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Environmental impacts of coal combustion: A risk approach to assessment of emissions

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ABSTRACT

This paper summarises some of the work performed in the Cooperative Research Centre for Coal in Sustainable Development (CCSD) on emissions from current power generation. A comprehensive approach was taken in the CCSD program to assessing environmental issues of concern for the power, and by implication the coal, industries. Here results of sampling on full scale operating plants are described, and detailed data on emission fluxes, particle size distributions, trace element concentrations as a function of particle size, and speciation of the trace elements are illustrated. The results show that particle capture in electrostatic precipitators (ESPs) is significantly less efficient than in fabric filters (FFs), particularly for submicron material, and that significant enrichment is observed in the finer particle sizes emitted from both ESPs and FFs. Results for the speciation of chromium, arsenic and selenium in coals, bottom ash and fly ash are also presented. The majority of chromium in fly ash is present in the less toxic Cr^{3+} form. Speciation of arsenic in feed coals is variable but the dominant form of As in fly ash is the less toxic As^{5+}.
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1. Introduction

Environmental regulations and agreements, enacted at both the national and international level, present a significant challenge to the future viability and operations of the coal and utility industries. Technological and political initiatives to mitigate or adapt to greenhouse gas emissions are likely to have a significant impact. It is likely that pressures to reduce emissions of carbon dioxide and pollutant gases and particles will intensify.

Electric power producers will also seek to significantly reduce emissions from conventional pulverised coal combustion and to develop new technologies such as integrated gasification combined cycle (IGCC), oxy-firing and pressurized fluidized bed combustion (PFBC) with inherently lower emissions.

Environmental concerns are already presenting, and will continue to present, challenges to the coal industry in reducing emissions. Environmental concerns presently associated with the use of coal include:

- gaseous and particulate emissions produced in the combustion process, notably NO_x, SO_2 and toxic trace elements;
- emissions of CO_2 and the implications of such emissions for global warming;
- emissions to air, water and land from operations associated with the mining of coal and the subsequent disposal of ash and spoil.

The industry needs to be well positioned to respond to these challenges, particularly in a more competitive environment where other fuels, such as gas and biomass, are being seriously considered as alternatives to coal.

There are also air quality issues that have the potential to impact on existing and future markets for thermal coals. These include emissions of fine particles and toxic compounds, and impacts of industrial NO_x on regional air quality.

During the past seven years the Australian Cooperative Research Centre for Coal in Sustainable Development (CCSD) developed a detailed program of research on emissions from current power generation. A comprehensive approach was taken to assessing environmental issues of concern for the power, and by implication the coal, industries. The objectives included:

- Development of a database of pollutant emissions (gas, particulate and trace toxic compounds) from Australian power stations, utilising validated sampling and analytical procedures.
- Measurement of trace element release and transformations from selected Australian coals.
- Development of techniques (correlations, predictors and models) which relate emissions to coal properties, furnace design and operating conditions.

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- Use of data and techniques generated above to develop reduction strategies for the environmental impacts of pollutant emissions to land, air and water from coal utilisation in current power plants where this is assessed as being required.
- Assess the policy settings which may be necessary to achieve improvements in the sustainability of coal combustion.

In this paper results for trace element emissions from full scale power plants and for speciation of trace element emissions are summarised. Further details are available in the CCSD reports and publications from which these results are drawn.

2. Observations of trace element enrichment at full scale and development of size dependent emission factors

Accurate and representative reporting of emissions of trace toxic species from all sources is a high priority in order for their environmental impact to be assessed. There are at least three options which could be considered for the reporting of emissions:

- Direct measurements of emissions at the location required to report.
- Modelling of emissions based on coal properties and composition, high temperature chemistry and the performance of particle collection devices.
- Development of a database of emissions based on measurements at a large number of plants, and the calculation of emission factors for emissions from this database.

The last approach is the one that has been adopted by many national pollutant inventories, drawing heavily on available US data and presents the best opportunity, currently, to develop procedures for estimating emissions of trace species from coal combustion plant. However there are a number of problems which emerge when the available data are considered. These include:

- data quality;
- data accessibility;
- data variability;
- data quantity.

In part, these problems are related to the complexity of the processes by which emissions of trace elements occur. For instance, the more volatile trace elements may be emitted in the gas phase or enriched on the fine (sub-micron) particulate fraction, and hence escape capture by electrostatic precipitators or bag filters. Alternatively, trace elements may reside in the fly ash collected by gas cleaning devices or in the bottom ashes or slags. Their ultimate fate, in the latter case, will depend on the utilisation and/or disposal options chosen for the ash or slag, and in many cases will be determined by the leachability of the trace elements.

Further complexity arises due to post-combustion reactions and transformations of trace elements which can play an important role in determining their deportment in combustion [1,2]. For example, Seames and Wendt [3,4] have established a relationship between the concentration of solid phase arsenic, selenium and cadmium to calcium in supermicron particles, suggesting formation of trace element/Ca complexes. Interactions with iron have also been reported, and possible control strategies using sorbents have been investigated [5,6]. Current knowledge of these processes is incomplete, and modelling or estimation techniques, which account for all these effects are still in the process of development.

The US data for trace elements has been critically reviewed by Helble [2]. He shows that the US databases provide information on coal rank, ash content, sulphur content, trace element concentrations, coal higher heating value, trace element emissions rate, and particle emissions rate. Emissions of individual trace elements are reported based on the following relationship:

$$E_i = A_{i,in}(1-\eta_i) = C_i(1-\eta_i)/H \tag{1}$$

where E_i is the emission on a mass per fuel energy content basis; $A_{i,in}$ is the concentration of the trace element i at the inlet to the particle collection device (mass per unit fuel energy content); C_i is the concentration (mass fraction) of trace element i in the coal on an as-received basis; η_i is the capture efficiency of trace element i in the particle collection device; and H is the higher heating value of the coal on an energy content per unit mass basis.

The particle collection efficiency, η, of a particle collection device is defined [2] as:

$$\eta = 1 - PM_{out}/PM_{in} \tag{2}$$

where $PM_{out,in}$ is the particulate matter concentration (mass per unit heat input) at the outlet or inlet to the particle collection device.

PM_{out} can be expressed [2] in terms of coal parameters as:

$$PM_{out} = PM_{in}(1-\eta) = f_a(1-\eta)/H \tag{3}$$

where f_a is the mass fraction of ash in the coal on an as-received basis.

Combination of Eqs. (1) and (3) gives an expression for trace element emissions as a function of measurable parameters:

$$E_i = \frac{C_i PM_{out}(1-\eta_i)}{f_a(1-\eta)} \tag{4}$$

However the broad range of trace element emissions observed at different plant has led to the development of a modified version of Eq. (4):

$$E_i = a_i\left[\frac{(C_i PM_{out})}{f_a}\right]^{b_i} \tag{5}$$

where a_i and b_i are empirical factors.

Eq. (5) is the form recommended by EPRI for interpretation of the DOE and PISCES data (see [2] and references quoted there). It is also the basis for the equations used in the Australian National Pollutant Inventory (NPI) workbook,[1] but it should be recognised that this is an empirical approach, and one which does not allow for the enrichment of many trace elements in the fine particle sizes. As these particles are more difficult to capture in electrostatic precipitators, this simplification may be significant. Helble [2] has developed a model, which includes trace element concentrations as a function of particle size, and size dependent particulate capture efficiencies. Using this model he is able to show that the predictions of emitted concentrations of volatile trace elements such as arsenic and selenium can be improved.

At present, data for Australian coals and facilities is not extensive enough for the refinements incorporated in Helble's model, and the approach used in the NPI, and based largely on US data, should be the preferred method for reporting emissions.

It had been known since the work of Davison et al. [7] that the fine particle fraction of fly ash could be enriched in trace elements compared with the fraction of trace elements in the parent coal. This is due to the volatilisation of some elements in the boiler and their subsequent condensation in the cooler sections of the flue gas stream. There has been considerable work investigating these observations for a variety of electrostatic precipitators (ESP) stations burning different coals [2]. These studies have found different behaviour for different elements and their transport

[1] See http://www.npi.gov.au/handbooks/approved_handbooks/ffossilfuel.html, accessed 11th September 2008.

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