

Comments for the Author, and replies to editor:

The concentrations are expressed in $\mu\text{g}/\text{m}^3$; however, it is unclear whether the air volumes used were volumes at ambient temperature and pressure or instead at standardized conditions; this should be indicated.

We have included the sentence at lines 179-180 "Time-integrated flow rates at ambient air pressure and temperature are used to define the sampled volume for aerosol concentrations reported in $\mu\text{g m}^{-3}$ ".

Lines 5-14: There are problems with the affiliation of some of the authors. For example, Kebin He is from China and not from Indonesia.

Thank you. These affiliations have been corrected.

Line 33: Replace "black carbon" by "equivalent black carbon".

Done.

Line 74: Replace "et al. 1994" by "et al., 1994".

Done.

Line 84: Replace ", however" by "; however,".

Done.

Lines 126-127: Replace "black carbon" by "equivalent black carbon".

Done

Line 142: Replace "457nm, 520nm, 634nm" by "457 nm, 520 nm, 634 nm".

Done

Line 156: Replace "et al (2015)" by "et al. (2015)".

Done

Line 164: Replace "burning," by "burning".

Done

Line 179: Replace "black carbon" by "equivalent black carbon".

Done

Line 189: First, as appears from Table 1, several more trace metals are measured than Zn, Mg, Fe, and Al; all measured trace metals should be listed here or the text should at least be modified. Secondly, Zn cannot really be called a crustal element. According to Mason (Principles of Geochemistry, 3rd ed., Wiley, 1966) the concentration of Zn in average crustal rock is only 70 ppm; there are more than 20 elements with higher concentrations in average crustal rock.

Line 191 has been rephrased to “One filter half is analyzed for crustal components Mg, Fe, and Al as well as trace elements Zn, V, Ni, Cu, As, Se, Ag, Cd, Sb, Ba, Ce and Pb.”

Lines 191-193: More information on the ICP-MS analysis is needed; a literature reference should at least be provided.

This section has been expanded to lines 195-201 “The acid/filter combination is boiled at 97C for 2 hours, and the liquid extract is submitted for quantitative analysis via inductively coupled plasma mass spectrometry (ICP-MS, Thermo Scientific X-Series 2), and follows standardized methodology as in Rice et al. (2012). The ICP-MS analysis is quantified via five concentrations (25, 50, 100, 250, and 500 ug/L) of a 25-element acidified stock solution. Three reference metal ions (45Sc, 115In, and 159Tb) are also used for atomic mass calibration. All ion mass signals are measured in triplicate, and the mean signal value is used for elemental quantification”.

Line 220: Replace "associate as" by "associated as".

Done

Line 239: The statement that "Trace elemental oxides are the summation of oxides for all measured ICP trace elements" is incorrect; Mg, Fe and Al are also determined by ICP-MS, but, according to Table 1, not included in TEO.

Line 247-48 has been rephrased to “Trace elemental oxides are the summation of estimated oxide mass for trace elements as measured by ICP-MS, and make up a negligible portion of total mass (< 1%).”

Line 325: Reference is made here to Sect. 4.9; however, there is no such section.

This has been corrected to “Sect. 3.9”

Line 362: Replace "is ongoing task" by "is an ongoing task".

Done

Line 409: Replace "account for" by "accounts for".

Done

Line 415: Should it not be "dependent" instead of "independent" here?

We have attempted to rephrase this sentence to “Although RM, as defined here, is not fully independent from measured ASO_4 , correlations between these two mass fractions imply related sources”.

Line 417: Replace "potassium K" by "K".

Done

Line 456: Replace "relate to" by "relates to".

Done

Line 469: Replace "than other" by "than in other".

Done

Line 470: Replace "to larger" by "to a larger".

Done

Line 480: Replace "than most" by "than for most".

Done.

Line 480: Replace "than either" by "than in either".

Done

Line 481: Replace "than Yang" by "than in Yang".

Done.

Line 482: Replace "than Oanh" by "than in Oanh".

Done.

Line 513: Replace "black carbon" by "equivalent black carbon".

Done

Line 560: Replace "salt" by "sea salt".

Done

Line 568: Replace "for two" by "for almost two".

Done

Line 652: Should it not be "Science and Technology" instead of "Science" here?

Corrected, thank you.

Line 658: Replace "al.(2010)" by "al. (2010)".

Done

Line 667: Replace "10m" by "10 m".

Done

Line 687: Replace "orgnic" by "organic".

Done

Line 689: Replace "the value" by "with the value".

Done

Section 6.2: It is unclear at which RH the PM2.5 data of the Beta Attenuation Monitor were taken. At dry conditions (RH=35%)? If not, how were they converted to dry conditions (RH=35%)? Clarification is definitely needed.

We have included at line 719 "The BAM instrument contains a drying column with a 35% humidity set point"

Line 711: Acronyms and abbreviations, here "BAM", should be explained (written full-out) when first used; presumably, "BAM" stands for "Beta Attenuation Monitor".

This has been now done at line 718.

Line 718: Replace "panel shows" by "panel in Figure 4 shows".

Done

Lines 723-724: The sentence starting with "The agreement" is redundant; it repeats essentially what is already said in line 711.

We have now deleted this line.

Line 739: Replace "black carbon" by "equivalent black carbon".

Done

Reference list (pages 21-28): Titles of journal articles should be in lower case instead of in Title Case; this comment applies, e.g., to Begum et al. (2012), Behera et al. (2010) and Bell et al. (2007), but also to several more references.

We have modified the use of capitalization as requested.

Lines 907-909: This reference should be moved down; it should come after "USEPA, 2014".

Done

Line 1001: Replace "Sci. , 326" by "Science, 326".

Done

Line 1168, within Table 1: It is stated in column 2 that the measurement method for the CM species is "ICP-MS & IC"; however, none of the three elements listed for CM in column 3 is determined by IC, they are measured by ICP-MS only.

This had referred to an older method. It is now updated and corrected to ICP-MS only.

Line 1176: Replace "Residue Matter" by "Residual Matter".

Done

Page 30, footnote of Table 3: Replace "Residue Matter" by "Residual Matter".

Done

Page 30, footnote of Table 3: It is unclear to what the "Geometric mean of ratio" applies.

Superscripts "a" and "b" have been added to table. They had been accidentally removed.

Page 31, caption of Figure 1: The figure shows data for "Ammonium Sulfate" not for "Ammoniated Sulfate" (ASO₄). If the figure is correct, the acronym "ASO₄" cannot be used in the caption; in addition, the text in lines 289-293 would also need to be revised.

In the caption of Figure 1, "ASO₄" has been replaced with "ammonium sulfate" and "ANO₃" has been replaced with "ammonium nitrate".

We have also replaced "ASO₄" with "ammonium sulfate" at line 301, and included a sentence at line 299-300 "The κ_v value for ammonium bisulfate is similar to the κ_v value of ammonium sulfate, which is adopted here for ASO₄"

Page 33, line 2 of figure caption: Replace "residual" by "residual matter".

Done.

Page 35, within Table A1: Replace "100 nm particle" by "100 nm particles".

Done

Variation in Global Chemical Composition of PM_{2.5}: Emerging Results from SPARTAN

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Abstract

The Surface PARTiculate mAtter Network (SPARTAN) is a long-term project that includes characterization of chemical and physical attributes of aerosols from filter samples collected worldwide. This manuscript discusses the ongoing efforts of SPARTAN to define and quantify major ions and trace metals found in fine particulate matter (PM_{2.5}). Our methods infer the spatial and temporal variability of PM_{2.5} in a cost-effective manner. Gravimetrically-weighed filters represent multi-day averages of PM_{2.5}, with a collocated nephelometer sampling air continuously. SPARTAN instruments are paired with AEROSOL ROBOTIC NETWORK (AERONET) sun photometers to better understand the relationship between ground-level PM_{2.5} and columnar aerosol optical depth (AOD).

We have examined the chemical composition of PM_{2.5} at 12 globally dispersed, densely populated urban locations and a site at Mammoth Cave (US) National Park used as a background comparison. Each SPARTAN location has so far been active between the years 2013 and 2016 over 2 to 26 month periods, with an average period of 12 months per site. These sites have collectively gathered over 10 site-years of quality aerosol data. The major PM_{2.5} constituents across all sites (relative contribution \pm SD) are ammoniated sulfate (20% \pm 11%), crustal material (13.4% \pm 9.9%), equivalent black carbon (11.9% \pm 8.4%), ammonium nitrate (4.7% \pm 3.0%), sea salt (2.3% \pm 1.6%), trace element oxides (1.0% \pm 1.1%), water (7.2% \pm 3.3%) at 35% RH, and residual matter (40% \pm 24%).

Analysis of filter samples reveals that several PM_{2.5} chemical components varied by more than an order of magnitude between sites. Ammoniated sulfate ranges from 1.1 $\mu\text{g m}^{-3}$ (Buenos Aires, Argentina) to 17 $\mu\text{g m}^{-3}$ (Kanpur, India [dry season]). Ammonium nitrate ranged from 0.2 $\mu\text{g m}^{-3}$ (Mammoth Cave, in summer) to 6.8 $\mu\text{g m}^{-3}$ (Kanpur, dry season). Equivalent black carbon ranged from 0.7 $\mu\text{g m}^{-3}$ (Mammoth Cave) to over 8 $\mu\text{g m}^{-3}$ (Dhaka, Bangladesh and Kanpur, India). Comparison of SPARTAN versus coincident measurements from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network at Mammoth Cave yielded a high degree of consistency for daily PM_{2.5} ($r^2 = 0.76$, slope = 1.12), daily sulfate ($r^2 = 0.86$, slope = 1.03) and mean fractions of all major PM_{2.5} components (within 6%). Major ions generally agree well with previous studies at the same urban locations (e.g. sulfate fractions agree within 4% for eight out of 11 collocation comparisons). Enhanced anthropogenic dust fractions in large urban areas (e.g. Singapore, Kanpur, Hanoi and Dhaka) are apparent from high Zn:Al ratios.

The expected water contribution to aerosols is calculated via the hygroscopicity parameter κ_v for each filter. Mean aggregate values ranged from 0.15 (Ilorin) to 0.28 (Rehovot). The all-site parameter mean is 0.20 ± 0.04 . Chemical composition and water retention in each filter measurement allows inference of hourly PM_{2.5} at 35% relative humidity by merging with nephelometer measurements. These hourly PM_{2.5} estimates compare favorably with a beta attenuation monitor (MetOne) at the nearby US embassy in Beijing, with a coefficient of variation $r^2 = 0.67$ ($n = 3167$), compared to $r^2 = 0.62$ when κ_v was not considered. SPARTAN continues to provide an open-access database of PM_{2.5} compositional filter information and hourly mass collected from a global federation of instruments.

60 1. Introduction

61

62 Fine particulate matter with a median aerodynamic diameter less than, or equal to, $2.5 \mu\text{m}$
63 ($\text{PM}_{2.5}$), is a robust indicator of premature mortality (Chen et al., 2008; Laden et al., 2006).
64 Research on long-term exposure to ambient $\text{PM}_{2.5}$ has documented serious adverse health effects,
65 including increased mortality from chronic cardiovascular disease, respiratory disease, and lung
66 cancer (WHO, 2005). Outdoor fine particulate matter ($\text{PM}_{2.5}$) is recognized as a significant air
67 pollutant, with an Air Quality Guideline set by the WHO at $10 \mu\text{g m}^{-3}$ annual average (WHO,
68 2006). Many regions of the world far exceed these long-term recommendations (Brauer et al.,
69 2015; van Donkelaar et al., 2015), and the impact on health is substantial. The 2013 Global
70 Burden of Disease estimated that outdoor $\text{PM}_{2.5}$ caused 2.9 million deaths (3 % of all deaths) and
71 70 million years of lost healthy life on a global scale (Forouzanfar et al., 2015). Atmospheric
72 aerosol is also the most uncertain agent contributing to radiative forcing of climate change
73 (IPCC, 2013). Aerosol mass and composition also play a critical role in atmospheric visibility
74 (Malm et al., 1994). Additional observations are needed to improve the concentration estimates
75 for $\text{PM}_{2.5}$ as a global risk factor, and to better understand the chemical components and sources
76 contributing to its formation.

77

78 The chemical composition of $\text{PM}_{2.5}$ offers valuable information to identify the
79 contributions of specific sources, and to understand aerosol properties and processes that could
80 affect health, climate and atmospheric conditions. Spatial mapping of aerosol type and
81 composition using satellite observations and chemical transport modelling can help elucidate the
82 global exposure burden of fine particulate matter composition (Kahn and Gaitley, 2015;
83 Lelieveld et al., 2015; Patadia et al., 2013; Philip et al., 2014a); however, ground-level sampling
84 remains necessary to evaluate these estimates and provide quantitative detail. Furthermore, the
85 long-term health impacts of specific chemical components are not well understood (e.g. Lepeule
86 et al., 2012). The health-related impacts of specific PM composition have been reviewed
87 previously (Lippmann, 2014). Although $\text{PM}_{2.5}$ composition can be implicated in the variance
88 observed in cardiovascular health effects, there is insufficient long-term $\text{PM}_{2.5}$ characterization
89 for adequate health impact assessments of specific aerosol mixtures (e.g. Bell et al., 2007). More
90 generally, urban $\text{PM}_{2.5}$ speciation is not yet consistently characterized on a global scale.
91 Continental sampling has been conducted in North America (Hand et al., 2012) and Europe
92 (Putaud et al., 2004, 2010), however there remains a need for a global network that consistently
93 measures $\text{PM}_{2.5}$ chemical composition in densely populated regions.

94

95 No global $\text{PM}_{2.5}$ protocol exists for relative humidity (RH) filter equilibration. The U.S.
96 EPA measurements are between 30-40% RH, European measurements are below 50% RH, and
97 different protocols exist elsewhere. Ambient humidity affects the relationship of dry $\text{PM}_{2.5}$ with
98 satellite observations of aerosol optical depth. Aerosol water also influences the relationship
99 between dry $\text{PM}_{2.5}$ and aerosol scatter. A large body of literature has examined the relationship
100 of aerosol composition with hygroscopicity (e.g. IMPROVE (Hand et al., 2012; IMPROVE,
101 2015), Chemical Species Network (CSN) (Chu, 2004; USEPA, 2015), ISORROPIA (Fountoukis
102 and Nenes, 2007), and Aerosol Inorganic Model (AIM) (Wexler and Clegg, 2002)). More
103 recently Petters and Kreidenweis (2007, 2008, 2013) have developed κ -Kohler theory, which
104 assigns individual hygroscopicity parameters κ to all major components, from insoluble crustal
105 materials to sea-salt. Mixed values can then be weighted by local aerosol composition.

106
107 Ground-based observations of $PM_{2.5}$ have insufficient coverage at the global scale to
108 provide assessment of long-term human exposure. Satellite remote sensing offers a promising
109 means of providing an extended temporal record to estimate population exposure to $PM_{2.5}$ on a
110 global scale, and especially for areas with limited ground-level $PM_{2.5}$ measurements (Brauer et
111 al., 2015; van Donkelaar et al., 2015). Even in areas where monitor density is high, satellite-
112 based estimates provide additional useful information on spatial and temporal patterns in air
113 pollution (Kloog et al., 2011, 2013; Lee et al., 2012). However, there are outstanding questions
114 about the accuracy and precision with which ground-level aerosol mass concentrations can be
115 inferred from satellite remote sensing. Standardized $PM_{2.5}$ measurements, collocated with
116 ground-based measurements of aerosol optical depth, are needed to evaluate and improve $PM_{2.5}$
117 estimates from satellite remote sensing. To meet these sampling needs, the ground-based
118 network SPARTAN (Surface PARTiculate mAtter Network) is designed to evaluate and enhance
119 satellite-based estimates of $PM_{2.5}$ by measuring fine particle aerosol concentrations and
120 composition continuously over multi-year periods at sites where aerosol optical depth is also
121 measured (Holben et al., 1998; Snider et al., 2015). The network includes air filter sampling and
122 nephelometers that together provide long-term and hourly $PM_{2.5}$ estimates at low RH (35%).
123

124 We discuss the ongoing efforts of the SPARTAN project to quantify major ions and trace
125 metals found in aerosols worldwide. Section 2 describes the methodology used to infer $PM_{2.5}$
126 composition. Section 3 defines categories of aerosol types (crustal and residual material,
127 equivalent black carbon, ammonium nitrate, ammoniated sulfate, sea salt, and trace metal oxides)
128 as a function of specific chemical species. Section 4 describes the implementation of sub-
129 saturated κ -Kohler theory to estimate aerosol water content based on aerosol compositional
130 information. Section 5 compares relative aerosol composition with that reported in available
131 literature, and assesses the general consistency of our findings across all sites. Section 6
132 evaluates hourly $PM_{2.5}$ estimates (35% RH) at Beijing with a beta attenuation monitor at the US
133 Embassy.
134

135 2. Overview of Methodology

136

137 SPARTAN has been collecting $PM_{2.5}$ on PTFE filters for at least two months, across 13
138 SPARTAN sites, between 2013 and 2016, with an average period of 12 months per site. Snider
139 et al. (2015) provide an overview of the SPARTAN PM observation network, the cost-effective
140 sampling methods employed and post sampling instrumental methods of analysis. Each site
141 utilizes a combination of continuous monitoring by nephelometry and mass concentration via
142 filter-based sampling. Nephelometer scatter is averaged to hourly intervals at three wavelengths
143 (457 nm, 520 nm, 634 nm), and converted to 550 nm via a fitted Angstrom exponent. Total
144 scatter is proportional to $PM_{2.5}$ mass and volume (Chow et al., 2006). Hence we provide dry
145 (35% RH) hourly $PM_{2.5}$ estimates by combining scatter at 550 nm at ambient RH with filter mass
146 and chemical composition information used to determine water content as described below.
147

148 Briefly, filter-based measurements are collected with an AirPhoton SS4i automated air
149 sampler. Each sampler houses a removable filter cartridge that protects seven sequentially active
150 25 mm diameter filters plus a field blank. Air samples first pass through a bug screen and then a
151 greased impactor plate to remove particles larger than 10 μm in diameter. Aerosols are collected

152 in sequence on a preweighed Nuclepore filter membrane (8 μm , SPI) that removes coarse-mode
153 aerosols with diameters from 2.5 - 10 μm in diameter (PM_{10}), while fine aerosols ($\text{PM}_{2.5}$) are then
154 collected on pre-weighed PTFE filters (2 μm , SKC). For each filter, sampling is timed at regular,
155 staggered 24-hour intervals throughout a 9-day period. Sampling ends for each filter at 09:00
156 when temperatures are low, to reduce loss of semi-volatile components. As described by Snider
157 *et al.* (2015), loss rates of ammonium nitrate during passive air flow were an order of magnitude
158 less than during active air flow. Thus the sampling protocol is designed to actively sample for
159 one diurnal cycle and to avoid daytime sampling after collecting nighttime PM. Following the
160 IMPROVE protocol (Hand and Malm, 2006), filters are transported at room temperature in
161 sealed containers between measurement sites and the central SPARTAN laboratory at Dalhousie
162 University, where analysis is conducted.

163
164 Site locations are designed to sample under a variety of conditions, including biomass
165 burning (e.g. West Africa and South America), biofuel emissions (e.g. South Asia), monsoonal
166 conditions (e.g. West Africa and Southeast Asia), suspended mineral dust (e.g. West Africa and
167 the Middle East) and urban crustal material. Each SPARTAN site provides a representative
168 example of local and regional conditions in highly populated areas. Site selection prioritizes
169 under-represented globally-dispersed, population-dense regions; no SPARTAN sites yet exist in
170 Europe. The sites of Atlanta and Mammoth Cave are included for instrument inter-comparison
171 purposes with other networks.

172 2.1. Filter weighing

173 Filters (PTFE, capillary) are both pre and post-weighed in triplicate using a Sartorius Ultramicro
174 balance with 0.1 μg precision. Gravimetric weighing is performed in a cleanroom facility at $35 \pm$
175 5% RH and 20-23°C. A total of 497 quality-controlled filters have been weighed across all
176 SPARTAN sites. The median collected material on sampled filters, as well as the lower and
177 upper quintiles (in parentheses), are 72 (42, 131) μg for Teflon and 90 (44, 154) μg for
178 Nuclepore. The combined uncertainty ($\pm 2\sigma$) of quality-assured single filter PM mass
179 measurements is $\pm 4.0 \mu\text{g}$. Time-integrated flow rates at ambient air pressure and temperature are
180 used to define the sampled volume for aerosol concentrations reported in $\mu\text{g}/\text{m}^3$. These filters are
181 subsequently analyzed for water-soluble ions, trace metals, and surface reflectance to obtain
182 equivalent black carbon.

183 2.2. Equivalent Black Carbon (EBC)

184 We define the equivalent black carbon (EBC) as the black carbon content of PTFE filters derived
185 via surface reflectance R using the Diffusion Systems Smoke Stain Reflectometer EEL 43M
186 (Quincey *et al.*, 2009) as further discussed in Sect. 4.6. We use the term equivalent black carbon
187 following the recommendation of Petzold *et al.* (2013) for data derived from optical absorption
188 methods.

189 2.3. Trace metals

190 To maximize the information extracted from the filters, each one is cut in half with a ceramic
191 blade following approaches similar to Zhang *et al.* (2013) and Gibson *et al.* (2009). One filter
192 half is analyzed for crustal components Mg, Fe, and Al as well as trace elements Zn, V, Ni, Cu,
193 As, Se, Ag, Cd, Sb, Ba, Ce and Pb. We first digest this filter half by adding it to 3.0 mL of 7%

194 trace metal grade nitric acid, similar to Fang et al. (2015). The acid/filter combination is boiled at
195 97C for 2 hours, and the liquid extract is submitted for quantitative analysis via inductively
196 coupled plasma mass spectrometry (ICP-MS, Thermo Scientific X-Series 2), and follows
197 standardized methodology as in Rice et al. (2012). The ICP-MS analysis is quantified via five
198 concentrations (25, 50, 100, 250, and 500 $\mu\text{g/L}$) of a 25-element acidified stock solution. Three
199 reference metal ions (^{45}Sc , ^{115}In , and ^{159}Tb) are also used for atomic mass calibration. All ion
200 mass signals are measured in triplicate, and the mean signal value is used for elemental
201 quantification.

202 2.4. Water soluble ions

203 Water-soluble ions NO_3^- , SO_4^{2-} , NH_4^+ , K^+ , Na^+ are detected using the second filter half. The filter
204 is spiked with 120 μL of isopropyl alcohol and immersed in 2.9 mL of 18 M Ω Milli-Q water.
205 Filters and liquid extracts are sonicated together for 25 min before being passed through a 0.45
206 μm membrane filter to remove larger matrix components. Extractions are analyzed by ion
207 chromatography (IC) via a Thermo Dionex ICS-1100 instrument (anions) and a Thermo Dionex
208 ICS-1000 (cations) instrument (Gibson et al., 2013a, 2013b).
209

210 3. $\text{PM}_{2.5}$ aerosol composition

211
212 Section 2 defined the methodology of basic physical and chemical properties obtained in
213 SPARTAN filters. Section 3 describes the chemical definitions used to infer each chemical
214 component as discussed in turn below. Table 1 contains a summary of equations and
215 accompanying references used to quantify SPARTAN $\text{PM}_{2.5}$ chemical composition.

216 3.1. Sea Salt (SS)

217 We take 10% of $[\text{Al}]$ to be associated with Na and remove this crustal sodium component
218 (Remoundaki et al., 2013). Sea salt is then represented as $2.54[\text{Na}^+]_{\text{ss}}$ to account for the
219 associated $[\text{Cl}^-]$ (Malm et al., 1994).

220 3.2. Ammonium nitrate (ANO_3)

221 We treat all nitrate as neutralized by ammonium as NH_4NO_3 . The corresponding mass of ANO_3
222 is a 1:1 molar ratio of $\text{NH}_4:\text{NO}_3$, or $1.29[\text{NO}_3^-]$ based on molecular weight.

223 3.3. Sodium sulfate (Na_2SO_4)

224 Sodium sulfate is treated as a fraction of measured sodium, $0.18[\text{Na}^+]_{\text{ss}}$ (Henning et al., 2003);
225 however, it contributes negligibly to total aerosol mass ($< 0.1\%$) at all sites.

226 3.4. Ammoniated sulfate (ASO_4)

227 Ammonium not associated with nitrate, and sulfate not associated with sodium, are assumed to
228 be associated as a mixture of NH_4HSO_4 and $(\text{NH}_4)_2\text{SO}_4$.

229 3.5. Crustal material (CM)

230 Crustal material consists of re-suspended road dust, desert dust, soil, and sand. Following the
231 elemental composition of natural desert dusts by Wang (2015), we generalize that natural CM is

232 approximately $10 \times [\text{Al} + \text{Fe} + \text{Mg}]$. Aluminum, iron, and magnesium are chosen due to their
 233 collectively consistent composition in natural mineral dust and frequency above detection limit
 234 ($> 95\%$). Silicon is not available. Titanium was found not to contribute significantly ($< 1\%$) to
 235 CM mass.

236 3.6. Equivalent Black Carbon (EBC)

237 The amount of EBC carbon (μg) is logarithmically related to concentration, as determined by
 238 relative surface reflectance R/R_0 . For a given exposed filter area, absorption cross-section and
 239 light path, reflectance is related to concentration via

$$[\text{EBC}] = \frac{-A}{qv} \ln\left(\frac{R}{R_0}\right) \quad \text{Eq. 1}$$

240 where v is volume of air (0.9 to 5.8 m^3), A is the filter surface area (3.1 cm^2), and q is the product
 241 of the effective reflectivity path p and mass-specific absorption cross section σ_{SSR} ($\text{cm}^2/\mu\text{g}$). The
 242 absorption coefficient σ_{SSR} used here is $0.06 \text{ cm}^2/\mu\text{g}$ based on prior literature (Barnard et al.,
 243 2008; Bond and Bergstrom, 2006), adjusted to the 620 nm detection peak of the SSR. The
 244 effective light path p here is taken to be 1.5 for our thick PTFE filters (e.g. Taha et al., 2007). We
 245 treat water uptake by EBC as negligible.

246 3.7. Trace elemental oxides (TEO)

247 Trace elemental oxides are the summation of estimated oxide mass for trace elements as
 248 measured by ICP-MS, and make up a negligible portion of total mass ($< 1\%$). We include these
 249 concentrations for completeness. Water uptake by TEO is treated as negligible.

250 3.8. Particle-bound water (PBW) associated with inorganics

251 We estimate the water-mass uptake for the inorganic chemical components of sea salt (SS),
 252 ammonium nitrate (ANO_3) and ammoniated sulfate (ASO_4). The mass of particle-bound water
 253 (PBW) associated with chemical component X is

$$\text{PBW}_X = [X] \kappa_{m,X} \frac{\text{RH}}{100 - \text{RH}} \quad \text{Eq. 2}$$

254 The total mass of inorganic (IN) PBW is then $\text{PBW}_{\text{IN}} = \sum_X \text{PBW}_X$.

255 3.9. Residual matter (RM)

256 Residual matter, which is treated as mainly organics, is estimated by subtracting dry inorganic
 257 mass (IN) and its associated water (referenced to our weighing conditions of $35 \pm 5\%$ RH) from
 258 total $\text{PM}_{2.5}$ mass:

$$\text{RM}_{35\%} = \text{PM}_{2.5,35\%} - [\text{IN}] - [\text{PBW}_{\text{IN}}] \quad \text{Eq. 3}$$

259 Negative $\text{RM}_{35\%}$ values are retained if reconstructed inorganic mass at 35% RH exceeds total
 260 $\text{PM}_{2.5}$ by less than 10% , otherwise values are flagged and excluded from the mass average.
 261 Negative values occur, on average, 2% of the time. Water-free RM (0% RH) is estimated by
 262 subtracting organic-associated PBW using an estimated hygroscopic parameter $\kappa_{m,\text{RM}} = 0.1$ as
 263 discussed in section 4.
 264

265 4. Aerosol hygroscopicity

266

267 We apply the single-parameter measure of aerosol hygroscopicity (κ) developed by Petters
268 and Kreidenweis (2007, 2008, 2013) to represent the contribution of water uptake by individual
269 components. The κ parameter is defined from 0 (insoluble materials) to greater than 1 for sea
270 salt. Although initially developed for supersaturated CCN conditions, hygroscopic parameters κ
271 have been more recently used in sub-saturated conditions (Chang et al., 2010; Dusek et al., 2011;
272 Giordano et al., 2013; Hersey et al., 2013). For particle diameters that dominate the mass fraction
273 of PM_{2.5} (larger than 50 nm), the difference in κ between CCN and sub-saturated aerosols is
274 small (Dusek et al., 2011). The water retention of internal mixtures of aerosol components is
275 often predicted within experimental error (Kreidenweis et al., 2008). Aged, polarized organic
276 material, which is a major component of PM_{2.5}, shows comparable growth factors both in super-
277 and sub-saturated regions (Rickards et al., 2013).

278

279 The volume hygroscopicity parameter κ_v is defined as a function of particle volume V and
280 water activity a_w

$$\frac{1}{a_w} = 1 + \kappa_v \frac{V_d}{V_w} \quad \text{Eq. 4}$$

281 where V_d and V_w are the dry particulate matter and water volumes, respectively. To a first-order
282 approximation $a_w = \text{RH}/100$. Aerosol volume growth is related via κ and RH by defining $f_v(\text{RH})$
283 as the humidity-dependent ratio of wet and dry aerosol volume:

$$f_v(\text{RH}) \equiv \frac{V_{tot}}{V_d} = \frac{V_d + V_w}{V_d} = a + \kappa_v \frac{\text{RH}}{100 - \text{RH}} \quad \text{Eq. 5}$$

284 Combining the previous equations and relating to a diameter D growth factor ($GF \equiv D/D_d$) yields

$$GF = \left(a + \kappa_v \frac{\text{RH}}{100 - \text{RH}} \right)^{1/3} \quad \text{Eq. 6}$$

285 where $a = 1$, except for sea salt as discussed in Sect. 3.1. Reliable estimates of κ_v are available
286 for individual components (*c.f.* Table 2).

287

288 The next sections outline how we apply κ to represent mass and volume hygroscopic growth
289 in major hygroscopic aerosol components. Four components directly contribute to water uptake:
290 ammonium nitrate (ANO₃), ammoniated sulfate (ASO₄), sea salt (SS), and organics. We treat
291 black carbon (EBC), crustal material (CM), and trace oxides (TEO) as non-hygroscopic. We
292 evaluated inorganic component growth curves using the AIM model (Wexler and Clegg, 2002)
293 for RH = 10 – 90% except for sea salt, which included RH = 0%. Hygroscopic parameters were
294 matched to modeled fits. Aerosols are treated as internally mixed, without deliquescence or
295 efflorescence points, as discussed further below.

296 4.1. Inorganic behavior

297 Figure 1 shows the hygroscopic growth for inorganics. The κ_v value of 0.51 for
298 ammonium sulfate best matches the AIM model over RH = 10-90% and is similar to the GF -

299 derived $\kappa_v = 0.53$ estimated by Petters and Kreidenweis (2007). The κ_v value for ammonium
300 bisulfate is similar to the κ_v value of ammonium sulfate, which is adopted here for ASO_4 . Our
301 AIM-derived ammonium nitrate growth curve is smaller than ammonium sulfate, at $\kappa_v = 0.41$.
302 Empirically both ammonium compounds share the same $GF = 1.6$ at $\text{RH} = 85\%$ (Sorooshian et
303 al., 2008), however ANO_3 is less hygroscopic at lower RH.

304
305 Sea salt accounts for a small fraction of aerosol mass over land, however its hydrophilic
306 nature makes it significant for water retention. A 1:1 volume ratio with water as RH approaches
307 0% (Kreidenweis et al., 2008) yields $a = 2$ (Eq. 2 and 3). A hygroscopic constant $\kappa_v = 1.5$ then
308 best fits AIM from the deliquescence point up to 90% RH.

309
310 We follow the widely used convention (e.g. Pitchford et al. (2007)) that $\text{PM}_{2.5}$ under
311 variable sub-saturated RH does not exhibit deliquescent phase transitions. There is compelling
312 evidence to adopt smooth hygroscopic growth curves. Various experiments show sub-
313 micrometer, internally mixed aerosols will not deliquesce as readily as pure compounds. For
314 example, Badger et al. (2006) observed ASO_4 aerosol deliquescence is clearly inhibited by the
315 presence of humic acids. A smooth growth curve has been observed over the range $\text{RH} = 10 -$
316 85% for ambient aerosols at Jungfraujoch (Swietlicki et al., 2008). Analysis of submicron aerosol
317 mixtures consisting of SS, ASO_4 , ANO_3 , and levoglucosan also showed no apparent phase
318 transition (Svenningsson et al., 2006).

319 4.2. Organic matter behavior

320 Identifying a representative organic hygroscopic parameter is challenging, as many volume
321 growth curves are available based on a variety of laboratory experiments and field campaigns.
322 Organic composition varies by site, and by season. The Appendix table A1 contains a collection
323 of hygroscopic parameters from the literature. Values for $\kappa_{v,OM}$ range from 0 to 0.2. We choose a
324 single $\kappa_{v,OM}$ value based on the oxygen/carbon ratio (O:C), which is a function of oxidation,
325 hence age of the organics. Generally O:C ratios are between 0.2 – 0.8 in urban environments
326 (Rickards et al., 2013). We select an O:C ratio of 0.5 to represent the populated nature of
327 SPARTAN sites (e.g. Robinson et al., 2013). This corresponds to an organic parameter of
328 $\kappa_{v,OM} = 0.1$ for a variety of organic mixtures (Jimenez et al., 2009).

329 4.3. Aerosol water in multi-component systems

330 Mass-based hygroscopic water uptake κ_m is more convenient than κ_v to estimate water
331 retention in gravimetric analysis. The parameters κ_v and κ_m are related by water-normalized
332 density, $\kappa_{m,X} = \kappa_{v,X}/\rho_X$. Table 2 contains κ_v values identified for major aerosol chemical
333 components and densities. For a multi-component system we estimate aerosol water mass using a
334 mass-weighted combination of κ_m values:

$$\kappa_{m,tot} = \frac{1}{M} \sum_X m_X \kappa_{m,X} \quad \text{Eq. 7}$$

335 Mass calculations are used to determine residual aerosol mass as described in Sect. 3.9.
336 Estimates of total water uptake by volume are applied to aerosol light scatter in Sect. 5. The
337 volume parameter $\kappa_{v,tot}$ is similarly determined by a linear combination of volume-weighted
338 components X (e.g. Bezantakos et al., 2013):

$$\kappa_{v,tot} = \frac{1}{V} \sum_X v_X \kappa_{v,X} \quad \text{Eq. 8}$$

339 The hygroscopic growth of ASO_4 and organic mixtures are treated as linear combinations of pure
 340 compounds (Robinson et al., 2013). Errors in aerosol water uptake are less significant in mixtures
 341 than for individual species due to dilution effects (Kreidenweis et al., 2008). For ambient aerosols,
 342 empirically measured $\kappa_{v,tot}$ usually lies between 0.14 and 0.39 (Carrico et al., 2010).
 343

344 4.4. Sources of Uncertainty

345 Uncertainty in atmospheric $\text{PM}_{2.5}$ concentrations can be separated into air volume and $\text{PM}_{2.5}$
 346 mass. We estimated total flow volume variance to be $\pm 10\%$, while 2σ pre and post gravimetric
 347 mass measurement varied by a combined $\pm 4 \mu\text{g}$. Characterization of hourly $\text{PM}_{2.5}$ uncertainties
 348 can be found in Appendix A2.
 349

350 Of concern is the loss of semivolatiles after sampling. In the laboratory semivolatile loss
 351 is inhibited by storing filters in closed containers. As discussed in Section 2, the sampling
 352 protocol is designed to minimize semi-volatile loss. We tested the retention of semivolatile
 353 material in the field by examining the trend in $\text{PM}_{2.5}$ and ANO_3 mass from the first filter sampled
 354 (54 day residence time in instrument) through the last filter sampled (negligible residence time in
 355 instrument). Statistically insignificant trends were found for both $\text{PM}_{2.5}$ ($-0.09 \pm 0.46 \mu\text{g m}^{-3}$
 356 $^3/\text{position}$) and ANO_3 ($0.06 \pm 0.15 \mu\text{g m}^{-3}/\text{position}$) providing confidence in retention of
 357 semivolatiles on filters in the cartridge.
 358

359 Other uncertainties include absolute equivalent black carbon mass due to the reflectivity
 360 path p ($\pm 30\%$) and absorption cross section σ ($\pm 30\%$), which combine to in quadrature $\pm 42\%$.
 361 Trace metal recovery yields were tested using a sequential second digestion with 20% nitric acid.
 362 Each acid-digested element was quantified by five dilutions of a 25 element standard (25 – 500
 363 ppb) plus three internal calibration metals (Sc, In, Tb). The elemental comparison of crustal
 364 materials varies regionally (Wang, 2015), which contributes to CM uncertainty of $\pm 30\%$ based
 365 on Al, Fe and Mg composition. Recovery of individual water-soluble elements was determined
 366 through 5-point anion and cation standards curves each with $r^2 > 98\%$ and $<10\%$ mass
 367 uncertainty for most elements at environmentally-relevant concentrations, including sulfate,
 368 nitrate, and ammonium. Based on lab filter spike tests, water-soluble ion extractions show $> 95\%$
 369 extraction efficiency. Uncertainties of water-soluble ion yields are generally $\pm 5\%$, except when
 370 close to limit of detection (approximately $0.1 \mu\text{g m}^{-3}$, depending on filter sampling duration).
 371 Errors in the component values affect our estimate of κ_v , which will affect the inferred aerosol
 372 water. Network evaluation **is an ongoing task** that will continue over time.
 373

374 5. Mass speciation results

375 5.1. Overview of $\text{PM}_{2.5}$ mass speciation

376 Gravimetrically-weighed $\text{PM}_{2.5}$ concentrations within the period June 2013 to February 2016
 377 span an order of magnitude, from under $10 \mu\text{g m}^{-3}$ (e.g. Atlanta) to almost $100 \mu\text{g m}^{-3}$ (Kanpur).

378 Sites include a variety of geographic regions including partial desert (Ilorin, Rehovot, Kanpur),
379 coastline (Buenos Aires, Singapore), and developing megacities (Dhaka). Table 3 and Figure 2
380 contain the resulting PM_{2.5} mass, composition, and location of each SPARTAN site. The mean
381 SPARTAN composition over all sampling sites in descending concentration is 40% RM
382 (primarily organic), 20% ASO₄, 13% CM, 12% EBC, 4.7% ANO₃, 2.3% SS and 1.0% TEO.
383

384 There is significant variation of relative and absolute speciation from these long-term
385 averages. ASO₄ concentrations range from 1 µg m⁻³ (Buenos Aires, summer) to 17 µg m⁻³
386 (Kanpur, dry season). The fraction of sulfate in PM_{2.5} exhibits much weaker spatial variation (10-
387 30%) as increases in ASO₄ coincide with increases in total PM_{2.5}. Hence locations with enhanced
388 sulfate tend to have enhancements in other aerosol components.
389

390 ANO₃ concentrations exhibit a larger spatial heterogeneity than sulfate. Absolute values
391 range over 30-fold, from 0.2 µg m⁻³ (Mammoth Cave, summer) to 6.8 µg m⁻³ (Kanpur, dry
392 season). Corresponding mass fractions are 7-8 % in Kanpur, Beijing, and Buenos Aires, and
393 below 2% in Bandung. This heterogeneity reflects large spatial and temporal variation in NH₃
394 and NO_x (NO + NO₂) sources. There were noticeable seasonal increases in ANO₃ during
395 wintertime periods in Beijing, Kanpur, and Dhaka, coinciding with lower temperatures.
396

397 CM concentrations span an order of magnitude from 1.0 µg m⁻³ (Atlanta) to 16 µg m⁻³
398 (Beijing). The fraction of CM in PM_{2.5} exhibits pronounced variation (5-25%). Except during
399 dust storms, CM does not show clear patterns of temporal or regional variation. This could be
400 explained by non-seasonal road dust, which may account for over 80% of CM in regions with
401 heavy urban traffic (Huang et al., 2015).
402

403 We used Zn:Al ratios to assess the relative importance of local road dust (*c.f.* Table 3).
404 Aluminum is mostly natural in origin (Zhang et al., 2006) whereas Zn is primarily from tire wear
405 (Begum et al., 2010; Councill et al., 2004). For example, ratios are above 3 for Dhaka and
406 Hanoi, but less than 0.3 for Mammoth Cave and South Dekalb site (Atlanta). In fine-mode
407 aerosols, the ratio tends to be highest in large cities distant from natural CM. In coarse-mode
408 aerosols, a low Zn:Al ratio (< 0.1) indicates the aerosol CM component is dominated by regional
409 dust.
410

411 Absolute EBC spans an eight-fold concentration range from 1.1 µg m⁻³ (Atlanta) to above 8
412 µg m⁻³ (Dhaka and Kanpur). Mass fractions of EBC ranged from 4% (Singapore) to 25%
413 (Manila). Trace element oxide (TEO) material is mainly composed of Zn, Pb, Ni, Cu, and Ba,
414 hence also derived mainly from anthropogenic sources. TEO contributes negligibly to total mass
415 (1%), as expected. Sea salt remains a consistently small contributor (2%) to total mass, except
416 for Buenos Aires and Rehovot (5-6%) due to coastal winds. Particle-bound water (PBW) mass at
417 35% humidity is determined from the growth parameter κ_m. PBW mass contribution is similar to
418 EBC (7%). At low humidity, the combined mass of ANO₃, EBC, TEO, sea salt, and PBW
419 **accounts for** 15-35 % of aerosol mass.
420

421 RM as inferred from mass reconstruction of inorganic compounds, PBW, and total filter-
422 weighed mass is implicitly treated as the organic aerosol mass fraction. In terms of relative
423 composition, RM spans a factor of two, from 30% mass in Buenos Aires to almost 60% in

424 Kanpur. Temporal changes in RM tend to coincide with increases in ASO_4 , with an all-site $r^2 =$
425 0.92. Although RM, as defined here, is not fully independent from measured ASO_4 , correlations
426 between these two mass fractions imply related sources.
427

428 We interpret the abundance of water-soluble K relative to Al as an indicator of wood smoke
429 (e.g. Munchak et al., 2011). K:Al ratios averaged over each site range from < 2 (Mammoth Cave,
430 Atlanta) to 16 (Kanpur), where combustion activity is apparent. Singapore was downwind of
431 significant Indonesian forest fire smoke during its sampling period of Aug-Nov 2015, averaging
432 to K:Al = 13. The correlation between K:Al and RM across all SPARTAN sites is $r^2 = 0.73$,
433 supporting the attribution of RM as mostly organic.
434

435 Across all sites, coarse and fine mode mass fractions are approximately equal (0.50), with
436 fractions ranging from below 0.40 (Hanoi, Buenos Aires, and Manila) to above 0.55 (e.g.
437 Bandung, Kanpur, Atlanta, Mammoth Cave). The two size modes can be temporally correlated
438 per site, though sometimes weakly, from $r^2 = 0.15$ (Hanoi) to $r^2 = 0.76$ (Rehovot). We observe
439 strong temporal correlations between sulfate and ammonium in $PM_{2.5}$ ($r^2 = 0.72 - 0.99$). Nitrate
440 and ammonium are less consistently related (Table 3), ranging from higher values in Singapore
441 ($r^2 = 0.66$), Kanpur ($r^2 = 0.58$), Beijing ($r^2 = 0.28$), to weaker values in Ilorin and Manila ($r^2 <$
442 0.1). The strength of correlations with ammonium could be influenced by excess ammonium
443 relative to sulfate. The $[NH_4^+]/[SO_4^{2-}]$ ratio in $PM_{2.5}$ is 2.6 in Kanpur and 1.3 in Ilorin.

444 5.2. Collocation overview

445 We compare SPARTAN $PM_{2.5}$ speciation with previous studies available from the literature
446 and focus on collocated relative $PM_{2.5}$ composition of major components within the last 10 years.
447 TEO is omitted due to lack of significant mass contribution. Aerosol water content is also
448 omitted as it was not directly measured in any of the collocation studies. If not provided, CM is
449 treated as defined in Sect 4.5 where possible. Organic mass (OM) to organic carbon (OC) ratios
450 are from Philip et al. (2014b) with updates from Canagaratna et al. (2015).
451

452 Figure 3 provides an overview of the comparison studies organized by SPARTAN data
453 availability. Only sampling at Mammoth Cave sampling was temporally coincident with the
454 comparison data. SPARTAN compositional information is generally consistent with previous
455 studies, considering inter-annual chemical variation and measurement uncertainty. For example,
456 both SPARTAN and comparative studies find that $PM_{2.5}$ is composed of between 10-30% ASO_4
457 and 5-20% CM for sampled sites. SPARTAN EBC mass fraction generally matches within 5
458 percentage points of collocated studies, except for Bandung and Kanpur. SPARTAN and prior
459 studies find that ANO_3 is usually a small fraction of total mass, except at Beijing and Kanpur (7-
460 8%) due to their high agricultural and industrial activity. All studies find that sea-salt is below
461 3% of total mass. SPARTAN-derived RM has potentially the largest potential error, yet typically
462 is consistent with the combined organic and unknown masses of other studies. This offers further
463 evidence that SPARTAN measurements of RM are predominantly organic in nature.
464

465 5.3. Individual site characteristics

466 Below we discuss each site in more detail. We also examine how our chemical composition
467 from a global array of sites relates to local anthropogenic activities and surrounding area.

468 References to land type at specific sites are derived from Latham et al. (2014), unless otherwise
469 indicated. The number of filters is given in parentheses.

470

471 **5.3.1 Beijing, China (n = 114)**

472 Beijing has attracted considerable attention for its air pollution (Chen et al., 2013).
473 Agricultural areas to the west and the Gobi Desert to the north surround the city's 19 million
474 dwellers. The SPARTAN air sampler is located on the Tsinghua University campus, 15 km
475 northwest of the downtown center. This is our longest-running site, with 2.5 years of near-
476 continuous sampling. It reports the third-highest PM_{2.5}, at 69 µg m⁻³, the third highest ASO₄ (12
477 µg m⁻³) and the highest CM (16 µg m⁻³) of all sites. The significant ANO₃ (5.5 µg m⁻³) reflects
478 significant urban NO_x near agricultural NH₃ sources. ANO₃ values were highest during winter,
479 as expected from ammonium-nitrate thermodynamics. A high CM component in the springtime
480 reflects regional, natural CM sources. The mean PM_{2.5} Zn:Al ratio is lower than in other large
481 cities (0.51) likely due to a larger fraction of natural dust sources and the sampling location in the
482 northwest quadrant of the city, upwind of many traffic sources. The lowest coarse-mode Zn:Al
483 mass ratios are observed in April 2014 (0.07) and April 2015 (0.06) during the annual Yellow
484 dust storm season. This is balanced by urban dust sources throughout the year, in agreement with
485 Lin et al. (2015) who found evidence of high CM in industrial areas of Beijing.

486

487 *Beijing Comparison:* Relative masses in Beijing compare well with previous studies.
488 SPARTAN ASO₄ (19%) is close to Yang et al. (2011) (17%) and Oanh et al. (2006) (20%)
489 and the RM of 37% is similar to combined OM (33 and 29%) and unknown fractions (10 and
490 24%) of comparison studies. SPARTAN ANO₃ concentrations (8.5%) are relatively higher
491 than for most other locations, though lower than in either previous study (11-12%), possibly
492 due to different sampling periods. CM is greater than in Yang et al. (2011) (25% vs. 19%),
493 and significantly higher than in Oanh et al. (2006) (5%), potentially due to a difference in
494 definitions.

495

496 **5.3.2 Bandung, Indonesia (n = 77)**

497 Bandung is located inland on western Java surrounded by a volcanic mountain range and
498 agriculture (e.g. tea plantations). The sampler is located on the Institute of Technology Bandung
499 campus, 5 km north of the city center. Almost two years of sampling have resulted in a mean
500 PM_{2.5} concentration of 31 µg m⁻³. Sea salt is low at this elevated (826 m) inland site. ANO₃ and
501 CM levels are also low, but RM is moderately high compared with other sites, at 55%. This
502 could be explained by large amounts of vegetative burning; organic PM_{2.5} mass fractions can rise
503 above 70% during combustion episodes (Fujii et al., 2014). Volcanic sources of sulfur, in
504 addition to industrial sources, may explain the relatively higher ASO₄ compared with Manila or
505 Dhaka (Lestari and Mauliadi, 2009). Influxes of volcanic dust from the Sinabang volcano from
506 August – September 2014 (2000 km northwest of Bandung) could explain why coarse-mode
507 Zn:Al ratios drop to 0.09 for this period compared to the annual mean of 0.21.

508

509 *Bandung Collocation:* Bandung is a volcanically active area, so that composition, in
510 particular ASO₄, differs due to naturally variable circumstances. SPARTAN ASO₄ (21%) is
511 higher than the 4% fraction reported by Lestari and Mauliadi (2009), but is identical with
512 measurements by Oanh et al. (2006). SPARTAN EBC (13%) is less than either previous
513 study (19% and 25%) and the more recent analysis of 19% BC (Santoso et al., 2013).

514 SPARTAN ANO₃ is 2% by mass, lower than measured by Oanh et al. (2006) (13%) but
515 similar to Lestari and Mauliadi (2009). Both of the earlier studies show lower RM fractions
516 (36%, and 42%) compared with 54% RM in this study.
517

518 **5.3.3 Manila, Philippines (n = 63)**

519 Manila is a coastal city located in Manila Bay, adjacent to the South China Sea and
520 surrounded by mountains. The sampling station, located at the Manila Observatory, is about 40
521 m higher in altitude than the central city. The PM_{2.5} concentrations at the observatory (18 µg m⁻³)
522 are expected to be lower than in the main city, but still influenced by vehicular traffic, fuel
523 combustion and industry (Cohen et al., 2009). Compared to the all-site average, the CM fraction
524 in Manila is typical (11%), but **equivalent black carbon** is twice as great (25%). The high EBC
525 agrees with previous observations, attributable to a relatively high use of diesel engines (Cohen
526 et al., 2002).
527

528 *Manila Collocation:* SPARTAN fractions of ASO₄ and EBC are similar to Cohen et al.
529 (2009). Our RM (43%) is lower than OM (57%), whereas SPARTAN CM was greater than
530 Cohen et al. (2009). These differences could reflect sampling differences, or emission
531 changes over the last decade.
532

533 **5.3.4 Dhaka, Bangladesh (n = 41)**

534 Dhaka is a densely populated city (17,000 persons/km²) in a densely populated country
535 (1,100 persons/km²). The sampler is situated in the heart of downtown Dhaka, on the University
536 of Dhaka rooftop, and is influenced by air masses from the Indo Gangetic Plain (Begum et al.,
537 2012). More than half the country is used for agricultural purposes (Ahmed, 2013). Local
538 contributing PM_{2.5} sources include coal and biomass burning, and heavy road traffic combustion
539 products and dust (Begum et al., 2010, 2012). PM_{2.5} concentrations are the fourth highest of any
540 SPARTAN site, at 52 µg m⁻³. Dhaka has the second-highest absolute EBC of any site, at 8.4 µg
541 m⁻³, which can be explained by the abundance of truck diesel engines (Begum et al., 2012). We
542 estimate 41% of PM_{2.5} in Dhaka is RM. Crop or bush burning on both local and regional scales
543 contribute significantly to organics (Begum et al., 2012). The high mean PM_{2.5} Zn:Al ratio of 3.4
544 reflects a large contribution from urban traffic.
545

546 **5.3.5 Ilorin, Nigeria (n = 40)**

547 Ilorin is located in a rural area with low-level agriculture and shrub vegetation. The sampler
548 is sited on the university campus, 15 km east of the city of 500,000 people. Aerosol loadings
549 have seasonal cycles from agricultural burning events and dust storms (Generoso et al., 2003).
550 The RM accounted for two thirds of total PM_{2.5} mass, among the largest, influenced by biomass
551 burning. There is evidence of biomass burning in the PM_{2.5} peak in late spring 2014, and again in
552 2015. Lower ASO₄ (12%) compared to other SPARTAN sites reflects the sparse surrounding
553 industry. CM levels are comparable to other locations, except during dust storms. During a dust
554 storm (between April 14th - May 2nd 2015), CM increased to two thirds of PM_{2.5} mass. The PM_c
555 Zn:Al ratio during the storm decreased to 0.01 versus 0.25 during non-storm days.
556

557 **5.3.6 Kanpur, India (n = 33)**

558 Kanpur is a city of 2.5 million people. The sampler is located at the IIT Kanpur campus
559 airstrip, about 10 km northwest of the city. The city lies in the Indo-Gangetic Plain, where

560 massive river floodplains are used for agricultural and industrial activity (Ram et al., 2012). We
561 sampled from December 2013 – May 2014, and September-November 2014, capturing one dry
562 season. SPARTAN-measured $PM_{2.5}$ for this period was $99 \mu g m^{-3}$, the highest of any SPARTAN
563 site, of which 59% is RM, 19% ASO_4 , and 7.4% ANO_3 . The absolute values of all three
564 components are also the highest among those measured. Molar $[NH_4^+]:[SO_4^{2-}]$ ratios are higher
565 in Kanpur (2.6) than elsewhere. High background ammonia has been observed in the region
566 from satellite (e.g. Clarisse et al., 2009) which could explain the high levels of ANO_3 . Wood
567 smoke is apparent from the high K:Al ratio (16), associated with organic matter burning during
568 winter dry months. We detected significant Zn concentrations (Zn:Al = 1.0), which is in
569 agreement with Misra et al. (2014) observations of a tripling of zinc during anthropogenic
570 sourced dust.

571
572 *Kanpur Collocation:* Relative fractions among the major species CM, sea salt, ASO_4 &
573 ANO_3 all match well with previous studies (Behera and Sharma, 2010; Chakraborty et al.,
574 2015; Ram et al., 2012) that also sampled during winter dry seasons. Chakraborty et al.
575 (2015) measured 70% organic mass composition and found a combined mass of 28% for
576 $ASO_4 + ANO_3$ compared to SPARTAN mass (26%). SPARTAN ASO_4 (19%) compares well
577 to 13% of Ram et al. (2012) and 18% for Behera and Sharma (2010), and ANO_3 (7.4%) is
578 close to previous values (6.1% and 6.6%). By comparison SPARTAN slightly overestimates
579 EBC by 4-6%. SPARTAN CM (4.8%) is lower than Behera and Sharma (2010) (10%).
580 Notably the combined OM + unknown fractions from these previous two studies account for
581 almost two thirds of aerosol mass, 58% for Behera and Sharma (2010) and 63% for Ram et
582 al. (2012), similar to our 59% RM estimate. SPARTAN $PM_{2.5}$ concentrations, as well as RM,
583 reach a maximum during the month of December. This is consistent with recent work
584 (Villalobos et al., 2015), who attribute this increase to agricultural burning and stagnant air.

585 586 **5.3.7 Buenos Aires, Argentina (n = 31)**

587 Buenos Aires has a metropolitan population of 12 million. SPARTAN instruments are
588 located on the urban CITEDEF campus 20 km west of the central downtown. The megacity, the
589 southernmost in our study, is surrounded by grassland and farming on the west and the Atlantic
590 Ocean on the east. The latter explains the relatively high proportion (6%) of sea salt. Total $PM_{2.5}$
591 ($10 \mu g m^{-3}$) and relative RM (31%) are low compared with other large metropolitan areas, likely
592 influenced by clean maritime air. In addition to sea salt and natural CM, the contribution of EBC
593 is 17%, which could reflect significant local truck diesel combustion (Jasan et al., 2009).

594 595 **5.3.8 Rehovot, Israel (n = 30)**

596 Rehovot is located on a four-story rooftop on the Weizmann Institute campus, 11 km from
597 the Mediterranean Sea and 20 km south of Tel Aviv. The city is surrounded by semi-arid, mixed-
598 use cropland, and the region experiences occasional Saharan desert dust outbreaks. Typical $PM_{2.5}$
599 concentrations are low ($16 \mu g m^{-3}$), with the composition in Rehovot consisting of 29% ASO_4 ,
600 and 20% CM. The RM fraction is smaller in Rehovot (16% total $PM_{2.5}$ mass) than at other
601 SPARTAN sites. Aerosol sources in Israel include agriculture, desert dust, traffic and coal-based
602 power plants (Graham et al., 2004). Relative sodium concentrations are high in Rehovot (4%),
603 similar to Buenos Aires and Ilorin, and may include a contribution from dust.

604

605 *Lag Ba'Omer festival:* We measured high ASO₄ concentrations on May 7-18, 2015, during
606 which time a large number of bonfires were lit nearby. During the festival, over 75% of total
607 aerosol mass came from ASO₄ + ANO₃, leading to a brief doubling of the hygroscopic
608 parameter κ_v . We observed a K:Al ratio of 38 for May 6th of the festival, the highest for any
609 single filter.

610
611 *Saharan dust storm:* We had the opportunity to measure a severe dust storm in Rehovot from
612 a filter sampling February 4-13, 2015. The coarse filter Zn:Al ratio dropped to 0.02 during
613 the Saharan dust storm from the typical value of 0.3. On the coarse filter we obtained an
614 absolute CM mass of 950 μg , which accounts for half of the collected mass during the storm.
615 13% of dust storm PM_c is combined sea salt, ANO₃, and ASO₄, leaving 35% RM. Although
616 this RM fraction may imply an incomplete CM extraction, it is possible that a significant
617 portion of desert dust carries adsorbed organic material (Falkovich et al., 2004).

618 619 **5.3.9 Mammoth Cave NP, US (n = 19)**

620 The Mammoth Cave sampling site straddles National Park mountainous terrain to the north
621 and east, with farmland to the south and west. It is about 35 km from the closest town, Bowling
622 Green, KY, with about 50,000 residents. Sources of PM are expected to be non-local, hence we
623 consider it our 'background' site.

624
625 *Mammoth Cave National Park Collocation:* This temporary SPARTAN site was deployed
626 for comparison with the IMPROVE network station (IMPROVE, 2015). Unique among our
627 sites, sampling was temporally coincident with IMPROVE's 1-in-3 day regimen. We
628 obtained quality-controlled samples from June-August 2014. Temporal variation in daily
629 values is consistent with IMPROVE for sulfate ($r^2 = 0.86$, slope = 1.03) and total mass of
630 PM_{2.5} ($r^2 = 0.76$, slope = 1.12). Differences between IMPROVE vs. SPARTAN are small for
631 ASO₄ (36% vs. 33%), ANO₃ (2.4% vs. 1.2%), CM (7% vs. 11%), and EBC (3.0% vs. 5.6%),
632 respectively. The combined OM + unknown + water fraction IMPROVE was 51%, similar to
633 the SPARTAN RM mass fraction of 49%.

634 635 **5.3.10 Atlanta, US (n = 13)**

636 Atlanta represents a major urban area in a developed country. The temporary SPARTAN site
637 was located at the South Dekalb supersite 15 km east of downtown Atlanta. Air sampling was
638 performed for a 4-month period spanning winter to spring 2014. Over the past 10 years
639 significant decreases in PM_{2.5} have been observed here and across the eastern United States
640 (Boys et al., 2014). The surrounding region is tree-covered or agricultural.

641
642 *Atlanta (South Dekalb) Collocation:* Co-sampled filters from the Atlanta CSN station
643 (USEPA, 2015) provide a comparison with the summer 2014 SPARTAN data. The EPA OM
644 fraction (43%) agrees well with the SPARTAN mean RM (48%). Crustal, SS, EBC and
645 ASO₄ are within 2% relative to total composition. SPARTAN component fractions in Atlanta
646 are also consistent with respect to Butler et al. (2003); components CM (12% vs. 10%),
647 ASO₄ (23% vs. 28%), ANO₃ (3.5% vs 4%) and RM and OM (48% vs 55%) closely match,
648 except for EBC (11% vs. 3%), perhaps reflecting different time periods.

649 650 **5.3.11 Singapore, Singapore (n = 12)**

651 Singapore is a densely populated coastal city-state at 7,770 people/km². The sampler is
652 located on a rooftop at the National University of Singapore (NUS), near the center of the city.
653 Transportation is mixed-use, including taxis, rail, and bicycles, which may help explain the
654 relatively low EBC and CM of 3%. Despite this, the Zn:Al ratio remains high at 1.5, implying a
655 dominant traffic-based contribution to CM. SPARTAN instruments have observed significant
656 biomass burning downwind from Indonesia, causing an increase in absolute PM_{2.5} from 32 in
657 August to 120 µg m⁻³ in September 2015, as well as an increase in RM from 44% to 62%. The
658 K:Al ratio steadily increased during this same period, from 7.2 (Jul 24 – Aug 2, 2015) to 17 – 24
659 (Aug 11 – Sept 25).

660
661

662 **5.3.12 Hanoi, Vietnam (n = 10)**

663 Hanoi is an inland megacity surrounded by grassland and agriculture. The sampler itself is on
664 a building rooftop at the Vietnam Academy of Science and Technology, 5 km northwest of the
665 city center. Motorbikes are the main forms of transportation downtown and the primary source of
666 mobile-based PM_{2.5} (Vu Van et al., 2013). In Hanoi, the PM_{2.5} Zn:Al ratio was 3.7, also the
667 highest of any SPARTAN site, indicative of significant traffic and tire wear.

668

669 *Hanoi Comparison:* SPARTAN PM_{2.5} composition is generally consistent with Cohen et al.
670 (2010). Slight differences are perhaps related to differences in sampling season and location.
671 SPARTAN sea salt fraction was larger (2.5% vs. 0.6%), but with a lower ASO₄ fraction
672 (17%) compared with Cohen et al. (2010) (29%). Sulfate tends to be lower in the spring-
673 summer seasons, coinciding with our measurement period, which may explain the
674 discrepancy. SPARTAN EBC (10%) is close to the Cohen et al. (2010) value of 8%, whereas
675 SPARTAN RM (51%) and CM (16%) masses are slightly higher.

676

677 **5.3.13 Pretoria, South Africa (n = 5)**

678 Pretoria is a high-altitude city (1300 m) surrounded by arid, low-intensity agriculture and
679 extensive grasslands. The SPARTAN sampler is located on a 10 m CSIR building rooftop 12 km
680 east of downtown area (*pop.* 700,000). Preliminary measurements of south-hemisphere
681 springtime show absolute PM_{2.5} concentrations to be low, at 6.4 µg m⁻³. There are significant
682 fractions of CM (22%) and EBC (22%), and low RM (14%). The PM_{2.5} Zn:Al ratio (0.69)
683 indicates vehicle traffic contributes to CM.

684 **6. Refining estimates of dry hourly PM_{2.5} using κ_v**

685 Our assessment of PM_{2.5} hygroscopicity is determined by site-specific chemical composition. We
686 then use the time-varying hygroscopicity to refine the PM_{2.5} values inferred from nephelometer
687 scatter.

688 **6.1. Relating PM_{2.5} composition to κ_v**

689 The outer pie charts of Figure 2 show the site-mean hygroscopic growth constant κ_v ,
690 surrounded by the water contributions at 35% RH. The major contributors to PBW are ASO₄,
691 ANO₃, RM, and sea salt, as inferred from the values listed in Table 2 and weighted by
692 composition as in Eq. 5. ASO₄ and RM contribute similarly to total aerosol water whereas ANO₃
693 contributes less to PM_{2.5} hygroscopicity due to its smaller mass. The contribution of sea salt to

694 hygroscopicity can be significant, and makes a dominant contribution in both Rehovot and
695 Buenos Aires.

696
697 The parameter κ_v , when averaged across all sites, is 0.20, matching the generic estimate
698 $\kappa_{v,tot} = 0.2$ applied in the initial SPARTAN study (Snider et al., 2015). Recently Brock et al.
699 (2016) estimate κ_v values between 0.15 and 0.25 for ambient aerosols with 50% organic
700 composition at subsaturated humidity. The local SPARTAN value in Atlanta (0.17) is consistent
701 with the value of 0.16 ± 0.07 by Padró et al. (2012) in Atlanta. We found significant long-term
702 differences in $\kappa_{v,tot}$ between cities, from 0.15 in Ilorin to 0.28 in Rehovot, and differences
703 between filters at single sites ($\sigma \sim 0.05$). There is little correlation of $\kappa_{v,tot}$ with changes in mass
704 ($r^2 < 0.01$). However, there are significant changes in $\kappa_{v,tot}$ due to seasonality and specific
705 events (e.g. dust storms, fires). In Beijing, aerosol hygroscopicity was 50% higher in mid
706 summer (August) due to increased sulfate, and in late winter (March) due to a relative increase in
707 sea salt. A summertime sulfate peak also agrees with observations by Yang et al. (2011). Table 3
708 shows the site-specific PBW in $PM_{2.5}$. At $RH = 35\%$, PBW ranges from $0.6 - 6 \mu g m^{-3}$,
709 comparable in absolute values to EBC. Above 80% RH PBW will account for more than half of
710 aerosol mass. Accounting for this water component in nephelometer scatter motivates the
711 following section.

712 6.2. Relating nephelometer scatter to dry (RH=35%) $PM_{2.5}$

713 We apply a temporally resolved, site-specific κ_v to refine our relationship between total
714 nephelometer scatter and $PM_{2.5}$. We calculate a 45-day running mean aerosol volume-weighted
715 κ_v at each SPARTAN site. We then use the hygroscopic growth factors to estimate dry hourly
716 $PM_{2.5}$ from hourly nephelometer measurements of ambient scatter and hourly measured RH.
717 Appendix A2 describes the procedure in more detail.

718
719 We compared our hourly $PM_{2.5}$ in Beijing with $PM_{2.5}$ measurements from a Beta Attenuation
720 Monitor (BAM, MetOne) at the US Embassy, located 15 km away. The BAM instrument
721 contains a drying column with a 35% humidity set point. The left panel of Figure 4 shows the
722 time series of hourly dry $PM_{2.5}$ concentrations predicted by SPARTAN during the summer.
723 Pronounced temporal variation is apparent, with $PM_{2.5}$ concentrations varying by more than an
724 order of magnitude. A high degree of consistency is found with the BAM ($r^2 = 0.67$). The
725 exclusion of water uptake in hourly $PM_{2.5}$ estimates (by setting all $\kappa_v = 0$) decreased hourly
726 correlations slightly to $r^2 = 0.62$. The average humidity in Beijing was 47% for the measurement
727 period, corresponding to a mean 17% volume contribution by water ($\kappa_v = 0.19$). Hygroscopic
728 growth should play a more significant role under more humid conditions (e.g. Manila and
729 Dhaka).

730
731 The right panel in Figure 4 shows daily-averaged $PM_{2.5}$ ($n = 148$). In 2014 there were 3167
732 coincidentally available hours with which to compare. The coefficient of variation for averaged
733 24-hour measurements remained high ($r^2 = 0.70$). There was a mean offset of $10 \mu g m^{-3}$.
734 However the slope is near unity (0.98), suggesting excellent proportionality between our
735 nephelometer and the BAM instrument for $PM_{2.5}$ concentrations below $200 \mu g m^{-3}$. Above this
736 concentration, nephelometer signals become non-linear. The agreement remained similar for
737 hourly values ($r^2 = 0.67$).

738

739 7. Conclusions

740

741 We have established a multi-country network where continuous monitoring with a 3-
742 wavelength nephelometer is combined with a single multi-day composite filter sample to provide
743 information on PM_{2.5}. Long-term average aerosol composition is inferred from the filters,
744 including equivalent black carbon, sea salt, crustal material, ammoniated sulfate, and ammonium
745 nitrate. This composition information was applied to calculate aerosol hygroscopicity, and in turn
746 the relation between aerosol scatter at ambient and controlled RH. These data provide a
747 consistent set of compositional measurements from 13 sites in 11 countries.

748

749 We report ongoing measurements of fine particulate matter (PM_{2.5}), including compositional
750 information, in 13 locations in two month or greater intervals all within a three-year span (2013-
751 2016). The mean composition averaged for all SPARTAN sites is ammoniated sulfate (20% ±
752 11%), crustal material (13.4% ± 9.9%), equivalent black carbon (11.9% ± 8.4%), ammonium
753 nitrate (4.7% ± 3.0%), sea salt (2.3% ± 1.6%), trace element oxides (1.0% ± 1.1%), water (7.2%
754 ± 3.3%) at 35% RH, and residual matter, which is probably primarily organic (40% ± 24%).

755

756 Analysis of filter samples reveals that several PM_{2.5} chemical components varied by more
757 than an order of magnitude between sites. Ammoniated sulfate ranged from 1 µg m⁻³ in Buenos
758 Aires to 17 µg m⁻³ in Kanpur (dry season). Ammonium nitrate ranged from 0.2 µg m⁻³
759 (Mammoth Cave, summertime) to 6.8 µg m⁻³ (Kanpur, dry season). Equivalent black carbon
760 ranged from 0.7 µg m⁻³ (Mammoth Cave) to 8 µg m⁻³ (Dhaka and Kanpur). Locations with
761 enhanced sulfate tend to have enhancements in other PM components. For example, ammoniated
762 sulfate and residual matter (probably organic) are highly correlated across sites ($r^2 = 0.92$).

763

764 Crustal material concentrations ranged from 1 µg m⁻³ (Atlanta) to 16 µg m⁻³ (Beijing).
765 Measuring Zn:Al ratios in PM_{2.5} was an effective way to determine anthropogenic contribution
766 to crustal material. Ratios larger than 0.5 identified sites with significant road dust contributions
767 (e.g. in Hanoi, Dhaka, Manila, and Kanpur). Some locations, such as Beijing and Buenos Aires,
768 had both high anthropogenic and natural crustal material. Low coarse Zn:Al ratios were apparent
769 during natural dust storms. Anthropogenic crustal material is an aerosol component neglected by
770 most global models and which may deserve more attention.

771

772 Potassium is a known marker for wood smoke. Enhanced K:Al ratios were found in
773 Singapore downwind of Indonesian forest fires, in Kanpur during the winter dry season from
774 agricultural burning, and in Rehovot during a bonfire festival. Furthermore, these ratios were
775 correlated with RM concentrations ($r^2 = 0.73$), supporting the attribution of RM as mostly
776 organic.

777

778 SPARTAN measurements generally agree well with previous collocated studies. SPARTAN
779 sulfate fractions are within 4% of fractions measured at eight of the ten collocated, though
780 temporally non-coincident, studies. Dedicated contemporaneous collocation with IMPROVE at
781 Mammoth Cave yielded a high degree of consistency with daily sulfate ($r^2 = 0.86$, slope = 1.03),
782 daily PM_{2.5} ($r^2 = 0.76$, slope = 1.12), and mean fractions for all major PM_{2.5} components (within

783 2%). Crustal material is typically consistent with the previous measurements, at 5-15%
784 composition. SPARTAN equivalent black carbon ranged broadly, from 3% (Singapore) to 25%
785 (Manila), and matched within a few percent of most previous works. Ammonium nitrate (4%)
786 generally matched other sites, though it was sometimes lower, as in Beijing and Atlanta. Sea-salt
787 was consistently low, as found in previous measurements. Sea salt fractions were highest in
788 Buenos Aires and Rehovot (6%), reflecting natural coastal aerosols. SPARTAN residual matter
789 is consistent with the combined organic and unknown masses. Comparing with collocated
790 measurements supports the expectation that most of the RM is partially organic. Residual matter
791 could also include unaccounted-for particle bound water, measurement error, and possibly
792 unmeasured inorganic materials.

793
794 We calculated the hygroscopic constant κ_v for individual $PM_{2.5}$ filters to estimate water at
795 variable humidity, and to infer wet and water-free residual matter. Based on a range of literature,
796 we treated residual matter as mostly organic, with constant $\kappa_{v, RM} = 0.1$. Residual matter and
797 ammoniated sulfate largely determined overall water uptake in aerosols. These individual
798 species, along with sea salt and ammonium nitrate, resulted in a mean mixed hygroscopic
799 constant of 0.20, implying that for many sites, water content above 80% RH will account for
800 more than half of aerosol mass. For cleanroom conditions of low humidity (35% RH), mean
801 water composition was estimated to be 7% by mass.

802
803 Water retention calculations allow for volumetric fluctuation estimates of aerosol water at
804 variable RH. We subtracted the water component to predict dry nephelometer scatter as a
805 function of time, anchored to filter masses at 35% RH. For Beijing, we assessed the consistency
806 of SPARTAN predictions of hourly $PM_{2.5}$ values with BAM measurements taken 15 km away,
807 and found temporal consistency ($r^2 = 0.67$), with a slope near unity (0.98). The explained
808 variance decreased to $r^2 = 0.62$ when setting $\kappa_v = 0$. This comparison tested both SPARTAN
809 instrumentation and our treatment of aerosol water uptake.

810
811 These measurements provide chemical and physical data for future research on $PM_{2.5}$.
812 Collocation with sun photometer measurements of AOD connects satellite observations to
813 ground-based measurements and provides information needed to evaluate chemical transport
814 model simulations of the $PM_{2.5}$ to AOD ratio. As sampling expands, SPARTAN will provide
815 long-term data on fine aerosol variability from around the world. Ongoing work includes an
816 analysis of trace metal concentrations and interpreting SPARTAN measurements with a
817 chemical transport model. The data are freely available as a public good at [www.spartan-](http://www.spartan-network.org)
818 [network.org](http://www.spartan-network.org). We welcome expressions of interest to join this grass-roots network.

819
820

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838 8. References

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1182 **Figures and Tables**

1183 **Table 1: Summary of speciation definitions**

Species (at 0% RH)	Measurement Method	Species Mass (μg). For concentrations, divide masses by sampling volume v	Reference
SS	IC (anion and cation)	$2.54[\text{Na}^+]_{\text{SS}}$, where $[\text{Na}^+]_{\text{SS}} = [\text{Na}^+]_{\text{tot}} - 0.1[\text{Al}]$	(Remoundaki et al., 2013) (Malm et al., 1994)
ANO ₃		$1.29[\text{NO}_3^-]$	(Malm et al., 1994)
ASO ₄		$[\text{SO}_4^{2-}]_{\text{non-ss}} + [\text{NH}_4^+] - 0.29[\text{NO}_3^-]$, where $[\text{SO}_4^{2-}]_{\text{non-ss}} = [\text{SO}_4^{2-}]_{\text{total}} - 0.12[\text{Na}^+]$	(Dabek-Zlotorzynska et al., 2011; Henning et al., 2003)
Na ₂ SO ₄		$0.18[\text{Na}^+]_{\text{SS}}$	
CM	ICP-MS	$10 \times ([\text{Al}] + [\text{Mg}] + [\text{Fe}])$	(Wang, 2015)
EBC	SSR	$20.7 \times \ln(R_o/R)$	(Taha et al., 2007)
TEO	ICP-MS	$1.47[\text{V}] + 1.27[\text{Ni}] + 1.25[\text{Cu}] + 1.24[\text{Zn}] + 1.32[\text{As}] + 1.2[\text{Se}] + 1.07[\text{Ag}] + 1.14[\text{Cd}] + 1.2[\text{Sb}] + 1.12[\text{Ba}] + 1.23[\text{Ce}] + 1.08[\text{Pb}]$	(Malm et al., 1994)
PBW _{inorg}	$\kappa_{m,X}$	$\sum_X [f_{m,X}(\text{RH}) - 1][X]$	(Kreidenweis et al., 2008)
PBW _{RM}		$\text{RM}(1 - 1/f_{m,\text{RM}})$	Table 2
RM(35%)	Mass Balance	$[\text{PM}_{2.5}] - \{[\text{EBC}] + [\text{CM}] + [\text{TEO}] + [\text{ANO}_3] + [\text{SS}] + [\text{ASO}_4] + [\text{Na}_2\text{SO}_4] + [\text{PBW}]_{\text{inorg}}\}$	This Study
RM(0%)	Mass Balance $\kappa_{m,\text{OM}} = 0.07$	$\text{RM}(35\%) - \text{PBW}_{\text{RM}}$	Organic growth factors: (Jimenez et al., 2009; Sun et al., 2011)

1184 **Species:** EBC = Equivalent black carbon, TEO = Trace metal oxides, CM = Crustal Material, ANO₃ = Ammonium
 1185 nitrate, ASO₄ = Ammoniated sulfate, PBW = particle-bound water, RM = residual matter (assumed representative of
 1186 organic matter), [X] = concentration of any hygroscopic species. **Measurement Instruments:** IC = Ion
 1187 Chromatography, ICP-MS Inductively coupled plasma mass spectrometry, SSR = Smoke Stain Reflectometer, $\kappa_{m,X}$
 1188 = single-parameter hygroscopicity by mass (Kreidenweis et al., 2008). RH = Relative Humidity,

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1190 **Table 2: κ -Kohler constants for volume (κ_v), mass (κ_m), and related quantities**

Compound [X]	$\kappa_{v,X}$	Approximate Density ($\rho_X/\rho_{\text{water}}$)	$\kappa_{m,X}$	PBW(% mass) at	
				RH = 35%	RH = 80%
Crustal	0	2.5 ^a	0	0	0
EBC	0	1.8 ^b	0	0	0
TEO	0	2.5	0	0	0
RM	0.1 ^c	1.4	0.07	2	12
ANO ₃	0.41	1.72	0.24	17	61
ASO ₄	0.51	1.76	0.29	15	56
Na ₂ SO ₄	0.68 ^d	2.68 ^d	0.25	12	50
SS	1.5 ^e	2.16	0.69	22	68

1191 PBW = Particle-bound water. EBC = Equivalent black carbon, TEO = Trace Element Oxides, RM = **Residual Matter**
 1192 (associated with organics), ANO₃ = ammonium nitrate, ASO₄ = Ammoniated sulfate. ^aWagner et al. (2009), ^bBond
 1193 and Bergstrom (2006), ^cAssuming an urban O:C ratio of 0.5 then $\kappa_{v,\text{OM}} = 0.1$ Jimenez et al. (2009), ^dPetters and
 1194 Kreidenweis (2007). ^eFitted using non-deliqesced, subsaturated AIM Model III values, plus 0% RH endpoint by
 1195 Kreidenweis et al. (2008).

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Table 3: PM_{2.5} composition and water content ($\mu\text{g m}^{-3}$) at each SPARTAN location.

City	Host Institute	Lat/Lon (°)	Elev./Inst. Elev. (m)	Filters (<i>n</i>)	ASO ₄	ANO ₃	CM	SS	EBC	TEO	RM	PBW 35%RH	ρ 0%RH (g/cm ³)	NO ₃ vs. NH ₄ ⁺ (r ²)	PM _{2.5}	PM _{2.5} /PM ₁₀	<i>K_{p,tot}</i>	PM _{2.5} K/Al	Zn/Al	Filter Sampling Period
Beijing	Tsinghua University	40.010, 116.333	60//7.5	114	12.0 ^a (7.9)	5.5 (6.4)	15.9 (8.8)	1.5 (2.1)	5.7 (3.4)	0.62 (0.51)	23.8 (18)	4.7 (2.8)	1.69	0.32	69.5 (2.5)	0.49	0.19	2.9	0.51	2013/06–2016/02
Bandung	ITB Bandung	-6.888, 107.610	826//20	77	6.0 (2.3)	0.7 (1.3)	2.5 (1.5)	0.3 (0.2)	3.7 (2.0)	0.14 (0.11)	16.0 (5.9)	1.9 (0.6)	1.55	0.06	31.4 (1.0)	0.58	0.17	6.8	0.52	2014/01–2015/11
Manila	Manila Observatory	14.635, 121.080	60//10	63	2.7 (1.5)	0.3 (0.2)	1.9 (1.0)	0.5 (0.4)	4.3 (3.3)	0.13 (0.13)	7.3 (3.5)	1.1 (0.5)	1.61	0.03	18.2 (0.8)	0.39	0.16	6.3	1.03	2014/02–
Dhaka	Dhaka University	23.728, 90.398	20//20	41	7.5 (4.3)	2.1 (1.8)	5.9 (4.0)	1.4 (1.7)	8.4 (5.1)	1.50 (1.46)	21.4 (16)	3.5 (2.2)	1.63	0.43	51.9 (3.7)	0.40	0.17	5.3	3.39	2014/05–2015/11
Ilorin	Ilorin University	8.484, 4.675	330//10	40	1.9 (0.8)	0.3 (0.1)	3.0 (2.2)	0.3 (0.4)	1.6 (0.8)	0.09 (0.07)	7.6 (3.8)	0.9 (0.4)	1.62	0.05	15.7 (0.8)	0.44	0.15	2.9	0.49	2014/03–
Kanpur	IIT Kanpur	26.519, 80.233	130//10	33	17.6 (12)	6.8 (5.3)	4.4 (2.3)	0.6 (0.3)	8.3 (4.7)	0.47 (0.36)	54.6 (33)	6.3 (3.6)	1.52	0.58	99.3 (9.1)	0.56	0.18	16.2	1.01	2015/10–2013/12
Buenos Aires	CITEDEF	-34.560, -58.506	25//7	31	1.1 (0.5)	0.8 (0.4)	2.2 (1.6)	0.6 (0.3)	1.7 (1.2)	0.12 (0.12)	3.1 (1.8)	0.9 (0.3)	1.70	0.28	10.1 (0.6)	0.39	0.19	2.7	0.44	2014/11–2014/10
Rehovot	Weizmann Institute	31.907, 34.810	20//10	30	4.7 (1.9)	0.9 (0.5)	3.3 (1.6)	0.7 (0.6)	2.2 (2.0)	0.12 (0.13)	2.6 (2.8)	1.6 (0.6)	1.79	0.01	16.1 (1.0)	0.40	0.28	2.7	0.40	2015/02–2016/02
Mammoth Cave NP	Mammoth Cave	37.132, -86.148	235//7	19	4.1 (2.4)	0.2 (0.1)	1.4 (1.4)	0.1 (0.1)	0.7 (0.4)	0.02 (0.03)	6.1 (4.3)	1.0 (0.5)	1.59	0.00	13.6 (1.8)	0.56	0.22	1.1	0.13	2014/04–2014/08
Atlanta	Emory University	33.688, -84.290	250//2	13	2.0 (0.9)	0.3 (0.1)	1.0 (0.4)	0.1 (0.1)	1.1 (1.0)	0.04 (0.02)	4.1 (1.8)	0.6 (0.2)	1.61	0.00	9.1 (0.7)	0.69	0.17	1.9	0.26	2014/01–2014/05
Singapore	NUS	1.298, 103.780	10//20	12	16.1 (6.5)	1.2 (0.9)	0.8 (0.3)	0.9 (0.4)	3.1 (2.7)	0.20 (0.16)	39.8 (29)	5.0 (2.4)	1.48	0.66	66.8 (11)	NA	0.21	13.2	1.53	2015/08–
Hanoi	Vietnam Acad. Sci.	21.048, 105.800	10//20	10	6.0 (2.1)	1.6 (0.4)	5.6 (5.4)	0.9 (0.2)	3.7 (2.1)	0.69 (0.43)	18.2 (7.8)	2.6 (0.7)	1.59	0.22	39.4 (3.9)	0.38	0.18	8.9	3.74	2015/12–2015/05
Pretoria	CSIR	-25.756, 28.280	1310//10	5	1.2 (1.6)	0.7 (0.3)	1.3 (1.8)	0.2 (0.1)	1.4 (0.9)	0.04 (0.04)	1.0 (0.7)	0.5 (0.4)	2.09	0.48	6.4 (2.3)	0.32	0.24	6.0	0.86	2015/09–2015/11
SPARTAN mean (% mass)	All sites			497	20 (11)%	4.7 (3.0)%	13.4 (9.9)%	2.3 (1.6)%	11.9 (8.4)%	1.0 (1.1)%	40 (24)%	7.2 (3.3)%	1.65	0.24	32.4 (2.9)	0.50	0.20	4.6 ^b	0.73 ^b	2013–2016

^aValues in parentheses are 1 σ standard deviations. RH = Relative Humidity, ANO₃ = ammonium nitrate, ASO₄ = Ammoniated sulfate, CM = Crustal material, EBC = Equivalent black carbon, TEO = Trace Element Oxides, RM = Residual Matter, PBW = Particle-bound water. Mean Na₂SO₄ was not significant (< 0.1 $\mu\text{g m}^{-3}$) at any SPARTAN site. ^bGeometric mean

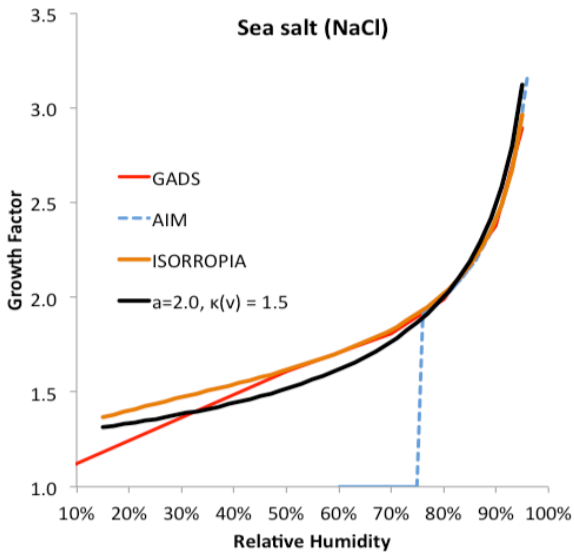
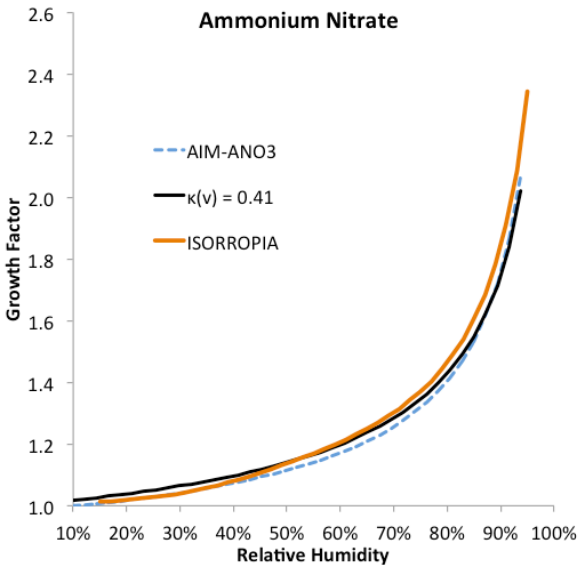
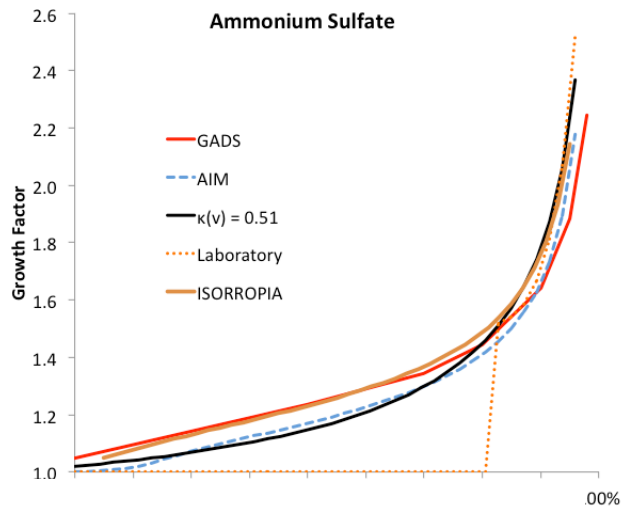


Figure 1: Hygroscopic growth factors for ammonium sulfate (top), ammonium nitrate (centre), and sea salt (bottom). GADS = Global Aerosol Dataset estimated from empirical data (Koepke et al., 1997). ISORROPIA = Aerosol thermodynamic model at T=298K (reverse mode) and assuming linear water/solvent volume additivity (Fountoukis and Nenes, 2007). AIM = Aerosol Inorganic Model calculated metastable growth for ammonium sulfate and ammonium nitrate at T=298K (Wexler and Clegg, 2002), Laboratory ammonium sulfate fit is $GF = 1.49 + 2.81 \cdot RH^{24.6}$ (with deliquescence at 80%) for bulk pure ammonium sulfate (Wise et al., 2003). All components are fit using Eq 6.

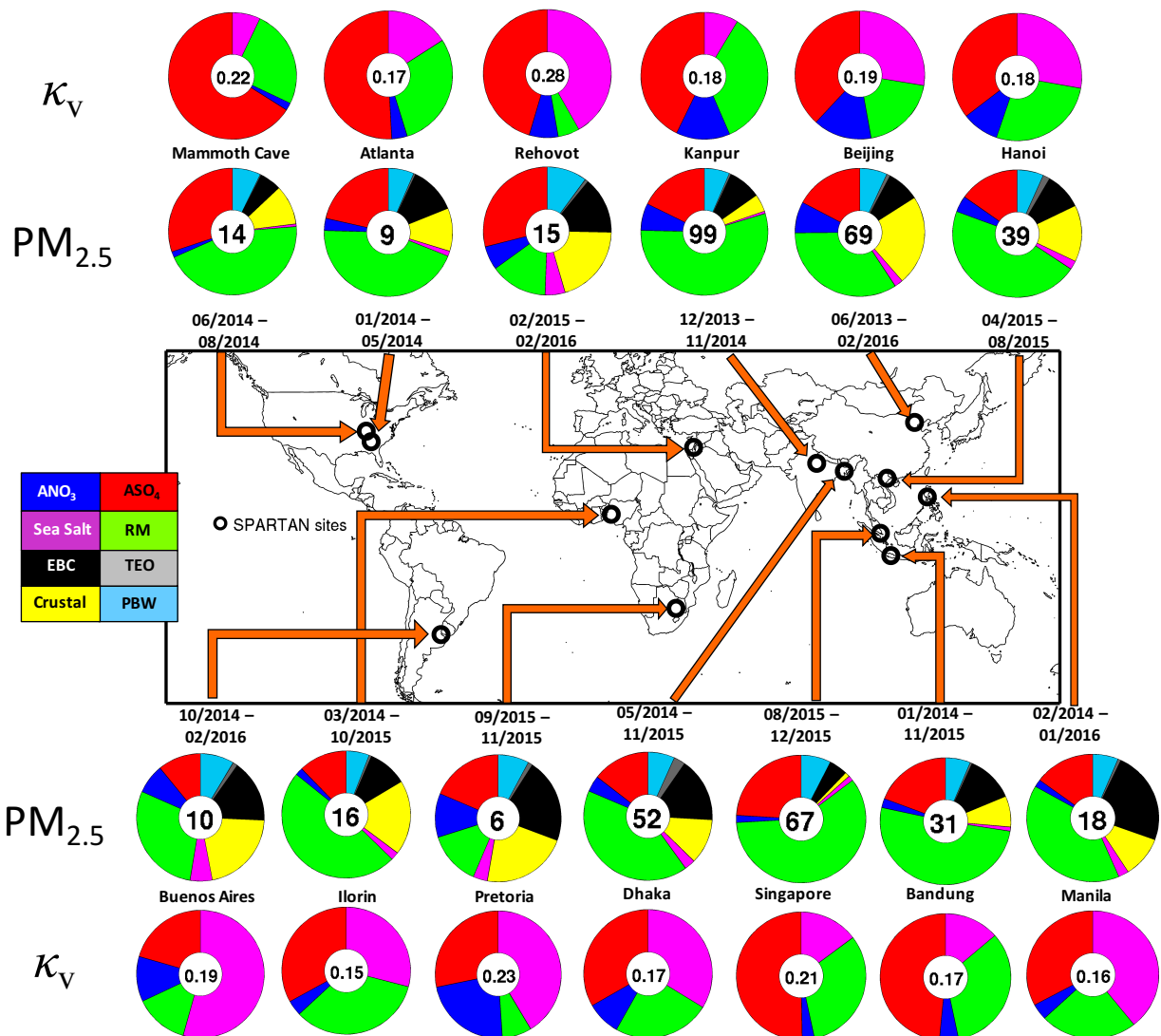




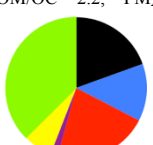
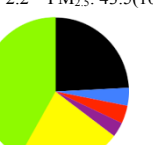

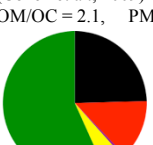

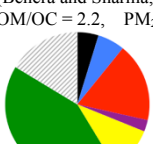
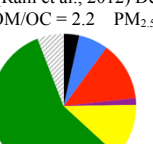
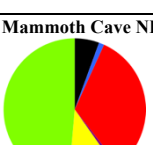
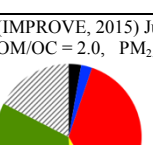
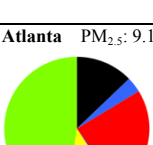
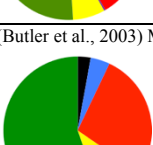
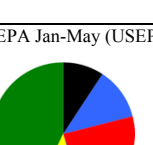
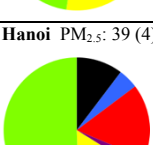
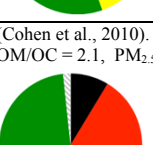


Figure 2: $PM_{2.5}$ mass (inner circle, $\mu g m^{-3}$) and composition mass fraction (filled colors) is shown in interior pie charts. Exterior pie charts contain site-mean κ_v surrounded by the relative contribution of PBW water at 35% RH.

Figure 3: Comparison of SPARTAN water-free aerosol composition with 11 collocated speciation studies. The numbers in parentheses show 1- σ deviations of averaged masses. The number of filters sampled is n . Dark green = organic, Light green = residual matter, black = equivalent black carbon, red = Ammoniated sulfate, blue = ammonium nitrate, purple = sea salt, yellow = crustal, and grey stripes = unknown. OM/OC ratios are from Philip et al. (2014b) and Canagaratna et al. (2015). Relative mass percentages are based on water-free aerosol components. SPARTAN percentages are renormalized to 100% after omission of species not found in comparison studies.

PM _{2.5} mass = $\mu\text{g m}^{-3}$ ($1\sigma/\sqrt{n}$), components = % (1σ)		
This study (total mass = $\mu\text{g m}^{-3}$)	Prior Study ($\mu\text{g m}^{-3}$)	Prior Study ($\mu\text{g m}^{-3}$)
Beijing PM _{2.5} : 69 (3), $n = 114$  <ul style="list-style-type: none"> 8.5 (10)% ANO₃, 19 (12)% ASO₄, 2.3 (3.3)% SS, 25 (14)% CM, 8.8 (5.3)% EBC, 37 (27)% RM 	(Yang et al., 2011) 2005-2006, OM/OC = 1.7, PM _{2.5} : 119(40)  <ul style="list-style-type: none"> 11 (7)% ANO₃, 17 (10)% ASO₄, 1.3 (0.6)% SS, 19 (3)% CM, 7 (5)% EC, 33 (16)% OM, 10 (10)% Unk 	(Oanh et al., 2006) 2001-2004, OM/OC = 1.7 PM _{2.5} : 136 (45)  <ul style="list-style-type: none"> 12 (1.5)% ANO₃, 20 (1.8)% ASO₄, 1.2 (1.2)% SS, 5 (3)% CM, 9 (7)% EBC, 29 (22)% OM, 24 (24)% Unk
Bandung PM _{2.5} : 31 (1), $n = 77$  <ul style="list-style-type: none"> 2.4 (1.4)% ANO₃, 21 (8)% ASO₄, 1.0 (0.3)% SS, 8.6 (4.1)% CM, 13 (4)% EBC, 55 (19)% RM 	(Oanh et al., 2006) 2001-2004, OM/OC = 2.2, PM _{2.5} : 45.5(10.6),  <ul style="list-style-type: none"> 13(4)% ANO₃, 21(3)% ASO₄, 1.6(0.2)% SS, 6.6(0.5)% CM, 19 (4)% EBC, 36(11)% RM 	(Lestari and Mauliadi, 2009) 2001- 2007, OM/OC = 2.2 PM _{2.5} : 43.5(10.5)  <ul style="list-style-type: none"> 4(6)% ANO₃, 4(4)% ASO₄, 3(2)% SS, 23(21)% CM, 24(14)% EBC, 42(35)% RM
Manila PM _{2.5} : 18 (1), $n = 63$  <ul style="list-style-type: none"> 1.8 (1.2)% ANO₃, 16 (9)% ASO₄, 2.9 (2.4)% SS, 11 (6)% CM, 25 (19)% EBC, 43 (21)% RM 	(Cohen et al., 2009) 2001-2007, OM/OC = 2.1, PM _{2.5} : 46 (19),  <ul style="list-style-type: none"> ANO₃ N/A 14 (9)% ASO₄, 0.6 (1.5)% SS, 5 (1.7)% CM, 25 (11)% EBC, 57(22)% OM, 	
Kanpur PM _{2.5} : 99 (9), $n = 33$  <ul style="list-style-type: none"> 7.4 (5.7)% ANO₃, 19 (13)% ASO₄, 0.7 (0.3)% SS, 4.8 (2.9)% CM, 9 (5.0)% EBC, 59 (35)% RM 	(Behera and Sharma, 2010) Oct. 2007 – Jan 2008, OM/OC = 2.2, PM _{2.5} : 172 (73),  <ul style="list-style-type: none"> 6.1 (1.3)% ANO₃, 18 (4)% ASO₄, 2.6 (0.6)% SS, 10 (3)% CM, 4.8 (1.1)% EC, 42 (9)% OM, 16 (10)% Unk 	(Ram et al., 2012) Dec 2008 – Feb 2009, OM/OC = 2.2 PM _{2.5} : 158 (47)  <ul style="list-style-type: none"> 6.6(4)% ANO₃, 13 (5)% ASO₄, 1.5 (0.9)% SS, 12 (6)% CM* 3 (1.1)% EC, 57 (23)% OM, 6 (24)% Unk *Assuming CM = [Ca]/0.034 (Wang, 2015)
Mammoth Cave NP PM _{2.5} : 13.6 (2), $n = 19$  <ul style="list-style-type: none"> 1.2 (1.0)% ANO₃, 33 (19)% ASO₄, 0.8 (0.8)% SS, 11 (11)% CM, 5.6 (3.2)% EBC, 49 (34)% RM 	(IMPROVE, 2015) June-Aug. 2014, OM/OC = 2.0, PM _{2.5} : 10.0 (5.8),  <ul style="list-style-type: none"> 2.4 (2.5)% ANO₃, 36 (17)% ASO₄, 0.3 (1.6)% SS, 7 (8)% CM, 3 (3)% EC, 34 (30)% OM, 17% Unk+H₂O 	
Atlanta PM _{2.5} : 9.1 (1), $n = 13$  <ul style="list-style-type: none"> 3.5 (1.2)% ANO₃, 23 (11)% ASO₄, 1.2 (1.2)% SS, 12 (4.7)% CM, 11 (2.6)% EBC, 48 (25)% RM 	(Butler et al., 2003) Mar. 1999 –2000 Feb, OM/OC = 2.0, PM _{2.5} : 24.2  <ul style="list-style-type: none"> 4 (0.2)% ANO₃, 28 (1.0)% ASO₄, 10 (0.8)% CM, 3 (0.2)% EC, 55 (5)% OM, 	EPA Jan-May (USEPA, 2015), OM/OC = 2.0 PM _{2.5} : 8.5  <ul style="list-style-type: none"> 12 (5)% ANO₃, 23 (15)% ASO₄, 1.4 (0.6)% SS 12 (5)% CM, 9.3 (5)% EC, 43 (36)% OM,
Hanoi PM _{2.5} : 39 (4), $n = 10$  <ul style="list-style-type: none"> 4.4 (1.1)% ANO₃, 17 (6)% ASO₄, 2.5 (0.6)% SS, 16 (15)% CM, 10 (5.8)% EBC, 51 (22)% RM 	(Cohen et al., 2010). 2001 –2008 OM/OC = 2.1, PM _{2.5} : 54 (33)  <ul style="list-style-type: none"> ANO₃ N/A 29 (20)% ASO₄, 0.6 (1.4)% SS 13 (7)% CM, 8 (3)% EBC, 40 (19)% OM, 2 (2)% Unk + ANO₃ 	

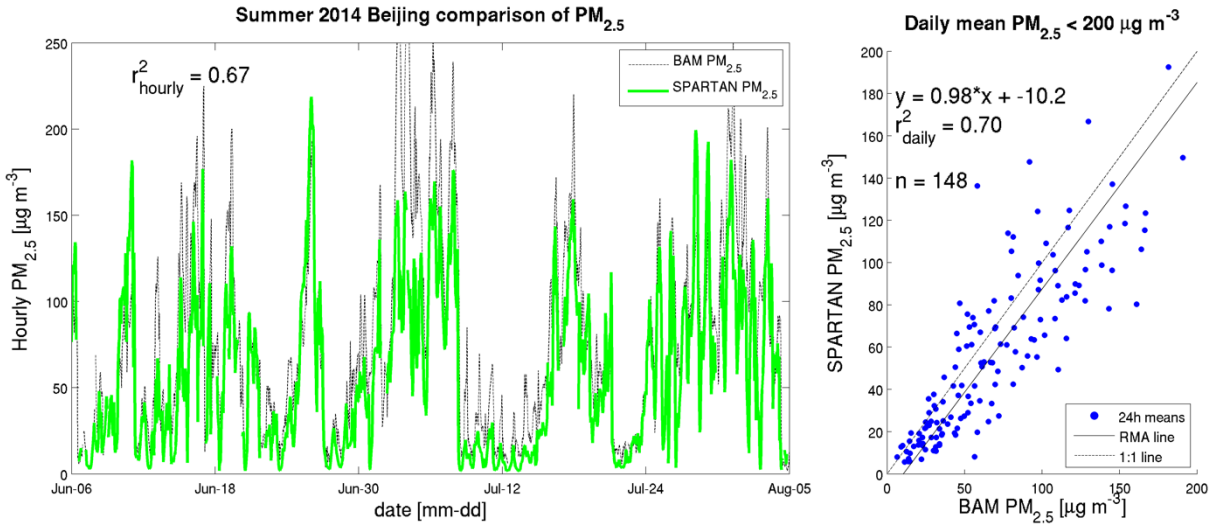


Figure 4: Left Hourly PM_{2.5} estimated from SPARTAN overlaid with a MetOne BAM-1020 (June-August 2014) at the Beijing US Embassy (15 km away). Right: 24-hour SPARTAN PM_{2.5} compared with BAM for the year 2014. Reduced major axis (RMA) slope and Pearson correlations for PM_{2.5} are given in inset.

Appendix:

Appendix A1:

Table A1: Hygroscopicity parameter κ , for various studies on organic material

κ , (OM)	Comments	Reference
0.045	Fitted to an aged organic mixture, subsaturated	(Varutbangkul et al., 2006)
0	IMPROVE network, subsaturated	(Hand and Malm, 2006)
0.10 ± 0.04	RH > 99%, fitted to SOA precursors	(Prenni et al., 2007)
-0.067 + 0.33(O:C)	Fitted, RH > 99%	(Jimenez et al., 2009)
0.29(O:C)	RH > 99%, 0.3 < O:C < 0.6	(Chang et al., 2010)
0.05	Best estimate from aged mixtures, subsaturated	(Dusek et al., 2011)
0.01 – 0.2	Field studies & smog chamber, subsaturated	(Duplissy et al., 2011)
0.16	RH > 99%	(Asa-Awuku et al., 2011)
0.05 – 0.13	Lab experiments, aged with H ₂ O ₂ and light; subsaturated	(Liu et al., 2012)
0.1	RH > 99%, D _{dry} < 100 nm	(Padró et al., 2012)
0.12ϵ_{WSOM}[#]	RH > 99%	(Lathem et al., 2013)
-0.005 + 0.19(O:C)	Fitted, RH > 99% 100 nm particles	(Rickards et al., 2013)
0.03, 0.1	HDTMA-measure, subsaturated	(Bezantakos et al., 2013)
0.1	Subsaturated	Selected for this study

[#] ϵ_{WSOM} = fraction of water-soluble organic material.

Appendix A2:

Dry aerosol scatter ($b_{sp,dry}$) is related to relative humidity (RH) by

$$b_{sp,dry} = \frac{b_{sp}(RH)}{f_v(RH)} \quad \text{Eq. A1}$$

Changes in scatter are also proportional to mass (Chow et al., 2006; Wang et al., 2010)

$$b_{sp,dry} = \alpha PM_{2.5,dry} \quad \text{Eq. A2}$$

where α ($m^2 g^{-1}$) is the mass scattering efficiency and a function of aerosol size distribution, effective radius, and dry composition. In this study we treat composition, density, and size distribution as constant over each of our 9-day intermittent sampling periods so that $\alpha \approx \langle \alpha \rangle_{9d}$. Under this assumption the predicted mass changes in low humidity (35% RH) are proportional to water-free aerosol scatter:

$$PM_{2.5,dry} = \langle PM_{2.5,dry} \rangle \frac{b_{sp,dry}}{\langle b_{sp,dry} \rangle} \quad \text{Eq. A3}$$

where $\langle \rangle$ indicates 9-day averages. The explicit compensation for aerosol water is then

$$[PM_{2.5,dry}] = \frac{\langle [PM_{2.5,dry}] \rangle}{\langle b_{sp}(RH)/f_v(RH) \rangle} \cdot \frac{b_{sp}(RH)}{f_v(RH)} \quad \text{Eq. A4}$$

where $[\]$ indicates concentration in $\mu g m^{-3}$. Uncertainties are a function of replicate weighing measurements ($\pm 4 \mu g$), flow volume ($\pm 10\%$), %RH (± 2.5), aerosol scatter ($\pm 5\%$), and κ_v (± 0.05).

$$\left(\frac{\delta[PM_{2.5,h}]}{[PM_{2.5,h}]} \right)^2 \approx \left(\frac{\delta PM_{2.5}}{PM_{2.5}} \right)^2 + \left(\frac{\delta V}{V} \right)^2 + \left(\frac{\delta b_{sp}}{b_{sp}} \right)^2 + \left(\frac{\delta f_v}{f_v} \right)^2 \quad \text{Eq. A5}$$

where

$$\left(\frac{\delta f_v}{f_v} \right)^2 = \frac{(f_v - 1)^2}{f_v^2} \left[\left(\frac{\delta \kappa}{\kappa} \right)^2 + \left(\frac{\delta RH}{RH \cdot (100 - RH)} \right)^2 \right] \quad \text{Eq. A6}$$

The average relative 2- σ $PM_{2.5}$ uncertainty was 26% for dry hourly predictions, increasing with higher RH cutoffs. A cut-off of RH = 80% has been applied to our data, above which hygroscopic uncertainties, as well as total water mass, dominate.