AN ASSESSMENT OF THE CONTRIBUTION OF COAL-FIRED POWER STATION EMISSIONS TO ATMOSPHERIC PARTICLE CONCENTRATIONS IN NEW SOUTH WALES

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Summary

In this study, state-of-the-art air quality assessment and modelling techniques were used to gain an understanding of the significance of emissions from coal-fired power stations located in New South Wales, Australia, to the occurrence of atmospheric particles (PM_{10} and $PM_{2.5}$) in the near-field of the power stations and regionally, including the Sydney urban area.

Using a conservative first-order reaction methodology combined with a comprehensive chemical transformation methodology the project has demonstrated that the contribution of NSW coal-fired power station emissions to urban fine particle concentrations is likely to be small and infrequent.

The study was funded by the state-owned electricity generating organisations, Delta Electricity, Eraring Energy and Macquarie Generation.

Keywords: Fine particles, power station emissions, inter regional transport, airshed modelling.

1 Introduction & Background

The Harvard Six Cities Study (Dockery 1993), which commenced in 1974, arguably laid the foundations for subsequent international research into the occurrence of atmospheric particles and health impacts. As a result of the Six Cities Study and the numerous studies which followed and continue today (see HEI 2002), the relationship between 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 Variation in 2003 (NEPC 2003) 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.

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 seven coal-fired power stations operating in three regions of NSW, or which form in the atmosphere from emissions from these facilities. The location of the power station regions in relation to the major urban areas of Newcastle, Sydney and Wollongong is shown in Figure 1.

Particulate collection systems on modern coalfired power stations are very effective in removing particles from the exhaust gases – however some technologies are relatively less effective in removing finer particles compared with coarse particles. Furthermore, unless flue gas scrubbing is employed the emission of sulfur and nitrogen oxides (NO_x) potentially leads to the formation of secondary particles in the atmosphere.

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.

The study was undertaken by a team comprising Malfroy Environmental Strategies, the CSIRO and Macquarie University.



Figure 1: The location of the power station regions in the Central Coast, the Hunter Valley and Western Coalfield Regions, in relation to the urban areas.

The focus of this paper is to present and discuss the results of the modelling assessment, which was undertaken by the CSIRO Divisions of Atmospheric Research and Energy Technology (Cope et al 2004).

2 Modelling of Particle Concentrations

Numerical modelling was used to estimate the ground level concentrations of $PM_{2.5}$ and PM_{10} resulting from the direct emission of particles from power station stacks (primary particle modelling), and from the in-plume conversion of gaseous species to ambient particles as the result of photochemical transformation processes (secondary particle modelling).

The modelling was undertaken for power stations located on the Central Coast, in the Hunter Valley and to the west of Sydney (See Figure 1).

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

- characterising emissions,
- understanding atmospheric formation and removal processes and
- understanding the relative impacts from a large number of very different sources.

Accordingly, in each phase of the modelling, it was attempted to take a conservative approach in order to assess the potential maximum possible impacts from power station emissions on particle concentrations.

3.1 Primary particle modelling

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 hour and annual average $PM_{2.5}$ and PM_{10} concentrations were estimated.

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. Initial and boundary conditions for the meteorological fields were provided by a large scale analysis, generated by the Bureau of Meteorology. 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).

The simulation of $PM_{2.5}$ and PM_{10} primary emissions was 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.

3.1.1 TAPM Domains

The TAPM simulations were 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. The regional grid was designed to capture the combined impact of all the power station sources, particularly within the Sydney metropolitan region.

3.1.2 Source and Run-Time Definitions

The prescribed emissions of primary particles from the power stations were estimated using individual power station characteristics: Generating capacity in megawatts (MW), conversion efficiency (% on a generated basis), average specific energy of the coal burnt (MJ/kg), average ash of the coal burnt (%), the fly ash fraction of the ash generated and the gross collection efficiency of the particulate collection device. USEPA emission factors were used to divide the particle emission into PM₁₀ and PM_{2.5} size fractions (USEPA 2003).

Relevant source characteristics such as stack height, efflux temperature and velocity for each power station were also obtained.

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

representative of 2002. Power stations were assumed to be running at continuous maximum load throughout the modelled year.

3.2 Secondary particle modelling

There are three major pathways for secondary particle production from power station emissions;

- Near-field conversion of sulfur trioxide (SO₃) / sulfuric acid (H₂SO₄) emissions to sulfate
- Gas–phase conversion of SO₂, and NO_x to sulfate and nitrate particles,
- Aqueous phase conversion of SO₂, and NO_x to sulfates and nitrates with subsequent evaporation resulting in sulfate and nitrate particles.

The complexities of modelling aqueous–phase secondary aerosol production precluded it from inclusion in the current study. Under some situations this could result in an underestimation of secondary particle production.

In the case of the near-field and regional modelling presented in the next sections, the mass of secondary particle matter was 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 $[(NH_4)_2SO_4]$, and as ammonium nitrate (NH_4NO_3) .

It was further assumed that all secondary particles reside in the $PM_{2.5}$ size fraction – an assumption which is well founded for $(NH_4)_2SO_4$ particles which are known to be in the order of 1 μ m in diameter, but conservative for NH_4NO_3 , a significant fraction of which may be larger than $PM_{2.5}$.

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.

4.2.1 First–order chemical transformation modelling methodology

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.

Conversion rates were developed through consideration of the literature (i.e. Hewitt 2001) and through a numerical modelling study in which the dispersion and chemical transformation of power station NO_x and SO_2 within the Sydney region was simulated using a high resolution Lagrangian wall model (Lilley and Cope 2005) 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 under 34 scenarios.

The first-order methodology considered only power station emissions, and further the rates developed were 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 these considerations, it is likely that the secondary particle results generated by the first-order methodology are conservative.

TAPM was run for one year (2002) using the first-order chemical transformation methodology and the results examined to identify worst-case days for more detailed modelling using the Comprehensive Chemical Transformation Approach.

4.2.2 Comprehensive Chemical Transformation Approach

A pre-release version of TAPM-Chemical Transport Model (CTM) was 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 Metropolitan Air Quality Study (MAQS) 1992 emissions inventory (Carnovale 1996), 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 sulfur oxidation (SO₂ to SO₃) 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 SO₂). As a consequence, the approach used in the Australian Air Quality Forecasting emissions inventory¹ was adopted which assigns 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 discussed below. Primary particle emissions were not included in the TAPM-CTM simulations.

¹ (<u>http://www.dar.csiro.au/publications/manins_2002axviii.pdf</u>)

The model was run using a uniform background ammonia concentration. This background was used as a surrogate for the ammonia sources within the region as 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.

The power station gaseous emissions were estimated as follows:

- SO₃ / H₂SO₄ emissions were estimated according to the National Pollutant Inventory (NPI) methodology, based on the sulfur content of coal burnt.
- SO₂ emissions were conservatively estimated assuming 100% emission of coal sulfur as SO₂ at full load operation.
- NO_x emissions were estimated from previously developed NO_x concentration, load relationships developed for each power station (Nelson et al 2002). It was assumed that the NO_x emitted was 10% dioxide and 90% nitric oxide, on a molar basis.

3 Results

This section provides an overview of some of the more significant results generated from the modelling of primary and secondary particles in the near-field of the power stations (within several kilometres) and regionally, which includes the Sydney urban area.

4.1 Primary particle results

Primary particle results for all three regions are summarised in Table 1, along with maximum predictions for the Sydney urban region. In each region the highest 24 h average concentrations are predicted to occur within 10 km of the power stations. 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 suggests that the annual average concentrations are dominated by near-source convective fumigation events, and that the spatial scales for plume touch-down are smaller that the computational grid size (i.e. less than 2 km).

4.2 Secondary particle results

Results are presented for the production of nearfield sulfate from emissions of SO_3 /H2SO₄ and for the regional production of sulfates and nitrates from SO_2 and NOx emissions.

4.2.1 From emissions of SO₃ /H₂SO₄

Predictions for near-field $(NH_4)_2SO_4$ concentrations are shown in Table 2. The concentrations are similar to, or greater than, the PM_{2.5} concentrations arising from primary particle emissions (Table1), with the Hunter Valley group of stations resulting in the largest predicted concentrations.

Table 2: Peak ground-level secondary $PM_{2.5}$ concentrations (24h and annual average) in the near-field of each power station region for emissions of SO_3 /H₂SO₄ from the 3 power station groups.

	Peak 24h average(µg m ⁻³)	Annual average (µg m ⁻³)
Central Coast	0.9	0.2
Hunter Valley	6.5	1.3
Western	1.4	0.3

4.2.2 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 due to the Sydney urban plume.

Table 1: Peak ground-level $PM_{2.5}$ and PM_{10} concentrations (24h and annual average) in the near-field of each power station region and in Sydney for primary particle emissions from the 3 power station groups.

	Peak 24h average		Annual average	
	PrimaryPM2.5 (µg m ⁻³)	PrimaryPM10 (µg m ⁻³)	PrimaryPM2.5 (μg m ⁻³)	PrimaryPM10 (μg m ⁻³)
Central Coast	2.3	5.3	0.4	0.9
Hunter Valley	2.6	4.5	0.5	0.9
Western	0.5	1.2	0.1	0.3
Sydney maxima	< 1.0	< 1.	< 0.1	< 0.1

<u>**12**</u> <u>**March 2002.**</u> TAPM-CTM concentration predictions for the combined ammonium sulfate and ammonium nitrate concentrations for 12 March 2002 are shown in Figure 2 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 μ g m⁻³. A combined peak concentration of 1 – 2 μ g m⁻³ is predicted for the Sydney region.

The meteorological conditions predicted for this day lead to the highest 24 h secondary particle concentrations for the modelled year (2002). These results may be contrasted with the observed concentration range in the NSW Department of Environment Conservation's monitoring network of $10 - 14 \ \mu g \ m^3$ (PM_{2.5}) and $17 - 29 \ \mu g \ m^3$ (PM₁₀) for this day. The observed concentrations obviously include particles other than sulfate and nitrate, which were the only particles modelled.

Considering Figure 2 further it can be seen that power station emissions are predicted to contribute a maximum of about 2.0 μ 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 μ g m⁻³ (which is a factor of 5–6 lower than that predicted by the first-order reaction rate methodology).

<u>**4**</u> June 2002 In addition to the March event considered above, a worst-case winter day was identified from the first-order results.

In considering the results, shown in Figure 3, 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 PM_{2.5} and PM₁₀ concentrations for the Sydney region are 10 -19 μ g m⁻³ and 12 – 20 μ g m⁻³ respectively. Combined peak 24 h ammonium sulfate/nitrate aerosol concentrations shown in Figure 3 for the combined urban-power station plume (left plot) is 8 μ g m⁻³. This is more than double that predicted for the summer case. Power station emissions are predicted to generate up to 4 μ g m⁻³ of ammonium nitrate/sulfate, well away from the urban area (right side). 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 0.1–2 μ g m⁻³ of ammonium sulfate and nitrate.

5 Discussion

Given the uncertainties inevitably involved with a "pilot" study for which there are no established precedents / protocols to follow, the assessment of the potential contribution of power station emissions to atmospheric particle concentrations 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



Figure 2:: Peak combined 24 h sulfate + nitrate concentrations (as $[NH_4]_2SO_4$ and NH_4NO_3) for 12th March 2002. **left**- power station and urban sources; **right**- power stations only.



Figure 3: Peak combined 24 h sulfate + nitrate concentrations (as $[NH_4]_2SO_4$ and NH_4NO_3) for 4th June 2002. **left**- power station and urban sources; **right**- power stations only

- The reporting of sulfates and nitrates in the fully neutralised form (ammonium sulfate and ammonium nitrate)
- Emitted sulfur trioxide / sulfuric acid was assumed to be fully neutralised by ammonia to ammonium sulfate in the near-field.

As noted in section 3.2, it was not possible to model the formation of secondary aerosols resulting from aqueous phase (cloud) processes. A review of wet and dry deposition studies in NSW undertaken by CSIRO (Ayers et al 1995, 1997) would suggest that the exclusion of this pathway is unlikely to significantly alter the results obtained.

With respect to the near-field modelling, predicted peak 24 hour average $PM_{2.5}$ concentrations were 8, 13 and 36% of the ANEPM advisory reporting standard of 25 µg 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 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 very few occurrences of elevated concentrations on an annual basis. Annual average PM_{2.5} results in the near-field of all three regions tend to be well within the ANEPM reporting standard. This observation is again consistent with a highly skewed distribution in which a few observations are significantly elevated above a (low) mean value.

At the regional scale, 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 μ g m⁻³, less than 10% of the ANEPM advisory reporting standard.

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 analysed. The frequency distribution of these 24 hour averages shows a rapid drop from maximum values, with the 90th percentile and median percentile concentrations being 40% and 15% of the maximum concentration respectively, indicating that the predicted highest power station contributions are likely to occur infrequently.

In summary, the results from the conservative first-order methodology combined with the comprehensive CTM methodology results suggest that power station contributions to urban particle levels are likely to be small and infrequent, a conclusion which is consistent with the findings of the earlier IRTAPS study (Nelson et al 2002) which examined the contribution of power station NO_x emissions to urban smog (ozone and nitrogen dioxide).

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, should the need arise:

- A well-designed ambient monitoring program in the near-field of each power station region.
- Confirmation of the percentage of sulfur emitted as SO₃/H₂SO₄ by each power station.

- Confirmation of the percentage of primary particles emitted as PM_{2.5} and PM₁₀.
- Regional ammonia measurements would contribute to the more accurate representation of aerosol chemistry
- TAPM-CTM modelling on a number of days on which particle levels were elevated in the major urban area.
- Consideration could be given to including aqueous phase sulfate production in TAPM-CTM.
- Further consideration of techniques which might enable a power station "finger print" to be detected in a collected sample of particle matter.

6 Acknowledgements

The authors thank the project sponsors, Delta Electricity, Eraring Energy and Macquarie Generation for their permission to publish the results from the project in this paper.

We acknowledge the Department of Environment and Conservation for permission to use the MAQS inventory in the TAPM-CTM modelling and for providing PM_{10} and $PM_{2.5}$ data from the monitoring network.

Finally, we thank our CSIRO colleagues Bill Lilley, Mary Edwards, Peter Hurley, Merched Azzi and John Carras for their contributions to the project

7 References

Ayers G.P., Gillett R. W., Selleck P. W., Marshall J. C., 1995. *COMRAIN Study*. Final Report to Pacific Power from the Environmental Consulting Research Unit Division of Atmospheric Research CSIRO

Ayers G.P., Gillett R. W., Selleck P. W., Powell J. C., 1997. *Central Coast and Hunter Valley Deposition Study*. Final Report to Macquarie Generation, Delta Electricity and Pacific Power from the Environmental Consulting Research Unit Division of Atmospheric Research CSIRO

Ayers G. and Granek H., 1997. *Deposition Modelling in the Hunter Valley/Central Coast Region of NSW*. Final Report to Macquarie Generation, Delta Electricity and Pacific Power by the Environmental Consulting and Research Unit, CSIRO Atmospheric Research, 42p

Carnovale, F., Tilly, K., Stuart, A., Carvalho, C., Summers, M. & Eriksen, P., 1996, '*Metropolitan air quality study: Air emissions inventory*', Final report to NSW EPA

Cope M, Lilley B, Azzi M, Edwards M, Hurley P, Carras J (2004) *NSW Electricity Generators Fine*

Particle Study – Modelling Component. CSIRO Investigation Report ET/IR 735.

Dockery D, Pope CA, Xu X et al (1993) *An* association between air pollution and mortality in six US cities. N. Engl. J. Med. 329, 1753-59.

HEI 2002 Understanding the health effects of components of the particulate matter mix: progress and next steps. HEI Perspectives, April 2002, Health Effects Institute, Boston, 20 pp.

Hewitt C.N. (2001). *The atmospheric chemistry of sulphur and nitrogen in power station plumes.* Atmospheric Environment 35, 1155-1170.

Hurley P. 2002, *The Air Pollution Model* (*TAPM*) *Version 2. Part 1: Technical Description*. CSIRO Atmospheric Research Technical Paper No. 55. See www.dar.csiro.au/TAPM.

Lilley W., and Cope M., 2005. Development of a high resolution chemically reactive near-field dispersion model for assessing the impact of motor vehicle emissions. Part 1: model description and validation. Proceedings of 17th International Clean Air Conference, Hobart Australia.

Lurmann, F.W., Carter, W.P. and Coyner, L.A. 1987, 'A surrogate species chemical reaction mechanism for urban scale air quality simulation models'. Final Report to U.S. Environment Protection Agency under Contract Number 68-02-4104.

Manins P., Ayers G., Bohm M., Raupach M., Williams D., Carras J., 1996. *Hunter Valley dry deposition study*. A report to Pacific Power CSIRO Ref: SB/1/217. 175p

Nelson P, Azzi M, Cope M, Lilley W, Carras J.,, Hurley P., Hyde R. 2002, 'Inter-Regional Transport Of Air Pollutants Study (IRTAPS).' CSIRO Energy Technology Investigation Report Et/IR 570R Prepared for Pacific Power International on behalf of: Eraring Energy, Delta Electricity & Macquarie Generation

www.dar.csiro.au/publications/cope_2003a.pdf NEPC (1998) Ambient Air Quality National Environment Protection Measure (NEPM). National Environment Protection Council, Available at http://www.ephc.gov.au /nepms/air/air nepm.html

NEPC 2003 Variation to the Ambient Air Quality National Environment Protection Measure (NEPM). National Environment Protection Council, Available at

www.ephc.gov.au/nepms/air/air_variation.html

Saxena P., Hudischewskyj B., Seigneur C. and Seinfeld J., 1986. A comparative study of equilibrium approaches to the chemical characterisation of secondary aerosols. Atmospheric Environment, 20(7): 1471–1483.

United States Environmental Protection Agency (2003) Compilation of Air Pollutant Emission Factors AP-42, Fifth edition