	Unknown (1 – 2 in UK)	Jones & Harrison (2006)
Anthropogenic secondary PM	2 – 4 (no data separating NZ and international sources)	Wilton <i>et al.</i> (2007)
Oceanic sulphate	Up to 0.4	Allen <i>et al.</i> (1997)
Remote background aerosol	0.1	Allen <i>et al.</i> (1997), Nyeki <i>et al.</i> (2005)

Table 2: typical source contributions to short-term PM₁₀ concentrations in New Zealand.

Component	Typical short-term concentrations / $\mu\text{g m}^{-3}$	Data Source
Sea salt	2 - 20 (in high winds) Up to ~ 100 (highly exposed site)	Wilton <i>et al.</i> (2007), Davy <i>et al.</i> (2007), Kocak <i>et al.</i> 2007
Local soils and dust	unknown	
Coarse Australian desert dust	Up to 30	Marx <i>et al.</i> (2005)
Volcanic dust	unknown	
Biogenic dusts	unknown	

On a short-term basis, however, of the order of weeks, days or even hours, other sources can become dominant. This is particularly the case for sea spray and the transport of coarser dust suspended in dust storms in Australia. However, the concentration of finer Australian dusts also varies on this scale due to variability in trajectories and the temporal and spatial variability of precipitation scavenging.

These spatial and temporal relationships are summarised in Table 3.

Table 3. Summary of the temporal and spatial variability in a range of contributors to regional background in New Zealand.

	constant	Regular (seasonal)	episodic
Spatially even	Remote background nss-sulphate	Oceanic nss-sulphate (summer peak), LRT from SE Australia (summer peak), LRT from western Australia (winter peak)	LRT from Australia
Regionally variable		Fine Australian dusts (spring and autumn peaks)	Fine & Coarse Australian dusts
localised		pollen	Sea spray, volcanic dust, forest fires, local dusts

3.2 Sea salt

Over 2.5 million New Zealanders live in town and cities within 20 km of the coast. Sea spray is generated by two mechanisms. On the large-scale the most significant is from breaking whitecaps in open ocean leading to PM₁₀ concentrations above the south Pacific of typically 10 – 20 µg m⁻³ (Grini *et al.*, 2002). Upon transport over land, sea salt is deposited and removed from the atmosphere leading to a rapidly decreasing concentration in the first ~5 km from the coast, beyond which the gradient flattens out. Further inland concentrations are dependent upon climate, topography and land-use. Typically, concentrations in the 5 km coastal band are elevated by 30 – 100 % compared to inland.

Within a few hundred metres of the coast generation and short-scale transport of larger spray droplets from surf breaking dominates and concentrations can be elevated by an order of magnitude compare to inland. Here long-term elevations of several $\mu\text{g m}^{-3}$ are likely with short-term peaks of tens of $\mu\text{g m}^{-3}$ associated with high winds. Specific quantification is acutely dependent upon local factors – principally wind speed, but also the potential for surf production, land-use, climate and fetch limitations (e.g. sheltering from offshore islands), thus preventing a generalised quantification without further investigation. Although peak values well in excess of $20 \mu\text{g m}^{-3}$ may occur these are strongly dependent upon wind speed and are thus rarely sustained for more than a few hours, except in very exposed locations.

Several particle compositional analysis studies in Auckland in the first half of the current decade identified sea salt as the primary component of PM_{10} by mass (Wang & Shooter, 2001, Wang *et al.*, 2005, Wang & Shooter, 2005), predominantly in the coarse mode. The recent source apportionment data showed that sea salt was the dominant source at in Hastings except in winter, was the dominant source across Auckland in summer, and was a comparable source to motor vehicles and biomass burning in spring and autumn. The time series of reconstituted sea salt contribution to PM_{10} in Hastings illustrated no clear seasonal pattern and there was an apparent randomness in the sea salt signal, which may be expected due to its sensitive dependence on wind speed and many other factors. This randomness in the real world must be borne in mind when long-term average or ‘typical’ values are sought or used. Despite the strong traffic influence at Khyber Pass Road, the 2006 source apportionment study attributed peak summer PM_{10} concentrations there to marine aerosol.

Table 4: average sea spray concentrations in the recent source apportionment studies.

	Hastings	Kingsland	Queen Street	Takapuna	Khyber Pass Rd	Penrose
Annual mean sea spray PM_{10} / $\mu\text{g m}^{-3}$	3.9	5.5	4.7	5.0	5.4	5.4
Shortest distance to coast / km	8	5	<1	2	3	7

The interquartile range in the sea salt contribution to PM_{10} at Hastings was $1.7 - 5.2 \mu\text{g m}^{-3}$. This contribution exceeded $10 \mu\text{g m}^{-3}$ on 5 out of 121 days, peaking at $20 \mu\text{g m}^{-3}$.

Despite sea salt's large contribution to PM_{10} the results of this study generally indicated that peak PM_{10} values across Auckland are currently related primarily to high anthropogenic emission and poor dispersion, i.e. low winds – just the kind of conditions in which we would expect the sea salt contribution to be minimal.

Recommendation 2

Air quality assessment of sites within 5 km of the coast should consider the impact of sea spray. Background PM_{10} is likely to have a directional dependence due to the influence of wind speed-dependent sea spray in onshore winds. In non-urban areas monitoring data may be filtered between onshore and offshore winds or by wind direction generally, to identify this variation. Onshore winds may not necessarily be taken as the worst-case, if offshore winds carry high contribution from an inland source.

Recommendation 3

PM_{10} monitoring sites within 200 m of the coast cannot be considered representative of locations further inland, or whole airsheds, nor are they particularly stable due to rapidly changing locally elevated surf-generated sea spray. Such sites should be avoided for the purpose of NES compliance monitoring unless the local contribution of sea salt is explicitly to be included.

3.3 Soil and dust

There is minimal data in New Zealand (and limited data globally) regarding the contribution of soils and fine mineral dust. Numerous natural, agricultural and other anthropogenic activities (such as quarrying, mining and construction) crush soils and minerals into particles below $10\ \mu\text{m}$, and also resuspend them into the atmosphere. Effects are highly variable and mostly localised, and thus very difficult to predict.

Vehicle ‘trackout’ can transport this material away from the source area. A recent study has suggested a less sporadic long-term transport of fine dust from Australia contributing $\sim 5 \mu\text{g m}^{-3}$ to PM_{10} in New Zealand, but this cannot be verified by direct measurement.

Soil profiles were reported for the source apportionment studies with mean concentrations of $1.8 \mu\text{g m}^{-3}$ (interquartile range of $0.5 - 2.5 \mu\text{g m}^{-3}$) at Hastings. There was a weak seasonal variation at Hastings with maximum contribution in spring and minimum in winter. The 2006 Auckland study reported mean PM_{10} concentrations from $1 - 2 \mu\text{g m}^{-3}$. A ‘construction’ profile was identified at Queen Street

3.4 Sulphate

Long-range transport of aged anthropogenic emissions (sulphates and nitrates) principally from Australia occurs sporadically. The east coast of South Island is relatively protected by precipitation scavenging over the Southern Alps. There is not yet any study or data describing the impact of precursor emissions in one New Zealand airshed contributing to secondary PM_{10} in another airshed, thus the significance of this pathway is unknown.

Lesser sources of sulphate include a natural oceanic source arising from the precursor emission of dimethyl sulphide by phytoplankton. This emission is seasonal and is stronger at lower latitudes. Concentrations in New Zealand are likely to peak in summer and in northerly winds at around $0.4 \mu\text{g m}^{-3}$.

An underlying well-mixed aged remote background aerosol based on sulphate, black carbon and complex aged organic compounds is present at all times with little temporal or spatial variation. The limited data available suggest that it contributes approximately $0.1 \mu\text{g m}^{-3}$.

Several studies have suggested that secondary PM_{10} may form from locally emitted precursors, but this remains to be confirmed. The Hastings source apportionment report acknowledged uncertainty over the origin of the sulphate, considering local industrial and traffic sources as one scenario (including direct sulphate emissions from fertilizer manufacture), or remote background sources (sea spray, DMS) as an alternative, but in either case the contribution was sufficiently small in comparison to other local sources that its control may not be the priority.

Concentrations of sulphate identified by receptor modelling were significantly higher at Kowhai than at Hastings (table 5), which may suggest the importance of local secondary formation.

Table 5: annual mean sulphate contribution to PM₁₀ from source apportionment studies

	Hastings	Kowhai
Annual mean sulphate PM ₁₀	0.8 µg m ⁻³	2.1 µg m ⁻³

The 2006 Auckland study found average values for sulphate across the 5 sites ranging from 1.3 – 1.6 µg m⁻³. This study was able to resolve non-sea-salt sulphate independently of total sulphate, which averaged 0.76 – 0.81 µg m⁻³ and 1.0 at the industry-influenced Penrose site. Concentrations peaked at all sites in the summer.

3.5 Contributions of profiles to background PM₁₀ and NES exceedences

Counting sea salt and soil as background, the contribution of background aerosol to PM₁₀ in Hastings in winter was estimated to be between 13 % and 15 %. However, on days when the NES was exceeded, it was estimated that background accounted for just 8 % of PM₁₀.

In Hastings, on days when the reconstructed total PM₁₀ exceeded 50 µg m⁻³, the mean sea salt, soil and sulphate contributions were 3.1, 1.9 and 1.0 µg m⁻³ respectively.

In Kowhai, if sulphate is included in the background estimate, the average background contribution was 59 %, ranging from 31 % in winter to 75 % in summer.

The total contribution of marine aerosol and ‘background’ (the sum of marine aerosol, sulphate and soil – which will include both anthropogenic and natural sources) to PM₁₀ in the 2006 Auckland study is shown in table 6. It should be noted that the Kingsland site is relatively central and residential compared to the other sites which are more representative of localised sources (principally traffic). Thus, Kingsland better fits the description of an ‘urban background site’ as defined in chapter 2, and represents a much larger spatial area. The lower background source contributions at other sites reflects higher contribution from other, local sources.

Table 6: Total contribution of marine aerosol and ‘background’ (the sum of marine aerosol, sulphate and soil) to PM₁₀ in the 2006 Auckland study.

	Marine aerosol	background
Kingsland	46 %	55 %
Takapuna	36 %	52 %
Queen Street	20 %	28 %
Khyber Pass Road	29 %	42 %

There were no exceedences of the NES recorded at Kowhai during the sampling period but two high concentration ($>30 \mu\text{g m}^{-3}$) PM₁₀ events were examined by Wilton *et al.* (2007), one in winter, one in summer. The winter event was dominated by ‘biomass burning’ (attributed to domestic heating) which contributed some 73 % of the total. Background sources, including sulphate contributed 15 % of the total. The summer event, in contrast was dominated by natural sources - a north-easterly wind brought a marine aerosol with sea salt and sulphate contributing 58 % and 29 % respectively.

No exceedences of the PM₁₀ NES were observed during filter sample days at the 5 sites in the 2006 Auckland study. The Regional Air Quality Target (RAQT) for PM_{2.5} was exceeded on 8 occasions at Queen Street. On one occasion, in January, marine aerosol contributed 66 % and sulphate 12 %. On an event in June, marine aerosol contributed 14 % and sulphate 4 %. On the other 6 occasions background sources were negligible. The RAQT for PM_{2.5} was exceeded 7 times at Khyber Pass Road, which is more distant from the coast than Queen Street. On no occasion was marine aerosol a significant contributor. Sulphate made a small, but potentially significant contribution ($> 10 \%$) on 4 out of the 7 occasions, peaking at 32 % on an exceedence in September. Neither marine aerosol nor sulphate were significant on the 7 days during which the RAQT for PM_{2.5} was exceeded at Penrose.

4. Other background PM₁₀ data sources

- Three alternative sources of background PM₁₀ data are presented: previously published summary estimates, regression of monitoring data and background monitoring site data (including filtered urban-edge data).
- Although a range of estimates are apparent, most of the approaches tend towards long-term averages of 8 – 10 µg m⁻³ in non-coastal locations.

4.1 Summary estimates

The draft *Good Practice Guide on Assessing Discharges to Air from Industry* (MfE, 2006) provides a table of indicative ‘worst-case’ background concentrations for use in urban and rural areas. For rural areas a single PM₁₀ value of **15 µg m⁻³** applicable to the whole country is suggested based on maximum values in monitoring data where no major upwind sources are identified. However, it must be remembered that this is a screening approach and this value may be excessively conservative in many cases.

The FRST Programme report: ‘*Straight and curved line paths (SLiPs & CLiPs): Developing the targets and predicting the compliance*’ (Fisher, *et al.*, 2005) recommended the use of a value of **10 µg m⁻³** in the absence of more specific local information:

“The focus of the Regulations is on “worst” cases. On this basis, opting for a background of 10 µg m⁻³ is considered appropriate. Concentrations could be slightly higher or lower, depending on the region. Background could easily be a lot higher in specific one-off circumstances, such as bush fires – but this is accounted for in the one allowable exceedence in the Regulations – so this aspect is not considered further.

The actual peak background PM₁₀ concentration anywhere in New Zealand is very unlikely to be much less than 5 µg m⁻³ (except perhaps in the Central Otago high country), and also very unlikely to be much greater than 15 µg m⁻³ (except in areas that are very close to exposed coasts following a very windy period, or are very dusty, or are severely affected by pollen).”

In the recently completed HAPiNZ project (Fisher *et al.*, 2007) long-term average background concentrations were estimated for the major urban areas based on an

analysis of 10 – 12 monitors on urban fringes and a further 10 short-term measurement campaigns. Six bands were presented, as shown in Table 7.

Table 7. Long term average background concentrations for six locations in NZ (source Fisher *et al* 2007)

Category	example	Background PM ₁₀ / $\mu\text{g m}^{-3}$
Inland (low population density)	Masterton	2
Urban flat	Most of Christchurch & Auckland	4
Urban valley	Wellington	6
Coast – not exposed	Gisborne	2
Coast – exposed	Nelson	8
Coast – highly exposed	Kaikoura	16

4.2 Monitoring data regression

Where monitoring data is available, a simple approach to estimating the long-term background contribution to PM₁₀ in an urban area may be provided by plotting PM₁₀ concentrations against CO. The approach is based upon the assumption that local emissions dominate over non-local and that local anthropogenic emissions are principally those due to common combustion sources. Thus the non-background contributions to PM₁₀ and CO will rise and fall in proportion such that when one is plotted against the other the points may tend towards a straight line with an intercept corresponding to the long-term average background PM₁₀, strictly that component of PM₁₀ that is not related to CO sources.

An example is provided of data from Masterton in Figure 4 (from Xie *et al.*, 2006). The intercept represents the average PM₁₀ concentration when there are no combustion contributions. i.e., zero CO concentration. This results in an average background PM₁₀ concentration of $8.8 \pm 0.9 \mu\text{g m}^{-3}$ (the uncertainty is the 95% confidence interval), which accounts for contributions from sea salt, windblown dust and secondary particulate from May-August 2004 (i.e. winter only).

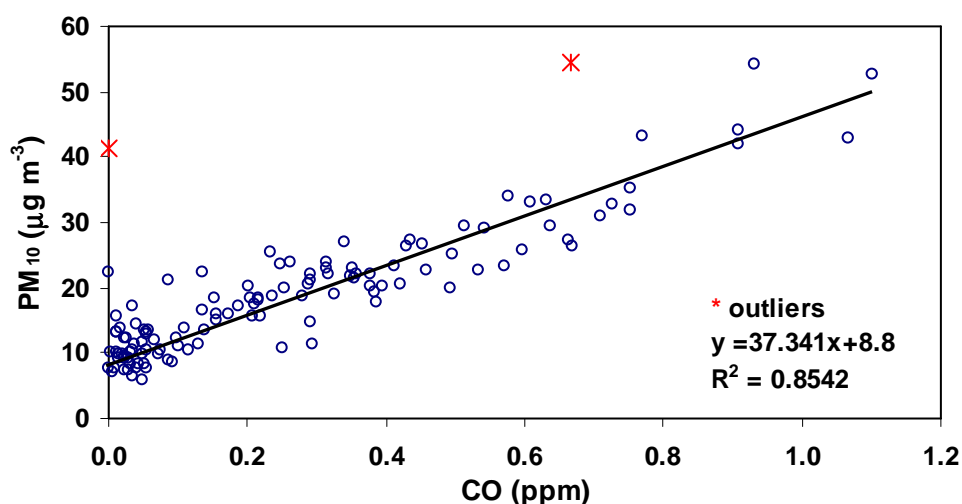
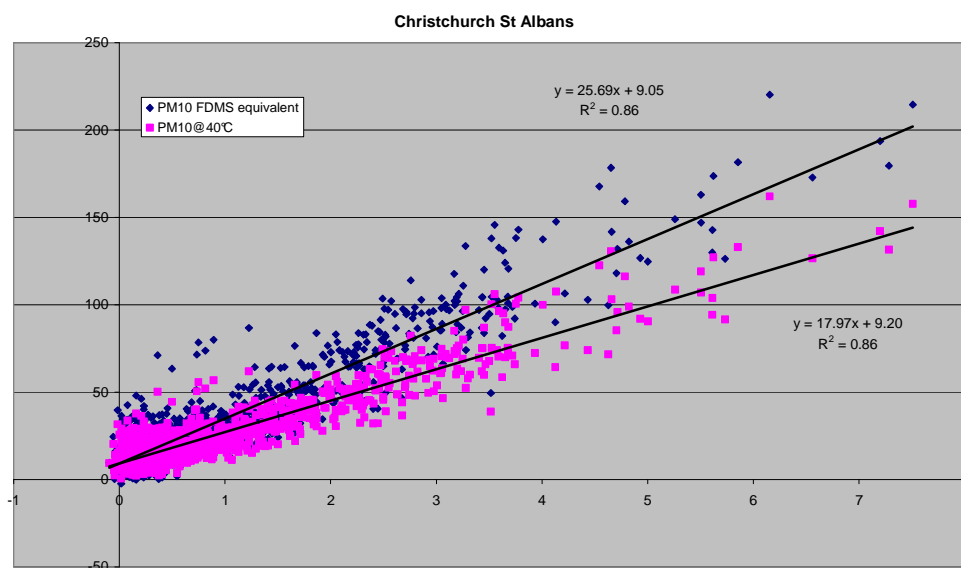


Figure 4. Linear reduced major axis regression of 24-hour average CO against PM₁₀ for Masterton for May-August 2004. Two outliers, identified as studentized residuals greater than 4.0 or smaller than -4.0, are excluded.

Figure 5 (a-c) (provided by Environment Canterbury) displays 24 hour average PM₁₀ versus CO from monitoring data in three Canterbury towns. Estimated background PM₁₀ is 9.0 – 9.2 μg m⁻³ in Christchurch, 11 – 11.5 μg m⁻³ in Ashburton and 11.3 – 11.9 μg m⁻³ in Timaru.



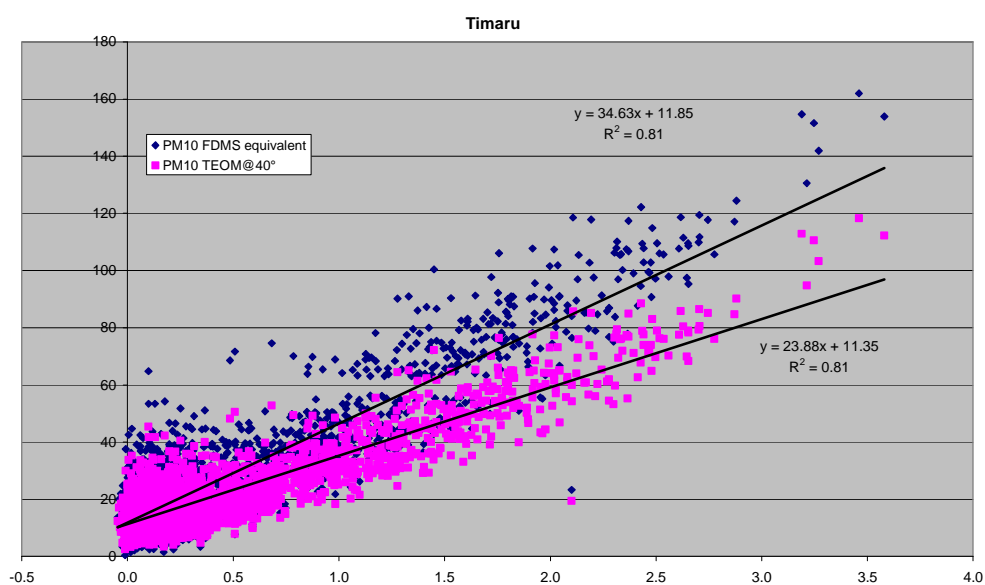
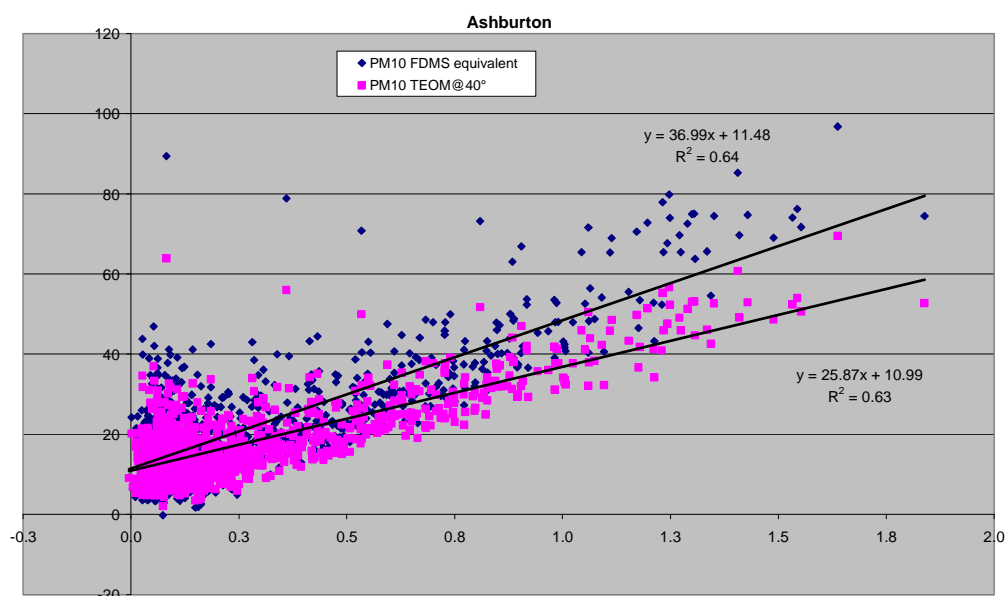


Figure 5: Linear regression of 24-hour average PM₁₀ against CO for a) Christchurch, b) Ashburton, c) Timaru from 1995 to 2005 (supplied by Environment Canterbury).

The slope of these lines should represent the relative contribution of domestic over traffic sources. This relationship is less likely to be linear in areas where non-combustion sources of PM₁₀ are significant, especially those with wind-driven sources. Natural aerosols can come from biomass burning but these tend to be episodic. Low concentrations of CO and combustion-sourced PM₁₀ occur in high

winds, but such winds will increase the emission and transport of sea spray and dust from soils, and will keep resuspended road dusts in the atmosphere increasing their ambient concentration. CO concentrations are also generally low relative to the sensitivity of conventional instrumentation. Thus we may expect this method to be less successful in windy, dusty, coastal or low locations with low CO concentrations.

4.3 Background monitoring sites

New Zealand has only one fixed PM₁₀ monitoring site which is in a genuinely regional background location, i.e. in a non-urban location unaffected by local sources. This is the Pongakawa site operated by Environment Bay of Plenty. PM₁₀ has been monitored at this site since 1997. This site is in open fields, 10 km from the coast and 35 km from both Tauranga and Rotorua. A summary of observations at this site is presented in table 8. In recent years annual mean concentrations have tended to a value around **9 µg m⁻³**. We are unable to state without further analysis whether the falling trend over the first six years of monitoring was due to reductions in natural or anthropogenic emissions. Over the entire dataset from December 1997 – September 2007 (inclusive), the 95th percentile was 20 µg m⁻³.

Table 8: summary of PM₁₀ concentrations (µg m⁻³) observed at the regional background site at Pongakawa (data courtesy of Environment Bay of Plenty).

Year	Median	Mean	Max
1997	13.4	13.3	17.5
1998	13.0	12.1	44.5
1999	9.9	11.2	27.6
2000	11.0	13.1	44.8
2001	10.5	11.5	49.1
2002	9.7	10.0	25.0
2003	6.8	8.1	30.8
2004	9.1	9.6	21.6
2005	7.3	8.7	32.1
2006	7.9	9.2	22.9

4.4 Filtering urban-edge monitoring data

In a very limited number of locations (see Table 9) monitoring data may be available from locations which are only partially affected by local urban emissions and which may be used to give some indicative information about regional backgrounds. This is the case for sites on the urban periphery which detect urban emissions in a restricted arc of wind directions. In this case the PM₁₀ data should be filtered according to wind direction. This is an imperfect approach and the following considerations should be applied:

- apply wind direction data that is as representative as possible of the monitoring site.
- attempt to identify wind directions, or meteorological conditions in which air flow at the PM₁₀ and meteorological monitoring sites are decoupled, or when local flows dominate, for example drainage flows in a local basin. Depending upon the scale of the effect periods in which these conditions occur may need to be filtered out.

Table 9: A non-exhaustive list of some urban-edge PM₁₀ monitoring sites

Region	Site	Measurements	notes
Northland	'Marsden Point'	PM ₁₀ , SO ₂	E of Whangarei and Marsden power station
Auckland	Pukekohe	PM ₁₀ , NO _x , O ₃	See below
	Kumeu	PM ₁₀	road-affected, especially on south side – use with care
Gisborne	Gisborne Airport	PM ₁₀	On W edge of Gisborne
Marlborough	Brooklyn Drive*	PM ₁₀	On E edge of Blenheim

* short-term site.

The Pukekohe (Cronin Road) site is operated by Auckland Regional Council. It is in a rural site approximately 2 km from the western edge of the town of Pukekohe (pop. 21,500), 40 km south of central Auckland (see Figures 6 & 7). Although ozone

measurements have been made at the site for many years, PM₁₀ monitoring began in 2005 and NO_x in 2006. Over the period for which PM₁₀ data exists (until the end of 2006) meteorological observations have indicated three clearly predominant wind directions. In one of those directions (95 – 125°) the Pukekohe site is downwind of Pukekohe town and that data has been removed from this analysis. The directions, the corresponding mean 24 hr PM₁₀ in each direction, and the mean wind speed as measured at Pukekohe are presented in Table 10. It can be seen that PM₁₀ is significantly higher in WSW winds, for which there are no significant upwind urban sources. The occurrence of higher winds in this direction suggests that this increase is due to wind-driven resuspension of coarse particles (most likely sea salt and/or soil).

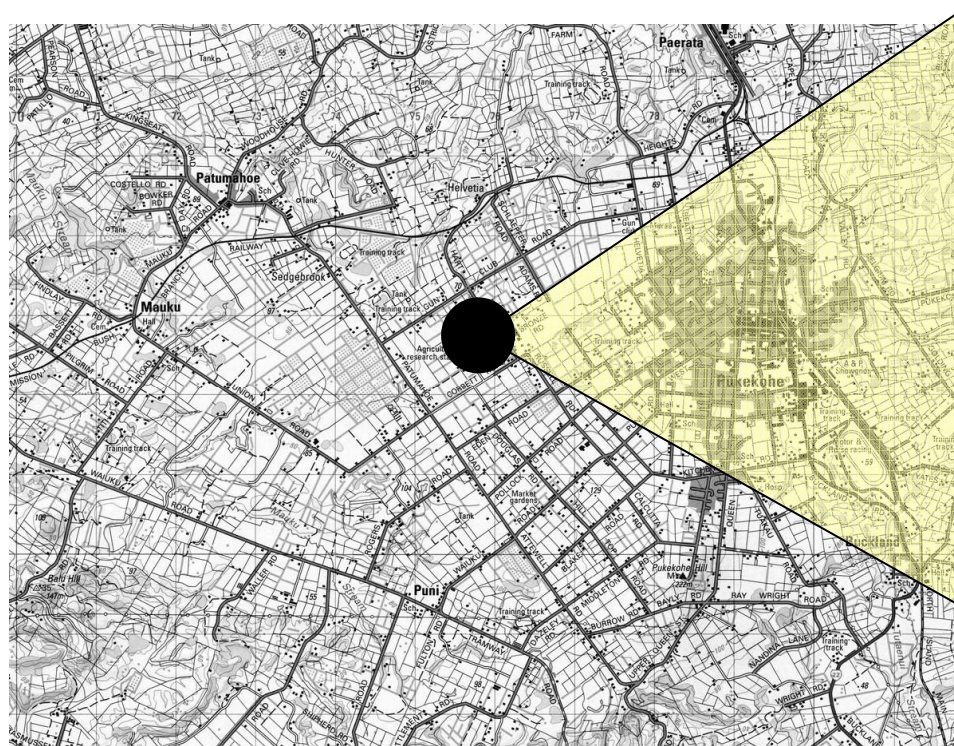


Figure 6: Location of Pukekohe site (black circle) and wind sector for which the site is downwind of Pukekohe town (shaded)

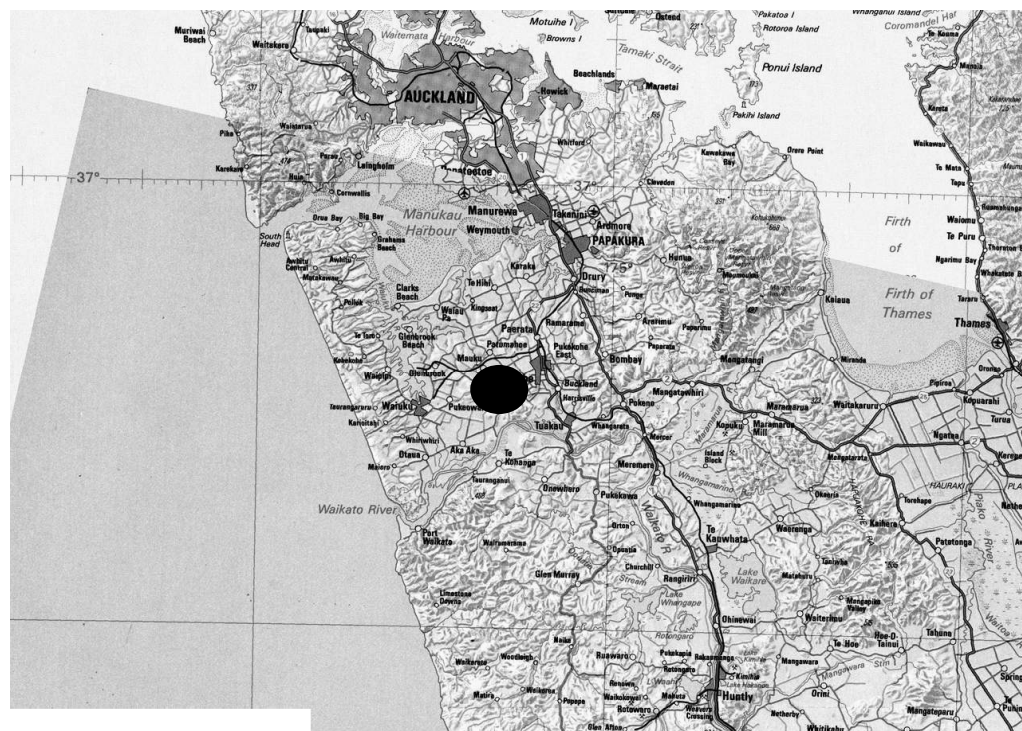


Figure 7: Location of Pukekohe site (black circle) on large scale.

Table 10: Mean PM_{10} and wind speed in the three predominant wind directions at the Pukekohe monitoring site (ARC) with periods when the site is downwind of Pukekohe town removed.

Wind sector	Mean PM_{10} / $\mu g m^{-3}$	Mean wind speed* / $m s^{-1}$	notes
WSW (215 – 285°)	14.1	2.9	
NE (5 – 55°)	10.4	1.9	Auckland & Papakura upwind
SE (125 – 145°)	10.3	2.5	

*mean wind speed as recorded on 10 m meteorological mast at the monitoring site.

4.5 Discussion – comparison of background PM_{10} estimates

The estimate of mean background PM_{10} derived by data regression for Masterton ($8.8 \mu g m^{-3}$) is considerably larger than that suggested by the HAPiNZ summary table ($2 \mu g m^{-3}$) suggesting weaknesses in the HAPiNZ values. The Pongakawa dataset (10 km from the coast) shows that 24 hour concentrations have exceeded $20 \mu g m^{-3}$ on 5 % of

occasions, whereas the end-user guidance report (Fisher *et al.*, 2005) suggested background concentrations are very unlikely to exceed $15 \mu\text{g m}^{-3}$ at non-coastal locations. The significance of such high peak background concentrations is dependent upon the dominating background source and whether there is temporal coincidence of peak background and primary emissions and poor dispersion. Where such peaks are caused by wind-driven resuspension (including sea salt generation) such temporal coincidence is unlikely.

We have reviewed several different approaches to quantifying background PM_{10} in New Zealand. Table 11 summarises four of the key approaches and the resulting predicted **long-term** means. Despite the large short-term and spatial variabilities and uncertainties it is noteworthy that four different approaches in different locations have predicted similar numbers. These values all appear to confirm that the value of $10 \mu\text{g m}^{-3}$ suggested as a nationally applicable estimate by Fisher *et al.* (2005) in the absence of more detailed information, is reasonable, except at exposed coastal sites where it will be an under-estimate.

Table 11: summary of different estimates of long-term average background PM_{10} in New Zealand.

Technique	Location	Mean PM_{10}
Receptor modelling	Hastings	8
Receptor modelling	Kingsland	8.7
Receptor modelling	Takapuna	9.2
Receptor modelling	Queen Street	8.4
Receptor modelling	Khyber Pass Road	9.6
PM_{10} – CO regression	Masterton	8.8
PM_{10} – CO regression	Christchurch	9.1
PM_{10} – CO regression	Ashburton	11.2
PM_{10} – CO regression	Timaru	11.6
Regional background monitoring	Pongakawa (Hawke's Bay)	~ 9 since 2003
Filtered rural background monitoring	Pukekohe (Auckland)	10 – 14 depending on wind direction

Recommendation 4

As a first estimate of long-term average regional background PM₁₀ in New Zealand we find that a value of 10 µg m⁻³ is reasonable at a non-coastal site.

5. Conclusions

5.1 General conclusions

Several definitions of ‘background’ have been found in common use. We find that no one definition is superior or preferable to any other. Consequently, it is recommended that reports and documents clearly define background when the term is used.

Broadly speaking, PM₁₀ consists of three components:

1. Local anthropogenic emissions. These are mostly derived from combustion, mostly occur in the fine mode, and their concentrations peak in high-emission/low-dispersion conditions, principally in low winds in winter, and especially at night in locations where domestic wood-burning for heating is prevalent.
2. Mineral dusts and sea salt in the coarse mode. Emissions are either natural or anthropogenic (e.g. construction, quarrying, road and vehicle wear), but a common factor is that their atmospheric concentrations peak in high winds, in direct contrast to the local fine mode emissions. Variation is rapid and localised.
3. Fine mode particles transported to an airshed from afar. This component is generally the smallest of the three, has some seasonal pattern, but a fair degree of unpredictability.

The latter two components are generally considered to be part of the background. Due to the different emission and transport processes involved each component rarely peaks simultaneously.

Coastal sites can have very high background concentrations due to high sea salt loadings. Long-term background measurements of the order of 15 µg m⁻³ have been made at Christchurch (in onshore winds) and Kaikoura with a strong spatial gradient reported in Christchurch.

The relative impacts of emission cuts can only be predicted if the contribution to concentrations of other emissions, including natural, non-inventory and non-local, can be quantified and understood. As the New Zealand AQNES for PM₁₀ specifies an averaging time of 24 hours, an understanding of the temporal variability of the

background is crucial. The key question is how much do background sources contribute to concentrations on those days when the NES is exceeded?

This question is addressed in Wilton *et al.* (2007) and Davy *et al.* (2007). In most areas of New Zealand it appears that NES exceedences occur on days when the background component is minimal. The opposite relationships between sea salt and combustion emissions with wind speed have led to similar conclusions in the UK where it was concluded that removal of the contribution of sea salt from PM₁₀ would make only minimal difference to the number of exceedences in London (Jones & Harrison, 2006).

Source apportionment studies using receptor modelling indicate that in summer, non-anthropogenic sources can constitute up to 80% of the measured PM₁₀ although on high pollution days in winter the value tends to be much lower, e.g. 10 - 20% in Masterton, 15% in Auckland or 8 - 10% in Hastings.

Estimates of long-term average regional background PM₁₀ have been compared from three methods (background and filtered monitor data, regression of PM₁₀ against CO monitor data, and receptor modelling) across 5 locations (all on North Island). All but one analysis suggested a value in the range 8 – 12 µg m⁻³, with a higher range at Pukekohe of 10 – 14 µg m⁻³ potentially linked to higher wind-driven resuspension in south-westerlies.

5.2 Recommendations for future monitoring

As this report has made clear, our knowledge of background PM₁₀ in New Zealand is limited by the paucity of appropriate monitoring data. Monitoring in more regional background sites, such as Pongakawa, is highly recommended, even if only on a limited survey basis. An alternative approach to determining background concentrations may be to employ methods which are specific to the expected dominant sources or chemical constituents. For example, in coastal locations where sea spray is expected to be the dominant contributor measurements of sodium or chloride ions may provide a more direct measurement of this background, or at least an independent method for verifying estimates derived from PM₁₀ monitoring. Further estimates of background PM₁₀ could be derived from regression methods if more PM₁₀ monitors were co-located with CO monitors. Again, limited surveys, while inferior to permanent installations, may still be preferable to no data at all.

5.3 Recommendations for future research

We have reported the successful use of regression of PM₁₀ monitor data against co-located CO data to derive an estimate of background PM₁₀. In principle, the same approach should be applicable to any quantifiable substance other than CO which has a dominant anthropogenic combustion source, e.g. NO_x, PM₁, particle number concentrations. To date we are unaware of any attempt to test the applicability of such data, but recommend that such an attempt is made.

There is much still to be done to quantify and predict the contribution of background sources to PM₁₀ in New Zealand. This report has mostly focussed on long-term averages, but there is a need to understand more about the temporal variabilities, so that the nature of ‘worst-cases’ can be better understood. This is especially the case in terms of some highly variable and unpredictable components. In general fine particles tend to be emitted on a more continuous basis and some of their sources are more widely distributed in space (e.g. oceanic sulphates). They have lower deposition velocities giving them longer atmospheric residence times. This leads to them being relatively ubiquitous in the New Zealand atmosphere, subject to seasonal and some random temporal variation, but reasonably well described by seasonal or annual means or median concentrations. In contrast, emission of coarse particles is much more localised and sporadic with a more binary nature (‘off’ or ‘on’). Deposition velocities are higher, residence times shorter, and transport distances much shorter. Consequently the impacts on ambient concentrations are much more localised, sensitive to local-scale influences (micro-climates, land-use, topography) and much harder to predict. In this sense a process-based model for prediction would need to be very sophisticated and would require, and be sensitively dependent upon, a large amount of input data. It may be that a more probabilistic approach based on monitoring and/or source apportionment data is a more suitable means of dealing with these impacts.

Recommendation 5

Further PM₁₀ monitoring at regional background sites (such as Pongakawa) is highly recommended.

Recommendation 6

Further co-located monitoring of CO with PM₁₀ monitoring is recommended. The use of regression with NO_x or measures of combustion-related particles should be researched.

6. Technical Annex – Review of sources and transport of regional background PM₁₀

6.1 Sea spray

6.1.1 Source variability

Globally, sea spray is believed to be the largest natural source of atmospheric particles, and perhaps the largest source of any type. The process of bubble bursting in the ocean projects particles of salt water over a range of sizes into the marine atmosphere. Many of these particles are small enough to remain suspended and if the relative humidity is low enough (approximately < 75 %) then they may evaporate and crystallize. The net emission rate is a function of wind speed, as is the size distribution of the particles generated, with higher winds leading to the suspension of more large particles leading to a rapid increase in the mass concentration. Some studies have reported threshold wind speeds, with 3 m s⁻¹ being an indicative value, although values over 7 m s⁻¹ have been reported (Meira *et al.*, 2006). Emission is also enhanced at the coast through wave breaking, especially in high-energy conditions.

6.1.2 Inland transport & deposition

Deposition of sea salt, as with all particles, is a function of particle size. The larger particles, suspended at the coast will rapidly deposit within the first few hundred metres due to sedimentation, impaction and interception. Finer particles will be transported progressively further, so that the size distribution, and net deposition velocity of the marine aerosol both change with distance inland. A small number of research campaigns have measured sea salt (or chloride) deposition within a few kilometres of a coastline. Typically the results have been summarised using parametric models that are generally of the form of an exponential decay in deposition (and hence by implication, surface-level concentrations) with distance and an exponential relationship with wind speed (e.g. Gustafsson & Franzen, 1996). Beyond a kilometre or so from the coast the coarser particles emitted at the coastline have mostly been removed leaving only finer particles generated both at the coast and from greater distances out into the sea or ocean. Meira *et al.* (2006) summarise a number of other studies that found that marine salt concentrations drop by 85 – 95 % in the first 500 m from the coastline. To date such field studies have not considered the effect of topography or varying vegetation, although modelling (e.g. Cole *et al.*, 2003) has indicated that increased surface roughness (which may be represented by both trees and buildings) leads to strongly enhanced deposition. Forested and urban surfaces are aerodynamically rougher than grass or crops and this variation in roughness may have

a seasonal component. This is significant in Auckland due not only to its urban fabric, but also because much of the city is separated from a major sea spray source (the Piha coast) by the forested Waitakere Ranges.

Furthermore, sea salt particles may be removed by rain and wet deposition processes. In relative humidities in the range 50 – 70 % the salt is more likely to be ‘wetted’ and settle gravitationally, or be removed by rainfall. This effect of humidity is non-linear and very significant at high humidities (Cole *et al.*, 2003).

Decay in sea salt concentrations at greater distances inland (tens or hundreds of kilometres) has barely been studied. However, in one study contours and three transects in Na^+ and Cl^- ions over western Europe were plotted by kriging of data from 91 monitoring sites in Belgium, the Netherlands, France, Germany and Spain (Delalieux *et al.*, 2006). In this study the immediate coastal enhancement was not included. Generally logarithmically decaying functions were fitted to the constructed transects across Belgium, France and Spain. The Cl^- concentration decreased by 90 % after about 560 km in Belgium, 390 km in France and 290 km in Spain. Although all locations in New Zealand are less than 150 km from the coast this is by the shortest route, not necessarily the route taken by prevailing winds. South-westerly winds are common in South Island, which is up to 800 km long in this direction.

Modelling has suggested that wind speed is the most important factor in determining inland sea salt concentrations with concentrations decaying fairly rapidly over the first ~5 km with a weaker linear gradient developing after ~10 km such that concentrations have approximately halved that at 1 km after ~50 km (Cole *et al.*, 2003).

6.1.3 Consequences of sea spray emission and dispersion for background PM_{10}

Values and gradients in the contribution of sea salt to PM_{10} are likely to be largest in coastal zones. This is highly significant for New Zealand as most of the population live near the coast. For example, 70 % of the New Zealand population live in the 20 largest towns or cities. Of these 20 cities, 17 (covering 90 % of this urban population) are on the coast, giving a coastal population of over 2.5 million. This sea salt contribution, and its spatial gradient, are highly sensitive to wind speed and are also moderated by wind residence time. Thus it changes substantially on a sub-daily timescale.

The strong gradients in coastal areas have great significance when interpreting monitor data. For example, in Auckland the Takapuna, Queen Street and Botany Downs monitors are all within 3 km of the coast, and the Orewa monitor is only 0.4 km from

an ocean-facing beach known for its surf. However, Auckland's complex coastal morphology made up of the Pacific coast (partly sheltered by the islands of the Hauraki Gulf), the Tasman Sea coast (from which Auckland is partially sheltered by the Waitakere Ranges), and two large harbours will lead to a highly spatially as well as temporally variant emission field for sea spray that will be very challenging to describe in any detail. The variation in deposition due to land-use and topography adds another layer of complication. These considerations apply equally to Wellington, well-known for its windiness.

In light of this it may be seen how limited any long-term, widely applied estimate of regional background must be. If we are to adopt a long-term average background value we must always remain aware that it will under-estimate the true background value within ~ 1 km of the coast (and especially within a few hundred metres) due to this localised sea spray component.

6.1.4 NZ data and studies

The simplest way of estimating the combined contribution of sea spray and long-range transport to regional background in New Zealand is to make measurements at a coastal site. Measurements at Kaikoura have indicated some of the highest PM₁₀ concentrations in New Zealand when urban sources are discounted. A mean background concentration of 16 µg m⁻³ has been estimated from these measurements. We have taken the one year of continuous hourly PM₁₀ observations (Feb 2002 – Feb 2003, provided courtesy of Environment Canterbury) and binned the data as a function of wind speed (measured at the monitoring site in bin widths of 0.2 m s⁻¹). We have also separated hours of onshore and offshore winds. For each wind speed bin we have calculated the mean PM₁₀ for onshore and offshore winds separately. The result is shown in Figure 8. Despite increased scatter at high wind speeds (when there are fewer observations leading to weaker statistics) this shows three key features:

1. an expected decrease in PM₁₀ with wind speed for low winds, representing dispersion of a local source,
2. an increase in PM₁₀ with increasing wind speed above 3 or 4 m s⁻¹,
3. consistently higher PM₁₀ in onshore winds compared to offshore (6 – 10 µg m⁻³ at low winds, going over 10 µg m⁻³ at higher winds).

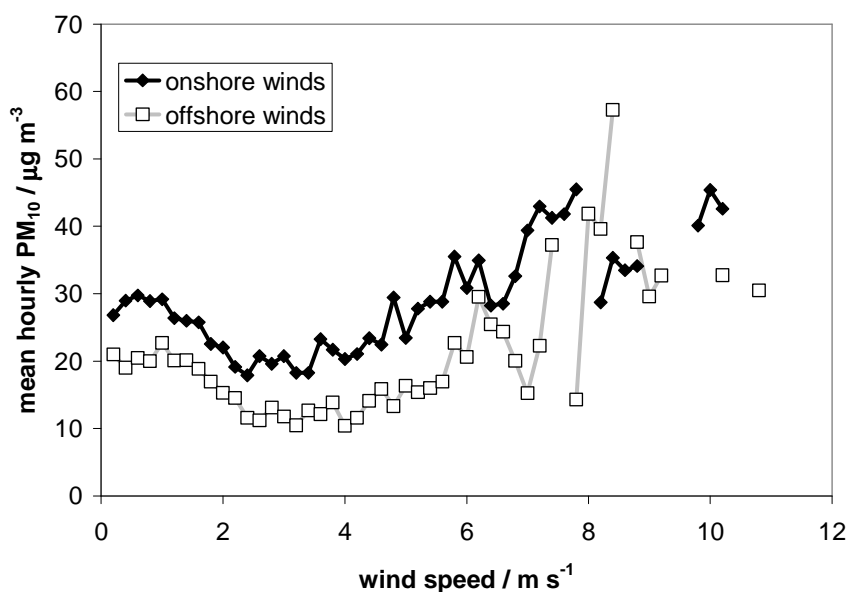


Figure 8: Mean hourly PM₁₀ for binned wind speeds observed over a year at Kaikoura (2002-3) in onshore and offshore winds (Data courtesy of Environment Canterbury).

Where possible, PM data should be filtered according to wind direction, although this may not be satisfactory in terms of 24 hour samples due to the regular variation in wind direction on shorter timescales. One simple alternative is to measure PM on an hourly or finer timescale using a continuous method, although this prevents the analysis of filter samples which may be required if it is desired to identify various components such as chloride or sulphate ions.

This approach was recently implemented and augmented by another approach by Clark *et al.* (2007). The SampleMaster 7000 (Harrison *et al.*, 2007) combines 7 gravimetric samplers and 7 sampling heads, with one of six activated depending on the wind direction, and the seventh activated in calm conditions. Thus each sample is associated with one wind direction range only. This arrangement was operated at New Brighton, Christchurch, 100 – 200 m from the beach in winter 2006 and summer 2006 – 7. A very strong sea spray signal was identified with mean summer PM₁₀ concentrations of 15 µg m⁻³ at the coast compared to 5 µg m⁻³ in offshore winds. The expected strong spatial gradient progressing inland above was suggested by a summer mean concentration of 11 µg m⁻³ at Coles Place, 8.4 km inland.

6.2 Mineral dust

6.2.1 Source areas

Anecdotally it has long been known that many parts of Australia are source areas for the resuspension of soil dusts by the action of the wind. However, exact delineation of source areas is difficult to achieve. Areas prone to dust storms are understandably poorly populated and difficult to travel across. Dust storms present an extreme environmental challenge to the operation and maintenance of instrumentation. Consequently desert areas are relatively data-poor.

Recently satellite-based remote sensing has been employed to provide a large-scale spatial coverage unavailable through ground-based measurements. A key investigation was that by Prospero *et al.* (2002) who used the Total Ozone Mapping Spectrometer instrument on the Nimbus 2 satellite to map in detail the major sources of dust around the globe, and characterised the common geography of those sources. They confirmed the expected general importance of aridity, but also showed how the presence of some water or water in the recent geologic past is crucial. They also showed that local topography also plays a very important role. Strong source areas tend to occur in topographical lows, consistent with alluvial catchments. One may expect central Australia to have many strong dust sources, as much of the continent is desert. However, the TOMS analysis showed that relative to deserts elsewhere this was not the case. Consistent with results from the northern hemisphere, it was proposed that this was due to the long-term aridity of the continent, leading to dust deposits having long been removed, and the low topography bereft of any basins likely to capture alluvial deposits.

Localised suspension processes include whirlwinds and thunderstorm outflows. However, the principal mechanism that leads to the long-range transport of desert dust in Australia is believed to be the passage of cold fronts. However, dust emission tends to peak many months after peak rainfall and this is believed to be due to the slow desiccation of wetted soils and the gradual loss of soil integrity provided by vegetation (Zender & Kwon, 2005).

This was explored in more local detail by Ekström *et al.* (2004) who produced a climatology of Australian dust storms based on monitoring data from 1960 to 1999. Two storm zones were defined. The stronger source (in terms of frequency) was a continental interior zone that experiences maximum dust storm activity in spring and summer. The storms are largely precipitated by westerly non-rainfall cold fronts and are terminated by the onset of the wet summer monsoon. In summer the focus of dust storm activity shifts to the near-coastal zone with maximum activity (albeit at a lower

overall frequency) in summer and autumn, especially in the west, southwest and south where summers are particularly dry.

Inter-annual variation can be considerable. As dust suspension requires the desiccation of sediments, plus, especially in the case of Australia, the regeneration of sediment supply to alluvial beds by seasonal ephemeral river flow, inter-annual variations in rainfall and drought strongly influence dust storm occurrence and severity. Consequently dust storm activity and source areas can change in El Niño years.

6.2.2 Trans-Tasman transport

Most of the evidence for trans-Tasman transport of dust comes from the identification of distinctively red dusts on snowpack and especially glaciers in the Southern Alps. Local dust is generally grey, although a recent study indicated that apparently grey dusts can contain 30 – 60 % Australian material (Marx *et al.*, 2005).

Dust haze is generally not observed in New Zealand. However, one of the largest recorded events was in April – May 1997 when a dust haze was reported over much of the country for several days giving rise to a weekly average concentration of $30 \mu\text{g m}^{-3}$ attributable to Australian dust alone (Marx *et al.*, 2005).

Prediction of dust transport impacts in New Zealand is particularly difficult due to its sporadic nature and it has been noted that not all cold front passages and associated dust storms in Australia lead to trans-Tasman dust transport (McGowan *et al.* 2000).

As cold front passage is the principal mechanism transporting dust across the Tasman Sea it is also quite probable that much of that dust is removed by precipitation en route. Furthermore, much of the dust that does reach New Zealand will be forced to rise by the steep western slopes of the Southern Alps increasing the probability of interception and impaction on surfaces and wet deposition in precipitation or orographic interception.

Marx *et al.* (2005) have developed a new technique for specifically identifying Australian dust deposited in New Zealand by determining ^{210}Pb radioactivity in dust samples. Weekly measurements were made from dust sampled at six remote sites in South Island including 12 years worth of samples thus providing an unusually high quality insight into long-term averages, seasonal patterns and weekly variability. A key aspect of this study, compared to previous studies of dust deposits in New Zealand or dust storms in Australia was its ability to look beyond these high profile events and identify the far more frequent transport of dust that is rarely observed by other means.

The average concentration of Australian dust in New Zealand was estimated as $5.3 \mu\text{g m}^{-3}$, while the median was $4.6 \mu\text{g m}^{-3}$. Such values indicate that the transport of this finer dust is much more significant in the long-term than individual dust storm events. Dust deposition was shown to be three to four times higher in Greymouth and New Plymouth compared to Christchurch and Dunedin due to precipitation scavenging associated with upland areas, and double that in areas with minimal orographic precipitation (Auckland, Invercargill and Kaitaia).

Australian dust concentrations in New Zealand peaked in autumn-winter, despite the maximum frequency of dust storms and westerly winds occurring in spring. The key may be that this ^{210}Pb technique is best suited to identifying sub-micron particles, which tend to be resuspended more easily, more often and in lighter winds than the coarser dust particles. Emission of these particles therefore begins earlier after the re-charging of alluvial beds by the summer monsoon, leading to a maximum emission in autumn. This decreases towards late spring as the fine particles are depleted, just at the time that cold front passage and higher wind speeds begin to initiate dust storms that resuspend the remaining coarser material. Thus whereas Australian climate and weather control coarse dust emission, fine dust emission is more strongly related to sediment availability.

6.2.3 New Zealand mineral dust

Dust deposition rates have been measured on the east side of the Southern Alps (McGowan, 1996), but also on the west (Marx & McGowan, 2005). Deposition rates on the west side were an order of magnitude lower than on the east, which is unsurprising considering the much higher rainfall and humidity there on the west side and rain shadow on the east. Dust sources are predominantly wide braided alluvial beds and resuspension is driven by downslope föhn winds (warm dry downslope winds commonly exceeding 15 m s^{-1}) as well as synoptic-scale troughs with non-precipitating cold fronts. Contributions to ambient concentrations were not reported in these studies, however their effects are believed to be localised.

6.3 Other dusts

6.3.1 Anthropogenic resuspension of dusts

Several anthropogenic activities create, resuspend and redistribute dust. Quarrying and mining generate large quantities of dust, although much of it is coarse and deposits to the surface locally (e.g. Pless-Mullooli *et al.*, 2000). These activities are largely carried out far from population centres, but construction is inevitably concentrated in built-up

areas. Most large construction projects are now required to have comprehensive systems in place to reduce dust emission, but the local impact of construction can still be considerable. Several studies have shown local PM₁₀ measurements enhanced by 10 µg m⁻³ or more in the vicinity of urban construction sites (Muleski *et al.*, 2005, Muir *et al.*, 2006). However, each site is unique, and the different stages of construction will emit different amounts and types of dust and predicting the impact before monitoring is nearly impossible.

The localised impact of these activities is compromised by track-out. This is the term used to describe the mud, soil and dust carried away from quarries, mines, construction sites and other dusty locales by vehicles, especially trucks. This can transport soils onto road surfaces which, when dry, are available for resuspension. Repeated disturbance by vehicles further crushes these particles to smaller sizes further aiding their resuspension. While methods are employed in the relevant industries to minimise track-out to a large degree it is unavoidable, but very difficult to quantify (Kinsey *et al.*, 2004).

Trackout dust joins the road dust and vehicle wear products on the road surface. This can be augmented wherever traction salt is added to a road surface. Although the salt crystals are large over a few days they become crushed until some are of resuspendible PM₁₀ size. Rainwater can also wash soil onto the road which, when the dried, can form a reservoir of resuspendible material.

The resuspension process is driven by natural turbulence in the wind, but in the case of road dust the action of the traffic greatly increases the emission rate. Traffic directly suspends wear products (primarily from brake pads and tyres) as well as depositing them on the road. Dust is resuspended by the direct action of tyre-road contact and by the turbulence induced by a moving vehicle. Emission is much stronger for larger vehicles and is increased at higher vehicle speeds. Mitigation is very difficult to achieve. The effectiveness of road sweeping is in doubt, as is road washing (although some road coatings do seem promising in reducing resuspension – see Norman & Johansson, 2006). Other than that only reducing the size of vehicles, reducing the number of HDVs, reducing traffic speeds and reducing total traffic appear to be potentially effective, although we are aware of no studies that have evaluated these strategies in practice.

6.3.2 Biogenic dusts

There are multiple biogenic particles in the atmosphere from land-based sources. Most of these are coarse in size and their emission is governed by biological cycles and

wind-driven resuspension. This includes rusts, dander, fungal spores and bacteria. Pollen is generally larger than 10 μm (except for manuka pollen) and so not directly relevant to PM_{10} , however pollen is regularly crushed by various processes, especially anthropogenic ones, such that pollen fragments do often fall into the PM_{10} size fraction. Research into biological particles suspended in the atmosphere is globally relatively immature and the contribution of such material to PM_{10} in New Zealand is unknown. A comprehensive review of the interaction between bioaerosols and meteorology is provided by Jones & Harrison (2004).

6.3.3 Volcanic dust

Globally, volcanoes are one of the largest natural sources of aerosol into the atmosphere, especially for sulphur species (Graf *et al.*, 1997). These emissions are generally remote and form part of the global remote background so that their influence on PM_{10} in New Zealand is generally indistinguishable from the remote background and probably negligibly small, especially in relation to concentrations often observed in urban areas.

Large-scale volcanic eruptions in New Zealand are relatively rare events. Mt Ruapehu probably represents the highest risk from ashfall to the New Zealand population. There is an immediate danger associated with large fallout, but also from fine ash that may have been transported large distances before reaching the surface. In the prevailing winds any plume from Mt Ruapehu would tend to be transported towards the Bay of Plenty or Hawke's Bay and the Pacific Ocean in general, impacting relatively small populations. Throughout the 1995/1996 eruptions, reports of ash fall were made in Wanganui, Taupo and Napier. During one event in mid-June 1996, the weather system resulted in the plume passing over Auckland. The plume was visible from elevated viewing positions and Auckland International Airport was closed in order to avoid damage to the aeroplanes. However, there was insufficient coarse material to form a visible ash layer on the ground surface and unfortunately, no PM_{10} monitors were in operation at the time. However, the meteorology and carbon monoxide concentrations recorded at the time suggested strong temperature inversion conditions and peak CO concentrations, irrespective of the volcano. The mortality rate for respiratory causes at Auckland hospital more than doubled in the days following this eruption event. Investigations are underway to determine whether this increase can be attributed to the volcanic ash, or was simply due to weather conditions conducive to high urban air pollution levels. (Dirks, K.N., personal communication). Work is also underway to develop a modelling methodology to be able to determine the contribution of sporadic sources (such as those from volcanoes) to urban air pollution (Dirks, K.N., personal communication) using data from Mt Etna in Italy that

erupts regularly. Elevated PM₁₀ concentrations through the re-suspension of fine volcanic dust by the wind or anthropogenic sources is also a concern (see above).

Minor eruption events may not be within the realm of air quality management, except in the case of the impact on communities within close proximity to active volcanoes such as the residents of the Bay of Plenty that are influenced by the eruption of White Island. In this case, residents may be subjected to regular locally-elevated sulphate levels (see also section 6.6.1 below).

6.4 Intra-national transport

New Zealand is still a relatively under-populated country. Urban areas take up only 3 % of the total land area and most atmospheric emissions are densely concentrated in a few areas. Consequently, the transport of material from one airshed to another, or the formation of secondary PM in one New Zealand airshed originating from emissions of precursors in another is unlikely to be a dominant source of background PM, but also cannot be ruled out. In the case of Auckland the prevailing wind directions are southwesterly and northeasterly. Open ocean lies in both of these directions leading to the conventional wisdom that Auckland is neither a source nor a receptor for intra-national transport. However, airmasses may arrive in Auckland on a southwesterly wind along trajectories that may have taken them along the west coast of South and North Island, and the same wind direction describes a line roughly linking Invercargill, Dunedin, Christchurch, Wellington, Palmerston North, Hastings and Napier. However, we know of no evidence that shows that emissions from any one of these cities are significantly affecting concentrations in others. Measurements at Baring Head (Allen *et al.*, 1997) did indicate that some sulphate measured there was related to SO₂ emitted within New Zealand, although a more specific source could not be specified.

6.5 Non-mineral long-range transport

6.5.1 Sources of sulphate

Measurements at Baring Head (e.g. Allen *et al.*, 1997), across the South Pacific and Southern Oceans have shown that the aerosol transported to New Zealand is representative of both natural and anthropogenic emissions from the nearby oceans, Australia and beyond. Principally long-range transport consists of a constant remote background component, augmented by a seasonally-varying natural marine source and a larger but more sporadic anthropogenic plume transport principally from Australia.

6.5.2 Source areas of long-range transport to New Zealand

The geographical origin of emissions in other parts of the world that are transported to New Zealand can be investigated by modelling of the movement of airmass trajectories. A study by Sturman *et al.* (1997) described many of the common atmospheric trajectories that may deliver fine particulate matter to the Tasman Sea and New Zealand in January and July. In July large numbers of trajectories to the mid Tasman Sea originated in Australia, but a greater number passed to the south of Australia and this number increased in January. Transport from southern Africa was indicated in the winter with 22 % of trajectories leaving central southern Africa arriving in the mid Tasman Sea after an average of 16 days, and 50 % of trajectories arriving in the mid Tasman Sea having passed south of Madagascar a fortnight before. This route was found to be insignificant in the summer.

Transport from western Australia (Perth) to New Zealand was found to be very limited in summer, but accounted for a third of all plumes in July with a transport time of less than a week. From Sydney 83 % of all summer low-level eastwards plumes passed over New Zealand 5 days later. In winter the dominant trajectory passes north of North Island after 3 days.

From the point of view of back-trajectories plotted from Auckland it was found that most airmasses originate from south of Australia, but transport from Australia is more common in winter and sources in western Australia are more common at this time of year. This seasonal distinction was even stronger for trajectories arriving in Christchurch. In general it should be noted that the Southern Alps act like a filter removing a lot of dust and hygroscopic aerosols (especially sulphates) through precipitation scavenging, preventing much of this material arriving in the airsheds of the east coast of South Island, so that for instance we may expect Christchurch to be less impacted by long-range transport than Auckland or Hamilton.

The principal significance of these analyses for air quality management in New Zealand is firstly that long-range transport of continental and anthropogenic background emissions from Australia is more common in the winter, at the time when local domestic heating emissions peak and NES compliance is most difficult to achieve. Secondly, trajectory analysis highlights the highly sporadic nature of long-range transport impacts and its susceptibility to subtle changes in the climate.

6.5.3 Remote and marine background

Sources and nature

On average, the particles making up PM₁₀ have an atmospheric lifetime of about a week, but some particles will remain suspended for much longer. On these timescales particles will travel many thousands of kilometres, and even encircle the globe such that they become very well mixed. They are also not inert, but will undergo many chemical and physical changes (generally termed ‘ageing’) such that they are physically and chemically different to the particles that were originally emitted. Thus, it is very difficult to ascribe their source. However, globally, significant sources of atmospheric particles include soil particles, desert dust, sea spray, volcanic dust, natural biomass burning and biogenic secondary organic aerosols. To this we may add particles emitted as a result of human activity – principally due to combustion, but also resuspension of dusts by traffic, quarrying, mining, and construction. Human indirectly contribute to dust emission by degrading soils and disturbing soil crusts.

Measurements in the remote background

Measurements at remote background locations have revealed that, as well as mineral dusts, particles consist of complex humic-like organic substances and also black carbon (Van Dingenen *et al* 2004, Putaud *et al* 2004) and various studies have reported values of up to 94 ng m⁻³ for black carbon concentrations in Spitsbergen (see e.g. Nyeki *et al* 2005 and references therein).

Assessment of the concentrations and composition of the marine background aerosol (DMS, nitrate, non-sea-salt sulphate, methanesulphonate, ammonium) have been made from measurements during 1991 and 1992 at Baring Head on the Cook Strait coast approximately 15 km southeast of Wellington (Allen *et al.*, 1997). Of the components listed, non-sea-salt sulphate was clearly dominant, especially in those periods when the back trajectory indicated a clean marine air mass origin (i.e. southerlies) during which concentrations were of the order of 0.1 – 0.2 µg m⁻³. Concentrations measured elsewhere in the marine environment of the southern hemisphere have tended to similar values indicating weak spatial gradients.

Remote sulphates

Research in the 1990s based on measurements at Baring Head and the Antarctic indicated that, despite the dominance of the marine DMS source of sulphate (see

below) a proportion of the non-sea-salt sulphate could not be explained by the known oxidation mechanisms of DMS (Allen *et al.*, 1997). This non-DMS component was of a low but relatively constant concentration indicating its remote source. Measurements during the First Aerosol Characterisation Experiment (ACE-1) in the Southern Ocean found that 10 – 45 % of sulphate particles were internally mixed with elemental carbon (Posfai *et al.*, 1999). Volcanic emissions of sulphur exceed biomass burning by 2.7 times in the southern hemisphere (Graf *et al.*, 1997) and it has been estimated that volcanic sulphur contributes 6 – 11 % of remote background nssS. Some relatively simple assessments have indicated that transport of sulphur from continental sources in southern Africa and south America may augment emissions from Australia to provide this continuous remote nssS background concentration encircling much of the southern hemisphere. Allen *et al.* (1997) estimated that this remote background nssS had a concentration of $0.1 \mu\text{g m}^{-3}$.

Dimethyl sulphide

Sulphate arising from the emission and oxidation of dimethyl sulphide can be considered to augment this remote background component. Biological production of gaseous Dimethyl sulphide (DMS) in the ocean and its transfer across the sea-atmosphere interface is the largest source of sulphate aerosol in the marine atmosphere. Studies such as those at Baring Head (e.g. Allen *et al.*, 1997) have found a seasonal pattern with higher concentrations of MSA and nssS in summer consistent with the summer blooming of DMS-producing phytoplankton, and a reduction almost to zero in winter. However, as well as a seasonal pattern, the concentration of this DMS-source sulphate is dependent upon the latitude through which the air mass has passed. For example, Allen *et al.* (1997) reported a value of $0.4 \mu\text{g m}^{-3}$ in air masses that had crossed the Tasman Sea compared to $0.08 \mu\text{g m}^{-3}$ for air masses arriving in New Zealand from the higher latitudes of the Southern Ocean.

6.6 What are the outstanding questions?

6.6.1 The source of the sulphate

An unanswered question in the recent source apportionment study reports is the origin of the sulphate. Sulphate made a small contribution in Hastings (4 % of PM_{10}) and a slightly larger proportional contribution in Auckland (3 - 7 %). For $\text{PM}_{2.5}$ sulphate contributed on average 10% in Hastings and 8 - 17 % in Auckland (with a seasonal peak of 35 % in summer). The Auckland contributions are quite substantial, so it is

worth asking to what degree this sulphate source is a) international, b) within New Zealand, c) local? That the concentrations peak in summer may suggest a significant role for photochemistry. However, the seasonal variability in the marine dimethyl sulphide source may also make a contribution (order of $<0.5 \mu\text{g m}^{-3}$ according to the data from Allen *et al.*, 1997). This seasonality was not evident at Hastings.

In general sulphates form through gas to particle conversion of sulphur dioxide to sulphuric acid and neutralisation by ammonia. This process typically occurs in urban plumes at the rate of around 1 % hour⁻¹, such that an air mass will have travelled 1000 km (or the length of New Zealand) before a substantial amount of SO₂ has been converted into sulphate. However, this rate can be much more rapid in exceptionally high concentrations, and a few industrial processes directly emit sulphates, such as copper smelting and fertilizer manufacture.

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 in a process that may be rapid but is not always observed. 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.

Wang and Shooter (2002) compared levels of sulphate in Auckland and Christchurch during limited sampling in summer and winter, and also differentiated between daytime and nighttime levels. Non-sea-salt sulphate was higher in the nighttime in Christchurch, but higher in the daytime in Auckland. The authors looked to differences in local emission to explain this discrepancy, citing higher vehicle emissions and daytime photochemical oxidants in Auckland and higher nocturnal domestic emissions in Christchurch, thus implicitly assuming that at least some secondary sulphate production was occurring in the same airshed as the precursor emissions. Further evidence for this was the increase in fine sulphate concentrations in winter in Christchurch when PM_{2.5} is dominated by domestic heating emissions, compared to spring and summer, in contrast to other studies mentioned in this report that found no seasonal bias, or a weak summertime peak. Fine mode concentrations in Christchurch were very approximately double those in Auckland despite its smaller concentration. This is due to its climate, which is generally colder due to its location, but also more prone to low winds, drainage flows and nocturnal inversions, which tend to significantly reduce dispersion. Thus rather than being rapidly advected away from the airshed, emissions can remain in a stagnant air mass in which pollutants accumulate thus increasing secondary particulate production rates leading to the in-

situ production of principally nitrates, but potentially also sulphates. Whether such in-situ production is significant in Auckland or other New Zealand airsheds is currently unknown. Later measurements in Auckland (Wang & Shooter, 2005) indicated a very small contribution ($< 0.2 \mu\text{g m}^{-3}$) of ammonium sulphate to PM_{10} in Auckland. This is much lower than the 'sulphate' contribution from the recent source apportionment study, but is consistent with the strong identification of the sulphate profile with aged marine aerosol.

Shipping has been identified as a major source of unregulated SO_2 emissions elsewhere, leading to plumes in which sulphate is formed and transported on the wind often towards the land (Lu *et al.*, 2006). Such emissions are inconsistently reported in emission inventories. For instance the Auckland emission inventory includes estimates of emissions of key pollutants, including PM_{10} , $\text{PM}_{2.5}$ and SO_2 , for ships in berth and at sea. However the impact of plumes advected to land, and the contribution of this source to background PM levels, has not been considered. The shipping source was not explicitly identified in the recent source apportionment studies in Auckland or Hastings (Wilton *et al.*, 2007). This sector could be significant in that it represents a controllable sector.

The report for the 2006 Auckland study used conditional probability function analysis (CPF) to determine any wind directions associated with a higher probability of observing an increased signal for each source profile. Using the 5 sites these were then triangulated to try to identify a spatial source for each profile. For sulphates, the Kingsland, Khyber Pass Road and Takapuna analyses together suggested a source around the Port of Auckland and the shipping lane exiting the Waitemata Harbour. Conventionally it would not normally be considered that the atmospheric transport time between the Port and the monitors is sufficient to allow SO_2 from ship emissions to form sulphates. However, potentially high concentrations at emission, high sulphur fuels, heterogeneous chemical interaction with sea spray and local land/sea breeze recirculations may individually, or in combination, provide an explanation for part of the observed sulphate. These issues are currently being researched within the FRST Programme and will be reported in a separate report.

An intra-national natural source of sulphate of episodic importance may be volcanic emissions of SO_2 . The most recent evidence of this is a period of elevated sulphate in September 2006 which was observed in the source apportionment data across all Auckland sites. Peak sulphate rose to $10 \mu\text{g m}^{-3}$ of PM_{10} at Takapuna (compared to an autumn average of $\sim 1 \mu\text{g m}^{-3}$), $6 \mu\text{g m}^{-3}$ of $\text{PM}_{2.5}$ at Khyber Pass Road and $2 - 4 \mu\text{g m}^{-3}$ of $\text{PM}_{2.5}$ at Queen Street, Kingsland and Penrose (compared to autumn averages of $\sim 1 \mu\text{g m}^{-3}$). The accompanying report used back-trajectory analysis to relate this event

to the combination of a period of increased volcanic SO₂ emissions (2 – 3 times the norm) from White Island in the Bay of Plenty and meteorological conditions resulting in the transport of that material to Auckland via the Hauraki Gulf, with a transit time of 1 – 2 days (Davy *et al.*, 2007).

6.6.2 How to deal with episodic emissions/transport

Many of the natural sources are coarse (rural dust, desert dust, sea spray, volcanic dust, pollen, rusts, dander), and so are some of the anthropogenic ones (quarry dust, road dust). Their transport distances are generally very short and their emission very sporadic. For these particles annual means are of minimal value, as is any assessment on more than 1 km scale. However, this is less of a problem for finer particles. There is still a sporadic, and hence probabilistic element to forest fires and regional transport, but less so for photochemical secondary particles. A probabilistic approach may be more appropriate.

6.6.3 Nitrate, bound water and secondary organic aerosol (SOA)

Few studies in New Zealand to date have reported on contributions from nitrates or strongly bound water, which is generally associated with nitrates and sulphates. A study in the UK (60 – 90 km from the coast) found bound water to be a larger contributor to background PM₁₀ than either sea salt or secondary organic aerosol, accounting for 2.4 – 2.9 µg m⁻³ on average (Jones & Harrison, 2006).

The presence of nitrates in PM₁₀ is generally ascribed to long-range transport due to the timescales involved in the formation of nitrate from gaseous precursors. However, nitrates form approximately 5 times faster than sulphates and in-situ formation of nitrate in urban airsheds, especially at night, has been observed. In the long-term it is believed that this local production is relatively minor (e.g. Putaud *et al.*, 2004), but it could be significant in low dispersion, high pollution scenarios when NES exceedance is possible.

Wang & Shooter (2002) found nitrate concentrations to be approximately half of those of sulphate in both Auckland and Christchurch with slightly higher concentrations at night, although it is unclear if this was due to higher rates of formation. It should also be noted that losses of ammonium nitrate through volatilisation may have been substantial in this study where no denuder system was used. Concentrations were significantly higher in Christchurch compared to Auckland, but again it is unclear if this is due to differences in dispersion, advection sources or local production.

Nitrates have been found to be associated with coarse sea salt or soil aerosol, showing a summertime peak in Auckland (Wang & Shooter, 2005). Correlations of coarse nitrate with HONO and NH₃ indicate a combined traffic exhaust and road dust source of coarse nitrate (Wang & Shooter, 2005).

The contribution of secondary organic aerosol to PM₁₀ in New Zealand is unknown. Wang *et al.* (2005a) estimated that one-fifth to one-quarter of the organic carbon measured in the winter atmospheres of Auckland and Christchurch was secondary in nature, and suggested *in-situ* SOA formation occurred in the Christchurch airshed in winter. Emission rates are also unknown. Around 25 % of New Zealand's land area is forested, so emissions of biogenic precursors are likely to be highly significant, especially for monoterpenes from coniferous trees, which are known to be particularly potent precursors. A study in the UK (Jones & Harrison, 2006) estimated SOA (which was assumed to be 100 % biogenic) contributed a mean of 1.2 – 1.7 µg m⁻³ to PM₁₀ in southern England (the UK has a forested area of 5 %, but Europe as a whole has a forested area of over 40 %). Current work in the boreal forests of northern Europe and eucalypt forests of Australia suggest that SOA from forests may be greatly underestimated (Tanja Suni, University of Helsinki, personal communication).

It is often assumed that most sulphate and nitrate in the continental atmosphere will be neutralised by ammonium ions derived from land-based ammonia emissions. Ammonia is the most common atmospheric base and is generally ubiquitous in vegetated and farmed regions of the world. Its principal source is animal urine. However, modern vehicles have been shown to emit ammonia and there are a number of other industrial and anthropogenic sources. However, it has been shown that the oceans contain a biogenic source of ammonia, generally sufficient to neutralise the sulphate that also arises from a biogenic marine source (DMS). It has been shown in Europe how control of ammonia emissions is as vital as controls on NO_x and SO₂ emissions in reducing secondary PM; however, to date the role of ammonia emissions, both rural and urban, in determining background PM₁₀ has not been investigated.

6.6.4 Sea breeze and local recirculation

Recirculation is known to be important in the Mediterranean (e.g. Rodriguez *et al.*). Whether it is important in New Zealand, for example in Auckland or the east coast of the South Island is not known.

We know that layering (especially nocturnal) occurs in Christchurch where a layer of air containing daytime emissions is undercut by highly polluted katabatic drainage flows (McKendry *et al.*, 2004).

These effects can bring polluted air into an airshed from outside (even if it did originate in the airshed itself) so can add to the background in as yet unquantified ways.

The role of such local recirculations is being investigated within the FRST programme and will be discussed in a separate report.

7. Appendix – Notes from the Background PM Workshops held at NIWA in May 2007

Notes compiled by Guy Coulson with contributions from Ian Longley.

Two focus groups were held to help assess the current state of understanding and priorities for the future of the subject of background concentrations of PM₁₀ in New Zealand.

Attendees

Auckland 28 th May 2007	Christchurch 29 th May 2007
Amy Clore, MWH Camilla Needham, BECA Jenny Simpson, Tonkin & Taylor Janet Petersen, Auckland RC Gavin Fisher, Endpoint Shanju Xie, Auckland RC Andrew Curtis, URS Paul Baynham, Northland RC Ian Longley, NIWA Gus Olivares, NIWA Guy Coulson, NIWA	Peyman Zavar-Reza, Uni. Canterbury Perry Davy, GNS Emily Wilton, Environet Neil Gimson, Golder Associates Teresa Aberkane, ECan Tim Mallett, ECan Jeff Bluett, NIWA Guy Coulson, NIWA

It was appreciated by all that the definition of background varied with the use to which it was being put and although it would be useful for us all to agree on a standard definition, or set of definitions, the word is so entrenched in each context that it would be hard to change. Suggestions of modifiers such as “Natural background” or “Urban background” were made but they were thought unlikely to change things. Therefore it is probably simpler for it to be defined by the user depending on the context.

In general there were three different definitions of background (with variations).

1. the purely natural background (+ long range transport)
2. the part that’s not accounted for in emissions inventories (and can’t be managed)
3. the part that’s not of immediate interest

From an emissions inventory perspective “natural” and “background” are simply types of source. Background becomes everything that can’t be accounted for in the inventory. This is then dependent on the accuracy of all the other emissions factors. Implicitly, definition 2 includes anthropogenic emissions that can be managed, but just aren’t in emission inventories, which may vary between councils.

We can have a ‘background source’ and a ‘background concentration’ (i.e. component of concentration attributable to the background source).

In definition 3 the ‘background sources’ do not necessarily comply with definitions 1 and 2 (e.g. domestic heating emissions in a road project resource consent).

The main use for each type of background is;

1. research and airshed modelling – closing the gap between models and monitoring
2. air quality management – NES compliance
3. air quality assessment and resource consent

The value of a single “Official” value for the background concentration as in the UK was not considered helpful because of the multiple uses. There were, however, calls for varying degrees of officially sanctioned numbers or methods for background estimation. These ranged from including a range of indicative concentrations in the good practice guides to modelling and assessment to maps of the contribution of different aerosol types to the total PM₁₀ at a km and daily resolution mostly for resource consent and assessment purposes but also for NES compliance.

One of the biggest problems appears to be resolution, both temporal and spatial since different background sources and concentrations vary on very different timescales. Annual averages (as used to estimate the official background for the UK) were generally not considered high enough resolution. It was acknowledged that getting higher resolution for PM₁₀ is problematic so it was considered that annual would be a good place to start before refining the estimates to seasonal, monthly and (eventually) daily. Maybe an annual, or winter & summer values for photochemical secondary PM is useful

High spatial resolution is probably only necessary in the major urban areas, although there may be some rural industrial sites where it could be helpful. How high a resolution? A 1 km grid would be nice but again is probably ambitious, so a coarser resolution grid or even an airshed would be the starting point.

Many of the natural sources are coarse (rural dust, desert dust, sea spray, volcanic dust, pollen, rusts, dander), and so are some of the anthropogenic ones (quarry dust, road dust). Their transport distances are generally very short and their emission very sporadic. For these particles annual means are meaningless, as is any assessment on more than 1 km scale. However, this is less of a problem for finer particles. There is still a sporadic, and hence probabilistic element to forest fires and regional transport, but less so for photochemical secondaries or anthropogenic combustion sources as defined by definition 2.

For AQ management there is a need to estimate background as a starting point for straight line paths to NES compliance.

Possible methods

A table of values for different locations as used in the HAPiNZ project.

An empirical model that includes rainfall and wind (and perhaps land use and vehicles). This could simply be a lookup table

The contribution of non-combustion sources can be estimated by plotting CO and PM₁₀ as in the Masterton paper, although this is unlikely to work as well in windy, dusty, coastal or low CO locations

There are many monitors at rural industrial sites that could provide data for background estimation. (Although guidance for the industrial companies concerned on where to place monitors to make this useful would be helpful)

Place monitoring sites on the coast in rural locations away from anthropogenic sources, where the aerosol can only come from the sea (marine background) or the land (rural background)

Source apportionment studies are being carried out in several locations now. From these we will be able to give estimates of the contribution from different sources to the total PM₁₀ in (at least) an airshed. Therefore the contribution of background (regardless of which type of background is required) can be apportioned to a measured total PM₁₀ concentration. (Although this still requires an understanding of how representative any given monitor is)

Use of modelling to provide baseline datasets for different locations.

Recommendations

Track down industry datasets for rural concentrations

Provide some indicative numbers to include in the various modelling GPGs (eg from source apportionment results)

Provide “simple” management tool (probably empirical and based on lookup tables). Airshed modelling could play a role, but a simpler, perhaps GIS-based approach may be preferable

Use the FRST “Clean air” programme to provide a case study for the method or methods that we think would work.

Develop an annual model before trying to improve the resolution. Resolution can be improved by using models to interpolate.

What happens next?

From the results of the source apportionment studies, we intend to publish a report aimed at air resource users which,

- Defines “background” air pollution and considers vehicle, industry, domestic and natural sources
- Presents an analysis of air quality and source apportionment monitoring data that aims to determine the contribution of background air pollution to urban air quality.
- Provides a method by which background air quality can be estimated for the major urban areas in New Zealand

This consultation exercise will be used to help define the concepts of background and their uses. These in turn will be used to identify methods that can be used by end users in a range of circumstances.

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