



## Full Length Article

# Hydrogen chloride emissions from combustion of raw and torrefied biomass



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## HIGHLIGHTS

- Torrefied biomass was found to have much lower chlorine content than raw biomass.
- Torrefaction was found to preferentially reduce the chlorine content of low alkali biomass.
- Most of the biomass chlorine was released as HCl during torrefaction.
- HCl emissions from burning torrefied biomass were significantly lower than those from raw biomass.
- HCl emissions from burning low-alkali torrefied biomass were drastically lower than those from raw biomass.

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## ABSTRACT

Elevated emissions of hydrogen chloride (HCl) from combustion of biomass in utility boilers is a major issue as it can cause corrosion and, in combination with the high alkali content often encountered in these fuels, it can also deposit molten alkali chloride salts on the boiler's water tubes. Such deposition can impede heat transfer and cause further corrosion. This work torrefied and then burned herbaceous biomass (corn straw) as well as crop-derived biomass (olive residue and corn-based Distillers Dried Grains with Solubles, DDGS), all pulverized in the size range of 75–150  $\mu\text{m}$ . It monitored the HCl emissions from torrefaction of biomass and, subsequently, the comparative HCl emissions from combustion of both raw and torrefied biomass. Results showed that during torrefaction most of the chlorine of biomass was released in the gas phase, predominately as HCl. Consequentially, combustion of torrefied biomass, which contained less chlorine than raw biomass, generated significantly lower HCl emissions than raw biomass, particularly so for biomass of low alkali content. This observation complements previous findings in this laboratory that torrefied biomass also generated lower  $\text{SO}_2$  emissions than raw biomass, albeit by a smaller factor. Both of these findings enhance the appeal of torrefied biomass as a substitute fuel in utility boilers.

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## 1. Introduction

Renewable energy (i.e., energy collected from resources that are naturally replenished on a human timescale, such as sunlight, wind, rain, biomass, tides, waves, and geothermal heat) is the world's fastest-growing energy source, having experienced growth 2.6% per year [1]. Particularly, the use of renewable fuels (biomass, biofuels etc.) is being projected to increase with an even faster pace (by 6.6% per year) [2]. Renewable energy accounted for 10% of the total energy consumption in the US in 2015; whereas bioenergy (wood, biofuels, biomass waste) accounted for nearly half of that,

i.e., 5% of the total energy consumption [3]. The corresponding numbers in Germany, UK and China were 6.5%, 7% and 2% of the total energy consumption, respectively, in 2015 [4–6]. The European Union has set a target of obtaining 20% of its total primary energy consumption from renewable sources by the year 2020 [7], including bioenergy. There have been a number of applications globally where renewable biomass has been co-fired with coal in existing power plants, please see Refs. [8–12]. There have also been efforts to generate electric power on a large scale from neat biomass in dedicated boilers, as currently exemplified in the operation of the DRAX power generation plant in UK [13].

As renewable biomass may be considered to be an abundant and nearly  $\text{CO}_2$ -neutral energy source, it can be used to generate electricity by direct combustion [14]. In such applications,

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however, raw biomass experiences some disadvantages as a fuel because of its high moisture content, low heating value, low combustion efficiency, resistance to pulverization and high price of transportation per unit energy content [15]. Besides, there are additional important issues associated with the combustion of biomass in utility boilers, those of surface deposition by slagging and fouling, and corrosion. The likelihood of corrosion depends significantly on the content of chlorine in the fuel, even though chlorine accounts for a minor fraction of the chemical composition in almost all solid fuels, including coals and biomass [16–18]. Chlorine concentrations in the biomass fuels are associated with the nutrient cycle and the living portion of the biomass material. Generally speaking, the chlorine content of wood is low, typically lower than that of coal, whereas the chlorine content of herbaceous biomass, fruit and crop residues, is much higher, typically higher than that of coal [16]. This is because the chlorine concentration in crop-related biomass is heavily influenced by harvest time and fertilization practices. Chlorine is largely problematic in combustion and can even lead to the formation of health hazardous polychlorinated dioxins and furans [19].

Biomass with high chlorine content is an undesirable fuel in power plants, although it can also bring some benefits such as oxidizing mercury and thereby facilitating its capture and control [20]. During combustion, fuel-bound chlorine may be released as HCl, Cl<sub>2</sub>, or alkali chlorides, mainly potassium chloride (KCl, with a melting point of 770 °C), sodium chloride (NaCl, with a melting point of 801 °C) and calcium chloride (CaCl<sub>2</sub> with a melting point of 772 °C) [21]. Several studies that investigated emissions of inorganic chlorine from biomass combustion [22–24] found that HCl and particulate chlorine are the dominant products. HCl is a significant contributor to corrosion in the boiler and, if released to the atmosphere, it contributes to acid rain [25]. Moreover, the formation of the aforesaid alkali chloride salts can generate sticky corrosive deposits causing localized damage to boiler water tubes and other surfaces [26]. When biomass is burned, slag formation in the radiative section of a boiler may significantly increase in comparison to coal. In addition, convective section fouling is also a troublesome biomass ash-related problem, even if slower than slagging, because of the relatively high alkali metal content of biomass. Both deposition phenomena, can result in increased flue gas temperatures and concomitant boiler efficiency losses [27]. To mitigate boiler deposition and corrosion, past research has focused on taking measures for addressing these issues, such as pre-treating the fuel, limiting the amount of biomass in the fuel blend to low percentages, using low chlorine and low alkali content biomass, or implementing sorbent injection in the furnace [28–36]. For instance, Obernberger et al. [28] and Jensen et al. [29] suggested washing of the fuel as an efficient pre-treatment measure to reduce chlorine concentration in biomass. Aho and Ferrer [30] co-fired high chlorine content biomass (meat and bone meal) with coal and reported on the importance of coal ash composition (aluminosilicates, sulfur) in preventing chlorine deposition on boiler surfaces. Coda et al. [31] suggested to add aluminum-containing additives (kaolin, bauxite and fly ash from a pulverized coal plant) and limestone to biomass to liberate Cl from alkali chlorides which, otherwise, can produce corrosive deposits. Others found that co-firing coals (Federal with 0.14% Cl, Pittsburgh No. 8 with 0.09% Cl and Eastern Kentucky with 0.17% Cl) with low-content chlorine biomass (wood with <0.01% Cl) can be beneficial in actually reducing the HCl emissions of the coals [32,33].

As low chlorine biomass is preferred in power generation, attention can be paid to literature reports showing that torrefaction reduces the chlorine content of biomass. Torrefaction of biomass is a mild thermal pre-treatment method, performed at low temperatures (typically 200–300 °C) in the absence of oxygen, i.e., it is mild pyrolysis. Such a process releases moisture, organic

compounds, CO and CO<sub>2</sub> [37]. Such torrefaction gas can be collected and burned to partially offset the energy consumption during the torrefaction process [37,38]. Torrefaction can alleviate many of the notable shortcomings of raw biomass, such as its high moisture content and associated low heating value, poor grindability, biodegradability (i.e., propensity to rot), etc. [39]. Björkman et al. [40] reported that 20–50% of the chlorine is released during low temperature pyrolysis (400 °C) of biomass. Keipi et al. [41] also observed that torrefaction reduced the chlorine content of eight biomass samples by 50% or more. Toptas et al. [42] confirmed that the torrefaction process releases a significant amount of chlorine in biomass as HCl and other compounds. Regarding those other compounds, Shang et al. [43] detected significant amounts of methyl chloride (CH<sub>3</sub>Cl) in the volatile torrefaction products. Moreover, Saleh et al. [44] reported that the release of chlorine during biomass torrefaction and pyrolysis peaked with about 60–70% at 350 °C. From the view-point of corrosion/deposition, the release of chlorine species in low-temperature brick-surfaced torrefaction furnaces may be less problematic than the release of such species in high-temperature metal-surfaced boilers.

This investigation examined the comparative emissions of HCl from combustion of both torrefied biomass and of their raw biomass precursors under high-temperature, high-heating-rate conditions, pertinent to those found in utility boilers. Two different types of biomass were burned, herbaceous (corn-straw) and crop-derived (rice husk and corn-DDGS). This investigation complements previous work in this laboratory, which burned the same three biomass types, among others, in the same furnace (albeit fitted with a different sampling train) under similar conditions, and compared the sulfur oxide and nitrogen oxide emissions of raw and torrefied biomass [45]. In that work, SO<sub>2</sub> emission factors of torrefied biomass were found to be lower than those from their respective raw biomass precursors, even if some torrefied biomass types contained higher sulfur mass fractions than their raw biomass precursors. Parenthetically, no significant differences were detected in NO<sub>x</sub> emission factors of raw and torrefied biomass [45].

## 2. Experimental apparatus and procedure

Three different types of biomass, corn straw, corn-based DDGS and olive residue, were selected for these experiments based on their relative high chlorine content. Corn straw was supplied by the Harbin Institute of Technology, China; olive residue samples were supplied by ELCOGAS, S.A., which is an IGCC power station located in Puertollano (Ciudad Real, Spain) [46]; and corn-based DDGS (Distiller's Dried Grains with Solubles) was provided by a North American ethanol-producing company. All of the fuels were dried, ground or chopped in a household blender, and sieved to obtain the size cut of 75–150 μm. Batch torrefaction of all samples was carried out in a horizontal muffle furnace, depicted in Fig. 1a, in the absence of oxygen at 275 °C for 30 min. Optical microscope photographs of the fuel particles are included in Table 1, together with the compositions of the fuels, given on a dry basis. The proximate analysis of the fuels was performed based on GB/T 212-2008 Chinese standards in an electric oven (5E-DHG) and muffle furnace (5E-MF6000). Ultimate analysis for carbon, hydrogen and nitrogen were carried out by an elemental analyzer (CHN2200) according to GB/T 30733-2014 standards, whereas sulfur analysis was carried out in a Coulomb fixed sulfur analyzer (5E-8S) based on the GB/T 214-2007 standard. The heating values of the biomass fuels were measured with a calorimeter (5E-AC/PL) according to GB/T 213-2008.

Both the raw and torrefied biomass samples were injected and burned in an ATS electrically-heated, laminar-flow, alumina drop-tube furnace (DTF) with a 25 cm long radiation zone [47], which is

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