



Measurements of  
non-volatile aerosols  
with a VTDMA

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# Measurements of non-volatile aerosols with a VTDMA and their correlations with carbonaceous aerosols in Guangzhou, China

H. H. Y. Cheung<sup>1</sup>, H. B. Tan<sup>2</sup>, H. B. Xu<sup>3</sup>, F. Li<sup>2</sup>, C. Wu<sup>1</sup>, J. Z. Yu<sup>1,4</sup>, and C. K. Chan<sup>1,5</sup>

<sup>1</sup>Division of Environment, Hong Kong University of Science and Technology, Hong Kong, China

<sup>2</sup>Institute of Tropical and Marine Meteorology, China Meteorological Administration, Guangzhou, China

<sup>3</sup>Sun Yat-sen University, Guangzhou, China

<sup>4</sup>Department of Chemistry, Hong Kong University of Science and Technology, Hong Kong, China

<sup>5</sup>Department of Chemical and Biomolecular Engineering, Hong Kong University of Science and Technology, Hong Kong, China

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Correspondence to: H. B. Tan (hbtan@grmc.gov.cn)  
and C. K. Chan (keckchan@ust.hk)

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## Abstract

Simultaneous measurements of aerosols of varying volatilities and carbonaceous matters at an urban site of Guangzhou, China were conducted in February and March 2014 using a Volatility Tandem Differential Mobility Analyzer (VTDMA) and an Organic Carbon/Elemental Carbon (OC/EC) Analyzer. In VTDMA, selected aerosols of 40 to 300 nm in mobility diameter were heated at elevated temperatures up to 300 °C and the size distributions of the residual particles were measured. Size dependent distributions were observed for both non-volatile and volatile materials (VM). The 40 nm particles were dominated by particles that completely vaporized (CV) at 300 °C, with an average number fraction of about 0.4. Particles larger than 80 nm were dominated by the medium and low volatility (MV and LV, respectively) fractions, with average number fractions of 0.5 and 0.15 respectively. VM did not contribute separately to number fraction but contributed to over 50 % of the total volume fraction for all sizes. In both number and volume fractions, diurnal variation was only observed for the high volatility (HV) group, CV and VM in 40 nm particles, likely because these particles were related to fresh emissions. The little diurnal variation of larger particles could be attributed to non-locally aged aerosols. Closure analysis between the residual mass of LV + MV and mass of EC or EC + OC<sub>2-4</sub> (sum of EC, OC<sub>2</sub>, OC<sub>3</sub>, and OC<sub>4</sub> of the OC/EC Analyzer) suggests that non-volatile materials measured by the VTDMA likely contain less volatile OC.

## 1 Introduction

Carbonaceous aerosols comprising organic carbon (OC) and elemental carbon (EC) or black carbon (BC) are one of the major light absorption constituents and are abundant in particulate matter (PM) (Rosen et al., 1978; Hansen et al., 1984; Japar et al., 1986; Chow et al., 1993; Horvath, 1993; Liousse et al., 1993; Fuller et al., 1999; Putaud et al., 2010). In China, the worsening of visibility degradation associated with PM is

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of increasing concern in recent years. In particular, numerous studies on air pollution were carried out in different cities in China including the Pearl River Delta (PRD) region which is a fast-developing economic zone (Cheng et al., 2006; Wu et al., 2007; Andreae et al., 2008; Chan and Yao, 2008; Gnauk et al., 2008; Tan et al., 2013a). In 2007, the mass concentrations of EC and OC at an urban Guangzhou (GZ) site were reported to be 6.8 to 9.4 and 6.6 to 22.5  $\mu\text{g m}^{-3}$  respectively (Yu et al., 2010).

Soot particles are often characterized in terms of EC and BC, depending on whether they are measured thermally or optically (Penner and Novakov, 1996; Lavanchy et al., 1999; Cheng et al., 2011 and references therein). Their optical properties are distinct when they are freshly produced (Novakov et al., 2003). After aging processes such as cloud processing, reaction with other species and coagulation, their structure, shape, size, mixing state and thus optical properties change (Horvath, 1993; Liousse et al., 1993; Ghazi and Olfert, 2012). EC is typically measured by thermal method such as the OC/EC Analyzer (Chow et al., 2007), BC are optically measured using instruments such as aethalometer (Hansen et al., 1984), Multi-Angle Absorption Photometer (MAAP) (Petzold and Schönlinner, 2004) and Particle Soot Absorption Photometer (PSAP) (Virkkula et al., 2005). However, it is not possible to retrieve the mixing state of soot particles from above techniques. To determine the mixing state of soot, Single Particle Soot Photometer (SP2) (Stephens et al., 2003), Soot Particle Aerosol Mass Spectrometer (SP-AMS) (Onasch et al., 2012) and Volatility Tandem Differential Mobility Analyzer (VTDMA) (Philippin et al., 2004) have been used.

Ambient aerosols have varying volatility properties based on their chemical compositions. VTDMA was first introduced by Rader and McMurry (1986) to study the behavior of aerosols upon thermal treatment. Philippin et al. (2004) later developed a VTDMA which is capable of evaporating volatile materials (VM) in aerosols at temperatures up to 300 °C. Non-volatile materials, such as EC and sea salt, can internally mix with (or be coated with) VM, which evaporate during thermal treatment at elevated temperatures. Based on the relative abundance of the non-volatile materials to VM, these internally mixed ambient aerosols can be categorized into three groups, namely the



volatile materials derived by the VTDMA and EC and OC measured by the OC/EC Analyzer to evaluate the significance of non-volatile OC in the VTDMA measurements.

## 2 Methodology

### 2.1 Experimental

#### 2.1.1 Measurement details

The campaign was taken place at the China Meteorological Administration (CMA) Atmospheric Watch Network (CAWNET) Station in Panyu, Guangzhou, China in summer from July to September 2013 and winter from 6 February to 21 March 2014, which is operated by the Institute of Tropical and Marine Meteorology (ITMM) of the CMA. The Panyu station is located at the center of the PRD region and on the top of Dazhengang Mountain (23°00' N, 113°21' E) with an altitude of about 150 m (Fig. S1 in the Supplement) (Tan et al., 2013a). It is about 120 m above the city average elevation and is surrounded by residential neighborhoods with no significant industrial pollution sources nearby. Measurements of particle number size distributions, volatility, mass concentrations of EC and OC were made in winter from 6 February to 21 March 2014. Some of the measurements were not made continuously due to maintenance work and hence only periods with both VTDMA and OC/EC measurements were analyzed.

#### 2.1.2 VTDMA measurements

We used a custom-made VTDMA based on a Hygroscopic TDMA system developed in ITMM (Tan et al., 2013b), with the humidifier between the two DMAs replaced by a heated tube which effect evaporation of volatile materials. In our VTDMA system shown in Fig. 1, ambient aerosols sampled by a PM<sub>2.5</sub> inlet first passed through a dryer at relative humidity below 20 %. The dry aerosols then passed through a neutralizer and entered the first differential mobility analyzer (DMA<sub>1</sub>) (Stream 1) to produce

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mono-disperse aerosols of diameter between 40 and 300 nm,  $D_0$ . The mono-disperse aerosols went either path (a) or (b) in Fig. 1 after leaving DMA<sub>1</sub>. In path (a), they were directed to a condensation particle counter (CPC, TSI Model 3772) to obtain particle counts,  $N_{D_0}$ . The particle number size distribution of the ambient aerosols,  $dN/d\log D_p$ , was also measured by varying the DMA<sub>1</sub> voltage (SMPS scan). Afterwards, the mono-disperse aerosols were directed via path (b) to a heated tube for volatility measurement (V-Mode) sequentially at 25, 100 and 300 °C.

Upon heating at 100 °C and beyond, volatile components of the particles such as sulfate, nitrate and volatile organics vaporize, resulting in the shrinking of particles and a new size distribution,  $dN'/d\log D_p$  of the remaining particles, i.e. the non-volatile materials in the LV/MV/HV aerosols (hereafter called residuals). These size distributions of six selected diameters from DMA<sub>1</sub> (40, 80, 100, 150, 200 and 300 nm) were measured by DMA<sub>2</sub> and CPC before they were heated at another temperature. Overall it took around one and a half to two hours to complete a cycle of measurements which consisted of SMPS scans and V-Mode measurements at 25, 100 and 300 °C. Hereafter, notations with the superscript prime refer to the LV/MV/HV residuals measured by DMA<sub>2</sub> and CPC after heating, while the corresponding ones without the prime refer to the LV/MV/HV residuals in ambient aerosols prior to heating.

Figure 2 illustrates how thermal treatment in the VTDMA affects the size distributions of the ambient aerosols. At each selected diameter  $D_0$  (and at each temperature) in DMA<sub>1</sub> in the VTDMA, the particles include CV particles (purple) and LV, MV and HV particles that have VM (orange) internally mixed with the LV (blue), MV (green) and HV (red) residuals. After heating, the remaining particles form LV, MV and HV residuals without any CV or VM. Evaporation of VM and CV does not change the number concentrations of LV, MV and HV particles.

A volatility shrinkage factor, VSF, is defined as the ratio of particle diameter after heating,  $D_p$ , to that before heating,  $D_0$ , to indicate the size reduction of the ambient particles (Eq. 1). The value of VSF is always smaller than or equal to one, depending

on the amount of volatile materials vaporized.

$$\text{VSF} = \frac{D_p}{D_0} \quad (1)$$

The VSF range for LV, MV and HV are larger than 0.9, between 0.4 and 0.9 and less than 0.4, respectively (Fig. 2) (Wehner et al., 2004, 2009).

Residual particles are either freshly emitted soot particles or other non-volatile materials such as sea salt or crustal particles, which do not shrink significantly during heating (LV particles), or particles that exhibit a larger size change due to evaporation of volatile materials (MV and HV particles). Some volatile materials, such as sulfate and nitrate, are formed via chemical and/or photochemical reactions. They mix internally with the non-volatile materials during atmospheric aging process and evaporate at 300 °C in the VTDMA. Most of the organic species, including primary and secondary, also evaporated at this temperature. In urban areas, the majority of the non-volatile mass in submicron particles can be assumed to be soot (Kondo et al., 2006; Kuwata et al., 2007).

### 2.1.3 OC/EC measurements

A semi-continuous Sunset OC/EC Analyzer (Model 4) was used to measure PM<sub>2.5</sub> mass concentrations of organic carbon and elemental carbon,  $m_{\text{OC}}$  and  $m_{\text{EC}}$  respectively, on an hourly basis (Turpin et al., 1990; Birch and Cary, 1996; Wu et al., 2012). The OC/EC Analyzer adopts the ACE-Asia protocol (a NIOSH-derived protocol), where OC evaporates at four set temperatures of 310, 475, 615 and 870 °C, and EC is combusted at temperature above 550 °C (Schauer et al., 2003). Based on volatility and refractoriness, the OC contents are named OC<sub>1</sub> to OC<sub>4</sub> with OC<sub>1</sub> being most volatile. In the VTDMA measurements, there were volatile or semi-volatile OC which vaporize at 300 °C or below. These vaporized OC are assumed to be OC<sub>1</sub>, which vaporizes at 310 °C, although this OC/EC set temperature is slightly higher than the set temperature of 300 °C in the VTDMA. With this assumption, the residuals of the VTDMA at 300 °C

(LV and MV residuals) are postulated to consist of (1) OC<sub>2</sub> to OC<sub>4</sub>, which vaporize at 475 °C and above, and (2) EC and other refractory PM components. We have ignored the HV residuals as their contributions are insignificant when compared to LV and MV residuals (see later). We will conduct a mass closure analysis based on the VTDMA and OC/EC measurements to examine this assumption.

## 2.2 Data analysis

### 2.2.1 Number fractions

The number fractions of LV, MV and HV residuals ( $\Phi'_{N,LV}$ ,  $\Phi'_{N,MV}$  and  $\Phi'_{N,HV}$ , with their sum equals unity) in Stream 2 on Fig. 1 were obtained from  $dN'/d\log D_p$  measured by the CPC. However, these fractions are not the actual number fractions of LV, MV and HV residuals ( $\Phi_{N,LV}$ ,  $\Phi_{N,MV}$  and  $\Phi_{N,HV}$ ) of the selected particles before heating because they have not taken into account the CV particles and particle diffusional and thermophoretic losses. The number fraction of CV ( $\Phi_{N,CV}$ ) is first obtained by considering the number fractions due to the residuals ( $1 - \Phi_{N,CV}$ ) and the number concentrations at a selected diameter  $D_0$  before heating ( $N_{D_0}$ ) and after heating ( $N'$ ):

$$N_{D_0} \times \eta_{D_0} \times (1 - \Phi_{N,CV}) = N' \quad (2)$$

where  $\eta_{D_0}$  is the transport efficiency of particles.

Equation (2) assumes that  $\eta$  is the same for LV, MV and HV particles.  $\eta$  accounts for particle loss between DMA<sub>1</sub> and DMA<sub>2</sub> due to diffusion and thermophoretic forces (Philippin et al., 2004), and it varies with particle size and heating temperature.  $\eta$  at each particle diameter and VTDMA temperature was determined by laboratory calibrations with sodium chloride (NaCl) particles, which do not evaporate (i.e.,  $\Phi_{N,CV} = 0$ ) at the heating temperatures used in our experiments. From the known  $\eta$  and field measurements  $N_{D_0}$  and  $N'$ ,  $\Phi_{N,CV}$  was obtained from Eq. (2). Afterwards,  $\Phi_{N,LV}$ ,  $\Phi_{N,MV}$  and  $\Phi_{N,HV}$  were obtained by renormalizing  $\Phi'_{N,LV}$ ,  $\Phi'_{N,MV}$  and  $\Phi'_{N,HV}$  with  $(1 - \Phi_{N,CV})$  so that the sum of  $\Phi_{N,LV}$ ,  $\Phi_{N,MV}$ ,  $\Phi_{N,HV}$  and  $\Phi_{N,CV}$  equals to unity.

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## 2.2.2 Volume fractions

The volume fractions of LV, MV, HV residuals and CV ( $\Phi_{V,LV}$ ,  $\Phi_{V,MV}$ ,  $\Phi_{V,HV}$  and  $\Phi_{V,CV}$ ) at each selected diameter  $D_0$  are defined as the ratios of the volume of LV, MV, HV residuals and CV to the total volume of the mono-disperse particles before heating. By assuming that the residuals are in spherical shape,  $\Phi_{V,LV}$ ,  $\Phi_{V,MV}$  and  $\Phi_{V,HV}$  can be calculated by:

$$\Phi_{V,i} = \frac{N_i \times \frac{\pi}{6} D_{p,i}^3}{N_{D_0} \times \frac{\pi}{6} D_0^3} = \Phi_{N,i} \times \frac{D_{p,i}^3}{D_0^3} \quad (3)$$

where  $N_i$  and  $D_{p,i}$  are the number concentration and mean residual diameter of  $i = LV, MV$  or  $HV$  residuals.

For LV particles, it is assumed that  $D_0$  and mean  $D_p$  are the same and hence  $\Phi_{V,LV}$  is the same as  $\Phi_{N,LV}$ . For MV and HV particles, the mean  $D_p$  is smaller than  $D_0$  due to the evaporation of volatile materials. The number weighted mean residual diameter ( $D_p$ ) is calculated by:

$$D_{p,i} = \frac{\sum_j D_{p,i} \times N_{i,j}}{N_i} \quad (4)$$

where  $D_{p,i}$  and  $N_{i,j}$  are the residual diameter and number concentration of  $i = MV$  or  $HV$  at the 75 diameter bins ( $j$ ) of VSF, respectively.

The volume fractions of the evaporated materials are calculated from the volume fractions of the residuals. The calculation for  $\Phi_{V,CV}$  is similar to that for  $\Phi_{V,LV}$ . Since the particle has completely vaporized, the vaporized volume is equivalent to the volume of the original particle. Hence,  $\Phi_{V,CV}$  is the same as  $\Phi_{N,CV}$ :

$$\Phi_{V,CV} = \frac{N_{CV} \times \frac{\pi}{6} D_{p,CV}^3}{N_{D_0} \times \frac{\pi}{6} D_0^3} = \Phi_{N,CV} \quad (5)$$

where  $D_{p,CV} = D_0$ . Since the sum of the total volume fraction of CV, VM and the residuals of LV, MV, HV equals to unity,  $\Phi_{V,VM}$  was obtained after the above volume fractions were calculated. Furthermore, we define the volume ratio of the residuals to their host particles for MV and HV ( $\Theta_{V,MV}$  and  $\Theta_{V,HV}$ ) for our discussions later:

$$\Theta_{V,i} = \frac{N_i \times \frac{\pi}{6} D_{p,i}^3}{N_i \times \frac{\pi}{6} D_0^3} = \frac{D_{p,i}^3}{D_0^3} \quad (6)$$

where  $N_i$  and  $D_{p,i}$  are the number concentration and mean residual diameter of  $i = MV$  or  $HV$  after heating, respectively.

### 2.2.3 Particle size distributions of number, volume and mass concentrations of LV, MV and HV

Due to the differences in the size cuts of the VTDMA and the OC/EC Analyzer, log-normal fits extrapolated to  $5 \mu\text{m}$  were applied to the particle number size distributions of the residuals of LV, MV and HV ( $dN/d\log D_{p,i}$ , where  $i = LV, MV$  or  $HV$ ) to estimate the volume and then mass concentrations (calculated later) of the ambient aerosols for comparison with  $\text{PM}_{2.5}$  OC/EC measurements. The volume size distributions ( $dV/d\log D_{p,i}$ ) are calculated by:

$$\frac{dV}{d\log D_{p,i}} = \frac{dN}{d\log D_{p,i}} \times \frac{\pi}{6} D_{p,i}^3 \quad (7)$$

where  $D_{p,i}$  is the mean residual diameter as defined in Sect. 2.2.2.

Volume ( $V$ ) concentrations of LV, MV and HV residuals can then be calculated by integrating the area under the fitted curves. As we only focus on LV and MV, densities of  $1.0 \text{ g cm}^{-3}$  (Hitzenberger et al., 1999) and  $1.5 \text{ g cm}^{-3}$  are applied to  $V_{LV}$  and  $V_{MV}$  to obtain mass ( $m$ ) concentrations of LV and MV, respectively. The choice of the densities is based on the assumption that LV and MV are dominated by soot and non-volatile OC, respectively.



are likely more affected by meteorology. Yu et al. (2010) reported that the mass median aerodynamic diameter of urban EC in the condensation mode in Guangzhou is about 380 nm.

Figure 4 shows the time series data of  $N_{LV}$ ,  $N_{MV}$ ,  $N_{HV}$ ,  $\Phi_{N,CV}$ ,  $\Phi_{N,LV}$ ,  $\Phi_{N,MV}$  and  $\Phi_{N,HV}$ , together with the mass concentration of  $PM_{2.5}$ . For 40 nm particles (Fig. 4a and c), in both number concentrations and fractions, HV and MV were comparable and LV was very low. There was relatively little temporal variation of their concentrations and number fractions throughout the whole period. CV had a large fraction, varying between 0.2 and 0.6. For 300 nm particles (Fig. 4b and d), CV, HV and LV fractions were small.  $N_{MV}$  tracks reasonably well with  $PM_{2.5}$  mass because of its dominance in the number concentrations. Internally mixed soot particles, manifested as MV particles, play an important role in Guangzhou, especially during polluted days. CV had a rather stable fraction of about 0.15, much smaller than that for 40 nm particles.

Figure 5 shows the diurnal variation of the number fractions. No obvious pattern of LV, MV, HV and CV was observed for particles larger than 80 nm. For 40 nm particles, clear maximum and minimum of the CV fraction are observed at 08:00 a.m. and 01:00 p.m., respectively. Furthermore, the trend of CV is opposite to those of HV and MV. For MV and HV, the average number concentrations increased since 08:00 a.m. and reached their maxima at 01:00 p.m. The increase of CV in 40 nm particles and to a lesser extent of LV in larger particles in the morning (Fig. 5) is consistent with traffic pattern, where freshly emitted volatile and non-volatile materials, likely OC and EC, are externally mixed and contributed to CV and LV, respectively. As time progresses in a day, the highly volatile species (CV) which were freshly emitted in the morning, may evaporate and react to form less volatile particles and become VM instead of CV (Robinson et al., 2007). Alternatively, these CV particles could also coagulate with smaller particles to form VM containing particles. Less fresh emissions with more CV particles turning into VM on MV and HV particles can explain the trend that the number concentration and fraction of CV decreased while those of MV and HV increased. VM does not contribute to number fraction and its significance and diurnal variation will be discussed later in

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volume fraction. The lack of diurnal trends for particles larger than 80 nm suggests that they are dominated by aged particles accumulation likely due to low mixing height in the winter. Freshly emitted particles have little contributions to the number fractions.

### 3.2 Volume fractions

5 The average and diurnal variation of volume fractions of CV, the residuals of LV, MV, and HV, and VM are shown in Fig. 6 and 7. Overall, the volume fractions of CV and VM are about 0.15 and 0.6, respectively (Fig. 6), indicating that most of the original volume of the ambient particles has evaporated at 300 °C. For 40 nm particles, CV and VM altogether contribute to almost 90 % of the total volume.  $\Phi_{V,LV}$ ,  $\Phi_{V,MV}$  and  $\Phi_{V,HV}$  are 0.15, 0.1 and less than 0.01, respectively. Size dependence of  $\Phi_{V,CV}$ ,  $\Phi_{V,LV}$ ,  $\Phi_{V,MV}$  and  $\Phi_{V,HV}$  are similar to number fractions:  $\Phi_{V,CV}$  in 40 nm particles almost doubles and  $\Phi_{V,LV}$  and  $\Phi_{V,MV}$  are only half of those in 80 nm or larger particles, and is more dominant in larger size. CV are mostly found and hence contributed to  $\Phi_{V,CV}$  in smaller particles while non-volatile materials contribute to  $\Phi_{V,LV}$  in the larger particles.

15 The diurnal variation of  $\Phi_{V,CV}$ ,  $\Phi_{V,LV}$ ,  $\Phi_{V,MV}$ ,  $\Phi_{V,HV}$ ,  $\Phi_{V,VM}$ ,  $\Theta_{V,MV}$  and  $\Theta_{V,HV}$  (the volume ratio of the residuals to their host particles for MV and HV, respectively) are shown in Fig. 7. Similar to the diurnal trends of the number fractions in Fig. 5, there was no significant variation of  $\Phi_{V,CV}$ ,  $\Phi_{V,LV}$ ,  $\Phi_{V,MV}$ , and  $\Phi_{V,HV}$  for all particle sizes except for 40 nm particles, where  $\Phi_{V,VM}$  shows a trend opposite to  $\Phi_{V,CV}$ . The opposite trends of VM and CV can be explained by the trends of the number concentrations of MV + HV and CV. When there are more MV and HV particles, volume fraction of VM increases. Based on  $\Theta_{V,MV}$  and  $\Theta_{V,HV}$ , about 70 to 80 % of  $\Phi_{V,VM}$  is from MV and there is no size dependence.

25 There was no obvious diurnal variation of  $\Theta_{V,MV}$  and  $\Theta_{V,HV}$  for all sizes (except for a minor peak at about 06:00 p.m. for  $\Theta_{V,MV}$ , which may be related to traffic), even for 40 nm particles where there is an obvious diurnal trend of  $\Phi_{V,VM}$ , which is the volume ratio of volatile materials internally mixed with non-volatile components to total volume before heating. The increase in  $\Phi_{V,VM}$  after the morning rush hours is likely attributed

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to the increase of MV particles ( $N_{MV}$ ) (Fig. 5) instead of changes in the amount of VM on the MV or HV residuals since  $\Theta_{V,MV}$  and  $\Theta_{V,HV}$  are relatively constant.

Together with the lack of size dependence and diurnal variation on the different volatility components for particles of 80 nm or larger, the high volume contribution of VM and association with MV particles suggest that aerosols sampled at the Panyu station are moderately aged with a significant amount of volatile materials in this campaign.

### 3.3 Closure analysis for LV, MV and OC/EC at 300 °C

Closure analysis of EC or EC + OC<sub>2-4</sub> (the sum EC, OC<sub>2</sub>, OC<sub>3</sub>, and OC<sub>4</sub>) and the residuals of LV + MV is conducted (Fig. 8). Good correlations ( $R^2 > 0.9$ ) for both EC and EC + OC<sub>2-4</sub> with LV + MV were obtained. Nonetheless, the slope for  $m_{LV+MV}$  to  $m_{EC}$  (2.94) is more than two times of that for  $m_{LV+MV}$  to  $m_{EC+OC_{2-4}}$  (1.22), indicating that EC alone cannot account for the mass of LV + MV. Including non-volatile OC give better mass closure for LV + MV and this observation supports our initial postulation that LV and MV may contain non-volatile OC. However, mass of EC + OC<sub>2-4</sub> do not explain all the mass of LV + MV. A possible explanation could be that the vaporizing temperatures of some OC<sub>1</sub> are close to the upper limit (310 °C), hence they were not completely vaporized in the heated tube and remained in LV + MV. The presence of other refractory materials, as well as errors during the measurements, could also be a reason for the difference. The extrapolated lognormal fitting of the size distribution of nonvolatile particles can cause errors if the mode diameter of the fitting is beyond the range of measurements of the VTDMA.

It should be noted that the heating environment in a VTDMA is different from that of an OC/EC Analyzer. In the VTDMA, aerosols were heated in the presence of oxygen, while in the OC/EC Analyzer, samples were heated in the presence of helium for OC. In the presence of oxygen, it is possible that OC<sub>2-4</sub> in the aerosols may have evaporated in the VTDMA even at 300 °C. Further study is needed to quantify the effect of oxygen on the oxidation of OC in the VTDMA.

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## 4 Conclusions

This study presents the first VTDMA measurements in the PRD, China. The measured volatility characteristics of ambient aerosols reflect the difference of the freshly emitted 40 nm particles and the aged bigger ones (80 to 300 nm). The 40 nm particles had 50 % of particles that completely evaporated at 300 °C. Particles larger than 80 nm were more aged and contained volatile components (VM) internally mixed with non-volatile residuals (MV) and VM contributed to over 50 % of the total volume fraction for all particles. From the mass closure analysis of the VTDMA data and OC/EC data, it is likely that the non-volatile residuals at 300 °C observed in VTDMA also contained some less volatile OC (OC<sub>2-4</sub>). Cheng et al. (2009) performed an aerosol optical closure to investigate the aerosol light absorption coefficient and single scattering albedo using the soot mixing states determined by a VTDMA in Yufa, Beijing. It is possible that some less volatile OCs (OC<sub>2-4</sub>) also absorb light.

The mixing state is a key parameter for accurately assessing the climatic impacts of soot particles (Jacobson, 2001). In this study, we found that externally mixed soot particles contribute to about 10 % for 40 nm particles and about 15 % for 80 nm and larger particles in number concentration. Such external mixing needs to be accounted for in assessing climatic impacts of soot particles in the region.

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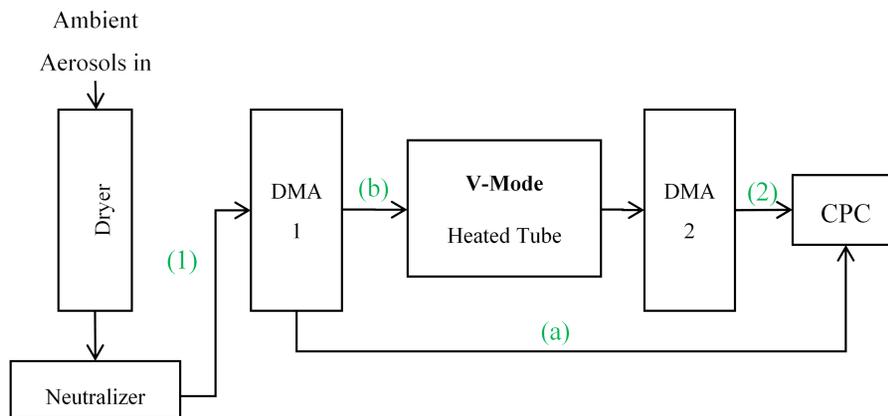
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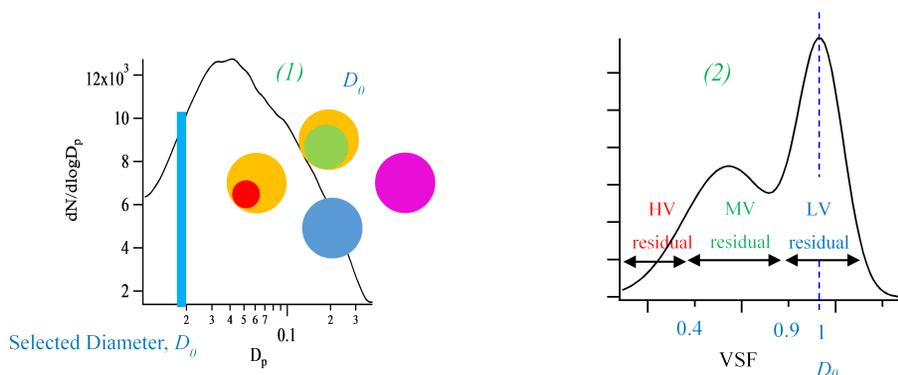
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**Figure 1.** Schematic diagram of the volatility tandem differential mobility analyzer (VTDMA).[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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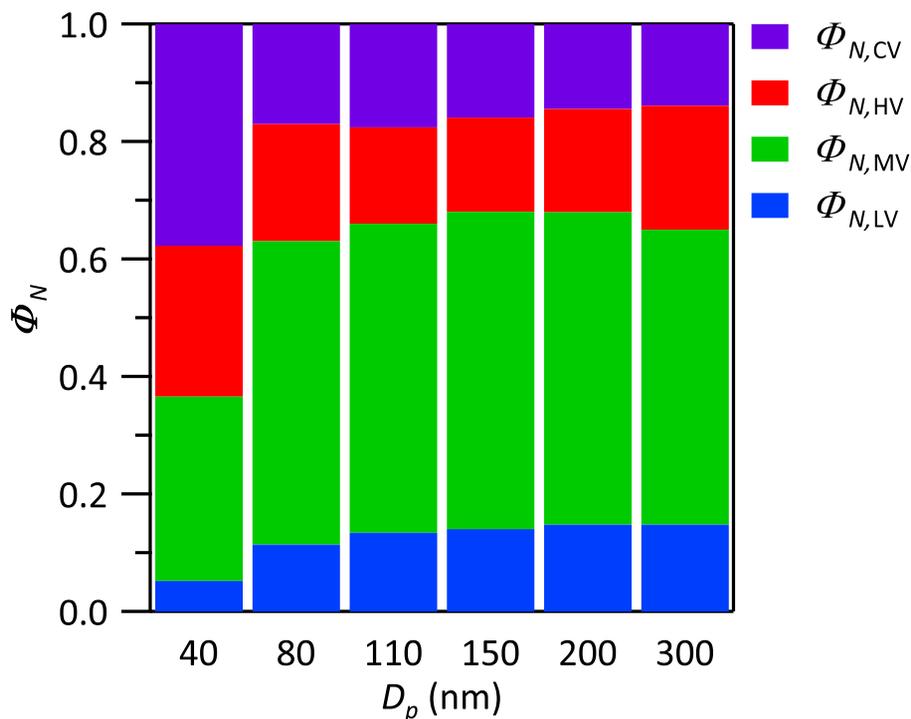
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**Figure 2.** Examples of particle size distributions of (left) ambient aerosols before entering DMA<sub>1</sub> and (right) residuals of the size-selected particles ( $D_0$ ) after heating. The left and right distributions correspond to (1) and (2) in Fig. 1 respectively. Residuals are divided into several groups – LV (blue), MV (green) and HV (red) – based on their volatilities. CV (purple) and VM (orange) are completely vaporized and hence not measured as residuals. VM appears as coating for illustration purposes only. It does not necessarily reflect the morphology of the particles.

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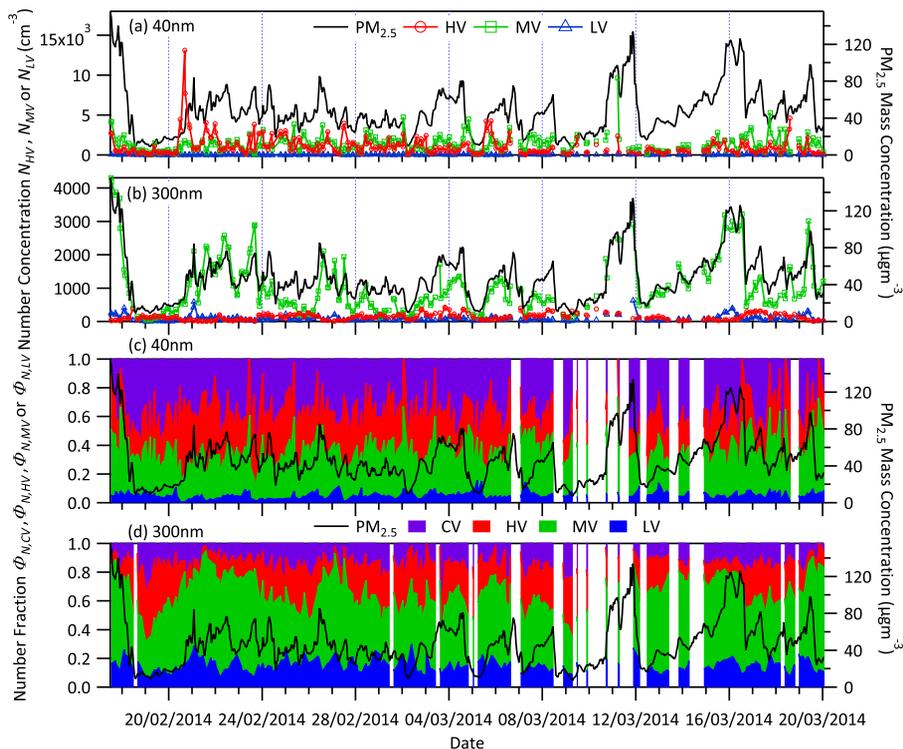


**Figure 3.** Average number fractions of CV (purple), HV (red), MV (green) and LV (blue) particles of six selected particle dry diameters (40, 80, 110, 150, 200 and 300 nm) in February and March 2014.

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**Figure 4.** Time series of number concentrations of (a) 40 nm and (b) 300 nm HV (circle, red), MV (square, green), and LV (triangle, blue) particles; number fractions of (c) 40 nm and (d) 300 nm CV (purple), HV (red), MV (green) and LV (blue) particles during the measurement period from 17 February to 20 March 2014. Time series of mass concentrations of PM<sub>2.5</sub> (solid line, black) are plotted on the right axis of each plot.

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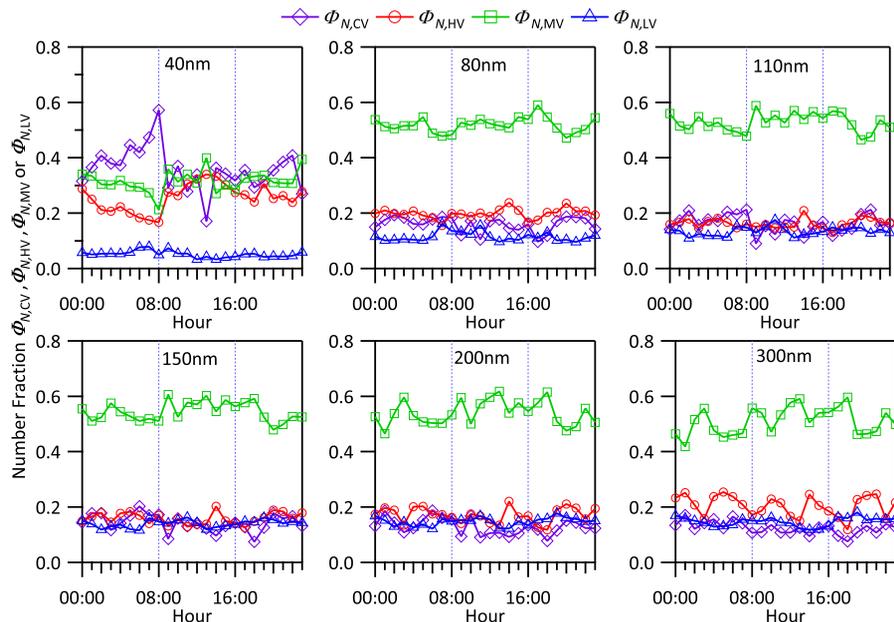
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**Figure 5.** Diurnal variation in number fractions of CV (diamond, purple), HV (circle, red), MV (square, green) and LV (triangle, blue) particles that are 40, 80, 110, 150, 200 and 300 nm in dry size.

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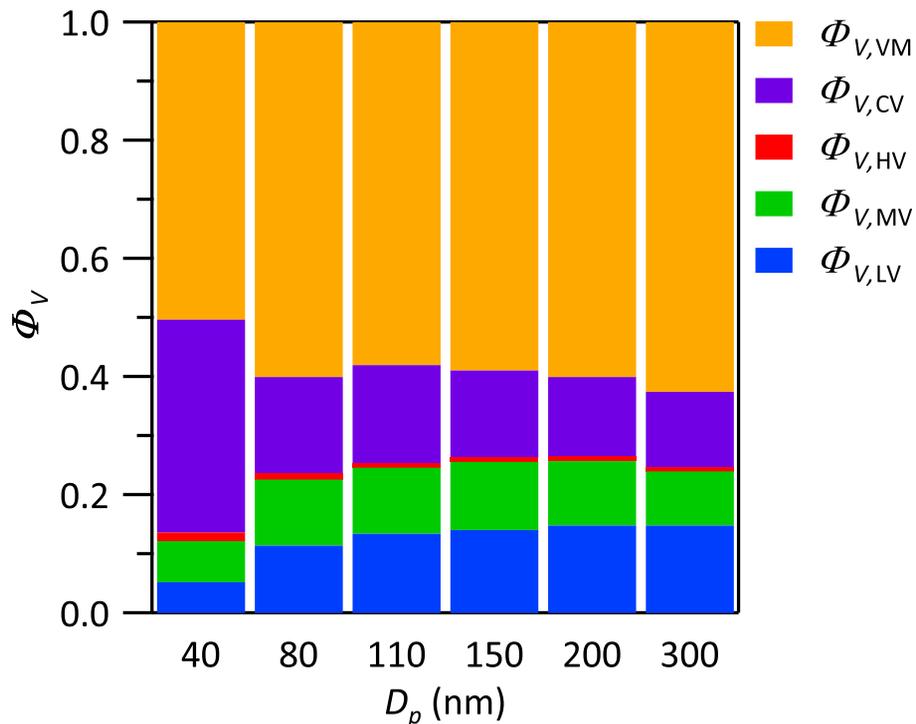
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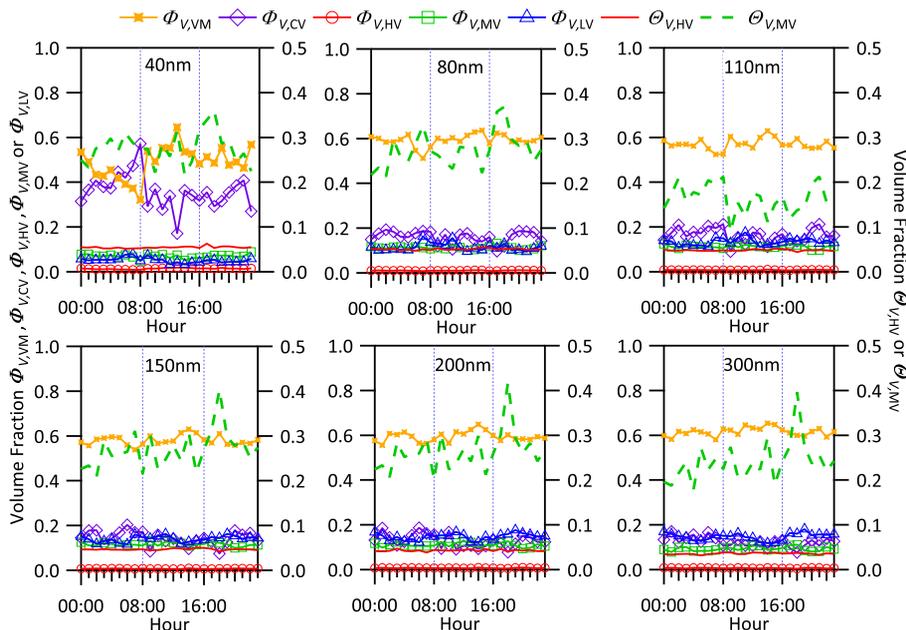


**Figure 6.** Overall average volume fractions of VM (orange), CV (purple), HV (red), MV (green), and LV (blue) particles as a function of particle diameter.

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**Figure 7.** Diurnal variation in (left axis)  $\Phi_{V,VM}$  (cross, orange),  $\Phi_{V,CV}$  (diamond, purple),  $\Phi_{V,HV}$  (circle, red),  $\Phi_{V,MV}$  (square, green),  $\Phi_{V,LV}$  (triangle, blue), (right axis)  $\Theta_{V,HV}$  (solid line, red) and  $\Theta_{V,MV}$  (dashed line, green) particles that are 40, 80, 110, 150, 200 and 300 nm in dry size.

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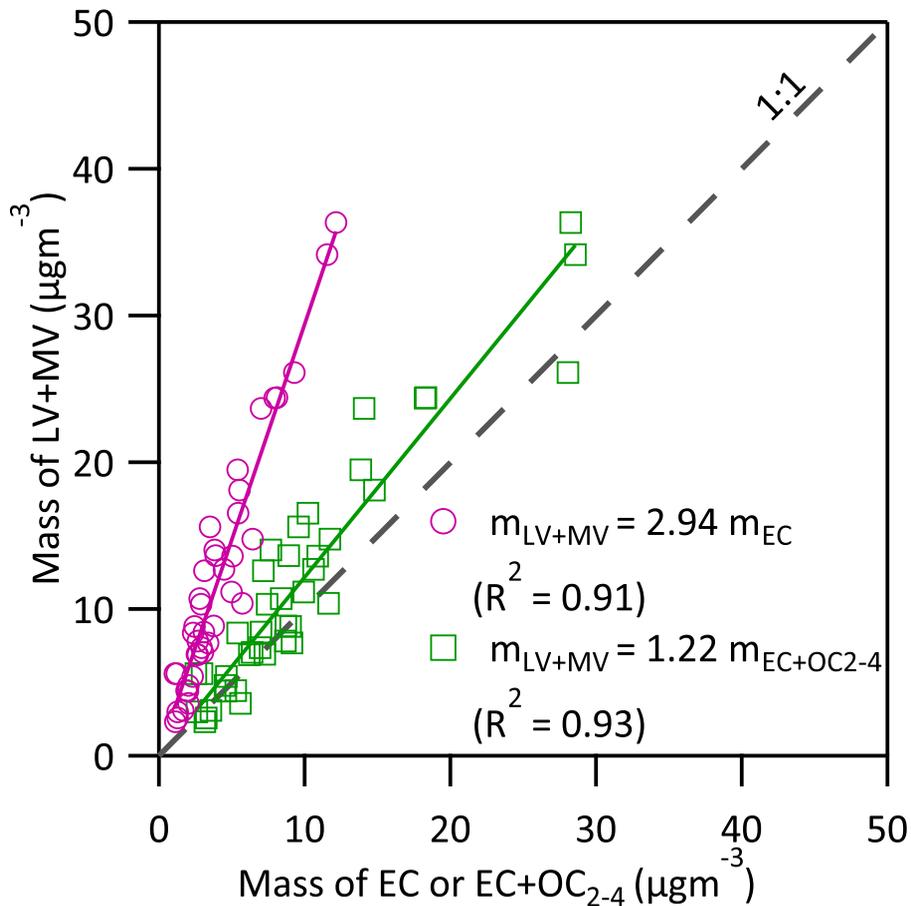
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**Figure 8.** Closure analysis of the total mass of LV and MV (LV + MV) from VTDMA at 300 °C and measured mass of EC (circle, purple) or total of EC and OC<sub>2-4</sub> (square, green) from the OC/EC Analyzer.