

An Assessment of the Contribution of Coal-Fired Power Station Emissions to Atmospheric Particle Concentrations in NSW

A report prepared for:

Delta Electricity, Eraring Energy and Macquarie Generation

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EXECUTIVE SUMMARY

The current study was commissioned by the three New South Wales (NSW) state-owned, coal-fired electricity generating organisations to assess the potential environmental and health implications arising from fine particles, either directly emitted from the seven coal-fired power stations in NSW, or which form in the atmosphere from emissions from these facilities.

The relationship between fine particle levels in the atmosphere and human health impacts has been the subject of significant scientific research and regulatory development over the past couple of decades. Despite these developments, the current understanding of the formation and occurrence of fine particles in Australian airsheds is poor, including a dearth of information on the relationships between sources of fine particles (and their precursors) and ambient concentrations. Investigation and management of atmospheric particles is more complex and difficult than required for the “simpler” gas-phase air pollutants, such as sulfur dioxide, as in most circumstances a collected sample of atmospheric particles will consist of a diverse and variable mixture of elements and compounds arising from potentially many different sources – both natural and anthropogenic in origin. Size variability adds to the complexity of the particle issue, as size plays a crucial role in the atmospheric lifetime and properties, and in the environmental and health impacts of particles.

Although modern industrial particle collection systems are very effective in removing particles from the exhaust gases of facilities, they are generally less effective in removing finer particles than coarse particles. Furthermore, unless flue gas scrubbing is employed the emission of sulfur and nitrogen oxides potentially leads to the formation of secondary particles in the atmosphere.

This study reviews the state-of-knowledge with respect to atmospheric particles, quantifies the emission of particles from power stations and reviews the literature on the formation of secondary fine particles formed from emissions of sulfur and nitrogen oxides. Two different numerical modelling approaches are used to quantify the atmospheric concentrations of PM₁₀ and PM_{2.5} (particles with a diameter of less than 10 micrometres and 2.5 micrometres, respectively) in different regions of NSW resulting from power station operations.

The report provides estimates of ground level concentrations of PM_{2.5} and PM₁₀ from the direct emission of particles and from the in-plume conversion of gaseous sulfur and nitrogen oxides to ambient particles as the result of photochemical transformation. Estimated ground level concentrations arising from power station emissions are placed in context with other emission sources and are compared with current air quality levels and existing and proposed air quality goals and standards.

The major outcomes arising from the study are presented below:

- 1 While the causal mechanism(s) responsible for the relationships between atmospheric particles and health effects observed in epidemiological studies have not been definitively established, there is strong support for the view that finer particles are more strongly associated with adverse health outcomes than coarser particles.
- 2 The association between mortality and other health effects and *particle mass* show relatively consistent magnitudes of effects. There is considerably less agreement and consistency in the results from studies which have examined associations between health impacts and the *components* of the fine particles.
- 3 The frequency distributions of ground-level concentrations associated with power station emissions suggests that the **near-field occurrence** of elevated particle concentrations is likely to be infrequent.
- 4 Predicted **peak 24 hour average PM_{2.5}** concentrations were 8, 13 and 36% of the National Environment Protection Measure (NEPM) Advisory Reporting Standard of $25 \mu\text{g m}^{-3}$ in the Western, Central Coast and Hunter Valley regions, respectively. It was estimated that ammonium sulfate formed from the emission of sulfur trioxide could contribute up to about 70% of these peak near-field results.
- 5 Predicted **peak 24 hour average PM₁₀** concentrations were 5, 12 and 22% of the NEPM standard of $50 \mu\text{g m}^{-3}$ in the Western, Central Coast and Hunter Valley regions, respectively. It was estimated that ammonium sulfate formed from the emission of sulfur trioxide could contribute up to about 60% of these peak near-field results.
- 6 Predicted **annual average PM_{2.5}** concentrations in the three generating regions were in the range $0.4 - 2.0 \mu\text{g m}^{-3}$. The upper limit of this range is equivalent to 25% of the relevant NEPM Advisory Reporting Standard.
- 7 In areas removed from urban emissions, power station emissions were predicted to contribute up to $3 - 4 \mu\text{g m}^{-3}$ of secondary particles to peak 24 hour PM_{2.5} concentrations
- 8 The results from the modelling of “worst-case” days suggest that the power station contribution to urban fine particle concentrations is small. Further, an analysis of one year’s modelling results indicates that these small power station contributions to urban fine particle concentrations are likely to be infrequent.
- 9 The “**worst-case” contribution** of power station emissions to **24 hour average PM_{2.5}** in the Sydney urban area was estimated to be $2 \mu\text{g m}^{-3} - 8\%$ of the relevant NEPM Advisory Reporting Standard.
- 10 The predicted contribution of power station emissions to **annual average concentrations of PM_{2.5} in Sydney**, using a conservative methodology, was $0.3 \mu\text{g m}^{-3}$ – about 4% of the relevant NEPM Advisory Reporting Standard.

The project has met its overall aim to provide robust, credible information on the potential atmospheric concentrations of fine particles from power station operations. A number of specific issues have been identified during the course of the project for which additional research may lead to improved characterisation of the contribution made by power station emissions to fine particle concentrations in the near-field and regionally.

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	Appendix 1. CSIRO Report included as a separate volume	

1 INTRODUCTION

The relationship between fine particle levels in the atmosphere and human health impacts has been the subject of significant scientific research and regulatory development over the past couple of decades.

The Harvard Six Cities Study (Dockery et al, 1993), which commenced in 1974, arguably laid the foundations for subsequent research on particulate matter and health impacts around the world. The investigation was designed initially as a study of the effects of ambient sulfur oxides and particulate matter on the respiratory health of children and adults living in six cities in the eastern United States. The health of participants in each community was followed for 12 years with concurrent air pollution measurements. The objectives of the study were expanded to incorporate new air pollution measurement technologies, to collect new types of health data, and to assess the health effects of exposure to indoor as well as outdoor ambient pollution.

As a result of the Six Cities Study and the numerous studies which followed and continue today, the relationship between acute health impacts and the atmospheric concentration of particles with an aerodynamic diameter less than 10 micrometres (PM_{10}) and less than 2.5 micrometres ($PM_{2.5}$) is now recognised in environmental goals and standards in many countries, including Australia.

The Australian Ambient Air Quality National Environment Protection Measure (ANEPM) includes standards for both PM_{10} and $PM_{2.5}$. The discussion document associated with the ANEPM acknowledged that the current understanding of the formation and occurrence of fine particles in Australian airsheds is poor, including a dearth of information on the relationships between sources of fine particles (and their precursors) and ambient concentrations.

It should be noted that PM_{10} includes both combustion related particles which tend to be less than 2.5 micrometres (μg) in diameter ($PM_{2.5}$) and mechanically generated particles which tend to be greater than $PM_{2.5}$. In most scientific discussions only particles less than $PM_{2.5}$ are regarded as “fine” (in some circles fine particles are those less than PM_1 – as discussed in Section 2), while particles greater than $PM_{2.5}$ are regarded as coarse. It is now generally recognised that health effects are more strongly correlated with the fine rather than coarse particle concentrations.

Atmospheric particles, unlike most other air pollutants, cannot simply be described by a chemical formula, such as SO_2 (sulfur dioxide) or HF (hydrogen fluoride), for example. In most circumstances a collected sample of atmospheric particles will consist of a diverse and variable mixture of elements and compounds arising from potentially many different sources – both natural and anthropogenic in origin. Added to this complexity is the potential for fine particles ($PM_{2.5}$) to form in the atmosphere from the emission of primary pollutants – the formation of sulfate particles from gaseous sulfur dioxide being a well recognised example. Finally, these fine particles have the potential to form and remain suspended in the atmosphere for many hundreds of kilometres from the point of emission. These factors make the investigation and management of particles much more complex and difficult than the “simpler” air pollutants, like sulfur dioxide.

The most recent scientific investigations into particles and health impacts have focused on 3 main areas:

- The effect of fine particles (PM_{2.5} and less), as most of the early data is from studies which measured PM₁₀ and to a lesser extent, PM_{2.5}
- The relationship between particle levels averaged over time periods longer than 24 hours and health impacts and,
- The mechanism(s) via which particles might be causing the observed health impacts, which could be related to particle mass (which is the most commonly used descriptor for particles), particle number, particle chemistry or even particle morphology (shape). As discussed in Section 2, current research is addressing the fact that fine PM mass may be an indicator (but not a cause) of adverse effects associated with air pollution – other pollutants or PM components that co vary with PM mass may be the underlying cause.

1.1 Aim and Relevance of the Study

The current study was commissioned by the three New South Wales state owned, coal-fired electricity generating organisations to address the potential environmental and health implications arising from fine particles, either directly emitted from the seven coal-fired power stations in NSW, or which form in the atmosphere from emissions from these facilities.

Modern industrial particulate collection systems are very effective in removing particles from the exhaust gases of facilities – however some control technologies are relatively less effective in removing finer particles compared with coarse particles. Furthermore, unless flue gas scrubbing is employed the potential exists for the formation of secondary particles from sulfur dioxide and nitrogen oxide emissions.

Hence, coal fired electricity generation is a potentially significant source of fine particles in the atmosphere, due to:

- The emission of ash particles which pass through the particulate collection system and,
- The formation of secondary particles, sulfates and nitrates in particular, from the emission of sulfur and nitrogen oxides.

The current project was devised in response to the above considerations and has as its principal aim to:

provide the NSW coal-fired electricity generators with robust, credible information on the potential atmospheric concentrations of fine particles from power station operations. This will enable the electricity generating organisations to respond and contribute to policy developments in a well-informed, constructive manner.

It is recognised that there are uncertainties relevant to every stage of assessing the implications of fine particle emissions from coal-fired power stations, including:

- Characterising emissions,
- Understanding atmospheric formation and removal processes,
- Understanding the relative impacts from a large number of very different sources

Notwithstanding the uncertainties involved, it is considered possible to undertake a practical investigation, using available local and international data, and a suitably conservative approach to provide scientifically robust, credible information on the potential particle concentrations resulting from power station emissions, in both an absolute sense and relative to other significant sources.

The study has identified areas where the results could be strengthened (made less conservative) *if, and when*, the need arises in the future due to:

- Proposed regulatory developments
- Community concerns
- The initial assessment indicating some reasonable cause for more detailed investigation

1.2 Scope of the Study

The project consists of a number of tasks:

1.2.1 Task 1: Review

Scope - *to provide the context for the study – why are fine particles a health issue, what is known about their formation and occurrence, what are regulators doing about fine particles.*

1.2.2 Task 2: Characterise particle emission and formation processes

Scope – *to quantify the emission and formation of fine particles from power stations - **what** is known about fine particle emissions from coal fired power stations, what is known about the formation and deposition of fine particles in the atmosphere*

1.2.3 Task 3: Modelling assessment

Scope – *to quantify the atmospheric concentrations of fine particles in different regions of NSW due to power station operations –using emission rates from task 2, model the transport of power station emissions, hour by hour, over a full year and calculate atmospheric concentrations.*

1.2.4 Task 4: Synthesis

Scope – *to bring the results from the other tasks together and discuss power station emissions in context with other emission sources, existing air quality levels and existing and proposed air quality goals/standards.*

Should the results indicate a need, consideration would be given to possible further relevant work, such as a measurement and or analytical programs which could enable the occurrence of

fine particles arising from power station emissions to be more accurately quantified and/or assessed relative to other major sources.

1.3 The Project Team

The project team consisted of essentially the same personnel that successfully undertook the Inter Regional Transport of Air Pollutants Study (IRTAPS):

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Peter Nelson	Graduate School of the Environment, Macquarie University
Martin Cope	CSIRO Energy Technology and Atmospheric Research
Bill Lilley	CSIRO Energy Technology
Merched Azzi	CSIRO Energy Technology
Mary Edwards	CSIRO Atmospheric Research
Peter Hurley	CSIRO Atmospheric Research
John Carras	CSIRO Energy Technology

1.4 Structure of the Report

Section 2 of the report addresses Scope Item 1, providing a review of recent research into the health and environmental implications of fine particles, an overview of particle formation and occurrence and a summary of legislative responses to the issue. This section also reviews available local studies and monitoring data which assist in providing context for the modelled predictions.

Section 3 of the report provides a detailed summary of the modelling component of the project (Scope Item 3). CSIRO's detailed report on the modelling component is included as Appendix I to the report.

Scope Item 2, the characterisation of particle emissions and formation processes is partly addressed in both Sections 2 and 3 of the report.

Results arising from the project are discussed in Section 4; options which might enhance, or provide greater certainty to, the preliminary results generated in this study are considered. The major conclusions arising from the project are presented in Section 5.

Throughout the report, and particularly in the modelling component, care is exercised in explicitly acknowledging all assumptions employed in undertaking the assessment and limitations inherent in the approach adopted.

2 REVIEW

Particles in the atmosphere arise from a great variety of natural sources (windborne dust, sea spray, volcanoes and bushfires for example) and anthropogenic activities (combustion of fuels for transport, industry and domestic heating, non-combustion industrial activities and agricultural activities for example).

The potential environmental effects of atmospheric particles depend significantly on their characteristics, including size and chemical composition, which in turn depend significantly on how the particles were formed.

This section provides a brief overview of how atmospheric particles (or aerosols¹) form, how they are removed from the atmosphere, their chemistry and potential health and environmental impacts.

2.1 Particle Formation, Characteristics and Removal Processes

2.1.1 Formation and size

Particles can be directly emitted into the atmosphere (primary particles or primary aerosol) or formed in the atmosphere by gas-to-particle conversion processes (secondary particles or secondary aerosol). In this project the most important secondary aerosols considered are sulfate and nitrate, which form from the emission of sulfur and nitrogen oxides, respectively. Section 2.7 provides an overview of the potentially important chemical pathways involved in secondary aerosol formation.

Atmospheric aerosols are generally considered to be particles in the size range from a few nanometres (nm) to tens of micrometres (μm) in diameter. Once airborne, particles can change their size and chemical composition by condensation (or evaporation) of gas-phase species, by coagulation with other particles, by chemical reaction, or by activation, in the presence of water concentrations exceeding supersaturation, to become fog or cloud droplets.

Particles in the size range of 10 to 100 micrometers are on the large end of the particle size scale of interest in the field of air pollution. The USEPA has adopted the following four terms for categorising particles of different sizes.

Table 2.1: USEPA terminology for particle sizes.

<i>EPA Description</i>	<i>Particle size diameter μm</i>
Super-coarse	> 10
Coarse	> 2.5 and ≤ 10
Fine	> 0.1 and ≤ 2.5
Ultra-fine	≤ 0.1

¹ Aerosol is technically defined as a suspension of fine solid or liquid particles in a gas whereas more common usage often refers to the solid component only

This report uses “PM” to refer to particulate matter inclusive of all size fractions and adopts the above USEPA definitions, which are generally consistent with accepted international scientific definitions of size fractions. It should be noted that these definitions may be inconsistent with some other local definitions under which PM₁₀ may be considered as “fine”.

The principal source of ultra-fine and fine particles are emissions from combustion processes, whereas coarse particles tend to be formed from the mechanical processes which breakdown material from a variety of sources into dust. Particles larger than a few tens of μm also tend to have a limited residence time in the atmosphere due to gravitational settling and deposition.

Figure 2.1 shows that the distribution of particles measured in urban air falls into three main modes based on their aerodynamic diameter², which in turn depends on formation processes.

- Nuclei mode – smaller than about 0.1 μm .
- Accumulation mode – between 0.1 and $\sim 2.5 \mu\text{m}$.
- Coarse mode – larger than $\sim 2.5 \mu\text{m}$.

The nuclei mode extending from about 0.005 to 0.1 μm diameter accounts for the majority of particles by number; because of their small size, ultra-fines seldom account for more than a few percent of the total mass of airborne particles. Particles in the nuclei mode are formed from condensation of hot vapours during combustion processes and from the nucleation of atmospheric species to form fresh particles.

Particles in the nucleation mode do not last long in the atmosphere, perhaps having residence times measured in minutes, as they coagulate (two or more particles combining) or condense (gas molecules condensing onto a solid particle). They are always present at some level, however, as they are constantly being generated by emissions from combustion processes. Lidia Morawska and colleagues (Morawska 2004) has recently completed a comprehensive review of the health impacts of ultra-fine ($< 0.1 \mu\text{m}$ diameter) particles, which also includes a comprehensive review of the sources and characteristics of atmospheric particles.

The accumulation mode, extending from about 0.1 to about 2.5 μm usually accounts for most of the aerosol surface area and a substantial part of the aerosol mass. The source of particles in the accumulation mode is the coagulation of nuclei mode particles and from condensation of vapours onto existing particles, causing them to grow into this size range. The accumulation mode is so-named because particle removal mechanisms are least efficient in this regime, allowing particles to accumulate in the atmosphere, having atmospheric residence times measured in weeks. An

² The term "aerodynamic diameter" has been developed by aerosol physicists in order to provide a simple means of categorizing the sizes of particles having different shapes and densities with a single dimension. The **aerodynamic diameter** is the diameter of a spherical particle having a density of 1 gm/cm^3 that has the same inertial properties [i.e. terminal settling velocity in the gas as the particle of interest.]

important mechanism for removal of this size range of particles is scavenging in water droplets and subsequent precipitation processes.

Since the sources contributing to the formation of particles in the ultra-fine and coarse particle size ranges are different, correlation between fine and coarse particles is frequently poor.

Figure 2.1 also shows alternative particle size definitions used in health studies. Ultra-fine particles correspond to particles in the nuclei mode. Fine particles include both nuclei and accumulation mode particles. Also shown is the relationship between the current “regulatory” size categories (PM_{10} and $PM_{2.5}$) and the formation modes. The following section on PM and health effects discusses “inhalable”, “thoracic” and “respirable” particles in relation to different sizes and their ability to enter the human respiratory system.

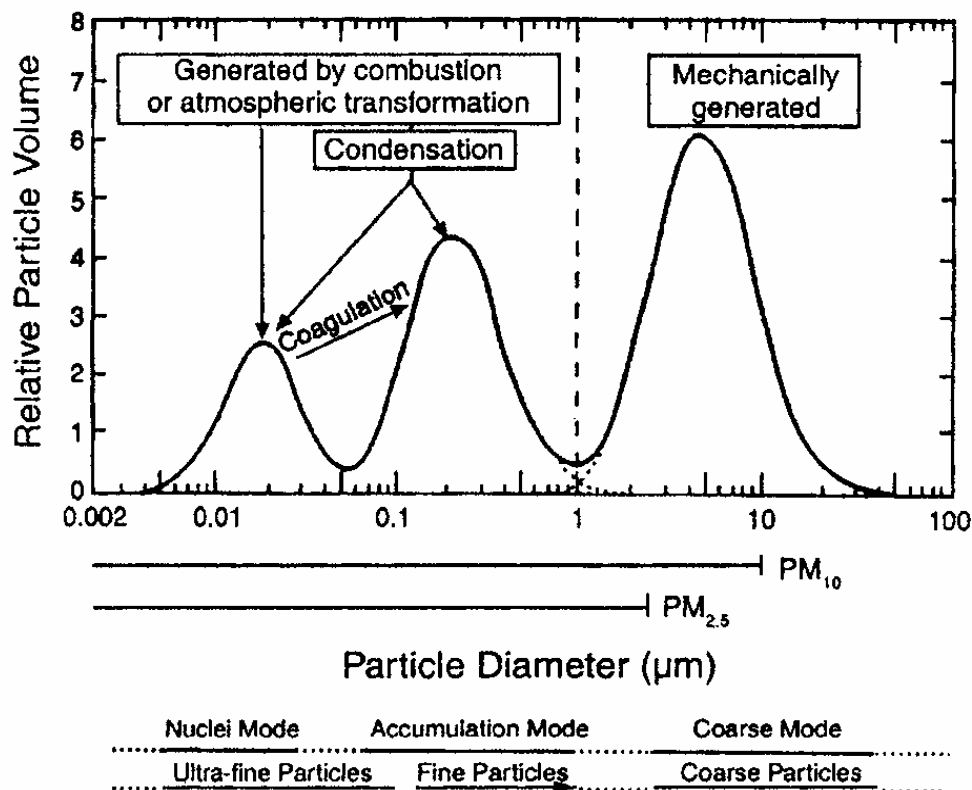


Figure 2.1: Typical distribution of three sizes or modes of particles and how different definitions of particle size relate to these definitions. (Source: HEI April 2002)

Figure 2.1 also illustrates that the largest particles ($> 1 \mu m$) form the highest proportion of the mass of particles in the atmosphere; the smallest particles comprise only 1 – 8 % of this mass. However, it should be recognised that while low in mass, ultra-fine particles are present in very high numbers and as is discussed in Section 2.2 the size of particles has a significant bearing on

their potential health impacts, which has lead to some criticism of current regulatory standards which are based on mass concentration.

2.1.2 Removal processes

As discussed above, the ultra-fine particles have very short residence times in the atmosphere as they tend to quickly form “larger” particles via coagulation and / or condensation processes.

Fine and coarse particles are removed from the atmosphere by two processes:

- Dry deposition –gravitational settling and turbulent transfer to the Earth’s surface.
- Wet deposition – incorporation of particles into cloud droplets and transfer to the surface in rain events.

2.1.3 Particle composition

The composition of PM can vary greatly spatially and temporally due to the diversity of sources as well as climatic factors which will affect the formation of secondary PM as well as the residence time of PM.

In general, the composition of coarse and super coarse particles consists mainly of crustal-derived minerals, biological material (pollen, for example) and sea salt. By contrast, ultra-fine and fine fractions are composed of products of combustion, including: elemental carbon, and a complex mixture of organic compounds and sulfates and nitrates, which may be neutralised by the ammonium ion (NH_4^+). Some metals may also be associated with the fine PM.

Information on the composition of PM in NSW is presented in Section 2.6

2.1.4 Sources of particles

As discussed in the introduction to this section particles are emitted by a diversity of natural and anthropogenic sources. Source strengths of both natural and anthropogenic particles can vary significantly both temporally and spatially.

Table 2.2 shows an estimate of aerosol particle emissions on a global scale. Several points are worth noting from the data presented:

- While the natural emission and formation of particles of all sizes dwarfs the contribution from anthropogenic sources on a global scale, anthropogenic sources of fine particles are greater than natural sources.
- Natural particles are dominantly $> 1 \mu\text{m}$
- Anthropogenic particles are dominantly $< 1 \mu\text{m}$
- Secondary sulfate is all fine whereas secondary nitrate can occur in both fine and coarse modes.

As is often the case with air pollutants, anthropogenic emissions have the potential to cause adverse impacts as their atmospheric concentrations can often be significant in populated areas. Concentrations of concern are most likely to occur under adverse weather conditions, such as ground-based inversions and light winds. In contrast, emissions from natural sources tend to be dispersed over much larger scales, but still may be associated with health impacts under adverse weather conditions.

Table 2.2: Global emission estimates for major aerosol types in the 1980s. (Source: Seinfeld and Pandis 1998)

SOURCE	FLUX, Mt / year ¹	Particle size category ²
NATURAL		
Primary		
Soil dust (mineral aerosol)	1,500	Mainly coarse
Sea salt	1,300	Coarse
Volcanic dust	30	Coarse
Biologic debris	50	Coarse
Secondary		
Sulfates from biogenic gases	130	Fine
Sulfates from volcanic SO ₂	20	Fine
Organic from biogenic VOC ³	60	Fine
Nitrates from NO _x	30	Fine and coarse
Total	3,100	
ANTHROPOGENIC		
Primary		
Industrial / mining dust	100	Fine and coarse
Soot (elemental carbon) from fossil fuels	10	Mainly fine
Secondary		
Sulfates	190	Fine
Biomass burning	90	Fine
Nitrates	50	Mainly coarse
Organic from anthropogenic VOCs	10	Fine
Total	450	
TOTAL	3,600	

1. Fine and coarse refer to mean particle diameter below and above 1 µm, respectively.
2. Reported as “best” estimates, with significant variability between low and high estimates particularly for natural sources.
3. Volatile organic compounds

Table 2.3 shows that in urban areas of Australia the estimated contribution from different anthropogenic source categories to particle emissions varies significantly. It should be noted that data in this table do not include secondary particles, which can be difficult to estimate in an

airshed. Furthermore, the significant and perhaps anomalous, differences between source contributions in the different airsheds would suggest that caution is required in making generalisations and comparisons without first understanding the underlying causes of the differences reported.

Table 2.3: Particle emissions from various source types (yearly average, as % of total) (NEPC, 1998)

Airshed	Mobile sources	Industrial Sources	Area-based sources
Sydney	30	34	36
MAQS Region ³	16	67	16
Port Philip region Vic.	16	10	74
SE Queensland	18	65	17
Perth-Kwinana	8	68	24
Port Pirie SA	2	94	4
Launceston Tas	1	2	97

2.2 PM and Health Impacts

This section provides a review of information on the relationships between particulate matter and health impacts. It starts by presenting a brief overview of how PM interacts with the respiratory system and includes information on what is currently known about the significance of PM characteristics, including size and chemical components.

2.2.1 PM interaction with the respiratory system – An overview

The respiratory system is very efficient at clearing itself of most inhaled particles. Many particles, particularly the larger ones, are trapped and expelled by the body's first line of defence which is a barrier of cells and fluids which prevent the particles entering the tissues of the body. Fluid secretions, such as mucous lining the airways and ciliated cells are important elements in this system.

Figure 2.2 provides a general representation of how particles of varying sizes interact with the major components of the human respiratory system. Particles of a diameter above 50 µm are rarely inhaled at all, as the air velocity of inspiration is usually too slow for them to be sucked into the nose.

Figure 2.2 shows that PM above about 10 µm in diameter is trapped in the nose and throat from where it is either expelled or swallowed.

Generally, for particles smaller than 10 µm, penetration increases with decreasing particle size. The upper size limit for penetration into the alveoli of the lung is considered to be approximately 10 µm, but most of the particles of this size will be deposited in the bronchial tree from where they can generally be moved back into the throat by mucociliary action and then expelled.

³ Metropolitan Air Quality Study Region of NSW, which includes the Newcastle, Sydney and Wollongong urban regions.

Maximum alveolar penetration and deposition occurs with particles less than about $3\text{ }\mu\text{m}$ diameter. The fine particles can penetrate into the lung and if reaching the alveoli can potentially enter the bloodstream.

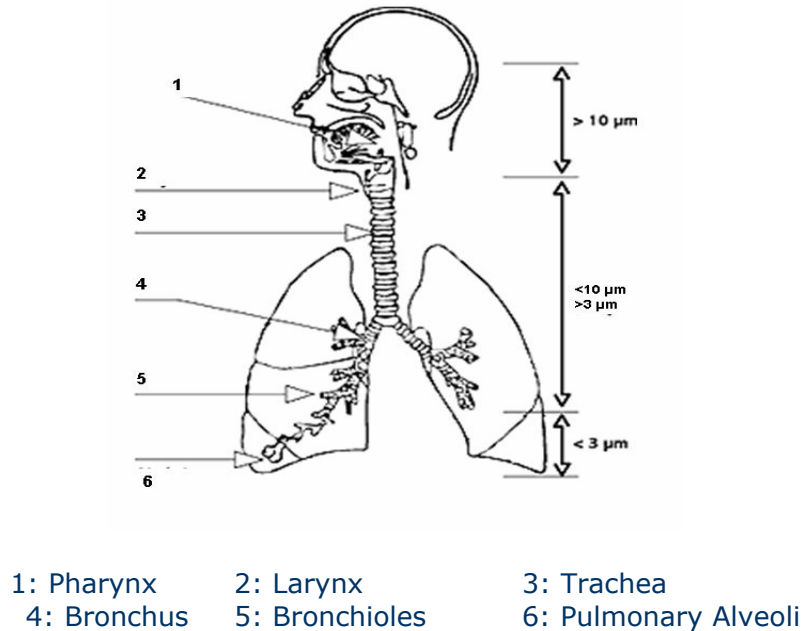


Figure 2.2: General representation of the interaction of PM of different sizes and the human respiratory system.

At this point a second line of defence comes into play; “scavenger” cells ingest the foreign material and attempt to destroy it. The most important scavenger cells are macrophages (white blood cells that reside in the tissues and air spaces of the lungs) and neutrophils (white blood cells found in the bloodstream). If this defence layer is overwhelmed, lymphocytes (another type of white blood cell) and the proteins that they synthesise become involved which may result in an inflammatory response and epithelial cell damage, changes in blood properties and possibly changes in the nervous system’s control of breathing and heart pattern.

Particle deposition in the airways can trigger events in many different cells, potentially resulting in changes in tissues and organs at sites progressively further from in the initial stimulus. These defence mechanisms are normal responses in healthy individuals but they may lead to adverse impacts.

In the 1980s and early 1990s, committees from the International Organization for Standardization (ISO), the American Conference of Governmental Industrial Hygienists (ACGIH), and the Comité Européen de Normalisation (CEN) developed internationally accepted definitions of what the sampling criteria for PM should be, based on understanding of the interaction of PM with the respiratory system. These committees agreed that health-related sampling should be based on one or more of three progressively-finer size fractions: inhalable, thoracic, and respirable.

Inhalable: The inhalable fraction is the mass fraction of particles which can be inhaled by nose or mouth. Since there are no experimental data on inhalable particles with an aerodynamic diameter of $>100\text{ }\mu\text{m}$, particles $>100\text{ }\mu\text{m}$ are not included in the inhalable convention.

Thoracic: The thoracic fraction is that portion of the inhalable particles that pass the larynx and penetrate into the conducting airways (trachea, bifurcations) and the bronchial region of the lung. The median value of the particle size is $11.64\text{ }\mu\text{m}$ with a geometric standard deviation (GSD) of $1.5\text{ }\mu\text{m}$. It has been shown that 50% of the particles in air with an aerodynamic diameter of $10\text{ }\mu\text{m}$ belong to the thoracic fraction; ($D_{50} = 10\text{ }\mu\text{m}$).

Respirable: The respirable fraction is the portion of inhalable particles that enter the deepest part of the lung, the non-ciliated alveoli. The median value is $4.25\text{ }\mu\text{m}$ with a GSD of $1.5\text{ }\mu\text{m}$. It has been shown that 50% of the particles with an aerodynamic diameter of $4\text{ }\mu\text{m}$ belong to the respirable fraction ($D_{50} = 4\text{ }\mu\text{m}$).

Two additional size fractions are sometimes discussed:

Extrathoracic: The extrathoracic fraction of inhaled particles are those that fail to penetrate beyond the larynx. The extrathoracic fraction is obtained by subtracting the thoracic fraction from the inhalable.

Tracheobronchial: The tracheobronchial fraction of inhaled particles are those that penetrate beyond the larynx but fail to reach the alveoli. The tracheobronchial fraction is obtained by subtracting the respirable fraction from the thoracic.

It is important to note that these size conventions are only approximations to the behavior of particles in the human respiratory tract of healthy adults. Actual particle penetration and deposition will depend on the physical variations in individuals, breathing rate, and on whether one is breathing through the nose or mouth.

2.2.2 Impacts and effects of PM_{10} and $\text{PM}_{2.5}$

The relationship between exposure to air pollutants and potential health impacts has been recognised for many years, at least since increasing industrial development in Europe resulted in large increases in emissions of black smoke and acid gases. The quantitative relationship between extreme air pollution events and excess mortality has also been established for around 50 years, since the famous “London Smog” of 1952. In that event, a strong rise in air pollution levels, particularly particles and SO_2 , was followed by sharp increases in mortality and morbidity. A recent re-analysis (Bell and Davis, 2001) of the London Smog estimates that about 12,000 excess deaths occurred from December 1952 through February 1953 because of acute and persisting effects of the event. Pollution levels during the London smog were 5 - 19 times above current UK and other international regulatory standards and guidelines (Bell and Davis, 2001) and were similar to current levels in some rapidly developing regions. Figure 2.3 shows the relationship between mortality and SO_2 for the London Smog (from Bell and Davis, 2001).

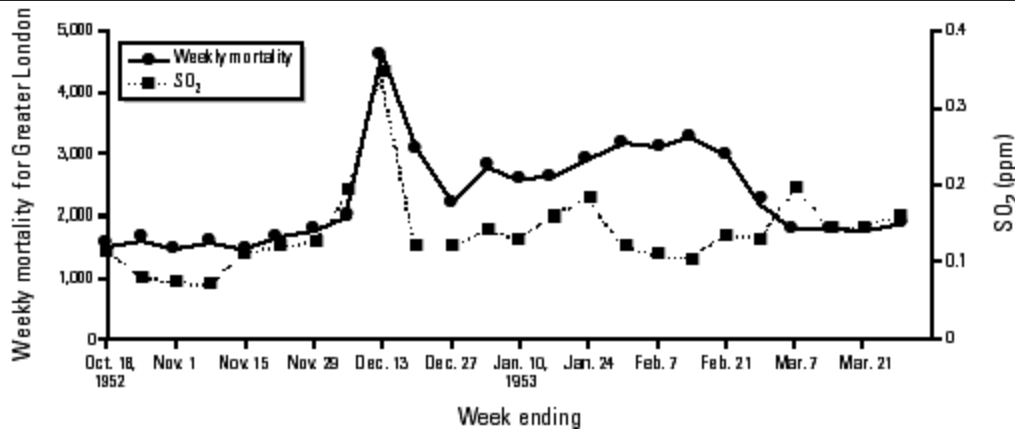


Figure 2.3: The relationship between mortality and air pollution during, and following the London “Smog” Event of 1952.

Effects of long term exposure to lower levels of pollutants are clearly more difficult to establish, in part because of the difficulties in separating the impacts of confounding factors on health outcomes, such as weather, lifestyle and occupation. However recent epidemiological research, based on long term observations in cities in the developed world, has consistently revealed an association between air pollution and human health indicators.

In particular, statistical analyses of urban air pollution worldwide have revealed a correlation between PM concentrations and short term impacts on health (Dockery et al, 1993; Wilson and Spengler, 1996; HEI, 2002). Recent results (Pope et al, 2002) have extended these findings to long term impacts. For example, Pope et al (2002) found that each $10 \mu\text{g m}^{-3}$ increase in the concentration of fine particles ($\text{PM}_{2.5}$) was associated with an 8% increased risk of lung cancer mortality. A similar magnitude of impacts has been observed worldwide, including in Sydney (Morgan *et al*, 1998).

Figure 2.4 presents a summary of the magnitude of the effects observed for PM_{10} from a number of studies (summarised from HEI 2001,2002, 2003). The figure shows good agreement between results obtained in the United States, Europe and from a “meta-analysis” of 29 studies in 23 locations in Europe and North and South America. In some contrast are the revised results for the 90 United States cities which are discussed later in this section.

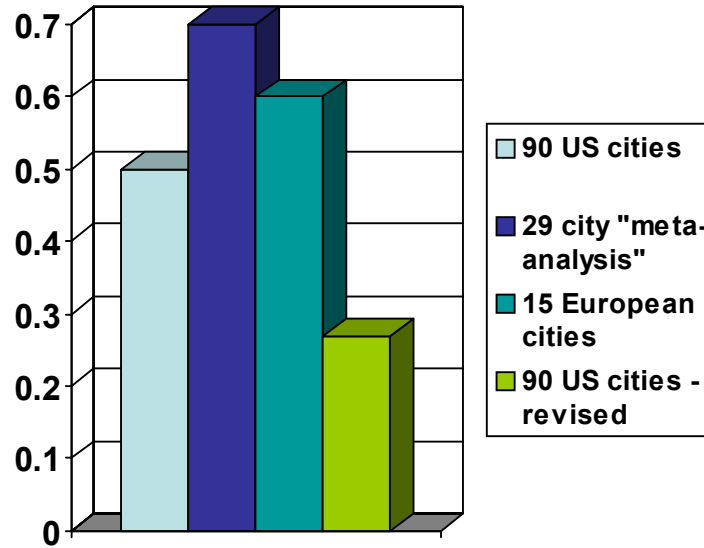


Figure 2.4: Percent excess mortality for 10 µg m⁻³ increase in PM₁₀ (summarised from HEI, 2001, 2002 and 2003).

Table 2.4 provides a summary of data arising from recent studies into PM_{2.5} concentrations and a range of short-term and long-term health end points. Table 2.5 summarises health endpoints in relation to PM_{2.5} in a number of Australian capital cities.

In summary, these studies suggest that atmospheric particles have substantial impacts on human health with more recent data indicating PM_{2.5} has more significant impacts than PM₁₀. A feature of the many studies which have now been conducted in many different countries is the convergence of the results obtained.

Detailed analyses of the data reveal a range of effects, as seen in the tables, and these include (Brasseur *et al*, 2003):

- Increased daily mortality; including all cause, respiratory and cardiovascular deaths
- Increased rates of hospital admission with respiratory diagnoses, including visits to emergency departments
- Exacerbation of asthma; including asthmatic attacks, fluctuations in the prevalence of bronchodilator use, emergency department visits, and hospital admissions
- Increases in respiratory symptom reports; including lower and upper respiratory and cough
- Decreases in lung function; including forced expiratory volume and peak expiratory flow reductions.

Table 2.4: PM_{2.5} studies and dose response relationships (NEPC 2002)

Health Endpoint	Age Group	Dose-response % increase per 10 µg m ⁻³ increase in PM _{2.5}	95% Confidence interval
Short-term effects 24-hour)			
<u>Mortality</u>			
All cause	All ages	2.3	1.3 – 3.3
Respiratory	All ages	8.6	5.2 – 12.4
Cardiovascular	All ages	1	0.15 – 1.9
<u>Hospital Admissions</u>			
Asthma	All ages	2.6	1 – 4.2
Cardiovascular disease	Elderly	1.7	1- 2.4
COPD ⁴	Elderly	2.6	0.4 – 4.8
Long-term effects (annual average)			
<u>Mortality</u>			
All cause	All ages	6	3 - 11
Lung cancer		14	4- 23
Cardiopulmonary disease		9	3 - 16

Table 2.5: Health effects (number of events) attributable to current levels of PM_{2.5} in Sydney, Melbourne, Brisbane and Perth (NEPC 2002)

	ShortTerm Health Endpoint						Long Term Health Endpoint		
	<i>Mortality</i>			<i>Hospital Admissions</i>			<i>Mortality</i>		
	All cause	Respira-tory	Cardio-vascular	Asthma	Cardio-vascular disease	COPD	All cause	Lung cancer	Cardio-pulmonary disease
Sydney	274	81	55	157	246	58	699	88	527
Melbourne	207	60	41	78	157	15	524	58	316
Brisbane	97	32	20	37	63	10	226	26	143
Perth	52	19	10	27	50	10	142	20	97
TOTAL	632	193	127	302	523	94	1611	195	1096
Including 2001 major bushfires									
Sydney	290	85	58	167	262	61	743	93	560
Brisbane	99	33	21	41	71	11	252	29	160

According to Morawska (2004) the number of studies addressing the association between ambient ultra-fine concentrations and mortality or morbidity is relatively small (8) compared with larger sized particles. The studies are limited to the investigation of acute health effects due to short-term exposure. Morawska summarises the findings from these studies which include:

⁴ Chronic obstructive pulmonary disease

- Mortality data suggest that fine particles have immediate health effects whereas ultra-fine particles have more delayed effects. Immediate effects seem to be related to respiratory disease whereas delayed effects are based on an increase in cardiovascular disease mortality.
- There is an indication that the acute effects of the number of ultra-fine particles on respiratory health are stronger than those based on the mass of the fine particles.

Uniquely for a “criteria” air pollutant, the interpretation of these epidemiological studies has not indicated a threshold below which no effects occur. On this basis, the World Health Organisation (WHO) decided not to recommend a health goal for particulate matter, at this stage, on the grounds that “*The available information does not allow a judgement to be made of concentrations below which no effects would be expected.*”

WHO has developed guidelines for PM but these guidelines do not include a limit or target value(s) (WHO, 2000). The summary provided by WHO (2000) states:

The weight of evidence from numerous epidemiological studies on short-term responses points clearly and consistently to associations between concentrations of particulate matter and adverse effects on human health at low levels of exposure commonly encountered in developed countries. The database does not, however, enable the derivation of specific guideline values at present. Most of the information that is currently available comes from studies in which particles in air have been measured as PM₁₀. There is now also a sizeable body of information on fine particulate matter (PM_{2.5}), and the latest studies are showing that this is generally a better predictor of health effects than PM₁₀. Evidence is also emerging that constituents of PM_{2.5} such as sulfates and particle strong acidity are sometimes even better predictors of health effects than PM_{2.5} per se.

The large body of information on studies relating day-to-day variations in particulate matter to day-to-day variations in health provides quantitative estimates of the effects of particulate matter that are generally consistent. Effects on mortality, respiratory and cardiovascular hospital admissions as well as other health variables have been observed at levels well below 100 µg m⁻³, expressed as a daily average PM₁₀ concentration. For this reason, no guideline value for short-term average concentrations is recommended either.

There have, however, been some recent developments in the analysis of PM and health effects data which has resulted in a revision of the magnitude of the effects. In addition, there has also been recent evidence for a threshold below which some effects are not observed.

In 2002, researchers at John Hopkins University and at Health Canada identified problems with the statistical model used in the majority of the studies of health effects (HEI, 2003). In response, much of the statistical data in the US were re-analysed using modified procedures which addressed the identified problems. The conclusions from this re-analysis (HEI, 2003) included the following points:

- In general, the estimates of effects decreased substantially, but the qualitative conclusions did not change.
- Across the 90 cities included in the Health Effects Institute (HEI) funded National Morbidity, Mortality and Air Pollution Study (NMMAPS), the revised mean effect on mortality decreased from 0.41% increase per 10 $\mu\text{g}/\text{m}^3$ increase in PM_{10} concentration to 0.21-0.27%, an overall decrease of nearly 50%.
- Smaller decreases in effects (8-10%) were found in estimates for hospitalisations for cardiovascular diseases and for chronic obstructive pulmonary diseases.
- The effect on pneumonia hospitalisations was substantially reduced

The issue of thresholds has also been extensively examined recently. The HEI summary (HEI, 2004) of recent work in this area makes the following points:

- Earlier results from most statistical studies of association between PM and health impacts, suggested a linear relation between concentration and daily mortality over the entire range of ambient PM_{10} concentrations in the continental US. That is, there appeared to be no concentration (or threshold) beneath which adverse events were not observed.
- In the most recent work the concentration-mortality relation was evaluated for the largest 20 US cities, using methods that address the shortcomings in the statistical methods alluded to above.

Based on the result, the researchers conclude that:

- For total mortality and cardiovascular-respiratory mortality there was no evidence for a threshold down to concentrations as low as 10 $\mu\text{g m}^{-3}$, and conclude that linear models without a threshold were appropriate for assessing effects
- For mortality from other causes, however, there was little evidence of an effect until PM_{10} concentrations were above 50 $\mu\text{g m}^{-3}$, and conclude that a threshold model would be reasonable for assessing the effect of PM_{10} on other cause mortality

In summary, these recent results summarised by the HEI reports (HEI, 2003; 2004) have resulted in a reduction in the estimates of the effects, and provide some evidence for a threshold for some effects.

2.2.3 Current studies – Composition of ambient particulate matter and health effects

Section 2.1.3 described the variable composition of particulate matter. It is now well-established that the fine ($\text{PM}_{2.5}$) fraction of ambient PM largely consists of carbon (elemental and organic), metals, sulfate, and nitrate. The relative contributions of these components varies spatially and temporally and will be determined, inter alia, by proximity to sources, time of day and year, and other factors.

Recent reviews (Lighty et al, 2000; Jacobson et al, 2000; Monn, 2001) have summarised the current understanding in the area of PM composition and health effects and what is currently known about the size and composition of combustion aerosols and the organic fraction, and also of the spatial variability in composition. It is beyond the scope of this report to review this information, since much relates to urban aerosols produced from combustion of liquid fuels and

wood, and to secondary organic aerosols. Some comments on the composition and formation mechanisms for particles from coal combustion are, however, relevant. These are given in a following section.

There remains intense activity in the area of PM and health effects, and particularly in investigating causal relations between fine particle composition and health effects. The introduction to Okeson *et al* (2003) summarises recent studies, with a particular emphasis on combustion generated fine particles. Briefly, the key issues are considered to be:

- The magnitude of the impact of PM on human health depends on PM mass, size distribution, composition (polyaromatic hydrocarbons and metals such as Fe, V and Zn), the presence of biogenic components (endotoxins, pollens, bacteria, viruses) and other factors.
- Particle size distribution “*appears to have a modulating effect on the degree of toxicity beyond that anticipated due merely to deposition issues.*” – as discussed earlier, there is evidence of increasing toxicity with decreasing particle size.
- Relative roles for soluble and insoluble components of PM require further elucidation.

A major study of atmospheric concentrations, exposure assessment and health impacts is being conducted by the Electric Power Research Institute (EPRI) in the Aerosol Research and Inhalation Epidemiology Study (ARIES). An overview of ARIES is provided in a fact sheet available from the EPRI web site (EPRI, 2004). The objectives of ARIES are to:

- Investigate via epidemiology and exposure studies associations between air quality and human health
- Provide input for consideration of the National Ambient Air Quality Standard (NAAQS) and for subsequent development of State Implementation Plans (SIPs)

What distinguishes ARIES from predecessor studies is its focus on an unprecedented range of potential agents in the air, including Volatile Organic Compounds (VOCs), aeroallergens, and specific PM components, in addition to PM mass (the basis for most previous investigations).

The factsheet (EPRI, 2004) also summarises results to date in ARIES. Of note are the following findings:

- Daily mortality results show that the best model fits for all-cause mortality in those aged 65 and older are observed for CO, NO₂, PM_{2.5}, coarse PM, SO₂ and ozone, followed by elemental and organic carbon.
- Hospital emergency department visits for cardiovascular disease are associated with NO₂, CO, elemental carbon (EC) and organic carbon (OC).
- Hospital emergency department visits for respiratory visits are associated with PM₁₀, PM_{2.5}, PM_{2.5} – water soluble metals, NO₂, CO and SO₂

Studies of the associations between, on the one hand, mortality and other health effects, and, on the other, particle mass, show relatively consistent magnitudes of effects (as seen, for example, in Figure 2.4). In some of these studies, these associations have also been explored for components of the fine particles, but in this case there is significantly less agreement and consistency. In the original “Six Cities” Study (Dockery et al, 1993), it is reported that: “Mortality was most strongly associated with air pollution fine particulates, including sulfates”. HEI reanalysis (2001) of the Six Cities Study results replicated the original results as well as assuring the quality of the data. HEI identified relatively robust associations of mortality with two indices of particle matter (PM_{2.5} and sulfate) and with SO₂.

The data from the “Six Cities” Study was also used to examine associations between mortality and fine PM from different sources (Laden et al, 2000). In this work chemical tracers were used to attribute source contributions to PM. Lead was used as a tracer for motor vehicles; selenium for coal combustion sources, and silicon as an indicator of crustal material. On this basis, crustal material was found to be not associated; motor vehicle sources were found to be responsible for 3.4% per 10 µg/m³ increase in PM_{2.5}; and coal combustion sources were found to be responsible for 1.1% per 10 µg/m³ increase in PM_{2.5}.

By contrast, recent results of the ARIES study (Klemm et al, 2004), discussed above, reveal a somewhat different picture. These results, derived from a very extensive field sampling campaign in Atlanta, show that the pollutants most strongly associated with observed health effects appear to be CO and carbon-containing particles. However the details of the effects are complex, and clearly require additional work in other locations to derive more general conclusions.

The Health Effects Institute (HEI) is also sponsoring work examining causal relationships between health impacts and exposure to PM. Of particular relevance to FIPARTS, is a study by Aust and co-workers (Aust *et al*, 2002) on the effects of metals bound to PM on lung cells. The basis for this study was the hypothesis that inhaled coal fly ash could be a health hazard because metals solubilized from fly ash within lung cells may cause toxic reactions. It was found (Aust *et al*, 2002) that:

- Coal fly ash particles entered lung cells and stimulated synthesis of the protein ferritin. Ferritin binds iron and is produced in response to increasing iron levels;
- The presence of ferritin indicates that iron was released intra-cellularly, and was available to provoke an inflammatory response by forming reactive oxygen species.
- There was indirect evidence for formation of intracellular reactive oxygen species since lung epithelial cells exposed to coal fly ash synthesized the inflammatory mediator interleukin-8.

The investigators concluded that there is a plausible connection between the intracellular release of a transition metal from particles, formation of reactive oxygen species, and lung inflammation.

Table 2.6 provides an overview of current understanding of the biological effects of the various components of the PM. It is clear from this brief summary that the inter-related effects of particle size, composition and other characteristics and health effects are not yet completely

understood, which in part may be attributable to the significant temporal and spatial variation observed in PM characteristics.

Table 2.6: Chemical components of PM₁₀ and their biological effects (HEI April 2002)

Component	Major subcomponents	Described Biologic Effects
Metals	Iron, vanadium, nickel, copper, platinum and others	Can trigger inflammation, cause DNA damage, and alter cell permeability by inducing production of reactive oxygen species (particularly hydroxyl free radicals) in tissues.
Organic compounds	Many are adsorbed onto particles; some volatile or semi volatile organic species from particles themselves.	Some may cause mutations, some may cause cancer, and others can act as irritants and can induce allergic reactions.
Biologic origin	Viruses, bacteria and their endotoxins, animal and plant debris (such as pollen fragments) and fungal spores.	Plant pollens can trigger allergic responses in the airways of sensitive individuals; viruses and bacteria can provoke immune defence responses in the airways.
Ions	Sulfate (usually as ammonium sulfate) Nitrate (usually as ammonium / sodium nitrate) Acidity (H ⁺)	Sulfuric acid at relatively high concentrations can impair mucociliary clearance and increase airway resistance in people with asthma; acidity may change the solubility (and availability of metals and other compounds adsorbed onto particles).
Reactive gases	Ozone, peroxides, aldehydes	May adsorb onto particles and be transported into lower airways, causing injury.
Particle core	Carbonaceous material	Carbon induces lung irritation, epithelial cell proliferation, and fibrosis after long-term exposure.

2.3 Health Costs Attributable to PM

Significant controversies and uncertainties pervade attempts to estimate human health costs associated with air pollution in “dollar terms”, not-the-least of which involve attempts to value a shortened life-span or pain and suffering by individuals and their families. Notwithstanding the significant difficulties, estimates of the cost of the health impacts associated with air pollution are undertaken, the most notable probably being the USEPA (1997) study “Benefits and Costs of the Clean Air Act”. These studies suggest that a significant component (several percent) of Gross Domestic Product (GDP) can be attributed to the population's exposure to fine particles (see also, London and Romieu, 2000 who provide an analysis for three European countries).

The assessments undertaken for the development of the Ambient Air NEPM (NEPC 1998, 2002) suggested that in Australia compliance with the NEPM PM₁₀ standard might avert health costs accounting for approximately 0.7% of GDP. Note that complying with the NEPM would not avert all health costs associated with PM₁₀ as the standard is not set at a “no-effect” level.

In 2002, a more detailed estimate of health costs was undertaken for the development of the NEPM PM_{2.5} Variation. This study suggested averted health costs gained by compliance with the PM_{2.5} reporting standard would be in the order of 0.6% of GDP taking into account morbidity costs only. Costs associated with mortality were not included due to reasons discussed above, but if included could push the total costs out to something like 10 billion dollars, or approximately 1.5% of GDP, depending on the methodology used. Note again, that the full costs would be greater than this figure, as the NEPM standard is not set at “no-effect” level.

2.4 PM and Environmental Impacts

2.4.1 Visibility, Climate, Ecosystems

As well as the effects on human health discussed in the previous section, fine particles also have potentially significant environmental impacts on visibility, climate and atmospheric properties and more broadly on ecosystems through deposition processes.

Fine particles, or aerosols, in the particle size range 0.2 - 2 µm, are the major contributors to visible light scattering in the atmosphere which determines visibility, a key indicator of air quality to the general public. Fine PM can cause a significant decrease in visibility resulting in a loss of visual amenity, economic, social and other impacts, the potential significance of which are notably demonstrated by the establishment of the Grand Canyon Visibility Transport Commission in the USA to advise the U.S. Environmental Protection Agency on strategies for protecting visual air quality at national parks and wilderness areas on the Colorado Plateau (Grand Canyon Visibility Transport Commission 1996).

In addition, it is now recognised that natural and anthropogenic aerosols play a substantial role in the radiative properties of the atmosphere (Brasseur *et al*, 2003). It is calculated that the largest negative forcing (ie cooling) is due to aerosol particles of human origin (IPCC, 2001).

The text by Brasseur *et al* (2003) provides an excellent overview of the current understanding of the impacts of aerosols on climate and atmospheric processes. Key issues summarised in that reference include:

- Effects of aerosols on the radiation in the atmosphere:
 - direct radiative effects, where aerosols both scatter and absorb incoming solar radiation
 - indirect effects, which refers to potential changes to cloud properties at the global scale due to perturbations (man-made) in the concentrations and properties of particles that form cloud drops or ice crystals. These effects are the most uncertain of the radiative forcing mechanisms.
- Aerosol effects on actinic flux, where aerosols scatter and absorb incoming UV radiation; which can have effects on photolysis rates and hence atmospheric chemistry.
- Aerosols can act as sources or sinks of reactive trace gases.

Components of atmospheric aerosols are also known to affect ecosystems, both directly and indirectly. The impacts may range from elimination of sensitive species and altered productivity to changes in the relative abundance of plant and animal species (Brasseur *et al*, 2003). Direct effects include deposition of acidic particles; indirect impacts may result in changes in the chemical balance of soils, lakes and surface ocean waters following the deposition of acidifying and reducing substances, such as sulphates, nitrates and ammonium. The nitrogen-containing species may also have impacts on the overall nutrient load in catchments or surface waters.

Local studies by CSIRO suggest that the aerosol contribution to total acid deposition is “inconsequential” compared with the wet and dry deposition fluxes and that total deposition fluxes are generally below levels which would result in long-term adverse environmental impacts (Ayers *et al* 1995, 1997).

2.5 Air Quality Guidelines and Standards

While the health evidence would appear to provide compelling reasons for regulatory actions to control and limit atmospheric concentrations of fine particles, the prompt development of regulatory measures has been tempered by a number of issues, including:

- Debate over the magnitude of the association between PM concentrations and health effects, including the recent reductions in the estimated effects, as a result of reanalysis of the health data.
- Uncertainty over the issue of threshold concentration(s).
- The absence of clear identification of cause and effect relationships.
- The contribution of natural sources to particle levels (bushfires, dust-storms for example).
- Uncertainty over the relative contributions from many anthropogenic sources to atmospheric concentrations.
- Uncertainty over the most cost-effective means of controlling atmospheric concentrations.

As noted in the section on health effects (Section 2.2) the World Health Organisation has decided not to recommend a health goal for particulate matter, at this stage, on the grounds that “*The available information does not allow a judgement to be made of concentrations below which no effects would be expected.*”

While recognising the uncertainties regarding PM and health impacts, regulatory agencies which are charged with the task of managing air quality are being required to develop air quality guidelines and standards for particulate matter. On the information currently available, setting an air quality standard for PM involves accepting a level of risk, in that it is not possible to set the standard at a “no-effect” level. Accepting a degree of risk is inherent in the normal standard setting process, but in the case of particles is more problematic because of the association between particle concentrations at very low concentrations and increased morbidity and mortality.

Table 2.7, Table 2.8, Table 2.9 and Table 2.10 show ambient air quality standards and guidelines adopted or proposed for a number of countries / jurisdictions, including Australia. As might be

expected, given the difficulties and uncertainties involved, there is considerable variation between the approaches adopted in different jurisdictions. The tables indicate that the PM_{2.5} advisory reporting standards adopted in the Air NEPM (Table 2.9) are significantly lower (more stringent) than those adopted in the United States.

Table 2.7: New ambient air quality standards in Europe for PM₁₀ (EC, 1997)

Target Date	Averaging time	Limit ($\mu\text{g m}^{-3}$)	Comment
Stage 1			
2005	24-hour	50	Not to be exceeded more than 25 times per year
2005	Annual	30	
Stage 2			
2010	24-hour	50	Not to be exceeded more than 25 times per year
2010	Annual	20	

Table 2.8: National ambient air quality standards in the USA (and California)

Status	Size	Averaging time	Standard $\mu\text{g m}^{-3}$	Comment
Retained	PM ₁₀	Annual	50	Averaged over 3 years
Current	PM ₁₀	24-hour	150	No more than one exceedence per year averaged over 3 years
Revised	PM ₁₀	24-hour	150	99 th percentile of 24-hour values in a year averaged over 3 years
New	PM _{2.5}	Annual	15	3-year average and spatial averaging
New	PM ₁₀	24-hour	65	98 th percentile of 24-hour values in a year at highest monitor averaged over 3 years
	PM ₁₀	Annual	30	California geometric mean
	PM ₁₀	24-hour	50	California

The health based PM standards are currently being reconsidered in the US, based on a review of recent health information. It is proposed that a new standard will be introduced for “coarse” particles ($>2.5 \mu\text{m}$ and less than $10 \mu\text{m}$) in response to criticisms that PM_{2.5} is a subset of PM₁₀. In time, the current PM₁₀ standard would be phased out. It is also understood that there are currently no moves to consider the introduction of a PM₁ standard in the US (Mike Myers personnel communication). The results from the US review are likely to have a significant bearing on the review of the Air NEPM, which is scheduled to commence in 2005.

In Australia the setting of standards is managed by the National Environment Protection Council (NEPC) under the National Environment Protection Measure (NEPM) process. The Ambient Air Quality NEPM (NEPC, 1998) was made in 1998. It includes a standard for Particles as PM₁₀ of $50 \mu\text{g m}^{-3}$ averaged over one day, and with a maximum of 5 exceedences per year.

Table 2.9: Australian guidelines for PM as promulgated in the air quality NEPM and variation (NEPC 1998, 2003)

Status	Size	Averaging time	Standard $\mu\text{g m}^{-3}$	Comment
Compliance standard	PM ₁₀	24-hour	50	No more than 5 exceedences per year
Advisory standard	PM _{2.5}	24-hour	25	Goal is to gather sufficient data nationally to facilitate a review of the Advisory Reporting Standards as part of the review of this Measure scheduled to commence in 2005
Advisory standard	PM _{2.5}	Annual	8	

Table 2.10: Other PM guidelines and standards

Country	Size	Averaging Time	Limit / standard $\mu\text{g m}^{-3}$	Comment
New Zealand	PM ₁₀	Annual	40	
	PM _{2.5}	24-hour	25	Not a standard – “monitoring value” for assessing data.
	PM ₁₀	24-hour	120	
United Kingdom	PM ₁₀	24-hour	50	Guideline – running average
Japan	PM ₁₀	24-hour	100	Standard
Hong Kong	PM ₁₀	Annual	55	
Hong Kong	PM ₁₀	24-hour	180	
Canada (except Quebec)	PM _{2.5}	24-hour	30	Based on 98 th percentile ambient measurement annually.

In response to the health evidence discussed above, NEPC made a Variation to the Ambient Air Quality NEPM in May 2003 (NEPC, 2003), introducing advisory reporting standards for PM_{2.5}. The variation will be used to inform a review of the NEPM which will commence in 2005. Table 2.9 summarises the Australian legislative position.

The procedure followed for the development of the Air NEPMs in Australia was based on a risk assessment approach, and included components of:

- Issues identification
- Hazard identification, and health endpoints
- Dose-response relationships
- Exposure assessments
- Risk characterisation, and associated health costs
- Setting of the proposed standard

Figure 2.5 and Figure 2.6 obtained from the the Impact Statement for the PM_{2.5} Variation (NEPC 2002) present an estimation of the short and long term health endpoints avoided for a number of PM_{2.5} concentration levels considered in the standard setting process.

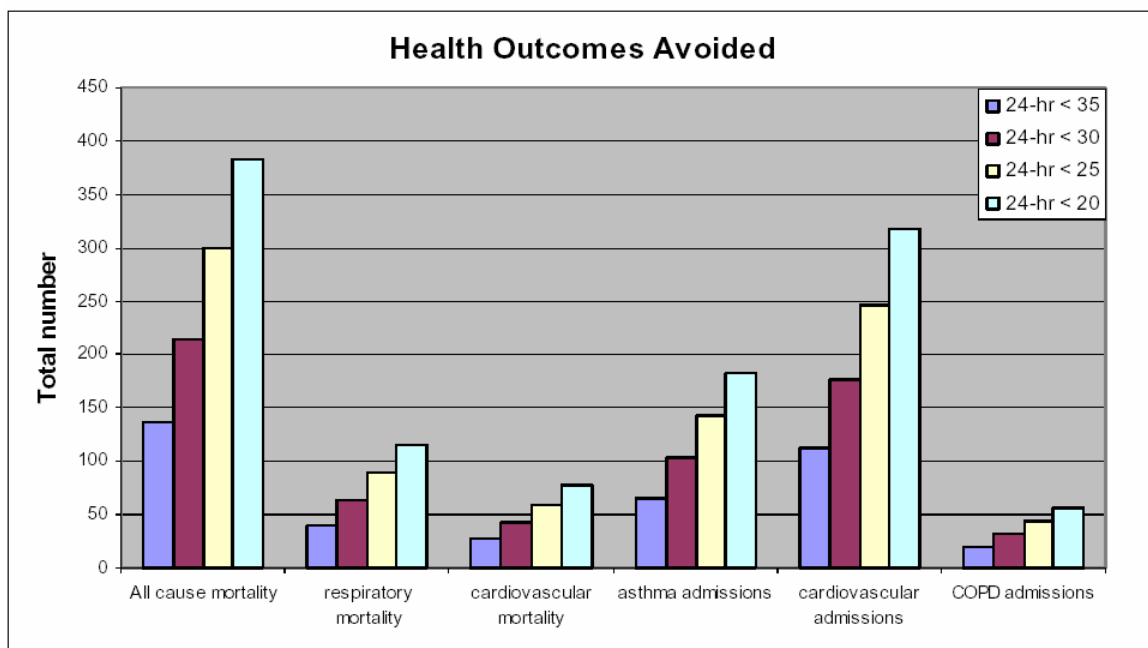


Figure 2.5: Short-term health outcomes avoided for different PM_{2.5} standards (NEPC, 2002).

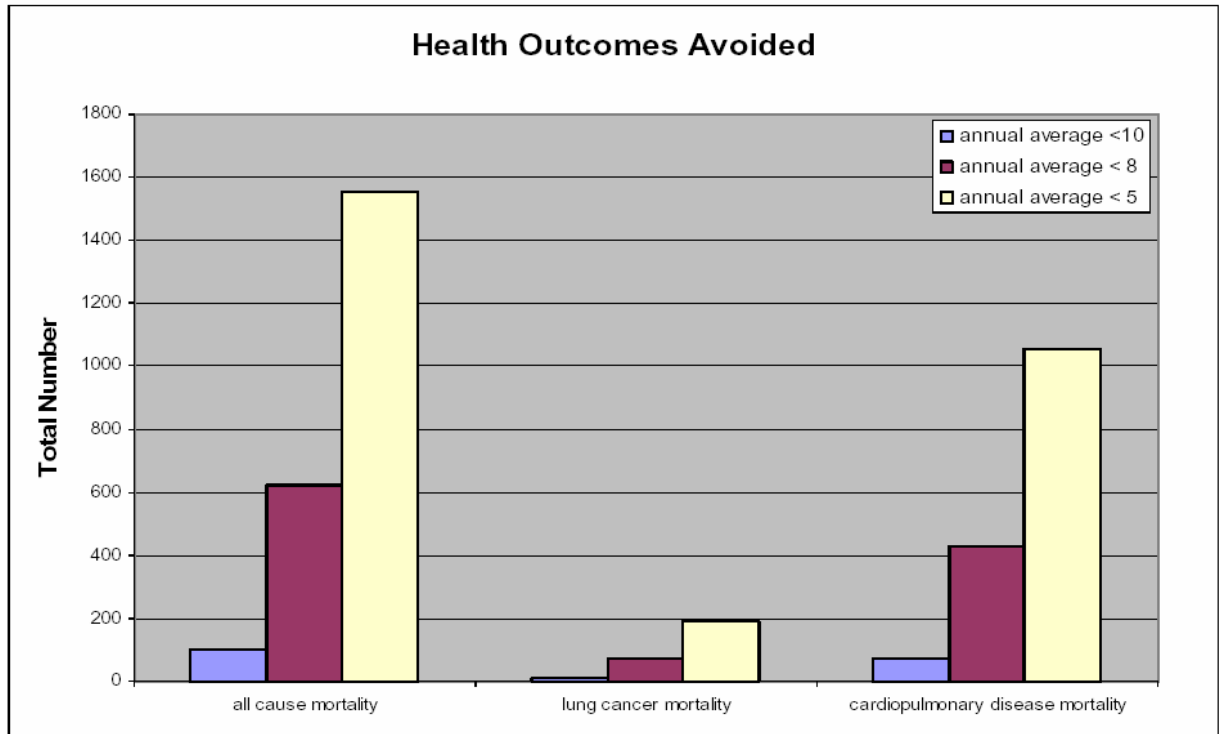


Figure 2.6: Short-term health outcomes avoided for different PM_{2.5} standards considered (NEPC, 2002).

2.6 PM Measurements and Studies

Many studies have been undertaken, or are in progress, in many countries, which are directed at better understanding the occurrence and sources of particulate matter in the atmosphere. Due to the variability in the characteristics and occurrence of PM, results are not necessarily applicable beyond the area in which they were obtained. It is fortunate that there have been a number of relevant studies undertaken in Australia and NSW in the recent past, (Gras et al, 1992; Cohen, 1999; Gras, 1996; Ayers et al, 1999 ANSTO 1995). Summaries of some of these studies are presented in this section. Additionally, Commonwealth and NSW State of the Environment Reports provide summary information on the current state-of-knowledge on the occurrence of PM in urban and regional airsheds. These studies have provided important information on the overall concentration and composition of fine particulate material in urban, and, to some extent, non-urban locations. In addition, detailed measurements of the size characteristics of particles (Morawska et al, 2001; Thomas and Morawska, 2002) have been performed in a number of airsheds.

2.6.1 State of the Environment Reporting

Commonwealth and NSW State of the Environment Reports (Aust. State of Environment Committee 2001, NSW DEC 2003) provide summary information on the current knowledge on the occurrence of PM in urban and regional airsheds.

Figure 2.7 shows the number of days in NSW airsheds on which the NEPM PM_{10} standard was exceeded between 1994 and 2001. In commenting on these data the NSW Department of Environment and Conservation note that bushfires are responsible for most of the occasions when high levels of particle pollution are recorded. This is particularly evident in the data in Figure 2.7 for 1994 and 2001 when severe bushfires burned throughout the greater Sydney area. For the non-bushfire period the maximum value for 2001 was approximately 83% of the standard.

In the absence of bushfires, hazard reduction burning, domestic wood heating and diesel vehicles become the major sources of particles in urban areas. In regional centres exceedences of the standard can also occur as a result of bushfires as well as burning for hazard reduction or agricultural purposes.

Occasionally, widespread dust storms can also result in extreme particle levels. During one of these episodes in November 2002, the NEPM standard was exceeded in all regions where monitoring was carried out. Levels ranged from about 120% of the standard at Tamworth to about three times the standard at Wagga Wagga and five times the standard at Bathurst. Levels of about 160% of the standard were recorded in Sydney, the Illawarra, lower Hunter and Albury.

More detailed analysis of the PM_{10} and $PM_{2.5}$ data collected by the DEC's monitoring network is provided in Section 2.6.5

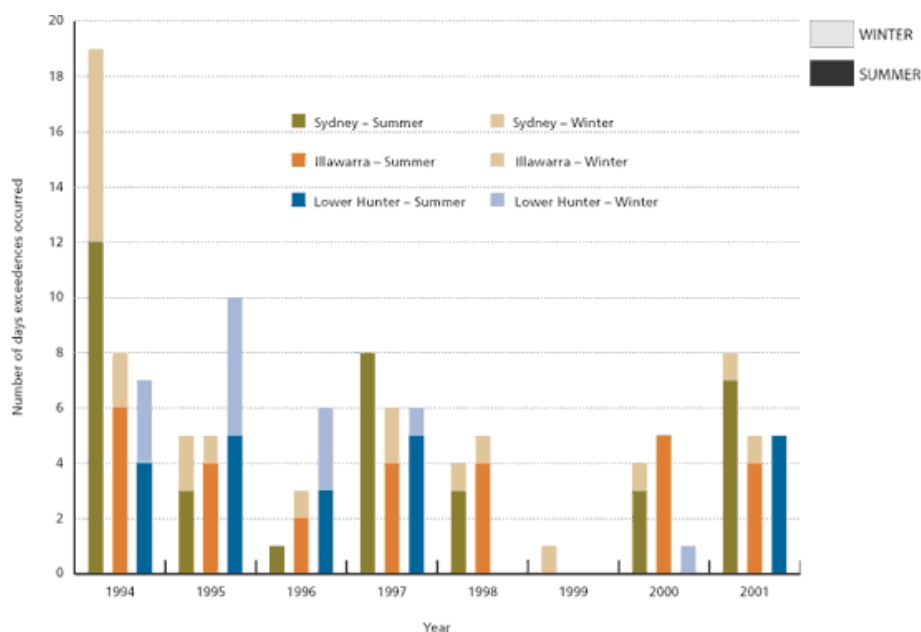


Figure 2.7: Number of days exceeding the NEPM PM_{10} standard in NSW airsheds. (DEC 2003).

2.6.2 ERDC $PM_{2.5}$ Study 1995

Between January 1992 and June 1993 $PM_{2.5}$ measurements were undertaken at 25 sites (24 in NSW and at Cape Grim, north-west Tasmania). The project was funded by the Energy Research and Development Corporation (ERDC) and was a collaborative investigation lead by the Australian Nuclear Science and Technology Organisation (ANSTO 1995).

The project's aims included the investigation of the relationships between fuel combustion and fine particle aerosols in urban and non urban environments.

Data collection on Teflon filters, twice per week over the period, resulted in over 5,000 filters being exposed over the monitoring period.

Ion beam analyses techniques were used to analyse exposed filters for 25 elements. The elemental results were used to estimate the relative contributions of the major constituents of the particulate matter to the total mass measured.

During 1992, 11 of the 25 ERDC monitoring sites returned total mass averages greater than what is now the NEPM advisory reporting standard of $8 \mu\text{g m}^{-3}$.

The estimated average composition of fine particles collected during the ERDC project was:

- 23% organic matter
- 23 % sulfate – assuming all particulate sulfur was in the form of ammonium sulfate.
- 22% soot (elemental carbon)
- 7% sea salt
- 6% soil
- 5% nitrate – assuming ammonium nitrate.
- 1% lead
- Usually 8 -10% water,
- trace elements and organic ions.

The annual average $\text{PM}_{2.5}$ concentrations at three sites in the vicinity of the coal-fired power stations and their ranking compared with the other ERDC sites are shown in Table 2.11. The table shows that data for individual sites can differ significantly from the “average” composition presented above, with Cullen Bullen showing a very high contribution from organic matter and Muswellbrook showing an elevated “ammonium sulfate” contribution compared with the average study data.

Table 2.11: Average $\text{PM}_{2.5}$ concentrations at ERDC sites in coal-fired power station regions and the estimated contribution from the major components.

<i>ERDC site</i>	<i>Average $\text{PM}_{2.5}$ concentration $\mu\text{g m}^{-3}$</i>	<i>Rank out of 25 sites</i>	<i>Percent Contribution</i>				
			<i>Organic matter</i>	<i>Ammon. sulfate</i>	<i>Soot</i>	<i>Sea Salt</i>	<i>Soil</i>
Muswellbrook Hunter Valley	6.5	15th	19	36	20	4	6
Doyalson Central Coast	6.1	19th	23	22	22	8	7
Cullen Bullen Western	9.7	7th	37	16	28	1	3

The “ammonium sulfate” contribution is based on the assumption that there is sufficient ammonia in the atmosphere for all the particulate sulfur captured on the filters to be in the form of ammonium sulfate. Ayers et al (see below) reported that sulfate ions contributed, on average 4.5% by weight to the $PM_{2.5}$ concentrations in Sydney, during a limited sampling period. The Ayers average sulfate contribution is significantly lower than reported in the ERDC study, even after allowing for the fact that the ERDC results assumed 100% neutralisation of sulfate by ammonia and Ayers et al reported ionic sulfate.

Additionally, the ratio of ammonium sulfate to ammonium nitrate reported in the ERDC study of between 5 and 10:1 is significantly greater than the sulfate to nitrate ratio reported by Ayers et al of about 2.5 (see Table 2.12). The ERDC report noted significant uncertainty in the nitrate results reported.

Figure 2.8 shows that ERDC sites with higher average mass concentrations generally also had higher concentrations of “ammonium sulfate”. Figure 2.9 however, shows that “ammonium sulfate” tended to contribute a smaller proportion (in the order of 20%) to the total $PM_{2.5}$ mass at sites recording higher total average $PM_{2.5}$ mass concentrations. These sites tended to be the more “urban” sites in the network. As the average mass concentrations fall from about $8 \mu g m^{-3}$ down to about $4 \mu g m^{-3}$, the percentage contribution of “ammonium sulfate” increases from about 20 to 35%. The sites showing lower average mass and higher “ammonium sulfate” contribution tend to be non-urban or regional sites in the network.

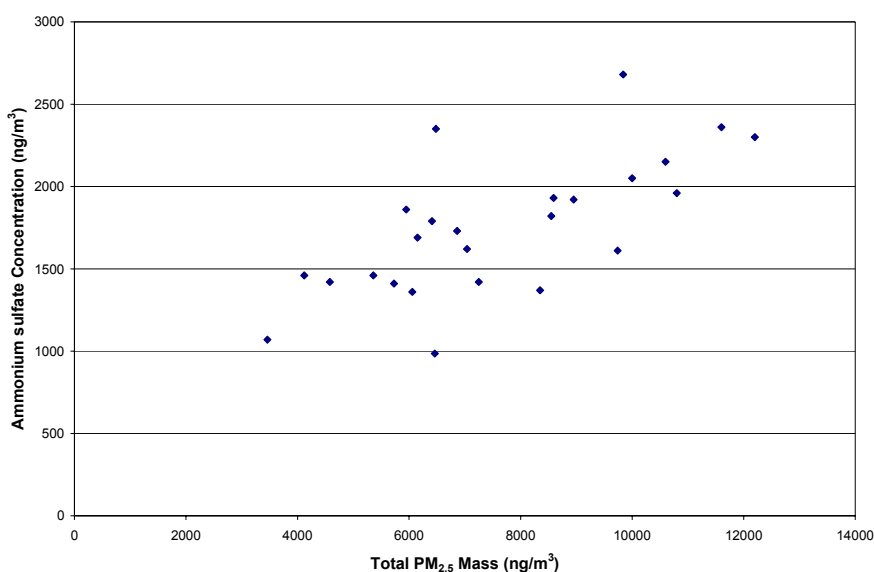


Figure 2.8: The relationship between average total $PM_{2.5}$ mass and average “ammonium sulfate” mass at the 25 ERDC sites in 1992.

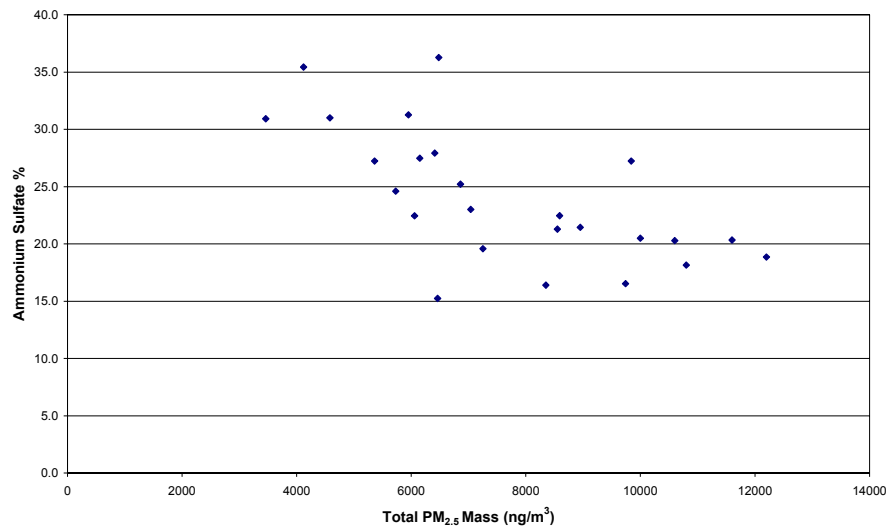


Figure 2.9: The percentage contribution of “ammonium sulfate” to average total PM_{2.5} mass at the 25 ERDC sites in 1992

Without access to the electronic data-sets it is not easy to undertake any further detailed analyses of the relationships between parameters in the ERDC data set or to investigate possible source contributions. However, with respect to the sulfur and “ammonium sulfate” data, a visual examination of the daily plots in the ERDC report suggest that elemental sulfur and “ammonium sulfate” as often as not, appeared to correlate poorly with total mass, a level of detail not revealed in the plots of average data shown in Figure 2.8. Figure 2.10 and Figure 2.11 show examples of the inconsistent relationship between 24 hour average mass and “ammonium sulfate” concentrations over six month periods at Muswellbrook and Campbelltown respectively.

While the ERDC data are useful in providing spatial information on the occurrence of PM_{2.5} over a significant part of NSW and an indication of the gross composition of the collected particle mass, it is concluded here that careful consideration of source characteristics and atmospheric processes (physical and chemical) are required in order to understand the contribution of particular sources to the occurrence of fine particles in NSW.

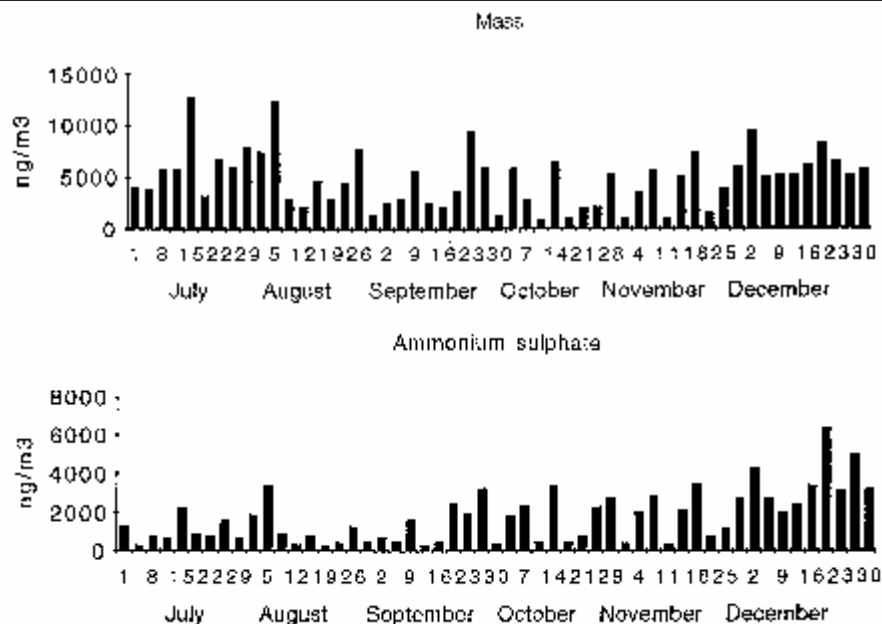


Figure 2.10: 24 hour average mass concentrations (top) and derived ammonium sulfate concentrations (bottom) measured at Muswellbrook between July and December 1992.

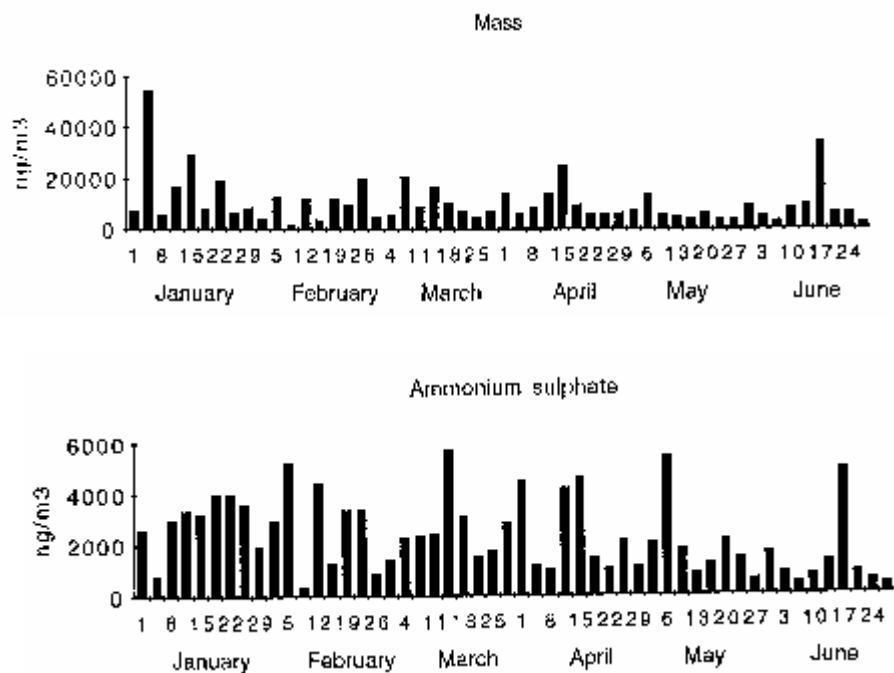


Figure 2.11: 24 hour average mass concentrations (top) and derived ammonium sulfate concentrations (bottom) measured at Campbelltown between January and June 1992.

2.6.3 Pilot Study: Chemical and Physical Properties of Australian Fine Particles

The Pilot Study, *Chemical and Physical Properties of Australian Fine Particles*, performed by CSIRO and ANSTO (Ayers *et al*, 1999), measured size and composition of particles in 6 Australian cities. The emphasis in this study was on comparison of instrumental techniques for fine particle characterisation. The study was of limited duration, but employed a comprehensive package of aerosol measurement equipment to generate a large database on a wide variety of chemical and physical properties. While the authors caution against drawing too many conclusions from this data set, the sulfate and nitrate content of the fine particle fractions is relevant to the current study. Table 2.12 summarises the results which vary noticeably between cities.

Table 2.12: Mean proportions of non-sea salt sulfate and nitrate in PM₁₀ and PM_{2.5} size fractions collected in six Australian cities (data from Ayers *et al*, 1999)

City	Size Fraction	Non-sea salt Sulfate Average proportion (wt%) of size fraction	Nitrate
Sydney	PM ₁₀	3.45	3.00
	PM _{2.5}	4.48	1.93
Melbourne	PM ₁₀	2.95	3.79
	PM _{2.5}	4.31	2.30
Brisbane	PM ₁₀	3.83	12.55
	PM _{2.5}	6.62	7.97
Canberra	PM ₁₀	3.01	2.99
	PM _{2.5}	3.65	1.45
Adelaide	PM ₁₀	0.65	2.87
	PM _{2.5}	1.0	2.18
Launceston	PM ₁₀	1.45	7.67
	PM _{2.5}	1.68	4.75

The data show that on average non-sea salt sulfate and nitrate in Sydney made up less than 7% by mass of the PM₁₀ and PM_{2.5} compared with about 25% reported in the ERDC study (section 2.6.2) which reported sulfate and nitrate as ammonium sulfate and ammonium nitrate rather than as the respective ions. Reporting the Pilot Study results in the same form as the ERDC results would suggest a contribution of about 10% from “ammonium sulfate” and “ammonium nitrate”- still significantly less than the ERDC estimated contribution.

Results cited in Seinfeld and Pandis (1998) included in Table 2.13 show aerosol concentration and composition data collected in different regions. The table shows that the average composition of non-urban and urban aerosols are roughly the same. The elemental carbon component, a direct indicator of anthropogenic combustion sources, is very low in remote area aerosols, but the sulfate component in remote aerosols is more comparable to the other regions. Note that Rubinox, California is located about 100 km east of Los Angeles.

The regional data shown in Table 2.13 appear to be more consistent with the ERDC sulfate and nitrate values rather than those reported by Ayers *et al* in the Pilot Study. Recall from the ERDC section that the urban sites and non-urban sites showed ammonium sulfate contributions of about

20% and up to 35% respectively. The Rubidoux results, which show lower sulfate and higher nitrate contributions would appear to be strongly influenced by the Los Angeles motor vehicle emissions, as might Sydney results, which suggests that further work is required to clarify the composition of the aerosol mass in Sydney and environs.

Expressing the percentage results in Table 2.13 as mass concentrations shows that while sulfate makes a relatively smaller percentage contribution to total PM mass at Rubidoux, on a mass concentration basis Rubidoux sulfate results are similar to those reported for non-urban sites and not dissimilar to urban results. The remote sulfate concentration of $\sim 1 \mu\text{g m}^{-3}$ is similar to the levels measured at the “cleanest” sites in the ERDC network.

Table 2.13: Mass concentrations and composition of tropospheric aerosols (Seinfeld and Pandis 1998)

Region	Mass $\mu\text{g/m}^3$	Percentage Composition (<i>derived mass concentrations in brackets</i> $\mu\text{g m}^{-3}$)				
		Carbon (elemental)	Carbon (organic)	NH_4^+	NO_3^-	SO_4^{2-}
Remote (11 areas)	4.8	0.3	11	7	3 (0.14)	22 (1.06)
Non-urban (14 areas)	15.0	5	24	11	4 (0.6)	37 (5.5)
Urban (19 areas)	32	9	31	8	6 (1.92)	28 (8.96)
Rubidoux, California	87.4	3	18	6	20 (17.4)	6 (5.24)

2.6.4 CRC for Coal in Sustainable Development (CCSD) Project

The contribution of power station emissions to fine particle concentrations in the Upper Hunter Valley was the subject of a PhD project (Hinkley, 2004; Hinkley *et al*, 2003), supported by the CRC for Coal in Sustainable Development.

Sampling was conducted at an existing monitoring site at Ravensworth, approximately 11 km to the SE of Bayswater and Liddell power stations. Primary particulate emissions were targeted as minimal gas to particle conversion was expected close to source. Samples were collected between June 2002 and March 2004 using a Burkard spore sampler to estimate the contribution of power station fly ash to total particulate mass. A cascade impactor was used to assess the contribution to aerosol chemistry and a Nanometer Aerosol Sampler was used to characterise the ultra fine particles ($<0.1 \mu\text{m}$) in the air.

The contribution of power station emissions to particle levels was targeted by conditional sampling which relied on sulfur dioxide measurements at the site being used to identify the presence of the power station plume at the site.

Results from the Burkard spore sampler indicated a maximum estimated contribution from power station fly ash emissions in the size range of 1 and 10 μm of $0.4 \mu\text{g m}^{-3}$, averaged over a period of less than one hour. The aerosol at the site was dominated by other sources such as windblown dust.

Characteristics of the ultra-fine PM were examined with a cascade impactor (giving size-segregated samples), chemical analysis and microscopic examination. These results showed that under elevated SO_2 conditions (i.e. when the power station plumes were being sampled) there was evidence for an increased contribution of sulfate species to the total submicron particles mass. The amount of sulfate observed was broadly consistent with emission estimates for sulfuric acid (H_2SO_4) from the power stations as reported in the National Pollutant Inventory (NPI). However secondary formation of sulfate from SO_2 emissions could not be completely discounted as a contributor to the observed sulfate results.

2.6.5 NSW Department of Environment and Conservation monitoring program

The NSW Department of Environment and Conservation (DEC) operates an extensive network of particle monitors in NSW, which until recently focused on PM_{10} in the Sydney – Newcastle - Illawarra urban areas. The introduction of the Air NEPM in 1998 saw this network extend beyond the area to regional centres, such as Armidale, Wagga Wagga and Bathurst.

The variation to the Air NEPM in 2003 imposed a modest requirement on jurisdictions to undertake $\text{PM}_{2.5}$ monitoring in addition to the existing PM_{10} monitoring requirements. The NSW DEC's $\text{PM}_{2.5}$ monitoring program however exceeds the requirements of the 2003 NEPM Variation.

PM_{10} is monitored at 17 sites and $\text{PM}_{2.5}$ at 9 sites. Both $\text{PM}_{2.5}$ and PM_{10} are monitored at 7 sites. Monitoring is undertaken continuously using TEOMS (Tapered Element Oscillating Microbalance).

Summaries of the PM_{10} and $\text{PM}_{2.5}$ data from the DEC monitoring network for the years 2001, 2002, 2003 are provided in Table 2.14 and Table 2.15. The data show that there is the potential for the PM_{10} standard to be exceeded at a number of sites in each year. Years with sites recording higher numbers of days above the standard, such as 2002, are likely to have been influenced by bushfires and / or dust events, as previously discussed.

Table 2.15 indicates that the $\text{PM}_{2.5}$ annual average advisory reporting standard was exceeded at all sites in all three years and that the $\text{PM}_{2.5}$ 24 hour average advisory reporting standard was frequently exceeded between 2001 – 2003. As is the case with PM_{10} , bushfire smoke can significantly impact $\text{PM}_{2.5}$ results, as is demonstrated by the 2002 results.

Table 2.14: NSW DEC PM₁₀ monitoring data summary

<i>Year</i>	<i>Number of days per site above the NEPM 24 hour average standard</i>	<i>Number of sites with more than 5 days above the NEPM standard</i>	<i>Range of Annual Averages</i> $\mu\text{g m}^{-3}$	<i>Range of maximum values</i> $\mu\text{g m}^{-3}$
2001	2 - 9	0	13.3 – 21.3	28.8 – 142.3
2002	4 - 37	14	18.7 – 28.9	72.6 – 258.2
2003	0 – 12	8	16.2 – 29.5	36.8 – 921.4

Table 2.15: NSW DEC PM_{2.5} monitoring data summary

<i>Year</i>	<i>% of sites exceeding the NEPM annual average advisory reporting standard</i>	<i>Number of days per site above the NEPM 24 hour average advisory reporting standard</i>	<i>Range of Annual Averages</i> $\mu\text{g m}^{-3}$	<i>Range of maximum values</i> $\mu\text{g m}^{-3}$
2001	100	2 - 9	9.1 – 12.3	41.6 – 118.6
2002	100	4 – 37	11.1 – 15.0	50.4 – 98.2
2003	100	0 - 12	9.3 – 13.6	14.1 – 112.5

Figure 2.12 shows the PM₁₀ concentration frequency distributions recorded at the DEC sites in 2001. The figure demonstrates the skewed nature of the distributions, which is also evident in 2002 and 2003 data. 90th percentile concentrations in 2001 were in the order of 20 – 30 $\mu\text{g m}^{-3}$ whereas 90th percentile concentrations in 2002, a year known to have experienced significant bushfire impacts, were 30 – 50 $\mu\text{g m}^{-3}$.

Figure 2.13 shows PM_{2.5} concentration frequency distributions recorded at the DEC sites in 2001. These distributions appear to be more highly skewed than the corresponding PM₁₀ distributions and also appear to show less variation between sites, particularly at concentrations less than the 90th percentile values.

90th percentile values of the PM_{2.5} concentrations show less annual variation than the PM₁₀ data. Generally 90th percentile PM_{2.5} concentrations were between 15 – 20 $\mu\text{g m}^{-3}$ in each of the three years.

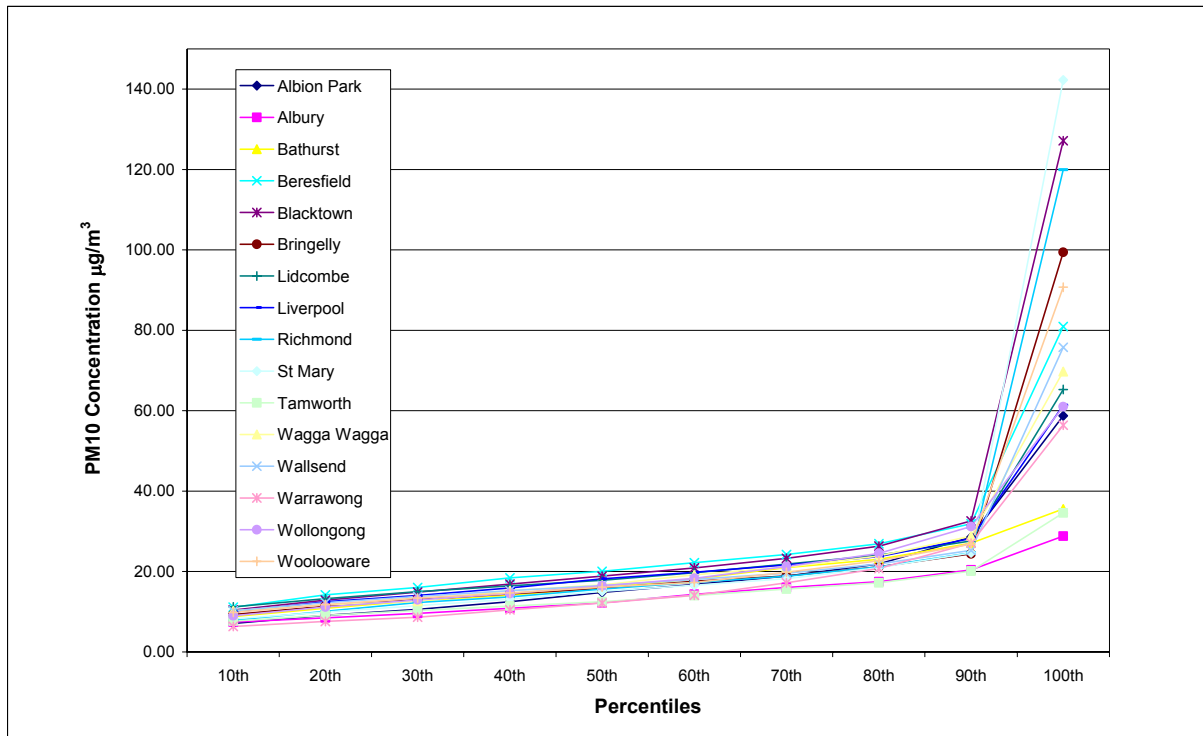


Figure 2.12: Frequency distributions of PM₁₀ concentrations at the DEC monitoring sites in 2001.

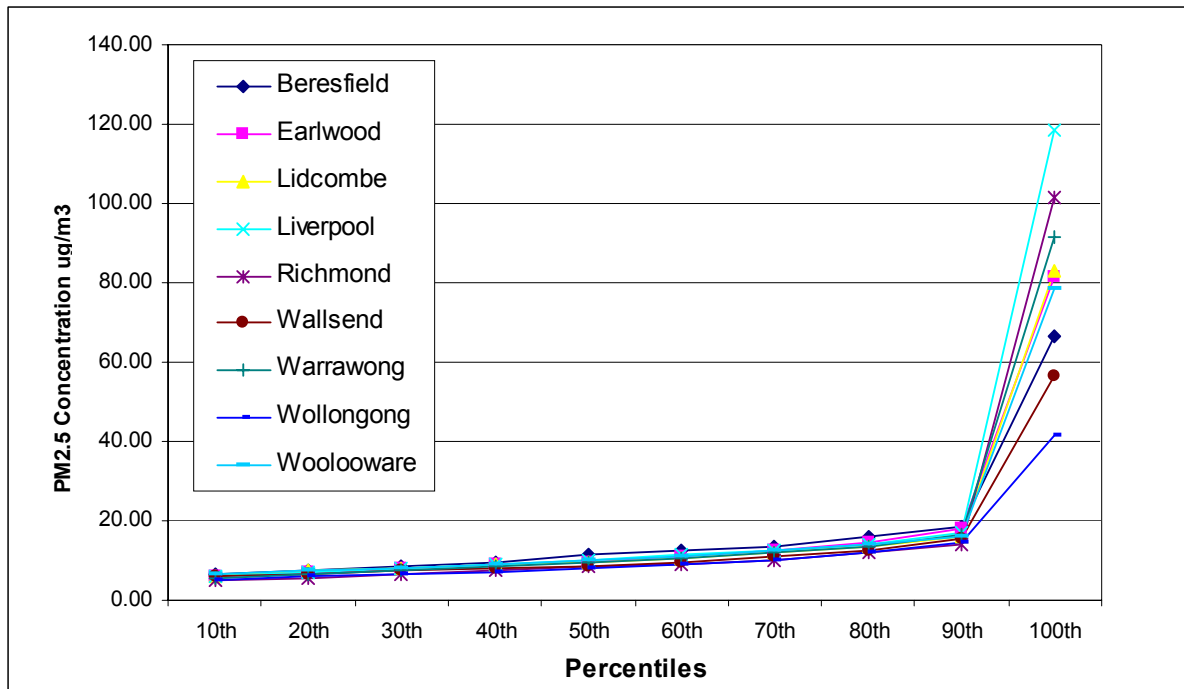


Figure 2.13: Frequency distributions of PM_{2.5} concentrations at the DEC monitoring sites in 2001.

The ratio of $PM_{2.5}$ to PM_{10} has also been determined for the DEC monitoring data. Figure 2.14 shows that the average site ratios are relatively constant from site to site and year to year, varying between about 0.5 and 0.6. Results for individual events days, can of course, vary significantly. On occasions, PM_{10} can be entirely $PM_{2.5}$ (ratio of 1). However, the $PM_{2.5}$ fraction of PM_{10} seldom falls below about 0.2

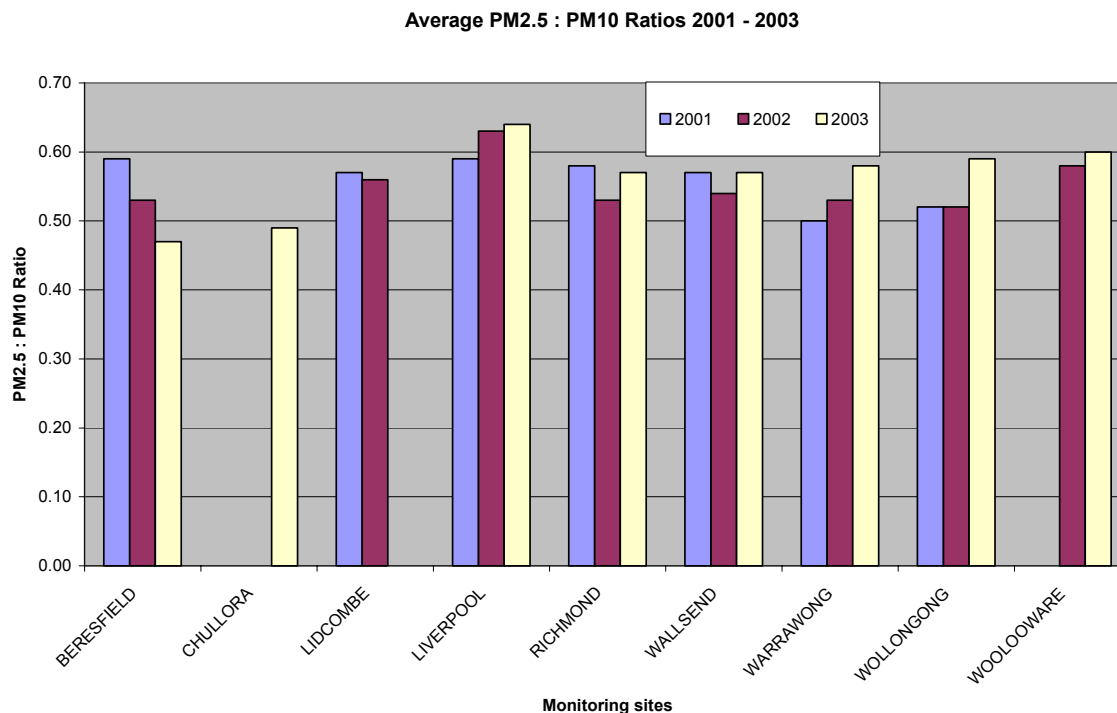


Figure 2.14: The ratio of $PM_{2.5}$ to PM_{10} at the DEC monitoring sites in 2001, 2002 and 2003.

2.6.6 Wyee monitoring

To date there has been relatively little routine monitoring of fine particles at sites in the vicinity of the power station emissions. However, Delta Electricity has undertaken routine monitoring at Wyee for several years. The monitoring site is:

- 6.5 km from Vales Point Power Station (VPPS) and downwind of the power station under ENE wind directions.
- 13 km from Eraring Power Station (EPS) and downwind of the power station under NNE wind directions.

While EPS is twice the size of VPPS, its fabric filters result in PM emissions which are about one-tenth of the VPPS emissions, which is equipped with electrostatic precipitators.

The monitoring program at Wyee has consisted of about one year of PM_{10} monitoring *followed by* over 1.5 years of $PM_{2.5}$ monitoring, together with measurements of wind speed and direction, SO_2 and NO_x .

Figure 2.15 shows the relationship between all PM results ($PM_{2.5}$ and PM_{10}) and wind direction measured at Wyee, based on hourly average data. The vertical black line in the figure is drawn at a wind direction of about 78 degrees, indicating the direction for which the Wyee monitor is directly downwind of VPPS. The dashed line is drawn at a wind direction of about 20 degrees, the direction for which the Wyee monitor is directly downwind of EPS. At this gross level of analysis, the figure would not appear to indicate a relationship between VPPS emissions and elevated PM measurements at Wyee.

To further examine the contribution of VPPS emissions to regional PM concentrations use has been made of the Wyee SO_2 measurements. VPPS along with Eraring (and Munmorah) Power Stations are the dominant emitters of SO_2 in the region. It can be assumed that any elevated SO_2 recorded at Wyee will have been emitted from one of the power stations. As previously discussed it is far more difficult to determine the source of measured PM due to the many and varied sources. So, here SO_2 is used to signal the presence of a power station plume at the monitoring site. The PM data coincidental with the elevated SO_2 concentrations may enable the power station contribution to PM to be estimated.

Figure 2.16 shows SO_2 plotted against wind direction at Wyee, again with lines indicating directions downwind of VPPS and EPS. The figure clearly shows elevated SO_2 concentrations at Wyee are associated with wind blowing from the direction of VPPS. The figure would also appear to indicate elevated SO_2 concentrations associated with wind blowing from the direction of EPS.

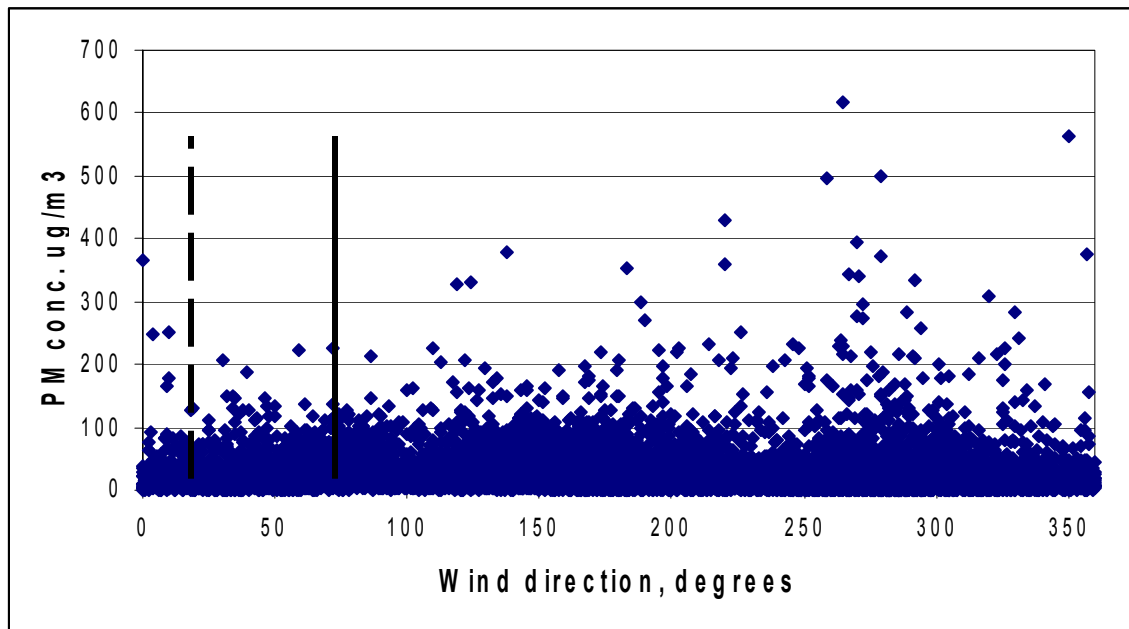


Figure 2.15: The relationship between wind direction and PM concentration at Wyee. The solid and dashed black lines indicate wind directions for which Wyee is directly downwind of VPPS and EPS respectively.

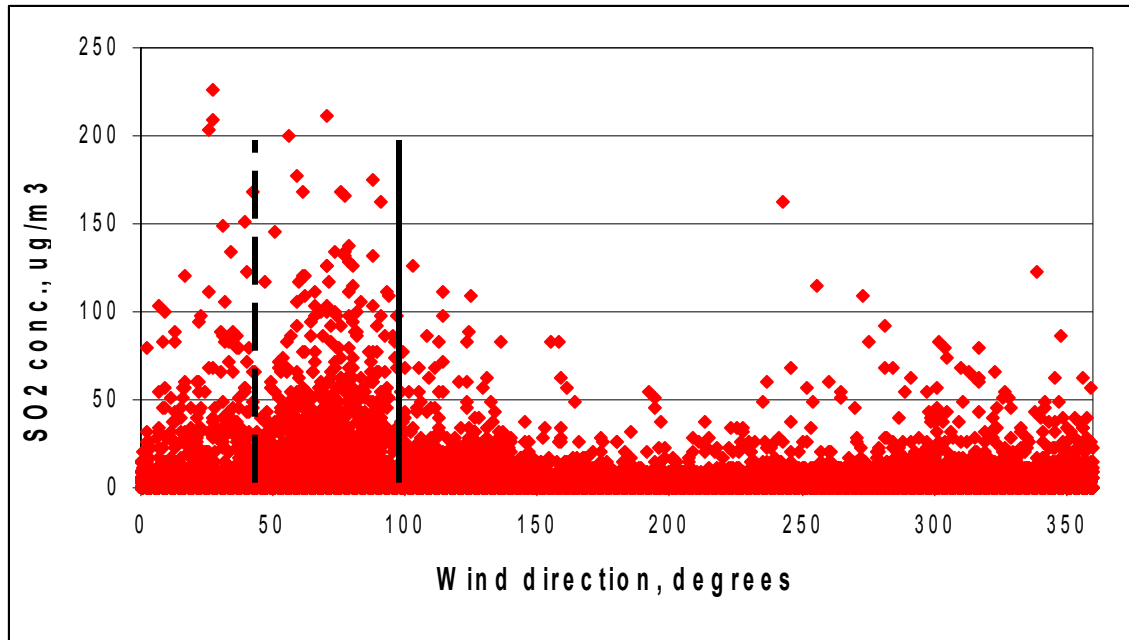


Figure 2.16: The relationship between wind direction and SO₂ concentrations at Wyee. The solid and dashed black lines indicate wind directions for which Wyee is directly downwind of VPPS and EPS respectively.

Figures 2.15 and 2.16 also suggest a source of PM and SO₂ from directions of about 250 – 350 degrees. As the Wyee site is within one kilometre of the F3 Freeway to the west it is likely that motor vehicles are a significant source of pollutants measured at Wyee.

Having established that SO₂ might be used as a signal for power station emissions, the PM data set were sorted by wind direction and SO₂ concentration. In the following analyses PM₁₀ and PM_{2.5} are examined separately and conditions under which Wyee is “downwind” of VPPS includes wind directions from 48 to 108 degrees, which includes winds 30 degrees either side of directly downwind (78 degrees) to account for plume spreading. This downwind data set has been further filtered to exclude all PM results when SO₂ concentrations were less than 30 µg m⁻³ in an attempt to exclude non-power station events from the final filtered data sets.

The results of the PM₁₀ and PM_{2.5} analyses are shown in Table 2.16, Table 2.17 and Figure 2.17 and Figure 2.18. The tabulated data indicate that when the VPPS plume is being measured at Wyee average PM concentrations are likely to be higher at Wyee than when the plume is not being measured at the site. The figures and tables however indicate a very weak, or non-existent, correlation between SO₂ and PM₁₀ and PM_{2.5} concentrations for the 103 and 155 “downwind” hours analysed, which suggests that the filtering process has not effectively removed the influence of non-power station PM sources.

Further restricting the PM₁₀ data set to events when SO₂ measured at Wyee was greater than 60 µg m⁻³ and wind directions were between 63 and 93 degrees increases both the average PM₁₀ concentration and the strength of the correlation between SO₂ and PM₁₀ concentrations, as shown

in Table 2.16, but a similar restriction on the PM_{2.5} data lowered the average concentration and did not improve the correlation between PM_{2.5} and SO₂ (Table 2.17).

It should be noted from this analysis that the filtered PM₁₀ and PM_{2.5} data sets which might indicate a VPPS contribution to PM₁₀ and PM_{2.5} at Wyee are restricted to about 1% of the 22,000 hours analysed.

An examination of the frequency distributions of PM₁₀ and PM_{2.5} (Figure 2.19 and Figure 2.20) indicate that higher “downwind” average concentrations indicated in Tables 2.16 and 2.17 are not due to the peak (> 90th percentile) concentrations, but rather due to slightly higher concentrations through the distribution from the 10th to 90th percentiles. Maximum concentrations of both PM₁₀ and PM_{2.5} are more likely to occur when Wyee is not downwind from VPPS.

Table 2.16: Analysis of PM₁₀ concentrations at Wyee under “downwind” of VPPS conditions.

	All Data	Downwind of VPPS(1)	Downwind of VPPS(2)
Number of records	8,334	103	23
Maximum hourly value (µg m ⁻³)	618	119	119
Average (µg m ⁻³)	24.4	35.2	41.0
Correlation coefficient SO ₂ and PM ₁₀		0.25	0.44

1. Wind directions between 48 and 108 and SO₂ greater than 30 µg/m³
2. Wind directions between 63 and 93 and SO₂ greater than 60 µg/m³

Table 2.17: Analysis of PM_{2.5} concentrations at Wyee under “downwind” of VPPS conditions.

	All Data	Downwind of VPPS(1)	Downwind of VPPS(2)
Number of records	14,131	151	50
Maximum hourly value (µg m ⁻³)	563	101	47
Average (µg m ⁻³)	11.7	16.6	11.3
Correlation coefficient SO ₂ and PM ₁₀		-0.1	-0.2

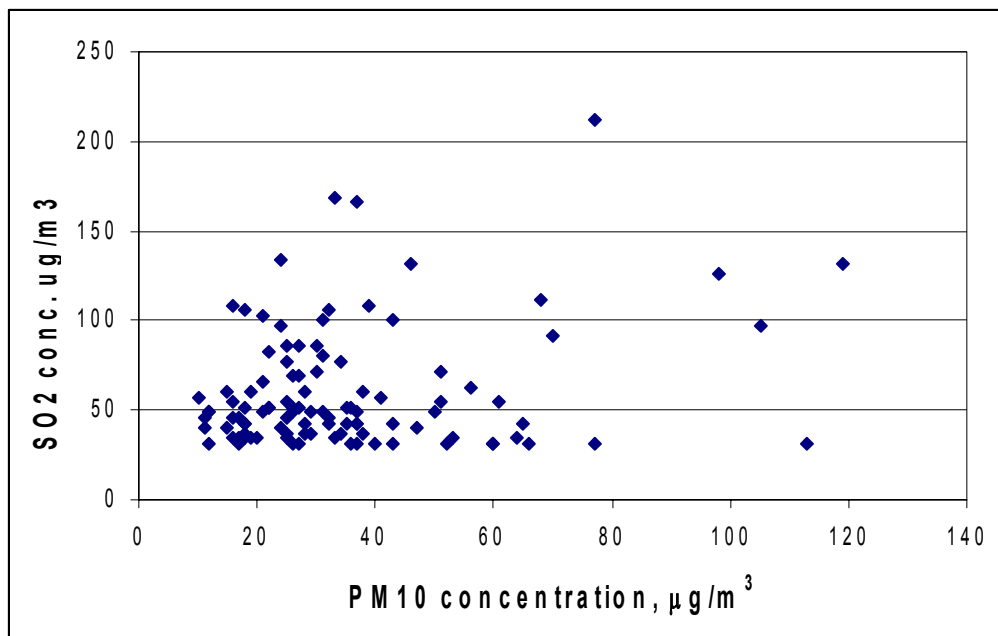


Figure 2.17: The relationship between PM_{10} and SO_2 at Wyee under “downwind” of VPPS conditions.

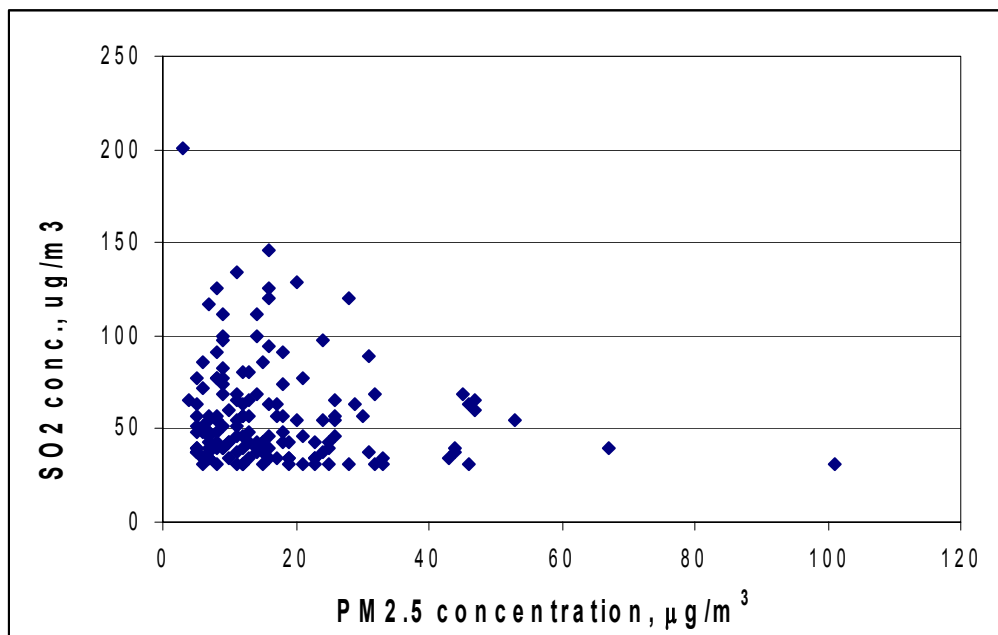


Figure 2.18: The relationship between $PM_{2.5}$ and SO_2 at Wyee under “downwind” of VPPS conditions

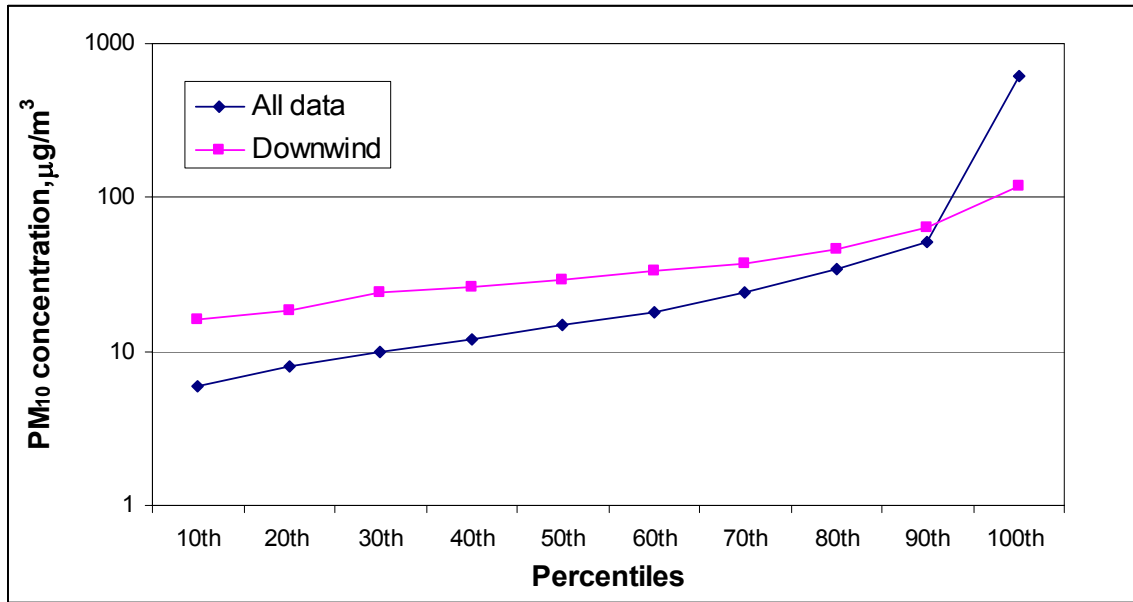


Figure 2.19: PM₁₀ frequency distributions for “All Data” and “Downwind Data”

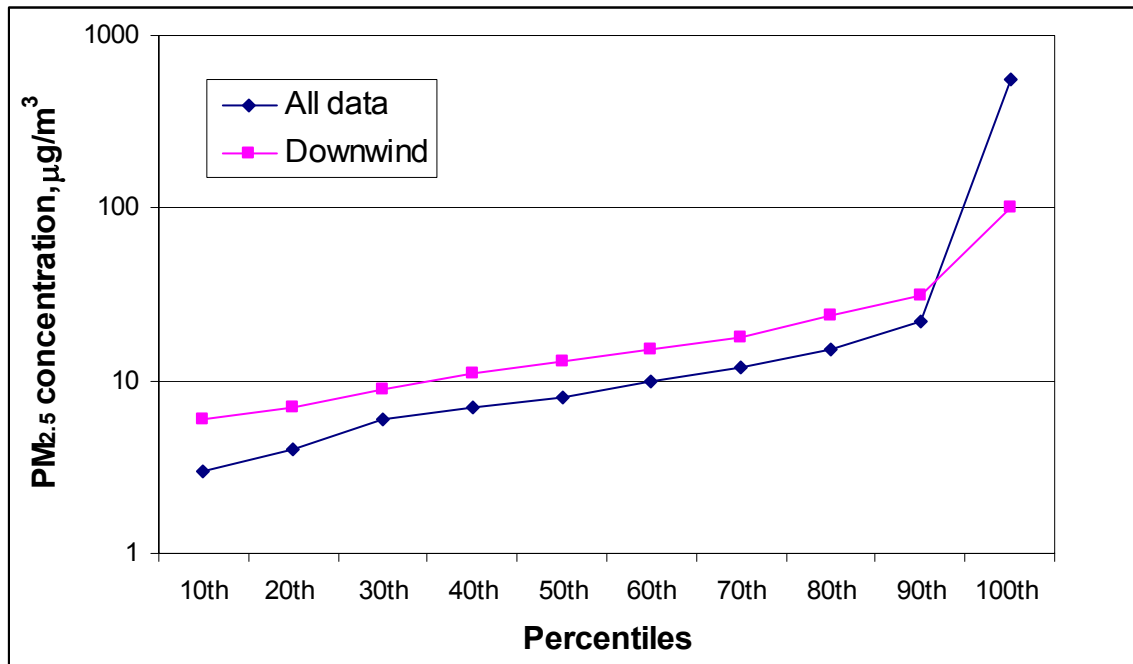


Figure 2.20: PM_{2.5} frequency distributions for “All Data” and “Downwind Data”

2.7 Coal fired Power Station emissions

To assess the potential impacts of power station emissions on particle levels in the atmosphere it is necessary to take into account the emission of primary particles and the formation of secondary particles from precursor emissions.

Primary particle emissions result from the small component of fly ash not trapped by bag filters and electrostatic precipitators, and may vary in size from much less than one μm in diameter to many tens of μm in diameter.

Secondary aerosol production from power station emissions form from the emission of sulfur and nitrogen oxides and can occur via three pathways which are discussed in this section.

This section also describes the method of estimating power station emissions of the relevant substances for the purposes of modelling the contribution of power station emissions to the atmospheric concentration of fine particle matter. In summary for each power station, emission rates for the following substances were estimated:

- Primary particles
 - $\text{PM}_{2.5}$ and PM_{10}
- Sulfur trioxide / sulfuric acid
- Sulfur dioxide
- Nitrogen oxides

2.7.1 Primary emissions – coal mineral matter derived particulate matter

Fine particles associated with power station emissions include direct emissions of non-combustible residues derived from the mineral matter in the coal, and indirect formation of sulfates and nitrates from emitted SO_2 and NO_x . In this section we consider available information on the direct emissions.

Particle size distributions of ash produced during coal combustion have long been known to be multi-modal in character. The majority of the ash is greater than 1 μm in size, and results from pulverized fuel burnout leading to an ash residue, and particle fragmentation processes. A much smaller proportion of fine material is also produced. The nature and composition of this material is of great importance, since:

- Collection efficiency in electrostatic precipitators (ESPs) is lowest for particles in the ultra fine 0.1 - 1 μm size range. This is illustrated in Figure 2.21 which shows data collected by the VTT Technical Research Centre of Finland (Jokiniemi 2003). The figure also illustrates that particle penetration through fabric filters is generally much lower and less size dependent than through ESPs.
- In the context of trace element deportment, trace elements have been observed to be enriched in the fine fraction (Linak et al 2002).

- The health effects observed recently for PM are associated most strongly with the fine fraction. Increasing toxicity with decreasing PM aerodynamic diameter has been reported (Okeson 2003)
- Light scattering is greatest for particulate material in the submicron range; hence visibility impacts are highest for this range.

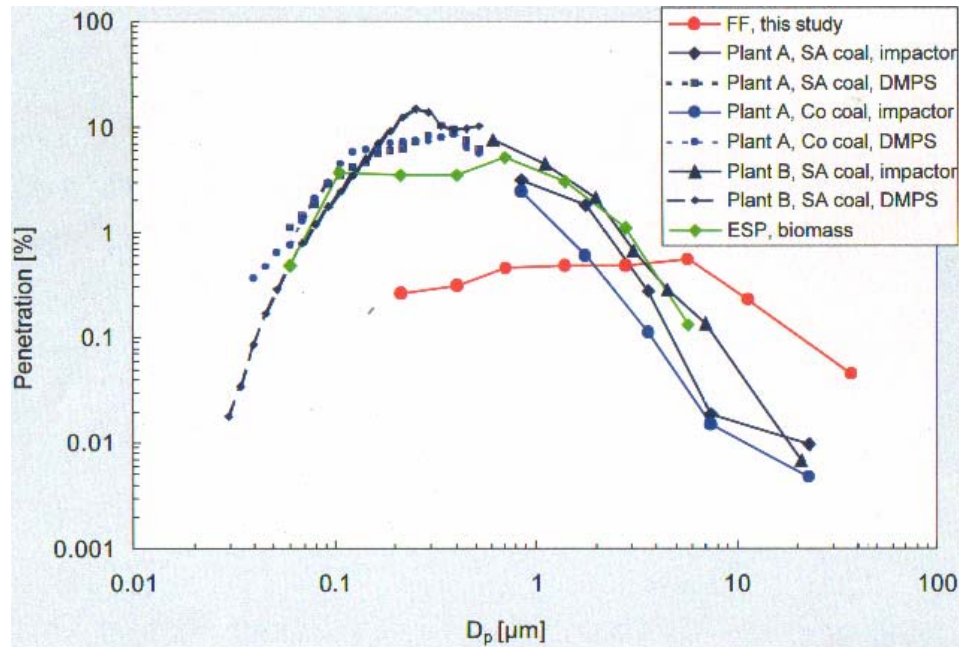


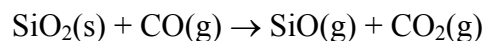
Figure 2.21: Fly ash particle penetration as a function of particle size for ESPs and FFs for various boilers and fuels (Jokiniemi 2003)

The nature of the fine particles released from coal-fired power stations has been studied for many years. Early work is summarized and reviewed by Damle *et al* (1982). More recent work at full scale (Kauppinen and Pakkanen, 1990) and at pilot scale (Galbreath *et al*, 2000) has extended the understanding of the size and composition as a function of size.

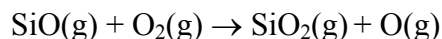
Submicron particulate material produced in combustion processes largely arises from vaporisation and condensation processes, although recent results (Linak *et al*, 2002) have revealed a contribution, probably from fragmentation processes, to particles in the 0.7 - 3.0 μm size range.

The main features of this mechanism are as follows:

- During combustion highly reducing conditions can exist inside coal particles
- Under these conditions refractory oxides can be reduced to more volatile suboxides or elements; for example, in the case of Si:



- The volatile species is transported away from the particle into the bulk gas where O_2 concentrations are significantly higher, and the suboxide or element is re-oxidised:



- Provided the vapour pressure of the oxide exceeds the saturated vapour pressure spontaneous condensation will occur, and nuclei of the submicron fume will be formed

This model was extended by Sarofim and co-workers (Quann and Sarofim, 1982) to account for observed distributions of inorganic species in the fine ash products from coal combustion; and further by Haynes (1999) who developed a detailed kinetic model for silica vaporization and condensation.

Submicron particle formation during combustion and post-combustion processes is important in trace element deportment. Trace toxic species such as arsenic and selenium can vaporise during combustion forming volatile combustion products. These species are often not present in sufficient concentrations to homogeneously condense into particles. Heterogeneous condensation of these species on existing particles is, in that case, more likely. Finer particles contribute relatively more to the available surface area than coarser particles, so enrichment of the more volatile trace elements in the fine particle fraction is often observed, as detailed above.

The deposition mechanism of the vaporised trace elements onto ash particle surfaces can result in a mechanism-specific correlation between trace element concentration and particle size. For example, deposition rate limited by ash particle surface reaction kinetics is governed by the reaction rate at the surface area, and leads to a $1/d_p$ dependence, where d_p is the particle diameter. Such relationships, together with ESP particle penetration information, and similar information for fabric filters, may be useful in developing practical models of trace element emission.

2.7.2 Secondary formation of sulfate from sulfur trioxide / sulfuric acid emissions

The generation of electricity from the combustion of sulfur containing fossil fuels results in the production of sulfur oxides, mostly sulfur dioxide but including lesser amounts of sulfur trioxide and sulfates. Sulfur trioxide may be converted to sulfuric acid prior to being emitted to the atmosphere and if emitted directly to the atmosphere is likely to be rapidly converted to sulfuric acid. Because of its low vapour pressure sulfuric acid rapidly condenses as sulfate onto existing aerosols, or, at sufficiently high concentrations, nucleates to create new aerosols. In the presence of excess ammonia, the sulfate will be neutralised to form ammonium sulfate, $[NH_4]_2SO_4$, particles, which are less than $1\ \mu m$ in diameter. This component of the secondary aerosol is likely to form close to the sources (i.e. in a similar pattern to the primary particle emissions).

If sulfur trioxide were produced in a stack *without* moisture present, sulfuric acid would not be produced. However, in stacks from fossil fuel combustion processes both water and sulfur trioxide are combustion products and they have a great affinity for each other – they react very rapidly to form sulfuric acid. In stacks from combustion processes in which there is moist air, (which includes water vapour) any reaction that forms sulfur trioxide can be considered equivalent to forming sulfuric acid.

From the above it is considered necessary to include the power station emissions of sulfur trioxide / sulfuric acid as precursors of fine particle matter in the atmosphere and to assess the potential contribution to atmospheric concentrations.

2.7.3 Secondary formation of sulfate and nitrate from SO₂ and NO_x

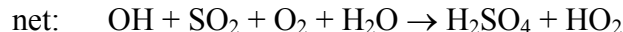
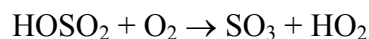
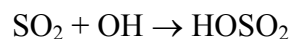
Fine particles can arise from so-called secondary processes in the atmosphere, in which particles are formed by physical and / or chemical transformations of precursor species such as SO₂, NO_x and organic species. Hence, in addition to the mechanisms discussed above, power station emissions of SO₂ and NO_x can also be converted to particulate phase sulfate and nitrate.

The processes by which the gas to particle conversions occur are complex, with both homogeneous (gas phase) and heterogeneous (aqueous phase) processes contributing.

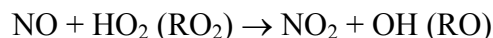
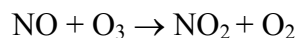
Gas phase oxidation

The following gas phase reactions have been identified as being most important (Brasseur et al 1999).

SO₂:



NO_x:



The rate at which oxidation occurs is a function of hydroxyl radical (OH) concentration, which in turn is strongly influenced by temperature and radiation, by the concentrations of NO_x, ozone (O₃), water vapour and volatile organic compounds (VOCs), the latter being emitted from many anthropogenic and natural sources (i.e. see Carnovale et al., 1996). It can be concluded that the rate of gas phase production of secondary particles within a power station plume is a strong function of the atmospheric environment into which the plume is emitted and transported. This is borne out by the literature review of Hewitt (2001) who documented observed gas phase oxidation rates for SO₂ in the range < 0.1–30% h⁻¹.

It should be noted also that, as the oxidation processes are primarily dependent on the production of OH radicals under sunlight, the formation of gas-phase sulfate effectively ceases at night time and the formation of gas-phase nitrate slows to much lower rates.

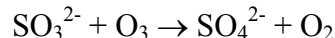
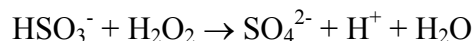
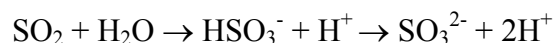
Reactions between excess ammonia, the major neutralizing species in the atmosphere, and the acid aerosols formed by the above gas-phase processes results in the formation of ammonium nitrate and ammonium sulfate. The sulfate is almost exclusively ultra fine material ($<1\ \mu\text{m}$), but the nitrate can partition between $\text{PM}_{2.5}$ and larger material.

Conversion of SO_2 and NO_x to sulfates and nitrates in power station plumes has been reviewed by Hewitt (2001) and a summary from that paper is presented later in this section (2.7.4).

Aqueous phase oxidation

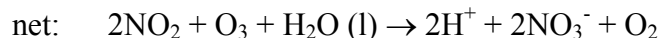
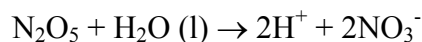
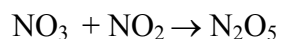
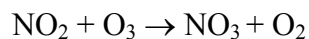
Sulfur dioxide and nitrogen oxides can also undergo aqueous-phase oxidation to sulfate [as S(VI)] and nitrate. When ozone or hydrogen peroxide (H_2O_2) are present in cloud water, aqueous-phase oxidations rates of SO_2 to SO_4^{2-} can be 100s of percent per hour. As a consequence, in-cloud aqueous-phase SO_2 oxidation is an efficient source of sulfate, which is then available to be deposited to the ground through wet deposition, or to condense and become sulfate particles if the cloud water evaporates. The relative contribution of gas phase and aqueous phase sulfate and nitrate production to the overall loading of particle mass in an airshed will depend strongly on the availability and characteristics of clouds, the likelihood that the clouds and the SO_2 and NO_x plumes will interact and the probability that rainout occurs.

SO_2 :



NO_x :

Gas phase followed by aqueous phase (in droplets, cloud particles):



In the context of the current study, modelling the complex processes involved in aqueous-phase SO_2 and NO_x oxidation and the subsequent potential deposition pathways has not been possible. Modelling of secondary aerosol production is limited to the gas-phase processes only. As a consequence, the predicted sulfate and nitrate loading resulting from SO_2 and NO_x oxidation within the power station plumes may be an underestimate under some circumstances.

While it is not possible to quantify the possible significance of the aqueous phase (and subsequent evaporation) contribution to secondary particle occurrence, extensive deposition measurements in regional NSW by CSIRO (Ayers 1995, 1997) have shown that dry deposition (of acidic gases) accounts for 70 - 80% of the total deposition flux, wet deposition contributes 20

– 30% and sulfate and nitrate aerosols are “inconsequential” to the total deposition fluxes. Qualitatively, this would suggest that the exclusion of the contribution of fine particles formed from the evaporation of cloud droplets containing sulfate and nitrate ions, is unlikely to lead to a significant change in the model predictions and that any such underestimate is likely to be more than offset by conservative assumptions adopted in other areas of the study, including:

- Full load, constant operation of all power stations
- Abundant ammonia
- Ammonium nitrate all in the fine fraction (see below).

Sulfate and nitrate particle sizes

Of relevance to the current study are the aerosol size fractions in which the sulfate and nitrate PM resides. In the case of sulfate, it may be expected (Seinfeld and Pandis, 1998) that it will reside in the fine fraction (i.e. less than 1 μm) as a result of gas-phase condensation or from heterogenous aqueous-phase reactions.

Nitrate PM has been observed to reside in both the fine and coarse particle fractions. Fine fraction ($< 1 \mu\text{m}$) nitrate aerosol is usually in the form ammonium nitrate and is a result of gas-phase condensation. Coarse fraction nitrate is formed from the reaction of nitric acid with sodium chloride or aerosol crustal material (i.e. sea salt and dust, Seinfeld and Pandis 1998). This is illustrated in Figure 2.22 in which the nitrate size distribution for Sydney has been plotted. The figure shows that 31% of the nitrate is in the $\text{PM}_{2.5}$ size fraction and that 54% is in the $\text{PM}_{2.5}$ to PM_{10} size fraction. 85% of the nitrate is therefore in the PM_{10} size fraction. (Personal communication, Melita Keywood, CSIRO). Thus a majority of the nitrate is in the coarse fraction ($> 2.5 \mu\text{m}$). The results presented in Figure 2.22 are representative of urban Sydney. It is not clear as to how representative these results may be of an elevated power station plume in which the degree of mixing with air containing ammonia, sea salt aerosol and crustal material may be reduced. In this study, the conservative assumption has been made that all of the nitrate aerosol will reside in the $\text{PM}_{2.5}$ size category. Note that this will leave the PM_{10} calculations unaffected given that $\text{PM}_{2.5}$ is also a sub-set of PM_{10} .

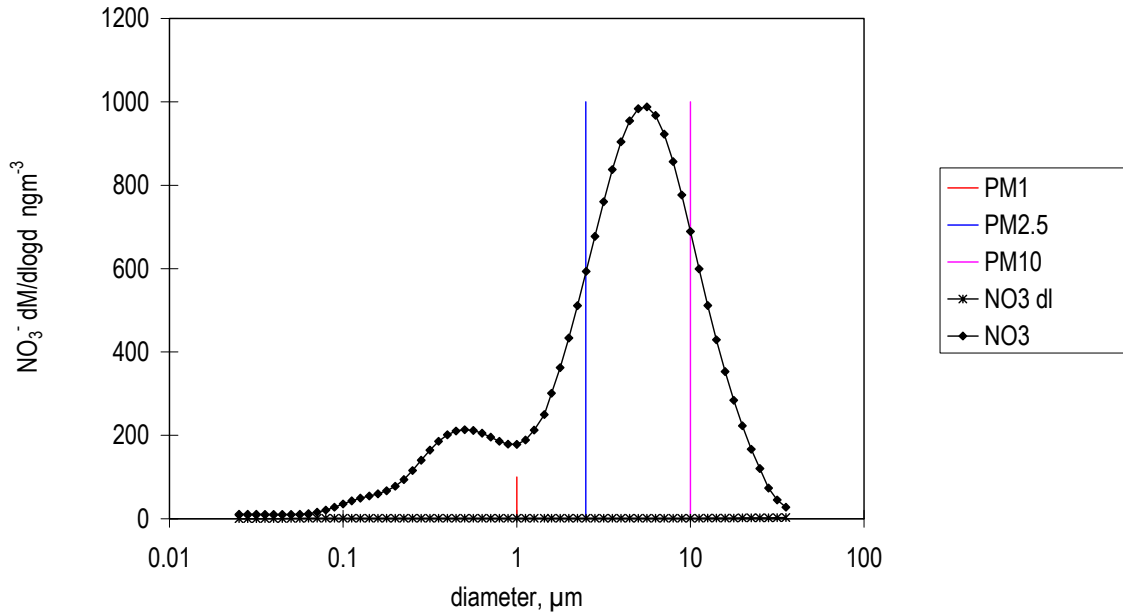


Figure 2.22: Size distribution of nitrate as observed in Sydney (Melita Keyword CSIRO; personal communication)

2.7.4 Summary of plume chemistry

The following summary of plume chemistry is taken from Hewitt (2001):

- In non-cloudy conditions, SO_2 removal in power station plumes occurs primarily during the daytime by reaction with the OH radical, whereas NO_x removal occurs both during the daytime, by fast reaction with OH, and more slowly at night-time by the $\text{NO}_3/\text{N}_2\text{O}_5$ pathway.
- In non-cloudy conditions NO_x removal will occur much more rapidly (~10 times faster) than SO_2 removal.
- In cloudy conditions, SO_2 will be removed rapidly by reactions in the aqueous water, but NO_x will not.
- The dry deposition velocity of SO_2 is greater than that of NO_x leading to more rapid removal of SO_2 by this process. Conversely, nitrate aerosol is likely to be removed more rapidly by dry deposition than the sulfate aerosol.
- These differences in removal rate will cause changes in the ratios of S and N concentrations with time of travel from the point of emission. In clear air, as the plume travels downwind the $\text{SO}_2:\text{NO}_x$ ratio will increase. In cloudy conditions, where aqueous phase reactions become important and photochemical processes less important, the SO_2 oxidation will proceed faster than NO_x oxidation and hence the $\text{SO}_2:\text{NO}_x$ ratio may decrease.
- SO_2 and NO_x removal rates will normally be lower in a plume than in background air, due to oxidant limitations, both in the gas and aerosol phases, with plume fringes offering an intermediate oxidant environment.

- Absolute oxidation rates of SO_2 and NO_x will vary with plume and background air composition and ambient conditions. In sunny conditions a maximum SO_2 conversion rate of around 3% per hour and a maximum NO_x conversion rate of 30% per hour might be expected. However lower rates may be expected in normal power station plumes as oxidant supply becomes diminished by consumption of O_3 (by reaction with NO_x), although the rate of oxidation of NO_x will remain ~ 10 times that of SO_2 in photochemically active conditions.

In addition to the above summary from Hewitt, it can also be noted that while the oxidation of NO_x proceeds faster than the oxidation of SO_2 in clear conditions, in the presence of available ammonia in the atmosphere ammonium sulfate is formed preferentially to ammonium nitrate. In ammonia limited conditions, sulfate aerosols can persist in the un-neutralised acid form while nitrate will remain in the gas phase.

2.7.5 Estimating power station emissions

This section describes the method of estimating power station emissions of the relevant substances for the purposes of modelling the contribution of power station emissions to the atmospheric concentration of PM_{10} and $\text{PM}_{2.5}$. For each power station, emission rates for the following substances were estimated:

- Primary particles
 - $\text{PM}_{2.5}$ and PM_{10}
- Sulfur trioxide / sulfuric acid
- Sulfur dioxide
- Nitrogen oxides

Primary particle emissions

Emissions of particulate matter from the seven coal fired power stations were estimated using individual power station characteristics:

- Station generating capacity in megawatts (MW).
- The station's conversion efficiency, on a generated basis (%).
- The average specific energy of the coal burnt (MJ/kg).
- The average ash content of the coal burnt (%)
- The fly ash fraction of the ash generated
- The gross collection efficiency of the particulate collection device.

It was conservatively assumed that all power stations were operating at maximum load throughout the year (2002) being modelled.

The above data provided estimates for primary PM emission from the individual power stations. The USEPA Compilation of Air Pollutant Emission Factors, AP-42 (USEPA 2003) was used to partition the total PM into relevant size fractions as included in Table 2.18.

Table 2.18: Partitioning of coal-fired power station primary particle emissions into size fractions.

Collection Device	% of PM in Size Category	
	PM _{2.5}	PM ₁₀
Electrostatic precipitator	29	67
Fabric filter	53	92

A summary of all emission rates for all power stations is provided in Table 2.19 at the end of this section.

Sulfuric acid emissions

Available emission testing⁵ data from NSW coal-fired power stations has shown considerable variability in the sulfur trioxide/sulfuric acid concentrations, although, on average, it would appear that sulfur trioxide/sulfuric acid emissions are about 1% of the sulfur dioxide emission.

Formation and emission of SO₃ from coal-fired plants has recently been reviewed by Fernando (2003). This review concludes that typical SO₃ concentrations from coal-fired plants are less than 5 ppm, depending on a range of variables including sulfur concentration, transition metals such as vanadium, nickel and iron which can act as catalysts, excess air levels and ash content and composition.

The USEPA Emergency Planning and Community Right-to-Know Act document - Guidance for Reporting Sulfuric Acid (USEPA 1998) includes emission factors for sulfuric acid based on the fuel sulfur content. The emission factor for coal, shown below, results in 1.9% of the sulfur in coal being emitted as sulfuric acid.

Coal combustion:

H_2SO_4 (kg/tonne) = 0.19 x S, where S = weight per cent sulfur in coal
Eg 2% Sulfur, S = 2

The advantage of using an emission factor over site specific testing is that the factor will result in temporal and spatial consistency in the reported amounts of the substance, whereas stack testing is likely to show considerable variability from test to test, which may be a function of test and analytical procedures, rather than due to fuel or operating parameters. The USEPA factor was used to provide estimates for the emission of sulfuric acid from the NSW power stations in the modelling component.

A summary of all emission rates for all power stations is provided in Table 2.19 at the end of this section.

⁵ Testing of sulfur trioxide/sulfuric acid mist.

Sulfur dioxide and nitrogen oxide emissions

SO₂ and NO_x emission rates were based on the assumption that all power stations were operating at full load throughout the year (2002) being modelled. As discussed above in relation to primary emissions, this is clearly a very conservative assumption.

Sulfur dioxide emission rates were calculated using the average sulfur content of coal delivered to each power station and assuming that all the sulfur was emitted as sulfur dioxide. This is a conservative assumption as it is known that a small fraction of the sulfur is retained in collected ash and further, as discussed above, some of the sulfur is emitted as sulfur trioxide / sulfuric acid.

Nitrogen oxide emissions rates previously developed for IRTAPS (Nelson 2002) were applied in the modelling of fine particles. The continuous full load assumption is more conservative for NO_x than it is for SO₂, as the mixing ratio (ppm) of NO_x often increases, sometimes significantly, as boilers approach maximum output. An example of this is shown in Figure 2.23. This relationship is due to the fact that part of the NO_x emitted is formed from nitrogen contained in combustion air (thermal NO_x) and due to turbulence disturbing the low NO_x reducing conditions when all burners are in operation. Hence, the increase in the mass of NO_x emitted with load is due to higher output (greater gas flows) combined with a higher NO_x concentration in each cubic metre of exhaust gas emitted. With no change in coal sulfur content, the mixing ratio of SO₂ in the flue gases stays relatively constant with load.

A summary of all emission rates for all power stations is provided in Table 2.19 at the end of this section.

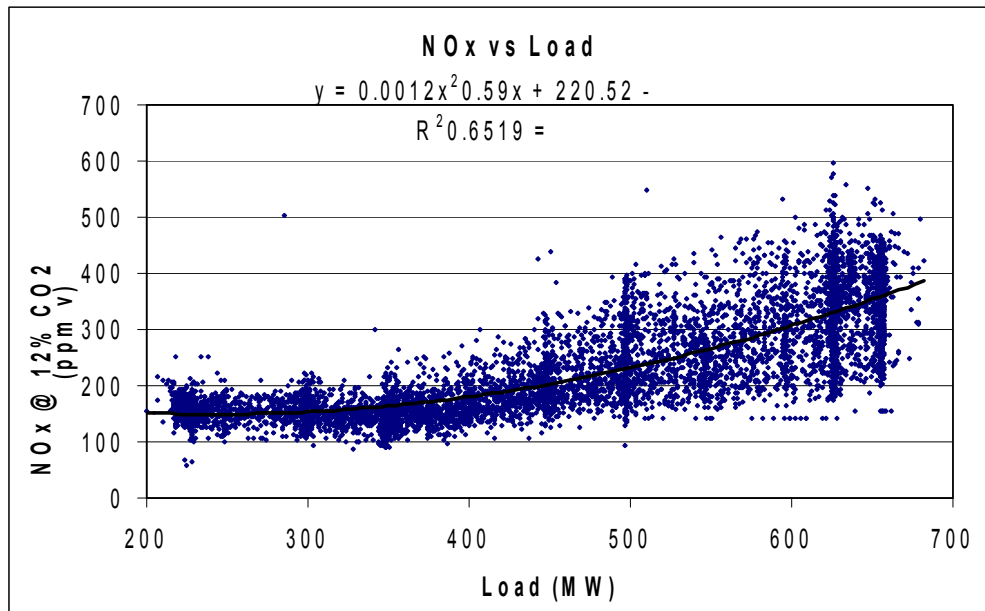


Figure 2.23: NO_x concentration versus load for a NSW generating unit.

Table 2.19: Source characteristics of all power stations considered in the TAPM modelling

<i>Source</i>	<i>Easting (m)</i>	<i>Northing (m)</i>	<i>Stack height (m)</i>	<i>Radius (m)</i>	<i>Plume velocity (m s⁻¹)</i>	<i>Plume temperature (K)</i>	<i>PM_{2.5} (g/s)</i>	<i>PM₁₀ (g/s)</i>	<i>NO_x (g/s)</i>	<i>SO₂ (g/s)</i>	<i>H₂SO₄ (g/s)</i>
Central Coast											
Eraring: stack 1	361839	6340550	200	5.24	23	403	13.13	22.73	947.61	1129.50	10.73
Eraring stack 2	361862	6340770	200	5.24	23	403	13.13	22.73	947.61	1129.50	10.73
Vales Point	364250	6329710	178	5.15	20	384	41.91	96.83	1119.70	1309.10	12.44
Munmorah	364090	6324100	155	3.95	18.2	403	7.77	13.49	323.33	467.50	4.44
Hunter Valley											
Liddell: stack 1	309700	6416310	168	4.34	22.2	396	8.60	14.93	611.11	1382.50	13.14
Liddell: stack 2	309720	6416460	168	4.34	22.2	396	8.60	14.93	611.11	1382.50	13.14
Bayswater stack 1	307210	6413800	250	5.28	23	403	10.34	17.98	828.67	2016.80	19.16
Bayswater stack 2	306950	6413910	250	5.28	23	403	10.34	17.98	828.67	2016.80	19.16
Western											
Mt Piper	223710	6304880	250	5.51	23	403	0.72	1.25	1152.79	1549.60	14.72
Wallerawang stack 1	228690	6300120	177	2.95	26.3	394	2.91	6.73	416.67	640.80	6.09
Wallerawang stack 2	228760	6300120	177	3.15	23.2	394	2.91	6.73	375.00	640.80	6.09

3 MODELLING OF PARTICLE CONCENTRATIONS

This section documents the outcomes of the numerical modelling component of the study, which was undertaken by the CSIRO Divisions of Energy Technology and Atmospheric Research. The numerical modelling was used to estimate the ground level concentrations of PM_{2.5} and PM₁₀ resulting from the direct emission of particles from power station stacks, and from the in-plume conversion of gaseous species to ambient particles as the result of photochemical transformation processes.

The modelling has been undertaken for power stations (see Figure 3.1) located on the Central Coast, in the Hunter Valley and to the west of Sydney. Two components of work have been undertaken.

1. Using an inventory of primary particle emissions for each of the power stations and a numerical meteorological-chemical transport model, the near field and regional (i.e. including Sydney) peak 24 h and annual average PM_{2.5} and PM₁₀ concentrations have been estimated.
2. For the task of estimating secondary particle concentrations, two different modelling approaches were considered in developing a preliminary estimate of the likely worst-case concentrations of ammonium sulfate and ammonium nitrate as a result of gas-phase oxidation processes.

The methodology and results from the primary particle modelling are presented in Section 3.1. Outcomes from the modelling of secondary particle production are discussed in Section 3.2. Section 3.3 provides a synthesis of the results arising from the primary and secondary particle modelling for the near-field and regionally. The CSIRO report including all results and more detailed descriptions of the methodologies employed is included in a separate volume to this report.

3.1 Primary Particle Modelling

The modelling of peak ground-level concentrations of primary particle emissions (i.e. fly ash) has been undertaken using TAPM (Hurley 2002), a combined weather prediction, chemical transport modelling system. TAPM solves the governing equations of mass, momentum, energy, moisture and tracer transport on a series of user defined, nested grids. Initial and boundary conditions for the meteorological fields are provided by a large scale analysis, generated by the Bureau of Meteorology. The system is able to simulate the transport of tracers, primary particles, and a simple photochemical system including sulfate and nitrate for a variety of source characteristics. The system is typically run for periods of days to months. A detailed technical description of the system is available from (http://www.dar.csiro.au/publications/hurley_2002a.pdf). Verification studies are documented in (http://www.dar.csiro.au/publications/hurley_2002b.pdf).

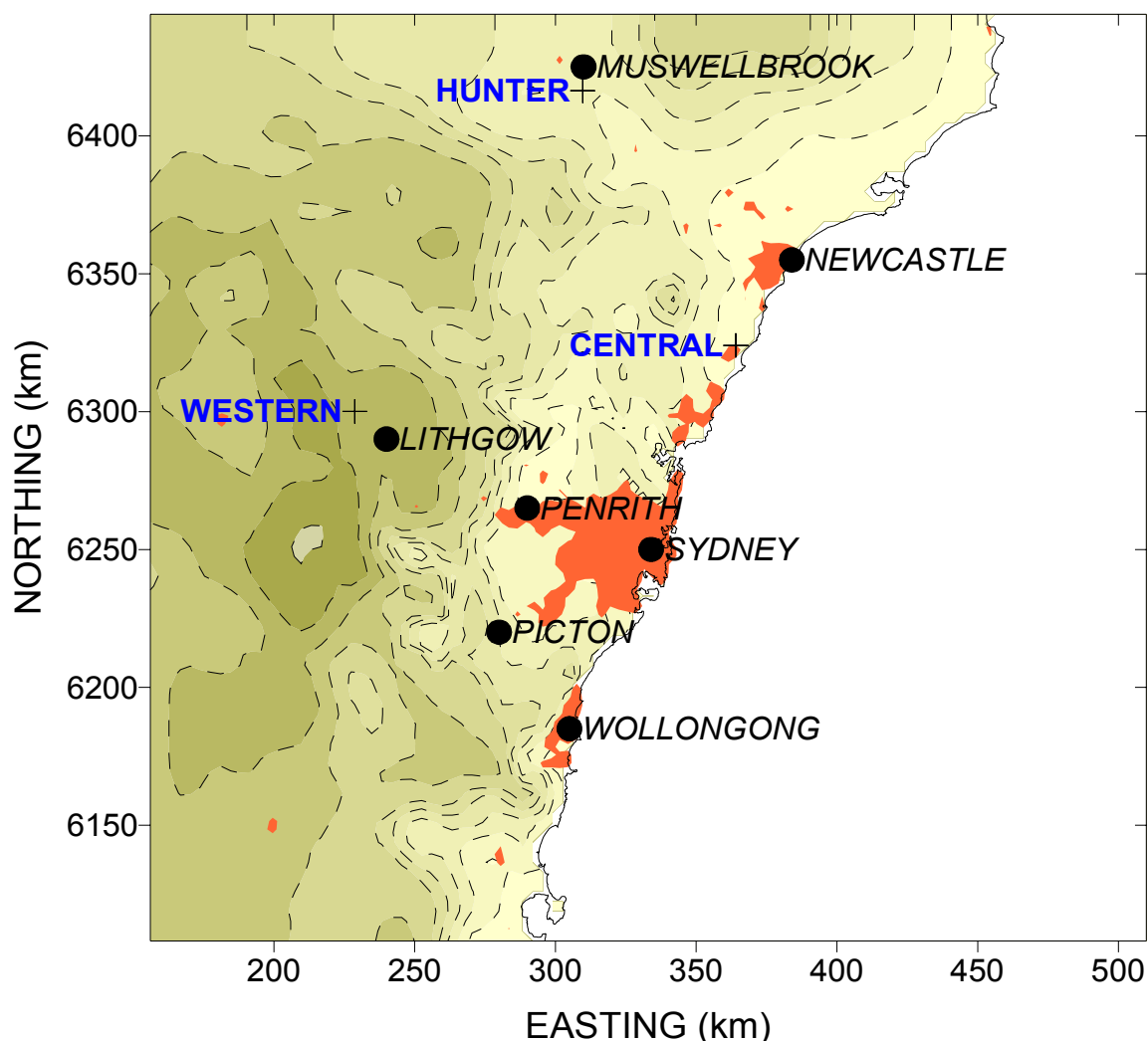


Figure 3.1: Map of the east coast of NSW, showing the location of the three groups of power stations modelled in this study: Hunter Valley, Central Coast and Western). Also shown are the topographic relief (dashed lines), selected population centres and regions where the population density exceeds 500 people per km² (orange).

The simulation of PM_{2.5} and PM₁₀ primary emissions has been undertaken by running TAPM in ‘tracer’ mode in which a pollutant is emitted, advected and diffused without undergoing the processes of wet and dry deposition and chemical transformation. TAPM has been used to predict peak 24 h and annual average concentrations of the two particle size fractions. Emissions from each power station group have been considered individually, and in the case of the regional modelling for all groups in combination, as well as individually.

3.1.1 TAPM domains

The TAPM simulations have been undertaken for three near-field grids and one regional grid. The near-field grids were configured to capture the regions of maximum primary particle matter (PM) impact within a ~20 km radius of each power station source group (Central Coast, Hunter Valley and Western). The regional grid has been designed to capture the combined impact of all the power station sources, particularly within the Sydney metropolitan region.

Each near-source grid system has been configured with 3 nested grids. For the meteorological simulations a grid spacing of 20, 6 and 2 km respectively has been selected with mesh sizes of 25 x 25 x 25 (x-y-sigma). The air quality simulations have been conducted on double resolution grids- 10, 3 and 1 km with mesh sizes of (41 x 41 x 25) points. The highest resolution meteorological and air quality grids for the three regions are shown in Figure 2 of Appendix 1.

A single grid system, which encompasses all of the power station groups, has been used for the regional modelling. The system consists of 2 nests (12 km and 4 km grid spacing) with mesh sizes of 75 x 75 x 25 nodes. The meteorological and air quality grids are identical. The inner (4 km) nest is shown in Figure 3 of Appendix 1.

3.1.2 Source and run-time definitions

The prescribed emissions of primary particles were discussed and presented in Section 2.7 and summarised in Table 2.19. Also shown in Table 2.19 are relevant source characteristics such as stack height, plume temperature and efflux velocity for each power station.

Each of the TAPM simulations was run for a 12 month period, using initial and boundary meteorological conditions which are representative of 2002.

3.1.3 Primary particle results

This section presents the predicted peak 24 h and annual average PM_{2.5} and PM₁₀ concentrations arising from primary PM emissions. Results are presented for the near-field and regional domains. It should be noted that the results are for power station primary PM emissions only; emissions from non-power sources are not included.

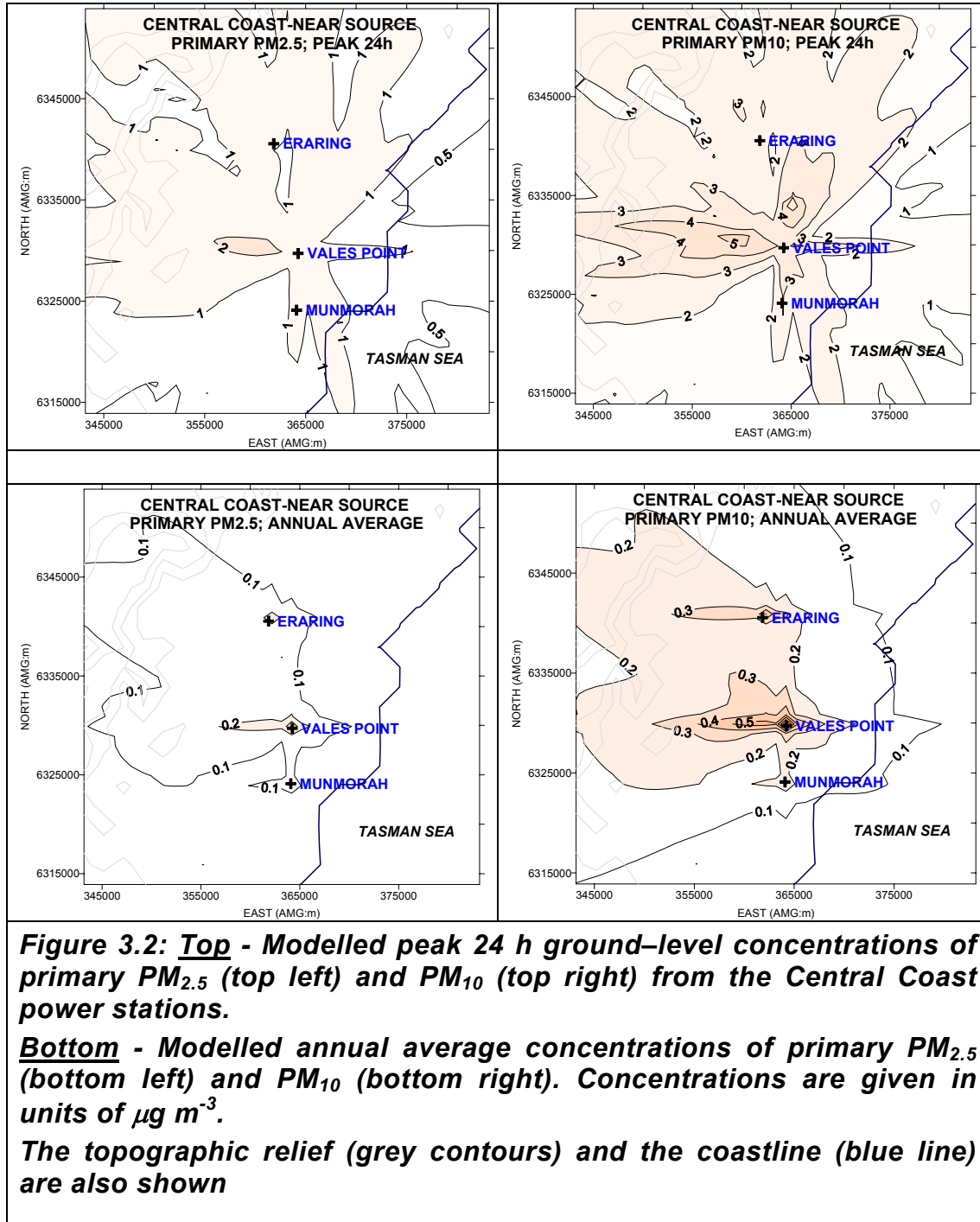
Some context as to the significance of the results may be gained by recalling that the National Environment Protection Measure (NEPM) for PM₁₀ is 50 µg m⁻³ (24 h average), and that the NEPM advisory reporting standards for PM_{2.5} are 25 µg m⁻³ (24 h average) and 8 µg m⁻³ (annual average).

Near-field primary particle results

Near-field results for each power station region are shown in Figure 3.2 (Central Coast), Figure 3.3 (Hunter Valley) and Figure 3.4 (Western). Results for all three regions are summarised in Table 3.1.

It can be seen that the highest PM 24 h average concentrations are predicted to occur within 10 km of the power station sites in all three regions. The peak annual average concentrations are also predicted to lie close to the sources, generally falling within the same computational cell as

the point of emission. This indicates that the annual average concentrations are dominated by near-source convective fumigation events, and that the spatial scales for plume touch-down are smaller than the computational grid size (i.e. less than 2 km).



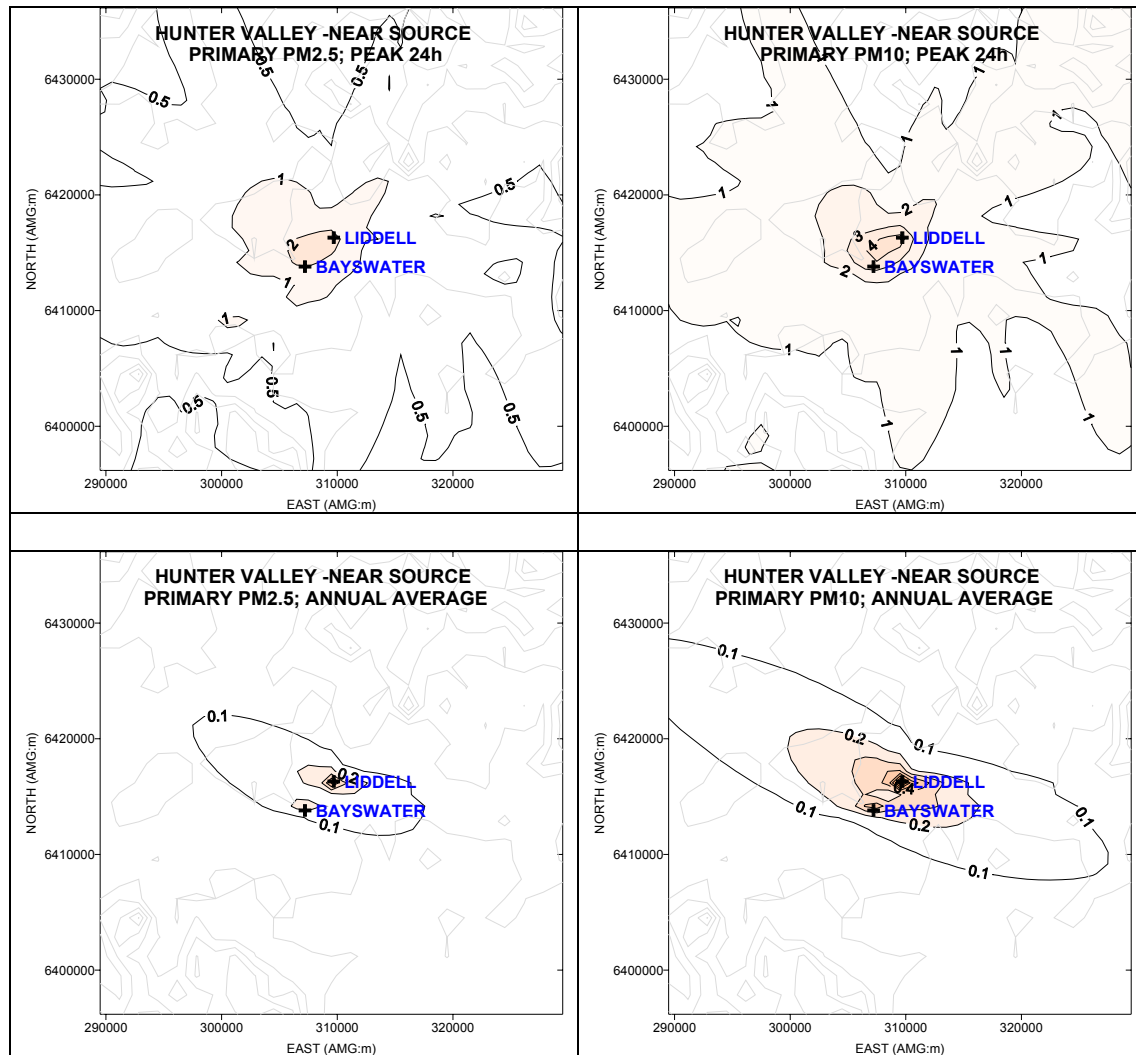


Figure 3.3: Top - Modelled peak 24 h ground-level concentrations of primary PM_{2.5} (top left) and PM₁₀ (top right) from the Hunter Valley power stations.

Bottom - Modelled annual average concentrations of primary PM_{2.5} (bottom left) and PM₁₀ (bottom right).

Concentrations are given in units of $\mu\text{g m}^{-3}$. The topographic relief (grey contours) are also shown.

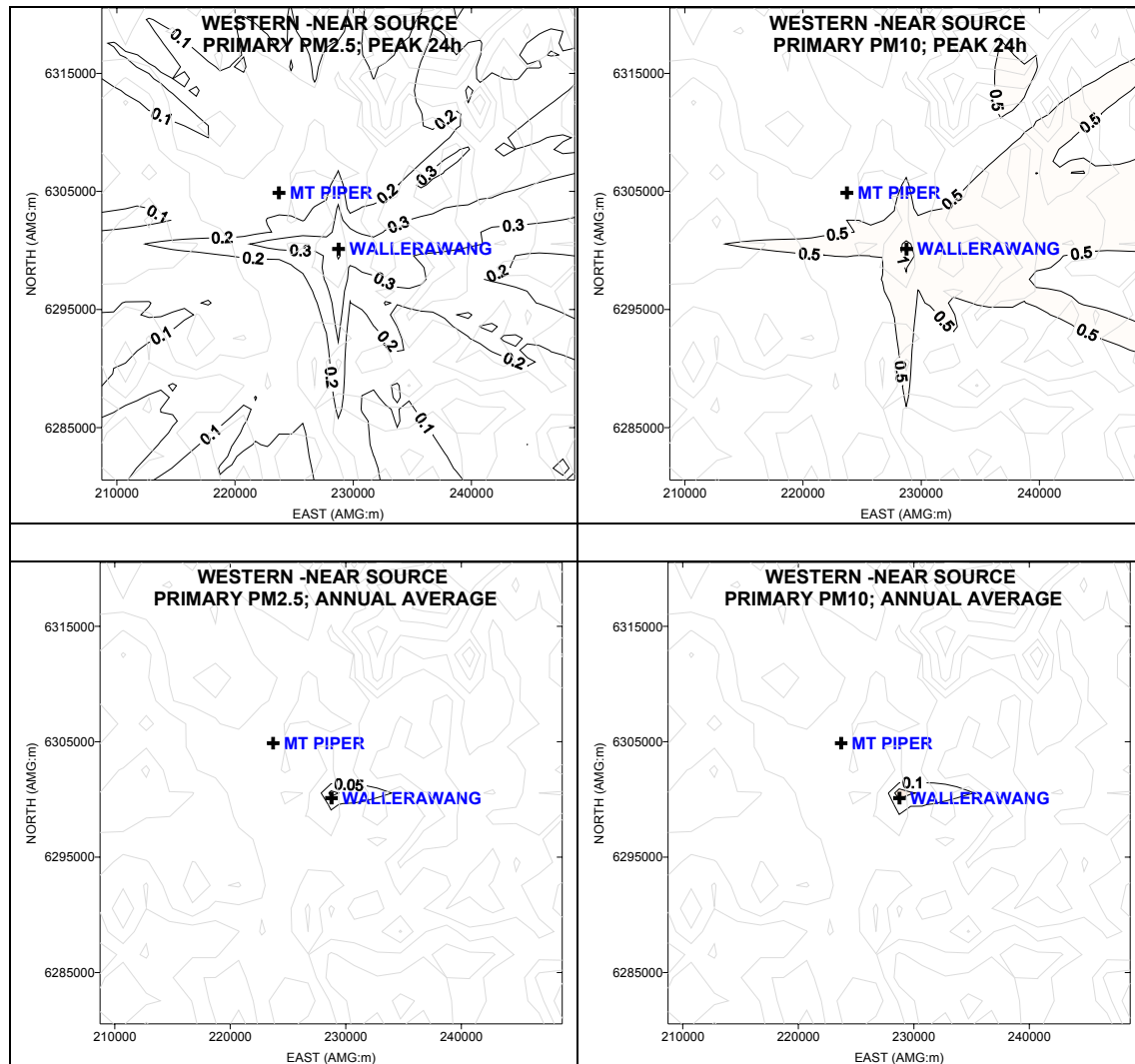


Figure 3.4: Top - Modelled peak 24 h ground-level concentrations of primary PM_{2.5} (top left) and PM₁₀ (top right) from the Western power stations.

Bottom - Modelled annual average concentrations of primary PM_{2.5} (bottom left) and PM₁₀ (bottom right). Concentrations are given in units of $\mu\text{g m}^3$.

The topographic relief (grey contours) are also shown.

In summarising the primary near-field results, the peak ground-level 24h PM_{2.5} and PM₁₀ concentrations are typically 10% or less of the associated NEPM standards in the Hunter Valley and Central Coast. The Western power station group is predicted to have the smallest impact, with peak 24h concentrations reaching 2 - 3% of the associated NEPMs. The maximum annual average PM_{2.5} concentrations are calculated to be 1 - 5% of the associated PM_{2.5} NEPM advisory reporting standard.

Some guidance as to the veracity of the Hunter Valley results is available from the observational work of Hinkley et al. (2003) who used a Burkard spore sampler to collect PM₁₀ particles at Ravensworth approximately 7 km to the south east of Bayswater and Liddell power stations. Samples were collected between May and December 2002 and the exposed tapes were cut into 1-day segments and analysed using a scanning electron microscope. The observed daily maximum PM₁₀ concentration was 0.33 $\mu\text{g m}^{-3}$ (0.31–0.88 95% confidence interval, averaged over a period of less than one hour). By contrast, the model predicted peak 24 h PM₁₀ concentrations of 1–2 $\mu\text{g m}^{-3}$ for the Ravensworth site, which while higher, are not inconsistent with the observed concentration peak when it is recalled that the modelling was undertaken for an entire 12 month period using maximum load operating conditions. (Note the peak predicted concentration at Ravensworth is lower than the regional maximum shown in Table 3.1).

Table 3.1: Modelled peak ground-level PM_{2.5} and PM₁₀ concentrations (24h and annual average) in the near-field for primary particle emissions from the major power station groups in NSW, operating at continuous maximum load.

<i>Power station group</i>	<i>Peak 24h average</i>		<i>Annual average</i>	
	<i>Primary PM_{2.5} ($\mu\text{g m}^{-3}$)</i>	<i>Primary PM₁₀ ($\mu\text{g m}^{-3}$)</i>	<i>Primary PM_{2.5} ($\mu\text{g m}^{-3}$)</i>	<i>Primary PM₁₀ ($\mu\text{g m}^{-3}$)</i>
<i>Near-field</i>				
<i>Central Coast</i>	2.3	5.3	0.4	0.9
<i>Hunter Valley</i>	2.6	4.5	0.5	0.9
<i>Western</i>	0.5	1.2	0.1	0.3

Regional primary particle results

Regional results for the three power station groups are shown in Figure 3.5 (24 h averages) and Figure 3.6 (annual averages). Note that the results from the individual power station groups have not been added, but rather plotted on the same figure) The peak regional-scale concentrations are lower than the near-field concentrations for both PM_{2.5} and PM₁₀ for both 24h and annual averaging times. This occurs because the peak concentrations for the primary particles all occur close to the source (i.e. well within the bounds of the near-field grids), and the coarser resolution (4 km vs. 1 km) regional grid is not able to resolve power station plumes as well in the near field.

Considering the plot of peak 24h PM_{10} (Figure 3.5- rhs), it can be seen that the Central Coast power station group is predicted to be the largest contributor of PM of the three power station groups.

The predicted regional distribution of annual average $\text{PM}_{2.5}$ and PM_{10} concentration is shown in Figure 3.6. Similarly to the 24 h predictions it can be seen that the highest concentrations occur close to the sources. Again, the Central Coast power station group is predicted to be the most significant of the three power station groups.

Figure 3.5 and Figure 3.6 indicate that for the Sydney region the primary particle emissions from the power stations are predicted to contribute

- Less than $1 \mu\text{g m}^{-3}$ to 24 h average PM_{10} and $\text{PM}_{2.5}$ concentrations
- Less than $0.1 \mu\text{g m}^{-3}$ to annual average PM_{10} and $\text{PM}_{2.5}$ concentrations

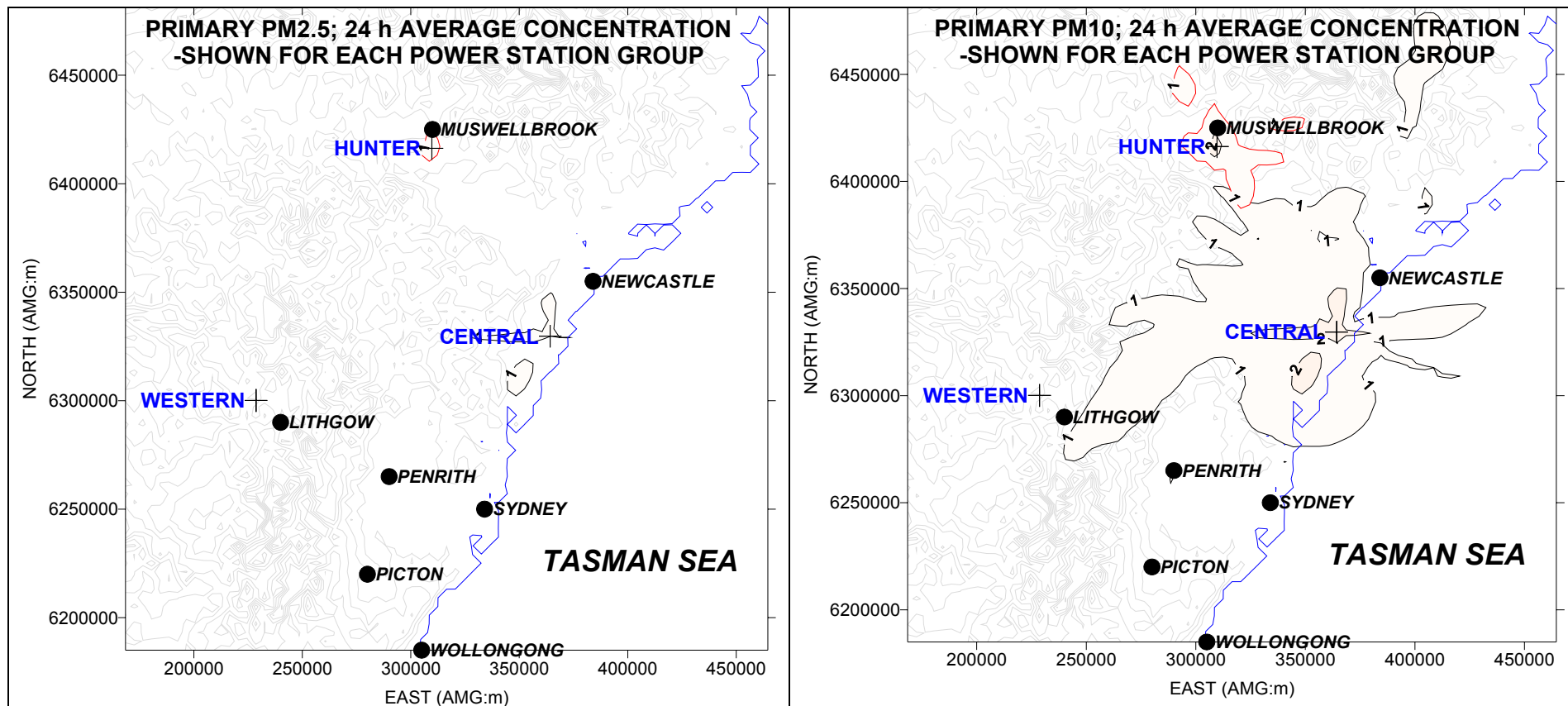


Figure 3.5: Modelled regional peak 24 h ground-level concentrations of primary PM_{2.5} (left) and PM₁₀ (right) from each of the power station groups (plotted for each group in isolation, not as the summation across all the groups) for the regional domain. Concentrations are shown in units of $\mu\text{g m}^{-3}$. Also shown is the topographic relief (grey contours) and the coastline (blue line). A minimum concentration isopleth of $1.0 \mu\text{g m}^{-3}$ has been displayed.

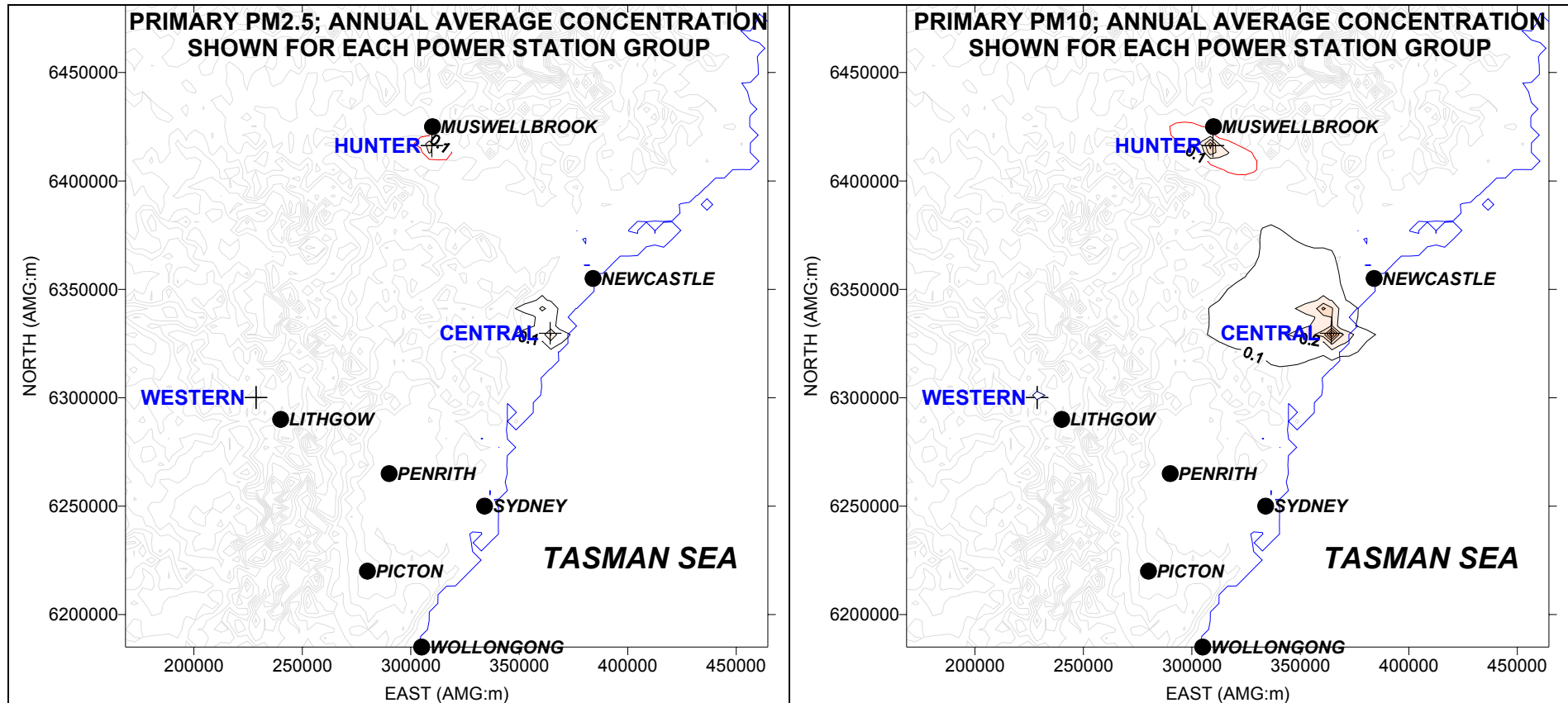


Figure 3.6: Modelled regional annual-average ground-level concentrations of primary PM_{2.5} (left) and PM₁₀ (right) from each of the power station groups (plotted for each group in isolation, not as the summation across all the groups) for the regional-scale domain. Concentrations are shown in units of $\mu\text{g m}^{-3}$. Also shown is the topographic relief (grey contours) and the coastline (blue line). A minimum concentration isopleth of $0.1 \mu\text{g m}^{-3}$ has been displayed.

3.2 Secondary Particle Modelling

As discussed earlier, there are three major pathways for secondary particle production from power station emissions; near-field conversion of H_2SO_4 emissions, gas-phase and aqueous phase particle production. In this section predictions of the sulfate concentrations resulting from the direct emissions of H_2SO_4 are presented followed by the outcomes of modelling gas-phase sulfate and nitrate production. Again it is noted that the complexities of modelling aqueous-phase secondary aerosol production has precluded it from inclusion in the current modelling study.

3.2.1 Sulfate and nitrate aerosol – neutralisation and size fractions.

In the case of the near-field and regional modelling presented in the next sections, the mass of secondary particle matter has been estimated by assuming that sufficient ammonia exists so as to enable all of the sulfate and nitrate to exist in the aerosol phase as ammonium sulfate $[(\text{NH}_4)_2\text{SO}_4]$, and as ammonium nitrate (NH_4NO_3) . Some guidance as to the conservatism of this approach may be gained by considering the outcome of a NH_3 sampling program which was conducted during the Hunter Valley Dry Deposition Study (Manins et al., 1996). During this study, weekly NH_3 concentrations were measured using a passive sampling device at a site in the upper Hunter Valley. Referring to Figure 3.7, it can be seen that concentration peaks of 8–15 ppb were observed during the first half of the observation period, while concentrations in the range 2–5 ppb were observed during the second half of the observation period. The annual average NH_3 concentration was 4.62 ppb. Assuming that this ammonia is available to produce ammonium sulfate or ammonium nitrate leads to an upper bound annual average concentration of $12.5 \mu\text{g m}^{-3}$ $(\text{NH}_4)_2\text{SO}_4$ or $15.2 \mu\text{g m}^{-3}$ of NH_4NO_3 .

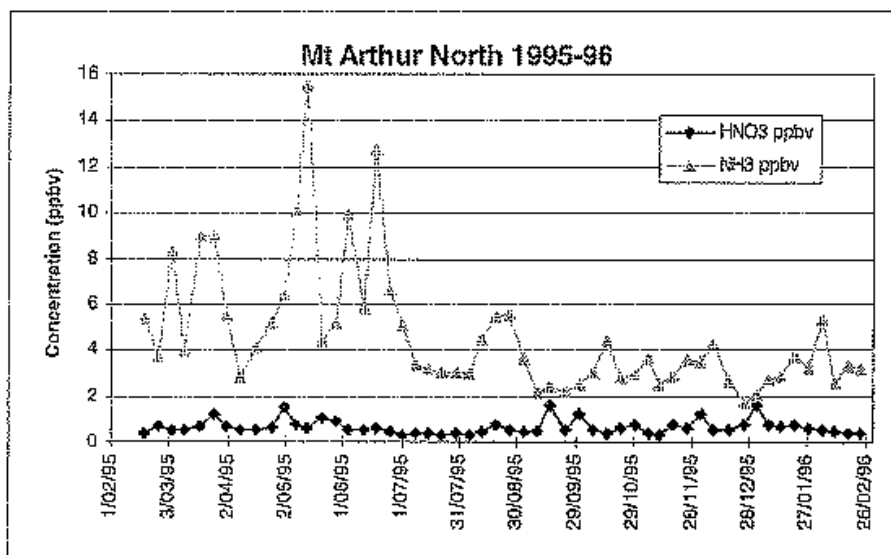


Figure 3.7: Time series concentrations of weekly average ammonia and nitric acid as observed in the upper Hunter Valley over the period February 1995 – February 1996.

As discussed in Section 2.7 it is assumed that all ammonium sulfate and ammonium nitrate will reside in the $< 1.0 \mu\text{m}$ size fraction. As shown in that section this is likely to be a conservative assumption for ammonium nitrate, which has been shown to exist in both the fine and coarse size fractions.

3.2.2 Near-field secondary sulfate formation from SO_3 / H_2SO_4 emissions

Table 2.19 shows that some of the power station sulfur emissions are in the form of sulfur trioxide / sulfuric acid. The low vapour pressure of this gas causes it to rapidly condense onto existing particles, or if present in sufficiently high concentrations, to nucleate into new particles. As noted above, we have made the assumption that ammonia exists in sufficient concentrations as to fully neutralise the sulfate to form ammonium sulfate and it is also assumed that the sulfate particles are fully neutralised within several kilometres of the power station stacks.

As discussed in the following sections, the aerosol production from H_2SO_4 emissions are combined with the secondary sulfate generated via the gas-phase oxidation pathways to provide a single figure for ammonium sulfate. In this section, however, results for near-field ammonium sulfate from the emission of H_2SO_4 are presented without consideration of gas phase conversion of emitted SO_2

Given the rapid rate of transport of H_2SO_4 from the gas phase to the aerosol phase, it may be assumed that near-field ammonia sulfate concentrations will be dominated by the conversion of H_2SO_4 emissions and will exhibit concentration peaks with a similar spatial distribution to the primary particle emissions. This assumption allows the peak ground-level near-field sulfate concentrations to be estimated from the peak near-field PM concentration predictions and the ratios of the emitted PM and H_2SO_4 . Note that this approach is not exact for the sources considered in this study because the ratio $\text{H}_2\text{SO}_4:\text{PM}_{2.5}/\text{PM}_{10}$ is not the same for all of the power stations within a given source group, mainly due to the significant difference in PM collection efficiencies between fabric filters and electrostatic precipitators. However, for the cases in which the ratios vary significantly between power stations in the region (i.e. for Central Coast and Western), we have selected the ratios for the power stations which contribute most to the primary PM concentration peaks, which are the ESP equipped stations.

The predictions for ammonium sulfate production from H_2SO_4 emissions are summarised in Table 3.2. In the majority of cases, the secondary aerosol component is predicted to generate ambient concentration peaks which are comparable to, or greater than, those resulting from the primary $\text{PM}_{2.5}$ emissions, which were presented in Table 3.1 and shown again in Table 3.2 for each region.

In particular, it can be seen that peak 24 h near-field $[\text{NH}_4]_2\text{SO}_4$ concentrations are predicted to lie in the range $1 - 7 \mu\text{g m}^{-3}$. The annual average near-field $[\text{NH}_4]_2\text{SO}_4$ concentrations are predicted to lie in the range $0.2 - 1.3 \mu\text{g m}^{-3}$. The most significant source group is the Hunter Valley, where peak 24 h, near-source $[\text{NH}_4]_2\text{SO}_4$ concentration is predicted to reach $6.5 \mu\text{g m}^{-3}$ and the annual average concentration is predicted reach $1.3 \mu\text{g m}^{-3}$. Combining these predictions of secondary aerosol with the primary PM predictions, shown in Table 3.1, yield total predicted $\text{PM}_{2.5}$ concentrations of 9.1 and $2.0 \mu\text{g m}^{-3}$ respectively for 24 h and annual average

concentrations from this source group (36% and 25% of the NEPM reporting standards for $PM_{2.5}$.)

The regional grid results, in brackets, are all generally less than the near-field results.

Table 3.2: Near-field (and regional grid) $[NH_4]_2SO_4$ concentrations estimated from predicted $PM_{2.5}$ concentrations and the ratio of emission of $PM_{2.5}$ and H_2SO_4 .

Source	$PM_{2.5}$ Emitted (g/s)	H_2SO_4 Emitted (g/s)	Ratio of $[NH_4]_2SO_4$ and $PM_{2.5}$ emitted	Predicted Peak primary 24 h $PM_{2.5}$ ¹ ($\mu g m^{-3}$)	Peak 24 h sulfate ² ($\mu g m^{-3}$)	Predicted annual primary $PM_{2.5}$ ¹ ($\mu g m^{-3}$)	Annual sulfate ² ($\mu g m^{-3}$)
Central Coast							
Eraring:	26.26	21.46	1.10	2.3 ³ (1.3)	0.9 (0.5)	0.4 (0.3)	0.2 ³ (0.15)
Vales Point	41.91	12.44	0.40				
Munmorah	7.77	4.44	1.20				
Hunter Valley							
Liddell	17.2	26.28	2.10	2.6 (1.4)	6.5 ⁴ (3.5)	0.5 (0.3)	1.25 ⁴ (0.8)
Bayswater	20.68	38.32	2.50				
Western							
Mt Piper	0.72	14.72	27.59	0.5 (0.4)	1.4 ⁵ (1.1)	0.1 (0.1)	0.3 (0.3)
Wallerawang	5.82	12.18	2.82				

¹As given in Table 3.1. ²As $[NH_4]_2SO_4$ equivalent; ³For peak downwind of Vales Point (Figure 3.2);

⁴Using Bayswater ratio; ⁵For peak downwind of Wallerawang (Figure 3.4).

With respect to the validity of the near-field ammonium sulfate predictions, it is noted that they are consistent with sulfate observations collected during the ERDC Aerosol Sampling Project (ANSTO et al. 1995), where fine particle sampling was conducted at Muswellbrook (approximately 12 km away from the closest power station in the Hunter Valley). Ammonium sulfate concentrations were deduced from sulfur concentrations by assuming that the sulfur as sulfate was fully neutralised by ammonia. A time series plot of the observed daily ammonium sulfate concentrations for the Muswellbrook site for the period July–December 1992 is shown in Figure 3.8. It can be seen that 24 h ammonium sulfate concentrations were observed to range between $< 1000 \text{ ng m}^{-3}$ ($< 1 \mu g m^{-3}$) to nearly 6000 ng m^{-3} ($6 \mu g m^{-3}$). This is consistent with the range of ammonium sulfate concentrations predicted by TAPM on the near-field grid, particularly when the distance between Muswellbrook and the power station sites is taken into account.

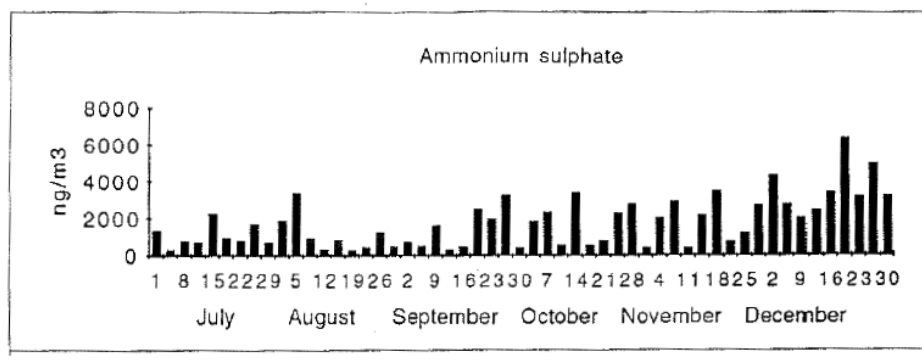


Figure 3.8: Time series plot (July–December 1992) of daily $[\text{NH}_4]_2\text{SO}_4$ concentrations (ng m^{-3}) for Muswellbrook in the Hunter Valley NSW. ANSTO et al. (1995).

3.2.3 Regional secondary sulfate and nitrate concentrations

In this section the regional scale impacts resulting from the in-plume production of ammonium sulfate and ammonium nitrate are considered. The gas phase production of sulfate and nitrate has been modelled using two levels of chemical modelling detail. The first and simpler approach uses a special version of TAPM's tracer mode in which NO_x loss and nitrate production, and SO_2 loss and sulfate production are coupled using first order conversion rates in which the generation rate of a product species (e.g. sulfate) is a function only of a single reactant species (e.g. SO_2). This approach is relatively computationally in-expensive to run, thus allowing the use of long term integrations of TAPM to estimate annual average particle concentrations, and the distribution of peak 24h average particle concentrations. However, the approach provides a simplified representation of the actual non-linear processes which control the generation of sulfate and nitrate from their parent species. A brief description of the method and the associated modelling outcomes are presented in Appendix 1.

The second approach uses a new version of TAPM; TAPM-CTM in which the TAPM meteorology has been coupled with a complex chemical transport model. Use of this system enables a more comprehensive treatment of the chemical and aerosol transformations to be specified. However, the method is computationally and data intensive, and thus has only been applied to a limited number of case study days. A discussion of the approach and the associated modelling outcomes is given after the next section and in Appendix 1.

First-order chemical transformation modelling methodology

As discussed above, this approach uses a special version of TAPM's tracer mode in which NO_x loss and nitrate production, and SO_2 loss and sulfate production are coupled using first order conversion rates. The mechanism was configured to allow separate conversion rates for the sulfate and nitrate system, and conversion rates which varied by day and night and seasonally.

Critical to this approach is the determination of rates for the conversion of NO_x and SO_2 to aerosol nitrate and sulfate which are representative of the conditions present in

the greater Sydney region. These conversion rates can then be applied in the numerical modelling and used to generate peak 24 h and annual average concentrations of PM_{2.5} and PM₁₀ in the Sydney region.

Conversion rates were developed through consideration of the literature (i.e. Hewitt 2001) details of which were discussed in Section 2.7 and through a numerical modelling study in which the dispersion and chemical transformation of NO_x and SO₂ within the Sydney regions power station plumes were simulated using a high resolution Lagrangian wall model (LWM; see Appendix 1). The Lagrangian simulations were undertaken for meteorological conditions conducive to photochemical smog production, and yielded estimates of plume-average first-order chemical production rates for nitric acid and sulfate production.

The LWM was run for 34 scenarios, covering a range of meteorological conditions, a range of background NO_x concentrations, a range of background VOC concentrations and speciation, for daylight and nocturnal conditions and for cases of deposition enabled and no deposition. This ensemble of runs was considered likely to cover the range of conditions for which gas phase conversion of the sulfur and nitrogen would occur within the greater Sydney region during summer conditions. Given the conversion rates increase with increasing temperature and radiation, it may be concluded that the derived conversion rates will represent an upper bound for the other seasons of the year.

A complete set of results is shown in Appendix 1, and the range of the peak results is shown in Table 3.3, categorised into species, day and night, deposition and no deposition.

Table 3.3: Modelled range of peak removal and production rates (% h⁻¹)

Scenario	Species			
	SO ₂ (% h)	SO ₄ ²⁻	NO _x	NO ₃ ⁻
<i>Day time</i>				
<i>Deposition on</i>	(-2) – (-4)	0.02 – 0.2	(-0.2) – (-4.0)	0.3 – 1.8
<i>Deposition off</i>	(-0.03) – (-0.7)	0.02 – 0.2	(-0.1) – (-2.5)	0.3 – 1.9
<i>Night time</i>				
<i>Deposition on</i>	(-0.1) – (-0.3)	< 0.01	(-0.1) – (-0.5)	0.06–0.15
<i>Deposition off</i>	< -0.01	< 0.01	(-0.1) – (-0.3)	0.07–0.2
Recommended Rates				
<i>Day time</i>		0.2% h ⁻¹		2% h ⁻¹
<i>Night time</i>		0% h ⁻¹		0.2% h ⁻¹

Also included in Table 3.3 are a set of recommended conversion rates. These are based on the review of Hewitt (2001), the modelling results presented in Table 3.3, and discussions between members of the project team. Note that while the recommended conversion rates lie at the low end of the range observed in numerous field experiments as documented in Hewitt 2001, they are approximately five times greater than the peak conversion rates derived from the Lagrangian Wall modelling.

Thus, it is considered that the recommended conversion rates are likely to be conservative for the meteorological and air quality conditions modelled in this study.

The TAPM configuration for the first-order chemical transformation simulations was identical to that used for the regional primary emission modelling. Thus the model was run for a period of 12 months (2002), using the 4 km spaced grid as shown in Appendix 1. Emissions of NO_x and SO_2 and H_2SO_4 as required for the prediction of sulfate and nitrate production are given in Table 2.19. Note that the emitted SO_3 was assumed to be instantaneously converted to H_2SO_4 and thence to ammonium sulfate. The first order conversion rates are those recommended in Table 3.3, modified on a monthly basis to account for the reduced rates of photochemical activity in the cooler, less sunny winter months (See Appendix 1). A monthly variation was calculated as the ratio of the monthly integrated smog produced equation (Johnson 1984), normalised by the monthly integrated smog produced equation for the most chemically reactive month (January). This resulted in January first-order rates being five times higher than the winter rates.

In the first-order modelling component only emissions from power stations were considered.

First-order regional modelling results.

The predicted peak 24h and annual average concentrations of secondary and combined primary and secondary concentrations are given in Table 3.4. The predicted regional spatial distributions of combined primary and secondary particle are given in Figure 3.9 and Figure 3.10. Note that the sulfate and nitrate components of the PM are represented as ammonium sulfate and ammonium nitrate equivalent.

From Table 3.4 it can be seen that the peak 24 h secondary particle concentrations are predicted to lie in the range $6\text{--}16\ \mu\text{g m}^{-3}$. This is 24 – 64% of the 24 h $\text{PM}_{2.5}$ NEPM. Concentrations are predicted to be highest in the Hunter Valley, followed by the Western region. When combined with the primary PM emissions, peak 24 h $\text{PM}_{2.5}$ concentrations are predicted to lie in the range $6\text{--}17\ \mu\text{g m}^{-3}$. When concentrations from all of the sources groups are combined on the regional TAPM grid, the peak 24 h $\text{PM}_{2.5}$ concentration is predicted to be $17.3\ \mu\text{g m}^{-3}$.

The regional spatial distribution of the predicted peak 24 h $\text{PM}_{2.5}$ concentration is shown in Figure 3.9 (left). It can be seen that the highest concentrations are predicted to lie close to the Hunter Valley power station group. Concentrations of more than $10\ \mu\text{g m}^{-3}$ are also predicted to occur close to the Western power station group. Considering the Hunter Valley again, and referring to Table 3.2 it can be seen that primary PM and ammonium sulfate generated from H_2SO_4 emissions are predicted to contribute about $5\ \mu\text{g m}^{-3}$ to near source $\text{PM}_{2.5}$ totals on the regional grid. Thus the majority of the peak 24 h $\text{PM}_{2.5}$ concentration within the vicinity of the Hunter Valley power station group is predicted to result from the secondary particle production, which given the proximity to the power stations and the time required for secondary reactions to proceed is perhaps an indication of the conservative nature of the first-order reaction methodology.

The power stations are predicted to generate peak 24 h $PM_{2.5}$ concentrations of 5 - 6 $\mu g m^{-3}$ within the Sydney region, which again, results mainly from secondary processes.

Table 3.4: Peak ground-level $PM_{2.5}$ and PM_{10} concentrations (24h and annual average) for secondary particles, and combined primary and secondary particles for the three power station groups

	24h average				Annual average		
	Secondary $PM_{2.5}$ ($\mu g m^{-3}$)	Total $PM_{2.5}$ ($\mu g m^{-3}$)	Total PM_{10} ($\mu g m^{-3}$)		Secondary $PM_{2.5}$ ($\mu g m^{-3}$)	Total $PM_{2.5}$ ($\mu g m^{-3}$)	Total PM_{10} ($\mu g m^{-3}$)
Regional							
Central Coast	5.5	5.9	6.4		0.5	0.8	1.1
Hunter Valley	15.9	16.7	17.3		1.2	1.5	1.8
Western	12.7	13.0	13.5		0.5	0.6	0.7
Combined		17.3	17.8			1.7	2.0
Sydney		5 - 6	5 - 6			0.2 - 0.3	0.2 - 0.3

The predicted peak 24 h PM_{10} concentrations are only marginally higher than the $PM_{2.5}$ predictions. The modelled differences are due to the fraction of primary particle emissions which lie in the 2.5–10 μm size fractions (*viz.* Table 2.19). As such, the power station contribution relative to the higher PM_{10} NEPM (50 $\mu g m^{-3}$) are reduced (12 – 34%) compared to $PM_{2.5}$.

The predicted spatial distribution of peak 24 h PM_{10} (Figure 3.9-right), is seen to be almost identical to that of the $PM_{2.5}$ distribution. This is to be expected given that the only significant differences are due to the small primary PM contribution near to each source group.

The predictions of peak annual average $PM_{2.5}$ (and PM_{10}) concentrations are given in Table 3.4 and shown in Figure 3.10. The highest annual average concentrations are relatively low, falling in the range 0.5 – 1.2 $\mu g m^{-3}$ for the secondary $PM_{2.5}$ (6 – 15% of the $PM_{2.5}$ NEPM); falling in the range 0.6 – 1.5 $\mu g m^{-3}$ for the combined primary and secondary $PM_{2.5}$. When all power station sources are considered in combination, a peak annual average $PM_{2.5}$ concentration of 1.7 $\mu g m^{-3}$ is predicted (21% of the NEPM Advisory Reporting Standard). Referring to Figure 3.10 (left), it can be seen that the highest annual average concentrations are located relatively close to each power station source group. Peak annual average $PM_{2.5}$ concentrations are predicted to fall within the range 0.2 – 0.3 $\mu g m^{-3}$ over the Sydney region (less than 5% of NEPM Advisory Reporting Standard).

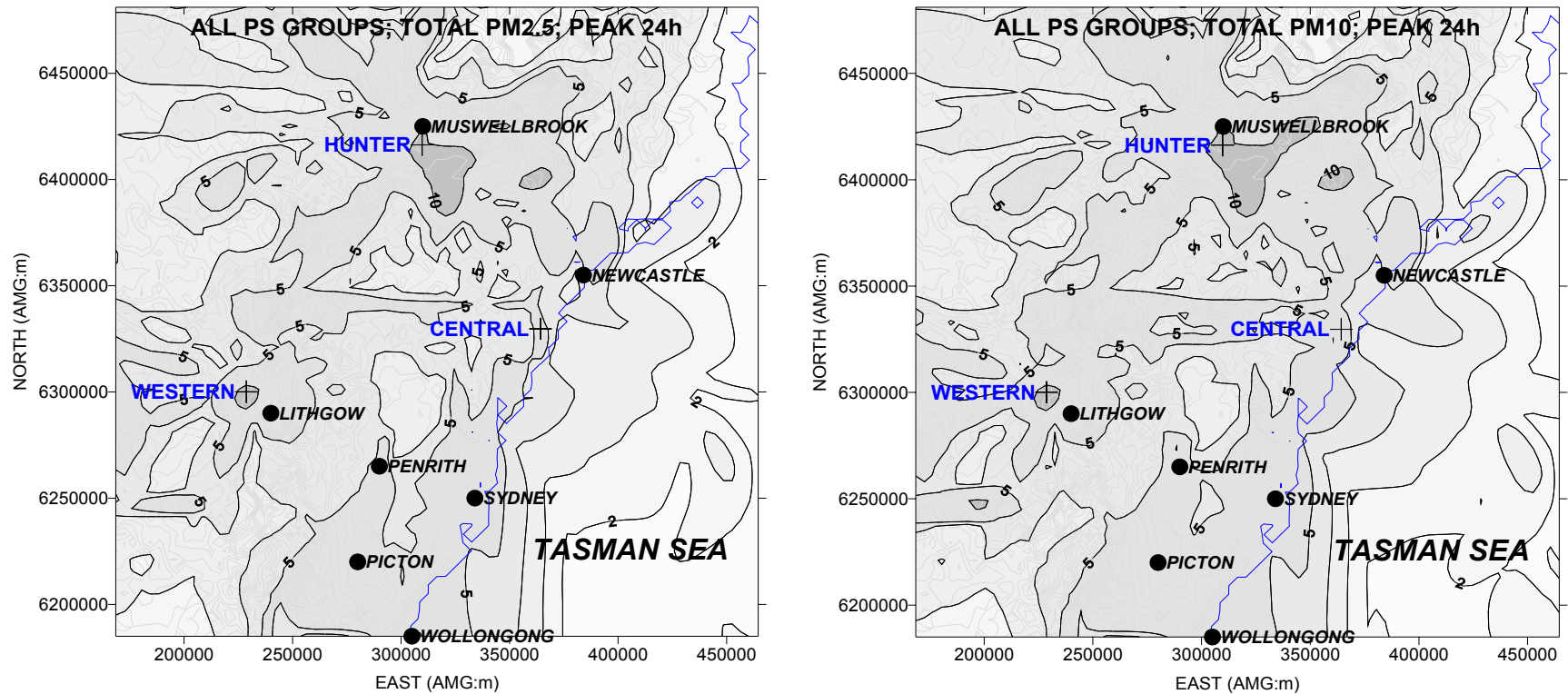


Figure 3.9: Top - modelled regional peak 24 h ground-level concentrations of total (primary and secondary) PM_{2.5} (left) and PM₁₀ (right) from all of the power stations. Concentrations are given in units of $\mu\text{g m}^{-3}$. The topographic relief (grey contours) and the coastline (blue line) are also shown.

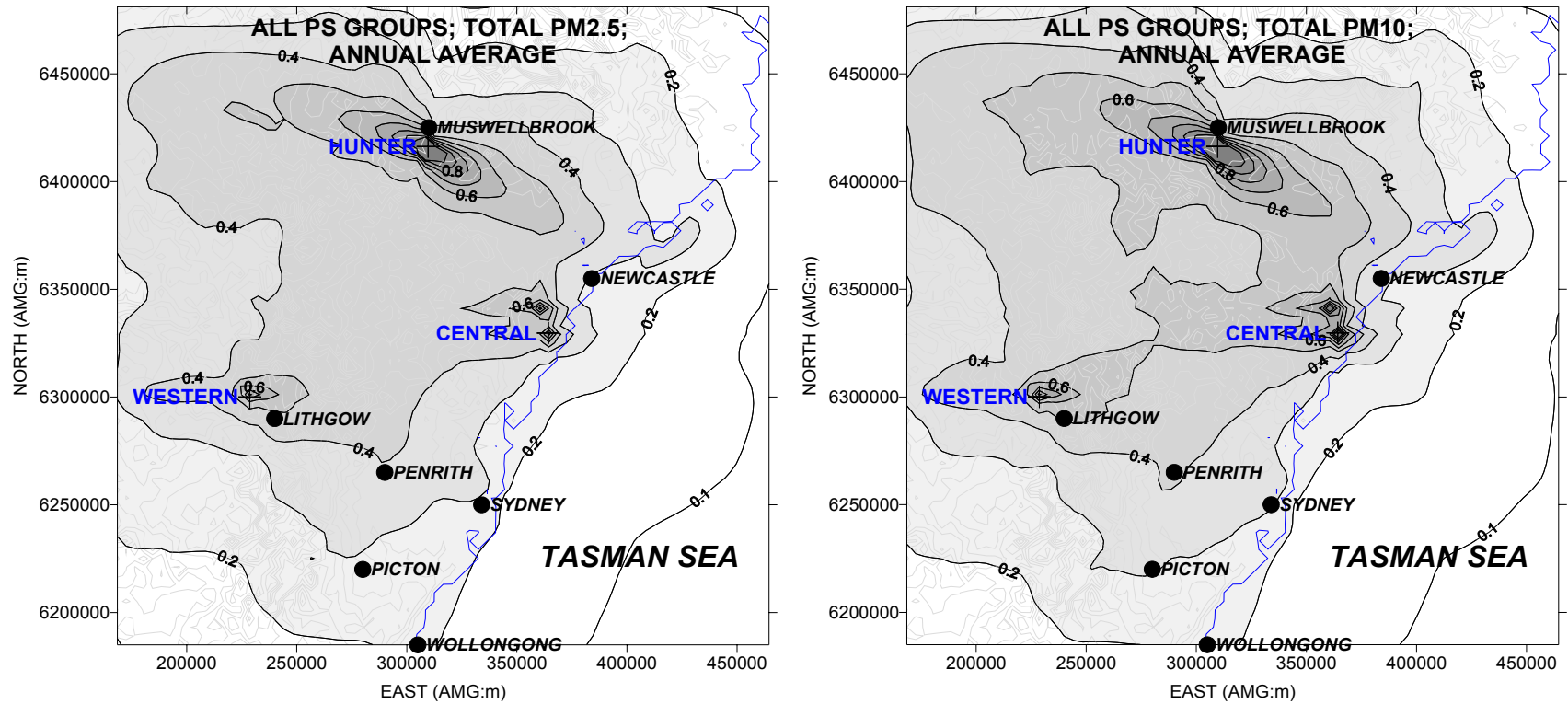


Figure 3.10: Top - modelled regional annual average ground-level concentrations of total (primary and secondary) PM_{2.5} (left) and PM₁₀ (right) from all of the power stations. Concentrations are given in units of $\mu\text{g m}^{-3}$. The topographic relief (grey contours) and the coastline (blue line) are also shown.

In order to undertake an initial assessment of the frequency with which power station emissions contribute to the formation of secondary sulfate and nitrate concentrations, the highest first-order predicted concentrations for each day of the modelled year (2002) were extracted and the results plotted as a frequency distribution in Figure 3.11. Note that the concentrations are normalised with respect to the maximum predicted 24 h concentration for the year, on the basis that the absolute first-order reaction rate results are considered to be overly conservative (as discussed in the next section). Also, the distribution contains the highest values which were predicted to occur anywhere in the regional grid on each day of the modelled year, rather than the daily concentrations predicted at an individual grid cell in the regional grid. Notwithstanding this conservatively derived distribution, Figure 3.11 exhibits a strongly skewed distribution – there is a rapid drop from maximum values, with the 90th percentile and median percentile concentrations being 40% and 15% of the maximum concentration respectively, indicating the highest predicted power station contributions are likely to occur infrequently.

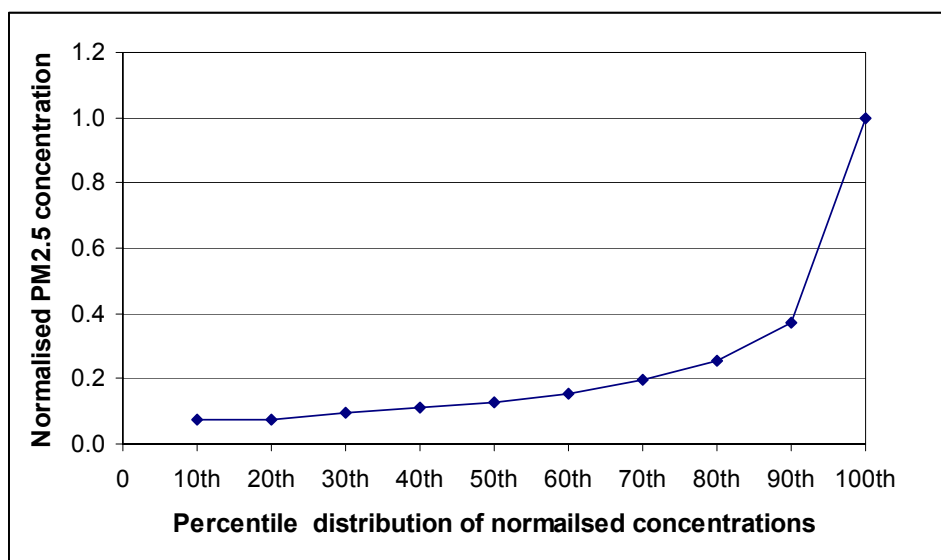


Figure 3.11: Normalised frequency distribution of power station emission contribution to regional secondary sulfate and nitrate.

Comprehensive Chemical Transformation Approach

In the previous section the use of first-order conversion rates led to the majority of the particle mass being predicted to result from secondary particle production. However, as discussed previously, the authors have reservations regarding the applicability of the first-order conversion rates, even though they fall towards the bottom of the range presented in Hewitt (2001). In particular, the rates are representative of the maximum values which often occur late in the day; rather than a sustained or average rate for the modelled meteorological and air quality conditions. Given this, it is likely that the secondary particle results presented in the previous section are conservative.

In order to investigate conversion rates further, a pre-release version of TAPM-CTM has been run for the worst-case summer and worst-case winter secondary aerosol days as predicted by TAPM using the first order conversion rates. TAPM-CTM includes a chemical transport module which is able to treat chemical transformation in a more

comprehensive manner than is available in TAPM. For the current study, TAPM-CTM was configured with the Lurmann, Carter, Coyner chemistry (Lurmann et al., 1987) and interfaced with the MARS (Saxena et al., 1986) aerosol module. MARS models the thermodynamic equilibrium of an ammonia, ammonium, sulfate, nitric acid, aerosol nitrate, aerosol water system.

The model was run with the MAQS 1992 emissions inventory (with year 2000 vehicle updates) for all surface anthropogenic sources. Biogenic emissions were also included. The use of the full MAQS inventory was required in order to predict background concentrations of nitric acid and $\text{SO}_2 \rightarrow \text{SO}_3$ oxidation as required by the MARS mechanism. Note that the inventory does not provide a speciation of sulfur emissions into gaseous and aerosol components (all SO_x is assumed to be emitted as SO_2). As a consequence, we have adopted the approach used in the Australian Air Quality Forecasting emissions inventory⁶ and assigned 3% (on a mass basis) of the emitted SO_x to SO_3 and 97% of the emitted SO_x to SO_2 . Point source emissions of NO_x , SO_2 and H_2SO_4 from each of the power stations are as given in Table 2.19. Primary particle emissions have not been included in the TAPM-CTM simulations.

The model was run using a uniform background ammonia concentration. This background has been used as a surrogate for the ammonia sources within the region (ammonia is not included within the current MAQS emissions inventory). An ammonia concentration of 5 ppb was chosen, this being close to the annual average concentration measured in the upper Hunter Valley in (1995–1996) during the Hunter Valley dry deposition study (Manins et al. 1996).

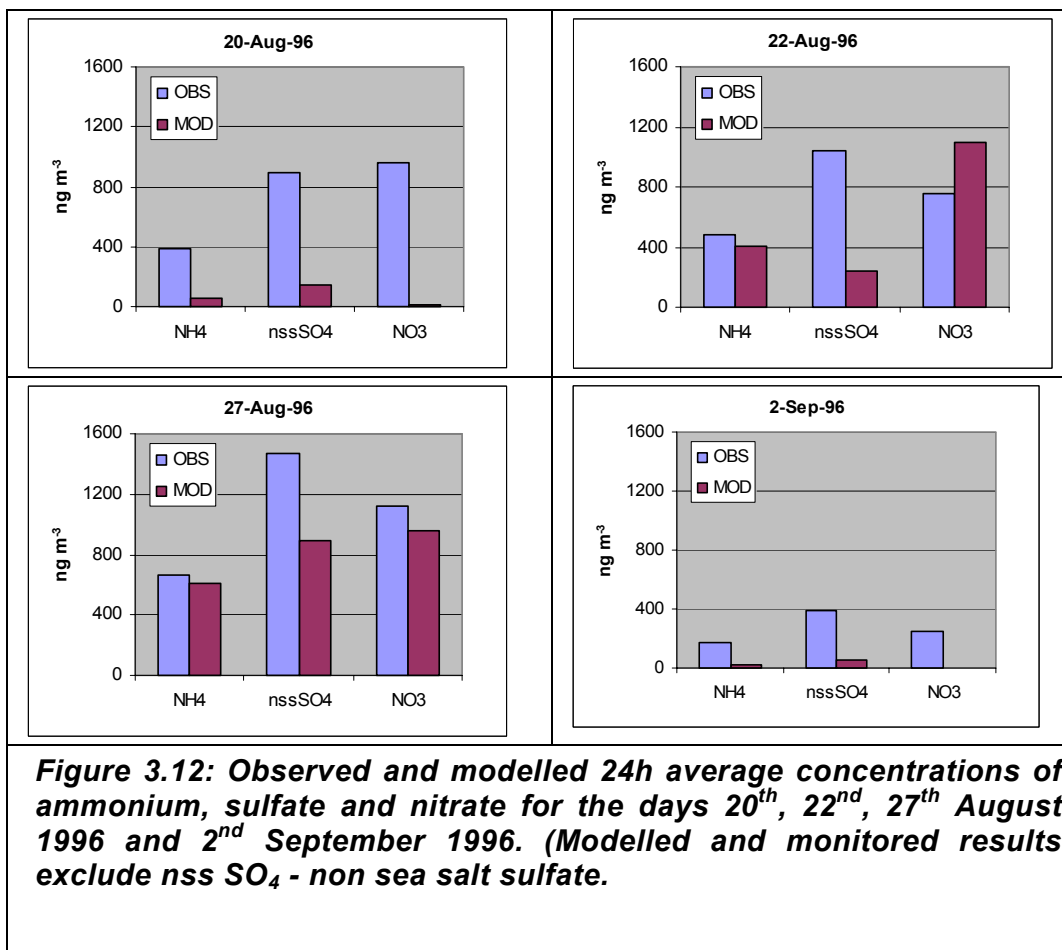
Simulations were undertaken using the same regional model domain as for the earlier TAPM simulations. For each scenario, the model was integrated for 48 hours, with the second 24 hours corresponding to the worst-case day. Emissions from all power station groups were included in a single run. In addition, scenarios were modelled in which the power station sources were omitted. The difference between the two emission scenarios then yielded the power station contribution.

Verification

In order to provide some confidence in the application of TAPM-CTM, predictions from the model have been compared against 24 h average concentrations of NH_4^+ , SO_4^{2-} and NO_3^- observed during a fine particle pilot study (Ayers et al. 1999 and Melita Keywood, personal communication). The observations were taken at Liverpool monitoring station in Sydney and covered the period 20, 22, 27 August 1996 and 2 September 1996. TAPM-CTM was integrated for four two-day periods commencing the day previous to each of the observation periods. The modelled and observed 24h average aerosol concentrations are shown in Figure 3.12. It can be seen that all of the aerosol components were under predicted by the model on 20th August. This case requires further investigation because the meteorological simulations generate strong winds for the majority of this 24 hour period. The model performs better on 22nd and 27th August, and under-predicts the (low) observed concentrations on 2nd September. For 22nd and 27th August, the model predicts the total concentrations of NH_4 and NO_3 with good skill. Sulfate is under predicted, an

⁶ (http://www.dar.csiro.au/publications/manins_2002axviii.pdf)

outcome which may arise from the omission of the domestic fuel combustion source group from the (summer) emissions inventory which was available for use with this study.



The model verification is still preliminary in nature (i.e. verification of the meteorological modelling component, and the gas-phase pollutants has not yet been undertaken). Nevertheless, the results are considered to be sufficiently promising as to provide confidence in the use of the modelling system for providing a preliminary assessment of the power station impacts.

Comprehensive regional modelling results

As noted above, TAPM-CTM was used to simulate a worst-case 24 h summer and worst-case 24 h winter particle episode. As these episodes have been selected from the first order rate TAPM modelling it should be noted that the days are likely to be worst-case with respect to power station impacts, but may not necessarily be worst-case with regards to impacts from the Sydney urban plume. Furthermore, in selecting these days, it is assumed that the TAPM first-order reaction methodology provides a sufficiently robust description of the particle dynamics as to be able to correctly identify the meteorological characteristics of these worst-case events.

12 March 2002. The meteorological conditions predicted for this day lead to the highest 24 h secondary particle concentrations for the modelled year (2002). The spatial distribution of peak 24 h concentration predicted by TAPM (with the first order methodology) is shown in Figure 3.9 and exhibits concentration peaks close to the sources in the Hunter Valley and near to the Western power stations. Use of the first-order reaction approach led to the prediction of peak 24 h $\text{PM}_{2.5}$ and PM_{10} concentrations of $17\text{--}18\ \mu\text{g m}^{-3}$ for this day on the regional grid. Primary particle emissions contributed $1\text{--}2\ \mu\text{g m}^{-3}$ and primary H_2SO_4 emissions contributed $3\text{--}4\ \mu\text{g m}^{-3}$ (ammonium sulfate equivalent). Within the Sydney region, peak $\text{PM}_{2.5}$ and PM_{10} concentrations from power station contributions of $5\text{--}6\ \mu\text{g m}^{-3}$ have been predicted using the first-order methodology). These results may be contrasted with the observed concentration range of $10\text{--}14\ \mu\text{g m}^{-3}$ ($\text{PM}_{2.5}$) and $17\text{--}29\ \mu\text{g m}^{-3}$ (PM_{10}) for this day. The observed concentrations obviously include the contribution from other anthropogenic sources and natural sources.

TAPM-CTM concentration predictions for the combined ammonium sulfate and ammonium nitrate concentrations for 12 March 2002 are shown in Figure 3.13 for urban and power station emissions (left-hand plot) and for power station emissions alone (right-hand plot). The predicted peak 24 h average total secondary sulfate/nitrate concentration is $3.4\ \mu\text{g m}^{-3}$. A combined peak concentration of $1\text{--}2\ \mu\text{g m}^{-3}$ is predicted for the Sydney region, of which power stations are predicted to contribute $0.1\text{--}1\ \mu\text{g m}^{-3}$. Note that Figure 3.14(right plot) is equivalent to the plot of worst case 24 h $\text{PM}_{2.5}$ plot shown in Figure 3.5 (with the exception that the Figure 3.5 also includes a small primary component ($< 2\ \mu\text{g m}^{-3}$)).

Considering Figure 3.13 further it can be seen that power station emissions are predicted to contribute a maximum of about $2.0\ \mu\text{g m}^{-3}$ to secondary sulfate/nitrate in the regional grid on this day, with maximum concentrations in Sydney of between 0.1 and $1\ \mu\text{g m}^{-3}$, which is a factor of $5\text{--}6$ lower than that predicted by the first-order reaction rate methodology. This supports the view that the secondary concentrations predicted using the first order approach are conservative. The use of smaller first-order conversion rates (rather than those based on the peak rates predicted with the Lagrangian Wall modelling) in the 12 month TAPM modelling may have resulted in a better level of agreement between the two approaches.

Of principal interest is the impact made by the power station emissions relative to the emissions from urban areas in the greater Sydney region. We can investigate the relative contribution of power station and urban sources to the ammonium sulfate/ammonium nitrate totals through the use of scatter plots. Shown in Figure 3.14 is a scatter plot of the modelled contribution of power station emissions to the ground-level 24 h concentrations of NO_3 and SO_4 and the summed concentrations of NH_4 , NO_3 and SO_4 .

Note that any points which lie along the 1:1 line correspond to regions in the modelling domain where power stations are the only source of particles. Points which lie along the horizontal axis correspond to regions in the airshed where non-power station sources (i.e. urban sources) are the only source of particles. Points which lie between the horizontal axis and the 1:1 line have contributions from both power stations and urban sources.

Considering the figures it can be seen that the model predicts peak 24 h NO_3 concentrations of up to $1 \mu\text{g m}^{-3}$, and that most of the points lie along the 1:1 line. This indicates that power station emissions are predicted to be the dominant contributor for the modelled meteorological conditions. This is consistent with the finding that the urban contribution is predicted to be minor for this event. Note that the selection of a day with minimum urban impact and maximum power station impact is an expected outcome of considering only power station sources in the first-order TAPM particle modelling.

Considering the SO_4 plot in Figure 3.14 it can be seen that the power stations are predicted to contribute the majority of the sulfate (up to $2.0 \mu\text{g m}^{-3}$). This appears to result primarily from the near-field conversion of H_2SO_4 to SO_4 as discussed in a previous section. Note that this near-field impact is smaller than the $3.5 \mu\text{g m}^{-3}$ which was estimated using the TAPM $\text{PM}_{2.5}$ modelling (Table 3.2), and likely results from the different plume rise algorithms used by the TAPM and CTM modelling systems.

The power station contributions to total ammonium sulfate/nitrate concentrations are shown in the right-hand plot of Figure 3.14. It can be seen that peak 24 h concentrations of up to $3 \mu\text{g m}^{-3}$ are predicted for these particle types. This maximum is predicted to occur outside of the Sydney region. Within the Sydney region, a maximum power station contribution of $\sim 1.0 \mu\text{g m}^{-3}$ is predicted. Power station emissions are predicted to be the major contributor to these concentrations (the urban contribution is predicted to occur below concentrations of about $2 \mu\text{g m}^{-3}$ for this event. Again, note that this concentration is a factor of 5–6 lower than that predicted using the first order reaction approach.

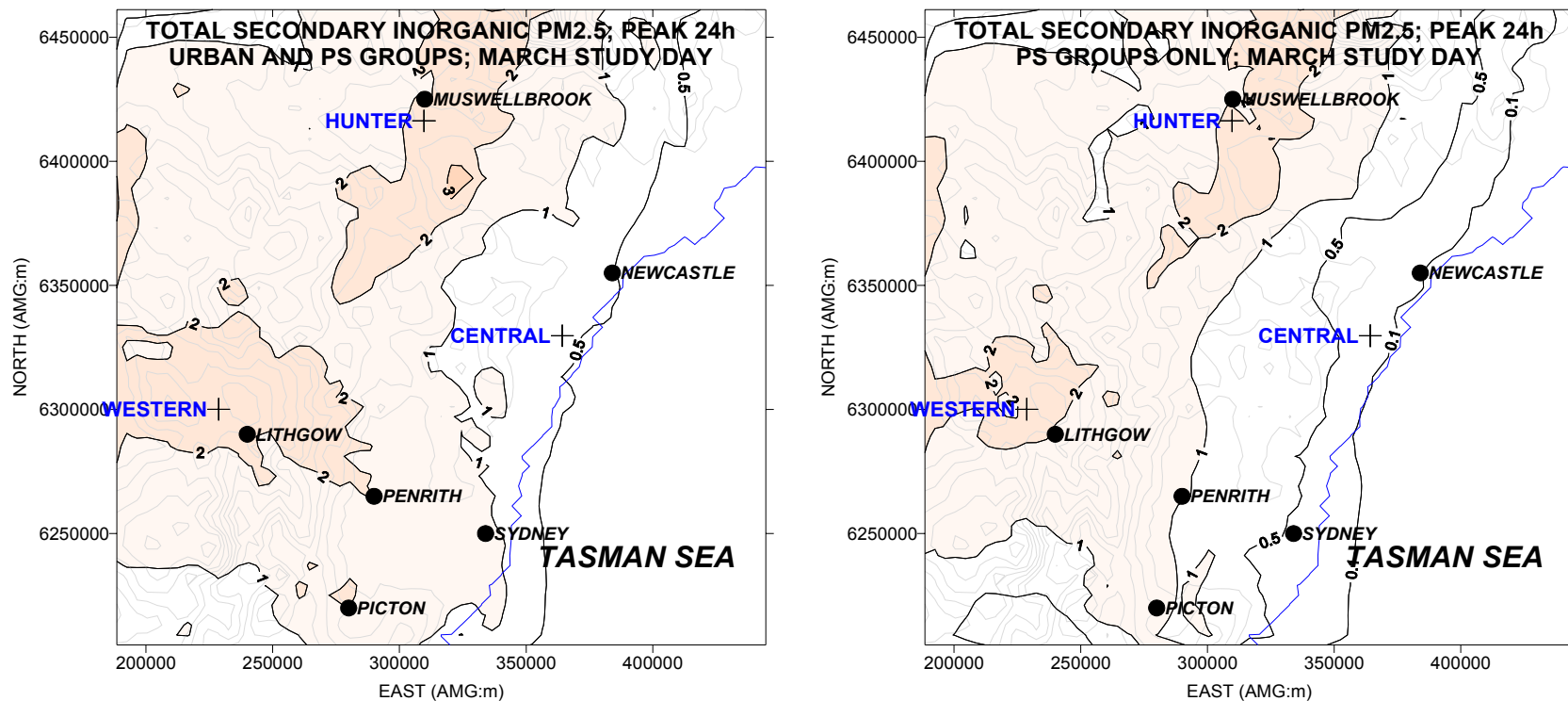


Figure 3.13: Peak combined 24 h sulfate + nitrate concentrations (as $[\text{NH}_4]_2\text{SO}_4$ and NH_4NO_3) for 11–12 March 2002. left- power station and urban sources; right- power stations only.

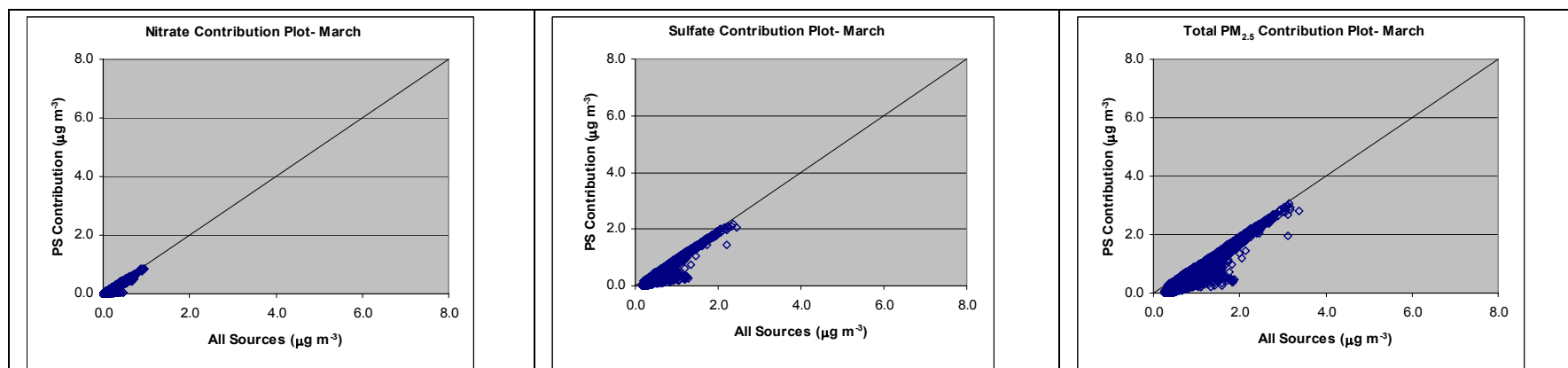


Figure 3.14: Scatter plots of the predicted contribution of power station emissions to total concentrations of NO_3^- , SO_4^{2-} and summed ammonium–nitrate/sulfate. The plots are of peak 24 h concentrations for every grid point in the modelling domain (surface level only) for 12–13/3/2002.

4 June 2002 In addition to the March event considered above, we used the annual TAPM runs to identify a worst-case winter day. TAPM-CTM was configured and exercised for this day (48 h simulation which included the previous day) in order to investigate how the secondary particle concentrations may vary between summer and winter meteorological conditions.

In considering the results, a significant outcome is that more secondary aerosol is predicted to be generated on this winter day in comparison to the March event described previously. In particular, the urban contribution is predicted to be significantly greater. This is a result of the lower levels of dispersion and stronger patterns of recirculation present during 3–4 June, which more than offsets the cooler temperatures and lower levels of radiation present during the period. Note that the range of observed $\text{PM}_{2.5}$ and PM_{10} concentrations for the Sydney region are $10\text{--}19\ \mu\text{g m}^{-3}$ and $12\text{--}20\ \mu\text{g m}^{-3}$ respectively.

Combined peak 24 h ammonium sulfate/nitrate aerosol concentrations are shown in Figure 3.15 for 4 June 2002. The predicted peak 24 h concentration for the combined urban-power station plume is $8\ \mu\text{g m}^{-3}$. This is more than double that predicted for the summer case. Power station emissions are predicted to generate up to $4\ \mu\text{g m}^{-3}$ of ammonium nitrate/sulfate, well away from the urban area. This is about a factor of four lower than the peak 24 h concentrations predicted using the first order rate TAPM simulations. Within the Sydney region, power stations are predicted to contribute $0.1\text{--}2\ \mu\text{g m}^{-3}$ of ammonium sulfate and nitrate.

As was done for the March event, the relative contribution of urban and power station sources to the ammonium sulfate/nitrate totals may be examined through the use of a scatter plot analysis (Figure 3.16). It can be seen that the model predicts peak 24 h NO_3^- concentrations of slightly less than $6\ \mu\text{g m}^{-3}$ for this June case. Power station emissions are predicted to contribute up to $2.0\ \mu\text{g m}^{-3}$ to NO_3^- totals in the range $0\text{--}2.0\ \mu\text{g m}^{-3}$, (occurring outside the urban areas) and up to $1\ \mu\text{g m}^{-3}$ to NO_3^- totals in the range $2.5\text{--}4.5\ \mu\text{g m}^{-3}$. Power stations are predicted to contribute less than $1\ \mu\text{g m}^{-3}$ to the peak urban concentrations of $5\text{--}6\ \mu\text{g m}^{-3}$.

Considering the SO_4^{2-} plots, it can be seen that the power stations are predicted to contribute comparable levels to the urban sources ($\sim 1.5\text{--}3\ \mu\text{g m}^{-3}$ each). In interpreting the sulfate plots, recall that the urban sulfate emissions may be underestimated for this winter event, because domestic fuel combustion has been omitted from the inventory.

The power station contribution to combined concentrations of ammonium sulfate/nitrate is shown in the right-hand plot of Figure 3.16, where it can be seen that 24 h ammonium sulfate/nitrate concentrations are predicted to peak at about $8\ \mu\text{g m}^{-3}$ (occurring well to the east of Sydney as shown in Figure 3.15). Power station plumes are predicted to contribute $< 1.0\ \mu\text{g m}^{-3}$ to this peak. However, the power station plumes are predicted to contribute $1\text{--}2\ \mu\text{g m}^{-3}$ to combined urban-power station aerosol peaks in the range $4\text{--}6\ \mu\text{g m}^{-3}$. Outside of the urban plume (i.e. along the 1:1 line), power station emissions are predicted to generate up to $4\ \mu\text{g m}^{-3}$ to the ammonium sulfate/nitrate aerosol mass.

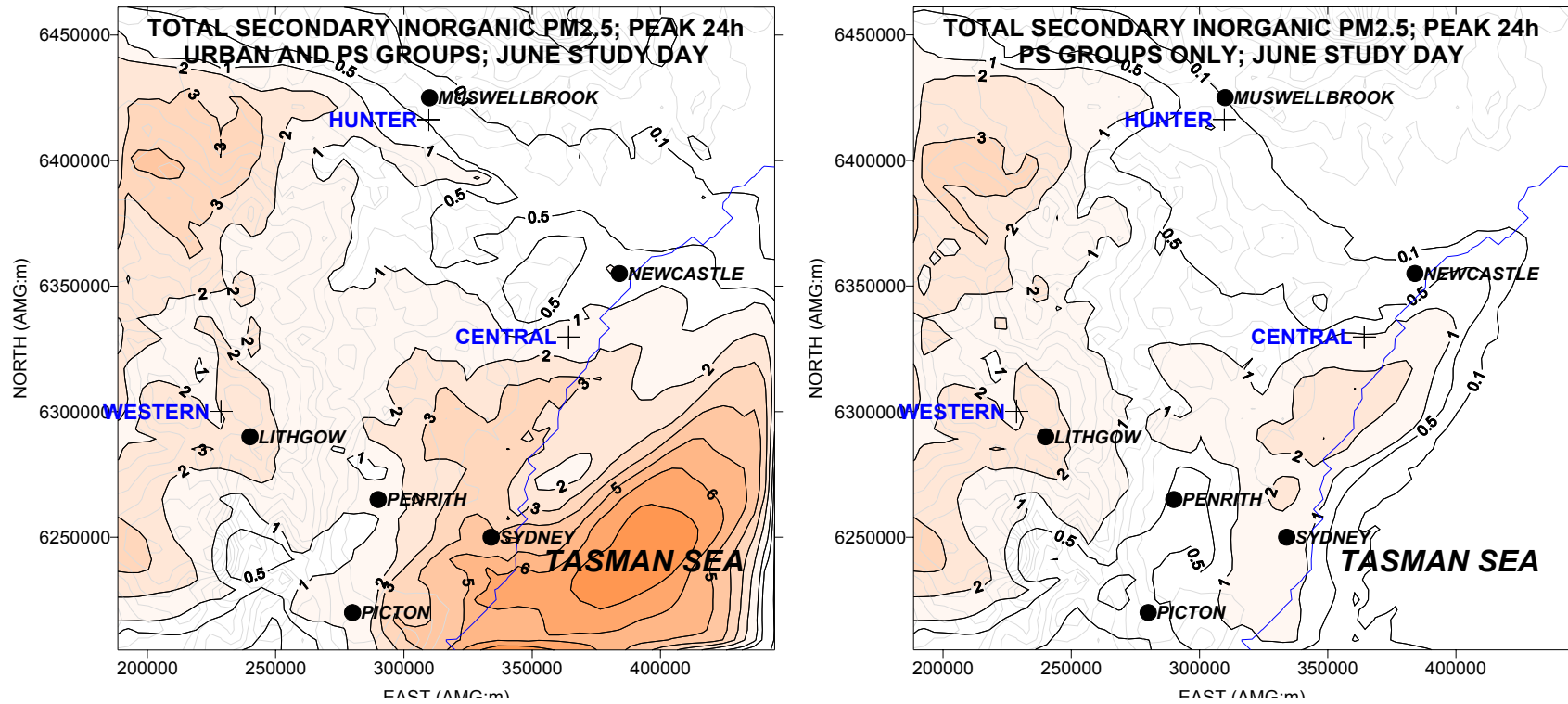


Figure 3.15: Peak combined 24 h sulfate + nitrate concentrations (as $[\text{NH}_4]_2\text{SO}_4$ and NH_4NO_3) for 3–4 June 2002. left- power station and urban sources; right- power stations only.

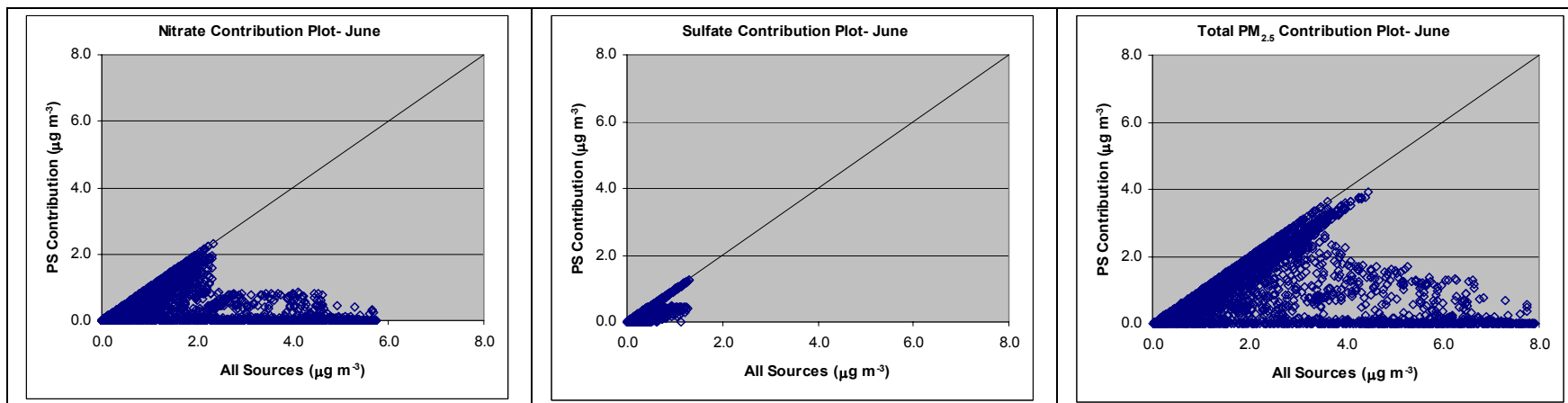


Figure 3.16: Scatter plots of the predicted contribution of power station emissions to total concentrations of NO_3^- , SO_4^{2-} and summed ammonium–nitrate/sulfate. The plots are of peak 24 h concentrations for every grid point in the modelling domain (surface level only) for 3–4/6/2002.

3.3 Synthesis of modelling results

The numerical modelling has considered three aspects of PM production from the power station groups.

1. Near-field and regional (including Sydney) concentrations of emitted primary particles mainly resulting from a small component of fly ash which has not been trapped by bag filters or electrostatic precipitators.

The outcomes of the primary particle modelling are summarised in Table 3.1 and indicate:

- Peak near-field $PM_{2.5}$ 24 h concentrations in the range $0.5 - 3 \mu g m^{-3}$ for the 3 power station groups and concentrations of $< 1 \mu g m^{-3}$ in the Sydney region.
- Peak 24 h PM_{10} concentrations are predicted in the range $1 - 5 \mu g m^{-3}$ in the near-field, and $< 1 \mu g m^{-3}$ in Sydney.
- Peak annual average $PM_{2.5}$ concentrations in the range $0.1 - 0.4 \mu g m^{-3}$ in the near-field and $< 0.1 \mu g m^{-3}$ in the Sydney region.
- Peak annual average concentrations of PM_{10} in the range $0.3 - 0.9 \mu g m^{-3}$ in the near-field and $< 0.1 \mu g m^{-3}$ in the Sydney region.

2. Near-field secondary sulfate concentrations resulting from the emission of SO_3 / H_2SO_4 .

Ammonium sulfate concentrations formed from the emission of sulfur trioxide / sulfuric acid are summarised in Table 3.2 and indicate:

- Predicted peak near-field, 24 hour ammonium sulfate concentrations in the range $1-7 \mu g m^{-3}$, suggesting that the emission and conversion of SO_3 has the potential to generate larger near-field $PM_{2.5}$ concentrations than the directly emitted fly ash.
- The combined impacts from the primary particle matter and the converted SO_3 leads to predicted near-field peak 24 hour $PM_{2.5}$ concentrations in the range $2 - 9 \mu g m^{-3}$, or up to 36% of the 24 hour $PM_{2.5}$ NEPM.
- The combined near-field annual average $PM_{2.5}$ concentrations in the range $0.4 - 2.0 \mu g m^{-3}$. The upper limit of this range is equivalent to 25% of the annual average $PM_{2.5}$ NEPM.

3. Gas-phase, secondary particle production (as ammonium nitrate and ammonium sulfate) from the oxidation of SO_2 and NO_x in power station plumes.

Two modelling approaches were used for this task:

12 month TAPM modelling with a conservative first-order reaction scheme.

This approach provided predictions for potential power station impacts in isolation from other sources. Results from this approach are summarised in Table 3.4 and indicate:

- Peak total (primary and secondary) 24 hour $\text{PM}_{2.5}$ concentrations of $6 - 17 \mu\text{g m}^{-3}$ and peak 24 hour PM_{10} concentrations of $6 - 18 \mu\text{g m}^{-3}$. The highest concentrations were predicted to occur within the Hunter Valley. Concentrations of $5 - 6 \mu\text{g m}^{-3}$ are predicted for the Sydney region
- Annual average $\text{PM}_{2.5}$ and PM_{10} concentrations in the range $1 - 2 \mu\text{g m}^{-3}$. The peak annual average concentrations are predicted to occur close to the power station sources. In Sydney, peak annual average concentrations are predicted to reach $0.2 - 0.3 \mu\text{g m}^{-3}$.

These first-order secondary particle concentration predictions are considered to be conservative and likely to be caused by the selection of maximum rather than average first-order conversion rates for SO_2 to SO_4 and NO_x to NO_3 .

Case-study modelling using TAPM-CTM and a thermodynamic steady-state aerosol mechanism.

To test the conservatism of the first-order methodology the predicted worst-case 24 hour period (plus a poor dispersion winter event) were re-modelled using TAPM-CTM and a comprehensive chemical transformation mechanism. This case-study modelling predicted that power station emissions contributed:

- $3 - 4 \mu\text{g m}^{-3}$ to peak 24 h ammonium sulfate + nitrate concentrations outside the urban area.
 - Up to about $2 \mu\text{g m}^{-3}$ to peak 24 h ammonium sulfate + nitrate concentrations within the Sydney region.
- Thus the comprehensive chemical modelling yielded concentration peaks which are about a factor of four lower than the peaks predicted by the first order reaction approach.

4 DISCUSSION

The issue of atmospheric particles (or aerosols) and coal-fired electricity generation is relevant, topical and important for several reasons:

- While the causal mechanism(s) for the observed relationships between PM and health effects have not been definitively established, there is strong support for the view that finer particles are more strongly associated with adverse health outcomes than coarser particles.
- Ambient air quality data from the urban and regional areas of NSW reveal frequent exceedences of the National Environment Protection Measure PM_{10} compliance standard and the more recent $\text{PM}_{2.5}$ advisory reporting standards. Bushfires and dust storms are known to contribute to elevated PM events.
- The NEPM process requires jurisdictions to develop air quality management plans in situations where NEPM goals / standards are not being achieved.
- A review of the Ambient Air NEPM is scheduled to commence in 2005.

- While there is a reasonable level of understanding of the emission sources of primary PM₁₀, understanding of the emission sources of primary PM_{2.5} and the formation of secondary PM in the atmosphere is poor. Secondary PM is likely to be well represented in the PM_{2.5} (fine) size fraction.
- Fine PM has the potential to remain suspended in the atmosphere for extended periods and therefore it is possible for these particles to be transported over large distances.
- Coal-fired power stations are significant emitters of both sulfur and nitrogen oxides into the Greater Metropolitan Region. National Pollutant Inventory figures indicate that electricity supply (which is dominated by the seven coal-fired power stations) contributed 87% and 48% of the sulfur oxides and nitrogen oxides respectively in NSW in 2003. The NPI further indicates that coal-fired power stations emit about 5% of the PM₁₀ in NSW.
- Coal-fired power stations have the potential to contribute to the formation of fine PM in the atmosphere from sulfur and nitrogen oxide gaseous emissions, as well as from the emission of primary PM.

The assessment undertaken in this project has addressed the potential contribution from coal-fired power stations to the atmospheric concentrations of PM. The assessment has been comprehensive, addressing:

- Both near-field and regional potential impacts, with consideration given to potential impacts in the Sydney urban region.
- Both short-term (24 hour average) and long term (annual average) concentrations.
- Both PM₁₀ and PM_{2.5}
- Primary emissions and the production of secondary aerosols in the atmosphere from the emission of sulfur and nitrogen oxides.

Given the uncertainties inevitably involved with a “pilot” study for which there are no ready established precedents / protocols to follow, the assessment has attempted to adopt a conservative approach, where possible. This involved adopting the following assumptions:

- All seven coal-fired power stations were operating continuously at maximum load throughout the year being modelled.
- All secondary nitrate occurs in the PM_{2.5} (fine) particle fraction.
- The selection of “worst-case” days for detailed assessment.
- The reporting of sulfates and nitrates in the fully neutralised form (ammonium sulfate and ammonium nitrate).
- Emitted sulfur trioxide / sulfuric acid is assumed to be fully neutralised by ammonia to ammonium sulfate in the near-field.

The report acknowledges that it has not been possible to model the formation of secondary aerosols resulting from aqueous phase (cloud) processes followed by the evaporation of cloud droplets, leaving behind aerosols. A review of wet and dry deposition studies in NSW undertaken by CSIRO would suggest that the exclusion of this pathway is unlikely to significantly alter the results obtained (Ayers et al 1995, 1997).

It is also possible that the use of TAPM has introduced a further degree of conservatism, at least in the near-field results, due to the model's tendency to over-predict both the mean and the infrequent, highest concentrations (Luhar et al 2004). TAPM's possible over-prediction was also demonstrated in an earlier local study (Carras et al 2002) in which TAPM's highest predicted concentrations in relation to emissions from coal-fired power stations on the Central Coast of NSW were significantly greater than observed concentrations from over ten years' of air quality monitoring in the region. Note that this result is, to some extent, dependent of the spacing of the computational grid used by TAPM. For example, a 500 metre horizontal grid spacing was adopted for the Carras et al (2002) study. In the case of the current study, a 1000 metre grid spacing was used. Thus any tendency to over predict may be offset, to some degree, by the coarser grid spacing.

With respect to the near-field modelling, predicted peak 24 hour average PM_{2.5} concentrations are 8, 13 and 36% of the NEPM advisory reporting standard of 25 $\mu\text{g m}^{-3}$ in the Western, Central Coast and Hunter Valley regions respectively. It was estimated that ammonium sulfate formed from the emission of sulfur trioxide could contribute up to about 70% of these peak near field results.

In relation to these elevated PM concentrations it is worth noting that frequency distributions of ground level concentrations resulting from emissions from tall stacks tend to be highly skewed, with most measurements being close to "baseline" and relatively few occurrences of elevated concentrations on an annual basis. Figure 4.1 shows a sulfur dioxide frequency distribution from the Wyee site, which was considered in Section 2. The figure clearly demonstrates the highly skewed nature of the data which is typical of distributions from tall-stack emissions. In addition to the likelihood that elevated ground level concentrations are infrequent, is the consideration that, as discussed above, TAPM may well be over-predicting these relatively few elevated occurrences.

The above considerations suggest that further field-based work may be beneficial in better understanding the occurrence of PM in the near-field of the power stations. This suggestion is developed further later in the discussion.

On an annual average basis, the near-field PM_{2.5} results, in all three regions, tend to be well within the NEPM reporting standard. This result is consistent with other air pollutants for which

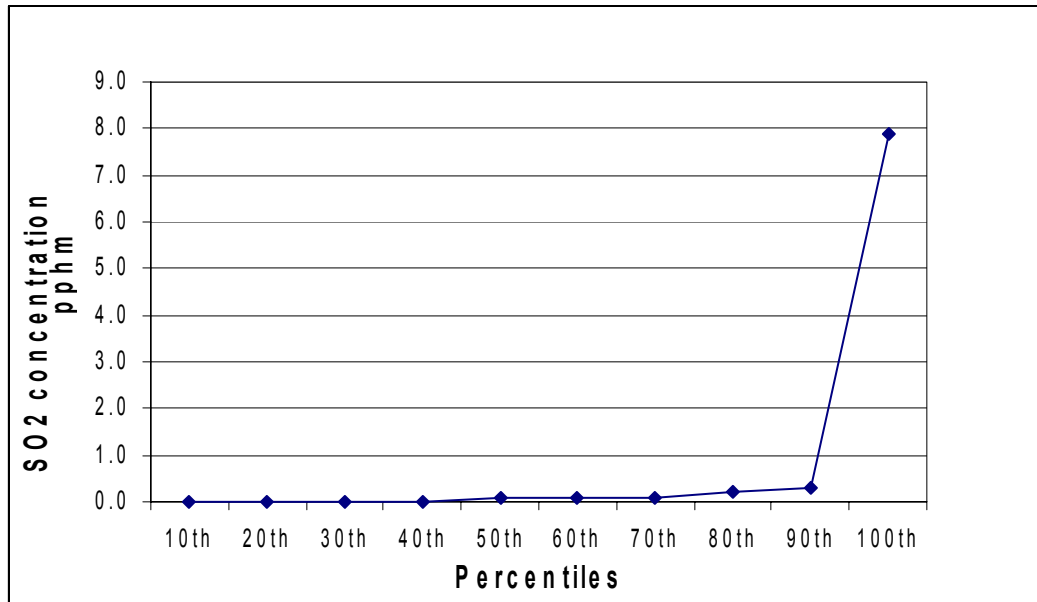


Figure 4.1: Frequency distribution of hourly average sulfur dioxide concentrations at Wyee.

short-term and long-term goals exists. In the case of SO₂, for example, it is not uncommon for the short-term goals to be approached or exceeded infrequently in the vicinity of strong point sources, while annual average concentrations are generally well below the annual goal. This observation is consistent with a highly skewed distribution in which a few observations are significantly elevated above a (low) mean value.

With respect to potential regional PM impacts, and particularly those in the large urban areas, days selected on the basis of possible “worst-case” power station impact in the Sydney region indicated a potential maximum power station contribution to PM_{2.5} 24 hour average concentrations of 2 µg m⁻³, less than 10% of the NEPM advisory reporting standard. This result suggests that power station contributions to urban PM₁₀ levels are likely to be small, (<10% of NEPM), a conclusion which is consistent with the earlier IRTAPS study (Malfroy et al 2003) which examined the contribution of power station NO_x emissions to urban smog (ozone and nitrogen dioxide). Further work would be required to quantify the potential incidence of the small power station contributions to urban PM concentrations.

Possible enhancements

The work undertaken for this project has identified a number of areas in which additional research may lead to improved characterisation of the contribution made by power station emissions to fine particle concentrations in the near-field and regionally. These options, some of which have been alluded to in the preceding discussion, are discussed briefly below:

- Undertake a well-designed monitoring program in the near-field of each power station region of PM₁₀ and PM_{2.5}. Examination of PM₁₀ and PM_{2.5} measurements in conjunction with other existing measurements, of SO₂ for example, might lead to better quantification of the contribution of power station emissions to particle concentrations in each region.

Consideration of the work by Hinkley in the Hunter Valley could assist in devising such a program.

- Near-field concentrations of sulfate resulting from the SO_3 emissions have been identified as a likely significant component of near-field fine particle concentrations from the power station groups. It is considered that there would be benefit in confirming the percentage of sulfur emitted as $\text{SO}_3/\text{H}_2\text{SO}_4$ by each power station. This could be determined from existing coal sulfur measurements, continuous measurements of SO_2 and periodic testing of $\text{SO}_3/\text{H}_2\text{SO}_4$, which are now routine at most power stations.
- Confirm the percentage of primary particles emitted as $\text{PM}_{2.5}$ and PM_{10} . The current project relied on data presented in the USEPA's AP-42 emission factor document. More representative local data may be available for some power stations, but it should be recognised that high quality size fractionated data are neither easy nor inexpensive to obtain.
- Ammonia is a key compound for neutralising sulfate and for transferring nitric acid from the gas phase to the aerosol phase. Ammonia measurements could be undertaken by inexpensive "passive" techniques and would contribute to the more accurate representation of aerosol chemistry in the TAPM-CTM model. (It is our understanding that the Department of Environment and Conservation is developing an ammonia emissions inventory for the greater Sydney region in a form suitable for input into air quality models).
- Undertake TAPM-CTM modelling on a further limited number of days to ascertain the power station contribution on days in which PM levels were elevated in the major urban area (avoiding days unduly influenced by bushfire smoke and or dust storms). This would complement the results for the two days modelled in the current study which were selected on the basis that they were "worst-case" in terms of power station contribution. This task could be most usefully undertaken when the DEC upgrade to the 1992 air emissions inventory is complete.
- Consideration could be given to extending the TAPM-CTM modelling approach to include aqueous phase sulfate production. This task would build on the work already undertaken by Ayers and Granek (1997) and by Hurley (2002).
- Undertake further consideration of techniques which might enable a power station "finger print" to be detected in a collected sample of PM.
- While an initial analysis of first-order reaction rate results indicates that small power station impacts on urban fine particle concentrations are likely to be infrequent, it is considered that this result might usefully be re-visited upon completion of the upgrade to the air emissions inventory for the region.
- Further work in assessing potential health impacts and associated costs due to population exposure to predicted particle concentrations in the region arising from the full range of emission sources would enable the relative impacts from power station emissions to be more fully considered.

5 CONCLUSIONS

- 1 While the causal mechanism(s) responsible for the relationships between atmospheric particles and health effects observed in epidemiological studies have not been definitively established, there is strong support for the view that finer particles are more strongly associated with adverse health outcomes than coarser particles.
- 2 The association between mortality and other health effects and *particle mass* show relatively consistent magnitudes of effects. There is considerably less agreement and consistency in the results from studies which have examined associations between health impacts and the *components* of the fine particles.
- 3 The frequency distributions of ground-level concentrations associated with power station emissions suggests that **the near-field occurrence** of elevated particle concentrations is likely to be infrequent.
- 4 Predicted **peak 24 hour average PM_{2.5}** concentrations were 8, 13 and 36% of the National Environment Protection Measure (NEPM) Advisory Reporting Standard of 25 $\mu\text{g m}^{-3}$ in the Western, Central Coast and Hunter Valley regions, respectively. It was estimated that ammonium sulfate formed from the emission of sulfur trioxide could contribute up to about 70% of these peak near-field results.
- 5 Predicted **peak 24 hour average PM₁₀** concentrations were 5, 12 and 22% of the NEPM standard of 50 $\mu\text{g m}^{-3}$ in the Western, Central Coast and Hunter Valley regions, respectively. It was estimate that ammonium sulfate formed from the emission of sulfur trioxide could contribute up to about 60% of these peak near-field results.
- 6 Predicted **annual average PM_{2.5}** concentrations in the three generating regions were in the range 0.4 – 2.0 $\mu\text{g m}^{-3}$. The upper limit of this range is equivalent to 25% of the relevant National Environment Protection Measure Advisory Reporting Standard.
- 7 In areas removed from urban emissions, power station emissions were predicted to contribute up to 3 – 4 $\mu\text{g m}^{-3}$ to peak 24 h ammonium sulfate + nitrate concentrations.
- 8 The results from the modelling of “worst-case” days suggest that **the power station contribution to urban fine particle concentrations is small**. An analysis of one year’s results indicates that these small **power station contributions to urban fine particle concentrations are likely to be infrequent**.
- 9 The “worst-case” contribution of power station emissions to **24 hour average PM_{2.5} in the Sydney urban area** was estimated to be 2 $\mu\text{g m}^{-3}$ – **8% of the relevant NEPM Advisory Reporting Standard**.
- 10 The predicted contribution of power station emissions to **annual average concentrations of PM_{2.5} in Sydney** was 0.3 $\mu\text{g m}^{-3}$ – about **4% of the relevant NEPM Advisory Reporting Standard**.

The project has met its overall aim to provide robust, credible information on the potential atmospheric concentrations of fine particles from power station operations. A number of specific

issues have been identified during the course of the project for which additional research may lead to improved characterisation of the contribution made by power station emissions to fine particle concentrations in the near-field and regionally.

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Appendix 1. CSIRO Report ET/IR 735: NSW Electricity Generators' Fine Particle Study – Modelling Component.

This report is provided as a separate volume to the main report.