## Development of an automated total carbon analyzer for atmospheric aerosols

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## Introduction

Increasing attention is being paid to carbonaceous aerosols, a major component in  $PM_{2.5}$ , owing to the importance of its complex effects over various geographical scales, such as its effects on human health, haze, the hydrological cycle, and climate change. However, the sources, atmospheric processes, and impacts of carbonaceous aerosols are not well understood; moreover, uncertainties in measurement methods tend to be high owing to the complexity of the physical/chemical properties of carbonaceous aerosols as compared with the properties of other components (e.g., inorganic ions and minerals) of  $PM_{2.5}$ .

Thermal–optical analysis (TOA) is a popular method for determining aerosol EC and OC collected on quartz filters. However, the temperature protocol (e.g., IMPROVE, NIOSH) and optical detection (transmittance, reflectance) adopted in TOA strongly affect the separation of OC and EC.

EC and OC concentrations at Fukue Island in Japan were measured from November to December in 2012 in a short-term campaign using a semi-continuous TOA analyzer (Sunset Laboratory). However, the liquid-based calibrations of the instrument using two thermal protocols, namely IMPROVE-like and Total-Carbon (only in  $O_2$ /He mixture), showed a difference of ca. 30%.

The purpose of this study is to develop an automated semi-continuous total carbon (TC, TC = EC + OC) analyzer employing a thermal technique and to minimize the uncertainties in the measurement of carbonaceous aerosol.

## **Experiments, Results, and Discussions**

Based on thermal desorption analysis, a semicontinuous automated analyzer for TC (TC\_JA) was developed in this study. One-step heating in 2% O<sub>2</sub>/He mixture up to 870 °C was employed to evolve carbonaceous compounds sampled on a quartz filter. TC concentrations were measured as CO<sub>2</sub> through a heated column with an oxidizing catalyst (MnO<sub>2</sub>) using an NDIR CO<sub>2</sub> analyzer (LI-840A).

TC concentrations in Yokosuka, Japan, were measured using the semi-continuous carbon analyzer (Sunset Laboratory) and the TC analyzer developed in this study, and comparisons were conducted during the period from Sep 2012 to Feb 2013. A  $PM_{2.5}$  sharp-cut cyclone (BGI) and a denuder (Sunset) were employed to remove any volatile organic compounds from the sample air.

Under almost the same thermal Total-Carbon protocols, the TC concentrations measured by the Sunset and newly developed instruments (TC\_Sunset and TC\_JA) were in good agreement, as shown in Figure 1. In the range below 4  $\mu$ g/m<sup>3</sup>, however, the TC\_Sunset/TC\_JA ratios were found to be concentration dependent, with higher ratios occurring in the lower concentration range. This result might indicate the better performance of the detection system as a result of the detector, theromogram data system (CDS plus, LAsoft), and/or gas dryer (Perma Pure) using Nafion membrane tubing, which continuously removes only water vapor from the gas streams.

In addition, TC\_IMPROVE (OC + EC) measured using the Sunset analyzer was compared with the TC\_JA. The concentrations with TC\_IMPROVE tended to be higher than those of TC\_JA (ca. 30% with a strong correlation). Identification of the exact origins of these differences is underway. These results will help to understand the differences in the liquid-based calibrations at Fukue.

We recommend that measurements by TOA should be undertaken using different protocols, such as IMPROVE-like and Total-Carbon protocols, for the accurate determination of EC and OC.

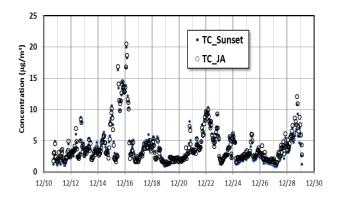


Figure 1 TC concentrations in Yokosuka, Japan, measured using the Sunset carbon analyzer and the TC analyzer developed in this study, Dec 2012

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