

SRC eucalypt combustion

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1. ABSTRACT

The combustion performance of stemwood from a purpose grown eucalypt, *E. nitens*, was evaluated. It had a high moisture content and was rich in bio-accumulated inorganics such as calcium and potassium. During combustion the inorganics underwent a complex set of transformations. Some of the major reactions were identified and shown to have important consequences in terms of overall combustion performance as judged by parameters including extent of ash slagging and fouling, and emissions of particulates, greenhouse gases and volatile toxic trace elements.

2. INTRODUCTION

As energy demands increase so does the need to ensure that new energy technologies offer improved efficiency and minimum environmental impact. Biomass combustion has the potential to play an increasing role in meeting these demands but variability in properties of biomass fuels is large and may significantly influence the efficiency and environmental impacts associated with its utilisation. In New Zealand it is estimated that woody biomass presently accounts for 4 to 5% of energy needs with the forest industry being the major consumer of woody biomass energy. It is expected that the energy requirements of the forest processing industry will increase over the medium term and this requirement, plus other environmental drivers, are likely to result in a steady increase in demand for bioenergy in this country.

It is becoming increasingly acknowledged that in order to understand biomass combustion and fully realise its potential benefits in the future energy mix it is essential to gain some fundamental knowledge of the relationship between fuel properties and combustion performance. Combustion performance may be evaluated in terms of a range of important efficiency and environmental parameters. These include the extent of ash deposition (slagging and fouling) — factors recognised [1-6] as crucial to process efficiencies and boiler design. Also included are environmental impact factors [7-10] such as particulate emission levels, emissions of toxic trace inorganics, and emission levels of greenhouse gases such as methane and nitrous oxide. Many of the observed efficiency and environmental parameters are largely determined by the nature and extent of chemical reactions occurring among the various inorganic phases present in the firebox [1,11-13]. The focus of this study was to identify some of the more significant of these and the mechanisms by which they exert their influence on combustion performance of a sample of stemwood from a purpose grown eucalypt, *E. nitens*.

3. EXPERIMENTAL

The stemwood sample of *E. nitens* was taken from a recently felled area. It was allowed to partially air dry and then hogged using a 25 hp hogger equipped with a screen and an opening of about 75 x 20 mm. The hogged material was mixed thoroughly and a screen analysis carried out using a Williams classifier.

Samples (50 to 60 kg) of the *E. nitens* were burned in a small scale combustor (Figure 1) of approximately 50 kW firing rate. Several grab samples of the feedstock fuel were taken during the course of the combustion trials, aggregated, mixed, sub-sampled and then analysed for a range of properties including moisture, ash, volatile and sulphur content using standard laboratory methods. Major ash constituent elements were determined by X-Ray Fluorescence (XRF) and trace element concentrations were measured by ICP-MS after microwave digestion of a sample finely ground in a zirconia ring mill.

Fuel was loaded into a hopper and fed by means of an auger into the furnace where it dropped 300 mm onto a dispersion cone. Primary air was supplied from beneath this cone in a way similar to that found in a Vekos stoker. No secondary air was used. A shallow restraining ring around the cone ensured that the fuel could not be blown away from the air supply. The combustor was mounted on a combustion rig comprising a convective cooling tube bank, stack, cyclone and associated sampling and monitoring equipment.

Furnace exit gases passed over the convective tube bank that controlled the stack sampling temperature. Particulate matter was removed by a high efficiency cyclone. Inlet air flow and stack gas flow were measured using orifice plates and thermocouples were used for temperature measurement. Flue gas was withdrawn and passed through an isokinetic quartz sampling train to a quartz disc filter where any remaining particulates were captured and weighed. The flue gas then entered a series of bubblers containing solutions to trap any volatile inorganic compounds remaining in flue gas. Included in the train was a solution of 5% nitric in 10% hydrogen peroxide to trap the toxic inorganics boron, arsenic and selenium, [14] and 4% potassium permanganate in 10% sulphuric acid [15] to trap mercury. Elemental concentrations of these species in the solutions were measured by ICP-MS and cold vapour atomic absorption.

Samples of flue gas were withdrawn from a point slightly below the isokinetic quartz line, passed through a drying tower and into a gas chromatography gas analyser where levels of oxygen, nitrogen, carbon monoxide, carbon dioxide, methane and nitrous oxide were measured. Flue gas was also drawn from a third sampling port, diluted with inert gas to lower dew point to below room temperature and passed to a pulsed UV fluorescence sulphur dioxide analyser.

After each run, samples of bottom and cyclone ash were recovered and weighed. Particle size sieve analyses were performed using a range of standard Endecotts sieves and ashes were analysed for major and trace elements using XRF and ICP-MS. Loss on ignition was measured by weighing an oven dry sample of ash in a porcelain crucible and igniting to constant weight at 570°C. Samples from a fouling probe inserted immediately prior to the convective tube bank were also recovered and weighed.

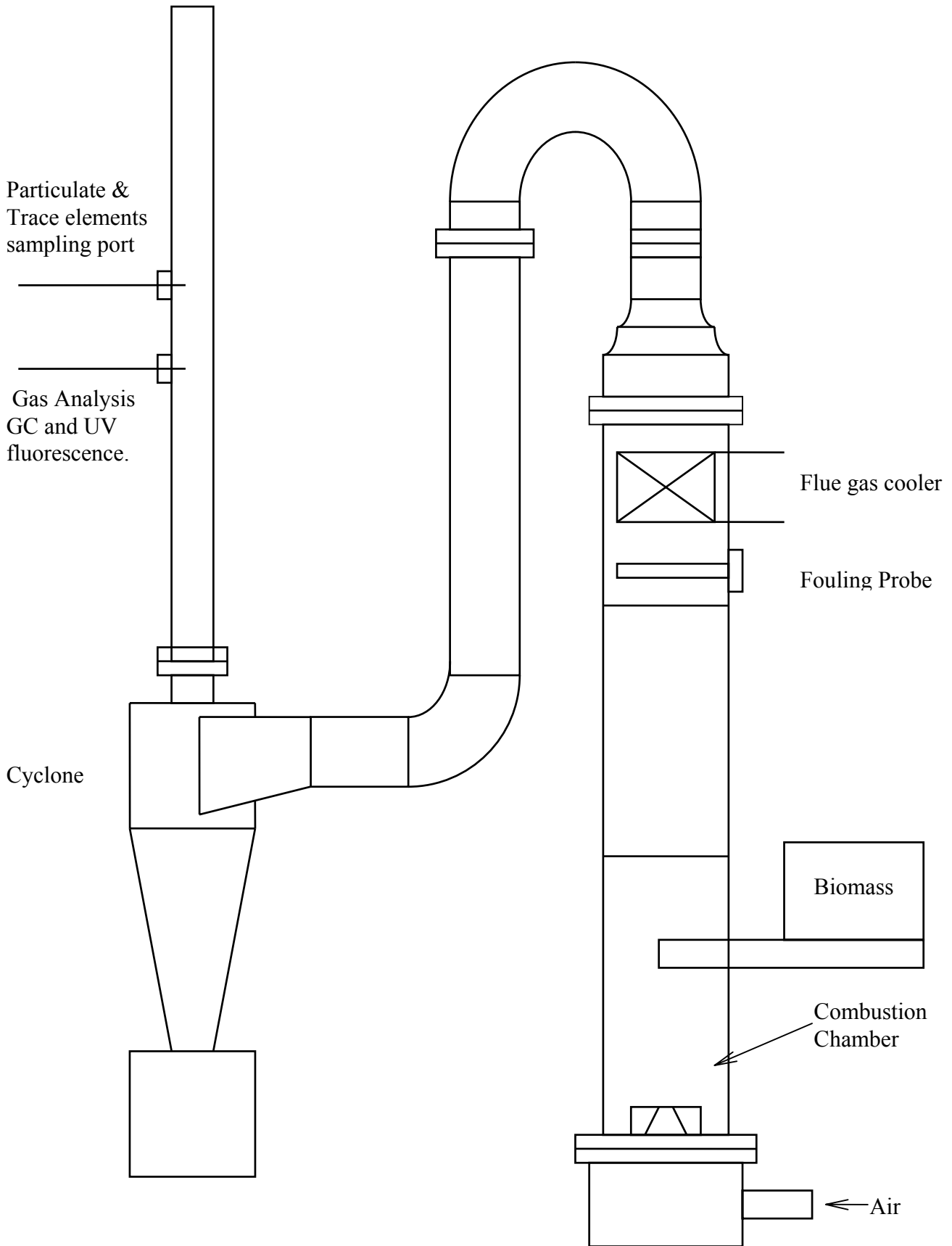


Figure 1. Biomass combustion rig

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A representative sample of the fuel was treated in an oxygen plasma to remove the organic material and generate low temperature ash (LTA) - a product intended to retain as closely as possible the inorganic material and crystalline phases present in the original fuel. The LTA was heated at 10°C per minute from ambient to 1100°C under reducing conditions in the sample pan of a differential thermal/thermogravimetric analyser (DTA/TGA) to determine temperatures where significant ash changes were taking place as evidenced either by weight losses and/or thermal events. Samples of LTA were then heated at 10°C per minute in a tube furnace to a series of temperatures chosen so as to fall either above or below points where ash changes were occurring. The resultant ashes were examined by Fourier Transform infra-red spectroscopy (FT-ir) and X-Ray Diffraction (XRD) in order to identify mineralogy changes undergone.

4. RESULTS AND DISCUSSION

Properties of the *E. nitens* fuel are shown in Tables 1 to 4. As received and fed, the fuel had a fairly high moisture content and low calorific value and the ash was dominated by bio-accumulated components such as calcium and potassium. The combustion data is shown in Tables 5 to 7.

Table 1. *E. nitens* properties

<i>Proximate analysis (wt %)</i>	
Moisture	51.4
Ash	0.69
Volatiles	39.1
Fixed carbon	8.8
Calorific Value (MJ/kg)	8.37
<i>Ultimate analysis (% dry basis)</i>	
Sulphur	0.02
Carbon	49.58
Hydrogen	5.82
Nitrogen	0.20
Oxygen (by difference)	42.96

Table 2. *E. nitens* ash properties

<i>Ash constituents (% oxide in ash)</i>	
SiO ₂	7.3
Al ₂ O ₃	5.0
Fe ₂ O ₃	8.2
CaO	36.3
MgO	8.5
Na ₂ O	3.8
K ₂ O	19.7
TiO ₂	0.35
MnO	1.9
Ash Fusion (redn, soft, °C)	1430

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Table 3. *E. nitens* particle size (wt % retained at size)

32 mm	4
19 mm	14
13 mm	35
6 mm	31
3 mm	6
< 3 mm	9

Table 4. *E. nitens* trace element concentrations (ppm in feedstock)

Arsenic	0.35
Boron	3.5
Selenium	0.11
Mercury	0.15
Chlorine	4900

Table 5. Combustion indicators

Total fuel consumed (kg)	68.5
Total bottom ash (kg)	0.495
Bottom ash > 1 mm (%)	Small
Unburnt carbon in bottom ash (%)	0.7
Total cyclone ash (kg)	0.044
Unburnt carbon in cyclone ash (%)	13.4
Total fouling probe ash (g)	0.90
Particulate emissions (mg/m ³ at 12% CO ₂)	191
Stack temperature (°C)	179

Table 6. Partitioning of selected volatile toxic trace inorganics

	Conc in bottom ash (mg/kg(%))	Conc. in cyclone ash (mg/kg (%))	Conc in vapour (µg/m³ (%))
Boron	260 (55)	720 (13)	63 (10)
Arsenic	39 (80)	58 (10)	0.1 (< 1)
Selenium	0.3 (2)	8.5 (12)	0.5 (3)
Mercury	nd	nd	12.7 (52)

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Table 7. Flue gas composition*

	<i>E. nitens</i>
Carbon dioxide (%)	10.6
Carbon monoxide (ppm@12% CO ₂)	2450
Oxygen (%)	10.5
Sulphur dioxide (ppm@ 12% CO ₂)	2.3
Nitrous oxide (ppm @ 12% CO ₂)	16
Methane (ppm @ 12% CO ₂)	86

* = Mean reading over course of experiment.

4.1 Inorganic transformations accompanying heating of *E. nitens* derived LTA

The LTA derived from *E. nitens* LTA was dominated by amorphous material and whewellite – mono-hydrated calcium oxalate – a phase commonly associated with plant material. A small silica signal was seen and no signals attributable to ordered aluminosilicate clay material were observed. Small amounts of potassium were observed in the form of arcanite (K₂SO₄) and nitre (KNO₃).

The combined FT-ir, XRD and thermal analysis investigation identified the following sequence of reactions on heating. Whewellite lost its water of crystallisation below 200°C and by 500°C had disappeared to be replaced by an XRD pattern consistent with a mixed potassium/calcium carbonate. Also present was sylvite (KCl) along with minor contributions from calcite (CaCO₃) and arcanite. By 1050°C the mixed carbonate had disappeared. So too had the calcite. It had been replaced by lime (CaO). A little oldhamite (CaS) and periclase (MgO) were identified. There were no signs of onset of melting.

Although the ash was low in aluminosilicates there was nevertheless a significant corresponding FT-ir signal and changes were seen between 500 and 1050°C. Most notable was the disappearance of the silica signal and formation of calcium aluminosilicates¹⁴ from the reaction of alumina and silica phases with calcium oxide - a reaction that becomes increasingly prevalent above 950°C. Because there was insufficient silica and alumina to react with all of the calcium oxide, a considerable quantity of this material remained at 1050°C.

Other complex transformations were undoubtedly taking place and several small and unassigned FT-ir and XRD peaks were observed. It is nevertheless informative to consider how these few identified chemical changes play a major role in influencing the combustion performance of the *E. nitens*.

4.2 The influence of identified inorganic transformations on slag formation

In terms of slag formation - ash deposition in the sections of the combustor close to the flame — the most significant ash chemistry relates to whewellite breakdown and subsequent reactions involving calcium. Although this includes formation of significant quantities of calcium oxide — a well known fluxing agent — the low firebox temperatures associated with combustion of this high moisture content fuel and the

small quantities of alumina and silica associated with the sample inhibit slag formation by this means. It was difficult to measure firebox temperatures accurately but these were consistent with a firebox temperature of below 950°C — the temperature at which calcium aluminosilicate formation becomes apparent. Essentially all of the bottom ash recovered from combustion of the *E. nitens* fuel passed through a 1mm sieve.

4.3 The influence of identified inorganic transformations on fouling formation

Fouling buildup is initiated in the cooler convective heat transfer sections of the plant by the deposition of a thin layer of material made up of condensed vapours. It usually involves the release of alkali metals (sodium and potassium) from the firebox in the form of oxides and chlorides which react primarily with sulphur gases to form alkali metal sulphates. These impact with the probe as sticky condensation products which then capture a high proportion of any subsequent impacting particles [1,13]. The formation of fouling may be inhibited by the presence of kaolinite, [19-23] a clay material, commonly found associated with coal deposits, that reacts with alkali metal oxides and prevents them leaving the firebox. These factors suggest that *E. nitens* with its high concentrations of potassium and chlorine and absence of observable quantities of kaolinite may be susceptible to fouling behaviour. Although it is low in sulphur concentration, calculations show that were all the sulphur in the feedstock to be released in the flue gas there may be sufficient quantities present for fouling deposition to occur. That it did not do so relates to the fact that a significant amount of the sulphur was captured by calcium oxide and retained in the bottom ash (see below).

4.4 The influence of identified inorganic transformations on partitioning behaviour of selected toxic volatile trace inorganic

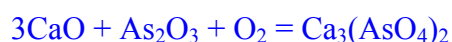
4.4.1 Boron. The transformations taking place between the major inorganics in the combustion zone not only affect the extent and type of ash deposition. They may also have a significant influence on the partitioning behaviour and emission levels of toxic volatile trace inorganic constituents. For example in combustion studies of high calcium sub-bituminous New Zealand coals under stoker conditions it is found that boron, which is normally regarded as either a Class III (vaporisation, non-condensation element) or Class II element (vaporised but condensed downstream and concentrated on fine grained particulates) [27] may show little of the expected partitioning between bottom ash and fly ash [28,29] with as little as 5% of the boron being recovered downstream of the combustor. This was ascribed to the reaction of calcium oxide with active alumina and silica phases to form calcium aluminosilicate matrices which are known [30,31] to have a very high affinity for boron incorporation. The reaction becomes increasingly prevalent as temperatures rise above 950°C [29,32].

The *E. nitens* sample has some of the requirements necessary to bring about boron capture in the bottom ash. There should be an abundance of calcium oxide as a result of the breakdown of the mixed potassium calcium carbonate and calcite. Alumina and silica phases are present although it is not certain if they are as active as those associated with coal combustion where they almost invariably arise in a highly disordered form from kaolinite dehydration and rearrangement. It is also unlikely that the temperature in the firebox reaches or maintains the 950°C required for significant calcium aluminosilicate formation. As a result, the boron behaves more like a typical

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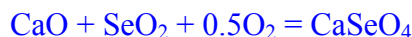
Class II volatile with a significant proportion (23%) being recovered downstream from the combustor either on cyclone ash or associated with flue gas (Table 6).

4.4.2 Arsenic. Arsenic is also usually classified as a Class II volatile. It leaves the firebox and is recovered primarily in association with fly ash particles from the colder parts of the system or, if the stack sampling temperature remains in excess of 193°C (the sublimation point of As₂O₃) a considerable percentage may end up in the stack gases [33]. The elevated moisture content of the *E. nitens* resulted in low temperatures at all points throughout the system including the stack sample point where a temperature of only 179°C was maintained. Essentially all of the arsenic volatilised from the firebox condensed onto fine particulates and less than 1% of the arsenic was recovered downstream of the firebox. In addition, at temperatures in excess of 500°C arsenate forming reactions such as



are known to occur [34]. Given that significant quantities of CaO are being generated during combustion it is highly probable that this reaction is occurring and further inhibiting arsenic release.

4.4.3 Selenium. It is well known that it is notoriously difficult to achieve good mass balances for selenium [34]. This study was no exception – recovery was poor (22%). Nevertheless it may be noted that some of the recovered selenium was found in the bottom ash, probably as a result of



analogous to the well established mechanism for sulphur retention (see below). The formation of calcium selenate [35,36] has been observed in studies on high calcium sub-bituminous coals and it appears that the high calcium ash of the *E. nitens* is allowing this behaviour to be mimicked .

4.4.4 Mercury. Mercury is well established [27,36] as a Class III volatile element. It behaved as expected with all recovered mercury being associated with flue gas.

4.4.5 Particulate emission levels. The concentration of particulates recovered from the flue gas sampling line (191 mg/m³) is a high result for this combustion rig. Typically, results of less than half this quantity are obtained. The increase is attributable mainly to the high moisture content of the fuel. It leads to less than optimal combustion conditions and generation of larger quantities of high carbon low-density soot particles than usual. These readily escape the cyclone which is designed for capture of higher density inorganic material.

4.4.6 Flue gas composition. The high moisture content of the *E. nitens* and the negative impact on combustion was also clearly shown in the comparative flue gas compositions. Appreciable quantities (average 16 ppm) of N₂O were observed – these are usually below 2 ppm for drier biomass fuels and coals. This is primarily a consequence of the low firebox temperatures achieved in the *E. nitens* burn. It is generally accepted [38] that increasing firebox temperatures leads to increased concentrations of hydrogen and

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hydroxy radicals in the gas phase and that both of these species very effectively reduce N_2O to N_2 .

Higher moisture content is also primarily responsible for the observed high CO and methane levels. These are well known indicators of poor combustion performance and are commonly found with high moisture content fuels. It should be noted that the CO levels remain consistently high throughout the burn whereas the levels of methane, the more readily combustible of the two gases, vary widely with a mean value of 86 ppm but at times peaking in excess of 250 ppm. This suggests that temperatures and conditions within the firebox are, on occasion, sufficient to combust much of the methane.

As mentioned above when considering chemical factors relating to fouling deposits, the emission levels of SO_2 were very low. Calculations show that approximately 10% of the total amount of available sulphur was escaping in the flue gas and the majority of the remainder was accounted for among the bottom and fly ashes. Given that lime is being generated during *E. nitens* combustion this is not too surprising. The well known [3,24,25] sulphur capture reaction



is likely to occur. Although the oxide has a preference for incorporation into silicate structures the low firebox temperatures and small concentrations of silicon and aluminium inhibit this process. The oxide remains available for sulphur capture. If temperatures in the firebox become too high [26] SO_2 may begin to be released again according to the reaction



However this reaction is not favoured as the high moisture content of the *E. nitens* fuel ensured that firebox temperatures remained low throughout the run. Indeed, as mentioned above, it appears that the conversion of calcium through to the oxide is only partial. It is nevertheless sufficient to capture much of the SO_2 liberated during combustion of this low sulphur fuel.

5. CONCLUSIONS

The chemical reactions occurring between inorganic species in the firebox have a significant impact on the combustion behaviour. The extent to which these reactions may proceed is strongly influenced by firebox temperature which is in turn influenced by the moisture content of the fuel. As can be seen from this present example, the interplay of factors can be very complex.

The sample considered has a high moisture content and, in all likelihood efforts would be made to lower this prior to combustion at an industrial site. If the *E. nitens* used in this evaluation is typical of a purpose grown crop (i.e., high levels of bio-accumulated species such as calcium and potassium and low silica and alumina) it may be expected that drying will lead to improved efficiencies and reduction of particulate emission levels and greenhouse

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gases. Reduced moisture content may lead to increased temperatures and thermal intensities within the firebox and it should be noted that this, coupled with the formation of a fluxing agent— calcium oxide – may encourage the onset of slagging behaviour. Off-setting this is the low level of aluminates and silicates available for fluxing. It is worth noting that in a subsequent series of combustion trials in which moisture content of the *E. nitens* was reduced to 27%, little increase in slag formation was observed while the expected improvements in efficiencies and reductions in emissions of particulates, greenhouse gases and toxic volatile trace metals were observed.

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