

Aerosol volatility measurement using an improved thermodenuder: Application to secondary organic aerosol

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Abstract

An improved thermodenuder is used to measure the volatility of secondary organic aerosol (SOA) produced during α -pinene/O₃ and α -pinene/NO_x photooxidation. The thermodenuder allows a wide range of aerosol residence times in the heated zone compared to existing systems avoiding the complications due to slow mass and heat transfer processes. The performance of the thermodenuder was tested using mono-disperse ammonium sulfate particles.

The volatility of SOA was investigated in the 50–220 °C temperature range. Almost 98% of the SOA volume generated from the α -pinene/O₃ reaction evaporated at 75 °C after 15.8 s in the heated zone. However, more than 50% of the particle mass did not volatilize at 100 °C when the residence time was reduced to 1.6 s. The SOA obtained from α -pinene/NO_x photooxidation showed similar volatility characteristics even after 10 h of “aging” in the smog chamber. The measured remaining aerosol mass after the particles pass through the thermodenuder is quite sensitive to their residence time in the heated zone of the system, for residence times of the order of seconds. Interpreting the remaining aerosol mass as non-volatile even when the thermodenuder operates at temperatures above 200 °C may be erroneous if low residence times (less than a few seconds) are used.

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1. Introduction

High concentrations of atmospheric aerosols have been linked to increased respiratory illness and mortality (Dockery et al., 1993; Pope et al., 2002). Atmospheric particles scatter and absorb visible light thus reducing visibility. Their direct interaction with solar radiation and their ability to act as cloud condensation nuclei makes atmospheric aerosol an important element of the energy balance of our planet (Twomey, 1974).

A significant fraction of the atmospheric aerosol mass is semivolatile: the corresponding compounds can exist in significant concentrations in both the gas and particulate phases. The most important semivolatile inorganic aerosol compounds are ammonium nitrate and ammonium chloride and their atmospheric behavior is relatively well understood (Seinfeld & Pandis, 1998). On the other hand, there is little known about the behavior of the hundreds of semivolatile

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organic components of atmospheric particles. Understanding the partitioning of these organic compounds between gas and aerosol phases has been recognized as one of the major challenges in our efforts to quantify the rates of formation of secondary organic particulate matter (Kanakidou et al., 2005).

Measurement of the volatility of atmospheric aerosol can provide indirect information about its chemical composition (Jennings & O'Dowd, 1990; Clarke, 1991, 1993; Brock, Hamill, Wilson, Honsson, & Chan, 1995; Orsini, Wiedensohler, & Stratmann, 1999; Sakurai et al., 2003; Philippin, Wiedensohler, & Stratmann, 2004; Kalberer et al., 2004; Wehner, Philippin, Wiedensohler, Scheer, & Vogt, 2004; Kuhn et al., 2005). The change in size of a polydisperse or monodisperse aerosol population can be measured at low cost, online, and can be easily coupled with standard methods for aerosol size-distribution measurements. Volatility tandem differential mobility analyzers (VTDMA) coupling a thermodenuder with a TDMA system are used to measure the change in size of a monodisperse aerosol population (Philippin et al., 1999). There are various research and commercial models of thermodenuders available. Two companies, Dekati Ltd. (Osuusmyllynkatu 13, FIN-33700 TAMPERE, FINLAND) and TSI Inc. (500 Cardigan Road, Shoreview, MN 55126-3996, U.S.A.) provide commercial thermo-denuders. The TSI system (TSI, Model 3065) is restricted to flow rates less than 3 L min^{-1} and has a maximum temperature of 400°C . The Dekati thermodenuder (Dekati ELA 111 and 230) allows higher flow rates $10\text{--}20 \text{ L min}^{-1}$ but its maximum temperature is 300°C . The commercial models allow relatively small residence times, e.g., 0.3 s for the Dekati thermodenuder and the 1–1.5 s for the TSI unit. Burtscher et al. (2001) developed and tested a research-grade thermodenuder primarily for field campaign combustion exhaust measurements improving upon previous designs. Wehner, Philippin, and Wiedensohler (2002) improved further the above thermodenuder design by adding insulation to the heating component to obtain stable temperature profiles and by increasing the residence time in the heating zone to 9 s (at a flowrate of 0.6 L min^{-1}).

Thermodenuders have been recently used in laboratory studies to provide additional information about the composition of SOA. Baltensperger et al. (2005) reported that 65%, 25% and 10% of the SOA from the photooxidation of α -pinene/ NO_x remained in the particulate phase after heating for 1.5 s at 100, 150 and 200°C , respectively. After 24 h of aging 10%, 60% and 80% of the SOA from photooxidation of trimethylbenzene evaporated at 100, 150 and 200°C , respectively (Kalberer et al., 2004). These authors provided strong evidence that the lower volatility of the aged secondary organic aerosol (SOA) was associated with the formation of oligomers.

The thermodenuders used in most previous studies have been designed for field deployment and therefore allow relatively short residence times of the aerosol in the heated zone (usually of the order of 1 s or less). The small residence time and the potential resistances to mass transfer of material from the particulate to the gas phase (e.g., diffusion in the particulate phase) may lead to an underestimation of the actual volatility of secondary organic particulate matter.

In this study, we investigate the performance of a laboratory thermodenuder that allows a wide range of residence times (up to 20 s) and temperatures. The system is applied to SOA produced from α -pinene/ O_3 and α -pinene/ NO_x photooxidation reactions in the Carnegie Mellon University smog chamber. The effect of the residence time in the thermodenuder on the volatility measurements of SOA is investigated.

2. Experimental description

2.1. Thermodenuder design

Fig. 1 shows a schematic of the thermodenuder used in this study. The basic design builds upon the ideas of Wehner et al. (2002) and Burtscher et al. (2001). The system consists of two parts; a heating section and an organic vapor adsorption and cooling system.

The heating part consists of three components: a furnace (Electro heat system Inc.), a stainless steel tube, and a heating controller. The furnace is used as a heater and an insulator. No additional insulation is necessary making the heating part of this design simpler. The furnace is operated using a digital controller (WATLOW, series 988) which controls temperatures of the aerosol exiting the heating zone approximately within 1°C . The thermodenuder temperature (in the heated and adsorption/cooling sections) remains stable (deviations of less than 1°C) for several hours at flow rates up to 5 L min^{-1} and heating zone temperatures from 50 to 400°C . The temperature profile inside a similar system has been investigated by Wehner et al. (2002). The heating tube is made of stainless steel with inner diameter of 3.5 cm and total length 83 cm; however, the actual heating zone has a length of 55 cm. At a flow rate of 1 L min^{-1} , the average residence time of the aerosol in the furnace is 15.8 s. This is significantly longer than most existing commercial and research-grade thermodenuders.

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