

## Supporting Information

### Contributions of condensable particulate matter to atmospheric organic aerosol over Japan

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Number of pages: 6

Number of figures 4

## **S1. Measurement of filterable and condensable PM from stationary combustion sources**

Figure S1 is a schematic diagram of the sampling methodology for filterable and condensable PM used by the Tokyo metropolis<sup>S1</sup>. Filterable particulate matter (PM) from stacks is collected with an Andersen stack sampler (AS-500; Tokyo Dylec, Tokyo, Japan) in accord with JIS K 0302. We replaced some separation plates with a spacer to collect PM<sub>2.5</sub> samples on backup filters (Teflon-membrane and quartz fiber filters). For the sampling of condensable-plus-filterable PM, exhaust gas was passed through a cyclone at a flow rate of 6 L/min to remove particles larger than 2.5 µm. The air stream was then passed through a diluter (FPS-4000, Dekati Ltd. Kangasala, Finland), where the sample air was diluted with dry, clean air by a factor of 20 and cooled to ambient temperature. The diluted air was then introduced into a glass chamber (16.7 L) with a residence time of >10 s. The dilution ratio and residence time were the same for ISO 25597. A pump with a flow rate of ~80 L/min was used to aspirate the sample air from the glass chamber, and PM was sampled on Teflon-membrane filters and quartz fiber filters.

The sampling methodology of the Ministry of Environment<sup>S2</sup> is similar to that of the Tokyo Metropolis<sup>S1</sup>, but they differ in several respects. The former methodology involves sampling of filterable PM with a cascade impactor (VI-PM2.5; Tokyo Dylec, Tokyo, Japan), in accord with JIS Z 7152, and sampling of condensable-plus-filterable PM by introducing diluted air into a glass chamber (6.7 L) with a residence time of >10 s (flow rate was 40 L/min).

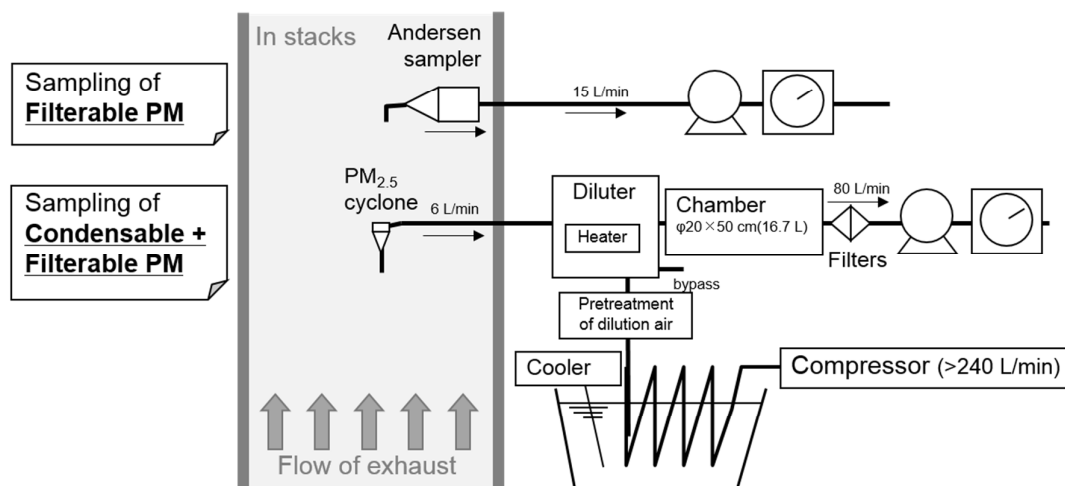
The sampled Teflon-membrane filters were dried for 1 hour at 110 °C and then cooled to ambient temperature in a desiccator. The PM<sub>2.5</sub> mass concentrations were determined from the weights of the sampled Teflon-membrane filters. Weighing was performed with an analytical balance (readability 1 µg) in a chamber where the temperature and relative humidity were controlled at 21.5±1.5 °C and 35±5%, respectively. The Teflon-membrane filters were conditioned in the chamber for >24 h before weighing. Concentrations of organic carbon and elemental carbon in the sampled quartz fiber filters were analyzed with a thermal/optical/reflectance carbon analyzer (DRI Model 2001, Desert Research Institute, Reno, NV) based on the protocol of Chow et al.<sup>S3</sup> The sampled quartz fiber filters were dried for 1 hour at 110 °C before the analysis.

## **References**

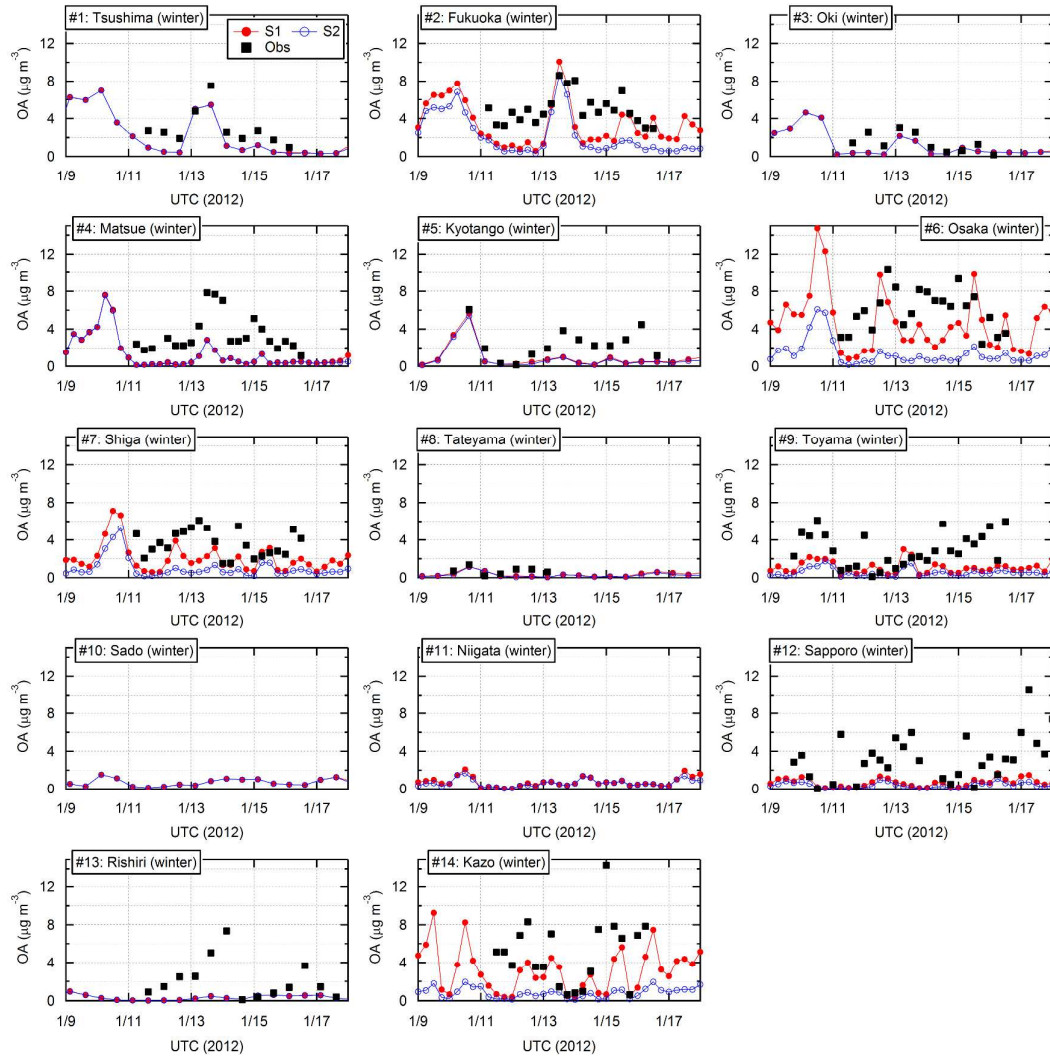
S1 Tokyo Metropolis, *Report of emission surveys of fine particulate matters (PM2.5)*; 2011;

[http://www.kankyo.metro.tokyo.jp/basic/conference/air/particulate\\_matter/study\\_committee\\_07.files/02\\_hassei.pdf](http://www.kankyo.metro.tokyo.jp/basic/conference/air/particulate_matter/study_committee_07.files/02_hassei.pdf).

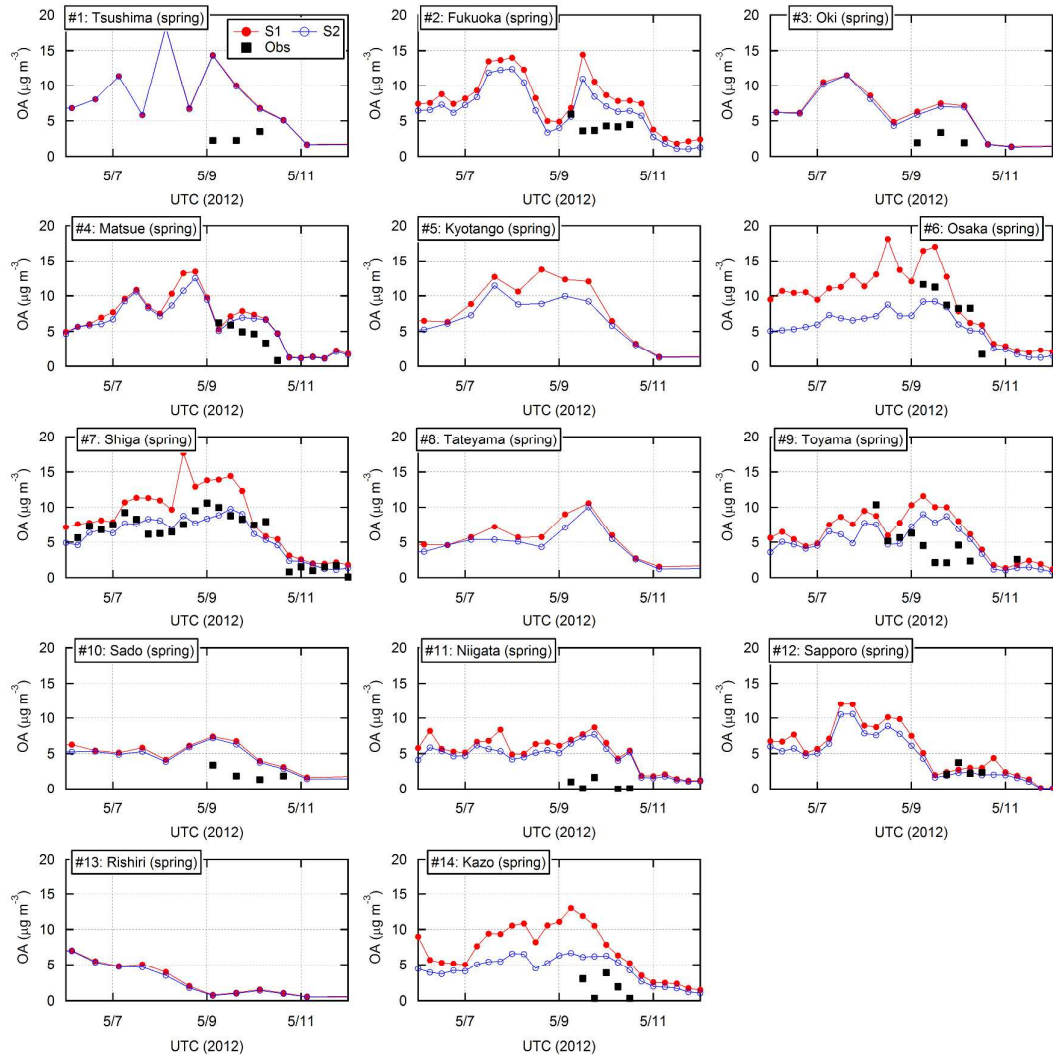
- S2 Ministry of the Environment, Japan, *Survey of stationary emission sources for PM<sub>2.5</sub> emission inventory and source profiles*; Tokyo, 2015.
- S3 Chow, J. C.; Watson, J. G.; Pritchett, L. C.; Pierson, W. R.; Frazier, C. A.; Purcell, R. G., The Dri Thermal Optical Reflectance Carbon Analysis System - Description, Evaluation and Applications in United-States Air-Quality Studies. *Atmospheric Environment Part a-General Topics* **1993**, 27, (8), 1185-1201.



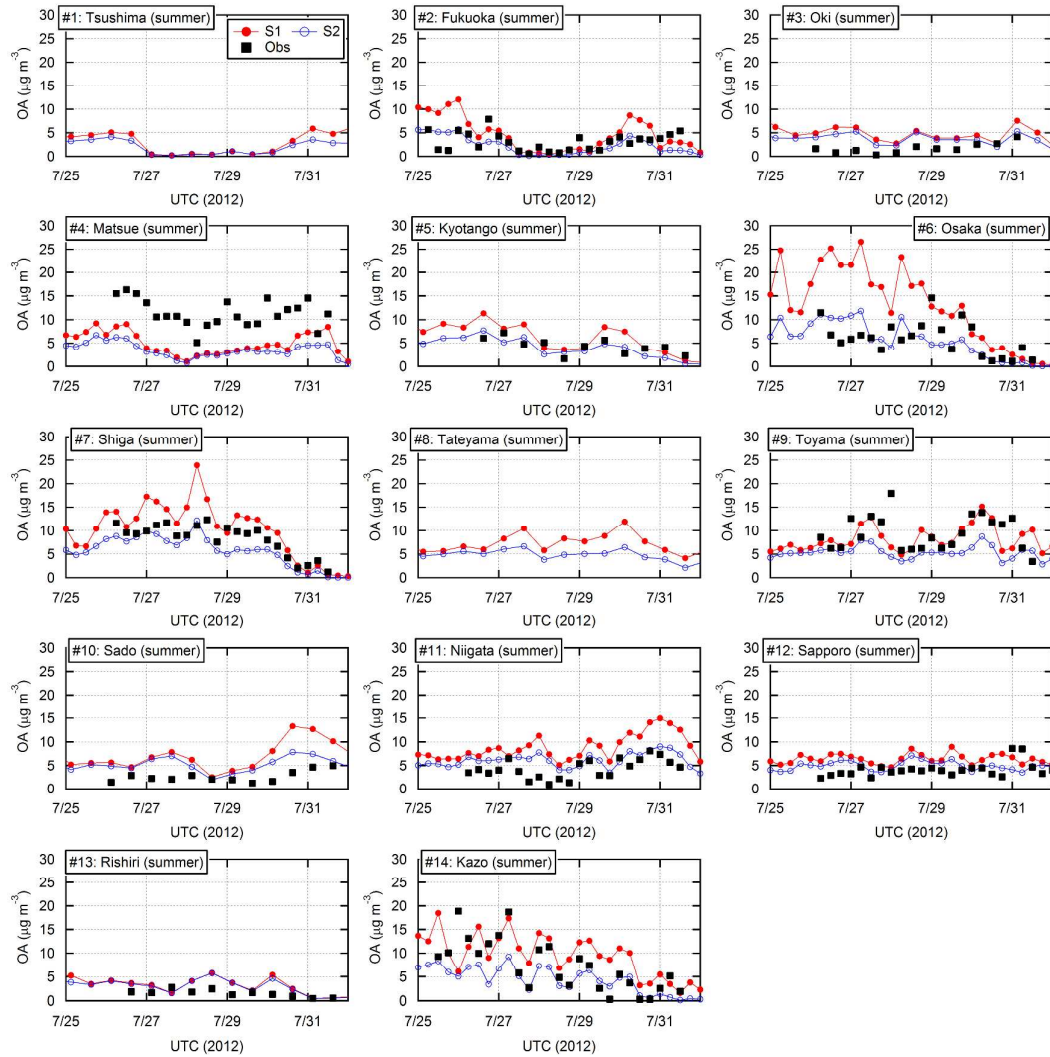
**Figure S1.** Schematic diagram of sampling methodology for filterable and condensable-plus-filterable PM<sup>S1</sup>.



**Figure S2.** Time series of observed and simulated OA concentrations at 14 observational stations in winter. The stations are classified as remote (#1, #3, #5, #8, #10, and #13), rural (#4, #7, #9, and #11), and urban (#2, #6, #12, and #14) sites.



**Figure S3.** Same as Figure S2, but for spring.



**Figure S4.** Same as Figure S2, but for summer.