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Emissions Performance of High Moisture Wood Fuels Burned in a Residential Stove

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Abstract

A study has been made of the effect of fuel moisture content on emissions from a wood burning domestic stove. Two fuel types were studied: beech which is a hardwood, and spruce which is a softwood. The moisture contents investigated were for a freshly felled wood, a seasoned wood and a kiln dried wood. The effect of the moisture measurement method was considered using a commercial electrical conductivity probe moisture meter which was compared with laboratory analysis by drying in an oven at 105°C. It was shown that the probe can significantly underestimate the actual moisture content in certain cases. Correlations were made of the burning rate, the Emission Factors for the formation of gaseous and particulate pollutants as a function of the moisture content. Also studied were the ratio of Black Carbon to Total Carbon (BC/TC). For this stove NO_x was only dependent on the fuel-nitrogen content and not on the moisture content.

Keywords

combustion, wood stove, moisture content, particulates

1. Introduction:

The use of wood as a fuel is widely used throughout the world, although the extent varies from country to country depending on their energy requirements and the supply situation. This trend in wood usage has increased in recent years and has resulted in high particulate and air pollution emissions within urban locations with consequential adverse effects on human health [1-2] and on climate forcing [3]. The same situation applies to the use of cookstoves in developing countries where often less attention is paid to the quality of the fuel. There is increasing concern over the impacts of wood stoves on air quality in the UK and worldwide, especially in terms of particulate pollution.

Recent improvements in stove design have resulted in a potential reduction in air pollution by improved control over the supply of air in the primary and secondary combustion chambers. However, increased particulate and pollution formation will result during cold-starting and reloading of fuel (refuelling). Thus lower emissions are dependent on operational practices but also on the fuel properties [4-6]. The use of pellet stoves and boilers and wood pellets with suitably defined fuel specifications has greatly improved the environmental impact in regions where it is economical to use these processed fuels. However in many cases logs are used and there are inadequacies in fuel properties with regard to size and moisture content (MC). High moisture contents contribute to limited thermal performance and enhanced pollutant emissions following slow or incomplete combustion, and some studies have been made of this aspect [6-9] .

The effect of biomass moisture content on combustion and emissions has been the subject of a number of investigations. However, a difficulty is that it can vary depending on the initial moisture content in the wood and the humidity of the surrounding air. In the case of logs, moisture can be lost or gained via the end grain so that not only is it being lost, or gained, but the moisture is unevenly distributed [10]. There is an issue that the practical

definition of moisture content into loosely defined categories-such as kiln dried, seasoned and freshly harvested, is at present insufficient to define the use of wood as a fuel. Some schemes have been implemented in which the wood is certified to have a moisture content less than 20%, but so far there is still limited data available from real stove studies about their impact.

A number of papers have studied emissions from the combustion of biomass with various moisture contents. Shen et al [11] observed higher emission factors for smoke and polycyclic aromatic hydrocarbons (PAH) for fuels with high moisture content. Bignal et al. [12] identified greater PAH and CO emissions in biomass fuels with high moisture content. It was also observed that the combustion of dry biomass may result in rapid burning resulting in excessive consumption of oxygen within the stove and consequential production of greater smoke concentrations. Chen et al. studied the effect of moisture on wildfires [13], however these combustion systems are different to that in a domestic stove.

In this study, the authors address the gaps in knowledge by measuring emissions from a wood burning stove which approximates real world conditions. The study determines the effects of the fuel moisture content on the burning rate and emission factors, using batch feeding in a single combustion chamber stove. In particular the work examines the particulate size and composition in terms of black carbon (BC), total carbon (TC) and brown carbon as well as organic carbon (OC). Wood stove users are often reliant on ‘probe’ moisture analysers for determining wood moisture content, the results given by one of these, an electrical conductivity measurement are compared against laboratory analysis using a drying oven.

2. Material and methods

2.1 Fuel Preparation

Two species of wood, beech (a hard wood) and spruce (a soft wood) were supplied as logs (by Certainly Wood, Hereford, UK) and each wood was supplied with three moisture contents (MC%); they were designated as (i) fresh cut, expected %MC >40%, (ii) seasoned, expected %MC 25-35% and (iii) kiln dried, expected <20% MC.

The logs were prepared as follows. The fuels were stored as 2.5 m cords in-situ for a period of weeks following felling and these provide the fresh cut samples. Following removal from the woodland, each of the cords were cut into 25cm logs and seasoned by storing outdoors for 24-48 days, noting that the weather had been frequently damp. The kiln dried logs were prepared by heating in a kiln for a period of 50 hours at ~100°C. After receipt, the logs were kept in a covered, outdoor stores. The bark was removed prior to all combustion experiments and laboratory analysis, and for combustion studies the dimensions were approximately 25 cm x 7 cm.

The wood moisture contents were determined using two techniques. A handheld two pin probe digital moisture meter (Dr Meter MD-812) was applied to both lengths of the 'end grain' and in two locations across the exposed centre of the log. The use of the moisture meter provides an approximate moisture concentration at a depth of 1-5mm depth, depending on the hardness of the wood (beech is much harder to penetrate). The meter is suitable for the range of 5-40% moisture content and is accurate to a resolution of 1% point. Measurements were made in a laboratory under conditions well within the operating specifications of 0-40°C and a relative humidity of 0-70%RH. The total moisture content was also determined using a technique based on the Oven Dry method (BS EN ISO 18134-1: 2015) on the logs being used for the combustion experiments. The temperature used during drying was such that the loss of volatile components, such as oils, in the wood is negligible.

2.2 Combustion Experiments

A fixed bed 5.7 kW maximum capacity Waterford Stanley Oisín multi-fuel domestic stove was operated under batch loading conditions previously described [14-16].

This approach approximates typical ‘real world’ conditions but in these experiments the flue gases from the stack entered a dilution tunnel. The stove was mounted on a balance in order to measure the burning rate. Sampling ports were positioned on the flue-stack 1.43m above the combustion chamber. A FTIR analyser (Gaset DX4000) with an integrated oxygen sensor was used for gaseous emission analysis. A pitot tube was used to measure the flue gas velocity. Particulate matter was sampled within the stack using a gravimetric method as described before, using a heated line at 120°C leading to a filter-block ($70 \pm 5^\circ\text{C}$) housing 50mm micro-quartz (Munktell) filter papers for sample collection for black carbon (EC) and organic carbon (OC) determination. Particulate matter (PM) concentrations and size-distribution were calculated post-dilution using a Dekati PM₁₀ Impactor set up to separate for $> \text{PM}_{10}$, $\text{PM}_{10} - \text{PM}_{2.5}$, $\text{PM}_{2.5} - \text{PM}_1$ and $< \text{PM}_1$. The flue-gas sampling rate for this was 10 ± 0.5 l/min and the temperature of the diluted air was 64°C. Greased 25mm aluminium foils were used for the collection of the particulate matter and an additional 47mm (Whatman GF/F) was placed at the base of the impactor for $< \text{PM}_1$ emission collection. The carbon monoxide concentrations pre and post-dilution were used to determine the dilution-factor in the dilution tunnel as CO is sufficiently unreactive at this temperature to permit it to be used in this way. Cross-contamination between experimental runs was minimised by removing soot from the stove and flue between experimental runs and cleaning the sampling probes and measurement equipment.

Each fuel was tested in duplicate, and each experiment consisted of two combustion cycles, a cold ignition cycle and a warm refuelling cycle. The cold start or ‘ignition batch’ was followed by a subsequent reload, this cycle was used for the measurement of gaseous and

particulate emissions during the phases, ignition, flaming and smouldering. Means of replicate tests are reported. Both fuel batches incorporated a measured fuel mass between 1.2 kg and 1.8 kg and ignition was achieved by means of kerosene-based firelighters (Zip High Performance) with a mass of 100g. Both firelighters and fuel were arranged in the stove in a reproducible geometrical arrangement for each of the experiments. Gaseous emissions were analysed by the FTIR using 60 s resolution. In addition, mass and differential pressure were measured at 120 s sampling resolution.

Emission factors (EF) for the different species (n), were determined as [species (n)] g/kg [fuel] using effectively the method described by Fachinger et al. [6] (although that paper uses thermal units). The emission factors obtained in this paper incorporate results over the whole combustion cycle and are sampled throughout the ignition, combustion and smouldering phases: initially they are obtained in mg/m³ dry flue gas at 0°C, 101.25 kPa and 13% O₂ and then converted to g/kg.

3. Experimental Results

3.1 Fuel analysis

Logs were selected in duplicate, split, milled and sieved to an approximate nominal top size of 1.0 mm. A representative sample was used for proximate and ultimate analyses using the appropriate British Standard methods, analysis was undertaken in triplicate and means are reported. Proximate analyses are given in Table 1 where the entries are on a dry basis and are accurate to ± 0.5 wt% and were obtained using milled samples. Ultimate analysis results given on a dry ash free basis and accurate to ± 0.2 wt% are also given in Table 1. The Higher Heating Values were calculated from the composition using the method given by Friedl et al [17]. They are accurate to $\pm 2\%$.

-Table 1 here-

As expected the three beech samples and the three spruce samples have similar elemental compositions. There are some variations due to the natural variability in samples taken from different parts of the tree; typically the moisture varies as discussed in the next section.

3.2 Fuel Moisture Contents (MC%):

The accurate determination of the moisture content in logs (MC wt%) is difficult and the two methods previously described were used. Firstly a commercial digital moisture probe was used as a method of estimating the fuel moisture content for each log consumed. This is the method typically available to stove users. Manual sampling of moisture content was determined across both the exposed centre of the log after splitting, which is the recommended measurement method, and within the end-grain. The results, some of which are at the extremity of the range of the meter, are summarised in Table 2. The errors shown there are the instrumental errors.

The second method was to use a drying oven in order to determine the total water content in the samples by heating whole logs at 105°C for 96 h; after this period of time the mass loss was constant within 2 %. This method is considered to give the actual moisture content for this research, but we estimate the overall error to be $\pm 5\%$. These results are shown in Table 2

-Table 2 here-

It is evident that the highest moisture values are those obtained by laboratory analysis using the Drying Oven method with the difference up to 100% between the two methods. The results for kiln dried logs are the only ones in agreement for the laboratory and probe tests. The outside measurements at a depth of 5mm were approximately similar, but the result for the end-grain were significantly lower. This is because the moisture in the logs is not

uniformly distributed throughout and this inhomogeneity is greater for the highest moisture logs that have tended to dry from the end surfaces first. The values used in this study for moisture content of the logs are those by the Oven Drying method. The differences between this and the outside measurements are significant and in the case of logs with high moisture content this will influence the combustion behaviour; the centre of the logs will remain cooler for much of the combustion cycle.

3.3 Burning rate studies

Each experimental run incorporated an ignition cycle, during which the ease of ignition was investigated, followed by a subsequent reload and emissions test cycle for particulate and pollutant emission analysis. Fuel consumption and burning characteristics were made at 120s increments and include visual recordings of the phase of combustion.

The cold start 'ignition batch' testing identifies significant difficulty in wet-fuel ignition when compared with the kiln-dried materials. Additional firelighter batches were often required following the end of flaming combustion while fuel material remained. Both seasoned and fresh-cut fuel presented difficulty in achieving fuel ignition; both kiln-dried beech and kiln-dried spruce required only the initial batch of firelighters. Typical results for the burning rate expressed as mass against time for beech wood samples are shown in Fig. 1; duplicate runs (numbered 1, 2) for the kiln dried, seasoned and freshly cut are given. The results for spruce are not shown but they behave in a similar way.

-Fig. 1 here-

The burning rates were quantified in terms of a characteristic burning time, t_b , of the exponential part of the combustion as $t_b = 1/b_c$, where b_c is the burning rate coefficient. The burning rate coefficient is a function of moisture, oxygen level, reactivity and the surface area. It is analogous to the function used in fluid bed combustion. The relationship between

moisture content and characteristic burning time (below) shows what one would expect: drier burns faster, but the relationship is more complex than just that. Fig. 2 shows the mathematical fit of the equations used in the case of all the samples.

-Fig.2 here-

Statistical analysis showed that the coefficients of determination, R^2 , for the exponential part of the curves are as follows: FCB: 0.998 and 0.999; KDB: 0.995 and 0.937; SB: 0.999 and 0.997; FCS: 0.961 and 0.997; SS: 0.999 and 0.964; KDS: 0.918 and 0.870. In most cases, the regression fit is good to very good. The exponential decay function is consistent with the expected mass loss during the period of consistent combustion. As a metric for comparing the behaviours of the various fuels, this is considered a reasonable criterion. It is recognised that the curve for KDS is a less good fit. However, we considered that they are not sufficiently divergent from the ‘exponential decay’ model to require altering model or the criterion which works well in all other cases.

Given the inherent variability in fuel moisture content, and resultant combustible fuel mass, a Characteristic Burning Time (CBT) (1/s), or the time required for the fuel batch to lose 37% mass, was used as a method of comparing burning rate independent of initial mass. Estimated values of CBT are plotted against fuel moisture content (%) so as to identify the effect of moisture content on burning rate. Fig. 3 presents the variability in CBT against moisture content (MC%) obtained following the application of linear regression and estimated moisture content at the time of the combustion experiment. Whilst the data points are few and scattered there is an apparent dependency in the cases studied here and this is indicated by applying a regression line. The linearity of the relationship should not be assumed from this and more evidence is required. The errors in the MC% are estimated to be $\pm 5\%$ and in the CBT $\pm 10\%$.

-Fig 3 here-

It is seen that the characteristic burning time increases with the moisture content but also that beech burns more rapidly than spruce and with approximately the same dependence on moisture content. Beech has only a slightly higher volatile content than spruce but as a hard wood has a significantly higher density (beech 721 kg/m³, spruce 450 kg/m³), and the thermal conductivity of beech is about three times that of spruce [18], although this does change as well with the moisture content. The increase in burning rate is consistent with the increased heat transfer rate due to the changes in the thermal conductivity.

-Fig. 3 here-

The temperature in the combustion chamber as a result of first gas phase flaming combustion and then smouldering bed combustion determines the characteristic burning times. These temperatures are reflected in the flue temperatures and the general behaviour has been observed by our previous studies with this stove and other research groups, most recently by Fachinger et al. [6]. At the beginning of the combustion cycle there is an initial increase in the burning rate and flue gas temperature resulting from the initial devolatilisation followed by combustion of the partially decomposed biomass and char during the smouldering phase. The emission of smoke is higher over this short initial flaming period.

There are differences in flue gas temperature for both beech and spruce fuels. Kiln dried fuels gave a much higher temperature, with peak conditions associated with a higher rate of combustion (Fig. 3). This is particularly the case during the early phases of combustion. The enhanced burning rates associated with the drier fuel types results in higher peaks in the flue gas temperature. Significant variation in temperature is identified between kiln dried and fresh cut fuels. Here kiln dried beech has initial flue gas temperatures of about 620°C declining to about 150°C at the end of the smouldering phase. In contrast, fresh cut beech has an initial temperature of about 325°C declining to the same final temperature, 150°C. Seasoned beech is intermediate in behaviour depending on the exact MC. Spruce,

which has a lower mass burning rate produces temperatures which are slightly lower during the initial flaming phase. Kiln dried spruce gives an initial peak temperature of 580°C eventually reducing to 150-180°C. Fresh cut spruce gives a higher temperature of 300°C and this is about halfway through the burning cycle; this is the result of the difficulty in the initial ignition process and the slow burning rate.

3.4 Gaseous Emissions Analysis

Emissions measurements were made for both the ignition batch and re-load batch cycles but only the reload batch results are used here. Data is presented in mg/m³ under a standardised condition and on a dry gas basis. The nature of the emission curves follows the conventional pattern observed by other research groups, for example, Fachinger et al. [6]. Mean values of the Emission Factors are given in Table 3 for all the fuels studied here. The estimated errors for the gaseous pollutants are $\pm 8\%$ and for the particulate matter (PM) $\pm 10\%$. It should be noted that trace species measured by FTIR and designated 'benzene' and 'CH₂O' may also include other similar species.

-Table 3 here-

The behaviour of some of the emission profiles from fuels with high moisture content does not follow the conventional pattern. The flaming combustion phase and the char combustion phase are far less distinct, and it was observed that some volatile material was released in spikes towards the end of the smouldering phase. The emissions of CH₂O and C₆H₆ can be measured throughout the cycle and there is an emphasis the emissions of unburned species towards the end of the cooler, smouldering phase. It is likely that the centre of wetter logs remains relatively cooler and unreacted biomass is 'trapped' there and decomposes later. The resulting low temperature pyrolysis of wood produces complex organic compounds such as PAH and Brown Carbon [15].

3.5 Particulate Matter Emissions

The results obtained by the Dekati Impactor in the dilution tunnel are shown in Fig. 4. Here the PM emissions are expressed in mass/m³ as measured directly by the Impactor. For each of the wood samples the experiments shown in Fig. 4 are in duplicate and the experimental errors are shown on the figures.

-Fig. 4 here-

It is seen that at the extremities of moisture content, i.e. very dry or high moisture -the beech produces more smoke than the spruce. The high particulate emission associated with the kiln dried wood was also associated with measurement of extremely low oxygen concentrations in the flue, indicating that the air flow was insufficient to permit complete combustion at this high burning rate. Beech was shown (Fig. 2) to burn more rapidly than spruce resulting in the different in the total particulate produced.

3.6 Particulate size distribution

The particle size distributions obtained using the Dekati Impactor are given in Table 4. It is clear that the majority (at least 92%) of the particulate matter on a mass basis is less than 1µm in diameter. The error here is ± 10%. The other fractions produced, PM₁-PM_{2.5}, PM_{2.5}-PM₁₀, >PM₁₀, are all less than 3 wt% which means that the mass collected is very small, and here the error is estimated to be ±30% The fact that most of the soot is < PM₁ has implications in relation to the number density of the soot and the effect on human health [1,19].

-Table 4 here-

3.7 Elemental Carbon/Total Carbon (EC:TC) Ratio

Particulate samples taken from the dilution tunnel using the Dekati Impactor for the PM_{10} fraction were collected on a filter paper and analysed for the ratio EC/TC, (Elemental Carbon/Total Carbon), by Sunset Laboratory Inc, a method used previously by us [15]. The results are given in Fig. 5 and Table 4. The experimental errors for EC/TC are $\pm 15\%$.

-Fig. 5 here-

It is apparent that the values for EC/TC are in the sequence kiln dried > seasoned > fresh cut, for both the beech and the spruce, but the values for spruce kiln dried are the highest than for beech. These results are slightly surprising because the fuel chemical analyses of the woods are similar; however the cellulose content for spruce and beech respectively are 45 and 37 wt %, for hemicellulose 27 and 35 and for lignin 27 and 21 %, which would explain the low values of EC/TC observed for beech (that is, high values of OC). The Modified Combustion Efficiency (MCE), which is given by the ratio $[\text{CO}_2]/[\text{CO} + \text{CO}_2]$, has been used as a measure of combustion inefficiency but more recently Pokhrel et al. [20] have shown that the ratio EC/OC provides a better correlation [20]. Since $\text{TC} = \text{EC} + \text{OC}$ then the data in in Fig. 5 shows the increase in combustion inefficiency as the moisture content increase for both woods. Shen et al have shown that there is a correlation between EF (PM) and OC [21].

Samples were also taken directly from the undiluted flue gases using the smoke meter and deposited on filter papers. Because the kiln dried beech burns the most rapidly, only four filters could be collected (one every 10 min); six filters were collected for the other two fuels for which the test lasted longer. It was seen for the kiln dried and the seasoned beech that the samples obtained during the initial flaming phase were black but those obtained during the later smouldering phase are slightly brown. These last samples are brown carbon as observed using this stove before [15]. Increasing the MC in the samples increases the combustion time

at lower fuel temperatures. Thus the Seasoned and Fresh Cut Wood samples produced increasingly greater amounts of 'Brown Carbon', and this is especially the case with the Fresh Cut wood, presumably caused by low temperature reactions inside the logs. This effect does not seem to have been observed before although Butcher et al. [22] observed that 42-77 mass % of wood fire soot could be extracted with benzene, and more recently Fachinger et al. [6] observed that the organics in PM₁ from wood stoves was 40%. Purvis et al. looked at organic speciation and concluded that the temperature at which the particle size sample is collected has a major impact on the measured distribution of organic material [23]. Burning high moisture wood has a deleterious effect on both environment and climate change because of both the increased emission of Black Carbon and also Brown and Organic Carbon.

4. Discussion

4.1 General features of the effects of moisture

A study has been made of the effects of moisture in lump firewood (small logs) under real world conditions, Firstly there are some interesting issues about measuring the moisture content because of the non-uniformity of the distribution of the moisture in the wood which causes uncertainties here as well as in the interpretation of emissions quoted in previous publications.

4.2 Comparison with other published results

The effects of moisture are complex affecting the burning rate and the formation of secondary combustion products. Emission factors are given for the gaseous and particulate species but there are only a few other studies in which the results can be directly compared. For example, Fachinger et al [6] made studies of EF for PM₁ from a wood stove but did not specifically give the moisture contents of the fuels studied and averaged results for a number

of woods which were in a narrow range; they also stated that very dry wood resulted in elevated particulate emissions. Other examples are Tihay-Felicelli et al., [24] using green waste, Chomane, et al. with rubber wood [25], Bahadori et al. with Bagasse [26] and Lu et al. using straw [27]. This work identified a reduction in total PAH combustion generated emissions from dried rice and bean straw following an increase in moisture content up to 30. This was attributed to there being a reduced combustion temperatures.

Of particular interest from both an environmental and a health point of view is the extent of the emission of particulate matter and associated organic matter. This has been the subject of a number of investigations, but there are difficulties in comparing experimental results from other research groups because of differences in the combustion units but also because of different methodologies used to collect sample. The particulate matter consists of carbonaceous soot particles together with a layer of PAH, associated particulate organic matter as well as fragments of ash and char. The amount of organic matter that is deposited on the collection filter depends on the temperature of the collection filter [28], which is 64°C in these experiments, but is often not specified. It is seen from the data for EC/TC in Table 4 that the amount of organic matter present is considerable, the values of EC/TC are low.

The results from the present experiments for the emission of total particulate matter from fuels with different moisture content are given in Fig.6. Also shown are data from other researchers where they have used stoves which have a similar design and method of operation to the present work. Data from Fachinger et al. [6] and Bafver et al. [29] as well as Mitchell et al. [14] using the same stove as the present experiments are given: in most cases results have had to be averaged. Shen et al. [11] have obtained data for different moisture levels (5-27%) for a range of woods (mainly Chinese Poplar) for a brick cooking stove but these are not included in Fig.6 because the stove type (designed for cooking) and PM sampling arrangements are different. However the results parallel our results in Fig. 6 but are ca. 20%

higher. NIWA [30] produced a report in 2007 showing that the in situ wet-weight particulate emission factor was 3.3 g/kg and was about three times smaller than the emission factor of 11 g/kg for pre-1994 wood burners. These results are constant over the range of 13-54 % moisture content. This is in contrast to the pre-1994 results which showed a strong parabolic variation with moisture content, and with the results in this paper and those of Shen et al. [11]. It raises an interesting issue about how the present results can be scaled and applied to other stove with different designs and thermal capacities.

-Fig. 6 here-

It is seen from present results in Fig. 6 and Table 4 the similarity of the behaviour of the particulate matter, benzene, which is an important intermediate in soot formation, formaldehyde and methane (which are indicators of the decomposition products from biomass). The EF for soot are in accord with previous investigations, namely that above 25% MC the smoke increases markedly [9, 29]. However this work indicates that below 5%MC here is no increase in smoke emission over the whole cycle, although there is for a short initial part of the cycle during ignition or refuelling. The main advantage of using very dry wood is because of the ease of ignition, but this comes at a cost, the energy required to dry the wood.

4.3 Effects on the combustion processes

Many studies have been undertaken with moisture contents between about 8 and 20%MC where the effect of moisture is not significant other than changing the heat available on a mass basis. The type of wood is also significant, whether it is hard or soft or indeed an agri-residue based fuel such as straw. Visually it is observed that if very dry log wood is ignited that this process occurs smoothly and transition to flaming combustion soon occurs. But

when the stove is refuelled with very dry wood it initially burns rapidly with the copious release of soot, and this process changes during the transition to flaming combustion. Very wet wood (>40% moisture) burns badly; moderately wet partially seasoned wood retains moisture within the wood which results in overlapping combustion stages as a result of delayed pyrolysis which forms decomposition products during smouldering, releasing CH₄, CH₂O, C₆H₆ and also forming brown carbon etc.

NO_x was not the primary focus of this study, since it is well established that at the low temperatures observed in this type of stoves, NO_x formation is dominated by fuel-N as discussed by Mitchell et al. [14] and the references listed in that paper. This is supported by the experimental results for spruce, which are within experimental error for all fuels. However, the results for beech suggest that moisture content may be having an impact on the nature of the emission of the nitrogen species. Both Chen et al. [13] and Shen et al. [21] found that moisture affects the partitioning of nitrogen between NO_x and NH₃, but further research is required to fully understand the impact of moisture on nitrogen partitioning amongst NO_x, NH₃ and N-PAH.

5. Conclusions

A study was undertaken to investigate the effect on the moisture content in the range 3.6 – 42.9 wt% on the combustion of wood logs in a domestic stove, and the following conclusions were made:

- i. The measurement of moisture in wood logs can lead to different results depending on the method used; analysis of the actual moisture contents determined by the Drying Oven method were different in some cases to those given by a handheld probe analyser. The results from the former were adopted.

- ii. Mass burning rates were a function of moisture content and wood type and have been quantified for the cases studied here.
- iii. The emissions of particulate matter over a whole combustion cycle were found to increase with increasing wood moisture in the range studied; this was paralleled by the emissions of benzene (and similar aromatic species) and formaldehyde.
- iv. Measurements of the ratio EC/TC (elemental to total carbon) indicate that the emissions of organic carbon decrease as the wood is drier.
- v. In all cases studied the particulate matter mainly consisted of PM₁. For both woods the PM₁ was higher for freshly cut wood than for kiln dried.
- vi. NO_x emissions were not seen to be affected by fuel moisture in this stove study, only by the fuel-N content.

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References

1. Bølling AK, Pagels J, Yttri KE, Barregard L, Sallsten G, Schwarze PE, Boman C. Health effects of residential wood smoke particles: the importance of combustion conditions and physicochemical particle properties. Part Fibre Toxicol 2009; 6: 29.
2. Orasche J, Schnelle-Kreis J, Schön C, Hartmann H, Ruppert H, Arteaga-Salas JM, Zimmermann R. Comparison of emissions from wood combustion. Part 2: Impact of

combustion conditions on emission factors and characteristics of particle-bound organic species and polycyclic aromatic hydrocarbon (PAH)-related toxicological potential. *Energy Fuels* 2013; 27: 1482–1491.

3. Bond TC, Doherty SJ, Fahey DW, Forster PM, Berntsen T, DeAngelo BJ, et al. Bounding the role of black carbon in the climate system: a scientific assessment. *J Geophys Res: Atmos* 2013; 118: 5380-5552.

4. L'Orange C, DeFoort M, Wilson B. 2012. Influence of testing parameters on biomass stove performance and development of an improved testing protocol. *E.S.D.* 2012; 16: 3-12.

5. Ozgen S, Caserini S, Galante S, Giugliano M, Morreale C. Emission factors from small scale appliances burning wood and pellets. *Atmos Environ.* 2014; 94: 144-153.

6. Fachinger F, Drewnick F, Giere R, Borrmann S. How the user can influence particulate emissions from residential wood and pellet stoves: Emission factors for different fuels and burning conditions. *Atmos Environ* 2017; 158: 216-226.

7. Shen GF, Wei S, Wei W, Zhang Y, Min Y, Wang B, et al. Emission factors, size distributions, and emission inventories of carbonaceous particulate matter from residential wood combustion in rural China. *Environ Sci Technol* 2012; 46: 4207-4214.

8. Orang N, Tran H. Effect of feedstock moisture content on biomass boiler operation. *Tappi Journal* 2015; 14: 629-637.

9. Magnone E, Park S-K, Park JH. Effects of moisture contents in the common oak on carbonaceous aerosols generated from combustion processes in an indoor wood stove. *Combust Sci Technol* 2016; 188: 982-996.

10. Jirjis R. Storage and drying of wood fuel. *Biomass Bioenergy* 1995; 9: 181-190.

11. Shen GF, Xue M, Wei S, Chen Y, Zhao Q, Li B. et al. Influence of fuel moisture, charge size, feeding rate and air ventilation conditions on the emissions of PM, OC, EC, parent

PAHs, and their derivatives from residential wood combustion. *J Environ Sci* 2013; 25: 1808-1816.

12. Bignal KT, Langridge S, Zhou JL. Release of polycyclic aromatic hydrocarbons, carbon monoxide and particulate matter from biomass combustion in a wood-fired boiler under varying boiler conditions. *Atmos Environ* 2008; 42: 8863-8871.

13. Chen L-W.A, Verburg P, Shackelford A, Zhu D, Susfalk R, Chow JC, Watson JG. Moisture effects on carbon and nitrogen emission from burning of wildland biomass. *Atmos Chem Phys* 2010; 10: 6617–6625.

14. Mitchell EJS, Lea-Langton AR, Jones JM, Williams A, Layden P, Johnson R. The impact of fuel properties on the emissions from the combustion of biomass and other solid fuels in a fixed bed domestic stove. *Fuel Process Technol* 2016; 142: 115-123.

15. Atiku FA, Mitchell EJS, Lea-Langton AR, Jones JM, Williams A, Bartle K.D. The impact of fuel properties on the composition of soot produced by the combustion of residential solid fuels in a domestic stove. *Fuel Process Technol* 2016; 151: 117-125.

16. Phillips D, Mitchell EJS, Lea-Langton AR, Parmar KR, Jones JM, Williams A. The use of conservation biomass feedstocks as potential bioenergy resources in the United Kingdom. *Bioresour Technol.* 2016; 212: 271–279.

17. Friedl A, Padouvas E, Rotter H, Varmuza K. Prediction of heating values of biomass fuel from elemental composition. *Anal Chim Acta* 2005; 554: 191-198.

18. Mason PE, Darvell LI, Jones JM, Williams A. Comparative study of the thermal conductivity of solid biomass fuels. *Energy Fuels* 2016; 30: 2158–2163.

19. Torveka T, Tissari J, Sippula O, Kaivosoja T, Leskinen J, Virén A, Lähde A, Jokiniemi J. Effect of wood combustion conditions on the morphology of freshly emitted fine particles. *Atmos Environ* 2014; 87: 65-76.

20. Pokhrel RP, Wagner NL, Langridge JM, Lack DA, Jayarathne T, Stone EA. et al. Parameterization of single-scattering albedo (SSA) and Absorption Ångström Exponent (AAE) with EC/OC for aerosol emissions from biomass burning. *Atmos Chem Phys* 2016; 16: 9549–9561.
21. Shen GF, Tao S, Wei S, Zhang Y, Wang R, Wang B, et al. Emissions of parent, nitro, and oxygenated polycyclic aromatic hydrocarbons from residential wood combustion in rural China *Environ Sci Technol* 2012; 46: 8123-8130.
22. Butcher SS, Sorenson EM. A study of wood stove particulate emissions. *J Air Poll Control Assoc* 1979; 29: 724-728.
23. Purvis CR, McCrillis RC, Kariher PH. Fine particulate matter (PM) and organic speciation of fireplace emissions. *Environ Sci Technol* 2000; 34: 1653-1658.
24. Tihay-Felicelli V, Santoni PA, Gerandi G, Barboni T. Smoke emissions due to burning of green waste in the Mediterranean area: Influence of fuel moisture content and fuel mass. *Atmos. Environ.* 2017; 159: 92-106.
25. Chomanee J, Tekasakul S, Tekasakul P, Furuuchi M, Otani Y. Effects of moisture content and burning period on concentration of smoke particles and particle-bound polycyclic aromatic hydrocarbons from rubber-wood combustion. *Aerosol Air Qual Res* 2009; 9: 404-411.
26. Bahadori A, Zahedi G, Zendehboudi S, Jamili A. Estimation of the effect of biomass moisture content on the direct combustion of sugarcane bagasse in boilers. *Int J Sustain Energy* 2014; 33: 349-356.
27. Lu H, Zhu L, Zhu N. Polycyclic aromatic hydrocarbon emission from straw burning and the influence of combustion parameters. *Atmos Environ* 2009; 43: 978-983.
28. Jones JM, Ross AB, Mitchell EJS, Lea-Langton AR, Williams A, Bartle KD. Organic carbon emissions from the co-firing of coal and wood in a fixed bed combustor. *Fuel* 2017; 195: 226-231.

29. Bafver LS, Lackner B, Tullin C, Berntsen M. Particle emissions from pellet stoves and modern and old-type wood stoves. *Biomass Bioenergy* 2011; 35: 3648-3655,

30. NIWA, Factors influencing particulate emissions from NES compliant woodburners in Nelson, Rotorua and Taumarunui 2007. National Institute of Water & Atmospheric Research Ltd, Auckland, New Zealand. March 2012.

Tables

Table 1

Proximate and Ultimate fuel analyses.

Fuel	Fresh cut beech	Seasoned beech	Kiln dried beech	Fresh cut spruce	Seasoned spruce	Kiln dried spruce
designation	FCB	SB	KDB	FCS	SS	KDS
VM (db)	85.0	84.8	86.1	85.0	83.5	84.2
FC (db)	14.7	14.7	13.7	14.8	16.2	15.8
Ash (db)	0.3	0.5	0.02	0.2	0.3	< 0.1
C (%) (daf)	49.9	48.9	49.0	50.0	50.9	49.9
H (%) (daf)	6.07	6.18	6.11	6.19	6.13	6.12
N (%) (daf)	0.12	0.08	0.09	0.08	0.08	0.08
S (%) (daf)	0.02	0.02	0.02	0.03	0.02	0.02
O (%) (daf)*	43.7	44.6	44.6	43.6	42.8	43.9
HHV (db) (MJ/kg)	19.8	19.3	19.5	19.9	20.2	19.9

db=dry basis, daf = dry ash free basis, O % calculated by difference.

Table 2

Comparison of Moisture Content measured by different methods.
Actual value compared with estimated using digital probe

Fuel	Actual MC% Heated 96 h in Drying Oven Error ± 5 %	MC%. Estimate 1. Digital probe, middle surface	MC%. Estimate 2. Digital Probe, average end of grain
FCB	36.4	29 ± 1	21 ± 2
SB	29.7	24 ± 1	19 ± 2
KDB	3.6	4 ± 1	3 ± 2
FCS	42.9	22 ± 1	21 ± 2
SS	39.8	22 ± 1	21 ± 2
KDS	7.1	5 ± 1	7 ± 2

Table 3

Overall cycle gaseous and total PM Emission Factors for the different woods.

The % moisture contents of the woods are shown in parenthesis.

Fuel	Emission Factor g/kg						
	CO	CH ₄	CH ₂ O	C ₆ H ₆	SO ₂	NO _x	PM
FCB (36.4%)	127.6	7.4	2.3	2.84	0.46	2.4	1.8
SB (29.7%)	96.8	3.4	1.26	1.46	0.51	1.41	0.5
KDB (3.6%)	70.0	0.74	0.15	0.35	0.24	1.11	1.0
FCS (42.9%)	106.9	5.23	1.70	2.2	0.86	0.80	1.9
SS (39.8%)	103.6	3.8	1.20	1.5	0.77	0.71	0.9
KDS (7.1%)	116.1	1.56	0.16	0.36	0.45	1.01	0.3

Table 4

Particle size distribution (mass %) and values of EC/TC for the different woods.

The % moisture contents of the different wood are shown in parenthesis.

Fuel	<PM ₁	PM ₁ -PM _{2.5}	PM _{2.5} -PM ₁₀	>PM ₁₀	EC/TC
FCB (36.4)	98.6	0.7	0.3	0.5	0.15
SB (29.7)	95.4	1.4	1.2	2.1	0.21
KDB (3.6)	92.6	3.9	1.8	1.7	0.28
FCS (42.9)	97.3	1.1	0.7	0.9	0.015
SS (39.8)	92.2	2.6	2.6	2.6	0.15
KDS (7.1)	92.5	3.1	3.0	1.4	0.51

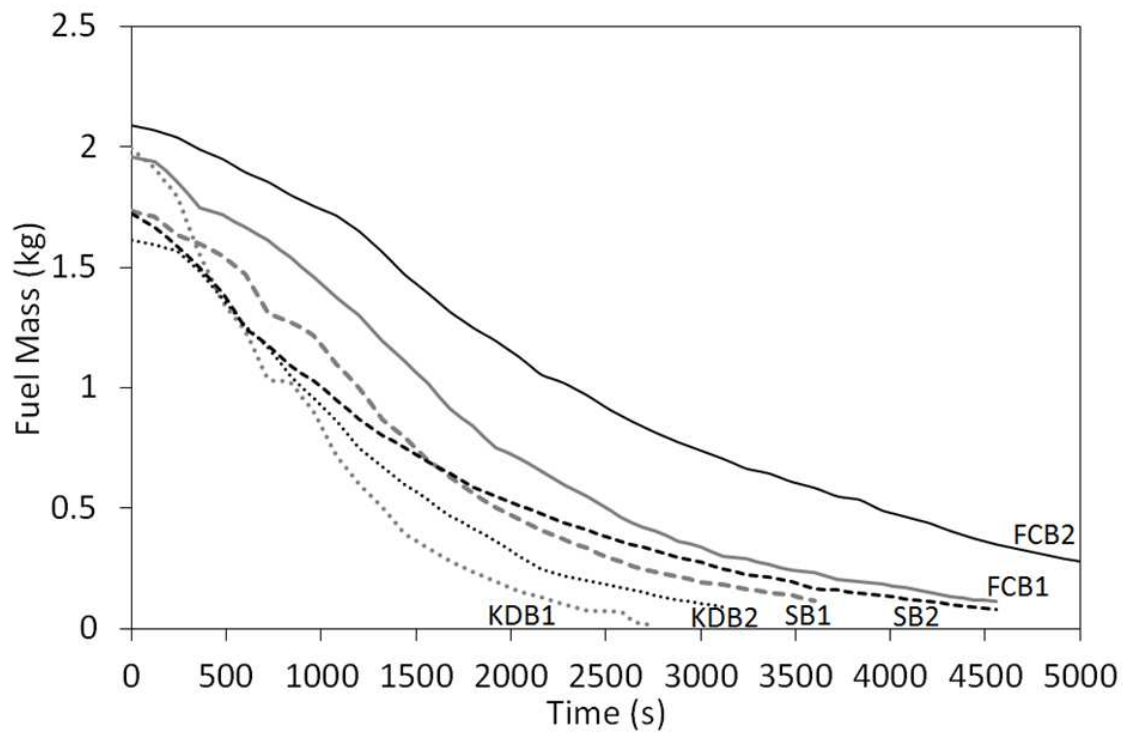


Fig. 1. Plot of fuel mass against burning time for the beech samples.

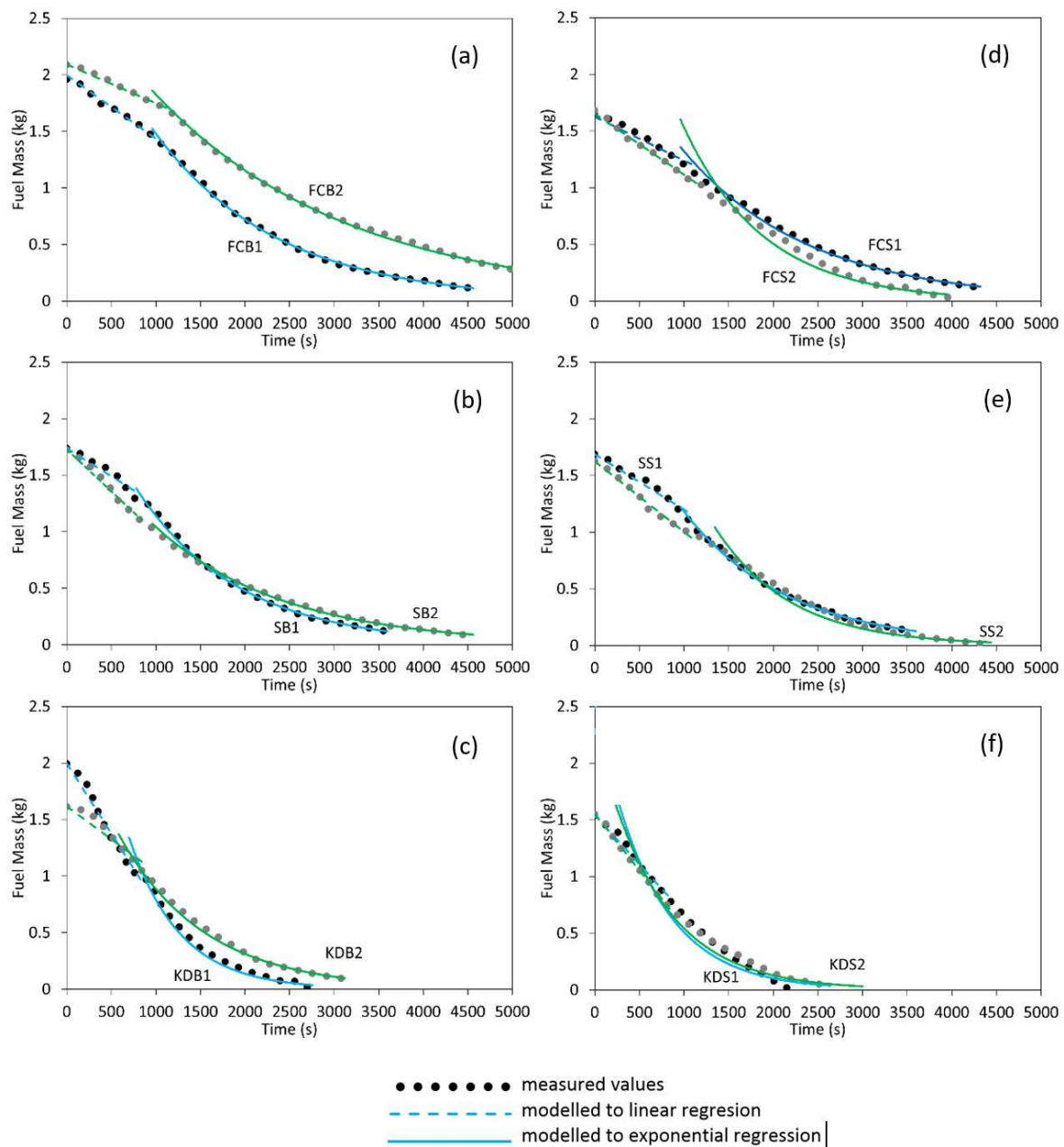


Fig. 2. Plots of the measured mass of the fuel throughout combustion with initial period of mass loss modelled to a linear function and the established combustion period modelled to an exponential function. The characteristic burning time is derived from the exponential function: (a) Fresh cut beech; (b) seasoned beech; (c) kiln dried beech; (d) fresh cut spruce; (e) seasoned spruce; (f) kiln dried spruce.

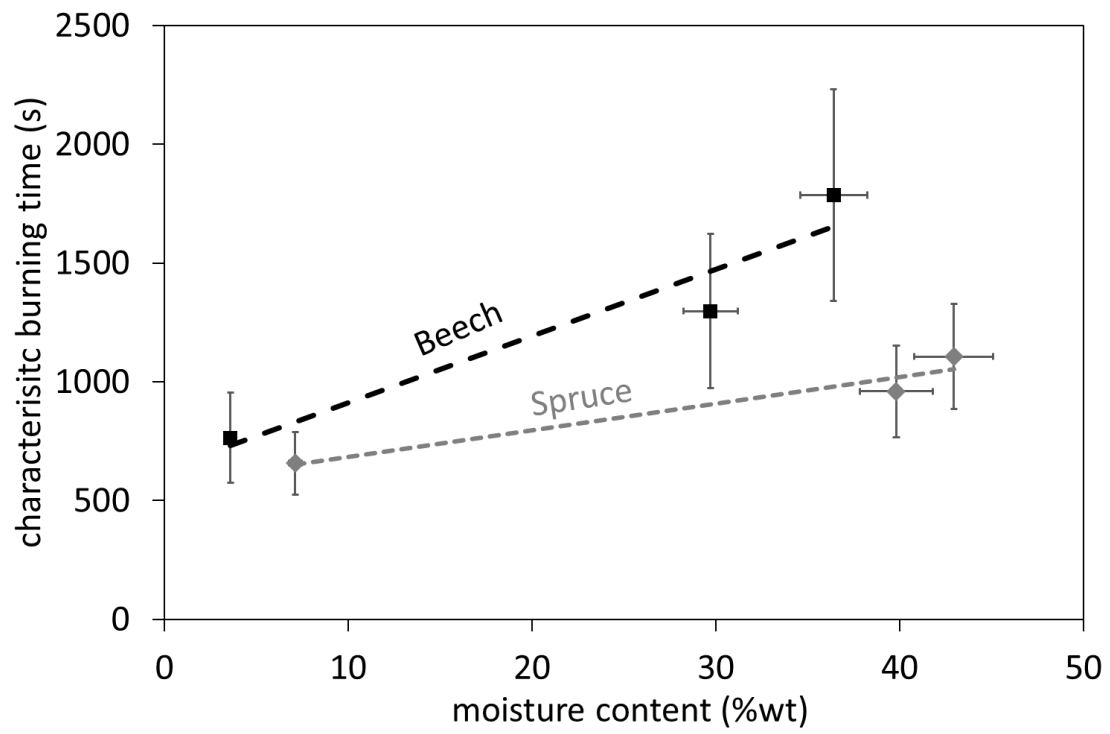


Fig. 3. Plot of characteristic burning time against moisture content for all the samples: ■, beech; ◇ spruce.

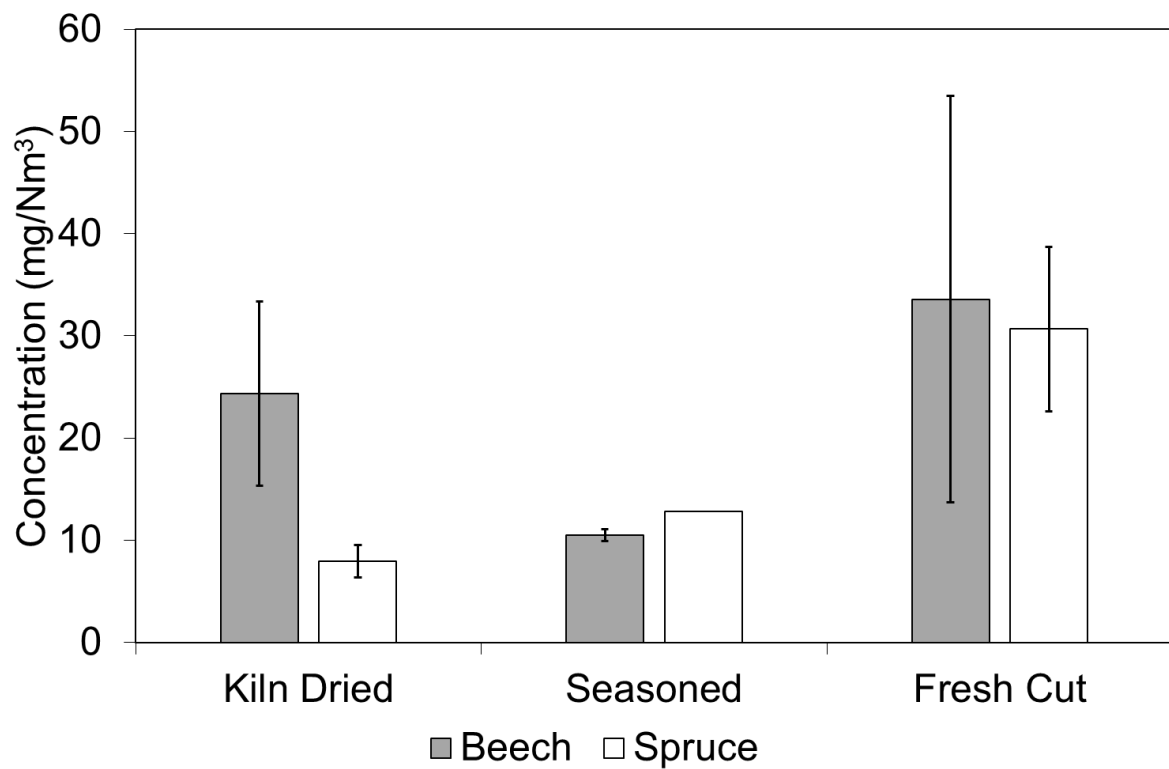


Fig. 4. Total particulate matter results obtained by the Dekati Impactor of the smoke concentrations (mg/Nm³) for refuelled cycles plotted for all fuels studied: ■ beech, □ spruce.

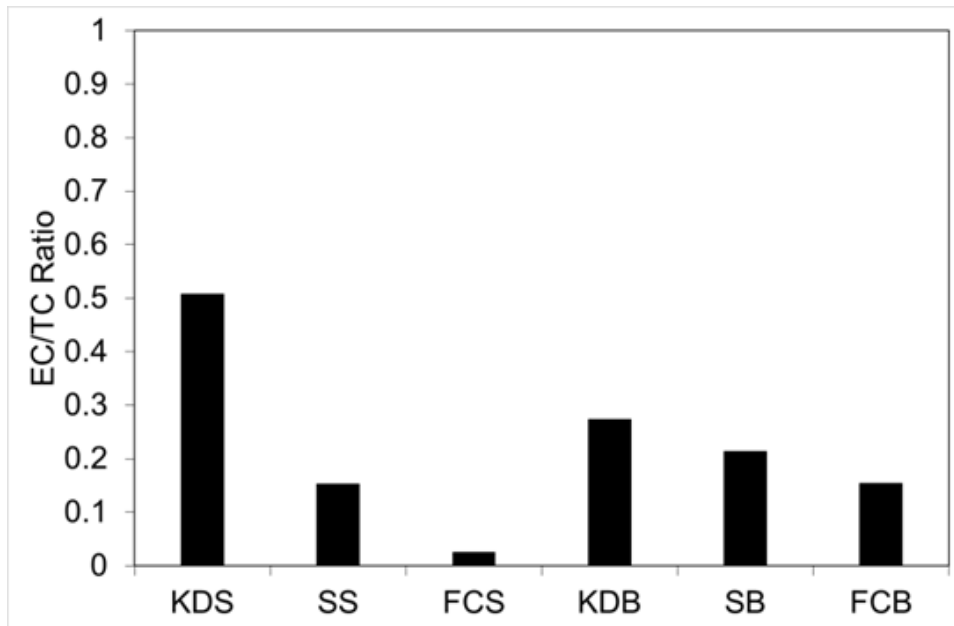


Fig.5. Measurements of the particulate EC/TC ratio for all samples over the whole combustion cycle taken in the dilution tunnel and using a refuelled batch.

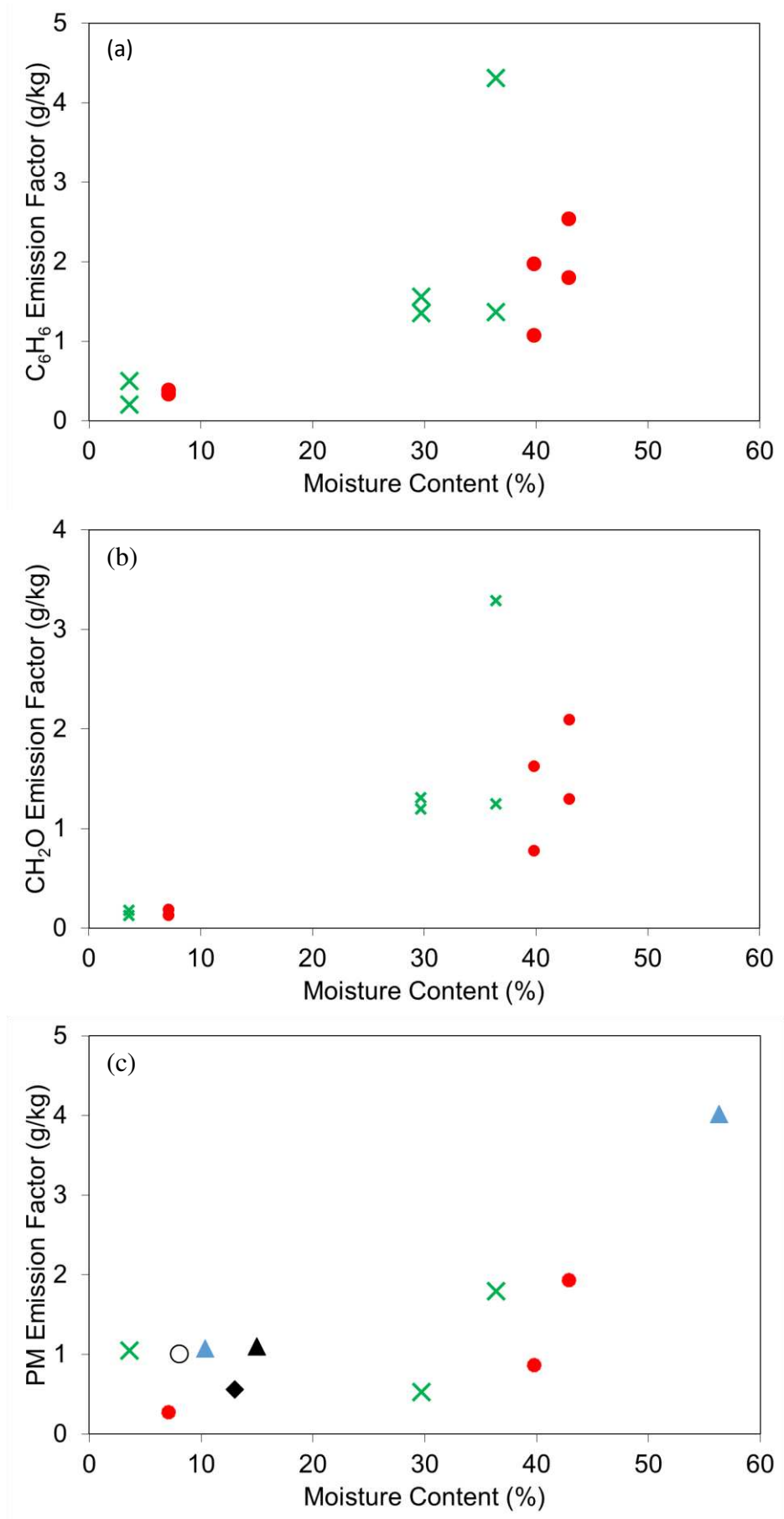


Fig. 6. Plot of the Emission Factors for (a) C₆H₆, (b) CH₂O and (c) total PM against Moisture Content: this work, x beech, ● spruce; ◆ Fachinger et al [6]; ▲ Bafver et al. [29]; ▲ Magnone et al. [9]; ○ Mitchell et al. [14].