Research article

NO and SO$_2$ emissions from combustion of raw and torrefied biomasses and their blends with lignite

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ABSTRACT

The impact of torrefaction on the NO and SO$_2$ emissions from combustion of biomass was investigated. Combustion experiments were carried out with two torrefied biomass fuels, i.e., poultry litter and olive tree pruning and their blends with lignite using a bench scale single particle reactor. For comparison, NO and SO$_2$ emissions from tests with untorrefied biomasses and their blends with lignite were also investigated. The total release of SO$_2$ and NO for each fuel was determined at three different temperatures: 900, 1000, and 1100 °C. The NO release from the untorrefied biomasses was found to be lower than those from torrefied biomasses, despite their higher fuel- N content. In case of co-combustion of both raw and torrefied biomass with lignite, the NO release was lower than the anticipated one. On the other hand, in the co-combustion experiments, blends with torrefied biomass showed a larger reduction in SO$_2$ release than the blends with raw biomass.

The study revealed that the SO$_2$ emissions from blends are not proportional to the mixing ratio of the fuels and to the emissions properties of the respective fuels. No clear correlation was detected between the NO$_x$ emissions and fuel-N content. In addition to the NO and SO$_2$ emissions, the sintering propensity of the ash residue were investigated using scanning electron microscopy (SEM).

1. Introduction

Stringent environmental legislations for poultry litter have led to the need of alternative waste management options such as biochemical and thermal processes (Dalolioa F.S. et al., 2017). Combustion, as a thermal process, is the easiest way to convert the waste to a sterile material with a significant reduction in volume by 80–95% and to recover energy (Pandey et al., 2016). In combustion of poultry litter waste, the major environmental and operating problems are associated with its high moisture, nitrogen content and composition of its inorganic contents (Di Gregorio et al., 2014). Polesek-Karczewska et al. (2018) also remarked that the main issue regarding the efficiency of poultry litter combustion was closely connected to a low bulk density of the fuel bed.

One option in thermal conversion of poultry litter is co-combustion. Co-combustion of biomass or waste with coal is a simple and economically feasible way of utilizing biomass and waste and for replacement of fossil fuels. Co-combustion of biomass or waste with coal is a simple and economically feasible way of utilizing biomass and waste for replacement of fossil fuels. The co-combustion of lignocellulosic biomass is increasingly gaining importance as it represents

a low cost, sustainable, and renewable energy option that can provide reduction in net CO$_2$, SO$_2$ and often NO$_x$ emissions (Sahu et al., 2014). Lower SO$_2$ emissions can be expected due to lower sulfur content of the biomass. In addition, the alkali in biomass should have the potential to retain sulfur in the ash as alkali sulfates. Zhang et al. (2013) observed significant reduction in SO$_2$ emission with an increased fraction of tobacco stem when blending with a high-sulfur bituminous coal. Similar results have been obtained for rice husk/coal blend (Huang et al., 2016), saw dust -coal blend (Kazagic and Smajevic, 2007) and agricultural wastes/coal blends (Narayanan and Natarajan, 2007). Despite its animalic origin, poultry litter waste is in many respects comparable with other biomass fuels. A number of co-combustion studies on poultry litter have been performed over the past decade. For instance, the influence of the combustion conditions on emission levels of pollutants such as SO$_2$ and NOx during co-combustion of chicken litter and peat in fluidized bed combustion was investigated by Henihan et al. (2003) and Abelha et al. (2003). Abelha et al. (2003) found that the main problem associated with the combustion of poultry litter was the high moisture content which influenced its feeding to the combustor. Li et al. (2008) investigated the effect of co-combustion of poultry litter and coal on emissions in a fluidized bed combustor. They observed that addition of

poultry litter into blend reduced the levels of SO_2. However, the level of NO either increased or decreased depending on the percentage of poultry litter in the blends. Junga et al. (2017) tested the technical and environmental performance of 10 kW understocker boiler during combustion of a blend of coal with laying hens mature. When 15% of the poultry litter was added to the coal, thermal output decreased (in about 20%) while CO and NO_x emission increased. The conversion of fuel blends of poultry litter and lignite has also been studied using thermogravimetric analysis (TGA). Yurdakul (2016) observed that with increasing coal content in the blend, the decomposition temperatures and burnout temperatures were shifted to higher temperatures, whereas combustion reactivity of the blends was decreased.

Although co-combustion is the least complicated and one of the most advantageous ways of utilizing biomass and waste in stationary energy conversion, there are technical challenges associated with co-combustion in existing coal power plants. The main challenges are poor grindability and low energy density of biomass. Torrefaction, i.e., mild heat treatment of biomass, is a promising way to improve fuel properties of biomass. Torrefaction of biomass can result in a less hydrophilic solid product with a higher heating value. In addition, the grindability may also be improved (Van der Stelt et al., 2011; Gil et al., 2015). Torrefaction has attracted significant interest in recent years. Most of the research has focused on the effect of process conditions and feedstock type on the product properties. A number of kinetic studies have also been undertaken in order to understand the combustion behavior of torrefied biomass. Despite the available knowledge on the combustion behavior of torrefied biomass, few studies have investigated poultry litter biochar behavior during combustion (Cimo et al., 2014; Novak et al., 2013). In addition, only few studies on the emissions from combustion of torrefied biomass have been reported in the literature (Li et al., 2012; Ndibe et al., 2015a,b). Moreover, one study has addressed the gas emissions from the combustion of poultry litter biochar (Mau and Gross, 2018). In that study, the gaseous emissions from biochar combustion were investigated with TGA–FTIR combustion experiments.

The European standard EN 14961-2 cannot fulfill the quality specifications for non-industrial pellets because of the specific characteristics of the raw material. It is clear that more research on residual biomass sources is needed to investigate their quality as fuels and the emissions generated during their combustion. In this study, differently from the studies in the literature, we investigated NO and SO_2 release from blends of lignite with four different biomasses: olive tree pruning and torrefied olive tree pruning, poultry litter and torrefied poultry litter. The study was performed using single particle reactor where single fuel pellets with different biomass to lignite ratio were combusted in a controlled gas atmosphere. In addition, the sintering propensity of the ash residues was analyzed using Scanning Electron Microscopy.

2. Materials and methods

2.1. Materials

The fuels used in this study included a Turkish lignite (from the Soma basin, Turkey), olive tree pruning (OP), torrefied olive tree pruning (OPB), poultry litter (PL) and torrefied poultry litter (PLB). The torrefied fuels were produced in a vertical furnace at 300 °C for a residence time of 30 min. Detailed information about the torrefaction process can be found in a previous work (Toptas et al., 2015). The properties of fuels are listed in Table 1.

Prior to combustion experiments, the fuels were dried in an oven and subsequently ground to a particle size less than 2 mm. Pellets were prepared from pure fuels and blends. The pellets were pressed using a manually operated hydraulic press. The diameter of the pellets was 8 mm. Instead of identical heights of the pellets, all pellets had a mass of 0.2 g. The blends of lignite and torrefied biomass and lignite and

Table 1
The properties of lignite, raw- and torrefed-biomass.

	Lignite	PL	OP	PLB	OPB
Proximate analysis (wt.% dry basis)					
Ash (at 550 °C)	11.5	8.2	3.3	12.8	4.1
Volatile matter	49.9	68.3	71.6	44.3	54.3
Fixed carbon	38.6	23.5	25.1	43.1	41.6
HHV, MJ kg^{-1}	25.1	14.4	17.8	22.9	22.3
Ultimate analysis (wt.% dry basis)					
C	63.92	35.7	43.47	57.39	54.79
H	4.25	5.27	5.78	4.40	5.35
N	1.51	9.61	1.29	5.88	1.62
S	1.11	0.24	0.18	0.48	0.27
O	17.71	40.98	45.98	19.05	33.87
Ash analysis (wt.% dry basis)					
Na_2O	0.76	5.11	2.09	4.92	1.95
K_2O	1.41	26.53	22.48	25.82	21.56
CaO	2.48	39.52	59.78	41.21	61.28
MgO	1.73	6.22	6.94	7.05	7.35
SiO_2	53.71	4.73	5.59	5.24	6.19
Al_2O_3	24.08	1.09	0.86	1.55	1.05

untreated biomass were prepared by physical mixing. Four different blends of lignite and untreated biomass were prepared. The mass fraction of lignite in these samples were 25 wt%, 50 wt% and 75 wt%, referred to as 3:1, 1:1, 1:3 in figures. In the pellets containing lignite and torrefied biomass, the mass ratios were 50%.

2.2. Combustion experiments

The combustion behavior of the fuels and their mixtures were investigated using a single particle reactor, which consists of a quartz tube reactor in an electrically heated ceramic furnace. Before the tests started, the reactor was heated to the temperature at which the combustion experiments were to be conducted. The sample was inserted into the reactor using a movable probe. In this way, the sample could be shifted from room temperature into the hot reactor environment within a second. The main part of the gas enters through an inlet at the bottom of the reactor. A smaller portion of the inlet gas enters at the level where the fuel sample is inserted. This gas is needed to cool the sample before entering the reactor as well as to keep the viewing windows clean. A total flow rate of 220 l/h was used in all experiments. The gas flows were controlled using mass flow rate controllers. The composition of the exiting gas was analyzed with commercial analyzers: an AO2020 Continuous Emissions Analyzer (ABB, Germany) was used for CO, CO_2, and SO_2 analysis; O_2 was analyzed using a 4900 Continuous Emissions Analyzer (Servomex, England); and NO_x was analyzed using a Model 20OEM chemiluminiscence analyzer (Teledyne, United States). Based on the time resolved gas measurements and the known total flow, the total amount of NO and SO_2 were calculated integrating over the whole combustion time. A detailed description of the reactor setup and the experimental procedure can be found in Karlström et al. (2015).

The combustion experiments were conducted at three different temperatures: 900 °C, 1000 °C and 1100 °C. The highest temperature corresponds to the maximum achievable temperature of the electrically heated reactor. The two other temperatures were selected to be wide apart enough to obtain a trend without an excessive amount of experiment. The lowest temperature corresponds approximately to the bed temperature in a fluidized bed combustor, whereas the temperature 1100 °C is similar to that in the free board (Vainio et al., 2012).

A mixture of 3 vol % O_2 in N_2 was chosen to avoid very high particle temperatures in comparison to the reactor temperature because of exothermal oxidation reactions. Using an oxygen rich gas mixture, such as synthetic air, easily leads to particle temperatures several hundreds degrees higher than the reactor set point, and correlating the results to the reactor temperature becomes questionable. In addition, a gas with low oxygen content is representative for conditions inside a fluidizing

bed, inside a flame as well as in the flue gas section of a boiler.

2.3. Characterization of fuels and ashes

Ultimate analysis was carried out using LECO CHNS 932 elemental analyzer according to ASTM D5291-96. The proximate analysis was done according to NREL/TP-510-42622 for ash analysis and ASTM D3175-89a for volatile matter. The higher heating values (HHV) of the biochars were calculated according to the formula proposed by Channiwala and Parikh (2002). The metal analysis of ashes in feedstocks was analyzed by Atomic Absorption Spectrometry (AAS) following acid digestion. The AAS was conducted using a novAA 300 (Analytik Jena,Germany). In addition, the residues from each combustion were analyzed by a Gemini 1530 scanning electron microscopy (SEM; LEO, Germany) equipped with an UltraDry Silicon Drift X-ray Detector (Thermo Scientific, USA).

3. Results and discussion

3.1. Fuel analysis

The volatile content of the olive pruning and the poultry litter is significantly higher than that of the lignite of the torrefied fuels. However, by torrefaction, the volatile content of olive pruning and the poultry litter becomes similar to that of the lignite.

Torrefaction of poultry litter decreased the nitrogen content considerably, whereas torrefaction of olive tree pruning slightly increased the nitrogen content. The way the nitrogen is bound in the biomass is one of the parameters affecting the partitioning of nitrogen between the volatiles and char during torrefaction (Darvell et al., 2012). A major nitrogen containing compound in poultry litter is urea. Due to the low decomposition temperature of urea, poultry litter releases nitrogen compounds at low temperatures (Whitely et al., 2006). Previous studies (Kim et al., 2009; Agblevor et al., 2010) related to pyrolysis of poultry litter also showed that most of nitrogen in poultry litter was released during the pyrolysis. Nevertheless, it should be noted that torrefied poultry litter still contain a considerable amount of nitrogen. On the other hand, sulfur behaved differently: most of the sulfur remained in the torrefied fuels.

3.2. Ash sintering behavior

In order to gain insight into the ash-sintering tendency, the residues from the combustion of all fuels at different temperatures were analyzed. The holding time varied between 10 and 35 min, partly because of differences in char reactivity. Fig. 1 displays SEM viewgraphs showing the morphology of the residues from the combustion of poultry litter, olive pruning and lignite at different temperatures. Although visually, residues presented a powder structure, the figures reveals that for biomass fuels, ash sintering started at 900 °C. The spherical shape and smooth surface imply that the ash has experienced a molten stage. At 1100 °C, the poultry litter residue presents a quite homogenous structure, probably due to an almost complete melting (Fernández et al., 2012). The ash from lignite shows different morphology, explained by its high Si and Al content.

SEM-EDX was used to analyze the composition of the ash residue. The result from this analysis, presented as oxides is shown in Fig. 2. The analysis revealed significant amounts of K and Ca and minor amount of Si in the residue from combustion of olive pruning and torrefied olive pruning. For poultry litter and torrefied poultry litter, the combustion residue contained significant amount P besides K and Ca. This indicates the presence of alkali phosphate besides alkali oxides, explaining the observed ash sintering. Although metals in ash are mainly in oxide forms, in case of poultry litter ash, P can be in the form of $Ca_5(PO_4)_3(OH)$ (Kaikake et al., 2009).

3.3. Gaseous emissions

To determine the time required for complete combustion for each fuel, a series of experiments with varying combustion time (15, 20, 25 and 30 min) and temperatures (900–1100 °C) were carried out. Based on the carbon oxides $(CO + CO_2)$ levels, 15 min, 20 min, 30 min reaction times for olive pruning, poultry litter, biochars/lignites, respectively, were taken as combustion times in the following experiments.

3.3.1. SO_2 emission

Fig. 3 shows SO_2 release during the first 200 s. It can be seen that SO_2 is mainly released during the devolatilization stage. It should be noted that the reported SO_2 levels are from tests using a single pellet and is not normalized to any residual O_2 level. Consequently, these emission levels are not transferable to a typical boiler.

Total emissions of SO_2 varied significantly depending on fuel type. One of the benefits of biomass combustion is the less SO_2 formation through a decrease in fuel bound sulfur (Ren et al., 2017).

It is interesting that although biomass is considered as viable option to decrease SO_2 emission, the highest SO_2 emissions were measured during combustion of olive pruning having the lowest S content (Fig. 4). There was no clear relation between the sulfur content of the fuel and the SO_2 emissions. This indicates that SO_2 emission depends on the amount and the composition of the mineral content and form of Sulphur in fuel rather than total sulphur content. To explain this contradiction, types of sulphur compounds (organic and inorganic sulphur) could be determined, besides total sulphur content of fuel. Sulphur in woody biomass is mainly present as organically-bound sulphur (Werkelin, 2008), which can be easily released. On the other hand, sulphur in coal exists mainly in inorganic form (ferrous sulphate, ferric sulphate and pyrite). During combustion, pyrite and ferrous sulphate usually forms in a gas that is rich in SO_2, whereas ferric sulphate usually forms in a gas that is rich in SO_3 (Hu et al., 2006). As based on above explanation, we can speculate that in our study, sulphur in lignite might be in ferric sulphate form.

It should be noted that SO_2 emissions from torrefied biomasses were lower than that from that from raw biomasses despite their higher sulfur content. The experiments revealed that the SO_2 release is temperature sensitive. The SO_2 release was lower at 900 °C compared to at the higher temperatures. This may be due to an increase in self-desulfurisation with rising temperature (Heschel et al., 1999). Similarly, Dayton et al. (1999), who investigated the release of sulfur during combustion of switchgrass, observed that SO_2 release is temperature dependent. Combining the results from the SO_2 measurements with the char yields, one can deduce that 50% and 10% of total sulfur in poultry litter and olive pruning, respectively, were released during torrefaction.

A comparison between the measured and anticipated SO_2 emissions from combustion of blends at 1000 °C is shown in Fig. 5. The anticipated emission is based on a purely additive behavior of the fuels. The figure reveals that the SO_2 emissions cannot be predicted based on the results obtained using pure fuel. For the blend containing 25% and 75% poultry litter, SO_2 emission increased compared with case of the lignite only, whereas blending poultry litter with the lignite at the ratio of 50% led to considerable decrease in SO_2 emission. As seen from Fig. 5, in the case of the blend containing 25% poultry litter, the experimentally obtained total SO_2 emission was much higher than the anticipated one. In contrast to poultry litter-lignite blends, addition of olive pruning to lignite at the ratio of 25% decreased the SO_2 emission compared with the lignite only case. In this case, the experimentally obtained total SO_2 emission was much lower than the anticipated one. The increasing of the olive tree pruning ratio in blend led to an increase in the total SO_2 emission. A similar behavior was observed by Wang et al. (2011) in their study of blends of wheat straw and coal. In the experiments using fuel blends, addition of torrefied biomass showed a larger reduction in SO_2 emission than addition of untreated biomass.

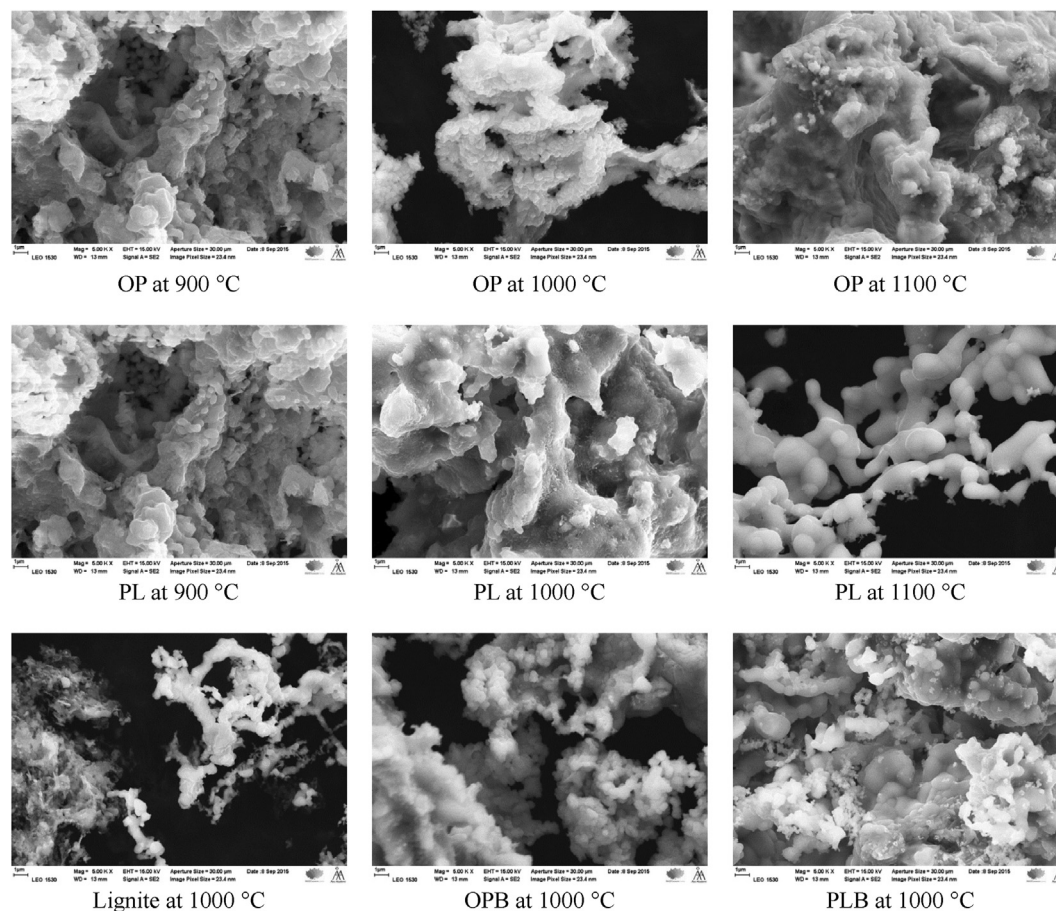In contrast to the blends containing untreated biomasses, the

Fig. 1. SEM images of residues from different combustion temperatures (Mag = 5.00 K X).
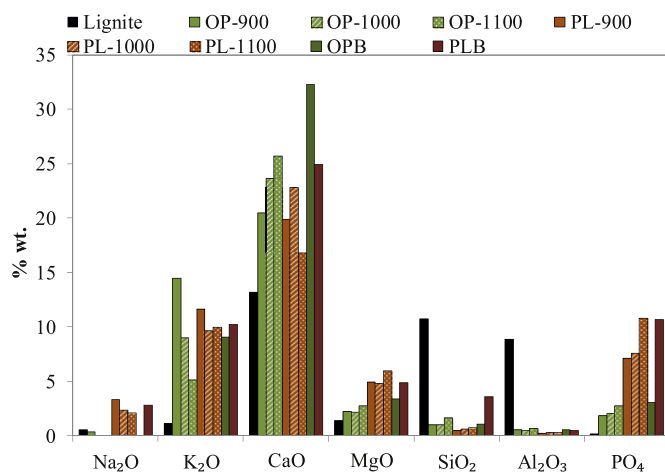

Fig. 2. EDX results of residues from different combustion temperatures.

blending of lignite with torrefied biomass significantly decreased the SO_2 emission compared with the lignite case. In addition, the experimentally observed emissions were lower than those anticipated based on a purely additive behavior. In spite of the higher sulfur content of the torrefied fuel, the blend of the torrefied fuel and the lignite released less SO_2 than the blend of the untreated fuel and the lignite. This may be due to the high calcium content of the torrefied biomass compared to the untreated biomass. As known, the addition of CaO can fix SO_2 from coal combustion as $CaSO_4$, that is decomposed at 1350–1400 °C.

3.3.2. NO emission

Fig. 6 shows the total NO emission obtained in the single particle experiments. Here too, the levels are not normalized to any specific O_2 level of the flue gas. In addition, the air to fuel ratio used in the experiments are not representative for a boiler. The total emissions of NO differed significantly from fuel to fuel. The NO emission is not solely depending on nitrogen content in fuel. For example, the nitrogen content of olive pruning was slightly lower than that of the lignite, but the total NO emission from olive pruning was higher than that from the lignite. The formation of NO is a complex process depending on fuel type and combustion conditions, such as temperature, air ratio, heating rate, particle size, etc. In solid fuel combustion systems, at temperatures below 1500 °C, the most important NO formation mechanism is that of oxidizing fuel bound nitrogen. Although high NO emission might be expected for fuels that contains a high amount of fuel bound nitrogen (Roy et al., 2013), the results obtained in previous studies (Tchapda and Pisupati, 2014; Winter et al., 1999) showed the mode that nitrogen is bound in the fuel is also important. Nitrogen compounds are present in coal in pyrrolic, pyridinic and amine forms, whereas in biomass in it can be present in a number of forms, such as proteins, amino acids, alkaloids, nucleic acid and chlorophyll (Glarborg et al., 2003). In combustion, the fuel-N is distributed between the volatiles and the char bound nitrogen (Glarborg et al., 2003). The inorganic species in fuel may also affect the fuel nitrogen conversion (Tchapda and Pisupati, 2014). It has been reported that the pyrolysis of nitrogen containing species in coals result in mainly HCN, while amine type functional groups (in biomass) would provide a possible source for NH_3 (Tchapda and Pisupati, 2014). Both, the HCN and NH_3 compounds are oxidized further to NO, NH_3 can also act as reducing agents similarly as in the thermal $DeNO_x$ process (Oliva et al., 2000). Winter et al. (1999) found that the fuel nitrogen
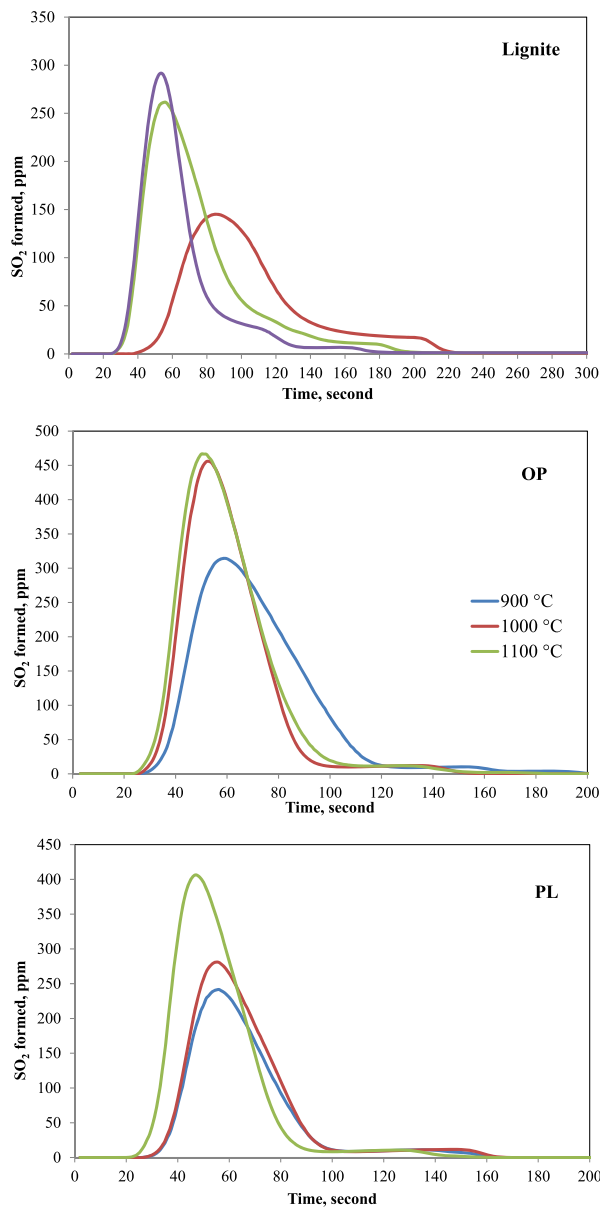
Fig. 3. SO₂ release profiles from combustion of olive tree purning (OP), poultry litter (PL) and lignite at different temperatures.
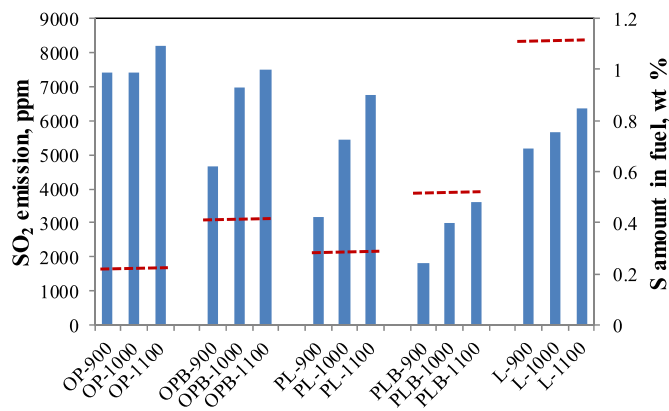

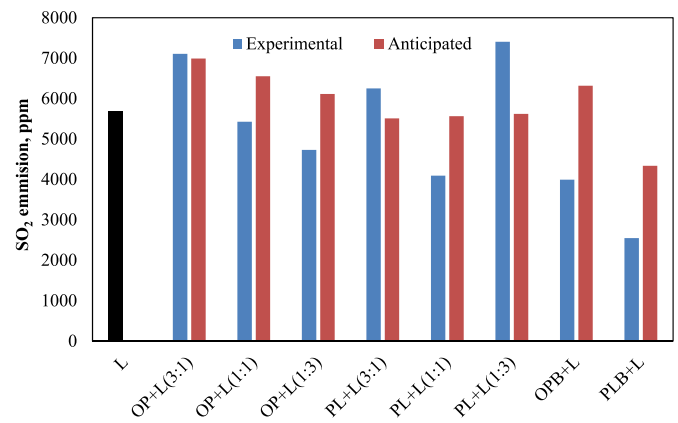**Fig. 4.** Total SO₂ emissions at different combustion temperature.

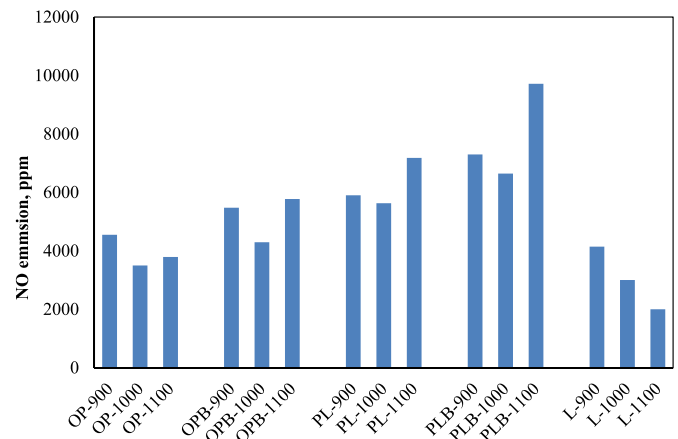**Fig. 5.** Total SO₂ emission from co-combustion at 1000 °C.

Fig. 6. Total NO emissions at different combustion temperatures.

conversion decreased with increasing nitrogen content and contributed this to the DeNOx process. Moreover, also Giuntoli et al. (2010) found that high fuel-N content could enhance De-NOx reaction. Similar result was also reported by Billen et al. (2015).

The influence of temperature on the NO emissions also varied with fuel. In the case of lignite, NO emission decreased with increasing combustion temperature. This result may be reasonable because of the increasing of the NO destruction reactions with NH₃ (Oliva et al., 2000). However, in case of biomass and torrefied biomasses, there is no linear correlation with temperature and the minimum NO emission was observed during combustion at 1000 °C.

The NO release from biomasses was lower than those from torrefied biomasses at all temperatures. The reason might be that raw biomass has higher volatile content which provides a reducing atmosphere during combustion. One of the most important finding in this study is that although the fuel bound nitrogen content in torrefied poultry litter was lower than in raw poultry litter, the NO emission from the combustion tests of torrefied poultry litter was higher. A possible reason is that the fuel-N functionality in the torrefied poultry litter is different from that in poultry litter. Most of volatile nitrogen in poultry litter seems to be released during torrefaction.

A comparison between the measured and anticipated NO emissions from combustion of blends at 1000 °C is shown in Fig. 7. In case of blends, the NO emissions increased as the poultry litter ratio increased. This is reasonable because the nitrogen content of poultry litter is higher than that of lignite. Consequently, NO emissions during co-combustion of lignite with poultry litter might be expected to be higher than the lignite only case. However, the NO emissions decreased slightly as the olive pruning ratio increased. It is noticeable that experimental NO emission values from both raw and torrefied biomass
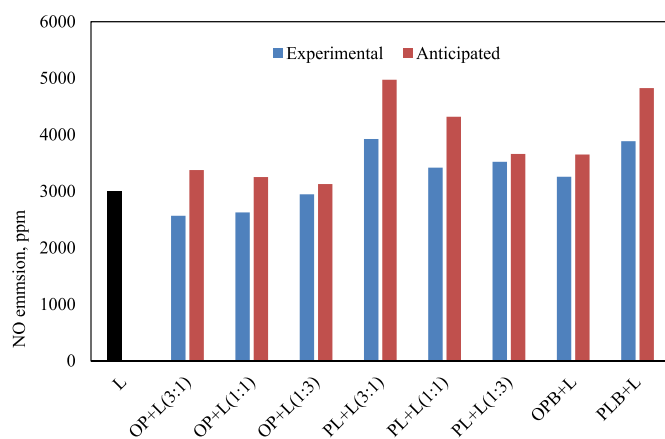
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Fig. 7. Total NO emission from co-combustion at 1000 °C.

containing blends were lower than the anticipated ones. A possible reason is the reactions between volatiles from biomass and from lignite, leading to the suppression of NO formation.

4. Conclusion

Gas emissions and combustion behavior of individual pellets of blends of lignite with olive tree pruning, torrefied olive tree pruning, poultry litter and torrefied poultry litter were investigated. The results were compared to those obtained using non-blended fuels. The SO_2 release from olive pruning was almost unaffected by the temperature in the range 900–1100 °C, whereas the other fuels all showed an increase in the SO_2 with increasing temperature. Although the torrefied biomasses had higher sulfur content, SO_2 emissions from the torrefied biomasses were lower than those of the untreated biomasses. Co-combustion of lignite and torrefied biomasses had a beneficial impact on SO_2 release. In contrast, no benefit of co-combustion on the NO release could be observed for blends of torrefied biomass with lignite. Although fuel bound nitrogen content in the torrefied poultry litter was lower than in the untreated poultry litter, NO emission from combustion of torrefied poultry litter was higher.

Acknowledgements

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