PARTICULATE MATTER FORMATION DURING CO-COMBUSTION OF AGRICULTURAL RESIDUES WITH TURKISH LIGNITE USING A DROP TUBE FURNACE

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ABSTRACT

This study investigates the particulate matter formation during combustion of olive residue, almond shell, and Tunçbilek lignite. Selected fuels (olive residue and Tunçbilek lignite) were also co-fired to evaluate the influence on particulate matter emission. Olive residue was ground to different size ranges (< 125 μm and 212–300 μm) to investigate the influence of the particle size and blended in different ratios of biomass / coal to analyse the interactions between fuels. Tests were performed in a drop tube furnace at high temperature (1000 ºC), with a high heating rate (~10^4 ºC/s), and short residence time (~3 s). Fuel was fed into the furnace at a low mass rate of 10 g/h using a syringe pump. Particulate matter was collected using a 3-stage stack impactor and categorized according to the aerodynamic diameters, PM2.5, PM2.5-10, and PM10. The results obtained included particle burnout, and particulate matter concentration. Particle burnout was above 95% for all studied fuels. Particulate matter emission depended greatly on the fuel and the blend. Olive residue presented the lowest values of PM2.5 (176 mg/g ash in fuel fed) compared with both almond shell (214 mg/g ash in fuel fed) and Tunçbilek lignite (286 mg/g ash in fuel fed). PM10 emission was particularly low for olive residue (~200 mg/g ash in fuel fed), whereas almond shell and Tunçbilek lignite showed similar values (~400 mg/g ash in fuel fed). Larger biomass particles resulted in unchanged particulate matter emissions. Co-firing of the olive residue with the Tunçbilek lignite resulted in lower PM2.5 (as compared to neat olive); higher PM2.5-10 (as compared to neat lignite); and lower PM10 (as compared to lignite). Blends of OR-TL in 25-75 ratio showed lower values of both PM2.5 and PM10 as compared with the 50-50 blends of the same fuels.

Keywords: Turkish lignite, Biomass, Co-firing, particulate matter, Drop tube furnace

1. INTRODUCTION

In Turkey, large reserves of indigenous lignite (15.6 billion tonnes) combined with an effort to decrease the energy dependence on imported fossil fuels have generated widespread incentives to increase the total installed capacity of lignite power plants three-fold by 2023 [1]. The burning of lignite coal presents several issues that require investigation such as the emission of pollutants (CO_2, NO_x, SO_x), particulate matter, slagging, and fouling. Particulate matter (PM) emission has contributed to an estimated 3.2 million premature deaths as a consequence of cardiovascular diseases and lung cancer [3]. Therefore, there is a need for further research on the formation of particulate matter in order to minimize its formation and design equipment suitable to capture PM.

The growing number of coal-biomass co-fired power plants [4] also raises concerns since studies on coal-biomass combustion with an emphasis on PM formation are still scarce. Some initial studies suggest that emission of submicron PM from biomass may be higher as compared to coal [5], and there are, additional concerns that the biomass-PM may interfere with the coal-PM precipitators. On the other hand, despite the high economic potential of 32.6 Mton per year, only about 0.1% of the electricity generation in Turkey was biomas-based in 2012, showing the great potential of co-firing for improving the efficiency of power generation [2].

For both coal [6–8] and biomass [9], the two main mechanisms of PM formation are (i) the solid particle pathway during which the excluded mineral matter is directly converted into coarse PM, and (ii) the

vaporization of mineral matter followed by conversion into fine PM during condensation. PM formation is therefore strongly dependent on the mineral content (included and/or excluded) [8,10], and on the fragmentation of char/ash particles at high temperatures [11,12]. Moreover, since coal and biomass typically differ in mineral content, the formation and characteristics of PM is strongly fuel-dependent [10,13–15]. The influence of coal rank on PM formation was investigated by Wen et al. [10] who observed that formation of both submicron and supermicron PM decreased with the increase in coal rank from lignite to coal, and coal to anthracite. Gao et al. [15] investigated mallee wood, bark, and leaf, and observed that the emissions from both bark and leaf were higher when compared to wood. Zellagui et al. [5] studied the co-firing of a woody biomass with different coals, and concluded that a ratio of 25 wt.% biomass minimized the formation of ultrafine PM0.1 and PM2.5.

In this context, this study investigates the particulate matter emission from the co-combustion of a Turkish lignite with two distinct agricultural residues, olive residue and almond shell, under high temperature and high heating rate conditions using a drop tube furnace.

2. MATERIALS AND METHODS

2.1. Fuel Samples

Three endogenous resources of Turkey were selected for this study: olive residue (OR), almond shell (AS), and Tunçbilek lignite (TL). Olive residue was supplied by the company Fora Zeytin. This company processes olives from the Havran, Balıkesir region, and delivered the olive residue ground to below 5 mm diameter and pre-dried to less than 10 wt.% moisture. Almond shell is an agricultural waste of almond production and was supplied as chunks by Bademder (cooperative from the region of Gülnar, Mersin). The lignite coal was supplied by the Turkish Coal Enterprises (TKI) from the mining region of Tunçbilek – Tavşanlı, Kütahya (annual tonnage of 7 Mt). The results of proximate, and ultimate analyses, inorganic species, and heating values can be found in Table 1. Fuels were first ground to a size smaller than 1 mm using a roll crusher, and ground further using a ring mill. All ground particles were sieved to a size cut < 125 µm and OR was additionally sieved to a larger particle size (212-300 µm) to assess the influence of particle size on the PM formation. All samples were dried overnight at around 100ºC before the combustion trials.

2.2. Experimental Methods

The experimental setup is schematically presented in Figure 1. The setup consists of a drop tube furnace, a feeding system, and a collection unit. The drop tube furnace (Protherm PTF 15.75.1000) is an electrically heated furnace with Si and $MoSi_2$ heating elements that can supply a maximum power of 7.5 kW and enable a maximum (constant) wall temperature of 1500ºC measured by an S-type thermocouple placed at the midpoint of the heated zone. An alumina tube (60% Al_2O_3) of 1500 mm length and 75 mm inner diameter was used to confine the reaction zone (1000 mm). The particles were fed from a glass vial by means of a syringe pump (Harvard Apparatus 22). The linear movement of the syringe pump was set to ensure that the distance between the particle bed and the tip of the capillary tube remained at ~3 mm along time. A 12 V vibrating motor was used to obtain a well-stirred particle bed, avoiding clogging in the capillary tube. Air was injected into the glass vial to force the particles into the capillary tube. Particles were injected downwards into the reaction zone of the furnace using a capillary tube of 2 mm inner diameter and a water-cooled injector.

The combustion of the particles in the drop tube furnace took place at a constant wall temperature of 1000ºC, and the heating rate was estimated to be on the order of 10^4 ºC/s. The total flow used, primary air + secondary air, was 20 L/min, which resulted in a residence time of ~3 s in the isothermal reaction zone. The mass flow of particles was kept low at 10±1 g/h to maintain lean combustion conditions and

to minimize interactions between particles (e.g. changes in the air-volatile ratio in the vicinity of the particles, or radiative heat exchange between particles).

Table 1. Elemental analysis of the studied samples

Parameter	Olive residue (OR)	Almond shell (AS)	Tunçbilek lignite (TL)
Proximate analysis, wt.% as received			
Moisture	5.9	8.5	2.81
Volatiles	71.9	73.0	31.1
Fixed carbon[a]	17.4	17.6	52.1
Ash	4.8	0.9	14.0
Ultimate analysis, wt.% dry basis			
C	46.6	45.4	61.8
H	6.4	6.3	5.6
N	0.6	0.4	2.7
S	-	-	1.5
O[a]	46.4	47.9	28.5
Inorganic species, wt.% dry basis			
Si	4.60	0.36	33.3
Al	1.76	0.09	12.9
Fe	7.95	1.41	22.3
Ca	26.4	52.1	1.91
Mg	1.23	0.13	1.73
Na	-	-	-
K	48.6	41.2	3.16
P	1.98	2.02	0.12
Ti	-	-	2.87
Cl	4.46	0.34	-
S	2.37	0.46	19.1
High heating value, MJ kg^{-1} [b]	18.6	18.0	27.8

[a]obtained by difference. [b]values obtained according to [16] and [17] for biomass and coal, respectively.

Particles were collected at the outlet of the drop tube furnace using a collection system that consisted of a 3-stage stack impactor (Tecora MSSI PM10-2.5 Multistage Stack Impactor) fitted with 47 mm quartz microfiber filters and a vacuum pump (Tecora Bravo Plus). Quartz filters were heat treated before sampling, according to the ASTM standard for coal ashing (D3174-12). Sampling was performed for a duration of 5-10 min, and the experiments demonstrated a good repeatability (see 98% confidence interval error bars in Figures 2 and 4). Collected particles were categorized in the sizes PM>10, PM2.5-10, and PM2.5 (PM>10 results are not presented herein). The burnout was determined for selected samples ashed in a muffle furnace in compliance with the ASTM standard D3174-12 [18] for coal and ASTM E1755-01 [19] for biomass, to determine the ash weight fraction. Subsequently, the particle burnout, ψ, was calculated in accordance with the procedure in the standard CEN/TS 14775 as follows.

$$\psi = \frac{1 - \frac{\omega_f}{\omega_x}}{1 - \omega_f} \tag{1}$$

In this equation, ω_f represents the ash weight fraction in the input fuel, and ω_x is the ash weight fraction in the collected sample. Burnout was above 95% for all fuels.
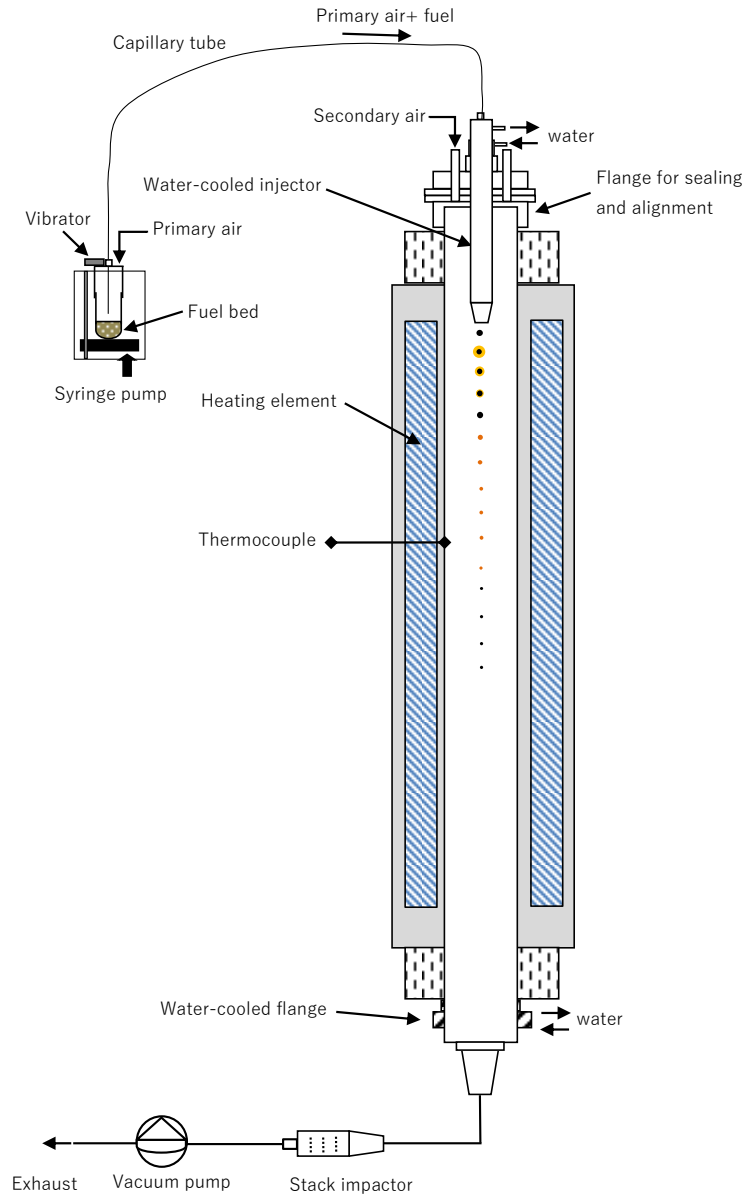
Figure 1. Schematic drawing of the drop tube furnace, feeding system, and collection unit

3. RESULTS AND DISCUSSION

3.1. Fuel Effect on PM Formation

The fuel type is known to have a strong effect on particulate matter formation [10,13–15]. In the present study, two biomass fuels and a lignite coal are compared in terms of submicron (PM2.5), PM2.5-10, and PM10 concentrations. All PM results are presented in Figure 2, and PM2.5 values are additionally presented in different basis in Table 2 to facilitate comparison with literature.

The concentration of PM2.5 was highest for TL (286 mg/g ash in fuel fed), followed by AS (214 mg/g) and OR (176 mg/g). PM2.5 for TL was higher (40 mg/g fuel fed) than the value found by Kazanç and Levendis [20] (~8 mg/g fuel fed) for a lignite coal from Texas (US). Similarly for PM10, Wen et al. [10] measured ~70 mg/g ash in fuel fed for a lignite coal, whereas herein the PM10 of TL was significantly

higher (435 mg/g ash in fuel fed). According to Wen et al. [10], there are two competing processes ruling the PM1-10 formation: mineral matter coalescence, and char fragmentation. The TL investigated has been observed in another work by the author [21] to undergo mild fragmentation under similar conditions; furthermore, in the same work [21] char combustion temperatures of ~1500 ºC were measured and albeit sufficient for the melting of Fe_2O_3 (melting temperature of 1565 ºC), are insufficient for the melting of SiO_2 (melting temperature of 1710ºC). The results obtained suggest that PM10 formation for TL was determined by char fragmentation.

PM2.5 values for OR and AS (see Table 2) were found to be in between the values reported by Ruscio et al. [22] for olive residue (~26 mg/g fuel) and corn residue (~3 mg/g fuel). TL exhibited a higher PM2.5 than that of OR and AS by 25-40 % (see Figure 2). The concentration of PM2.5 for biomass is due to the presence of alkali and alkali earth metals that present low vaporization point and tend to form particulate matter through the vaporization-condensation mechanism.

For PM2.5-10 the relationship between TL and OR is maintained, i.e. higher PM2.5-10 for TL, but not for TL and AS. Almond shell had PM2.5-10 of 187 mg/g of ash in fuel fed, whereas TL had 149 mg/g of ash in fuel fed. Almond shell had PM2.5-10 of 187 mg/g of ash in fuel fed, whereas TL had 149 mg/g of ash in fuel fed. In fact, PM2.5-10 for AS was ~%70 higher than that of OR. As observed by the author in another study [23], the interaction between aluminosilicates from lignite (33 wt.% of Si and 13 wt.% of Al, in ash) and calcium from almond shell (52 wt.% of Ca in ash) forms large gehlenite compounds which contribute to PM2.5-10. Due to the contribution of PM2.5-10 of AS, PM10 was similar to the value of TL (401 and 435 mg/g ash in fuel, respectively). Moreover, OR showed the lowest values of PM10 among the three fuels (233 mg / g ash in fuel fed).

It should be noted that due to the considerably different ash content of these two fuels (cf. Table 1), if the same PM2.5 emission was taken in fuel fed basis, the tendency would be inverted and coal PM2.5 would be greater than that of biomass (16 vs 9 mg/g fuel). This means that although the PM2.5 emission is lower for biomass; the lower ash content of biomass fuels also plays an important role and needs to be considered for biomass – coal co-firing applications.
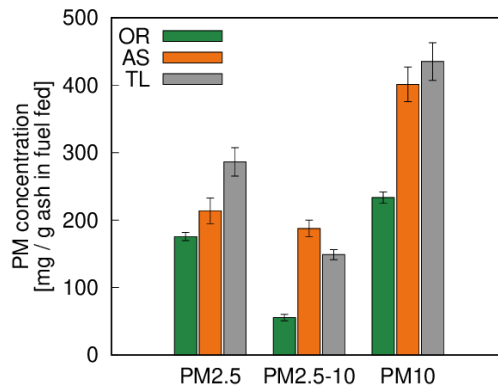

Figure 2. Particulate matter values of all neat fuels (OR – olive residue; AS – almond shell; TL – Tunçbilek lignite). All fuels < 125 μm. Vertical bars represent 98% confidence interval statistical error.

Table 2. PM2.5 concentration in different basis for OR, AS, and TL (all <125 μm)

concentration (basis)	OR	AS	TL
mg per m^3 of air	60	50	330
mg per g ash in fuel input	176	214	286
mg per g fuel input	9	8	40

3.2. Effect of particle size on PM formation

The effect of the particle size on PM formation was evaluated from comparison of two particle sizes of OR, specifically <125 µm and 212-300 µm. The results are presented in Fig. 3.

Particulate matter formation was lower for all ranges when using larger size particles. Large-size OR presented PM2.5 lower by 11%, and PM2.5-10 was 16% lower for small size OR. This illustrated the dependence of PM formation on the particle size. A likely explanation is the higher thermal inertia of the large particles that lowered the particle temperature during combustion and thus reducing the vaporization rate. Following the tendency of PM2.5 and PM2.5-10, PM10 underwent a reduction of 13%.
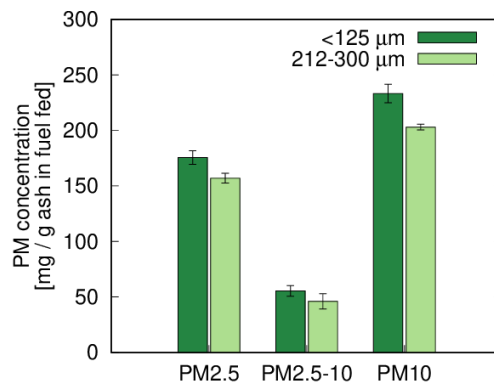

Figure 3. Particulate matter values of OR for two different size cuts (< 125 µm and 212-300 µm). Vertical bars represent 98% confidence interval statistical error.

3.3. Effect of co-Combustion on PM Formation

Since OR showed lower PM emissions among the two biomass fuels, biomass-coal co-firing trials were conducted for OR-TL blends. Both fuels were screened to < 125 µm and mechanically blended. Particulate matter concentration results are presented in Fig. 4. For 50-50 blends, PM2.5 resembled that of OR. In concentration basis (mg of PM per m^3 of air), PM2.5 of the blend (124 mg/m^3) was between OR (59.8 mg/m^3) and TL (329 mg/m^3). Moreover, PM2.5-10 of the same blend (166 mg / g ash in fuel fed) resembled the value of neat TL (149 mg/g ash in fuel fed) and was clearly above OR (56 mg/g ash in fuel fed).
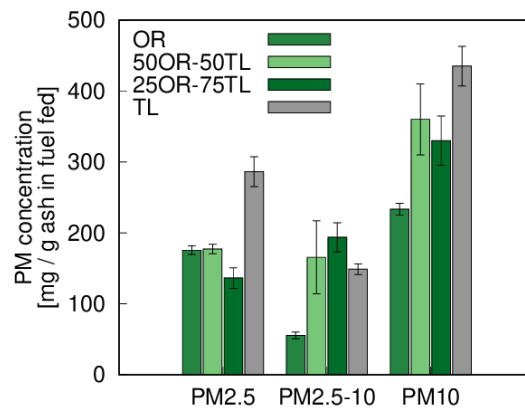

Figure 4. Particulate matter concentration for OR-TL blends. All fuels < 125 µm. Neat fuels are presented for comparison purposes. Vertical bars represent 98% confidence interval statistical error.

In summary, 25OR-75TL blends presented lower values of PM2.5 and PM10. As observed by Zellagui et al. [5], ratios of 25 wt.% of woody biomass in coal minimized submicron PM formation, whereas

values of 50 wt.% or 75 wt.% of biomass were closer to those of the neat biomass. The same trend was observed herein for OR-TL blends. Therefore, ratios of lower amount of biomass ought to be used in future investigation. Moreover, chemical analysis of the PM is necessary to infer on the mechanisms responsible for the high submicron and supermicron particles collected for the blends.

4. CONCLUSIONS

This study investigated the particulate matter (PM) formation during co-combustion of an abundant Turkish agricultural residue (olive residue - OR) with a Turkish lignite (Tunçbilek lignite - TL) currently used for power generation. Co-combustion trials were conducted at high temperature and high heating rate using a drop tube furnace. PM was categorised into PM2.5; PM2.5-10; and PM10. The main conclusions of this study can be summarized as follows:

- PM2.5 emission was lowest for olive residue (176 mg/g ash in fuel fed), followed by almond shell (214 mg/g ash in fuel fed) and Tunçbilek lignite (286 mg/g ash in fuel fed).

- PM10 emission was particularly low for olive residue (~200 mg/g ash in fuel fed), whereas almond shell and Tunçbilek lignite showed similar values (~400 mg/g ash in fuel fed).

- Particle size of olive residue did not have a significant effect on PM formation. This result is relevant since biomass-coal fired stations typically use biomass in particle sizes above those of coal. The results obtained herein suggest that the PM formation would not be significantly affected by the size of biomass. However, co-combustion trials using large-size biomass need to be conducted to ascertain this conclusion.

- Blends of OR-TL in 25-75 ratio showed lower values of both PM2.5 and PM10 as compared with 50-50 blends of the same fuels. This has practical implications in the sense that the equipment used to capture PM from lignite combustion can also be used for biomass-lignite co-firing since the share of PM2.5 is always lower for the blends.

ACKNOWLEDGEMENTS

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