



Qualitative Evaluation of m/z 85, 87, and 133 Signals in Organic Aerosol Mass Spectra of Fly Ash Produced by Coal Combustion

Satoshi Irei¹, Akio Shimono², Toshihide Hikida², Koji Kuramoto³, Yoshizo Suzuki³, Akinori Takami^{1*}

¹ National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki, 305-8506, Japan

² Shoreline Science Research, Inc., Tokyo 192-0045, Japan

³ Energy Technology Research Institute, National Institute of Advanced Industrial Science and Technology, 16-1, Onogawa, Tsukuba, Ibaraki, 305-8569, Japan

ABSTRACT

Quantitative estimation of particulate matter (PM) pollution from coal combustion, which is one of the major anthropogenic emission sources in China, is urgently needed to better understand transboundary pollution in the East Asian region. As a first step, we conducted laboratory experiments for the mass spectrometric characterization of fly ash from coal combustion using a fluidized bed reactor. Here, we report detection of notable signals at m/z 85, 87, and 133 in mass spectra for organic species obtained by an Aerodyne quadrupole aerosol mass spectrometer (AMS). Nine different coals, six of which were mined in north-east Asian region, were tested with three different combustion temperatures. The results showed that signals at m/z 85, 87, and 133 were significantly larger than those at the adjacent m/z , with similar observations having been made in different field studies carried out in western Japan. The m/z 85 to m/z 87 ratios were reproducible over the coals tested, suggesting the potential usefulness of these for fingerprinting coal combustion PM. The average ratio with the standard error of the mean was 2.8 ± 0.1 . While the m/z 133 to m/z 87 ratios varied more, the mean ratio with the standard error of the mean was 1.5 ± 0.2 , and the results were still reproducible. A comparison of the m/z 85 and 133 to m/z 87 ratios from other studies that also used AMS suggests that the ratios obtained in the current work are distinctive from those for vehicular emissions, plastic burning, and cooking emissions, but close to those for biomass burning. Despite this similarity, the results here still offer useful information for source identification of PM with the use of AMS measurements.

Keywords: Combustion aerosols; Emission characterization; Source profiles; Fly ash; Aerosol mass spectrometer.

INTRODUCTION

Transboundary pollution is a great concern in the countries of the East-Asian region in these days since air masses coming over the Chinese continent often carry desert dust and anthropogenic pollutants and the content in the air masses significantly influences the air quality of the neighboring countries. Fine particulate matter (PM) is one of the anthropogenic pollutants often long-range transported.

To date, we have observed chemical composition of fine PM at the Fukue and the Cape Hedo monitoring stations representing typical rural sites in the western Japan (Takami *et al.*, 2005; Takami *et al.*, 2007; Miyoshi *et al.*, 2013) using quadrupole aerosol mass spectrometers (AMS), which are a type of the commonly used instruments for high time

resolution measurements of fine PM pollution in these days. In our recent field studies in the western Japan, we observed unusually high loadings at m/z 85, 87 and 133 in organic aerosol mass spectra (Fig. 1), and these characteristic spectra were often found in the mass spectra for oxygenate organic aerosols extracted by PMF analysis. Takegawa *et al.* (2009) reported that remarkable signals at m/z 39, 41, 85, 87 and 133 in organic aerosol mass spectra were observed during the CAREBEIJIN campaign conducted in the summer of 2006 in China. The similar observations between their and our field studies are probably indicative for the influence of the transboundary pollution. Takegawa *et al.* (2009) speculated that the signals were likely ions of alkali metals such as, potassium (m/z 39 and 41 for its second dominant isotope), rubidium (m/z 85 and 87 for its second dominant isotope) and cesium (m/z 133) as well as that the source may be biomass/biofuel burning. Although their speculation is feasible, coal combustion, another possible source for these signals also needs to be evaluated.

Coal is one of the major fossil fuels used, and its consumption in China is continuously increasing since

* Corresponding author.

Tel.: +81-29-850-2509; Fax: +81-29-850-2579

E-mail address: takamia@nies.go.jp

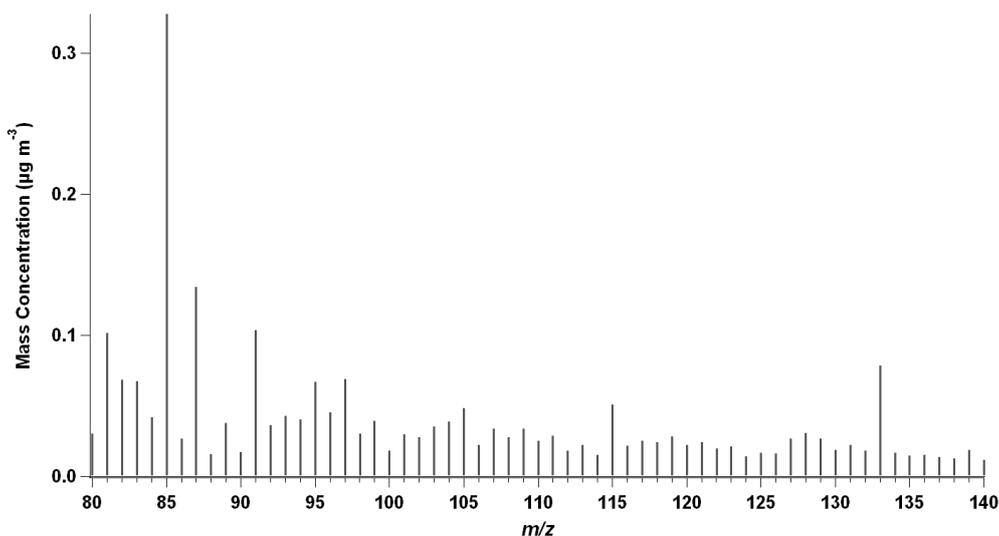


Fig. 1. Organic aerosol mass spectra observed in the field study in March 2009 at the Fukue monitoring station. *Spectra range shown is from m/z 80 to m/z 140.*

1980 (Ohara *et al.*, 2007) and accounts for nearly 50% of the world wide consumption today (U.S. Energy Information Administration, 2013). Thus, there must be significant impact of coal combustion on PM pollution transported from the Chinese continent. Although better understanding of its impact is urgent, no source characterization study for coal combustion using AMS has been reported to date.

The objective here is qualitative evaluation of m/z 85, 87, and 133 signals in organic aerosol mass spectra obtained by AMS analysis for sub-micron fly ash produced from coal combustion. Laboratory experiments of coal combustion were conducted using a fluidized bed reactor, and mass spectrometric analysis of sub-micron fly ash from coal combustion was carried out using an AMS.

EXPERIMENT

A fluidized bed reactor was used for particle generation from coal combustion (Fig. 2). The reactor consists of three main components: an electric heater (Trans Temp Transparent Tube Furnace, Thermcraft Inc.), a customary made 37 i.d. mm \times 350 mm height cylindrical quartz reaction vessel (Tokyo Motoyama Shokai), and a customary made screw feeder (Suzuki Shoko, Co. Ltd.). The electric furnace assists to raise temperature at which coal ignites. The reaction vessel was made of quartz and coated with thin gold to minimize radiative loss of heat. The bottom of the vessel was sealed with a fritted quartz plate. Approximately 4 cm top of the fritted glass was filled with 1 mm sieved silica sands. A mixture of pure oxygen (Tomoe Shokai, Corp.) and pure nitrogen gases (Tomoe Shokai, Corp.) was used for the combustion experiments. The gas mixture was supplied to the bottom of the vessel via the fritted quartz plate at the flow rate of 3.5 L/min. Prior to the combusting experiments, the oxygen content of the gas mixture was balanced to 21% by monitoring O_2 mixing ratio with a gas analyzer described later. Temperature and relative pressure inside the vessel were monitored by a thermocouple and a

water manometer. During the experiments, the pressure inside was maintained at approximately 1 kPa above the atmospheric pressure. The gas and fine particles from the coal combustion was exhausted from the top of the reaction vessel. 200 μm –100 μm sieved ground coal was used for the series of the coal combustion experiments. The ground coal was supplied from the upper side of the reactor vessel at the rate of 100 mg/min approximately. Unless otherwise noted, the combustion experiments were conducted under our standard condition, which was 5% residual O_2 under 1123 K combustion temperature. In addition to the experiments under the standard condition, dependence on combusting temperature was also tested.

The exhaust from the reactor was passed through a customary-made glass cyclone separator first for removing large size particles (i.e., particles of 20 μm or larger aerodynamic diameter). The outflow from the first cyclone was then branched into two, a flow to a gas reservoir for analysis of gas components and another to AMS. The former flow of approximately 1 L/min was consumed by the simultaneous analysis of mixing ratios for CO , CO_2 , SO_2 , NO , and O_2 using an infra-red gas analyzer (IR-400, Yokogawa Electric Corp.). The latter flow was passed through the second cyclone separator (URG-2000-30ED, URG Corp.) at the flow rate of 3 L/min for further size segregation of particles to smaller than 2.5 μm aerodynamic diameter (i.e., $PM_{2.5}$). The $PM_{2.5}$ was then measured by an AMS (Aerodyne Inc.) for its mass spectrometric analysis.

The details of the instrumentation and the method for quantitative analysis of non refractory components of fine particles ($\sim PM_{1.0}$) by AMS are described elsewhere (Jayne *et al.*, 2000; Jimenez *et al.*, 2003; Allan *et al.*, 2004). For our AMS measurements, the flash vaporizer temperature was set to 873 K, which approximately corresponds to the heater temperature under usual operation for field studies. The dwell time for each mode, the dead time between the two modes, and the data saving interval for both modes were 0.5 minutes, 0.05 minutes, and 2 minutes, respectively. Since

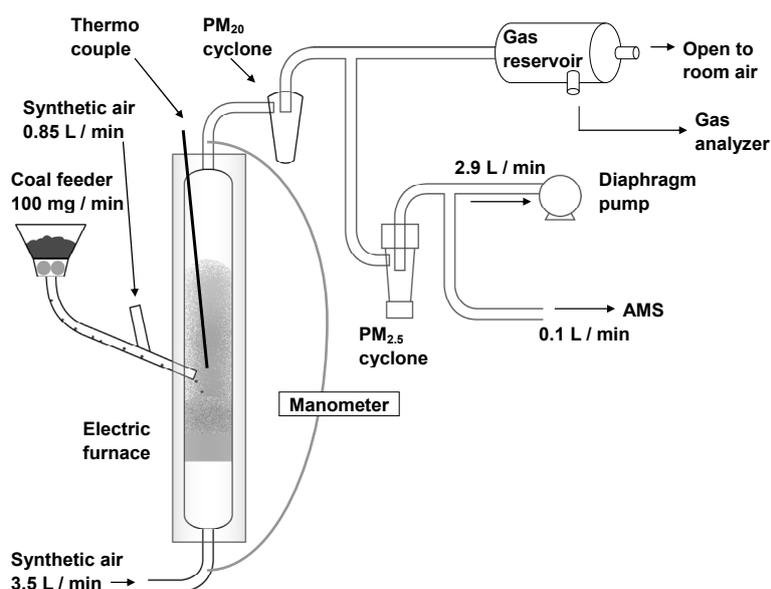


Fig. 2. Schematic setup for coal combustion experiments using fluidized bed reactor.

sensitivity of the AMS did not decrease significantly during the experiments, correction for the sensitivity deterioration was not made here.

Nine coals were chosen from three types (sub-bituminous, bituminous, and anthracite) for coal combustion experiments (Table 1). The 6 out of 8 were mined in the north-east Asian region (the coal ID from A to F), and the rest of 3 coals, G, H, I were in the south-east Asian, African, and Oceania regions, respectively.

RESULTS AND DISCUSSION

Coal Combustion Experiment

As an example, time series plot for combustion temperature and mixing ratios for O_2 , CO_2 , CO , NO , and SO_2 during the combustion experiment of the coal B is shown in Fig. 3. AMS measurements were performed as mixing ratios of the gaseous chemical species were almost under steady-state at 1023 K, 1123 K, and 1173 K, respectively. These measurement periods by the AMS were shown in vertical grey areas in Fig. 3. Measurement results showed that residual O_2 level during the combustion experiments was maintained between 5.7% and 5.2%, stable through the experiment. For trace gas species, higher combustion temperature, lower SO_2 and CO as well as higher NO and CO_2 mixing ratios. This is likely explained by that the coals were more well oxidized under higher temperature, resulting in lower mixing ratios of products that can be further oxidized, such as SO_2 and CO , and in higher mixing ratios of products that are stable oxidation state at the high temperature, such as NO and CO_2 . Larger fluctuation in mixing ratios for SO_2 and CO produced at the combustion temperature of 1023 K than those at 1123 K and 1173 K also suggests better oxidation at higher temperature.

Signals at m/z 85, 87, and 133

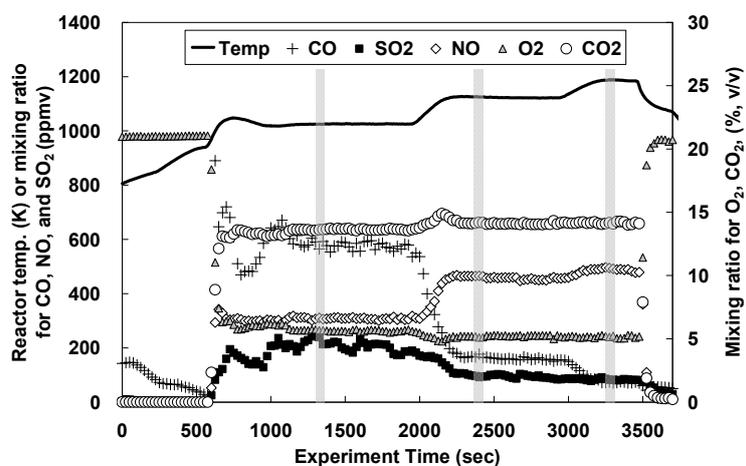
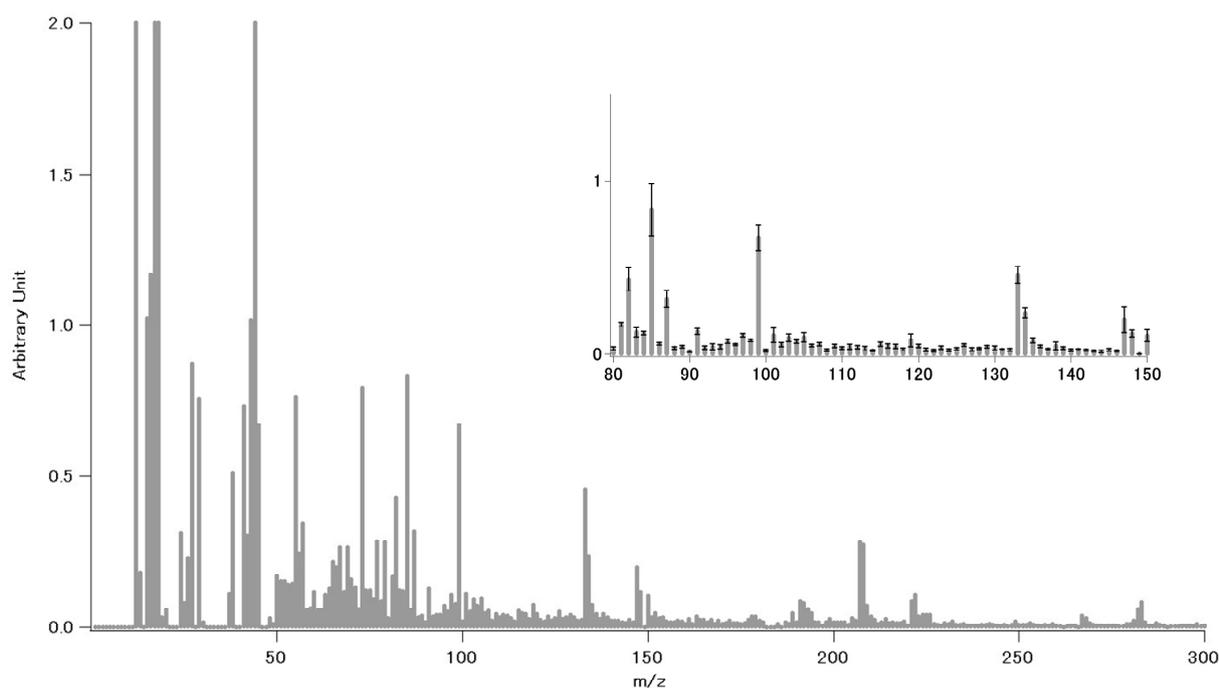
The blank measurements allowed us to confirm that

signals at m/z 85, 87 and 133 were low enough. Based on the blank measurements, detection limit (3 times the standard deviation) for m/z 85, 87, and 133 were 0.02, 0.01, 0.01 $\mu g/m^3$, respectively. Although results are not shown here, we confirmed that changing the heater temperature between 1173 K and 1023 K did not affect notable appearance of signals at m/z 85, 87, and 133.

As an example, the average organic aerosol mass spectra over 10 min. measurements (i.e., $n = 5$) from the combustion experiments of the coal A at 1123 K are shown in Fig. 4. Note that the error bars shown in Fig. 4 are standard errors of the mean based on the 5 replicate measurements (i.e., the standard deviation over the square root of 5). As seen, the signals at m/z 85, 87 and 133 were significantly larger than those of adjacent m/z , the similar observations in the field studies conducted before. The notably higher intensities at m/z 85, 87, and 133 than the signals at their adjacent m/z were observed in the organic mass spectra for all combustion experiments, although the magnitudes of the signal intensities considerably varied. As referred earlier, such remarkably high signals at m/z 85, 87, and 133 were also observed during our field studies in the western Japan (Fig. 1) as well as during the field studies conducted in Beijing, China (Takegawa *et al.*, 2009). Takegawa *et al.* (2009) speculated that the signals observed during the field studies above were probably alkali metals that can be detected by AMS, such as rubidium, cesium and their isotopes. This hypothesis seems to plausibly explain our field observations of the remarkable signals at m/z 85, 87, and 133 in the long-range transported particles that have undergone atmospheric oxidation or “aging” to some extent. If the signals were ions of organic substances, oxidation would alter the parent substances of these ions, resulted in variation of the signal strength over time. In future, further source studies combining element/compound specific analysis are inevitable for thorough evaluation of the parent substances giving the signals at m/z 85, 87, and 133. Nevertheless, this unique

Table 1. Number of coal combustion experiment tested.

Coal ID	Region of product	Type	Combustion temperature		
			1023 K	1123 K	1173 K
A	North east Asia	Bituminous	3	3	5
B	North east Asia	Bituminous	2	1	2
C	North east Asia	Anthracite	1	1	1
D	North east Asia	Anthracite	1	1	1
E	North east Asia	Bituminous	1	1	1
F	North east Asia	Bituminous	1	0	0
G	South east Asia	Sub-bituminous	1	1	2
H	Africa	Bituminous	1	1	2
I	Oceania	Bituminous	1	1	0

**Fig. 3.** Time series of combustion temperature and mixing ratios for O₂, CO₂, CO, NO, and SO₂ during the combustion experiment of the coal B. Three vertical grey bars in the figure indicate measurement periods by AMS at three different combustion temperatures.**Fig. 4.** Mean organic mass spectra ($n = 5$ or higher) of aerosol emitted from coal A combustion at 1123 K. The inset shown is the same mass spectra in expanded scale. The error bars shown in the inset are standard errors of the mean.

feature in organic mass spectra may potentially be used to identify PM from coal combustion.

As an example, temperature dependence of m/z 85, 87, and 133 signals tested with the coal A combustion is shown in Table 2. For evaluation of the temperature dependence, m/z 85 (or m/z 133) ratio to m/z 87 (referred to as 85/87 and 133/87 ratios hereafter) was used to cancel out variations of emission strength over the combustion experiments. As seen, the 85/87 ratios ranged from 2.61 to 3.21 with the mean \pm standard error of the mean (referred to as 1σ hereafter) of 2.7 ± 0.1 . The small 1σ values demonstrate that regardless of different combustion temperature the 85/87 ratios were reproducible as the combustion temperature was between 1023 K and 1173 K. Differences between the ratio for each experiment and the mean ratio were statistically insignificant except for Exp 5, and even the difference for Exp 5 is insignificant if 2σ , 95 percentile confidence level (95% CL), was considered. Therefore, we concluded that there was no combustion temperature dependence on the 85/87 ratios as the combustion temperature was between 1023 K and 1173 K. The mean value $\pm 1 \sigma$ of 2.7 ± 0.1 can represent the ratio of fly ash from the combustion of the coal A. The 133/87 ratios over the three different combustion temperatures varied more largely and randomly, ranging from 0.7 to 3 with the mean $\pm 1 \sigma$ of 1.3 ± 0.2 . Except for the ratio from the Exp 9, differences between each 133/87 ratio and the mean value were comparable with the propagated 1σ and smaller than the 95% CL, therefore, the ratios were reasonably averaged. Although the significant difference \pm the propagated 1σ for the Exp 9, -2.2 ± 0.7 , may be treated as an outlier, its influence to the mean value was small. We, therefore, included the result of Exp 9 for the calculation of the mean value and concluded that the overall mean $\pm 1 \sigma$ of 1.3 ± 0.2 also represents mean 133/87 ratio of fly ash from the combustion of the coal A. For other coals, the limited number of data sets obtained over different combustion temperatures was not enough to statistically evaluate

dependence of combustion temperature. However, the magnitude of the variations of the 85/87 and the 133/87 ratios was the similar to the experiments using the coal A. This implies that there was no such dependence for combustion of the other coals as well.

The analysis of different coals revealed that signals at m/z 85, 87, and 133 were notably higher than signals at their adjacent m/z in all the experiments of the other coal combustions. Their 85/87 and 133/87 ratios ranged from 1.9 to 4 and from 0.7 to 3, respectively (Table 3). The 85/87 ratios and the 133/87 ratios for the coal A to E, the north-eastern coals, were precise ($\sigma < 0.5$), while those for the coal G to I, the coals from other regions, varied more largely ($0.3 < \sigma < 2$). Note that the precision of the 85/87 and 133/87 ratios for the coal F is not known due to only the single measurement. A statistical test, one-sided analysis of variance (ANOVA), was performed to evaluate whether or not there were significant differences in ratio between the coals. The test resulted in that the F-value for the 85/87 ratios over all coals was 1.952. Meanwhile, the upper critical value of F (7, 28) at 0.05 of α was 2.388. The smaller F value than the upper critical value suggests that there was no significant differences between the 85/87 ratios. Similarly, the ANOVA was performed for the 133/87 ratios, and we found that there was no significant difference as well. Although the results of ANOVA exhibited that there was no significant difference between the 85/87 ratios as well as between the 133/87 ratios, we should note that one has to be careful with its interpretation since some coals have small data set. In overall, the 85/87 and 133/87 ratios $\pm 1 \sigma$ ($n = 27$) for the fly ash produced by combustions of variety of coals under our experimental conditions were 2.8 ± 0.1 and 1.5 ± 0.2 , respectively.

Comparison of m/z 85, 87, and 133 with Other Studies

85/87 ratios and 133/87 ratios from other source studies are listed in Table 4, together with the ratios observed in our

Table 2. Mean m/z 85 and 133 ratios to m/z 87 under different combustion temperature for combustion experiments of the coal A ($n = 5$ or more).

Experiment	Combustion Temp.	85/87 $\pm 1 \sigma^a$	Diff. ^d from the mean $\pm 1 \sigma^b$	133/87 $\pm 1 \sigma^a$	Diff. ^d from the mean $\pm 1 \sigma^b$
Exp 1	1123 K	2.7 \pm 0.6	0.0 \pm 0.6	0.7 \pm 0.2	0.6 \pm 0.3
Exp 2	1123 K	2.8 \pm 0.3	-0.1 \pm 0.3	1.3 \pm 0.2	0.0 \pm 0.3
Exp 3	1123 K	2.6 \pm 0.6	0.1 \pm 0.6	1.4 \pm 0.3	-0.1 \pm 0.4
Exp 4	1023 K	2.9 \pm 0.4	-0.2 \pm 0.4	0.9 \pm 0.2	0.4 \pm 0.3
Exp 5	1023 K	3.2 \pm 0.3	-0.5 \pm 0.3	1.1 \pm 0.1	0.2 \pm 0.3
Exp 6	1023 K	2.2 \pm 0.6	0.5 \pm 0.6	1.8 \pm 0.4	-0.5 \pm 0.5
Exp 7	1173 K	2.7 \pm 0.6	0.0 \pm 0.6	1.2 \pm 0.2	0.1 \pm 0.3
Exp 8	1173 K	2.8 \pm 0.9	-0.1 \pm 0.9	1.1 \pm 0.3	0.2 \pm 0.4
Exp 9	1173 K	2.7 \pm 0.7	0.0 \pm 0.7	3.5 \pm 0.7	-2.2 \pm 0.7
Exp 10	1173 K	2.1 \pm 0.9	0.6 \pm 0.9	1.4 \pm 0.6	-0.1 \pm 0.6
Exp 11	1173 K	2.6 \pm 0.4	0.1 \pm 0.4	0.8 \pm 0.1	0.5 \pm 0.3
Overall mean $\pm 1 \sigma^c$		2.7 \pm 0.1		1.3 \pm 0.2	

^a σ = standard errors of the mean from the replicate measurements ($n < 5$) during each experiment

^b σ = Propagated standard error of the difference

^c σ = standard deviation over square root of 11

^d Mean ratio – ratio of each Exp.

Table 3. Mean m/z 85 and m/z 133 ratios to m/z 87 for fly ash from combustion of different coals.

Coal ID	n	85/87 $\pm 1 \sigma^a$	133/87 $\pm 1 \sigma^a$
A	11	2.7 \pm 0.1	1.3 \pm 0.2
B	5	2.5 \pm 0.1	0.7 \pm 0.1
C	3	3.2 \pm 0.5	1.8 \pm 0.4
D	3	2.9 \pm 0.4	1.2 \pm 0.1
E	3	2.6 \pm 0.2	1.6 \pm 0.1
F	1	1.9	2.0
G	4	2.6 \pm 0.3	3 \pm 1
H	4	3 \pm 1	2 \pm 2
I	2	4 \pm 1	2 \pm 1
Overall mean $\pm 1 \sigma$		2.8 \pm 0.1	1.5 \pm 0.2

^a σ = standard deviation over square root of n.

Table 4. m/z 85 and 133 ratio to m/z 87 for fly ash from combustion of other materials.^a

Material	85/87	133/87
Biomass burning		
Oak	1.68	0.89
Musasa	0.44	0.10
Oak flame	1.65	0.93
Lignin powder	2.86	1.68
Savanna grass	3.30	1.37
Rice straw	1.86	1.12
Southern pine	1.96	0.92
Vehicular emission		
Diesel bus	23.71	4.35
Diesel track	17.31	2.97
Honda car	35.99	2.41
Other man-made emission		
Plastic	20.37	0.30
Chicken	39.87	1.55
Fatty burger	6.39	0.51
Lean burger	5.93	0.53
Paper	4.73	0.55
Salmon	4.35	0.86
Field Observation		
Fukue (episode Apr 9, 2009)	2.44	0.58
Hedo (episode Oct 18, 2009)	2.38	0.79

^aRatios for other emission sources were obtained from the AMS spectral database (<http://cires.colorado.edu/jimenez-group/AMSSd/>) by Ulbrich, I.M., Lechner, M. and Jimenez, J.L.

case studies at the Fukue and the Cape Hedo monitoring stations. As seen, the 85/87 ratios from the field studies conducted at Fukue and Cape Hedo are the similar to the ratio from the coal combustion studied here, while the 133/87 ratios observed in the field studies were almost a half. Unfortunately, our current knowledge is not enough to define whether the disagreement in the 133/87 ratios is due to uncertainties included in both the 133/87 ratios or really an indication of different origin. Alkali metal analysis including their isotopes may allow us to answer this open question. Compared to these ratios from the other source studies, the most of the 85/87 ratios except for some biomass

burning clearly differ from the ratio from this source study. Particularly many ratios for the vehicular emissions, which are expected to be another significant source of transboundary air pollution to Japan, are about 10 fold of magnitude higher. In contrast, the 85/87 and 133/87 ratios determined for the fly-ash from the coal combustion here are almost the same as those for burning of lignin powder and savanna grass. Thus, biomass burning Takegawa *et al.* (2009) suggested still remained as a possible source giving unique organic aerosol spectra with remarkable signals at m/z 85, 87, and 133. If these signals were of the alkali metals, such characteristic spectra would be remained in the PM, regardless of oxidation state.

CONCLUSION AND FUTURE WORK

We carried out laboratory experiments of coal combustion using a fluidized bed reactor to characterize organic mass spectra for PM from coal combustion. The study here focused detection of notably high loadings of m/z 85, 87, and 133. The results made us confirm the presence of remarkable signals (i.e., significantly higher signals than those at the adjacent m/z) at m/z 85, 87, and 133. The reproducible 85/87 and 133/87 ratios and its considerable difference from the ratio from other source studies indicate potential of m/z 85, 87, and 133 signals for identification of coal combustion PM. Further studies combining AMS measurements with bulk sample analysis for alkali metals will give characterization of the signals in more detail.

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