

Review 2:

This paper reports the development of an online monitoring system for PM_{2.5} composition. The unique feature for this system is the capability to measure more than 90% of PM_{2.5} mass with a high time resolution. This advance makes this system a powerful tool for understanding PM_{2.5} sources and deciding the corresponding control measures. Another novelty of this paper is the resolving of secondary organic aerosol (SOA) from the total PM_{2.5} mass by the usage of m/z 44, a good SOA tracer, in the PMF modelling. This system has been successfully applied in a megacity in China, with nine sources well identified, supporting its effectiveness and usefulness in PM_{2.5} control. Overall, I think it is a well-written paper with novelty and recommend its publication after considering the following concerns.

Response: Thank you.

Comments:

1. The author should make a clearer statement of the advantage of the new system compared to the separate instruments in the introduction part.

Response: Thanks for your comment. The statement of the new system has been added to the main text:

“There are two differences between the new online integrated system and the separate online instruments. On one hand, the new online integrated system used isokinetic sampling manifold and the same sampling head to ensure the reliability and comparability of synchronous sampling among different instruments. On the other hand, we integrated ACSM into the new system to measure OM to achieve PM_{2.5} mass closure better” (Line 62-66)

2. Line 31. “serious” might not be suitable to the current PM_{2.5} pollution status. A description of long-term problem should be better.

Response: Thank you for this comment and suggestion. we changed the sentence to:
“PM_{2.5} is a long-term problem in some cities or regions.” (Line 31)

3. Why Na was not measured? Could sea salt be a major source for PM_{2.5} in Shenzhen, a coastal city?

Response: We did not measure Na mainly for two reasons.

First, it is difficult to accurately measure the online integration of Na, Na has lighter atomic number, so the uncertainty of data measured by X-ray fluorescence method is larger. The ACSM evaporator was ~550°C, and some components such as NaNO₃, NaCl, and Mg(NO₃)₂ in the sea salt may not be detected due to the lack of gasification at this temperature. Although the instruments of ion chromatography, such as MARGA can realize Na⁺ online measurement, it is not suitable for integration due to its large volume and cumbersome operation.

Second, as the reviewer noted, sea salt represented by Na is not the main source of PM_{2.5} in Shenzhen, previous studies have shown that sea salt contributes less (1% to 3%) to PM_{2.5} (Huang et al., 2014; Huang et al., 2018).

We also pointed out it in the manuscript:

“The factor of sea salt cannot be identified in this study, because the measurement of its tracer (Na) is limited for X-ray fluorescence method and ACSM. However, the contribution of sea salt is little for Shenzhen (about 1% to 3%), and is not the main source of pollution.” (Line 310-312)

4. 2.4 Design of the data analytics platform. More details for data conversion of each instrument should be given.

Response: Thanks for your comments as it identifies a potential area for confusion of other readers. The details of data analytics platform have been clarified in the revised supplement.

“Design of the data analytics platform

The design solution of the data analytics platform was shown in Fig. S2, the data of three online instruments (ACSM, Xact-625, AE-31) were processed in same (.csv) format and saved in their respective local computer using data transmission software, and uploaded data to a same SQL Server remote database for data management. The database was based on the dedicated server of the integrated system. The data of SHARP-5030i can be directly connected to the server. In addition, an atmospheric environment data collection and processing system had been established. The data in the SQL Server database was called to achieve unified processing and display of integrated data.

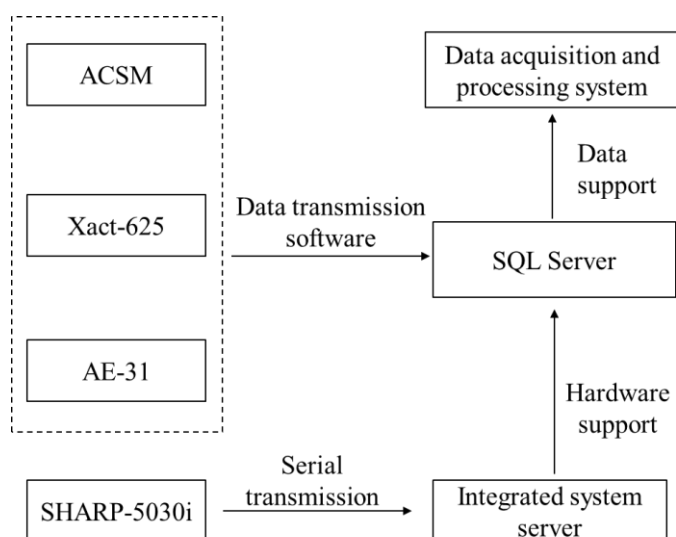


Figure S2. Design solution of data analytics platform”

5. Figure 2a. The mass closure is generally good. However, the authors should comment on some periods when a significant mass discrepancy appeared.

Response: As pointed out by the reviewer, the overall quality of the instrument is very good, with an average deviation of 6%, but the differences between reconstructed and measured are relatively large sometimes.

We further analyzed the 10/3 and 10/20, and think it may be related to the temperature. The temperature on October 3 is the highest during the observation period, and at

October 20, the temperature is at a relatively low level. The temperature has different effects on SHARP-5030i and integrated instruments, which results in different fitting effects to a certain extent. Of course, there may be other reasons, such as the measurement error of the integrated instruments, particle composition, temperature and relative humidity (Su et al., 2018; Zhang et al., 2017).

We also pointed out the reasons caused the underestimation in the manuscript:

“The average error margin of mass closure during the observation period is about 6%, which might be due to the measurement error of the integrated instruments, particle composition, temperature and relative humidity (Su et al., 2018; Zhang et al., 2017). A significant mass discrepancy between reconstructed and measured PM_{2.5} appeared in some periods (Fig. 2a). For example, the underestimation on October 3 and the overestimation on October 20 occurred when the temperature was the highest and the lowest during the observation period, respectively. Therefore, it was speculated that temperature might affect the composition of PM_{2.5}, causing the mass closure to deviate.” (Line 227-232)

6. Figure 4. The elements with low concentrations should be displayed with enlargement.

Response: We thank the reviewer for this suggestion. The figure is corrected as:

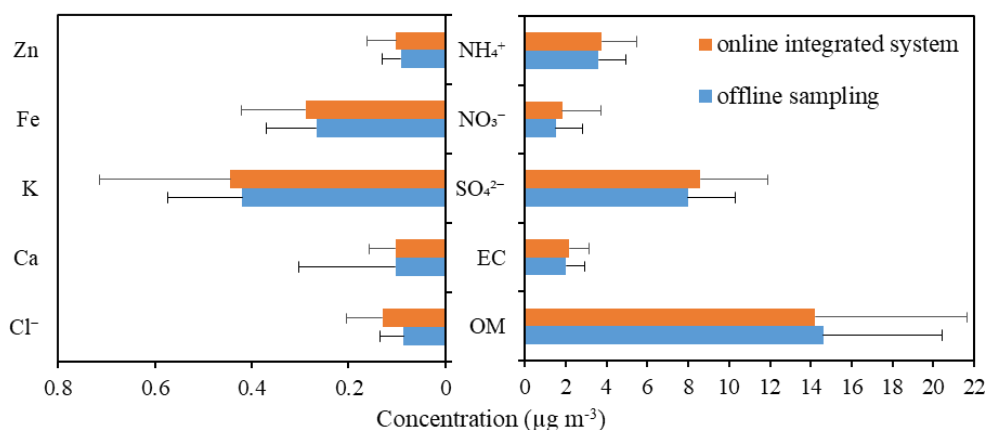


Figure 4. Concentrations of major chemical compositions of PM_{2.5} measured by online Integrated system and offline sampling. Note that the EC in the online integrated system

was referred to as BC. (Line 273)

7. Figure 7. There is a spike at 1:00 am for both biomass burning and fugitive dust.

Why?

Response: The spike of fugitive dust at 1:00 might be due to the influence of local sources. We carefully analyzed the time series of fugitive dust and biomass combustion sources, and found that the fugitive dust appeared a very high value at 1:00 on October 19, 25 and 28, which was significantly higher than the average dust concentration during the observation period, and made the average value of fugitive dust higher at 1:00. The tracer Ca also showed a higher value at the corresponding time, indicating that there were short-term dust emission activities. The biomass burning also appeared abnormally high value at 1:00 on October 2, relating to the firework shows on the China's National Day.

We added more specifically state the sources in manuscript:

“Analyzed with the time series of pollution sources (Fig. 5b), the results showed, the peaks around 22:00 and 1:00 of biomass burning was mainly due to the abnormally high source contribution, on the night of October 1, which is China's National Day, and there was a firework show 10 km away from the sampling site. Previous study has shown that the concentration of species (e.g. K, Ca, Cl⁻, NO₃⁻) in the PM_{2.5} would have greatly increase due to the fireworks (Tsai et al., 2012). The spike at 1:00 of fugitive dust and biomass burning mainly caused by several abnormally high values at 1:00, suggesting it might be due to the influence of local short-term activities.” (Line 355-361)

We added the standard deviation to the Fig. 7, so that readers can get more information from it.

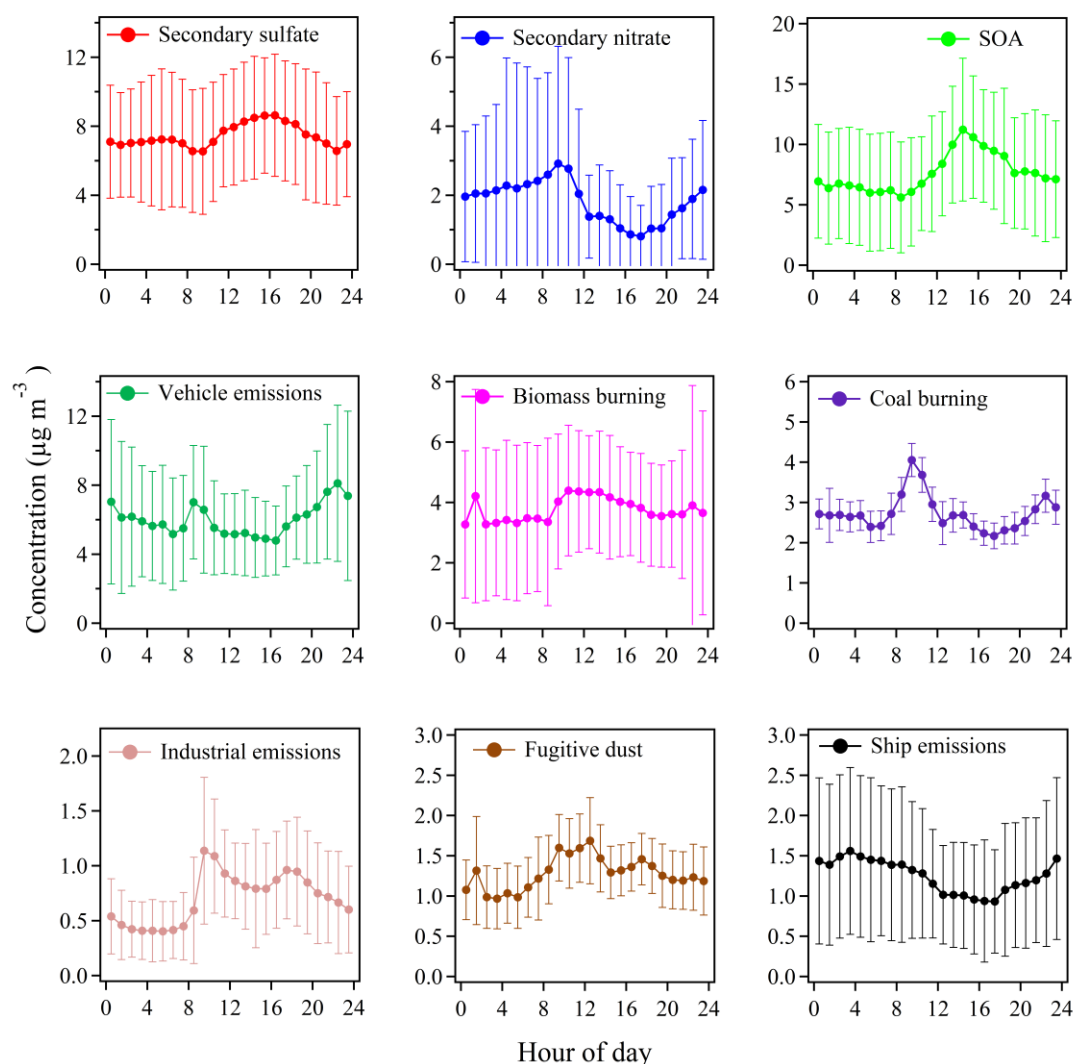


Figure 7. Diurnal variations of the nine sources resolved by ME-2. (Line 368)

8. Conclusions. For a technical paper, the prospects of further development of the new system should be given.

Response: We have mentioned the prospects, and added more explanation about this in the manuscript:

“The development and successful application of the online integrated system and source apportionment method suggested that they can be used for precise regulation of PM_{2.5}. (such as fireworks)” (Line 416-417)

“That is, the high time resolution source analysis of the new integrated system is helpful to study the variation of the primary and secondary sources of PM_{2.5} in the process of

heavy pollution, and to identify the key sources of heavy pollution and its mechanism, then to assess the impact of different sources. The system can run stably for a long time, and provides a key scientific basis for particulate matter control of China.” (Lines 417-422)

Reference

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