Atmos. Chem. Phys. Discuss., 13, C1035–C1042, 2013
www.atmos-chem-phys-discuss.net/13/C1035/2013/


Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Characterization of organic aerosol produced during pulverized coal combustion in a drop tube furnace" *by* X. Wang et al.

**X. Wang et al.**

xiaofeiwang@go.wustl.edu

Received and published: 5 April 2013

Response to Dr. Robert Healy's Comments

We thank Dr. Robert Healy for his very helpful comments. Our responses are listed below:

Comment 1: "This article presents some interesting conclusions regarding the chemical mixing state of particles produced from the combustion of pulverised coal. It would be useful, however, to expand the discussion to cover differences between the single particle mass spectra reported here and those obtained for power generation-related coal combustion and domestic coal combustion in other previous studies (Pekney et al., 2006; Bein et al., 2006; Bein et al., 2007; Healy et al., 2010). Ambient domestic coal and wood combustion particles have been separated using single particle mass spectrometry previously, and their assignments conïňĄrmed through combustion experiments (Healy et al., 2010). "

Response: We thank Dr. Robert Healy for providing these papers and will cite them and add the following discussions in our future revised manuscript. The studies on ambient aerosol at Pittsburgh (Pekney et al., 2006;Bein et al., 2006;Bein et al., 2007) suggested that the particles with the signal of Na/Si/K/Ca/Fe/Ga/Pb were associated with coal combustion emissions. However, Healy et al. (2010) showed that Ga and Fe were not present in their coal combustion aerosols. In our study, we did observe Na/K/Ca/Fe and Pb in a type of ambient aerosols, which is considered to be originated from coal combustion. Na/K/Ca/Fe are present in the average ATOFMS spectrum of this type (Figure 6A in our paper). And Figure R1 (shown below, with m/z from 150 to 250 Da) also clearly shows the presence of Pb. But we did not observe significant peak of silicates and Ga. In addition, Liu et al. (2003) reported that coal combustion aerosols contain the signal of Li, while both Healy et al. (2010) and our study did not find this signal. Thus, it seems that every study is different from each other. But, we think all of these studies are correct; and the differences do reflect the complicated nature of coal combustion.

Chemical compositions of coal combustion aerosols can be very different. There are 3 factors:

Firstly, coal is a complex mixture. It almost contains every elements in the periodic table. Different coal types have very different compositions. For example, Table R1 shows the major compositions of coals that used in our lab. There are large differences among these coals.

Secondly, combustion conditions play very important roles in particulate matter formation. The detailed formation mechanisms are described in some literatures (Haynes et al., 1982;Damle et al., 1982;Zhuang and Biswas, 2001;Suriyawong et al., 2006;Wang et al., 2013). In short, most of the submicrometer particles are formed via the metal-oxide vaporization-nucleation pathway (Damle et al., 1982), which includes: metal-oxides are reduced at the char surface and produce relatively more volatile metal or its sub-oxides into gas-phase. Then those vapors undergo rapid re-oxidation to form stable metal-oxide nuclei, and subsequent growth by coagulation and condensation. Supermicrometer particles are usually formed through burst of remaining parts of char particles. This pathway of fine particle formation is very sensitive to the combustion temperature and gas composition. For example, Suriyawong et al. (2006) found that formation rates of submicrometer silicate particles can vary several orders of magnitude in different combustion conditions. Therefore, the chemical composition of submicrometer particles can be very different when combustion condition is changed.

Thirdly, air pollution control devices (APCDs) can greatly affect emissions of particulate matter. Electrostatic precipitator (ESP) or fabric filter (FF) bag house are widely used for industrial coal boilers. An ESP can usually remove 99.9% of total particulate mass from coal combustion. But there are penetration windows for submicrometer particles (Li et al., 2009). These penetration window may change when operating conditions of ESPs are changed. FF bag house typically has higher particle removal than ESP (Shendrikar et al., 1983). Thus, different selections of APCDs can result in very different characteristics of particulate emissions.

In conclusion, the characteristics of coal combustion aerosol can be very different due to the differences in coal types, combustion conditions and applications of ACPDs. We do not think there will be a single universal tracer for coal combustion aerosol. Therefore, in order to identify them, more information will be needed. In this study, we want to use the organic signatures of coal combustion aerosol, as well as the inorganic signatures, coal usage in the region and satellite data to identify a group of ambient aerosol which was very likely from coal combustion.

Comment 2: “Potassium content was found to be very low in domestic coal combustion particles relative to wood combustion particles in that case. Domestic coal combustion particle composition has also been investigated more recently using an aerosol mass spectrometer (Dall'Osto et al., 2012)."

Response: Domestic coal combustion usually has a much lower combustion temperature than pulverized coal combustion. The lower temperature may inhibit the vaporization rate of metal elements, such as potassium. Thus the mass concentration of these elements should be much lower from domestic coal combustion than pulverized coal combustion. For pulverized coal combustion, Quann et al. (1982) found there was 2.5% of K2O in submicrometer ash particles, with a enrichment factor of 4.00. ATOFMS is extremely sensitive to potassium. This amount of potassium should lead to a dominant peak at m/z 39. Moreover, potassium content in coal varies a lot. For example, some potassium-enriched coals contain up to 22.6% of K2O in its ash (Zhou et al., 2010).

Dall'Osto et al. (2012) measured aerosol from domestic coal combustion using AMS. The result is quite interesting. It seems that their AMS spectrum from coal combustion is similar to this study. Better comparison can be achieved if we know the coal type (with proximate and ultimate analysis of coal) and combustion conditions (such as dimensions of the burner, combustion temperature and air flow rate).

Again, we thank Dr. Robert Healy for his very helpful comments.

References:

Bein, K. J., Zhao, Y., Pekney, N. J., Davidson, C. I., Johnston, M. V., and Wexler, A. S.: Identification of sources of atmospheric PM at the Pittsburgh Supersite - Part II: Quantitative comparisons of single particle, particle number, and particle mass measurements, Atmospheric Environment, 40, S424-S444, 10.1016/j.atmosenv.2006.01.064, 2006.

Bein, K. J., Zhao, Y., Johnston, M. V., and Wexler, A. S.: Identification of sources of



ACPD

13, C1035–C1042, 2013

atmospheric PM at the Pittsburgh supersite - Part III: Source characterization, Atmospheric Environment, 41, 3974-3992, 10.1016/j.atmosenv.2007.01.039, 2007.

Dall'Osto, M., Ovadnevaite, J., Ceburnis, D., Martin, D., Healy, R. M., O'Connor, I. P., Sodeau, J. R., Wenger, J. C., and O'Dowd, C.: Characterization of urban aerosol in Cork City (Ireland) using aerosol mass spectrometry, Atmos. Chem. Phys. Discuss., 12, 29657-29704, 10.5194/acpd-12-29657-2012, 2012.

Damle, A. S., Ensor, D. S., and Ranade, M. B.: Coal Combustion Aerosol Formation Mechanisms: A Review, Aerosol Sc. & Tech., 1, 119-133, 10.1080/02786828208958582, 1982. Haynes, B. S., Neville, M. N., 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.

Healy, R. M., Hellebust, S., Kourtchev, I., Allanic, A., O'Connor, I. P., Bell, J. M., Healy, D. A., Sodeau, J. R., and Wenger, J. C.: Source apportionment of PM2.5 in Cork Harbour, Ireland using a combination of single particle mass spectrometry and quantitative semi-continuous measurements, Atmospheric Chemistry and Physics, 10, 9593-9613, 10.5194/acp-10-9593-2010, 2010.

Li, Y., Surlyawong, A., Daukoru, M., Zhuang, Y., and Biswas, P.: Measurement and Capture of Fine and Ultrafine Particles from a Pilot-Scale Pulverized Coal Combustor with an Electrostatic Precipitator, J. Air Waste Manage. Assoc., 59, 553-559, 10.3155/1047-3289.59.5.553, 2009.

Liu, D.-Y., Wenzel, R. J., and Prather, K. A.: Aerosol time-of-flight mass spectrometry during the Atlanta Supersite Experiment: 1. Measurements, Journal of Geophysical Research: Atmospheres, 108, 8426, 10.1029/2001jd001562, 2003.

Pekney, N. J., Davidson, C. I., Bein, K. J., Wexler, A. S., and Johnston, M. V.: Identification of sources of atmospheric PM at the Pittsburgh Supersite, Part I: Single particle analysis and filter-based positive matrix factorization, Atmospheric Environment, 40, S411-S423, 10.1016/j.atmosenv.2005.12.072, 2006.

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.

Shendrikar, A. D., Ensor, D. S., Cowen, S. J., Woffinden, G. J., and McElroy, M. W.: Size-dependent penetration of trace elements through a utility baghouse, Atmospheric Environment (1967), 17, 1411-1421, 10.1016/0004-6981(83)90294-9, 1983.

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, 2006.

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. Zhou, K., Xu, M., Yu, D., Liu, X., Wen, C., Zhan, Z., and Yao, H.: Formation and Control of Fine Potassium-Enriched Particulates during Coal Combustion, Energy Fuels, 24, 6266-6274, 10.1021/ef101190x, 2010.

Zhuang, Y., and Biswas, P.: Submicrometer Particle Formation and Control in a Bench-Scale Pulverized Coal Combustor, Energy Fuels, 15, 510-516, 10.1021/ef000080s, 2001.

---

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 3345, 2013.

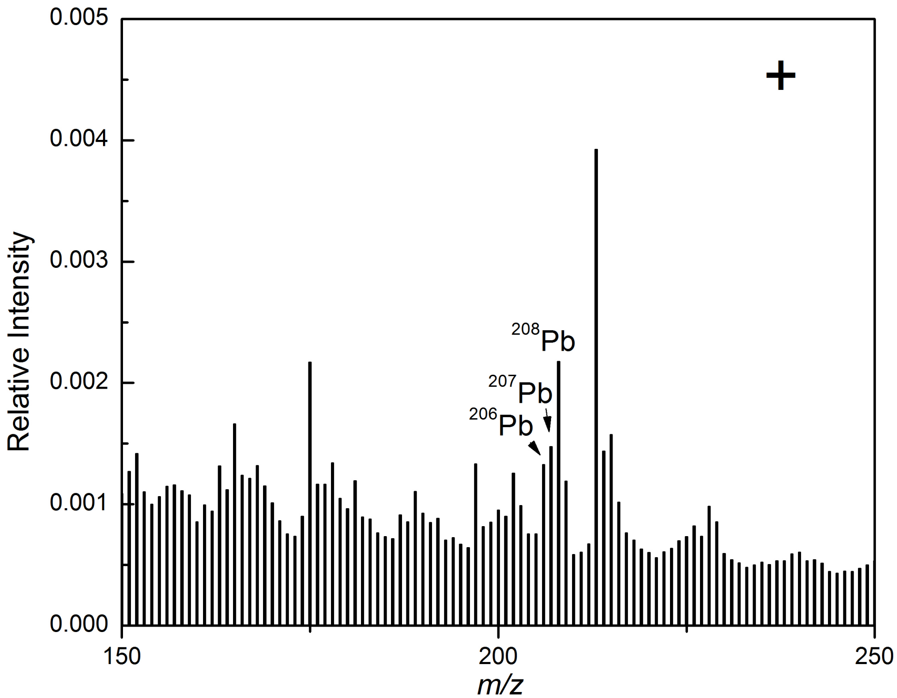


**Fig. 1.** Average mass spectra of a dominant type (coal) of ambient aerosol in Shanghai using ATOFMS (m/z from 150 to 250 Da)



ACPD
13, C1035–C1042, 2013



Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

| Coal Name | Proximate analysis | | | | Ultimate Analysis | | | | | | $Q_{net,d}$ | ASTM Rank |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | $Moisture_{ad}$ | $Ash_d$ | $VM_d$ | $FC_d$ | $S_d$ | $C_d$ | $H_d$ | $N_d$ | $O_d$ | $Cl_d$ | MJ/kg | |
| **American Coals** | | | | | | | | | | | | |
| Powder River Basin (PRB) Coal | 27.7* | 8.0 | 48.3 | 42.9 | 0.6 | 67.3 | 4.6 | 1.0 | 19.9 | 0.010 | 28.0 | Sub-bituminous |
| Illinois #6 Coal | 13.5* | 10.0 | 42.0 | 48.0 | 3.5 | 71.0 | 5.0 | 1.3 | 9.1 | 0.110 | 29.6 | Bituminous |
| **Chinese Coals** | | | | | | | | | | | | |
| S01 | 8.4 | 44.6 | 23.6 | 31.7 | 1.5 | 38.9 | 2.4 | 0.6 | 12.0 | 0.012 | 13.5 | Lignite |
| S02 | 17.8 | 33.3 | 27.0 | 39.7 | 0.6 | 49.2 | 2.8 | 0.9 | 13.3 | 0.014 | 17.4 | Lignite |
| S03 | 0.7 | 26.4 | 16.1 | 57.5 | 2.3 | 62.6 | 3.2 | 0.9 | 4.6 | 0.017 | 24.7 | Sub-bituminous |
| S04 | 3.8 | 23.0 | 10.5 | 66.5 | 2.8 | 67.0 | 2.6 | 1.0 | 3.7 | 0.031 | 26.0 | Anthracite |
| S05 | 0.9 | 21.9 | 20.5 | 57.6 | 2.0 | 67.2 | 3.9 | 1.0 | 4.1 | 0.039 | 27.1 | Bituminous |
| S06 | 4.5 | 30.6 | 9.6 | 59.8 | 2.9 | 61.1 | 1.9 | 0.7 | 2.9 | 0.047 | 23.2 | Anthracite |
| S07 | 9.1 | 24.8 | 27.8 | 47.4 | 0.5 | 57.7 | 3.5 | 0.9 | 12.6 | 0.361 | 21.9 | Sub-bituminous |
| S08 | 2.6 | 25.0 | 15.0 | 60.0 | 1.7 | 64.2 | 3.1 | 0.9 | 5.2 | 0.024 | 25.2 | Sub-bituminous |
| S09 | 4.1 | 30.5 | 24.3 | 45.2 | 0.5 | 55.7 | 3.3 | 0.9 | 9.2 | 0.072 | 21.8 | Sub-bituminous |
| S10 | 15.7 | 5.7 | 34.4 | 59.9 | 0.5 | 74.5 | 4.4 | 0.9 | 14.0 | 0.014 | 28.5 | Bituminous |
| S11 | 13.1 | 18.9 | 30.0 | 51.2 | 0.5 | 64.0 | 3.7 | 0.9 | 12.0 | 0.014 | 24.7 | Sub-bituminous |
| S12 | 10.2 | 8.0 | 36.3 | 55.8 | 0.5 | 71.8 | 4.5 | 0.9 | 14.3 | 0.009 | 27.9 | Bituminous |
| **Typical Indian Coals** | | | | | | | | | | | | |
| Typical (Low Grade, F) | 4.0 | 50.0 | 19.3 | 30.0 | 0.7 | 43.2 | 3.4 | 1.0 | - | - | 13.0 | Lignite |
| Typical (Medium Grade, D) | 15.0 | 30.0 | 19.8 | 50.0 | 0.2 | 31.8 | 2.4 | 0.8 | - | - | 21.0 | Sub-bituminous |
| **Selected Indian Coals** | | | | | | | | | | | | |
| Dadri | - | 38.2 | - | - | 0.5 | 40.3 | 4.2 | 0.9 | 15.9 | - | - | - |
| Rihand | - | 43.2 | - | - | 0.4 | 37.7 | 3.3 | 0.7 | 14.7 | - | - | - |
| Singrauli | - | 26.3 | - | - | 0.3 | 50.2 | 4.8 | 1.1 | 17.3 | - | - | - |
| Chandrapur | 5.0 | 47.0 | - | - | 0.8 | 37.7 | 2.7 | 1.1 | 5.8 | - | 15.3 | - |
| Dahanu | 5.9 | 38.5 | - | - | 0.4 | 42.4 | 3.7 | 0.8 | 14.2 | - | 16.7 | - |
| Nevyeli Lignite | 47.0 | 7.0 | - | - | 1.5 | 26.1 | 2.3 | 0.2 | 16.3 | - | 9.3 | - |
| Kutch Lignite | 36.0 | 15.0 | - | - | 2.3 | 28.3 | 3.0 | 0.9 | 13.9 | - | 12.1 | - |
| VM = Volatile Matter, FC - Fixed Carbon, ad air-dried, d dry, *as-received | | | | | | | | | | | | |

**Fig. 2.** Proximate and ultimate analysis of coals that used in Aerosol & Air Quality Research Laboratory (AAQRL) at Washington University in St. Louis