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Title:

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Date:

2022-04-01

Citation:

Garg, M., Silver, J. D., Schofield, R. & Ryan, R. G. (2022). Hourly emission inventories for air toxic emissions for eastern Australian electricity generators derived from energy distribution data. *International Journal of Environmental Science and Technology*, 19 (4), pp.2973-2992. <https://doi.org/10.1007/s13762-021-03429-5>.

Persistent Link:

<https://hdl.handle.net/11343/276269>

Hourly emission inventories for air toxic emissions for eastern Australian electricity generators derived from energy distribution data

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Published date: 1 June 2021

Abstract Emission inventories are a fundamental input for atmospheric chemical transport models in order to accurately simulate air pollution and its impacts. In Australia, emission inventories are currently not available for electricity generators at high temporal and spatial resolutions. In this study, a new high spatial and temporal resolution inventory for Australian power plant emissions has been developed. The emissions from coal and gas-fired power plants have been estimated using 5-minute electricity generation time-series data and emission factors. The emissions are estimated from 2010 to 2019 for sulfur dioxide (SO₂), nitrogen oxide (NO₂), particulate matter (PM) and mercury (Hg). We estimate that, for 2019, the coal-fired power plants in Australia emitted significant amounts of SO₂ (485.7 kTonne) and NO₂ (452 kTonne), as there are no sulfur dioxide or nitrogen oxide controls in place. In contrast, PM emissions are mitigated by particulate matter control devices and coal-fired power plants emit an estimated 27.48 and 13.46 kTonne of PM₁₀ and PM_{2.5}, respectively. Coal-fired power plants were responsible for an estimated 3.13 tonnes of Hg emissions. Gas-fired plant emissions are significantly lower than coal-fired power emissions for all pollutants. Temporal analysis showed that coal and gas-fired power plant emissions vary quite markedly during the day and over the week. There is a strong diurnal cycle in the gas-fired emissions, which if ignored by assuming constant emissions (such as is routinely done), will lead to significant errors in modelled air pollution.

Keywords Australian emissions, coal and gas-fired powered plants, high resolution data, air pollution

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1 Introduction

Air pollutants emitted from anthropogenic sources such as power plants, industries and vehicles have health (Jerrett et al., 2009; Beelen et al., 2014; Lelieveld et al., 2015; Xu et al., 2020), visibility and climate change impacts (Forster et al., 2007; Myhre et al., 2013; Von Schneidemesser et al., 2015). According to a recent global emission inventory (Crippa et al., 2018), the power generation sector was responsible for the highest proportion of anthropogenic air pollution in Australia, followed by industry, shipping, on-road transport vehicles and domestic solid fuel burning. Coal and gas-fired power plants have been estimated to collectively contribute 36% of NO₂ and 46% of SO₂ anthropogenic emissions in Australia (Crippa et al., 2018).

Australia is highly urbanized with greater than 85% of the population living in major cities (World Bank Organisation, 2020) which are in many cases geographically separate from major power generation hubs. However, emissions from power plants can be transported over long distances by winds. These emissions are subject to chemical reactions and conversion to the particulate phase resulting in secondary aerosols. Thus, while many Australian power stations are located hundreds of kilometres away from the nearest cities, their emissions can still have a significant impact on urban air quality and, hence, human and environmental health.

Studies have been conducted to investigate the impact of emissions from power plants on urban air quality in Australia. Cohen et al. (2012) analyzed PM_{2.5} sources for the greater Sydney metropolitan area using 11 years of mass spectrometry data. They showed that 26% of the PM_{2.5} mass fraction was ammonium sulfate produced from atmospheric oxidation of SO₂ to sulfate aerosol and using back-trajectory analyses showed that coal-fired power plants in New South Wales (NSW) were responsible for 14-18% of PM_{2.5} in Sydney. This study attributed between a third and a half of the sulfate component of PM_{2.5} to emissions from coal-fired power plants. Similarly, Victorian fossil-fueled power plants have been shown to affect the air quality both nearby and over significant distances. Aerosol speciation measurements made by ion beam analysis by Crawford et al. (2017) at Cape Grim Baseline Air Pollution Station, Tasmania, coupled with back-trajectories, showed evidence of long-range aerosol transport from emission sources on the Australian mainland, primarily the Victorian power stations in the Latrobe Valley. Ewald (2018) estimated the health impacts (104 premature deaths and 560 years of life lost annually) due to emissions from coal-fired power stations in NSW, using the Broome et al. (2015) dose-response relationships. The total annual cost due to mortality and disability adjusted life years was estimated to be \$539 million AUD per annum.

Electricity in Australia is produced by both non-renewable (coal and gas) and renewable sources (wind, solar, hydro). Coal and gas-fired power plants generate 62% and 21% of electricity respectively (AEMO, 2018). Electricity in Australia is distributed by network operators. The National Electricity Market (NEM) and Western Australia's South-West Interconnected System (SWIS) are the two largest electricity markets in Australia covering 86% and 8% of the demand respectively. The SWIS covers Western Australia while the NEM connects the eastern part of the country covering the states of Queensland, New South Wales, Victoria, South Australia, Tasmania and the Australian Capital Territory. This study fo-

79 cuses on the NEM, whose database provides electricity generation figures from
80 non-renewable (coal and gas) and renewable (solar, wind, hydro and biomass)
81 sources at a temporal resolution of 5 minutes. The NEM covers almost 93% and
82 55% of coal and gas electricity production in Australia respectively. Across the
83 NEM, brown and black coal contribute 24.8% and 52.4% of electricity generation
84 (AEMO, 2018). Brown coal is used in Victoria (it was also used in South Aus-
85 tralia), and black coal is used in New South Wales and Queensland, reflecting the
86 geographical distribution of these two coal types.

87 The impact of air pollutants on air quality and climate can be investigated us-
88 ing three-dimensional atmospheric chemical transport models which simulate the
89 fate of pollutants using meteorological data. Such models require emission invento-
90 ries, which are ideally accurate and at high spatiotemporal resolution, to simulate
91 air pollution and its impacts (Matthias et al., 2018; Vedrenne et al., 2016). Much
92 work has been done to build and improve regional emissions inventories for coun-
93 tries in the northern hemisphere by organisations such as the US Environmental
94 Protection Agency, the European Monitoring and Evaluation Programme, Envi-
95 ronment Canada, the Acid Deposition Monitoring Network in East Asia for China,
96 India and other Asian countries (Janssens-Maenhout et al., 2015; Crippa et al.,
97 2018; Lamarque et al., 2010). For other regions, including Australia, nationally
98 consistent gridded inventories are only available through global products, such
99 as the Emissions Database for Global Atmospheric Research (EDGAR) database
100 (Crippa et al., 2018).

101 Currently, for Australia, emissions from power plants are estimated both na-
102 tionally and regionally. The National Pollution Inventory (NPI) covers emissions
103 from industrial sources in Australia. Industrial emitters are obliged to report emis-
104 sions annually to the NPI which has developed a framework to estimate emissions
105 from Australian industries based on fuel consumption and technology used. The
106 industrial facilities report only the total emissions on a financial year (July to
107 June) basis. The best available regional air emission inventory is the NSW GMR
108 (New South Wales Greater Metropolitan Region) Air Emission Inventory for the
109 years 2003, 2008 and 2013, which also includes the regional emissions from cities
110 of Newcastle and Wollongong (EPA, 2012; DECC, 2019). All these inventories
111 provide annual estimates.

112 Many studies have adopted the National Pollution Inventory’s (NPI’s) reported
113 methods and data to assess the emissions from power plants and their impacts.
114 Nelson (2007) estimated improved mercury emission from stationary combustion
115 sources. They used both the top-down and bottom-up approaches (NPI method)
116 to quantify the emissions. Their two approaches led to similar estimates for the
117 mercury emissions of black coal power plants but found that the NPI method
118 underestimated the mercury emissions from brown coal. Nelson et al. (2012) es-
119 timated the mercury emissions from the Australian landmass using the results
120 and methodology from Nelson (2007). They found that 15 % of the total mercury
121 emissions are from coal-fired power plants in Australia. Tang and Mudd (2015)
122 analysed the NPI reported data for power plants in Australia. They provided in-
123 sights into the present scenarios of emission from the power plants in Australia.
124 They reported that the power plants are a significant source of F, HCL, NO_x,
125 PM_{2.5}, SO₂ and H₂SO₄ (ranging from 30 % to 90 % of emissions) compared with
126 emissions from other combustion sources in Australia. Strezov and Cho (2020) also
127 used the NPI database to quantify the environmental impacts of thermal power

128 plants in Australia using the “Recipe” methodology. They identified greenhouse
129 gases and air pollutants, including SO₂, NO_x and PM_{2.5} as significant pollutants
130 of concern from the coal and gas-fired power plants. They found that brown coal
131 had the most substantial adverse environmental impacts, followed by black coal.

132 State-based environmental agencies in Australia (e.g. the EPA Victoria and
133 NSW DPIE) employ fixed ambient air quality monitors in the vicinity of popu-
134 lated areas and industrial areas to check air pollution levels. These monitors have
135 sensors and provide continuous real-time data and valuable long-term trend data,
136 which helps environmental agencies develop air pollution control strategies. Also,
137 real-time data provides an early warning system to avoid any negative impacts
138 on the population living in the vicinity and help protect the surrounding environ-
139 ment. However, there is no publicly available real-time or long-term continuous
140 monitoring of emissions from anthropogenic sources in Australia. As a supplement
141 to such existing monitoring activities, this study focuses on a temporal emission
142 profile for power plants in Eastern Australian states.

143 Studies have found that modelled concentrations are sensitive to both tem-
144 poral and spatial distribution of the emission (Matthias et al., 2018; Vedrenne
145 et al., 2016; Wang et al., 2010, and references therein). Currently, high-resolution
146 inventory for power plants is not available for Australia and any other country in
147 the world. Thus, an emission inventory with high temporal data and an accurate
148 spatial representation of power plants will improve the air quality modeling.

149
150 Here we present a new detailed emission inventory for power plants in Australia
151 which can be used as an input for chemical transport models. It is characterised by
152 high temporal and spatial distribution of emissions. The emissions from coal and
153 gas-fired power plants in Eastern Australian states have been estimated using the
154 time-series of electricity generation data provided by the NEM. The pollutants es-
155 timated are sulfur dioxide (SO₂), nitrogen dioxide (NO₂), particulate matter (PM)
156 split into PM₁₀ and PM_{2.5} (with particles having an aerodynamic diameter less
157 than 10 and 2.5 μm, respectively) and mercury (Hg). The emission factors (EFs)
158 used in this study are estimated for each power plant. We discuss the emissions for
159 all coal and gas-fired power plants in each state with their temporal variation. This
160 method provides detailed emission time-series for every power plant. We discuss
161 the benefits of having an emission time-series relative to the annual average emis-
162 sions provided by inventories to date. Finally, we compare these new estimated
163 emissions with existing emission inventories provided by EDGAR and NPI.

164 2 Methods and Data

165 2.1 Conversion of electricity generation data to amount of fuel burnt

166 In this study, emissions from power plants are estimated using electricity genera-
167 tion data sampled at 5 minute intervals. Figure 1 shows the location of all the coal
168 and gas-fired power plants in the NEM data that are used in the analyses. Firstly,
169 electricity generation data from coal and gas-fired power plants is extracted from
170 the NEM database and is converted into fuel burnt and then plant specific emis-
171 sion factors (EFs) are used to estimate the emissions. Figure 2 shows the schematic

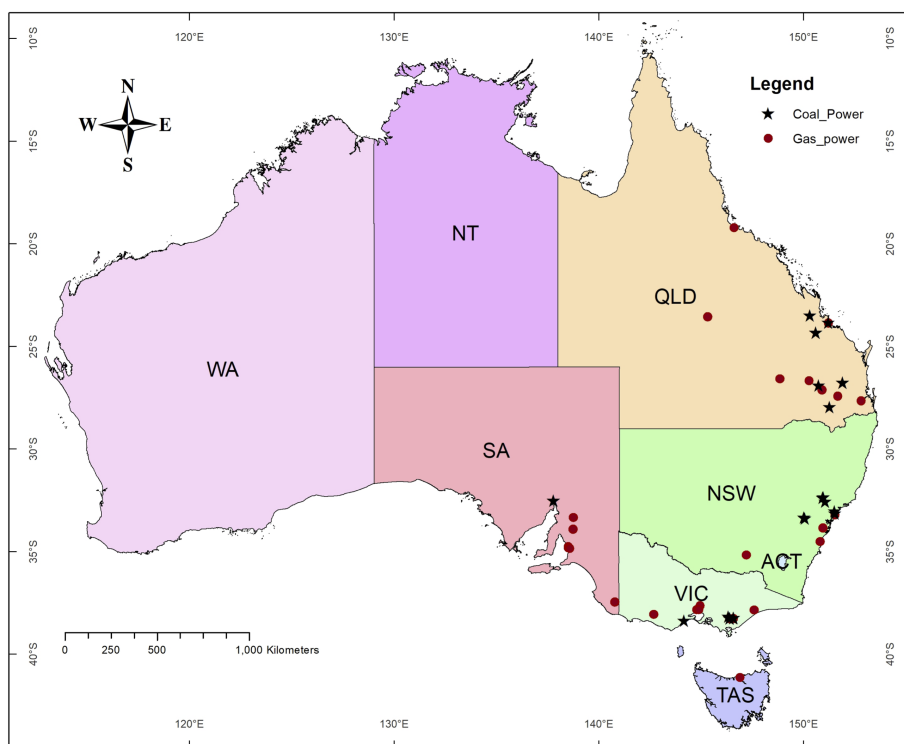


Fig. 1 Location of coal and gas-fired power plants in Queensland (QLD), New South Wales (NSW), South Australia (SA), Victoria (VIC) and Tasmania (TAS) covered by NEM. The Australian Capital Territory (ACT) is covered by the NEM but has no power plants. Western Australia (WA) and the Northern Territory (NT) are not covered by the National Electricity Market.

172 flowchart for emissions estimation from the coal and gas-fired power plants and
 173 the following sections details the complete methodology.

174 Electricity generation numbers (units of MWh) are converted to input heat
 175 energy (units of PJ/yr) generated from the fuel combustion using the plant's
 176 specific thermal efficiency (Eqn 1). The thermal efficiency of a plant is the ratio of
 177 energy produced to input heat energy from fuel combustion, given as a percentage.
 178 The input heat energy is converted to total fuel burnt using the higher heating
 179 value (also termed as the gross calorific value, GCV) of fuel (Eqn 1), which is
 180 defined as the amount of heat released when a unit mass of the fuel is burnt
 181 completely. The specific thermal efficiency values for coal and gas-fired power
 182 plants are accessed from the plant-specific details provided in the NEM metadata
 183 (see Tables 1 and 2) and specific higher heating values of fuel used (provided in
 184 Table 3) are adopted from the NPI methodology document (DSEWPC, 2012). We
 185 estimate the fuel burnt as:

$$FB = \frac{ELEC_{output} \times 3600}{(TE/100) \times (HHV/1000)} \quad (1)$$

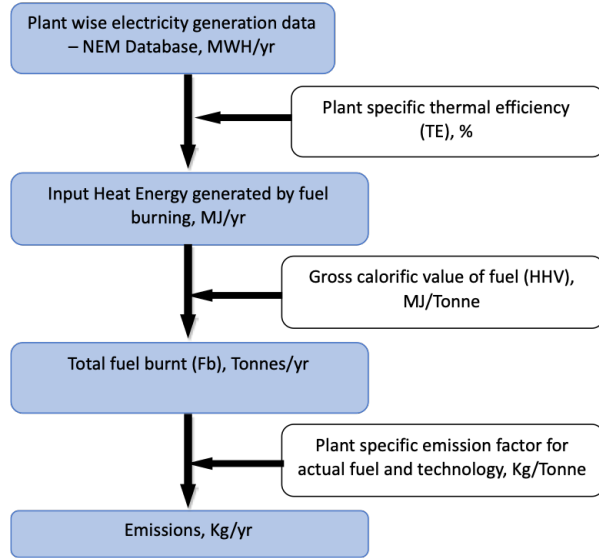


Fig. 2 Schematic flowchart for emissions estimation from the coal and gas-fired power plants. This flowchart refers to estimate the annual emission. To estimate the hourly emissions, the hourly electricity generation data was used instead of annual electricity generation data.

186 where FB is the fuel burnt (tonnes/yr), $ELEC_{output}$ is the electricity generated
 187 in a year (MWh/yr), TE is the thermal efficiency and HHV is the higher heat-
 188 ing value of fuel (MJ/kg). Multiplication by 3600 denotes the conversion between
 189 MWh and MJ and division by 1000 converts between kg and tonne.

190

191 2.2 Conversion of fuel burnt to emissions

192 Emissions are estimated by converting the fuel burnt into the emissions using the
 193 pollutant-specific emission factors (Eqn 2). Emission factors (EFs, expressed in
 194 kg/tonne) are defined as the mass of pollutant emitted per unit of activity, which
 195 in this case is taken to be the mass of fuel burnt. Finally, we estimate the emission
 196 as:

$$E_i = \frac{FB \times EF_i}{1000} \quad (2)$$

197 where E is the pollutant emission rate (tonnes/yr); FB is the fuel burnt (tonnes/yr);
 198 EF is emission factor for the pollutant (kg/tonne), i is the pollutant.

199 Plant-specific EFs developed from measurements are not available for all coal and
 200 gas-fired power plants in Australia. Instead, EFs used to estimate the emissions
 201 are derived using the NPI methodology (DSEWPC, 2012), containing comprehen-
 202 sive details of emissions factors and process information for air pollution source
 203 categories. EFs for various pollutants are derived using the facility-specific in-
 204 formation relating to fuel type (i.e., black, brown coal, oil), fuel characteristics,
 205 air pollution controlling devices (e.g. electrostatic precipitators, filter bags) and
 206 operating conditions (pulverized coal-fired in dry bottom boilers).

Table 1 Coal-fired power plant details. *Plant is no longer operational, NSW:New South Wales, VIC: Victoria, QLD: Queensland, SA: South Australia, BC: Black Coal, BrC: Brown Coal, FFB: Fabric Filter Bags, ESP: Electrostatic Precipitator, LNB: Low NO_x Burner, Con. Tech.:Control Technology (Source: NPI data), TE: Thermal Efficiency (%) (Source: NEM metadata).

Power Station	Region	Fuel Burnt	Con. Tech.	TE
Bayswater PS	NSW	BC	FFB, LNB	36
Eraring PS	NSW	BC	FFB, LNB	35
Mt Piper PS	NSW	BC	FFB, LNB	37
Liddell PS	NSW	BC	FFB, LNB	34
Vales Point B PS	NSW	BC	FFB, LNB	35
Kogan Creek PS	QLD	BC	FFB, LNB	39
Millmerran PS	QLD	BC	FFB, LNB	37
Callide C Nett Off	QLD	BC	FFB, LNB	37
Gladstone PS	QLD	BC	FFB, LNB	35
Tarong PS	QLD	BC	ESP, LNB	36
Stanwell PS	QLD	BC	ESP, LNB	36
Tarong North PS	QLD	BC	ESP, LNB	39
Callide PS	QLD	BC	ESP, LNB	37
Loy Yang B PS	VIC	BrC	ESP, LNB	27
AGL Loy Yang	VIC	BrC	ESP, LNB	27
Yallourn W PS	VIC	BrC	ESP, LNB	24
Redbank (*)	NSW	BC	FFB, LNB	33
Wallerawang (*)	NSW	BC	FFB, LNB	30
Morwell (*)	VIC	BrC	ESP, LNB	24
Anglesea (*)	VIC	BrC	ESP, LNB	27
Hazelwood (*)	VIC	BrC	ESP, LNB	22
Northern (*)	SA	BrC	ESP, LNB	28
Playford (*)	SA	BrC	ESP, LNB	28

207 2.3 Emission factors for Coal-fired power plants

208 2.3.1 SO₂ EF

209 Sulfur dioxide (SO₂) emissions are due to the conversion of sulfur content in the
 210 fuel to gaseous SO₂. Different fuels have different sulfur content, for example higher
 211 sulfur content is found in black coal than brown coal (Huleatt, 1991; Durie, 1992).
 212 The SO₂ emission factors are derived by sulfur content of the type of coal burned
 213 in different regions, given in Table 3, combustion technology and pollution control
 214 technology (Eqn. 3).

$$EF_{SO_2} = 1000 \times \frac{S}{100} \times \left(1 - \frac{CE}{100}\right) \times 2 \quad (3)$$

215 where S is the fuel sulfur content (%); CE is the efficiency of pollution control de-
 216 vice for pollutant i (%). Here multiplication by 1000 denotes the conversion from kg
 217 to tonne of fuel. As coal-fired electricity generators in Australia are not equipped
 218 with any SO₂ abatement technology, unabated emission factors for SO₂ emissions

Table 2 Gas-fired power plants details. NSW:New South Wales, VIC: Victoria, QLD: Queensland, SA: South Australia, TAS: Tasmania, OCGT: Open-Cycle Gas Turbine, CCGT: Closed-Cycle Gas Turbine, TE: Thermal Efficiency (%), Tech: Technology.

Power Station	Region	Tech	TE
Braemar	QLD	OCGT	30
Braemar 2	QLD	OCGT	30
Oakey	QLD	OCGT	33
Roma Gas	QLD	OCGT	30
Barcaldine	QLD	CCGT	46
Condamine	QLD	CCGT	48
Darling Downs	QLD	CCGT	46
Swanbank B & E	QLD	CCGT	47
Townsville	QLD	OCGT	46
Yarwun	QLD	CCGT	34
Colongra	NSW	OCGT	32
Uranquinty	NSW	OCGT	32
Smithfield Energy	NSW	CCGT	41
Tallawarra	NSW	CCGT	50
Somerton	VIC	OCGT	24
Bairnsdale	VIC	OCGT	34
Jeeralang A	VIC	OCGT	23
Jeeralang B	VIC	OCGT	23
Mortlake	VIC	OCGT	32
Valley	VIC	OCGT	24
Laverton North	VIC	OCGT	30
Newport	VIC	Steam	33
Osborne	SA	CCGT	42
Hallett	SA	OCGT	44
Dry Creek	SA	OCGT	26
Ladbroke Grove	SA	OCGT	30
Mintaro	SA	OCGT	28
Quarantine	SA	OCGT	32
Pelican Point	SA	CCGT	48
Torrens Island	SA	Steam	30
Torrens Island 2	SA	Steam	32
Tamar Valley	TAS	CCGT	48
Bell Bay Three	TAS	OCGT	29
Bell Bay Three	TAS	OCGT	28

219 are used.

220

221 2.3.2 PM EF

222 Emission factors for particulate matter (PM₁₀ and PM_{2.5}) have been calculated
 223 plant-wise using Eqn 4. Plant-specific details including operating conditions (pul-
 224 verized coal-fired in dry bottom boilers), fuel type burnt (black coal or brown

Table 3 Input parameters used to calculate the EFs for coal fired power plants. Uncertainty bounds are provided by the range. (a) - Huleatt (1991), (b) - Durie (1992); Perry et al. (1984), (c) - DSEWPC (2012), (d) - Meij (1994); USEPA (1995); Zhao et al. (2010) and references therein, (e) - USEPA (1995); Zhao et al. (2010) and references therein.

Parameters	Values (Range)
A- Ash Content in NSW coal (%) ^(a)	21.6 (17.4-25.8)
A- Ash Content in QLD coal (%) ^(a)	19.9 (11.7-28)
A- Ash Content in Brown coal (%) ^(b)	2.4 (1.3 -3.5)
S- Sulfur content in NSW coal (%) ^(a)	0.424 (.323-0.525)
S- Sulfur content in QLD coal (%) ^(a)	0.435 (0.145-0.725)
S- Sulfur content in Brown Coal (%) ^(b)	0.115 (0.09-0.14)
HHV of NSW coal (MJ/kg) ^(c)	24.585 (22.9 - 26.27)
HHV of QLD coal (MJ/kg) ^(c)	23.35 (19.3 - 27.4)
HHV of Brown coal (MJ/kg) ^(c)	9.48 (8.76 - 10.2)
HHV of Gas (MJ/Nm ³) ^(c)	38.8 (38 - 39.6)
CE - Efficiency of ESP (%) ^(d)	99.3 (99-99.6)
CE - Efficiency of FF (%) ^(e)	99.87 (99.76-99.99)

225 coal), fuel characteristics and control technologies are used (Table 1 and Table 3).

226

$$EF_{PM_y} = 1000 \times \frac{A}{100} \times \frac{F}{100} \times \left(1 - \frac{CE}{100}\right) \times f_y \quad (4)$$

227 where A is the ash content of fuel (%), F is the fly ash fraction (%), f is the
 228 particulate mass fraction by size and y is the particulate size (either 2.5 or 10
 229 μm in diameter). Here multiplication by 1000 relates to the conversion from kg to
 230 tonne of fuel.

231 Australian power plants are equipped with either fabric filter (FF) or electrostatic
 232 precipitator (ESP) to control particulate emissions (Nelson, 2007; Pavlish et al.,
 233 2003), with efficiency values given in Table 3. The fly ash fraction is 0.9 times the
 234 total ash for black and brown coal-fired power plants (USEPA, 1998). The particu-
 235 late mass fractions for PM_{10} and $\text{PM}_{2.5}$ to the filtered fly ash (after the particulate
 236 filtration devices) are determined as 0.67 and 0.29, respectively, for power plants
 237 equipped with ESPs and 0.92 and 0.53, respectively, for power plants equipped
 238 with FFs (DSEWPC, 2012).

239

240 2.3.3 NO_x EF

241 The facility-specific EFs for NO_2 (Table 4) are directly adopted from the NPI
 242 methodology document (DSEWPC, 2012). Generally, power plants emit oxides of
 243 nitrogen- NO and NO_2 , abbreviated as NO_x . The NPI has estimated the facility-
 244 specific NO_x emission factors by direct measurement and all NO_x is expressed in
 245 terms of NO_2 . NO_x emissions are process specific rather than fuel specific. The
 246 NO_x is formed during combustion by three mechanisms: (1) thermal NO_x , (2) fuel
 247 NO_x and (3) prompt NO_x (Muzio and Quartucy, 1997; Wendt, 1980; Miller and
 248 Bowman, 1989). Thermal NO_x forms from the oxidation of molecular nitrogen

249 in the combustion air, fuel NO_x from the oxidation of chemically-bound nitro-
 250 gen in the fuel, and prompt NO_x from the reaction between molecular nitrogen
 251 and hydrocarbon radicals. The first two contribute 20% and 80% of NO_x for-
 252 mation respectively in coal-fired power plants and prompt NO_x contributes only
 253 an insignificant fraction (Srivastava et al., 2005). In Australia, power plants are
 254 equipped with low NO_x burner (LNB) technology which controls the formation of
 255 both thermal and fuel NO_x.

256

Table 4 Emission factor of nitrogen dioxide (units of Tonne/Pj) for coal-fired power plant details, Source:-DSEWPC (2012).

Power Station	EF-NO ₂	Power Station	EF-NO ₂
Bayswater PS	220	Eraring PS	220
Mt Piper PS	220	Liddell PS	260
Vales Point B PS	260	Kogan Creek PS	523
Millmerran PS	523	Callide C Nett Off	523
Gladstone PS	523	Tarong PS	523
Stanwell PS	523	Tarong North PS	523
Callide PS	523	Loy Yang B PS	136
AGL Loy Yang	136	Yallourn W PS	106
Redbank PS	260	Wallerawang PS	260
Anglesea PS	151	Hazelwood PS	151
Northern PS	136	Playford PS	136
Morwell PS	151		

257 2.3.4 Hg EF

258 Mercury is emitted as elemental Hg from coal-fired power plants and is emitted
 259 from coal-fired power plants due to the presence of mercury in the coal. The
 260 Hg emission factors are adopted from Nelson (2007). The study (Nelson, 2007)
 261 derived the Hg emission factors for the Australian coal-fired power plants (Table
 262 5). The Hg emissions were assumed to be the function of Hg content in the coal
 263 and particulate control technology. Currently, Australian power plants are not
 264 equipped with specific control technologies to control mercury emissions although
 265 these mercury emissions are captured by ESPs and FFs. Nelson (2007) estimated
 266 the uncertainty of 75% in the mercury emissions which includes 50% uncertainty
 267 due to Hg content and 50% uncertainty due to control technologies (Nelson, 2007).
 268 Table 5 provides the emission factors used in our study. The emissions have also
 269 been estimated by using the NPI EFs. The NPI emission factors for black coal
 270 and brown coal are 0.042 and 0.026 g_{Hg} per tonne of coal burnt DSEWPC (2012),
 271 respectively, which are within the range of Hg EF provided by Nelson (2007).

Table 5 Mercury Emission factor (units of g tonne^{-1} of fuel burnt) (Source: Nelson (2007)).

Coal and particle capture type	Emission Factor
Black coal stations with ESPs	$0.05 \pm 75\%$
Black coal stations with FFB	$0.05 \pm 75\%$
Brown coal stations (only use ESPs)	$0.032 \pm 75\%$

2.4 Emission factors for gas-fired power plants

The list of gas-fired power plants is provided in Table 2. Emission factors used to calculate the emissions from gas-fired power plants are shown in Table 6. These emission factor values for NO_x as NO_2 , SO_2 , $\text{PM}_{2.5}$ and PM_{10} for gas-fired power plants are directly adopted from the NPI document (DSEWPC, 2012). The particulate emissions from gas-fired power plants are mostly in the finer fraction range i.e, in $\text{PM}_{2.5}$. Therefore, the EF values of PM_{10} and $\text{PM}_{2.5}$ for gas-fired power plants are assumed to be same (Pavri and Moore, 2001). Also, there is no PM control measure used for gas-fired plants in Australia. The Hg content in gas is negligible, therefore the Hg emissions from gas-fired power plants are not considered in the subsequent analyses.

Table 6 Emission factors for gas-fired power plants (DSEWPC (2012)). (a)- For All power stations except (b) & (c), (b) - For Newport Power Station, Victoria, (c) - For Torrens Island Power Station, South Australia.

Species	Emission Factor (kg/Pj)
NO_x	59000 (a), 117000 (b), 97000 (c)
SO_2	250 (a)
$\text{PM}_{2.5}$ & PM_{10}	3200 (a)

283

2.5 Uncertainty Estimates

The uncertainty around the central estimates of the emissions is calculated using a Monte Carlo simulation approach. In this study, uncertainty refers to the lower and upper bounds of a 95% confidence interval (CI) around a central estimate (Zhao et al., 2010; Liu et al., 2015). The Monte Carlo framework assumes a probability distribution for each of the input parameters. The normal distribution for all the input parameters is assumed. The upper and lower limits of the input parameters are considered to correspond to the 95% confidence interval (CI) of the distribution. The distribution of emission estimates was quantified by resampling input parameters following their respective distributions 10000 times.

The central value, lower and upper limits of the input variables (ash content, sulfur content, the higher heating value of fuel and control technologies efficiency for particulate matter) for coal-fired power plants are tabulated in Table 3. The uncertainty in the TE of the power plant is estimated to be the function of fuel

287

loading (Cardu and Baica, 1999; Sengupta et al., 2007), which refers to the rate of burning of fossil fuel (tonnes/hr). For coal-fired power plants, the uncertainty in TE based on fuel loading is taken as $\pm 4.2\%$ (Sengupta et al., 2007). For the gas-fired power plants, only uncertainties associated with variations in TE are considered. The uncertainty in TE of gas-fired power plants based on the fuel loading in open cycle gas turbines and combined cycle gas turbine are taken as $\pm 16.5\%$ and $\pm 4\%$, respectively (Jansohn, 2013). The uncertainty associated with SO_2 and $\text{PM}_{2.5}$ emission factors for gas-fired power plants are not included in the analyses due to a lack of underlying information such as gas composition and plant operating conditions.

2.6 Comparison with previous emission inventories

Finally, we have compared our estimated emissions with existing emission inventory covering the regional emission inventory i.e., National Pollution Inventory (NPI) and global emission inventory i.e., EDGARv4.3.2 estimates. Industries in Australia report their annual estimated or measured emissions of pollutants into the air, water and land to NPI. The companies estimate their emissions either based on the NPI methodology (DSEWPC, 2012) or measurements. But, the measurements may be limited to only a few time-points per year, and are in general not monitoring continually. For the comparison, we have used the air point emission data for the the financial years 2015-16 and 2016-17.

For the comparison with EDGAR inventory database, we have used EDGARv4.3.2 dataset (Crippa et al., 2018). The EDGAR emission inventory provides gridded emissions at a spatial resolution of approximately 10 km. The gridded emissions for the years 2011 and 2012 are compared with the total estimated emissions of power plants. For the comparison, the power plants' location were selected manually to map them onto the EDGAR inventory. Some power plants were in the same grid-cells or close to the EDGAR grid-cells where emissions were elevated but we found the EDGAR emission inventory to contain inaccuracies in terms of placement of some power plants.

3 Results and Discussion

We estimated the power plants based emission factors and emission intensity values for sulfur dioxide (SO_2), nitrogen dioxide (NO_2), particulate matter (PM) split into PM_{10} and $\text{PM}_{2.5}$ and mercury (Hg). The emissions are estimated and discussed for these pollutants from coal and gas power plants in Queensland (QLD), New South Wales (NSW), Victoria (VIC), South Australia (SA) and Tasmania (TAS) regions using the plant's based emission factors and time-series of electricity generation data provided by the NEM. Later, we discussed the emissions temporal variation and their comparison with existing emission inventories provided by EDGAR and NPI.

Table 7 Coal-fired power plant specific emission factors for PM₁₀, PM_{2.5}, SO₂, NO₂ and Hg (units of kg/tonnes: Emission (kg) per unit of coal burnt in tonnes).

Power Station	Region	PM ₁₀	PM _{2.5}	SO ₂	NO ₂	Hg
Bayswater	NSW	0.9 (0.4-1.4)	0.5 (0.2-0.8)	23.5 (17.6-29.4)	15.0 (13.8-16.2)	$5 \times 10^{-5} \pm 75\%$
Eraring	NSW	0.9 (0.4-1.5)	0.5 (0.2-0.9)	24.2 (18.1-30.2)	15.4 (14.2-16.7)	$5 \times 10^{-5} \pm 75\%$
Liddell	NSW	1.0 (0.4-1.5)	0.6 (0.2-0.9)	24.9 (18.7-31.1)	18.8 (17.3-20.3)	$5 \times 10^{-5} \pm 75\%$
Mt Piper	NSW	0.9 (0.4-1.4)	0.5 (0.2-0.8)	22.9 (17.2-28.6)	14.6 (13.4-15.8)	$5 \times 10^{-5} \pm 75\%$
Vales Point B	NSW	0.9 (0.4-1.5)	0.5 (0.2-0.9)	24.2 (18.1-30.2)	18.3 (16.8-19.7)	$5 \times 10^{-5} \pm 75\%$
Redbank	NSW	1 (0.4-1.6)	0.6 (0.2-0.9)	25.7 (19.2-32.1)	19.4 (17.8-20.9)	$5 \times 10^{-5} \pm 75\%$
Wallerawang C	NSW	1.1 (0.5-1.7)	0.6 (0.3-1)	28.2 (21.2-35.3)	21.3 (19.6-23)	$5 \times 10^{-5} \pm 75\%$
Callide C	QLD	0.9 (0.3-1.5)	0.5 (0.1-0.9)	23.3 (7.0-39.6)	32.9 (27.0-38.9)	$5 \times 10^{-5} \pm 75\%$
Callide	QLD	2.7 (0.7-4.7)	1.2 (0.3-2.0)	23.3 (7.0-39.6)	32.9 (27.0-38.9)	$5 \times 10^{-5} \pm 75\%$
Gladstone	QLD	0.9 (0.3-1.6)	0.5 (0.1-0.9)	24.7 (7.4-41.9)	34.8 (28.6-41.1)	$5 \times 10^{-5} \pm 75\%$
Kogan Creek	QLD	0.8 (0.2-1.5)	0.5 (0.1-0.8)	22.1 (6.7-37.6)	31.3 (25.6-36.9)	$5 \times 10^{-5} \pm 75\%$
Millmerran	QLD	0.9 (0.3-1.5)	0.5 (0.1-0.9)	23.3 (7-39.6)	32.9 (27-38.9)	$5 \times 10^{-5} \pm 75\%$
Stanwell	QLD	2.8 (0.7-4.8)	1.2 (0.3-2.1)	24 (7.2-40.7)	33.9 (27.8-39.9)	$5 \times 10^{-5} \pm 75\%$
Tarong North	QLD	2.5 (0.7-4.4)	1.1 (0.3-1.9)	22.1 (6.7-37.6)	31.3 (25.6-36.9)	$5 \times 10^{-5} \pm 75\%$
Tarong	QLD	2.8 (0.7-4.8)	1.2 (0.3-2.1)	24 (7.2-40.7)	33.9 (27.8-39.9)	$5 \times 10^{-5} \pm 75\%$
Loy Yang B	VIC	0.4 (0.1-0.8)	0.2 (0-0.3)	7.9 (6.7-9.1)	4.8 (4.4-5.2)	$3 \times 10^{-5} \pm 75\%$
Loy Yang A	VIC	0.4 (0.1-0.8)	0.2 (0-0.3)	7.9 (6.7-9.1)	4.8 (4.4-5.2)	$3 \times 10^{-5} \pm 75\%$
Yallourn W	VIC	0.5 (0.1-0.9)	0.2 (0.1-0.4)	8.9 (7.5-10.3)	4.2 (3.8-4.5)	$3 \times 10^{-5} \pm 75\%$
Morwell	VIC	0.5 (0.1-0.9)	0.2 (0.1-0.4)	8.9 (7.5-10.3)	6.0 (5.4-6.5)	$3 \times 10^{-5} \pm 75\%$
Anglesea	VIC	0.4 (0.1-0.8)	0.2 (0.1-0.3)	7.9 (6.7-9.1)	5.3 (4.8-5.8)	$3 \times 10^{-5} \pm 75\%$
Hazelwood	VIC	0.5 (0.1-1)	0.2 (0.1-0.4)	9.7 (8.2-11.2)	6.5 (5.9-7.1)	$3 \times 10^{-5} \pm 75\%$
Northern	SA	0.4 (0.1-0.7)	0.2 (0.1-0.3)	7.6 (6.5-8.8)	4.6 (4.2-5.0)	$3 \times 10^{-5} \pm 75\%$
Playford	SA	0.4 (0.1-0.7)	0.2 (0.1-0.3)	7.6 (6.5-8.8)	4.6 (4.2-5.0)	$3 \times 10^{-5} \pm 75\%$

3.1 Emission factors for coal-fired power plants

The coal-fired power plant emissions are estimated using EFs for sulfur dioxide (SO₂), nitrogen oxide (NO₂), particulate matter (PM) split into PM₁₀ and PM_{2.5} and mercury (Hg). The emission factors for SO₂, PM₁₀ and PM_{2.5} derived using Eqns 3-4 are tabulated in Table 7. Similar methods to estimate EFs have been used by several studies (Zhao et al., 2010; Liu et al., 2015; USEPA, 1998). The SO₂ EFs for black coal-fired power plants in NSW and QLD vary between 22.9-28.2 ($\pm 25\%$) kg/tonne and 22.1-24.7 ($\pm 70\%$) kg/tonne respectively. In contrast, SO₂ EFs for brown coal-fired power plants in VIC and SA vary between 7.9-9.7 ($\pm 16\%$) kg/tonne. The lower SO₂ EFs for brown coal is attributed to low sulfur content (0.115% in brown coal compared to around 0.430% in black coal).

The PM₁₀ and PM_{2.5} EFs for black coal-fired power plants, equipped with FF, in NSW and QLD vary between 0.8-1.1 ($\pm 57\%$) kg/tonne and 0.5-0.6 ($\pm 67\%$) kg/tonne, respectively. For black coal-fired power plants equipped with ESP in QLD, EFs vary between 2.5-2.8 ($\pm 74\%$) kg/tonne and 1.1-1.2 ($\pm 76\%$) kg/tonne respectively. This is consistent with FF controlling PM more effectively relative to ESP technology (99.87% compared to 99.3% as can be seen in Table 3).

Brown coal power plants in VIC and SA power plants equipped with ESP have PM₁₀ and PM_{2.5} EFs varying between 0.4 - 0.5 ($\pm 76\%$) kg/tonne and 0.2 (\pm

357 76%) kg/tonne respectively. For brown coal-fired power plants, the PM EFs in
358 terms of kg/tonne are less than from black-coal-fired power plants, even for the
359 plants equipped with FFs. This is due to the lower ash content and higher moisture
360 content of brown coal compared to black coal.

361 Despite having lower emissions per tonne of brown coal burnt, brown coal has
362 roughly one third the fuel calorific value of black coal, which means more fuel is
363 required to be burnt to produce the same amount of electricity. Hence, emissions
364 of all species per unit electricity production (or emission intensity) are increased in
365 case of brown coal. Table S1 shows the plant-specific emission intensity in terms of
366 kg emissions per unit electricity produced. It can be clearly seen that the PM₁₀ and
367 PM_{2.5} emission intensity values for brown coal-fired power plants are greater than
368 the black coal-fired power plants equipped with FFs. Similarly, the SO₂ emission
369 intensity values for brown coal-fired power plants (2.9-3.38 kg/MWh) are close to
370 those from black coal-fired power plants (3.45-4.14 kg/MWh).

371 3.2 Emissions from coal-fired power plants

372 The total annual emissions of NO₂, SO₂, PM₁₀, PM_{2.5} and Hg from coal-fired
373 power plants are estimated from 2010 to 2019 using the NEM data (Figure 3).
374 Coal-fired power plants collectively contribute the majority of NO₂ and SO₂ emis-
375 sions from the power sector. The estimates show that coal-fired power plants con-
376 tributed 452 kTonne NO₂ and 485.7 kTonne SO₂ emissions for the year 2019. The
377 maximum NO₂ emissions are from coal-fired power plants in QLD (257.19 kTonne)
378 for the year 2019, followed by NSW (136.58 kTonne), VIC (57.92 kTonne). There
379 were no coal-fired plants operating in SA in 2019 and hence zero emission from
380 coal-fired power plants in SA. The maximum SO₂ emissions are from coal-fired
381 power plants in NSW (199.69 kTonne) for the year 2019, followed by QLD (182.28
382 kTonne), VIC (103.74 kTonne). In contrast to gaseous emissions, particulate emis-
383 sions from coal fired power plants in Eastern Australia are lower than might be
384 expected. Coal fired power plants contributed to 27.48 kTonne PM₁₀ and 13.46
385 kTonne PM_{2.5} emissions. The particulate emissions are significantly reduced by
386 particulate matter control devices (i.e., fabric filter bags or electrostatic precipi-
387 tators) but there is no control technology used to limit SO₂ and NO₂ emissions
388 (Nelson, 2007; Pavlish et al., 2003).

389 State-wise estimates show that emissions from coal-fired power plants have re-
390 duced from the year 2010 to 2019 in all states within the NEM network except
391 QLD. An average across all power plants studied, the analysis shows that there is
392 a decrease of 8% NO₂, 14% SO₂ and 10% PM (both PM₁₀ and PM_{2.5}) emissions
393 from the year 2010 to 2019. The main reason for the decrease in the emission is
394 the closure of coal-fired power plants in SA and VIC regions.

395 The emissions from Victorian coal-fired power plants have significantly reduced
396 for NO₂, SO₂, PM₁₀ and PM_{2.5} by 42%, 38%, 39% and 39% respectively, from
397 the year 2010 to 2019. Most of the percentage drop in Victoria (2010-2019) ac-
398 tually occurred in 2017 which is largely due to the closure of Hazelwood power
399 plant in March 2017 (AEC, 2016). For NSW, an approximately 6% reduction in
400 emissions was observed over the years 2010 - 2019 and for SA, 100 % decrease in
401 emissions due to the closure of all coal based power plants (AEC, 2016). For QLD,
402 the emissions from coal-fired power plants have increased for NO_x, SO₂, PM₁₀ and

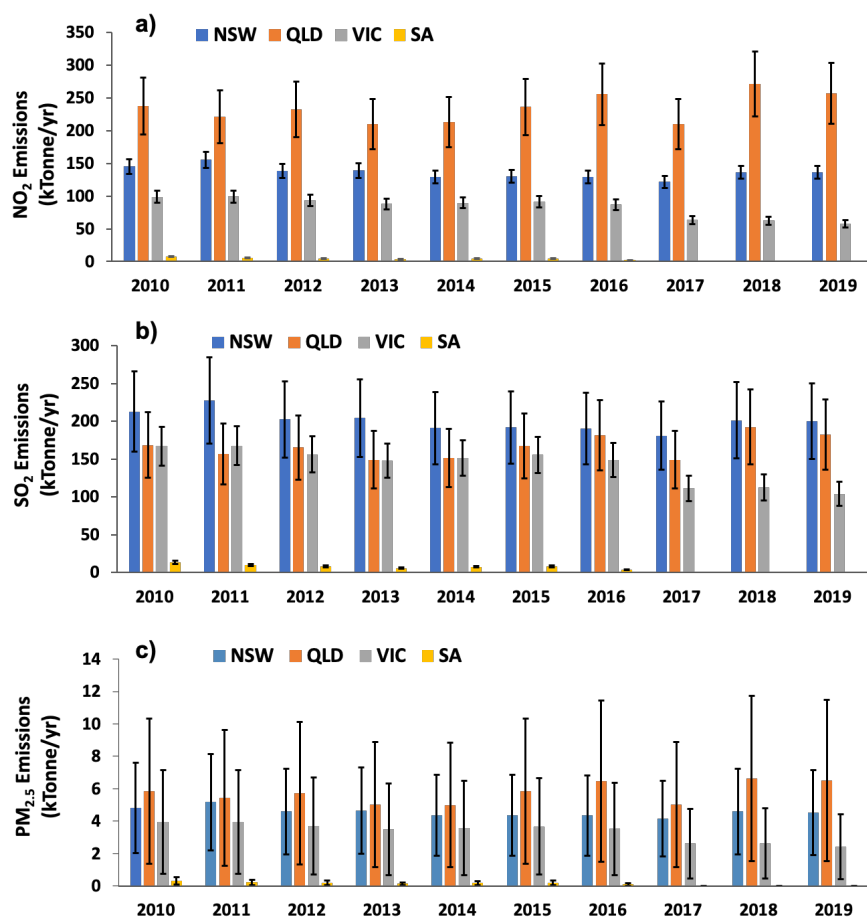


Fig. 3 Annual a) NO₂, b) SO₂ and c) PM_{2.5} emission with uncertainty from coal-fired power plants in the eastern Australia Energy Market calculated using the 5 min energy distribution data.

403 PM_{2.5} by 8%, 8%, 11%, and 11%, respectively. The emissions have increased from
 404 2016 to 2019 for most of the power plants in QLD because of the increase in elec-
 405 tricity demand which is attributed to population growth and change in weather
 406 conditions. More hot and cold days are experienced compared to previous years
 407 and hence more electricity is consumed to operate the air conditioners and heaters
 408 (EAAP, 2019).
 409

410 Figure 4 shows the estimated plant-specific NO_x and SO₂ emissions in QLD,
 411 NSW, SA and VIC. The maximum SO₂ emissions in year 2019 are from Eraring
 412 power station in NSW followed by Bayswater power plant in NSW. In QLD, Tarong
 413 power station has the highest SO₂ emissions followed by Stanwell and Gladstone
 414 power stations. In VIC, Loy Yang A emits the most SO₂, followed by Yallourn
 415 W power station and Loy Yang B power station. In contrast to SO₂, maximum

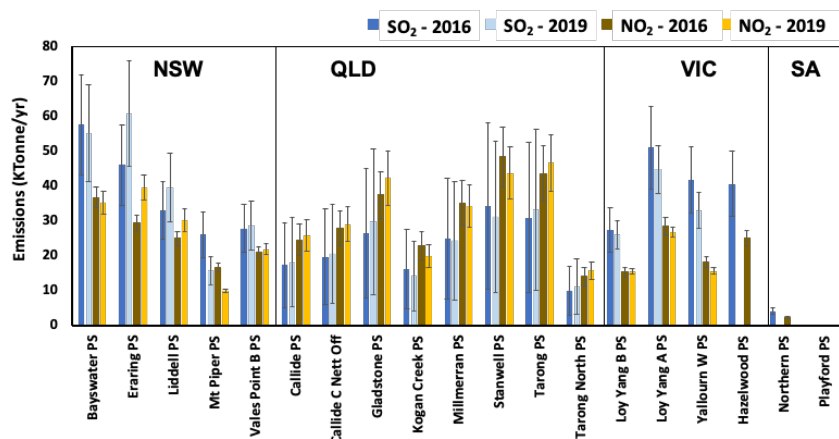


Fig. 4 Plant-wise annual NO_x as NO₂ and SO₂ emission for the year 2016 and 2019 from coal-fired power plants in the eastern Australia Energy Market calculated using the 5 min energy distribution data.

416 NO₂ emissions in year 2019 are from QLD from Tarong power station followed by
 417 Stanwell and Gladstone power station.

418 3.2.1 Mercury emissions from Coal-fired power plants

419 Mercury emissions are estimated using the EFs derived by Nelson (2007) (Table 5)
 420 and NPI EFs. Figure 5 shows plant-wise mercury emission from coal-fired power
 421 plants for the financial year 2015-16 and 2016-17. Using Nelson (2007) EFs, it
 422 is estimated that the total Hg emissions from coal-fired power plants are 3.59 tonnes
 423 for the year 2016-17. For 2019, the emissions have reduced to 3.13 tonnes due to the
 424 closure of the power plants, primarily the closure of Hazelwood power station in
 425 VIC in 2017. The 2019 plant-wise Hg emissions show that Victorian power plants
 426 have the largest emissions. Around 1.26 (0.42-2.4) tonnes of Hg are emitted from
 427 Victorian coal-fired power plants followed by 0.99 (0.29- 1.73) tonnes in NSW and
 428 0.88 (0.26-1.27) tonnes in QLD in 2015-2016. The plant with the highest emissions
 429 in Victoria is AGL Loy Yang followed by Energy Australia Yallourn and Loy Yang
 430 B power station.

431 Here, we compared the emissions estimated by NPI EFs with our derived EFs
 432 using Nelson (2007) study and energy generation data (shown in Figure 5). The
 433 emission factor estimates from Nelson (2007) with blue and red dots in Figure
 434 5 produce annual emissions the same range as the EF estimates from NPI. In
 435 the (Nelson, 2007) analysis, there was huge difference between the two estimates,
 436 however, in this work we find that the EFs for Hg have been updated by NPI and
 437 adjusted to EFs provided by Nelson (2007). Figure 5 also shows the NPI reported
 438 Hg emissions (indicated by the black and red bars). The comparison indicates
 439 that NPI reported emissions for QLD, VIC and SA, are within the range of our
 440 estimates. But for NSW, NPI reported emissions are negligible, and likely largely

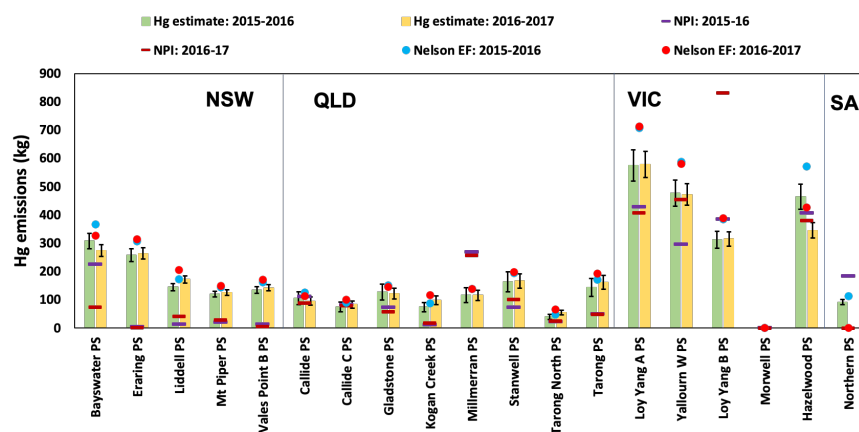


Fig. 5 Comparison of estimated Hg emissions using EFs established by (Nelson, 2007) (shown by bars with errors) and NPI method (shown by points) with NPI reported emissions (shown by line) for coal-fired power plants in the eastern Australia Energy Market for the financial year 2015-16 and 2016-17.

441 underestimated. This is likely because of the old Hg EF values are being used by
 442 industry which have not been updated with the latest EFs proposed by NPI.
 443

444 3.3 Emissions from gas-fired power plants

445 Figure 6 shows total estimated emissions of NO_x as NO_2 , SO_2 and $\text{PM}_{2.5}$ from
 446 gas-fired power plants from 2010 to 2019. Compared to coal-fired power plants, gas-
 447 fired power plants have lower emissions. The majority of emissions from gas-fired
 448 power plants are NO_2 (14.2 kTonne/yr) with SO_2 emissions being negligible (0.043
 449 kTonne/yr). The NO_x results from the formation of fuel NO_x in gas turbines,
 450 rather than thermal and prompt NO_x which are believed to be negligible in gas
 451 plants (Pavri and Moore, 2001). The source of SO_2 is the fuel sulfur content. The
 452 lower sulfur content in gaseous fuel leads to low SO_2 emissions.

453 Figure 6c shows that the estimated $\text{PM}_{2.5}$ emissions are very low (0.6 kTonne/yr).
 454 The low $\text{PM}_{2.5}$ emissions result from the negligible amount of unburnt fuel or fuel
 455 residue in contrast to the ash residue in coal burning. The PM mostly includes
 456 ash, ambient non-combustibles, and erosion and corrosion products and are influ-
 457 enced by the design of combustion system, fuel properties and combustor operating
 458 conditions (Pavri and Moore, 2001).

459 Figure 7 shows the estimated NO_2 emissions from the individual gas-fired
 460 power plants covered by the NEM. The maximum regional NO_x emissions are
 461 emitted from gas-fired power plants in SA (6.01 kTonnes/yr) followed by QLD
 462 (3.56 kTonnes/yr), VIC (2.6 kTonnes/yr), NSW (1.49 kTonnes/yr) and TAS (0.58
 463 kTonnes/yr) which is mostly due to the variation of gas-fired electricity production
 464 in different states. Yearly estimates for 2010 to 2019 show that emissions for NSW,
 465 QLD and TAS region have reduced by 21, 40 and 8% respectively, and for VIC and
 466 SA have increased by 330 and 10%, respectively. The reason for the decrease in

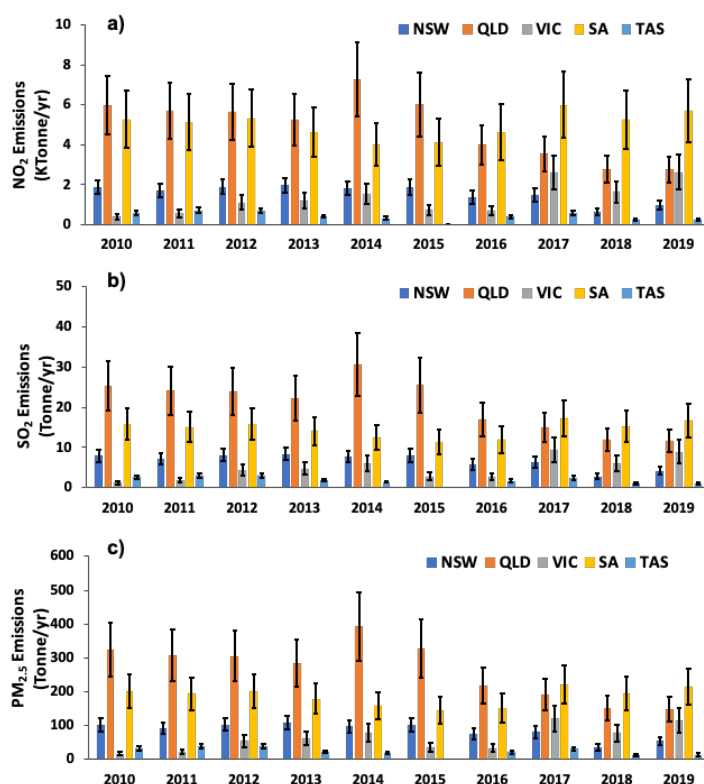


Fig. 6 Annual NO_x as NO_2 , SO_2 and $\text{PM}_{2.5}$ emission with uncertainty from gas-fired power plants in the eastern Australia Energy Market calculated using the 5 min energy distribution data from 2010 to 2019.

467 emissions from gas-fired power plants in NSW, QLD and TAS region is the increase
 468 in the production of electricity from renewable resources like hydropower and
 469 solar (rooftop) power plants (EAAP, 2019). There is also an increase of renewable
 470 resources of electricity generation in VIC and SA, but the decommissioning of
 471 some of the large coal-fired power plants in these states has shifted the burden to
 472 gas-fired power plants.

473 3.4 Temporal variation of emissions

474 To understand the average temporal distribution, both in terms of diurnal, weekly
 475 and seasonal patterns, normalised profiles were calculated for each hour-of-day,
 476 day-of-week and month-of-year. This type of information typically forms the basis
 477 of a temporal break-down of emissions used in chemical-transport modelling. This
 478 was done separately for each plant type (gas-fired and coal-fired plants) and for the
 479 individual power plants. Given the assumed linear relationship between emissions
 480 and electricity generation at these plants, this analysis was performed directly on
 481 the electricity production data.

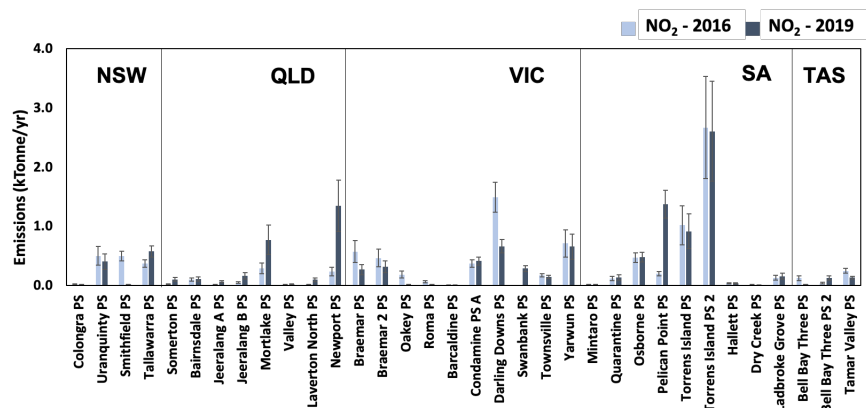


Fig. 7 Plant-wise annual NO_x emissions for the year 2016 and 2019 from gas-fired power plants in the eastern Australia Energy Market calculated using the 5 min energy distribution data.

482 The resulting temporal profiles on these different time-scales is presented in
 483 Figure 8. For coal-fired plants, power production and therefore emissions are near-
 484 constant throughout the 24-hour cycle, apart from a fall of 15-20% relative to the
 485 daily average between 02:00 h to 06:00 h, followed by two small peaks (Figure 8a).
 486 The first peak occurs between 08:00 h to 10:00 h (with an increase of 2-5%) while
 487 the second peak spans 18:00 h to 20:00 h hours (with an increase of 5-10% increase).
 488 In contrast, there is significantly more variation across the diurnal cycle for gas-
 489 fired power plants (Figure 8b). Emissions were negligible during the night, but
 490 showed a sudden increase in the morning (around 06:00-08:00 h), remaining rela-
 491 tively stable during the middle of the day (09:00-16:00 h), peaking in the evening
 492 (between 18:00-20:00 h) and then dropping down to near-zero levels (by 00:00 h).
 493 As with the diurnal distribution, the weekly emission profile for coal-fired power
 494 plants shows only minor variation, with weekend emissions being lower than week-
 495 day emissions by only 6-8% (Figure 8c). Again, the gas-fired power plants were
 496 substantially more variable, with emissions being lower on weekends by around
 497 60-70% (Figure 8d). When we average across the time-scale of a whole month,
 498 there was less of a difference between the gas- and coal-fired plants than on the
 499 shorter time-scales (Figure 8e,f). The seasonal emission profile for coal-fired power
 500 plants shows highest emissions in summer (Dec-Feb) and winter (Jun-Aug), with
 501 lower emissions observed in the months surrounding winter (Mar-May, Sep-Nov).
 502 In contrast, for gas-fired power plants no clear seasonal pattern was observed for
 503 monthly mean emissions.

504 Differences in the temporal variability of the emissions for coal- and gas-fired
 505 power plants can be attributed to their operating mechanisms. Coal-fired power
 506 plants are not turned off during the period of low electricity demand, as it takes
 507 several hours for these plants to reach their capacity. On the other hand, gas-fired
 508 power plants have short start-up times and their production can be quickly ramped
 509 up in minutes, thus they are turned off during periods of low demand. Being the
 510 cheaper source of electricity, coal-fired power plants are kept operational during

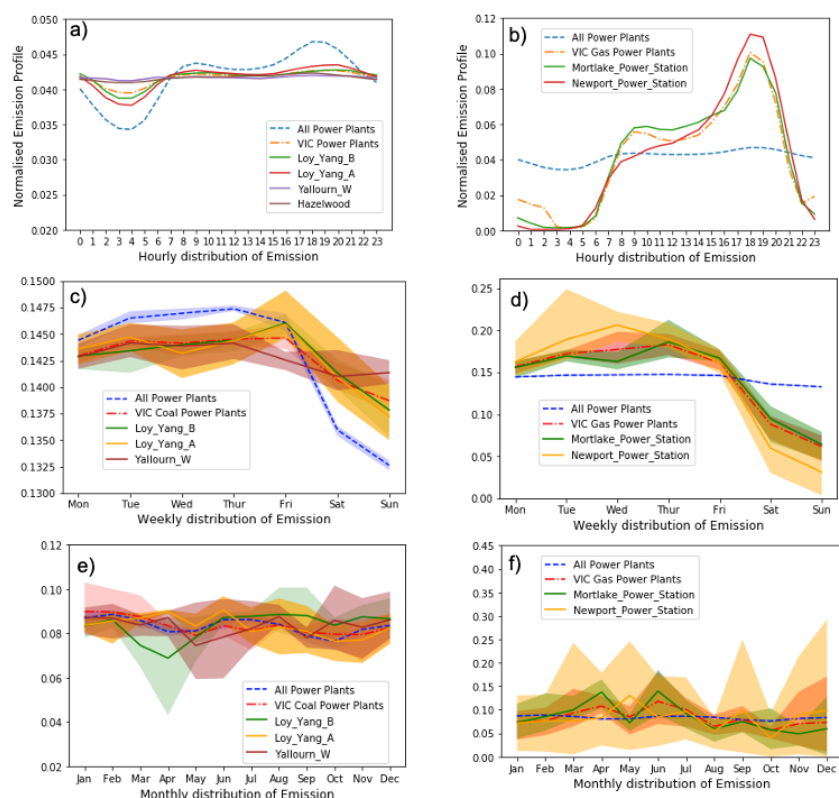


Fig. 8 Temporal variation of emissions from coal and gas fired power plants covering diurnal emission profile for (a) coal-fired and (b) gas-fired power plants, weekly emission profile for (c) coal-fired and (d) gas-fired power plants and monthly emission profile for (e) coal-fired and (f) gas-fired power plants.

511 low-demand periods to cover the supply.

512

513 Common practice for the temporal allocation of emissions in air quality mod-
 514 elling is to distribute an annual total based on diurnal, weekly and seasonal pat-
 515 terns of activity. Such activity patterns are typically based on averages, and the
 516 degree of accuracy of the resulting temporal distribution (accounting for all of
 517 these three cycles) is rarely quantified. The electricity generation data used here
 518 allows us to assess the likely errors resulting from this assumption. Figure 9 shows
 519 the normalised errors between the estimated hourly emissions and the correspond-
 520 ing time-series generated by convolving the average diurnal, weekly and seasonal
 521 profiles; the box-and-whisker plots summarise the distribution of normalised errors
 522 over plants. The normalized errors were an order of magnitude higher for gas-fired
 523 plants compared with coal-fired plants (24-1651% versus 13-54%). The analysis
 524 shows that models might result in a significant error in assessing the impact of
 525 emissions if gas-fired power plant emissions are inputted using an averaged emis-
 526 sion profile which may be based on electricity generation data from the whole
 527 power production sector (i.e. dominated by coal-fired power plants). The pre-

528 sented approach of emission estimation from electricity generation data provides
529 high temporal resolution emissions which is expected to improve the accuracy of
530 modelling results.

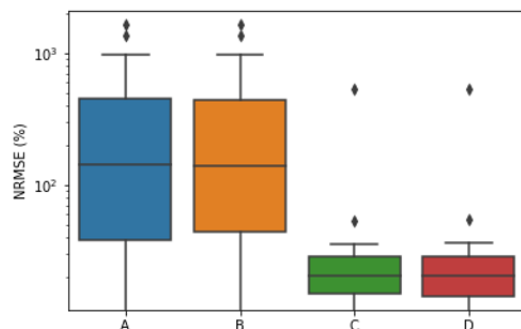


Fig. 9 The normalised root mean-squared error between the time-series of estimated emissions and the corresponding time-series based on the annual total and the mean diurnal, weekly and seasonal patterns. The box-and-whisker plots summarise the distribution of the normalised errors over plants. These errors are calculated separately for gas-fired power plants (A,B) and coal-fired plants (C,D); the dataset used to define the average profile was either all power plants (A,C) or only plants of the same fuel-source (B,D).

531 3.5 Uncertainty of emissions

532 After accounting for all the different sources of uncertainty, the resulting average
533 uncertainties for coal emissions are estimated as $\pm 13\%$ for NO_2 , $\pm 36\%$ for SO_2 ,
534 $\pm 68\%$ for PM_{10} , $\pm 70\%$ for $\text{PM}_{2.5}$ and $\pm 80\%$ for Hg. The variation in TE and
535 HHV is incorporated in all of the above uncertainty analysis. The higher uncer-
536 tainty in SO_2 emission estimates results from the additional incorporation of sulfur
537 content (S) while that in PM (split into PM_{10} and $\text{PM}_{2.5}$) emission estimates re-
538 sults due to the variation in ash content (A) and efficiency of particulate matter
539 control efficiency (CE), with the latter contributing the significantly. Similarly, for
540 Hg emissions, the variation is due to mercury content and efficiency of particulate
541 matter control efficiency. Hence, to further reduce uncertainty in the estimates,
542 we need a measurement of plant specific values for HHV, S content, A content,
543 Hg content and control efficiency (CE). The resulting average uncertainties for gas
544 emissions are estimated as $\pm 27\%$ for NO_2 , $\pm 30\%$ for SO_2 and $\pm 26\%$ for $\text{PM}_{2.5}$.
545 The uncertainty in gas emission estimates incorporates the variation in thermal
546 efficiency of power plants and heating value of the fuel. The variation of SO_2 and
547 $\text{PM}_{2.5}$ emission factors for gas-fired power plants are not included in the uncer-
548 tainty estimates due to lack of available data and the fact that gas-fired power
549 plants emit negligible SO_2 and $\text{PM}_{2.5}$.

550

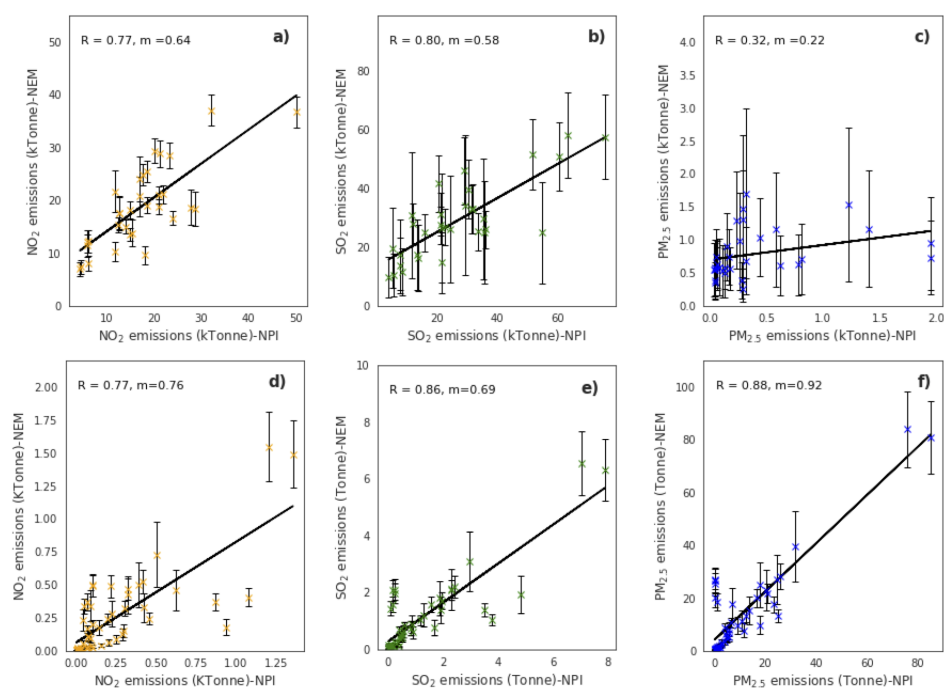


Fig. 10 Comparison between estimated emissions from coal (a - NO₂, b - SO_x and c - PM_{2.5} emissions) and gas (d - NO₂, e - SO₂ and f - PM_{2.5} emissions) fired power plants using this study's methodology (NEM) and NPI (National Pollution inventory) for the financial year 2015-16 and 2016-17. Here, R represents the correlation coefficient and m refers to the slope of the regression curve.

551 3.6 Comparison with annually reported National Pollution Inventory (NPI)

552 Figure 10 shows a comparison of estimated emissions from coal-fired power plants
 553 (Figure 10(a-c)) with NPI reported emissions for the financial years 2015-16 and
 554 2016-17. The comparison was performed separately for each region for both coal
 555 and gas-fired power plants. It can be seen that estimated average SO₂ and NO₂
 556 emissions from coal-fired power plants are well correlated within each region (with
 557 correlation coefficients varying between 0.6 and 0.95). The state-wise comparison
 558 is shown in Figure S1(a-c). The comparison shows NPI reported values for NO₂
 559 and SO₂ emissions in NSW and QLD are 12% and 4% lower, respectively, than the
 560 average estimated emissions from our work here, while in VIC the values are 4%
 561 higher. For PM_{2.5} emissions, the correlation coefficients vary between 0.32-0.67.
 562 For PM₁₀ and PM_{2.5} the NPI reported emissions in NSW, QLD and VIC, the
 563 values are 83, 43 and 57% lower, respectively, than the average estimated emis-
 564 sions in this paper. However, it can be seen that most of the NPI reported values
 565 are within the uncertainty range of the estimates quantified in this study. A likely
 566 explanation is that there are discrepancies in the methods used by the companies
 567 to calculate these emissions.

568

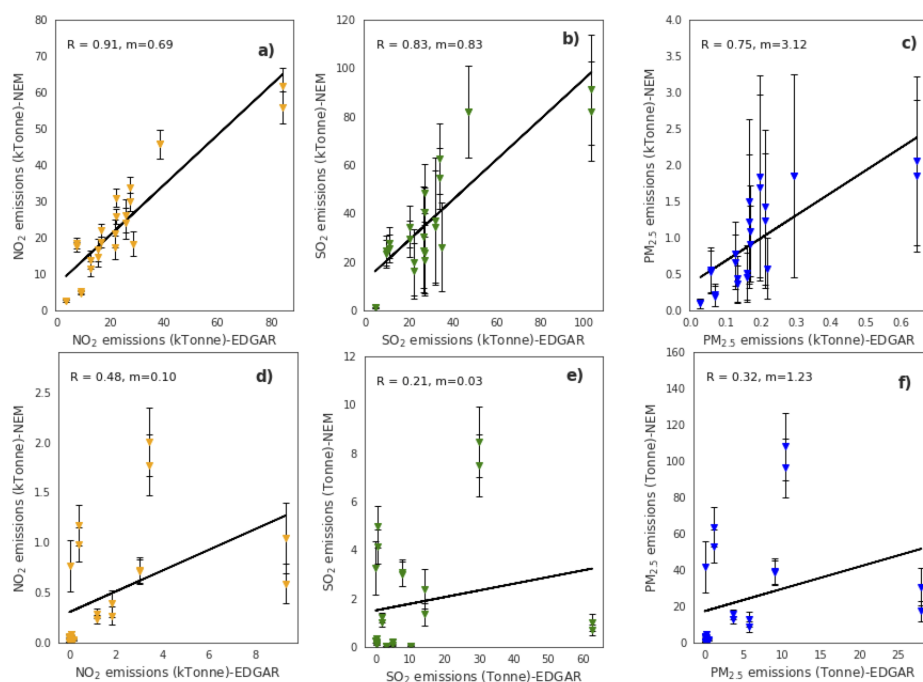


Fig. 11 Comparison between estimated emissions from coal (a - NO₂, b - SO_x and c - PM_{2.5} emissions) and gas (d - NO₂, e - SO₂ and f - PM_{2.5} emissions) fired power plants using this study's methodology (NEM) and EDGAR for the year 2011 and 2012. Here, R represents the correlation coefficient and m refers to the slope of the regression curve.

569 Figure 10(d-f) shows a comparison of estimated NO₂, SO₂ and PM_{2.5} from gas-
 570 fired power plants with NPI emissions reported for the financial years 2015-16 and
 571 2016-17. Our estimated SO₂, NO₂ and PM_{2.5} emissions from all gas-fired power
 572 plants are highly correlated (with R-value of 0.77, 0.86 and 0.88, respectively)
 573 with NPI reported emissions. For SO₂ emissions, comparison shows that the NPI
 574 reported emissions in NSW, VIC, QLD, SA and TAS are 135.8%, 2.2%, 12.8%,
 575 263.4% and 154.7% higher, respectively (see Figure S1-e). The reported values for
 576 NO₂ emissions in all states excepting QLD were higher (by 6-61%) (see Figure S1-
 577 d). For PM emissions, the values were lower (by 12- 67%) for all states except TAS
 578 (see Figure S1-f). For TAS, the values are 33.4% higher for both PM₁₀ and PM_{2.5}.
 579

580 The comparison shows that the NPI reported values are in reasonable agree-
 581 ment with our estimated emissions for both coal and gas-fired power plants. In
 582 case of discrepancy, the NPI reported emissions often fall within the calculated
 583 uncertainty range. However, there are still many other discrepancies in NPI re-
 584 ported emissions including the unreliability of companies self-reporting. We have
 585 found that companies are not consistent either in their application of the NPI's
 586 recommended methodology or with their independent methods to calculate their
 587 emissions. In some cases we found that the emissions are repeated identically over
 588 successive years. We found that details and metadata were missing, and the data
 589 reported lacked data quality checks (e.g., some cases were identified when coal-fired

590 power plants with reported emissions in successive years differing by more than
591 one order of magnitude). There is no system in place to ensure that these emis-
592 sions can be cross-validated by other techniques. The NPI estimates also lack any
593 details of seasonal or diurnal variation which is now a requirement of air pollution
594 modelling efforts.

595 3.7 Comparison with EDGAR

596 Figure 11(a-c) shows a comparison of estimated NO_2 , SO_2 and $\text{PM}_{2.5}$ from coal-
597 fired power plants with EDGAR emissions. Within each state, the comparison
598 showed a strong correlation for NO_2 (with correlation coefficients of 1.0, 0.81 and
599 0.99 for VIC, QLD and NSW, respectively) (see Figure S2-a), SO_2 (with correlation
600 coefficients of 1.0, 0.71 and 0.98 for VIC, QLD and NSW, respectively) (see Figure
601 S2-b) and $\text{PM}_{2.5}$ (with correlation coefficients of 1, 0.81 and 0.52 for VIC, QLD
602 and NSW, respectively) (see Figure S2-c) emissions from coal-fired power plants.
603 The comparison shows EDGAR emissions for NO_2 in NSW, QLD and SA are
604 4%, 10.5% and 80% higher, respectively, than the average estimated emissions in
605 our work here, and in VIC are 13% lower. For SO_2 , comparison shows EDGAR
606 emissions in NSW, VIC and SA are 14%, 80% and 59% lower, respectively, than the
607 average estimated emissions, and in QLD are 8% higher. This has implications for
608 Chemistry-Climate modelling using EDGAR emissions for the Australian region.

609 Figure 11(d-e) shows a comparison of estimated NO_2 , SO_2 and $\text{PM}_{2.5}$ from
610 gas-fired power plants with EDGAR emissions and the state-wise comparison is
611 shown in Figure 11(d-f). This comparison shows a poor correlation (with correlation
612 coefficients of 0.48, 0.21 and 0.32 for NO_2 , SO_2 and $\text{PM}_{2.5}$, respectively). This is
613 possibly related to inaccuracies in the EDGAR placement of some of the coal
614 and gas-fired power plants. There are more gas-fired plants, and we expect that
615 their locations are clustered. Also, in cases of ambiguity, plants can be sometimes
616 distinguished in the EDGAR grid-cells based on the relative size of their emissions.
617 However, the low emissions from gas-fired power plants made it difficult to identify
618 the correct corresponding grid-cells.

619 The analysis suggests that the emission factors used in EDGAR data for Aus-
620 tralian coal-fired and gas-fired power plants are the same for all regions i.e. region-
621 specific emission factors are not used. EDGAR (Crippa et al., 2018) uses inter-
622 national statistical data on fuel usage, economic activities, technology used and
623 emission abatement technologies to quantify the total emissions. The comparison
624 of EDGAR emissions with the power plant-specific annual fuel combustion shows
625 a strong correlation (with correlation coefficients more than 0.9) indicating that
626 the main difference is due to emission factors. The analyses showed that EFs used
627 in EDGAR (Crippa et al., 2018) estimates are lacking the Australia power plant's
628 specific details like the technology used, type of fuel burnt (brown coal or black
629 coal), fuel characteristics (ash content, sulfur content etc.) and emission abatement
630 technology (such as FF and ESP) used. Also, the diurnal variation of emissions is
631 not provided by EDGAR (Crippa et al., 2018) .

632 4 Conclusions

633 A new detailed emission inventory for coal and gas-fired power plants in eastern
634 Australia was developed to be used as input for air quality and chemical transport
635 models. The emissions are estimated by converting electricity generation data at
636 5-minute temporal resolution into fuel burnt and then derived plant-specific EFs
637 are used to estimate the emissions. This emission inventory therefore provides
638 plant specific EFs and emissions at the exact location of each power plant with
639 high temporal resolution. The temporal analyses of emissions shows the impor-
640 tance of considering high temporally resolved emission profiles. Comparison of our
641 estimated emissions with NPI and EDGAR shows that there are significant errors
642 and deficiencies in the NPI and EDGAR estimates. In particular, the NPI reported
643 emissions suggest that more stringent checks may be warranted on the quality and
644 consistency of emissions data reported by companies. The EDGAR provided emis-
645 sions at coarser spatial resolution and lacked the EF details used in our emissions
646 estimates. Also, both NPI and EDGAR estimates lack high temporal distribution
647 of emissions from plants, which is essential for modelling. The emissions provided
648 by our methodology can be further improved if plant-specific emission factors can
649 be estimated through direct measurement. The emissions and emission profiles
650 estimated in this study using real-time electricity generation data is expected to
651 improve air quality and chemical transport modelling. The presented method of
652 using high temporal resolution data can be extended to other parts of the world
653 for an improved analysis of the fate of pollutants able to be estimated by simu-
654 lations. Similarly, this method can be applied to estimate the emissions of other
655 pollutants and greenhouse gases from power plants, when appropriate electricity
656 generation data is available. In future work, these estimates will be used to model
657 the emission from both coal and gas-fired power plants and compared with surface
658 observational data.

659

660 5 Availability of data and materials

661 The datasets generated during the current study are available in the PANGAEA
662 repository, [<https://www.pangaea.de/tok/97ba6fe716625d601585573684b3fef15a83afec>]

663 **Acknowledgements** The authors acknowledge support from the Australian Research Coun-
664 cil (ARC) Centre of Excellence for Climate Extremes (CE170100023) and the Australian Re-
665 search Council's Discovery project: Tackling Atmospheric Chemistry Grand Challenges in the
666 Southern Hemisphere (DP160101598). MG is also supported by the Australian Government
667 Research Training Program Scholarship. This research was undertaken with the assistance of
668 resources and services from the National Computational Infrastructure (NCI) - under project
669 q90, which is supported by the Australian Government.

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838 **Supplementary information****Table S1** Emission Intensity for coal fired power plants (units of kg/MWH: Emission (kg) per unit of electricity produced in MWH).

Power Station	Region	PM ₁₀	PM _{2.5}	SO ₂	NO ₂	Hg
Bayswater	NSW	0.13 (0.06-0.21)	0.08 (0.03-0.12)	3.45 (2.58-4.31)	2.2 (2-2.4)	$1.71 \times 10^{-5} \pm 75\%$
Eraring	NSW	0.14 (0.06-0.22)	0.08 (0.03-0.13)	3.54 (2.66-4.43)	2.3 (2.1-2.4)	$1.76 \times 10^{-5} \pm 75\%$
Liddell	NSW	0.14 (0.06-0.22)	0.08 (0.04-0.13)	3.65 (2.74-4.56)	2.8 (2.5-3)	$1.81 \times 10^{-5} \pm 75\%$
Mt Piper	NSW	0.13 (0.06-0.2)	0.08 (0.03-0.12)	3.35 (2.51-4.19)	2.1 (2-2.3)	$1.66 \times 10^{-5} \pm 75\%$
Vales Point B	NSW	0.14 (0.06-0.22)	0.08 (0.03-0.13)	3.54 (2.66-4.43)	2.7 (2.5-2.9)	$1.76 \times 10^{-5} \pm 75\%$
Redbank	NSW	0.15 (0.06-0.23)	0.08 (0.04-0.13)	3.76 (2.82-4.7)	2.8 (2.6-3.1)	$1.66 \times 10^{-5} \pm 75\%$
Wallerawang C	NSW	0.16 (0.07-0.25)	0.09 (0.04-0.15)	4.14 (3.1-5.17)	3.1 (2.9-3.4)	$1.76 \times 10^{-5} \pm 75\%$
Callide C Nett Off	QLD	0.14 (0.04-0.24)	0.08 (0.02-0.14)	3.6 (1.09-6.12)	5.1 (4.2-6)	$1.75 \times 10^{-5} \pm 75\%$
Callide	QLD	0.41 (0.11-0.72)	0.18 (0.04-0.31)	3.6 (1.09-6.12)	5.1 (4.2-6)	$1.75 \times 10^{-5} \pm 75\%$
Gladstone	QLD	0.15 (0.04-0.25)	0.08 (0.02-0.14)	3.81 (1.15-6.47)	5.4 (4.4-6.3)	$1.85 \times 10^{-5} \pm 75\%$
Kogan Creek	QLD	0.13 (0.04-0.22)	0.07 (0.02-0.13)	3.42 (1.03-5.81)	4.8 (4-5.7)	$1.66 \times 10^{-5} \pm 75\%$
Millmerran Power Plant	QLD	0.14 (0.04-0.24)	0.08 (0.02-0.14)	3.6 (1.09-6.12)	5.1 (4.2-6)	$1.75 \times 10^{-5} \pm 75\%$
Stanwell	QLD	0.43 (0.11-0.74)	0.18 (0.05-0.32)	3.7 (1.12-6.29)	5.2 (4.3-6.2)	$1.80 \times 10^{-5} \pm 75\%$
Tarong North	QLD	0.39 (0.1-0.68)	0.17 (0.04-0.3)	3.42 (1.03-5.81)	4.8 (4-5.7)	$1.80 \times 10^{-5} \pm 75\%$
Tarong	QLD	0.43 (0.11-0.74)	0.18 (0.05-0.32)	3.7 (1.12-6.29)	5.2 (4.3-6.2)	$1.66 \times 10^{-5} \pm 75\%$
Loy Yang B	VIC	0.17 (0.04-0.3)	0.07 (0.02-0.13)	3.01 (2.55-3.47)	1.8 (1.7-2)	$3.66 \times 10^{-5} \pm 75\%$
Loy Yang A	VIC	0.17 (0.04-0.3)	0.07 (0.02-0.13)	3.01 (2.55-3.47)	1.8 (1.7-2)	$3.66 \times 10^{-5} \pm 75\%$
Yallourn W	VIC	0.19 (0.05-0.33)	0.08 (0.02-0.14)	3.38 (2.86-3.9)	1.6 (1.5-1.7)	$4.12 \times 10^{-5} \pm 75\%$
Morwell	VIC	0.19 (0.05-0.33)	0.08 (0.02-0.14)	3.38 (2.86-3.9)	2.3 (2.1-2.5)	$4.12 \times 10^{-5} \pm 75\%$
Anglesea	VIC	0.17 (0.04-0.3)	0.07 (0.02-0.13)	3.01 (2.55-3.47)	2 (1.8-2.2)	$4.12 \times 10^{-5} \pm 75\%$
Hazelwood	VIC	0.21 (0.05-0.36)	0.09 (0.02-0.16)	3.69 (3.12-4.26)	2.5 (2.3-2.7)	$4.49 \times 10^{-5} \pm 75\%$
Northern	SA	0.16 (0.04-0.28)	0.07 (0.02-0.12)	2.9 (2.45-3.34)	1.7 (1.6-1.9)	$3.53 \times 10^{-5} \pm 75\%$
Playford	SA	0.16 (0.04-0.28)	0.07 (0.02-0.12)	2.9 (2.45-3.34)	1.7 (1.6-1.9)	$3.53 \times 10^{-5} \pm 75\%$

Table S2 Emission Intensity for gas fired power plants (units of kg/MWH: Emission (kg) per unit of electricity produced in MWH).

Power Station	Region	PM ₁₀	PM _{2.5}	SO ₂	NO ₂
Colongra	NSW	0.036 (0.024-0.048)	0.036 (0.024-0.048)	0.003 (0.002-0.004)	0.664 (0.449-0.879)
Uranquinty	NSW	0.036 (0.024-0.048)	0.036 (0.024-0.048)	0.003 (0.002-0.004)	0.664 (0.449-0.879)
Smithfield	NSW	0.028 (0.023-0.033)	0.028 (0.023-0.033)	0.002 (0.002-0.003)	0.518 (0.43-0.606)
Tallawarra	NSW	0.023 (0.019-0.027)	0.023 (0.019-0.027)	0.002 (0.001-0.002)	0.425 (0.353-0.497)
Somerton	VIC	0.048 (0.032-0.064)	0.048 (0.032-0.064)	0.004 (0.003-0.005)	0.885 (0.598-1.172)
Bairnsdale	VIC	0.034 (0.023-0.045)	0.034 (0.023-0.045)	0.003 (0.002-0.004)	0.625 (0.422-0.827)
Jeeralang A	VIC	0.05 (0.034-0.066)	0.05 (0.034-0.066)	0.004 (0.003-0.005)	0.923 (0.624-1.223)
Jeeralang B	VIC	0.05 (0.034-0.066)	0.05 (0.034-0.066)	0.004 (0.003-0.005)	0.923 (0.624-1.223)
Mortlake	VIC	0.036 (0.024-0.048)	0.036 (0.024-0.048)	0.003 (0.002-0.004)	0.664 (0.449-0.879)
Valley Power	VIC	0.048 (0.032-0.064)	0.048 (0.032-0.064)	0.004 (0.003-0.005)	0.885 (0.598-1.172)
Laverton North	VIC	0.038 (0.026-0.051)	0.038 (0.026-0.051)	0.003 (0.002-0.004)	0.708 (0.479-0.937)
Newport	VIC	0.035 (0.024-0.046)	0.035 (0.024-0.046)	0.003 (0.002-0.004)	1.058 (0.715-1.401)
Barcaldine	QLD	0.041 (0.028-0.054)	0.041 (0.028-0.054)	0.003 (0.002-0.004)	0.759 (0.513-1.004)
Condamine A	QLD	0.024 (0.02-0.028)	0.024 (0.02-0.028)	0.002 (0.002-0.002)	0.443 (0.367-0.518)
Darling Downs	QLD	0.025 (0.021-0.029)	0.025 (0.021-0.029)	0.002 (0.002-0.002)	0.462 (0.383-0.54)
Swanbank B & E	QLD	0.025 (0.02-0.029)	0.025 (0.02-0.029)	0.002 (0.002-0.002)	0.452 (0.375-0.529)
Townsville	QLD	0.025 (0.021-0.029)	0.025 (0.021-0.029)	0.002 (0.002-0.002)	0.462 (0.383-0.54)
Yarwun	QLD	0.034 (0.023-0.045)	0.034 (0.023-0.045)	0.003 (0.002-0.004)	0.625 (0.422-0.827)
Braemar	QLD	0.038 (0.026-0.051)	0.038 (0.026-0.051)	0.003 (0.002-0.004)	0.708 (0.479-0.937)
Braemar 2	QLD	0.038 (0.026-0.051)	0.038 (0.026-0.051)	0.003 (0.002-0.004)	0.708 (0.479-0.937)
Oakey	QLD	0.035 (0.024-0.046)	0.035 (0.024-0.046)	0.003 (0.002-0.004)	0.644 (0.435-0.852)
Roma	QLD	0.038 (0.026-0.051)	0.038 (0.026-0.051)	0.003 (0.002-0.004)	0.708 (0.479-0.937)
Mintaro	SA	0.041 (0.028-0.054)	0.041 (0.028-0.054)	0.003 (0.002-0.004)	0.759 (0.513-1.004)
Quarantine	SA	0.036 (0.024-0.048)	0.036 (0.024-0.048)	0.003 (0.002-0.004)	0.664 (0.449-0.879)
Osborne	SA	0.027 (0.023-0.032)	0.027 (0.023-0.032)	0.002 (0.002-0.003)	0.506 (0.42-0.592)
Pelican Point	SA	0.024 (0.02-0.028)	0.024 (0.02-0.028)	0.002 (0.002-0.002)	0.443 (0.367-0.518)
Torrens Island	SA	0.038 (0.026-0.051)	0.038 (0.026-0.051)	0.003 (0.002-0.004)	1.404 (0.949-1.859)
Torrens Island 2	SA	0.036 (0.024-0.048)	0.036 (0.024-0.048)	0.003 (0.002-0.004)	1.316 (0.89-1.743)
Hallett	SA	0.048 (0.032-0.064)	0.048 (0.032-0.064)	0.004 (0.003-0.005)	0.885 (0.598-1.172)
Dry Creek	SA	0.044 (0.03-0.059)	0.044 (0.03-0.059)	0.003 (0.002-0.005)	0.817 (0.552-1.082)
Ladbroke Grove	SA	0.038 (0.026-0.051)	0.038 (0.026-0.051)	0.003 (0.002-0.004)	0.708 (0.479-0.937)
Bell Bay Three	TAS	0.04 (0.027-0.053)	0.04 (0.027-0.053)	0.003 (0.002-0.004)	0.732 (0.495-0.97)
Bell Bay Three 2	TAS	0.041 (0.028-0.054)	0.041 (0.028-0.054)	0.003 (0.002-0.004)	0.759 (0.513-1.004)
Tamar Valley	TAS	0.024 (0.02-0.028)	0.024 (0.028-0.002)	0.002 (0.002-0.443)	0.367 (0.518-0.02)

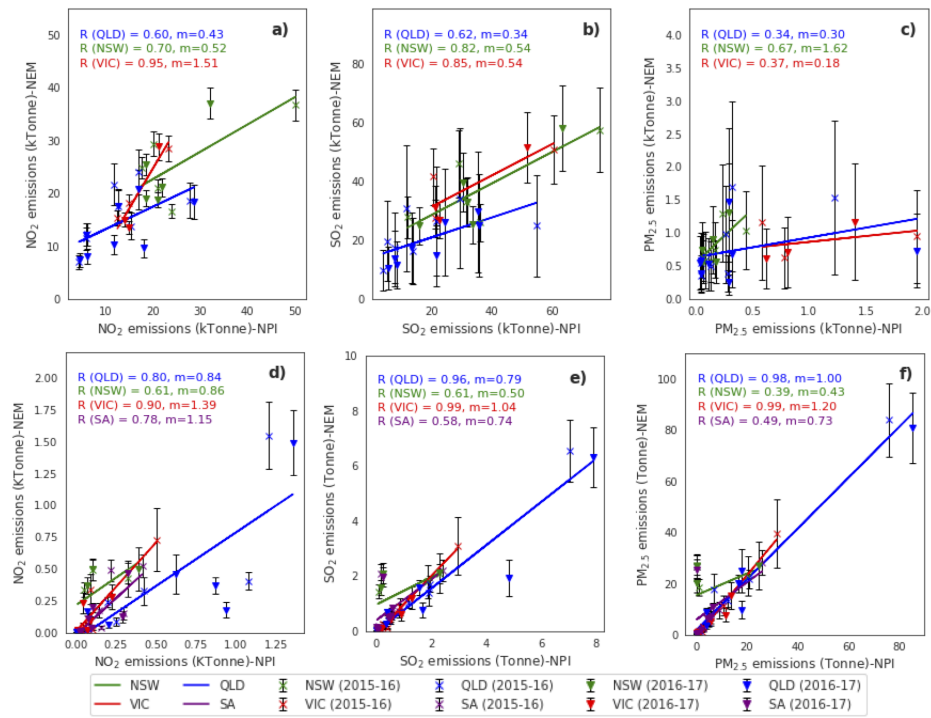


Fig. S1 State-wise comparison between estimated emissions from coal (a - NO₂, b - SO_x and c - PM_{2.5} emissions) and gas (d - NO₂, e - SO₂ and f - PM_{2.5} emissions) fired power plants using this study's methodology (NEM) and NPI (National Pollution inventory) for the financial year 2015-16 and 2016-17. Here, R represents the correlation coefficient and m refers to the slope of the regression curve.

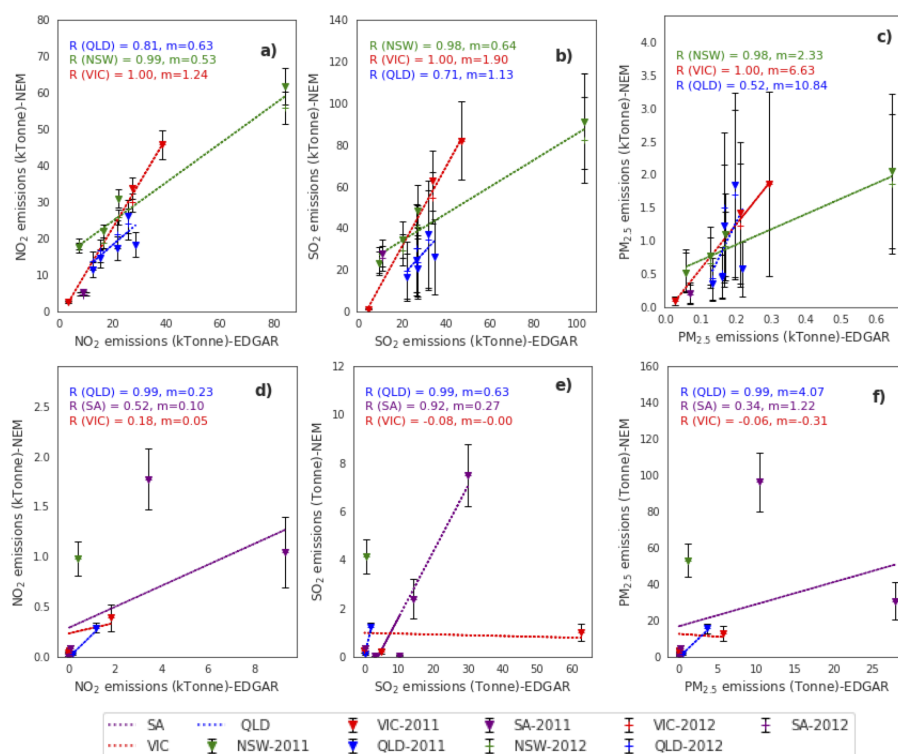


Fig. S2 State-wise comparison between estimated emissions from coal (a - NO₂, b - SO_x and c - PM_{2.5} emissions) and gas (d - NO₂, e - SO₂ and f - PM_{2.5} emissions) fired power plants using this study's methodology (NEM) and EDGAR for the year 2011 and 2012. Here, R represents the correlation coefficient and m refers to the slope of the regression curve.