Response to the Anonymous Referee #1

We thank the Anonymous Referee #1 for his/her very helpful comments. Our responses are listed below:

Comments:

The paper of Wang et al. (2013) is very timely - it attempts to characterise the organic aerosols produced during coal combustion. The abstract is also very interesting, reporting coal combustion is a main source of aerosols, which can be wrongly assigned to biomass burning.

Response: We thank the Anonymous Referee #1 for his/her comments. As he/she said, this work is very timely, because there are few studies that have reported AMS/ATOFMS data directly measured in exhausts from pulverized coal combustors. The scientific community need these source data to determine origins of atmospheric aerosols.

Comments:

However, the mass spectrometry analysis of this study is very poor. High resolution mass spectra of AMS are not presented, and the presentation of the ATOFMS data is also not good enough for ACP standard.

I suggest to deeply revise the current manuscript - which is not suitable for ACP at this stage.

Response: We politely disagree and feel that AMS data presented are sufficient and in support of some of our findings. This study is focused on a detailed characterization of fine particles generated during pulverized coal combustion using advanced techniques including aerosol mass spectrometry. We found some similarities in the characteristics of the aerosol from coal and biomass combustors. We feel may lead to incorrect apportionment of coal combustion particles to biomass burning sources. A field study in Shanghai did find a major fraction of such an aerosol, very similar to biomass combustion aerosols. However, the detailed analysis strongly suggests that this type of aerosol was probably originated from coal combustors. It proves that the incorrect apportionment could occur in a field study. We think that the data presented in this paper is sufficient to serve this purpose. While we agree that we could do more complicated analyses of the AMS/ATOFMS data; we feel it is not necessary to support the main findings of this paper.

Comments:

I shall suggest the following:

-Introduction: The introduction is very short, and need to be expanded. There are dozens papers on biomass-coal-combustion AMS-ATOFMS datasets, which are not even mentioned in the current manuscript. Pg. 3347 line 4-5 is a repetition of previous page. The objective of the study is not clearly stated.

Response: We thank the reviewer for his/her suggestions. We will expand the introduction in the revised manuscript. There are many papers on biomass combustion; however very few (to the best of our knowledge) that report coal-combustion AMS-ATOFMS datasets. Dr. Robert Healy mentioned some of them (Dall'Osto et al., 2012;Healy et al., 2010;Pekney et al., 2006;Bein et al., 2006;Bein et al., 2007) in his short comments (http://www.atmos-chem-phys-discuss.net/13/C923/2013/acpd-13-C923-2013.pdf). We now discuss those papers in our revised introduction.

Most of those papers only report some experiments of domestic coal combustion, which are very different from industrial coal combustion. And the combustion conditions were not been well controlled. For example, Dall'Osto et al. (2012) did not report the coal types, gas flow rate and combustion temperature, which are the key parameters when studying combustion. In our paper, the experiments of pulverized coal combustion had been conducted in a drop-tube furnace, with strict control of the combustion condition very precisely. We will clearly state these points in our revised manuscript.

We will revise Pg. 3347 line 4-5 as follows:

The objective of this paper is to use some advanced techniques of aerosol mass spectrometries to characterize fine aerosol particles generated from pulverized coal combustion. Some similarity between coal combustion aerosol and biomass burning aerosol had been found, which may lead to incorrect apportionment of coal combustion particles to biomass burning sources. An example of field study (in Shanghai) was given. And a formation mechanism of organic aerosol was proposed.

Comments:

- *methodology. Running ART2a with a vigilant factor of 0.7 leads to very broad particle types, which I am not sure can be correctly classified. To my knowledge, most of ATOFMS studies in the literature are carried out with at least 0.85.*

Response: ART2a usually produce large number of particle types. A typical step after ART2a classification is to manually merge similar particle types into one (Moffet et al., 2008). We tried many values. A vigilant factor of 0.85 led to more than 2000 particle types for our data. It would require lots of work on manual merging similar groups. Thus, many subjective errors may be introduced during this process. To reduce such errors, we used a vigilant factor of 0.7 which resulted in about 500 particle types. Following this, manual merging was carried out.

Comments:

- *results:*

- *pg 3352 Figure S2 is by far not uni modal as stated, and actually several differences (see the fine and coarse modes) can be seen when the mentioned ratio is changed.*

Response: Below is Figure S2. The size range of Figure S2 is from 9.5 to 425 nm. In this size range, only one peak can be identified. Thus we called it uni-modal distribution. It is very possible that there are other peaks at larger sizes, as is well known from broader size range measurements. However, the focus in this paper was on fine particles whose diameters were less than 500 nm.

Yes, when oxygen/coal ratio was changed from 30.1 to 8.6, particle concentrations slightly increased, although it was not significant. We will use more accurate words to describe Figure S2 in the revised manuscript.

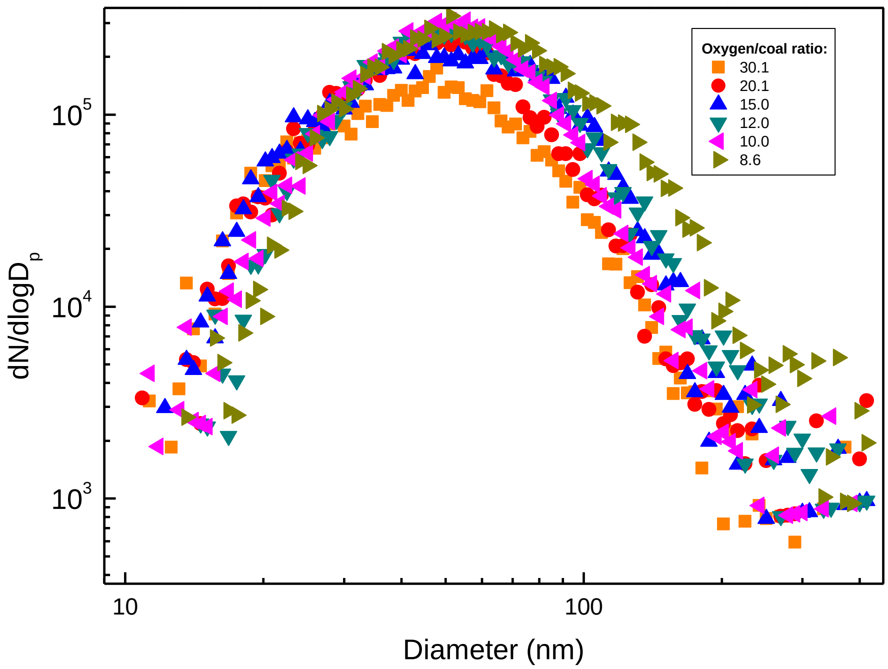

Figure S2. Size distribution of particles from coal combustion under various oxygen/coal ratios

Comments:

-pg 3354 Figure 3c. I am not sure this is valid, as it does not represents real ambient particles. This particle is simply a dust particle, with potassium,calcium and phospate.

Response: Figure 3C presents a source study. As stated in our paper, coal combustion particles collected on a quartz filter were extracted by pure water in an ultrasonic bath. The extract was atomized to produce droplets. A diffusion dryer was used to dry the droplets. The dried aerosol was introduced into the ATOFMS and the mass spectra were obtained (Figure 3c). In Figure 3C, the major finding is the presence of the peaks at m/z of -45, -59 and -73, which are known as biomass burning aerosol tracers.

Figure 3C is an average ATOFMS spectrum of coal combustion aerosols, which were collected from our drop-tube coal combustor.

To clarify, these are particles collected at the exit of the drop-tube coal combustor. The air used is laboratory grade, particle free air. Hence, there is no issue of ambient particles being in the sample.

Comments:

- *pg 3354 Fig S3 shows very large error bars. If t-test is applied, I feel the three bars are the same*

Response: We did the t-test. The p-value between pure air (0/100) and 40%N2 addition (40/60) is less than 0.0001, which is considered to be statistically significant. The p-values between (0/100) and (10/90), (10/90) and (20/80), (20/80) and (40/60) are 0.27, 0.50 and 0.04, respectively. Thus, generally speaking, the trend is significant: O/C ratio increased, when more N_2 was added into the system. We will add the above information and discussion in the revised manuscript.

Comments:

-pg 3368. what is the point of using HR-AMS if not presenting HR data? At this stage only a short weak description of mz 60 and 73 is carried out.

Response: We presented O/C ratios, which were calculated from HR data.

The reason for using HR data is that AMS cannot provide detailed molecular information on organics, while HR data can provide elemental compositions for each individual peak. This information can help in a better understanding of these organic compounds. However, in our case, we already had detailed molecular information by using GC-MS. A large group of organics were identified. An analysis on HR data may not further our understanding of organics produced from coal combustion. Thus, we did not include such analysis.

Comments:

-pg 3374. As pointed out by Dr. Healy (see short comment) - a more detailed description of the ATOFMS results should be attempted.

Response: Yes, we will add more discussions on ATOFMS results, as we stated in our response to Dr. Healy's comments.

First, we will add the discussion on comparison between our results and other studies (Bein et al., 2006;Bein et al., 2007;Dall'Osto et al., 2012;Healy et al., 2010;Liu et al., 2003;Pekney et al., 2006). These studies show that coal combustion aerosols from different studies have different ATOFMS spectrum patterns. It may reflect the complicated nature of coal combustion, since many factors, including coal types, combustion conditions and use of air pollution control devices (APCDs) have large impact on the resultant aerosol characteristics. We will discuss them in detail in our revised manuscript, since they are extremely important to understand the nature of coal combustion aerosols in the atmosphere.

Secondly, we will compare our results with the ambient data from other studies (Bein et al., 2006;Bein et al., 2007;Dall'Osto et al., 2012;Healy et al., 2010;Liu et al., 2003;Pekney et al., 2006). There is a new ACPD paper which describes a class of organic aerosols from coal combustion in Beijing, China (Sun et al., 2013). We will also compare our results with that study.

Again, we thank Anonymous Referee #1 for his/her very helpful comments. This has vastly improved our manuscript, and we look forward to it being accepted.

References:

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