

General Comments

This manuscript describes the design and operation of a TD-DMA-CIMS. While I would not call the technique novel (as thermal desorption-chemical ionisation mass spectrometry has been demonstrated and utilized in both laboratory and field studies) the authors have developed a new DMA enabling particle collection and heating in-situ and overall they do a good job characterising the instrument. My main concern, noted below, is that the authors do not compare their TD-DMA-CIMS implementation directly to prior TD-CIMS works and in their characterisation, they do not explain some critical assumptions regarding DMA characterisation.

Specific Comments:

1. The main issue I see is with a lack of comparison to prior TD-CIMS implementations. The authors do acknowledge the original TD-CIMS work (Voisin et al, 2003), but subsequent development and characterization efforts (Smith et al. 2004, McMurry et al. 2009) are overlooked. I think the authors need to make clearer in this study what is new/unique about their implementation of DMA-thermal desorption-chemical ionisation mass spectrometry. How does their limit of detection compare to prior implementations? I understand that the authors are able to make simultaneous particle and gas phase measurements, but I think this would also be possible with prior implementations of this technique.
2. Table 1 and lines 7-27. The table omits techniques where electrospray ionisation is utilized to generate ions. This is an important ionization technique which minimally fragments parent ions, and while it is sparingly used in aerosol science, there are a number of studies demonstrating its potential in aerosol analysis. I would recommend adding electrospray ionisation based methods to table 1 and mentioning them in the text as these methods can be applied to sub-30 nm particles. Specifically, He et al (2015) showed that nanoparticles can be collected electrostatically and then ions can be generated via electrospray, Horan et al (2017) showed that electrospray-like mass spectra can be collected for aerosol particles without the need for a distinct collection step, and SESI (secondary electrospray ionisation, sometimes called extractive electrospray ionisation), though it cannot distinguish between gas and particle phase, has been employed in several studies (Doezema et al. 2012, Gallimore and Kalberer 2013).
3. Equation (1) and the surrounding analysis. While the authors do note “Note that the aerosol coming from the DMA is not strictly monodisperse; instead the DMA provides a Gaussian-shaped size distribution,” it does not appear they account for this in their analysis or explain to readers why they do not need to account for this. Equation (1) is not the true transfer function of the DMA; this would only be the transfer function if the first DMA had extremely high resolution relative to the test DMA. Looking at the sheath flow rate employed in the TD-DMA relative to the aerosol flowrate, this is probably the case and the authors’ analysis is ultimately fine, but I would advise the authors to show this more clearly. Specifically, the number concentration of particles at the outlet of the first DMA is given by the equation:

$$N_2 = \int_0^{\infty} \frac{dn}{dd_p} \tau_G(d_p) dd_p$$

where $\tau_G(d_p)$ is the Grimm DMA transfer function/transmission function. For particles exiting the TD-DMA, the number concentration is:

$$N_1 = \int_0^\infty \frac{dn}{dd_p} \tau_G(d_p) \tau(d_p) dd_p$$

The ratio N_1/N_2 is hence not given by equation (1), but is:

$$\frac{N_1}{N_2} = \frac{\int_0^\infty \frac{dn}{dd_p} \tau_G(d_p) \tau(d_p) dd_p}{\int_0^\infty \frac{dn}{dd_p} \tau_G(d_p) dd_p} \neq \tau(d_p)$$

A common (reasonable) assumption is that $\frac{dn}{dd_p}$ is a constant over the region where $\tau_G(d_p)$ takes non-zero values. This leads to:

$$\frac{N_1}{N_2} = \frac{\int_0^\infty \tau_G(d_p) \tau(d_p) dd_p}{\int_0^\infty \tau_G(d_p) dd_p} \neq \tau(d_p)$$

Therefore, the method noted in the manuscript at present does not yield $\tau(d_p)$ unless $\tau_G(d_p)$ is significantly narrower (higher resolution) than $\tau(d_p)$. At an aerosol to sheath flow ratio of 3/5, I assume the assumption is reasonable, but does need to be justified or better yet, explicitly shown in the manuscript.

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