



### Supplement of

# Exploring the applicability and limitations of selected optical scattering instruments for PM mass measurement

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#### S1 Estimation of aerosol density used for SMPS

The density of ambient aerosol was calculated based on organic, SO<sub>4</sub>, NO<sub>3</sub>, and NH<sub>4</sub> mass concentration measured by AMS (Zhang et al. 2005). It assumes that the organic composition had an averaged density of 1.20 g cm<sup>-3</sup>, and that SO<sub>4</sub> and NO<sub>3</sub> were both components of salts, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (density 1.79g cm<sup>-3</sup>) and NH<sub>4</sub>NO<sub>3</sub> (density 1.72g cm<sup>-3</sup>) respectively (Lee et al. 2015). The averaged ambient density was then calculated by:

$$\rho_{ambient} = \frac{m_{Total}}{\frac{m_{organic}}{\rho_{organic}} + \frac{m_{SO_4}}{\rho_{(NH_4)_2 SO_4}} \times \frac{132}{96} + \frac{m_{NO_3}}{\rho_{NH_4 NO_3}} \times \frac{80}{62}}$$

Where  $\rho$  is density, and m is the mass concentration.

#### S2 Figures



Figure S1. The correlation between the reported mass concentration by optical sensors (DRX and PDR) and by TEOM.

For this size PSL, the reading of OPC-N2 is not included. The OPC-N2 did not produce a response for the 304 nm and smaller PSL particles because of its 380 nm lower detection limit.



Figure S2. (a) Results of a Mie scattering calculation for conditions appropriate to the DRX (black) and PDR (red); (b) Correlation between the relative flux with the ratio of optical instruments to TEOM (DRX: black; PDR: blue; and red points relates with the result of 90 nm PSL).



Figure S3. The normalized volume distribution of three aerosol groups. (a) Group 1, with low concentration dilutions. (b) Group 2, with medium concentration dilutions. (c) Group 3, with high concentration dilutions.

The volume concentration for each bin is divided by total integrated value for all bins to obtain normalized volume concentration in each bin, which shows the size distribution of different particles. Figure S3 shows the shift in volume distributions for liquid samples with increasing concentration dilution. As the concentration increases, the volume distribution shifted rightward towards larger particles.



Figure S4. Correlation between the readings of optical sensors and the reference TEOM measurement for 0.45 g  $L^{-1}$  sucrose (For convenient comparison, PDR data is multiplied by 3, while OPC-N2 data s multiplied by 50).





Figure S5. (a) A time series of 1-hour aerosol composition concentration and SMPS mass median diameter. (b) The mass fraction of different chemical species in the composition from 12/22/2016 to 01/07/2017 and from 09/27/2017 to 10/01/2017.



Figure S6. (a) A scatterplot showing correlation of SMPS measurements to concurrent AMS measurements, and (b) correlation of DRX measurements to concurrent PDR measurements.



Figure S7. (a) Scatterplot of organic mass fraction and the PDR/AMS ratio. Points are color-coded by aerosol median diameter. (b) Scatterplot and correlation of aerosol median diameter with the DRX/AMS ratio. Points are color-coded by organic mass fraction. (c)

## Scatterplot of organic mass fraction with the DRX/AMS ratio. Points are color-coded by aerosol median diameter. Points are sized by AMS mass concentration.

In Fig. S7b the correlation between aerosol median diameter with the DRX/AMS ratio was lower than with PDR/AMS, with the most deviated points the result of smaller diameter particles. One possible reason for the low correlation coefficient with DRX is that particles were with smaller size than other periods, and the aerosol with diameters smaller than 100 nm would cause significant bias to DRX, due to its size detection limit. After excluding the points with smaller size, the  $r^2$  increases to 0.52, which demonstrates the positive effect of aerosol size on optical instrument response.



Figure S8. (a) Averaged mass size distribution of long-distance transported aerosol composition, and (b) averaged mass size distribution of fresh aerosol composition.





0 hr forecast valid Mon 00z 2017-09-25



(d) 09/28/2017

0 hr forecast valid Wed 00z 2017-09-27

0 hr forecast valid Thu 00z 2017-09-28



(e) the map of fires and thermal anomalies from Terra and MODIS

Fig. S9. GEOS-5 forecasts for the spatial distribution of sulfate aerosol AOT over North America (https://portal.nccs.nasa.gov) on 09/24/2017, 09/25/2017, 09/27/2017, and 09/28/2017 (above four), and the map of fires and thermal anomalies from Terra and MODIS (https://worldview.earthdata.nasa.gov) on 09/23/2017

It is believed that the sulfate aerosol originated from the lower Mississippi Valley on 09/24/2017, as a result of multiple point-source fires clustered in the region. As time progresses, the sulfate aerosol distribution changed with synoptic downstream flow, and created the prominent band that passes over New York State on 09/28/2017. During this process, the aerosol was believed to have experienced long-distance transport.



Fig. S10. Time series of estimated aerosol refractive index estimated using the PDR and DRX.



Figure S11. (a) The correlation scatterplots of aerosol refractive index and the PDR/AMS ratio. (b) The correlation scatterplot of aerosol refractive index and the DRX/AMS ratio. All points are color-coded by aerosol median diameter, and sized by AMS mass concentration

#### S3 Tables

 Table S1. List of different dilution concentrations for each compound in the three test groups,

 and the volume median diameter of the generated size distribution.

Compound	Group 1	Group 2	Group 3
NaNO <sub>3</sub> (g $L^{-1}$ )	0.75	3	6
$(NH_4)_2SO_4 (g L^{-1})$	0.75	2.5	5
Sucrose (g L <sup>-1</sup> )	0.45	1.5	3
Adipic Acid (g L <sup>-1</sup> )	0.4	1.0	2.5
Median Diameter (nm)	153	202	231

Table S2. Calculated relative Mie scattering flux of the four particle species ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, NaNO<sub>3</sub>, sucrose, adipic acid) analyzed by the PDR and DRX, and different dilution concentrations (#1 indicates Group 1; #2 indicates Group 2; #3 indicates Group 3).

Relative flux	(NH4)2SO4	NaNO <sub>3</sub>	sucrose	adipic acid
DRX(#1)	3.37	2.56	3.34	2.50
DRX(#2)	4.72	5.04	5.04	3.19
DRX(#3)	5.41	5.54	5.54	3.72
PDR(#1)	0.79	0.59	0.86	0.69
PDR(#2)	1.31	0.93	1.45	0.96
PDR(#3)	1.59	1.03	1.67	1.24

Table S3. Reference table RF for different refractive indices (TP refers to the time point for each

hour of data, wavelength=880 nm, angle=90°)

	Index/RF	TP1	TP2	TP3	TP4	TP5	TP6	TP7	ТР
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1.2	0.40	0.39	0.39	0.40	0.40	0.41	0.41	
1.21	0.45	0.43	0.43	0.44	0.44	0.45	0.45	
1.22	0.49	0.47	0.47	0.48	0.49	0.49	0.50	
1.23	0.53	0.52	0.51	0.52	0.53	0.54	0.54	
1.24	0.58	0.56	0.56	0.57	0.58	0.59	0.59	
1.25	0.63	0.61	0.61	0.62	0.63	0.64	0.64	
1.75	6.04	5.85	5.73	5.91	6.01	6.06	6.11	
1.76	6.22	6.02	5.91	6.09	6.19	6.25	6.30	
1.77	6.41	6.21	6.08	6.28	6.37	6.43	6.49	
1.78	6.60	6.39	6.26	6.46	6.56	6.62	6.69	
1.79	6.79	6.58	6.45	6.65	6.75	6.82	6.88	
1.8	6.99	6.77	6.63	6.84	6.95	7.02	7.08	

#### **References:**

Zhang, Q., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., and Jimenez, J. L. Time-and size-resolved chemical composition of submicron particles in Pittsburgh: Implications for aerosol sources and processes. Journal of Geophysical Research: Atmospheres, 2005, 110(D7), 2005.

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