

SUPPORTING INFORMATION

Quantifying the Chemical Composition and Real-time Mass Loading of Nanoplastic

Particles (NPPs) in the Atmosphere using Aerosol Mass Spectrometry

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1 **1. Aerosol Mass Spectrometry and Potential Aerosol Mass reactor operation**

2 **conditions**

3 The Aerosol Mass Spectrometry (AMS) is the main instrument for collecting the mass
4 spectra of the nanoplastic particles in this study. Due to the aerodynamic lens and the chopper,
5 the AMS samples particles 10 million times more efficient than sampling gases.¹ Other
6 statistical tools, including the PMF, are used to further aid the data analysis of complicated
7 particle mass spectra collected from the AMS and described in Section S2 below.

8 The Potential Aerosol Mass (PAM) reactor is a horizontal 13 L copper cylindrical
9 chamber (46 cm long × 22 cm inner diameter) operated in continuous flow mode, and could
10 be used to generate Secondary Organic Aerosols (SOA) from Volatile Organic Compounds
11 (VOCs) with either the ozonolysis or photooxidation pathways.^{2,3} In this study, the SOA
12 particles are formed in the absence of any seed particles through OH radical or ozone oxidation.
13 Regarding OH radical oxidation, a 1 liter-per-minute (LPM) purified compressed air passing
14 through the deionized water was introduced into the PAM for the humidification. Another 1.5
15 LPM air flow was introduced into the homebuilt ozone generator and then entered the PAM.
16 The VOC precursors were introduced into the PAM via the syringe pump injection into a three-
17 neck round-bottom flask, which was carried out by a 1 LPM compressed air flow to the PAM.
18 The OH radicals were generated via the photolysis of ozone with UV lamps in the PAM. In
19 addition, 1 LPM flow of ozone was introduced into the PAM and the ozonolysis was conducted
20 under the dry condition. The UV lights in the PAM were turned off and the ozonolysis was the
21 predominated reaction under dark condition to form low volatile oxidation products, which
22 then undergo self-nucleation to form SOA particles.

23 **2. Positive matrix factorization (PMF) working principle**

24 PMF was first introduced in 1994 by Paatero and Tapper,⁴ and has been a useful tool
25 for resolving the time series data for mass spectrometer. The fundamental principle in PMF

26 analysis is that the measured data matrix of mass spectrum could be expressed by the
27 combination of selected number of factors based on mass conservation.⁵ The organic data
28 matrix *org* with the dimensions $t \times m$ is modelled according to Eq. S1:

$$29 \quad org_{i,j} = \sum_{p=1}^P ts_{ip}ms_{pj} + e_{ij} \quad (S1)$$

30 where P is the number of factors in the solution, $org_{i,j}$ is the signal of ion fragment j at time
31 step i in the organic data matrix, ts_{ip} is the concentration of factor p at the given time step i,
32 ms_{pj} is the fraction of ion fragment j in the mass spectrum for the particular factor p, and e_{ij}
33 is the residual not included by the solution for ion fragment j at time step i. With a given factor
34 number, the PMF solution provides a minimum summation of the weighted squared residuals
35 Q as indicated by Eq. S2 below:

$$36 \quad Q = \sum_{i=1}^t \sum_{j=1}^m (e_{ij}/\sigma_{ij})^2 \quad (S2)$$

37 where σ_{ij} is the estimated errors corresponding to the $org_{i,j}$. As the Q value also depends on
38 the dimensions of the data matrix and the number of factors chosen, normalizing the Q value
39 with the expected Q value (Q_{exp}), which is the degree of freedom of the solution, leads to a
40 useful diagnostic. The absolute Q/Q_{exp} value could be influenced by the unknown modelled
41 uncertainties, wrongly chosen number of factors, etc., and consequently, monitoring how the
42 Q/Q_{exp} varies among different iterations could assist in assessing the optimal solution.⁶

43 Ions with signal-to-noise ratio (S/N) lower than 0.2 were removed, and the ions with
44 S/N between 0.2 and 2 were downweighed by increasing the estimated error values.^{5,7} The
45 PMF solutions with one to ten factors are evaluated in this study. To further explore how
46 rotation and the random starting point would influence the uncertainty of the solutions,
47 different FPeak (from -1 to +1, step: 0.1), and SEED values (from 1 to 10, step: 1) were
48 adapted, as described by Zhang et al.⁸

49 **3. Detailed analysis procedures of ME-2**

50 As mentioned in the main text section 2.3, ME-2 allows for solution of S1 while using
 51 a prior information about component mass spectra or time series. In this work we utilize a
 52 reference input profile for the PS mass spectrum with a scalar a value that determines the extent
 53 to which the derived factor profile could vary from the input spectrum profile.^{6,9} The a value
 54 spans from 0 to 1 with an increment of 0.1, while the lower the a value, the more rigorous the
 55 constraint is. For example, if $a = 0.1$, for the profile of the extracted factor, the intensity of all
 56 the ions could vary as much as $\pm 10\%$ compared with the input reference spectrum. The ME-
 57 2 with the a value approach could provide a more complete exploration of the rotational
 58 ambiguity of the solution space.⁶

59 4. The relative ionization efficiency (RIE) of polystyrene nanoplastic particles

60 To develop the calibration curve of pure polystyrene (PS) nanoplastic particles (NPP),
 61 a mixing condensation particle counter (CPC, Model 1720, Brechtel) was deployed together
 62 with AMS. The aerosols generated are split into two lines for AMS and the CPC, and the
 63 detected ion rate in Hz (I) from AMS could be calibrated with the CPC measurements.^{10,11} The
 64 AMS ion signal was compared with the input number of molecules derived from the CPC
 65 particle number concentration and an assumed collection efficiency of 1 to obtain the ionization
 66 efficiency.

67 With ammonium nitrate as the standard, the relative ionization efficiency (RIE) for PS
 68 particles can be derived by comparing the ionization efficiency of the PS and ammonium nitrate.
 69 The detailed equation to calculate RIE is described in Eq. S1, Eq. S2, and Eq. S3 below:¹⁰

$$70 \quad C_S = \frac{10^{12} MW_S}{IE_S Q N_A} \sum_{all\ i} I_{S,i} \quad (S\ 1)$$

$$71 \quad \frac{IE_S}{MW_S} = RIE_S \frac{IE_{NO_3}}{MW_{NO_3}} \quad (S2)$$

$$72 \quad C_S = \frac{10^{12} MW_{NO_3}}{RIE_S IE_{NO_3} Q N_A} \sum_{all\ i} I_{S,i} \quad (S3)$$

73 where the C_S is the mass concentration of the PS particles in this study, Q is the
 74 volumetric sample flow rate in $\text{cm}^3 \text{s}^{-1}$, N_A is Avogadro number, and $\sum_{all\ i} I_{S,i}$ is the summation

75 of the detected ion rate in Hz for all fragment ions. IE_s and MW_s are the ionization efficiency
76 and molecular weight of the PS particles, respectively, and the IE_{NO_3} and MW_{NO_3} represent the
77 ionization efficiency and molecular weight of nitrate, respectively. The molecular weight of
78 polystyrene cancels out by combining Eqns. (S1) and (S2) to derive Eqns. (S3).

79 To further test the reproducibility of the decomposition products of PS NPPs, the
80 correlation values of the five mass spectra of PS NPPs collected during the five points
81 calibration was calculated and plotted in Figure S4 below, and it shows nearly identical mass
82 spectra. Besides, during the calibration experiment, the background blank samples were tested.
83 Below in Figure S5, S6, S7 show the AMS mass spectra collected during the nitrate calibration,
84 filtering period and the time series of tracer ions during the filtering period, which are 16 and
85 14 times lower than ambient-derived concentrations, respectively.

86 **5. The comparison of PS NPPs quantification with the AMS and pyrolysis-GC/MS**

87 To further validate the quantification of PS NPPs with the AMS, the pyrolysis GC-MS
88 analysis was also conducted following the established method.¹² PS NPPs standard were
89 collected by the AMS and onto a glass fiber filter (Cytiva, 0.7 μm particle retention)
90 simultaneously with 2 liter per minute flow rate for 77 minutes.

91 The pyrolysis GC-MS method was calibrated with the mass of PS NPP standards
92 ranging from 0.5 to 5 μg with the method established in previous study.¹² Briefly, the standards
93 were first injected in the pyrolysis chamber. Pyrolysis was performed at 700 $^{\circ}\text{C}$ for 1 min, and
94 thermodesorption was performed at 300 $^{\circ}\text{C}$ for 1 min. Gas phase pyrolysis products were
95 injected directly into a Shimadzu GCMS-QP2020NX (Shimadzu Corporation, Kyoto, Japan)
96 equipped with an RTX-1 capillarity column (30 m, 0.25 mm i.d., 25 μm film thickness). Column
97 flow was 1.1 mL/min with helium as the carrier gas at a split ratio of 5. The injection port
98 temperature was set at 310 $^{\circ}\text{C}$. GC oven started at 50 $^{\circ}\text{C}$ for 2 min and ramped to 180 $^{\circ}\text{C}$ at 15
99 $^{\circ}\text{C}/\text{min}$, increased to 310 $^{\circ}\text{C}$ at 5 $^{\circ}\text{C}/\text{min}$, and hold at 310 $^{\circ}\text{C}$ for 44 mins. Mass spectrometry

100 scanned from 50 to 600 m/z range with ion source temperature at 200 °C and interface
101 temperature at 280 °C. As shown below in Figure S8, the peak (~5 min) on the total ion
102 chromatogram (TIC) shows mass pattern matching to Styrene, a monomer of PS.¹³ The instrument
103 and membrane filter blanks do not show this PS peak on their chromatogram. The areas of the PS
104 peak (~5 min) on the TIC showed a linear relationship with the spiking masses, ($R^2 > 0.999$).
105 The linear relationship indicates the feasibility of quantification for PS MPs in test samples.

106 Two replicated sets of 16 circular pieces (1.9 mm diameter) with a total area of 0.454
107 cm² were cropped from the sample filter. Given that the effective filter area is 7.55 cm², the
108 mass of PS in each set of 16 circular pieces was projected to the mass of PS in the whole
109 sampling filter by multiplying 16.63 (the ratio of the sample area to the measured area). Each
110 set of 16 circular filter pieces was analyzed on a pyrolysis probe (CDS 6200, CDS Analytical,
111 LLC, Oxford, PA, USA) with the same pyrolysis and GC/MS methods from above. The mass
112 of PS NPPs in the 16 pieces of filter circles was estimated to be 0.57 μg, which corresponding
113 to 9.45 μg of PS collected on the filter and 61.36 μg/m³ in the air sample measured by
114 Pyrolysis-GC/MS. The mass concentration during the sampling time quantified by the AMS
115 was 60.78 μg/m³. The difference of PS concentration quantified by the AMS and the Pyrolysis-
116 GC/MS is shown to be 2%, further validating an accurate quantification of PS NPPs with the
117 AMS.

118 **6. The second ambient sampling**

119 The time series data for organic aerosols during the *ambient2* sampling period is shown
120 in Figure S14. Applying similar analysis procedures to *ambient1*, the best ME-2 solution for
121 *ambient2* sampling period has 3 factors, and they are identified as constrained PS factor, more-
122 oxidized OOA (MO-OOA), and hydrocarbon like organic aerosols (HOA). Again, the MO-
123 OOA factor has a higher f_{44} and O:C, and the HOA factor contains ions that have been
124 identified as markers of fresh fossil fuel combustion, including $C_nH_{2n-1}^+$ and $C_nH_{2n+1}^+$.^{14, 15}

125 **7. ME-2 analysis of synthetic data matrix**

126 To further validate the ability of ME-2 to quantify the concentration of ambient PS
127 NPPs, the organic data matrix was modified before reapplying the ME-2 analysis. First, all the
128 ME-2 derived factors were summed up except the extracted PS factor. The ME-2 analysis with
129 the synthetic organic data matrix as input did not extract any meaningful PS factor, as the 10-
130 minute moving average concentration has always been lower than the detection limit (12 ng/m³
131 for 10 minutes). Second, the PS section in the original organic matrix was substituted with a
132 fraction of the PS factor extracted by the original ME-2 analysis. 50%, 10%, 5%, and 2% of
133 the original PS factor were added to the organic matrix generated by summing all factors except
134 the original PS factor, and the ME-2 analysis was reapplied. The averaged PS concentration
135 derived from the ME-2 analysis with the synthetic data matrix during the intentional injection
136 period was 51%, 9%, 4%, and 2% of the original PS factor derived with the actual ambient
137 data matrix with their absolute mass loading in the table S1. The additional ME-2 analysis
138 successfully extracted a PS factor of around 16 ng/m³, which is lower than the averaged
139 concentration derived by the ME-2 for ambient sampling (30 ± 20 ng/m³). Such results further
140 validate the quantification of ambient PS NPPs with AMS and ME-2 analysis.

141 **8. The concentration of airborne microplastic particles from literatures**

142 In previous studies, the concentration of atmospheric microplastic particles was
143 reported in various units across different geographical areas. Herein, this section illustrates the
144 conversion of the previously reported concentrations into ng/m³, which is the unit of
145 measurement utilized in the present study, to provide a uniform basis for comparison. The
146 majority of prior research quantified the microplastics in terms of number concentration, i.e.
147 count per cubic meter.^{16,17,18,19} By assuming a sphere for the collected microplastics, with the
148 density and the midpoint of the reported size, the mass concentration of microplastics are
149 derived from the number concentration. Another study reported the concentration of

150 microplastics as a percentage of the total particles collected,²⁰ and considering our sampling
151 site, a total organic concentration of $6 \mu\text{g}/\text{m}^3$, which is the averaged concentration measured
152 during the sampling, is adopted for the derivation in the present study.

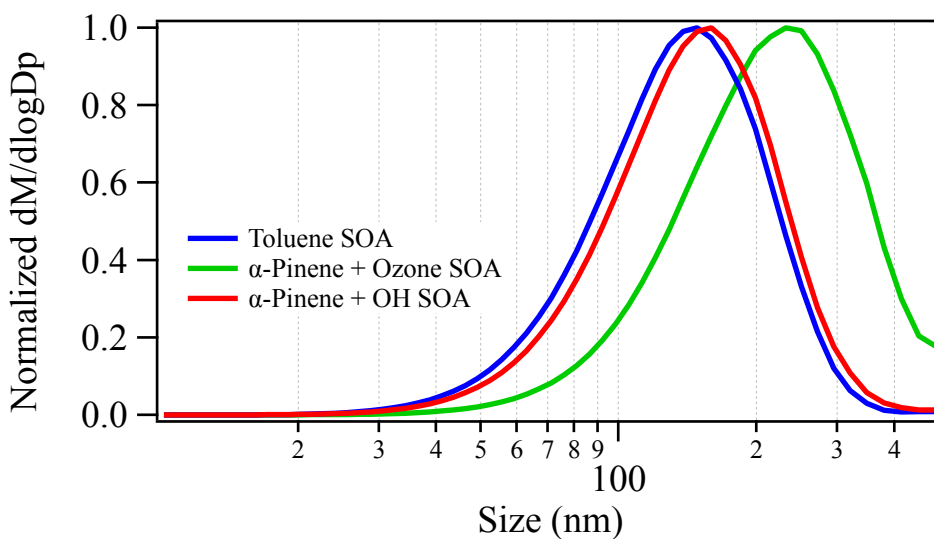
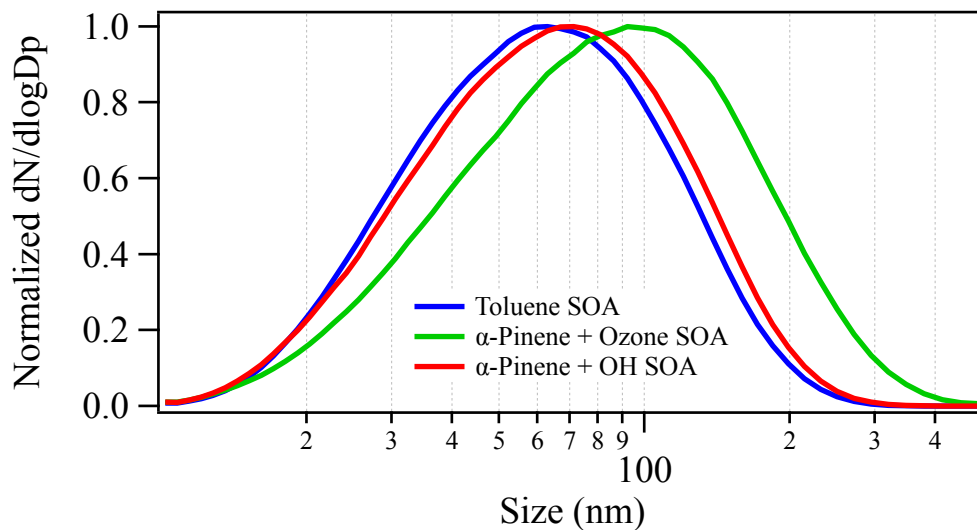
153 **9. Concentration of PS nanoplastic particles derived from the tracer ions**

154 As described in section 3.4, to further validate the feasibility of using tracer ions to
155 derive the abundance of PS nanoplastic particles, the concentrations from ME-2 and tracer ions
156 are assessed. The concentration is calculated based on the normalized intensity of the tracer
157 ion among the mass spectrum of the pure PS particle standard with the Eq. S4 below:

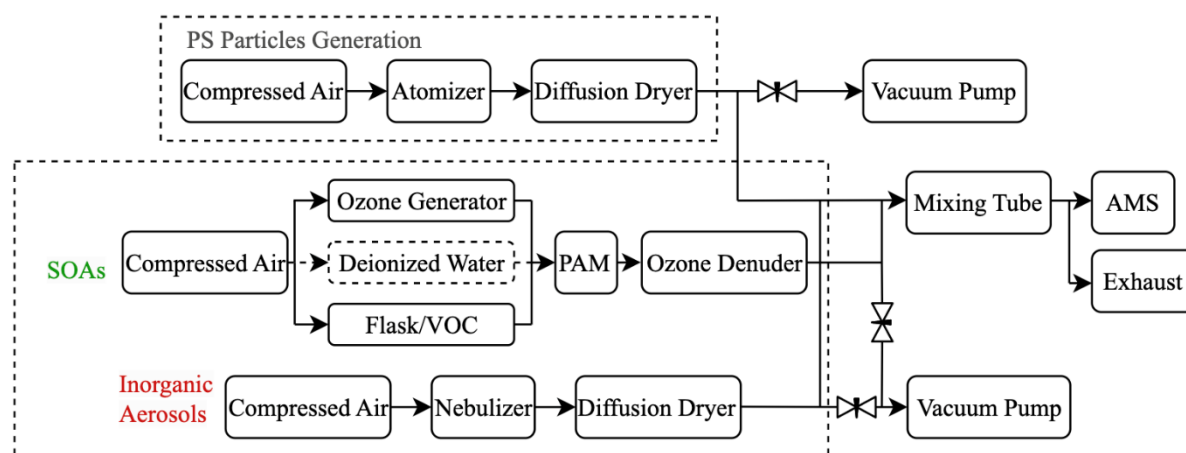
$$158 \quad [\text{PS}] = [\text{C}_8\text{H}_8^+]/0.06 \quad (\text{S4})$$

159 where [PS] is the concentration of PS nanoplastic particles in $\mu\text{g}/\text{m}^3$, $[\text{C}_8\text{H}_8^+]$ is the
160 concentration of C_8H_8^+ in $\mu\text{g}/\text{m}^3$, and 0.06 is the normalized intensity of C_8H_8^+ for the mass
161 spectrum of PS NPP standard.

162



167 **Figure S1:** The normalized number and mass distribution of self-nucleated SOAs generated
 168 through PAM.



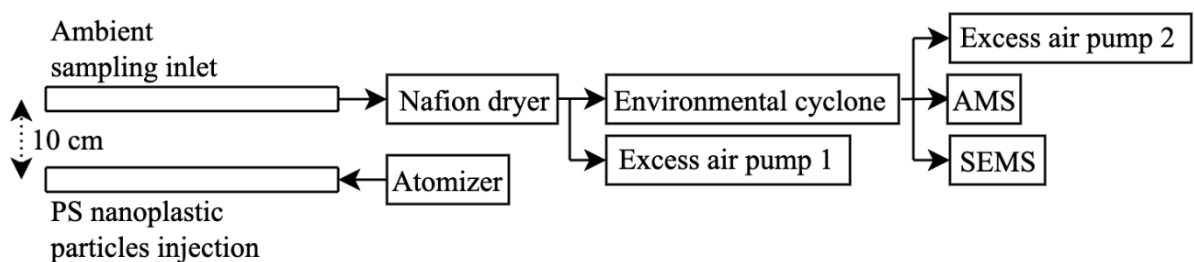
170 **Figure S2.** Experiments setup for the mixing of PS particles and SOAs/inorganic aerosols.

171 The top part of the setup demonstrated the PS particles generation, and the bottom part shows

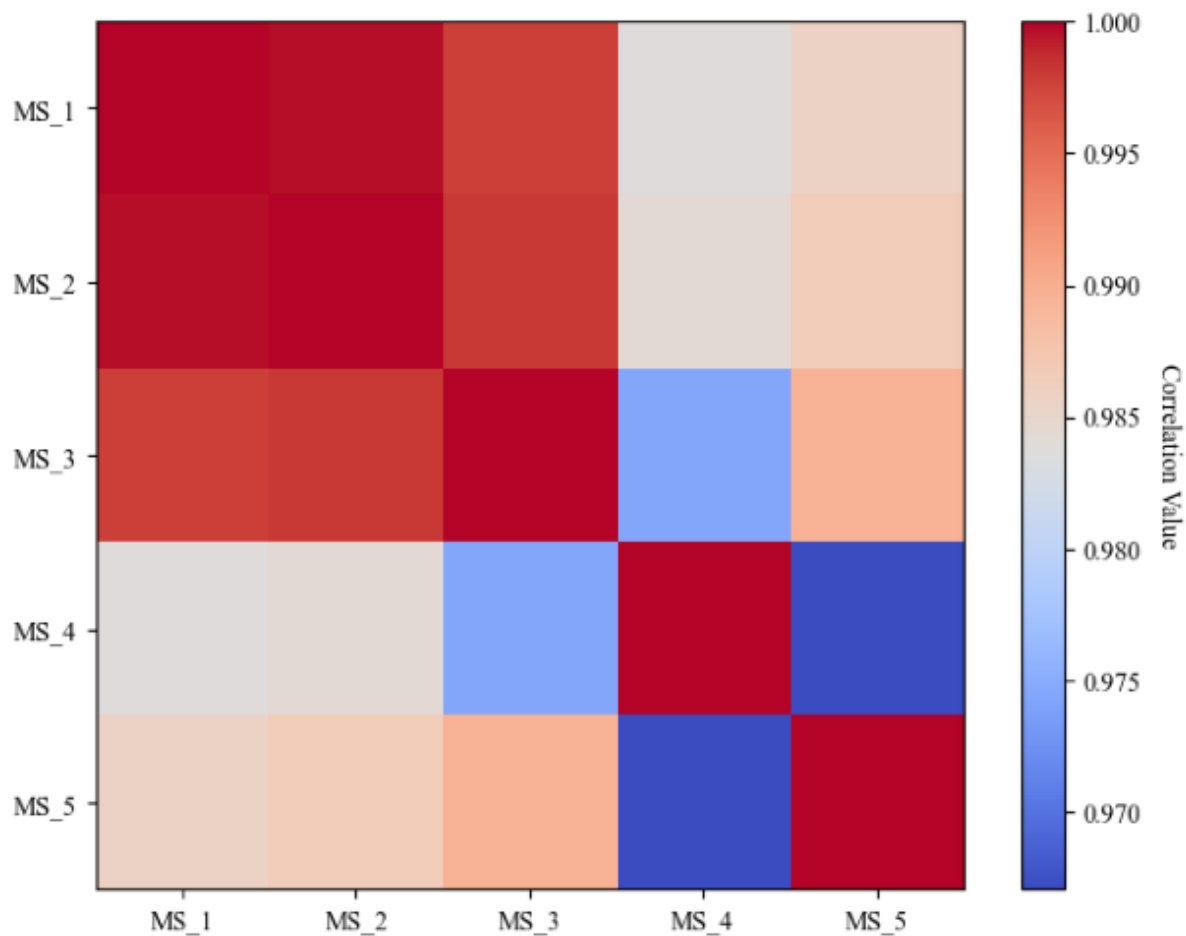
172 the SOAs and inorganic aerosols generation. For the SOAs generation, deionized water

173 line is only used during the OH radical oxidation pathway, whereas this line is replaced by

174 pure air during the ozonolysis pathway.



175 **Figure S3.** Schematic of ambient sampling setup. The excess air pump 1 drew 5 LPM of air
 176 to reduce wall loss of the submicron particles. The excess air pump 2 drew 1.6 LPM of air, to
 177 ensure the total flow passing through the environmental cyclone to be 2 liter per minute with
 178 a size cut of 1 μm .

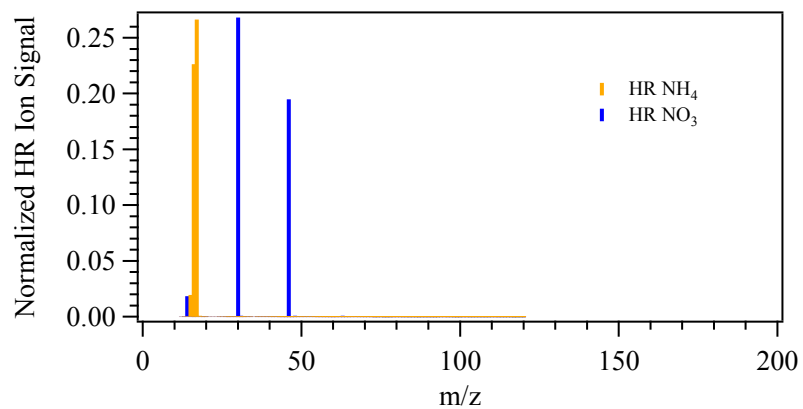


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180 **Figure S4.** The Pearson's correlation heatmap of the five mass spectra of PS NPPs collected

181 during the five-point calibration (MS_1 to MS_5).

182

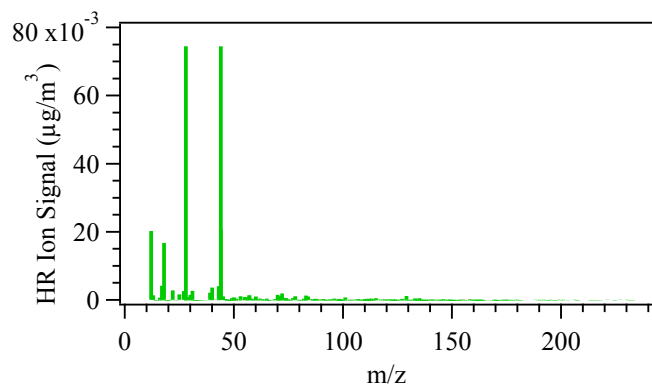


183

184 **Figure S5.** The mass spectrum of ammonium nitrate collected by the HR-ToF-AMS during

185 the calibration procedures.

186

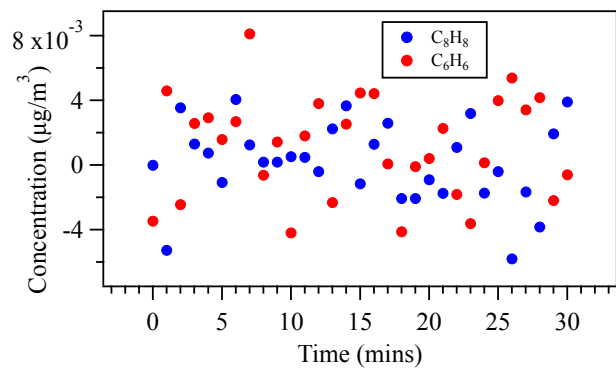


187

188 **Figure S6.** The mass spectrum collected from the HR-ToF-AMS when there was a filter

189 installed at the inlet.

190

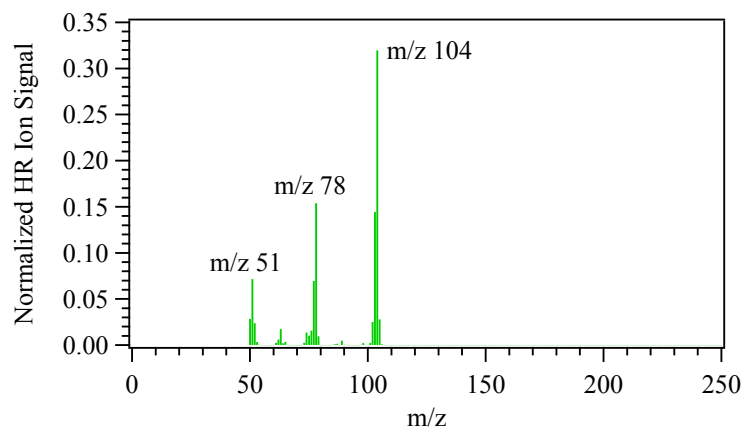


191

192 **Figure S7.** The derived concentrations of the tracer ions of PS NPPs when a filter was

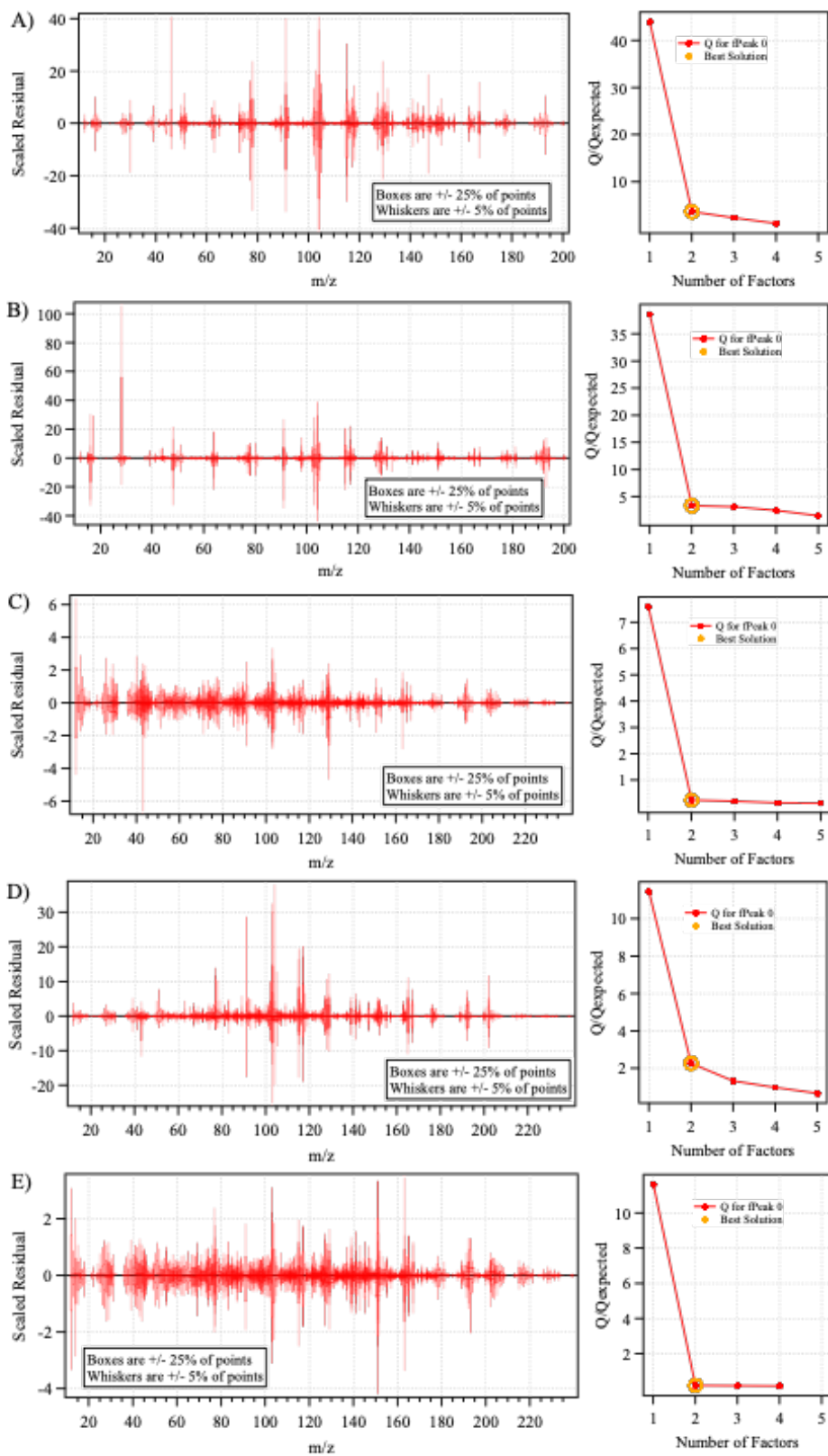
193 installed at the inlet of the HR-ToF-AMS.

194



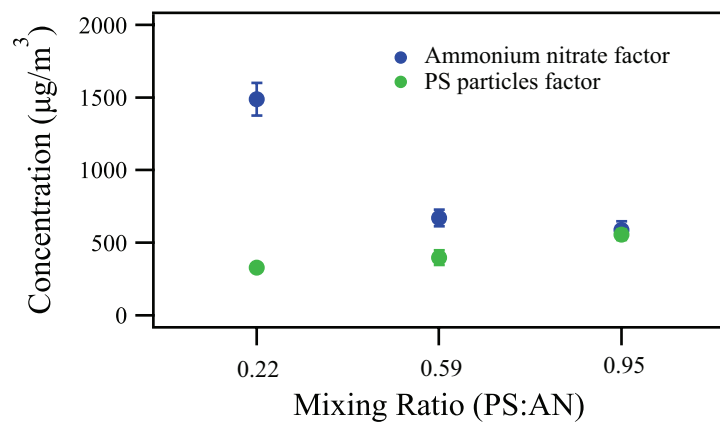
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196 **Figure S8.** Pyrolysis-GC/MS mass spectrum for pure PS NPPs.

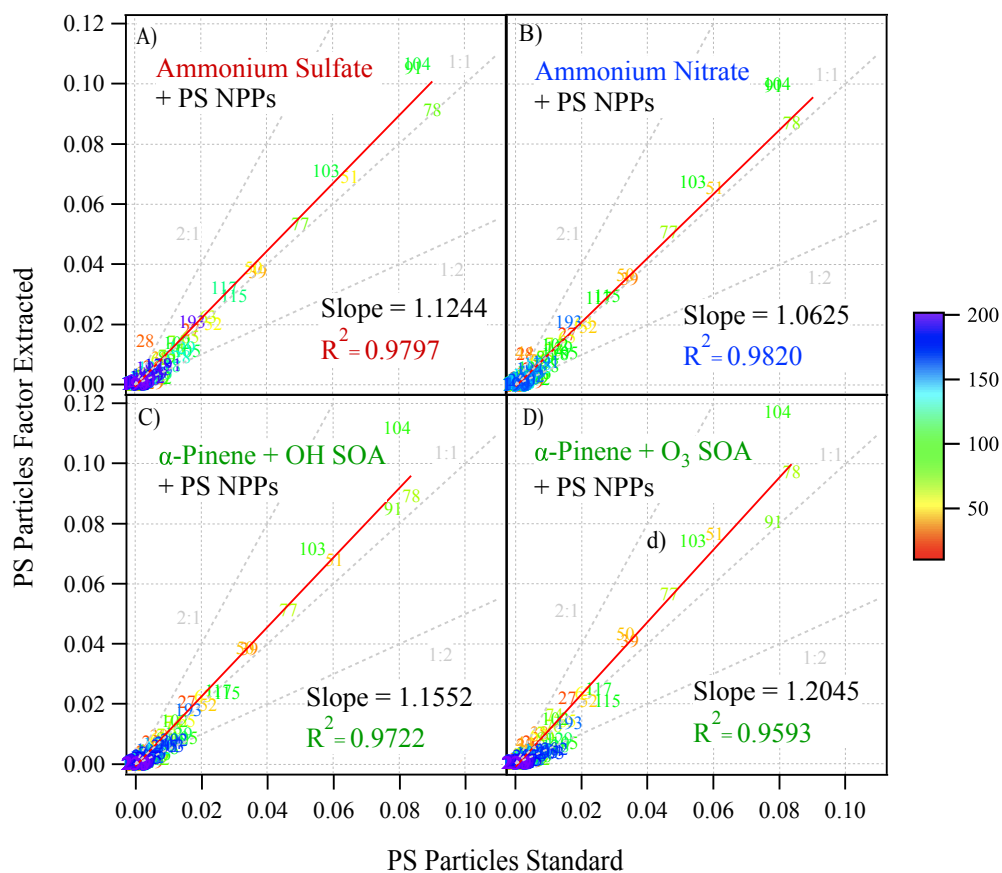


197 **Figure S9.** The scaled residual for the HR ions with two factors (right) and Q/Q_{exp} for the
 198 solution with different number of factors for different aerosol mixtures. A) ammonium

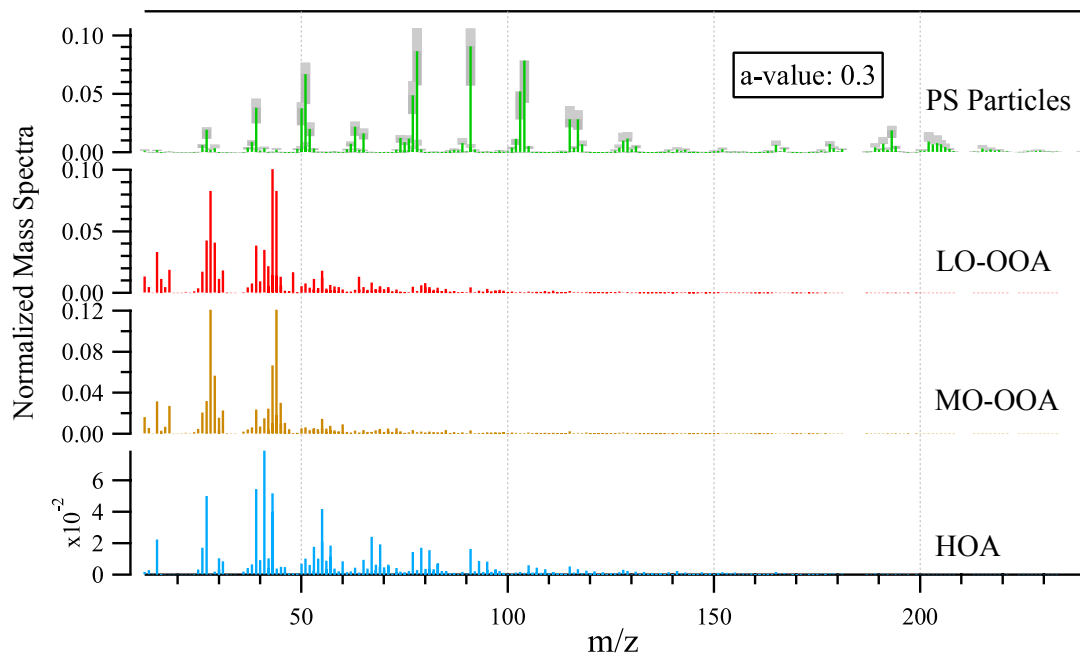
- 199 nitrate; B) ammonium sulfate; C) toluene with ozonolysis; D) α -pinene with ozonolysis and
- 200 OH oxidation; E) α -pinene with ozonolysis.



201 **Figure S10.** The time series of the two factors extracted by PMF for the mixture of PS
202 nanoplastic particles and ammonium nitrate inorganic aerosols.



203 **Figure S11.** Scatter plots of normalized HR organic ion signal of PS particles from the factor
 204 extracted from PMF (y axis) of the four binary mixture systems versus the pure PS standard
 205 (x axis). The four binary mixtures are PS particles mixed with particles comprised of A)
 206 ammonium sulfate; B) ammonium nitrate; C) α -pinene SOA from OH oxidation and
 207 ozonolysis; D) α -pinene SOA from ozonolysis alone. The color bar represents the m/z from
 208 the mass spectrum in the order from low to high values.



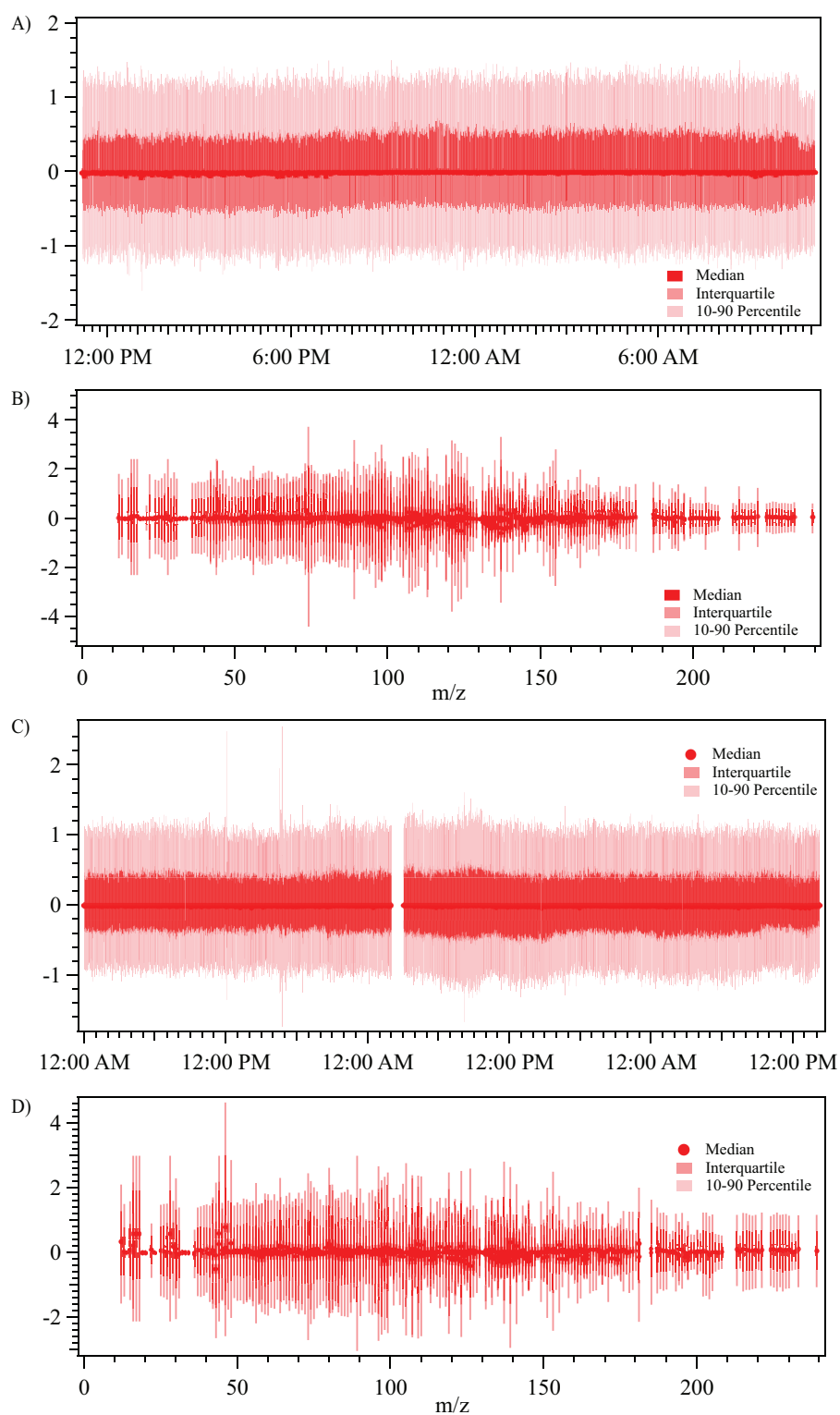
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210 **Figure S12.** The normalized mass spectra (pr nor) of each factor identified during ambient1

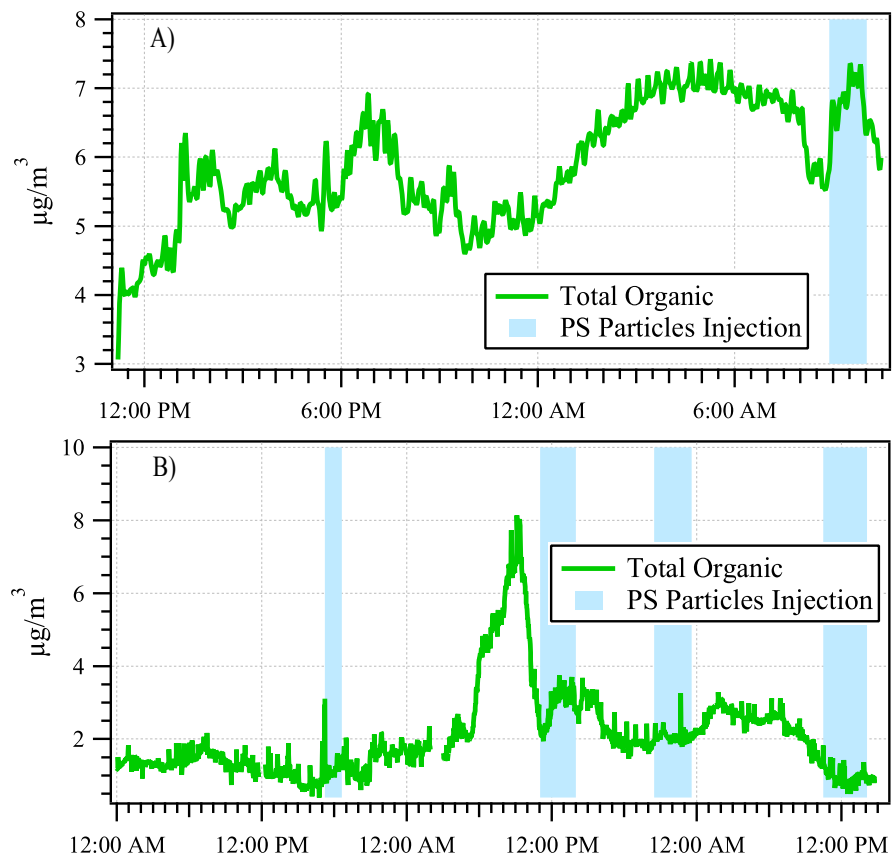
211 from the ME-2 analysis. The grey shaded areas denote the uncertainty of the mass spectra

212 with an a -value = 0.3.

213



214 **Figure S13.** The scaled residual for ambient samplings among the sampling time (A, C) and
 215 mass spectrum (B, D) to demonstrate optimum solution is achieved with minimum residual.
 216 While A and B are for *ambient1* and C and D are for *ambient2*.



217 **Figure S14.** The time series of total organic concentration during *ambient1* (A) and *ambient2*
 218 (B) sampling.

219 **Table S1.** The ratio of the PS factor added to the synthetic organic data matrix for ME-2
220 analysis to the total PS factor derived from the original ME-2 analysis, the ratio of the PS
221 factor derived from the corresponding ME-2 analysis to the total PS factor derived from the
222 original ME-2 analysis, and the averaged mass loading of the PS factor derived from the
223 corresponding ME-2 analysis during the intentional injection period.

Fraction of the PS factor added	Fraction of the PS factor derived	Averaged mass loading (ng/m ³)
100%	NA	654
50%	51%	333
10%	9%	61.5
5%	4%	26.9
2%	2%	16.2

224

225

226 **Table S2.** The averaged mass concentration and standard deviation of PS NPPs during
227 specific period and their corresponding p value from t-test. The 4-6 pm indicate the
228 background period, and the rest are the color-coded period in Figure 4A.

229

Time	4-6 pm	7-9 pm	9-11 pm	2-5 am	5-7 am
Mean Conc. ($\mu\text{g}/\text{m}^3$)	1.55×10^{-2}	1.78×10^{-2}	4.08×10^{-2}	4.05×10^{-2}	4.17×10^{-2}
SD	4.79×10^{-3}	7.27×10^{-3}	8.31×10^{-3}	7.80×10^{-3}	5.96×10^{-3}
p value	NA	4.44×10^{-3}	1.54×10^{-76}	2.34×10^{-79}	8.66×10^{-98}

230

231

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