## Supplement of Bimodal distribution of size-resolved particle effective density in a rural environment in the North China Plain

## Yaqing Zhou et al.

Correspondence to: Nan Ma (nan.ma@jnu.edu.cn) and Qiaoqiao Wang (q.wang2@outlook.com)

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Figure S1. Schematic diagram for particle effective density and PM<sub>1</sub> measurement.



Figure S2. Calibration results for DMA-CPMA-CPC system.



**Figure S3**. Particle effective density ( $\rho_{eff}$ ) distribution for (a) 50 nm, (b) 100 nm, (c) 150 nm, (d) 220 nm, and (e) 300 nm particles during the entire sampling period. The black lines and blue circles represent the geometric mean for the main-density mode ( $\bar{\rho}_{eff,main}$ ) and the sub-density mode ( $\bar{\rho}_{eff,sub}$ ) respectively at each size.



**Figure S4**. Trend lines of effective density ( $\rho_{eff}$ ) against mobility size for the entire sampling period, polluted and clean condition.



**Figure S5**. Diurnal cycle of (a) organics, nitrate, ammonium, sulfate, chloride and BC, (b) primary (POA) and secondary organic aerosols (SOA) mass fraction.



Figure S6. Diurnal cycle of the occurrence frequency of the sub-density mode at five mobility sizes.



**Figure S7**. Volume distribution of  $PM_{0.7}$  during (a) the entire sampling period, (b) polluted and (c) clean conditions. The shaded areas are volume distributions for particles below 300 nm, and vertical dotted lines are the peak positions of  $PM_{0.7}$  volume distribution.



**Figure S8**. (a) Mean square error (MSE) and R-square ( $R^2$ ) results from OA density sensitivity test. (b) Comparison of the average effective density of particles at 300 nm observed by DMA-CPMA-CPC ( $\bar{\rho}_{eff,tot,300nm}$ ) and ACSM-derived bulk effective density ( $\rho_{eff,ACSM}$ ). The colored dots in the right figure represent  $\rho_{eff,ACSM}$  based on different OA density assumption. Red line in the right figure is the line with slope of 1.



Figure S9. The mass fraction (top of the figure) and mass concentration (bottom of the figure) of  $PM_1$  chemical components under polluted and clean conditions.

size(nm)	$\bar{\rho}_{\rm eff,main}({ m g~cm^{-3}})$	size <sub>double</sub> <sup>a</sup> (nm)	$\rho_{\text{double}^{b}}(\text{g cm}^{-3})$	$k_{ m double}{}^{c}$
50	1.18	73	1.16	1.59
100	1.21	151	1.23	1.78
150	1.25	235	1.30	2.05
220	1.31	360	1.42	2.39
300	1.37	510	1.55	2.80

**Table S1**. Doubly charge estimation for five mobility size

<sup>a</sup> mobility size of doubly charged particles after penetrate DMA based on Boltzmann distribution <sup>b</sup> effective density at each size<sub>double</sub> retrieved from linear fit equation of  $\bar{\rho}_{eff,main}$ <sup>c</sup> the ratio of geometric mean of doubly charged mass to single charge mass in CPMA mass distribution