

Response to the Anonymous Referee #2

We thank Anonymous Referee #2 for his/her very helpful comments. Our responses are listed below:

Comments:

This paper describes the chemical characteristics of the organic aerosols produced during the coal combustion process. The authors present interesting and potentially exciting results with a proposed mechanism to explain their findings. I recommend this work for publication if the following points are resolved satisfactorily.

Response: We thank the Reviewer for this comment, and look forward to the paper being published.

(1)Page 3352, line19, the authors show “in a coal-fired power plant the typical oxygen/coal ratio is 1.2”, but in this study the ratio is ranging from 8.6 to 30 which is away from the typical ratio, reason and explanation are required for this choice.

Response: The reasoning for using a drop-tube furnace is to precisely control the combustion conditions, such as temperature and gas composition. The temperature of drop-tube furnace is precisely controlled by an electric heating element; and gas composition is well controlled by mass flow controllers. Thus the heat and CO₂ generated from coal combustion should not be too significant to change the temperature profile and gas composition in the furnace. Thus, a lower coal feed rate (higher oxygen/coal ratio) was used in our experiments. Due to practical limitations, if an oxygen/coal ratio of 1.2 is used, then it is difficult for us to control temperature, gas composition in the furnace. If the oxygen concentrations were lower, the chances of incomplete combustion are higher, and thus more organic aerosols would be expected.

It should be noted that most drop-tube studies of coal combustion have used similar conditions (Taylor and Flagan, 1981;Quann et al., 1982;Haynes et al., 1982;Suriyawong et al., 2006;Wang et al., 2013).

For better clarification, here is a brief description of our drop-tube furnace: A Lindberg/Blue M, Model HTF55342C Tube Furnace is a square-shaped electric heating device

used as a laboratory-scale coal combustor. It is approximately 43.2cm long, 88.9cm wide, and 40.6cm tall. The Lindberg/Blue M, Model HTF55342C Tube Furnace, as seen in Figure 1, consists of 6 parts: a controller, a furnace, an inlet manifold system, a ceramic tube, an outlet dilution system, and a coal feeding system.

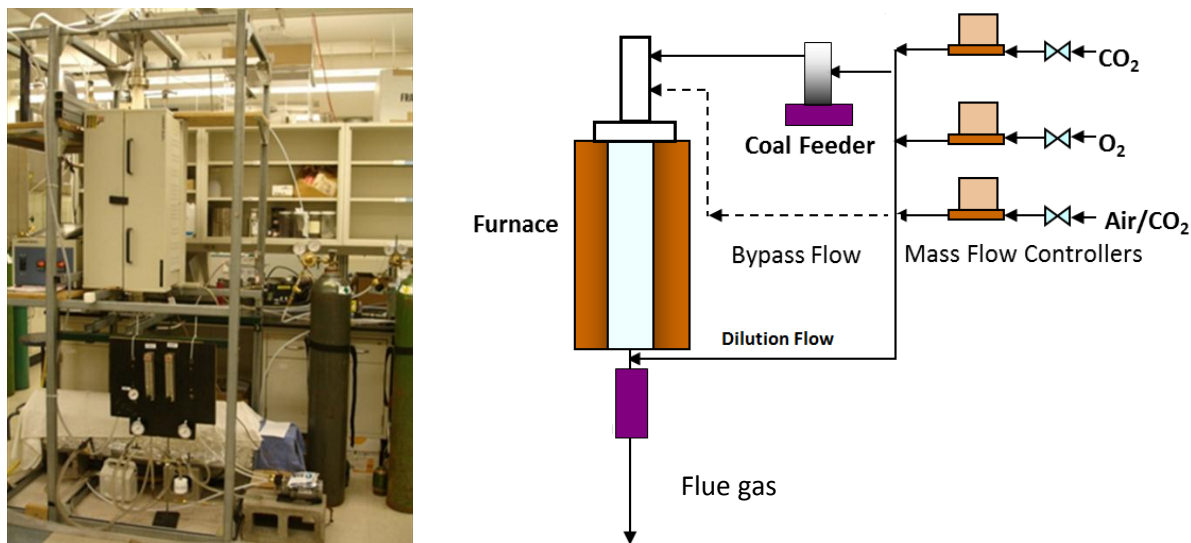


Figure 1. Picture and schematic drawing of the drop-tube furnace

Comments:

(2)Page 3354, line1-3, authors have used ATOFMS to measure the water soluble ions, such as K^+ of aerosols. I suppose ion chromatography is a better method used for the quantitative measurements of water soluble ions. Any specific reasons?

Response: ATOFMS cannot differentiate water-soluble potassium and non-water-soluble potassium. Thus, a monitor for aerosols & gasses in ambient air (MARGA) was used to detect water-soluble potassium in our field study. The focus of our study and the use of the ATOFMS in Figure 3C was to identify organic species signals from coal combustion aerosols: we found there were peaks at m/z of -45, -59 and -73 in ATOFMS spectra from coal ash. Those peaks are usually considered as the fragments of levoglucosan (Silva et al., 1999), which are the tracers of biomass burning aerosol. Thus this finding suggests that these three peaks may not be reliable

tracers of biomass burning aerosol, if coal combustion aerosols are also present around sampling site.

Comments:

(3)As the chemical composition of the coal varies from place to place, I miss the part where the authors have studied about the chemical composition for the coal used in this study, especially the potassium content.

Response: The proximate and ultimate analysis of PRB coal is shown below (Table 1 and 2). As we stated in the response to Dr. Healy’s comments, potassium content in coals vary greatly (up to 22.6 of K₂O in ash). And potassium is enriched in submicrometer particles after coal is combusted, due to the nucleation pathway of formation.

We thank Anonymous Referee #2 for his/her very helpful comments and suggestions.

Table 1. Proximate & ultimate analyses of Powder-River Basin (PRB) coal

Proximate Analysis (% wt)	
Volatile Matter ^a	48.3
Fixed Carbon ^a	42.9
Ash ^a	8.0
Moisture ^b	27.7
Lower Heating Value ^a (MJ/kg)	28.0
Ultimate Analysis (% wt)^a	
Carbon	67.3
Nitrogen	0.96
Hydrogen	4.58
Oxygen	19.9
Sulfur	0.57

Chlorine	0.01
Fluorine	0.796
^a - dry weight basis	^b - as-received basis

Table 2. Mineral composition of Powder-River Basin (PRB) coal

Mineral compound	Concentration (% wt)
SiO ₂	37.9
TiO ₂	1.2
Al ₂ O ₃	16.1
CaO	19.7
Fe ₂ O ₃	5.9
MgO	4.9
K ₂ O	0.6
Na ₂ O	1.4
P ₂ O ₅	1.7
Others	10.6

References:

Haynes, B. S., Neville, M., Quann, R. J., and Sarofim, A. F.: Factors Governing the Surface Enrichment of Fly-ash in Volatile Trace Species, *Journal of Colloid and Interface Science*, 87, 266-278, 1982.

Quann, R. J., Neville, M., Janghorbani, M., Mims, C. A., and Sarofim, A. F.: Mineral Matter and Trace-element Vaporization in a Laboratory-pulverized Coal Combustion System, *Environ. Sci. Technol.*, 16, 776-781, 1982.

Silva, P. J., Liu, D.-Y., Noble, C. A., and Prather, K. A.: Size and Chemical Characterization of Individual Particles Resulting from Biomass Burning of Local Southern California Species, *Environ. Sci. Technol.*, 33, 3068-3076, 10.1021/es980544p, 1999.

Suriyawong, A., Gamble, M., Lee, M.-H., Axelbaum, R., and Biswas, P.: Submicrometer Particle Formation and Mercury Speciation Under O₂-CO₂ Coal Combustion, *Energy Fuels*, 20, 2357-2363, 10.1021/ef060178s, 2006.

Taylor, D. D., and Flagan, R. C.: The Influence of Combustor Operation on Fine Particles from Coal Combustion, *Aerosol Sci. Technol.*, 1, 103-117, 10.1080/02786828208958581, 1981.

Wang, X., Michael Daukoru, S., Torkamani, S., Wang, W.-N., and Biswas, P.: Role of exhaust gas recycle on submicrometer particle formation during oxy-coal combustion, *Proc. Combust. Inst.*, 34, 3479-3487, <http://dx.doi.org/10.1016/j.proci.2012.07.049>, 2013.