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20 **Abstract**

21

22 The Kathmandu Valley in Nepal is a bowl-shaped urban basin in the Himalayan foothills
23 with a serious problem of fine particulate air pollution that impacts local health and impairs
24 visibility. Particulate carbon concentrations have reached severe levels that threaten the
25 health of 3.5 million local residents. Moreover, snow and ice on the Himalayan mountains
26 are melting as a result of additional warming due to particulate carbon, especially high black
27 carbon concentrations. To date, the sources of the Valley's particulate carbon and the impacts
28 of different sources on particulate carbon concentrations are not well understood. Thus,
29 before an effective control strategy can be developed, these particulate carbon sources must
30 be identified and quantified. Our study has found that the four primary sources of particulate
31 carbon in the Kathmandu Valley during winter are brick kilns, motor vehicles, fugitive soil
32 dust, and biomass/garbage burning. Their source contributions are quantified using a recently
33 developed new multivariate receptor model SMP. In contrast to other highly polluted areas
34 such as China, secondary contribution is almost negligible in Kathmandu Valley. Brick kilns
35 (40%), motor vehicles (37%) and biomass/garbage burning (22%) have been identified as the
36 major sources of elemental carbon (black carbon) in the Kathmandu Valley during winter,
37 while motor vehicles (47%), biomass/garbage burning (32%), and soil dust (13%) have been
38 identified as the most important sources of organic carbon. Our research indicates that
39 controlling emissions from motor vehicles, brick kilns, biomass/garbage burning, and soil
40 dust is essential for the mitigation of the particulate carbon that threatens public health,
41 impairs visibility, and influences climate warming within and downwind from the
42 Kathmandu Valley. In addition, this paper suggests several useful particulate carbon
43 mitigation methods that can be applied to Kathmandu Valley and other areas in South Asia
44 with similar sources and high particulate carbon concentrations.

45

46 **Key words:** PM₁₀, Particulate carbon, Source apportionment, SMP model, Kathmandu

47

48 1. Introduction

49

50 The Kathmandu Valley in Nepal is a bowl-shaped basin surrounded by mountains that is
51 home to seven UNESCO world heritage monuments, and the nation's capital city. According
52 to the 2011 census, the population of the Valley doubled from 1.6 million in 2001 to the
53 current population of 2.5 million residents, as well as around 1 million transient residents
54 (CBS, 2001; CBS, 2013). Rapid but unplanned growth has led to urban sprawl, and even
55 more rapid growth in economic activities and the vehicle fleet, including the use of small,
56 mostly diesel-fired power generators. In addition, issues of power shortages have led to
57 increased use of the power generators, as well as increased use of biomass burning and low-
58 grade coal in over 110 traditional brick kilns in the Valley. This has resulted in highly
59 elevated PM₁₀ concentrations, which threatens the health of local residents, deteriorates
60 visibility, damages crops, and affects climate warming (Giri et al., 2006).

61 PM₁₀ is a multicomponent air pollutant. It consists of inorganic compounds, organic and
62 elemental carbon, and trace metals. Its chemical composition depends on emission source
63 types. Total particulate carbon consists of two components, elemental carbon (EC, sometimes
64 called soot or black carbon) and organic carbon (OC). Particulate carbon is ubiquitous in
65 ambient air because it is a byproduct of incomplete combustion of fossil fuel and wood
66 products. It is directly emitted by various stationary and mobile sources, and indirectly
67 derived in the atmosphere from oxidation of gaseous volatile organic compounds. Our
68 research focused on particulate carbon because it is a major component of PM₁₀ observed in
69 the Kathmandu Valley (Shakya et al, 2010). Moreover, EC is toxic, has adverse health effects,
70 causes premature death, and reduces visibility (US EPA, 2012; WHO, 2012). In addition, EC
71 is a major source of global warming because it absorbs solar radiation and warms the air
72 (Bond et al., 2013; Jacobson, 2004; Ramana et al., 2010; Ramanathan and Carmichael, 2008);
73 it also contributes to the accelerated Himalayan cryosphere melting. OC contains toxic
74 organic compounds (Callén et al., 2011; Lundstedt et al., 2007; Pickering, 1999) and light-
75 absorbing brown carbons that warm the air (Andreae and Gelencser, 2006; Feng et al., 2013).

76 Therefore, mitigating elevated particulate carbon (both EC and OC) concentrations is
77 critical to improving health, visibility, and climate impacts in the Kathmandu Valley. In order
78 to develop the most effective mitigation strategy and control measures for the reduction of
79 particulate carbon, it is essential to identify the sources of particulate carbon and to quantify
80 their source contributions. While a general overview of different sources in the Kathmandu
81 Valley is existent, the exact source contributions were not known or quantified (Aryal et al.,

82 2008; Shakya et al., 2010). Typically, detailed air quality modeling is necessary to understand
83 and evaluate the relationship between emission sources and ambient particulate carbon
84 concentrations. Air quality models require extensive input of emissions and meteorological
85 data. However, emissions inventory in the Kathmandu Valley is not well understood and the
86 wind patterns and pollution transport pathways in the Kathmandu Valley can be quite
87 complex (Panday et al., 2009; Regmi et al., 2003). In this case, source contributions can be
88 calculated by a multivariate receptor model which does not require uncertain emissions and
89 complex wind data, but only requires measured ambient data.

90 In this source apportionment study, recently developed multivariate receptor model SMP
91 (Solver for Mixture Problem; Kim, 2013) is applied to PM₁₀ filter samples collected in the
92 Kathmandu Valley between December 2012 and February 2013. Four primary sources and
93 one secondary source are identified and their contributions to the measured PM₁₀ mass and
94 particulate carbon concentrations are quantified. These findings are expected to provide an
95 important scientific basis for developing and implementing effective air pollution control
96 strategies and mitigation methods for the Kathmandu Valley. These findings may also be
97 applicable to other South Asian countries with similar conditions. Furthermore, this study can
98 help reduce particulate concentrations if the findings are incorporated and utilized in policy
99 decisions targeting the major particulate sources in the Kathmandu Valley and other South
100 Asian countries. Therefore, the results of this study have the potential to benefit not only
101 residents of the Kathmandu Valley, but also nearby South Asian countries and nations across
102 the globe through a reduction in transported particulate carbons emanating from this region
103 and the concomitant reduced impact on climate change.

104 This paper begins with a description of measurement methods, followed by a brief
105 description of the multivariate receptor model SMP. Next, the SMP model estimated source
106 compositions and source contributions are discussed and summarized.

107

108 **2. Methods**

109

110 **2.1 Ambient measurements**

111

112 *Sampling site*

113 Kathmandu Valley is surrounded by mountains and hilly areas, ranging from 0.5 to 1.5 km
114 above the valley floor (Figure 1). Diurnal variation of the wind in the Kathmandu Valley
115 during winter dry season can be characterized as relatively calm during night and morning,

116 with weak easterly or southeasterly winds drifting toward Bode bringing the plumes from
117 brick kilns to the east, while relatively strong westerly winds blow urban emissions from
118 Kathmandu city towards the sampling site in the afternoon and until the evening (Panday and
119 Prinn, 2009; Regmi et al., 2003). Details of the measurement program can be found in the
120 Supplementary Information section.

121

122 ***Sampling and chemical analysis of PM₁₀***

123 Twenty four-hour PM₁₀ filter samples were collected daily between December 2012 and
124 February 2013 using a PM₁₀ sequential sampler placed on the roof of a building (15 m above
125 ground) at the supersite Bode, which is approximately 5 km east of the edge of the
126 Kathmandu Valley (Figure 1). Filters were changed daily at 09:00 local time. PM₁₀ sampling
127 was conducted for two intensive measurement periods: the first intensive sampling period
128 was between December 21, 2012 and January 3, 2013, and the second sampling period was
129 between February 13 and 21, 2013. PM₁₀ data was chemically analyzed for OC, EC, ions, and
130 trace metals at the NIER laboratories. Details of sampling and chemical analysis can be found
131 in the Supplementary Information section.

132

133 **2.2 Multivariate receptor modeling**

134

135 Ambient particulate concentrations (C) measured on a filter can be expressed as a linear
136 sum of products of two unknown variables; source contribution (S), and source composition
137 (A). Estimating these two unknown variables from one known measured concentration is
138 referred to as multivariate receptor modeling in aerosol source apportionment studies.
139 Details of multivariate receptor modeling including the SMP model (Kim, 2013), can be
140 found in the Supplementary Information section and Kim et al (2015).

141

142 **3. Results**

143

144 **3.1 Characteristics of PM₁₀ data**

145

146 Samples of PM₁₀ at Bode were collected as previously stated during the following two
147 winter measurement periods: December 21, 2012 - January 3, 2013, and February 13 - 21,
148 2013. PM₁₀ mass concentrations in the Kathmandu Valley often reach significantly high
149 levels in winter dry season because of a relatively low wind speed and low inversion layer

150 height, and also the additional emission sources (such as brick kilns) which are operated in
151 winter and pre-monsoon season only (January-April). Sharma et al. (2012) reported the
152 highest BC concentrations, and the lowest average wind speed and precipitation in winter.
153 For this reason, two intensive measurement periods in winter in the Kathmandu Valley were
154 chosen to characterize high particulate matter and quantify its source contributions and
155 thereby aid in the development of effective control strategies. The average PM₁₀ mass and
156 chemical species concentrations for these two sampling periods are summarized in Table 1
157 and displayed in Figure 2. Metals are assumed to be present as their major oxides (Table 1
158 and Figure 2); the OC concentration is multiplied by 1.4 to account for hydrogen and oxygen
159 present in the HCs and then converted to the concentrations of organic carbonaceous material
160 (OM). Figure 2 also shows the daily variation of chemical species compositions, where the
161 unexplained portion of the measured concentration is the difference between the measured
162 PM₁₀ concentration and the sum of the chemical species concentrations. In general, the mass
163 closure shows that the sum of the chemical species compositions is less than the measured
164 PM₁₀ mass concentration. There is a single observation on December 27, 2012, when the sum
165 of the chemical species compositions is greater than measured PM₁₀ concentrations. As
166 shown in Table 1, average PM₁₀ mass concentration is 132.0 $\mu\text{g m}^{-3}$ and 121.8 $\mu\text{g m}^{-3}$ for the
167 first and second sampling period, respectively; it is 127.7 $\mu\text{g m}^{-3}$ for both periods combined.
168 Figure 2 shows that OM (32.2%) and trace metals (metals consisting mostly of crustal
169 components; 31.7%) are the major chemical components that explain more than 60% of the
170 average total PM₁₀ concentration for the entire measurement periods. EC (7.9%), sulfate
171 (5.5%), nitrate (2.5%), ammonium (2.2%), chloride (1.6%), and other cations (sodium,
172 potassium, magnesium, and calcium; 1.4%) consist of the remaining PM₁₀. However, 15 % of
173 the total PM₁₀ mass concentration is unexplained. Meanwhile, the following factors can cause
174 this discrepancy between the measured and the constructed PM₁₀ mass concentration:
175 measurement errors in the total PM₁₀ mass and/or individual chemical species concentrations;
176 use of a relatively small multiplying factor to convert OC to OM; evaporation of semi-
177 volatile compounds during sampling and/or storage; and relatively high water content. There
178 is only a small difference in the average PM₁₀ concentrations for two intensive measurement
179 periods (10 $\mu\text{g m}^{-3}$) but there is a significant difference in compositions (shown in Figure 2):
180 OM and metals are higher in the first measurement period while EC and sulfate are higher in
181 the second period. This difference in chemical species concentrations is reflected in the
182 model-estimated source contributions and discussed in the source contributions section below.

183 In the Kathmandu Valley, secondary inorganic ion concentrations are relatively low (10%)
184 compared to other high fine particulate pollution areas (Fine et al., 2008; Huang et al., 2014;
185 Kim et al., 2000), whereas particulate carbon and trace metals concentrations are high. High
186 particulate carbon concentrations of OM and EC, which are characteristic of the Kathmandu
187 Valley, imply that primary combustion-related and/or secondary sources are dominant. The
188 high level of metals implies a primary fugitive soil dust source. Although we can infer
189 potential particulate carbon sources from the measured concentrations, it is unlikely to infer
190 how much of the measured concentrations are contributed from each particulate carbon
191 source. It is also not feasible to distinguish between primary or secondary contributions.
192 Therefore, to identify the PM₁₀ sources and quantify their contributions, the newly developed
193 multivariate receptor model SMP is applied and its model-estimated source compositions and
194 source contributions are discussed in the next two sections of this paper.

195 Nineteen samples in total were collected during the two sampling periods, which may
196 appear to be small for a multivariate receptor modeling analysis. The number of samples
197 ordinarily deemed adequate for multivariate analysis is a matter of number of degrees of
198 freedom per variable. For a multivariate receptor modeling application that uses a
199 multivariate statistical method such as PCA or regression, the adequate number of samples
200 for measurement is generally determined by an approach as suggested by Henry (1984).
201 While the number of samples collected for this study is less than suggested by Henry (1984),
202 the multivariate model SMP is not a statistical model, but rather a mathematical model that
203 uses non-linear programming, which does not require the same larger sample size. Eighteen
204 chemical species and 19 samples for this study are therefore sufficient to estimate source
205 compositions for five sources of 18 chemical species (and source contributions for five
206 sources over 19 sampling days).

207

208 **3.2 Identification of sources and their source characterization**

209

210 In general, source identification is achieved by inspecting model-estimated source
211 compositions with *a priori* knowledge and experience, which is always a challenging task. In
212 this study, five sources of fugitive soil dust, brick kilns, biomass/garbage burning, secondary,
213 and motor vehicles are identified by examining source compositions (summarized in Table 2;
214 displayed in Figure 3). The sum of the source compositions for each source is less than 1 as
215 was expected (Table 2). This indicates that model-estimated source compositions satisfy the
216 underlying mass balance equation of the receptor modeling.

217 The first source in Figure 3 is soil dust because model-estimated compositions show
218 typical characteristics of a primary fugitive soil dust, including relatively high compositions
219 of Si, Fe, and OC, and some fractions of NO₃, Ca, Ti, and Mn. As shown in Table 2, Si
220 composition of soil dust is 0.25, which is consistent with the typical range of the Si
221 composition for the earth's crust (McDonough, 2001; Taylor, 1964). Characteristics of soil
222 dust generated from paved roads, unpaved roads, construction activities and disturbed open
223 areas are similar and generally undistinguishable in the receptor modeling. In this study,
224 therefore, these geological sources are treated as a single source category of soil dust.

225 The second source is brick kilns, characterized by relatively high compositions in EC, OC,
226 SO₄, Si, and Fe. Brick manufacturing uses sulfur containing coal, typically mixed with
227 biomass, as fuel to bake bricks. As a result, this activity releases a group of chemical species
228 associated with raw brick material and burning of sulfur containing fuel. Clay is a raw
229 material used for brick production, which has the same chemical compositions (Si, Fe, and
230 OC) as the fugitive soil dust source explained above. Burning of sulfur containing coal as fuel
231 to bake bricks is reflected in the estimated source compositions as relatively high fractions of
232 EC, OC, and SO₄.

233 Source compositions of the third source are characterized by high OC and relatively high
234 EC, NH₄, Cl, and a small amount of Zn. NH₄, Cl, and Zn are generally considered as marker
235 species of a waste burning source, and OC and EC are considered as markers for a biomass
236 burning source. These two groups of species appear together in the compositions of the third
237 source. This implies that these two groups are correlated and cannot be separated into two
238 individual sources with the current limited data, and/or these two sources are located so close
239 to each other that they behave as if they were a single source. Often the garbage fires in the
240 Kathmandu Valley have a combination of organic/farm waste, as well as paper and plastics.
241 Consequently, the third source is named the biomass/garbage burning source.

242 The fourth source is characterized by NO₃, SO₄, NH₄ and OC, and this implies a secondary
243 source. Secondary chemical species such as NH₄, NO₃, SO₄, and secondary organic aerosols
244 (SOA) are not directly emitted, but rather formed in the atmosphere by chemical reactions
245 from gaseous precursor compounds; these secondary species then get transported together to
246 the receptor sampling site. Therefore, from the receptor point of view these secondary species
247 appear to be coming from the same secondary source. The secondary source in the
248 Kathmandu Valley seems to be dominated by ammonium sulfate while the SOA fraction is
249 small and ammonium nitrate appears in a negligible amount. High ammonium sulfate
250 fractions in the secondary source may be associated with the SO₂ compound emitted from the

251 brick manufacturing that are scattered in the valley's agriculture fields, and NH₃ emitted from
252 agricultural activities.

253 The last source shown in Figure 3 is a motor vehicle source, which shows large fractions of
254 OC, EC, Si and Fe. Si, Fe and Ca are the marker species of fugitive soil dust as explained
255 above. In general, these soil components appear in the source compositions together with the
256 motor vehicle source because fugitive soil dust on the road is re-suspended in the air through
257 motor vehicle traffic. Then vehicle exhaust and fugitive soil dust are mixed in the air and
258 reach the receptor site at the same time. Therefore, from the receptor perspective, mixed
259 vehicle exhaust and fugitive soil dust are not differentiable and appears as a single motor
260 vehicle source. A study of emissions from on-road traffic fleets of motorcycles and public
261 transport vehicles (*e.g.*, buses, taxis, three-wheelers and vans) was conducted in 2010 in the
262 Kathmandu Valley (Shrestha et al., 2013). The Shrestha study found that diesel-powered
263 buses are a dominant contributor to PM, BC and OC emissions.

264

265 **3.3 Source contributions**

266

267 The comparison of measured and model-estimated PM₁₀ mass is shown in Figure 4.
268 Model-estimated PM₁₀ mass is a sum of the estimated source contributions from five sources;
269 measured and model-estimated total PM₁₀ mass shows a good correlation and satisfies the
270 total mass constraint FNPC5, as expected (Figure 4). The SMP model-estimated daily source
271 contributions are summarized in Table 3 and Figure 5. Average source contributions for each
272 sampling period and both periods together are summarized in Table 4 and displayed in
273 Figures 6a and 6b.

274 As explained in the previous section (Table 3 and Figure 5), the first and second sampling
275 periods display a distinct variation in the total PM₁₀ mass and chemical species
276 concentrations and consequently in their source contributions as well. OM and metals are
277 observed to be high in the first period whereas EC and sulfate are high in the second period.
278 This difference in chemical compositions of OM, EC, metals and sulfate between the two
279 periods is reflected in the model-estimated source contributions (Table 1 and Figure 2).
280 Motor vehicles and biomass/garbage burning sources are the main sources of primary OM
281 whereas brick kilns, motor vehicles and biomass/garbage burning sources are the major
282 sources of EC. Sulfate is the major component of the secondary source and metals are the
283 dominant fraction of fugitive soil dust. Table 3 and Figure 5 show that the source contribution
284 is solely from motor vehicles on December 27, 2012. As explained in section 3.2, the motor

285 vehicle source defined in this study is a mix of motor vehicle exhaust and fugitive soil dust.
286 As a result, motor vehicle source contributions are not only from motor vehicles but also
287 from soil dust. This is shown in the measured chemical species concentrations (Figure 2a).
288 OM, EC and metals in Figure 2a explain a majority of the measured concentration, which is
289 explained solely by the motor vehicle source with no room for contributions from other
290 sources. Also, as explained in Section 3.1, the sum of the chemical species concentrations is
291 greater than the measured PM₁₀ concentration on December 27, 2012, which could have
292 resulted in overestimation of the motor vehicle contribution or underestimation of other
293 source contributions. As shown in Figure 6a, source contributions from motor vehicles and
294 soil dust sources are higher in the first period. In particular, the soil dust contribution is
295 almost three times higher in the first period. Figure 6a also shows that the brick kilns and
296 secondary source contribution is higher in the second period. These source contributions are
297 consistent with the variation of measured species concentrations between the two periods and
298 explain them well.

299 As shown in Figure 6b, local primary source contributions of soil dust (45.12 $\mu\text{g m}^{-3}$, 35%),
300 motor vehicles (43.39 $\mu\text{g m}^{-3}$, 34%), biomass/garbage burning (28.78 $\mu\text{g m}^{-3}$, 23%), and brick
301 kilns (7.86 $\mu\text{g m}^{-3}$, 6%) explain almost all (98%) of the measured PM₁₀ concentration in the
302 first period; only 2% is explained by secondary sources. In the second period, brick kilns
303 (32.53 $\mu\text{g m}^{-3}$, 28%) are revealed as the largest primary source contributor to the measured
304 PM₁₀ concentration, with motor vehicles (29.98 $\mu\text{g m}^{-3}$, 26%) as the second largest
305 contributor, followed by biomass/garbage burning (28.55 $\mu\text{g m}^{-3}$, 24%), and soil dust (14.22
306 $\mu\text{g m}^{-3}$, 12%). Notably, the soil dust contribution was considerably less in the second
307 sampling period. Factors that could have influenced the decrease of soil dust contribution are
308 described as follows. Two precipitation events were observed on February 16 and 17, 2013
309 during the second sampling period. The rain effect appears to have lasted through February
310 18, 2013, which still shows a relatively low total mass concentration. Furthermore, wind
311 speed and wind direction for the two sampling periods showed slightly different patterns. In
312 the first period, higher wind speed and lower precipitation were observed, which are
313 consistent with the findings of Sharma et al. (2012), and wind direction was more westerly
314 which normally brings polluted air masses from urban areas of Kathmandu metropolitan city
315 and Lalitpur sub-metropolitan city to the sampling site. Therefore, lower wind speed and
316 more precipitation during the second sampling period resulted in a decreased soil dust
317 contribution. In addition, winds blew with no dominant wind direction in the second

318 sampling period and this could have resulted in a lower soil dust contribution. It is worth
319 stating here that the sampling site is located in a mixed agricultural-residential setting. The
320 primary and secondary source contributions in the second period explain 90% and 10%,
321 respectively of the measured PM₁₀ concentration. Ninety-five percent of the average PM₁₀
322 concentration during both periods is attributed to local primary sources: motor vehicles
323 (37.74 µg m⁻³, 31%), soil dust (32.11 µg m⁻³, 26%), biomass/garbage burning (28.68 µg m⁻³,
324 23%), and brick kilns (18.25 µg m⁻³, 15%), while 5% is attributed to a secondary source.

325 The particulate carbon comprises 26 to 62% of the total measured PM₁₀ concentration in
326 the Kathmandu Valley. Therefore, it is important that the sources of particulate OC and EC
327 are identified and their contributions quantified. Figure 6c illustrates OC source contributions
328 to the measured OC concentrations for the first sampling period, second period and also both
329 periods combined. As shown in Figure 6c, relative contributions of motor vehicles and soil
330 dust sources to OC decreased in the second period while those of brick kilns,
331 biomass/garbage burning and secondary contributions increased. Motor vehicles (15.96 µg m⁻³
332 ³, 51%) and biomass/garbage burning (9.59 µg m⁻³, 30%) explain 81% of the measured OC
333 and the rest is explained by soil dust (5.17 µg m⁻³, 16%), brick kilns (0.63 µg m⁻³, 2%), and
334 secondary source (0.32 µg m⁻³, 1%) in the first sampling period. In the second sampling
335 period, motor vehicles (11.03 µg m⁻³, 41%) and biomass/garbage burning (9.51 µg m⁻³, 36%)
336 sources contribute 77% to the OC mass concentration while the remaining OC concentration
337 is explained by brick kilns (2.60 µg m⁻³, 10%), secondary source (1.77 µg m⁻³, 7%), and soil
338 dust (1.63 µg m⁻³, 6%). For both period combined, motor vehicles (13.88 µg m⁻³, 47%) and
339 biomass/garbage burning (9.56 µg m⁻³, 32%) sources contribute almost 80% of the average
340 OC mass concentration. The remaining OC is explained by soil dust (3.68 µg m⁻³, 13%),
341 brick kilns (1.46 µg m⁻³, 5%), and secondary source (0.93 µg m⁻³, 3%).

342 Figure 6d shows EC source contributions to the measured EC concentration for the first
343 period, second period and both periods, respectively. Relative contributions of EC from
344 motor vehicles and biomass/garbage burning sources decreased in the second period whereas
345 those of brick kilns contribution increased by a factor of almost 3. In the first period, motor
346 vehicles (4.31 µg m⁻³, 51%) and biomass/garbage burning (2.29 µg m⁻³, 27%) explain 78% of
347 the measured EC while the remaining EC is explained by brick kilns (1.78 µg m⁻³, 21%) and
348 soil dust (0.07 µg m⁻³, 1%), and there is no secondary contribution. In the second period,
349 brick kilns (7.38 µg m⁻³, 58%), motor vehicles (2.98 µg m⁻³, 24%) and biomass/garbage

350 burning ($2.28 \mu\text{g m}^{-3}$, 18%) sources explain all of the EC mass concentration. For the average
351 EC concentrations of both periods, three major sources for EC are brick kilns ($4.14 \mu\text{g m}^{-3}$,
352 40%), motor vehicles ($3.75 \mu\text{g m}^{-3}$, 37%), and biomass/garbage burning ($2.29 \mu\text{g m}^{-3}$, 22%).
353 Soil dust contribution to EC is a negligible amount of only 1%. As explained in the previous
354 paragraph, the first and second periods show large variations of particulate carbon
355 contributions. In general, most of the brick kilns operate from January to April each year.
356 However, during the sampling periods of this study, it was observed that firing of brick kilns
357 increased suddenly from January 1, 2013. Firing of all 110 plus brick kilns in the valley was
358 completed and all kilns became operational by January 20, 2013 (personal communication
359 with the Chairperson of the Federation of Nepalese Brick Industries). High EC and sulfate
360 concentrations observed in the second period (Table 1 and Figure 2) match well with high
361 brick kiln contributions as shown in Figures 6a and 6d.

362

363 **4. Discussion**

364

365 Analysis of the Kathmandu Valley PM_{10} source apportionment study indicates that four
366 primary local sources (motor vehicles, soil dust, biomass/garbage burning, and brick kilns)
367 are responsible for 95% of the PM_{10} concentrations. It also shows that motor vehicles,
368 biomass/garbage burning, and soil dust explain more than 90% of observed OC, whereas
369 brick kilns, motor vehicles, and biomass/garbage burning sources contribute to 99% of EC.
370 Therefore, emission control strategies to mitigate particulate carbon and PM_{10} in the
371 Kathmandu Valley should focus on emission reductions from these four primary sources to
372 be most effective. Compared to the control of secondary particulates, primary particulates are
373 relatively easy to control because emission reductions from primary emission sources would
374 linearly reduce ambient particulate concentrations.

375 The biomass/garbage burning source inferred from the SMP model is a mix of two sources
376 mostly derived from the burning of either garbage and agricultural residue and/or other
377 biomass on the street or in residences or in the industries such as brick factories. To reduce
378 emissions from open garbage burning which is common in the valley especially in winter,
379 open burning of both household wastes, including garden waste and agricultural residue
380 burning should be banned, and a garbage collection system made more efficient in the valley.

381 As brick kilns are furnaces used to bake bricks by burning coal and/or wood, these are one
382 of the major sources of EC (and not just in the Kathmandu Valley, but over large parts of

383 South Asia). Joshi and Dudani (2008) found emissions from brick kilns to adversely impact
384 the health of children attending a nearby school. This demonstrates the importance of
385 reducing the emission of EC, a toxic chemical and a primary indicator of adverse health
386 effects, from brick kilns. Options to reduce emissions from the existing bull's trench brick
387 kilns include the following: optimizing airflow and fuel in existing kilns to improve
388 combustion efficiency such as adoption of zig-zag firing; switching to more capital intensive
389 but cleaner brick making technologies; and switching to alternative building materials to
390 reduce the dependence on baked clay bricks.

391 Motor vehicles are the most important source of OC in the Kathmandu Valley. Reducing
392 emissions from vehicles can be accomplished in a number of ways, such as an improved
393 inspection and proper engine maintenance, retrofitting the existing diesel-powered vehicles
394 by fitting diesel particulate filter (DPF), cracking down on overloading of trucks and buses,
395 and designing a transportation network that prioritizes mass public transport and non-
396 motorized transport (rather than the current growth in motor cycles and cars).

397 Finally, reduction of fugitive soil dust, contributing 13% of the OC, is also essential to
398 improve organic particulate carbon air quality in the Kathmandu Valley. In general, fugitive
399 soil dust is generated from disturbed open areas, construction activities, uncovered open
400 storage piles and paved or unpaved roads. This source is relatively easy to control by
401 applying Best Available Control Measures (BACM; SCAQMD, 2005). For example, to
402 stabilize fugitive soil dust from disturbed open areas, control measures such as proper paving
403 of the road, foot path and parking lots, re-vegetation, chemical stabilizer or water can be
404 applied to the disturbed areas. Control measures such as watering or sweeping throughout
405 construction sites can be used to stabilize soils from construction activities. Likewise, street
406 cleaning and watering can be used to reduce soil dust emissions from the paved roads, as well
407 as paving or applying chemical stabilizer on unpaved roads.

408 Mitigation of four primary particulate carbon sources in the Kathmandu Valley will
409 substantially improve the health of residents, improve visibility, and slow down local and
410 regional climate change. Successful mitigation of particulate carbon in the Kathmandu Valley
411 would also set a positive example for other South Asia countries that are experiencing
412 similarly high primary particulate carbon concentrations.

413

414 **5. Summary and Conclusions**

415

416 The international SusKat-ABC air pollution measurement campaign took place in the
417 Kathmandu Valley and surrounding regions in Nepal between December 2012 and June 2013.
418 PM₁₀ filter sampling was conducted at the Bode super site during the following two periods:
419 December 21, 2012 to January 3, 2013, and February 13, 2013 to February 21, 2013. For both
420 sampling periods, high particulate carbon and low secondary inorganic ions in the
421 Kathmandu Valley are found in the PM₁₀ filter samples. The average PM₁₀ mass
422 concentrations for these two measurement periods are 132.0 $\mu\text{g m}^{-3}$ and 121.8 $\mu\text{g m}^{-3}$ for the
423 first and second period, respectively, and 127.7 $\mu\text{g m}^{-3}$ for both periods combined.

424 The two measurement periods show a distinct variation in PM₁₀ chemical species
425 compositions. Despite very similar PM₁₀ mass concentrations, OC and trace metals are higher
426 in the first sampling period while EC and sulfate are higher in the second sampling period.
427 Large variation in the chemical compositions led to large variations in source contributions.
428 Five sources (fugitive soil dust, brick kilns, biomass/garbage burning, secondary, and motor
429 vehicles) are identified. Source contributions from motor vehicles and soil dust sources are
430 higher in the first period and brick kilns and secondary source contributions are higher in the
431 second period. For both measurement periods, 95% of the average PM₁₀ concentration is
432 attributed to local primary sources, motor vehicles (31%), soil dust (26%), biomass/garbage
433 burning (23%), and brick kilns (15%), while only 5% is attributed to a secondary source.

434 Motor vehicles and biomass/garbage burning sources are the main sources of primary OC
435 whereas brick kilns, motor vehicles and biomass/garbage burning sources are the major
436 sources of EC. Ninety-percent of the average OC concentration of both sampling periods is
437 explained by the three primary local sources, motor vehicles (47%), biomass/garbage-burning
438 (32%) and soil dust (13%), while brick kilns (5%) and secondary source (3%) explained the
439 remaining OC concentration. The average EC concentration of both periods is attributed to
440 the following three major sources: brick kilns (40%), motor vehicles (37%), and
441 biomass/garbage burning (22%). The soil dust contribution to EC (1%) was a negligible
442 amount.

443 In this study, it was not possible to differentiate the motor vehicles between gasoline and
444 diesel vehicles. Similarly, the biomass/garbage burning source was not able to be separated
445 into biomass burning and garbage burning sources. In future studies, it would be crucial to
446 measure individual organic compounds along with OC/EC, ions and metals. Such individual
447 organic compounds can be used as tracers for specific sources. This would likely enable us to
448 separate contributions from gasoline and diesel vehicles. Such a study might also help

449 elucidate the specific source contributions for biomass and garbage burning sources.
450 Therefore, future study with more organic compounds as tracers would greatly help better
451 quantification of relative contributions of these sources, and aid the establishment of effective
452 strategies and actions to control particulate matters (particularly carbonaceous aerosols) in the
453 Kathmandu Valley.

454

455

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553

554 **Table 1.** Average chemical species concentrations ($\mu\text{g m}^{-3}$) of PM_{10} measured in the
 555 Kathmandu Valley.

	1 st period	2 nd period	Both Periods
OM	44.12	36.87	41.06
EC	8.33	12.57	10.11
NH_4^+	1.68	4.35	2.80
NO_3^-	3.89	2.34	3.23
SO_4^{2-}	3.47	11.84	7.00
Cl^-	1.99	2.25	2.10
Other Cations	1.95	1.66	1.82
Metals	48.21	29.70	40.41
Unexplained	18.34	20.21	19.13
Total Mass	131.97	121.77	127.68

556

557 **Table 2.** SMP model-estimated source compositions for each source.

	Soil Dust	Brick Kiln	Biomass/Garbage Burning	Secondary	Motor Vehicle
NH ₄	0.0059	0.0582	0.0246	0.1577	0.0000
NO ₃	0.0371	0.0000	0.0464	0.0003	0.0208
SO ₄	0.0073	0.1548	0.0206	0.5001	0.0089
Na	0.0040	0.0010	0.0078	0.0000	0.0000
Cl	0.0037	0.0247	0.0547	0.0000	0.0042
K	0.0080	0.0133	0.0108	0.0000	0.0010
CA	0.0088	0.0097	0.0145	0.0000	0.0032
OC	0.1145	0.0798	0.3333	0.1476	0.3679
EC	0.0016	0.2268	0.0797	0.0000	0.0994
Si	0.2506	0.1090	0.0301	0.0000	0.1654
Ti	0.0069	0.0026	0.0111	0.0000	0.0000
Mn	0.0022	0.0000	0.0099	0.0000	0.0000
Fe	0.0470	0.0139	0.0068	0.0000	0.0213
Ni	0.0012	0.0000	0.0085	0.0000	0.0000
Cu	0.0013	0.0000	0.0085	0.0000	0.0000
Zn	0.0016	0.0002	0.0109	0.0000	0.0000
Br	0.0007	0.0002	0.0074	0.0000	0.0000
Pb	0.0011	0.0000	0.0076	0.0000	0.0000
Sum	0.5035	0.6942	0.6932	0.8057	0.6921

558

559 **Table 3.** SMP model-estimated source contributions ($\mu\text{g m}^{-3}$) for each source.

	Soil Dust	Brick Kiln	Biomass/Garbage Burning	Secondary	Motor Vehicle
21-Dec-2012	78.27	2.58	21.38	5.96	24.51
22-Dec-2012	22.06	5.48	37.08	8.29	29.92
23-Dec-2012	98.01	27.74	33.88	0.00	25.13
24-Dec-2012	53.46	8.95	22.77	1.85	43.01
25-Dec-2012	32.45	4.54	33.94	1.43	35.17
26-Dec-2012	57.90	2.07	11.27	1.84	48.22
27-Dec-2012	0.00	0.00	0.00	0.00	150.58
28-Dec-2012	47.79	0.00	43.13	2.27	28.19
29-Dec-2012	65.18	10.03	48.10	1.24	0.00
02-Jan-2013	31.30	9.31	28.87	0.87	38.41
03-Jan-2013	9.88	15.77	36.14	0.00	54.13
13-Feb-2013	0.00	69.52	11.15	0.00	90.53
14-Feb-2013	39.60	35.43	30.15	10.80	60.99
15-Feb-2013	9.01	12.85	11.04	9.70	44.37
16-Feb-2013	0.00	11.66	7.43	4.04	9.14
17-Feb-2013	0.00	22.13	10.20	11.71	20.25
18-Feb-2013	3.68	25.03	28.22	14.77	14.52
20-Feb-2013	24.83	33.77	76.08	22.42	0.00
21-Feb-2013	36.66	49.83	54.11	22.40	0.00

560

561 **Table 4.** Average source contributions ($\mu\text{g m}^{-3}$) for each period and both periods.

	Soil Dust	Brick Kiln	Biomass/Garbage Burning	Secondary	Motor Vehicle
1 st Period	45.12	7.86	28.78	2.16	43.39
2 nd Period	14.22	32.53	28.55	11.98	29.98
Both Periods	32.11	18.25	28.68	6.29	37.74

562

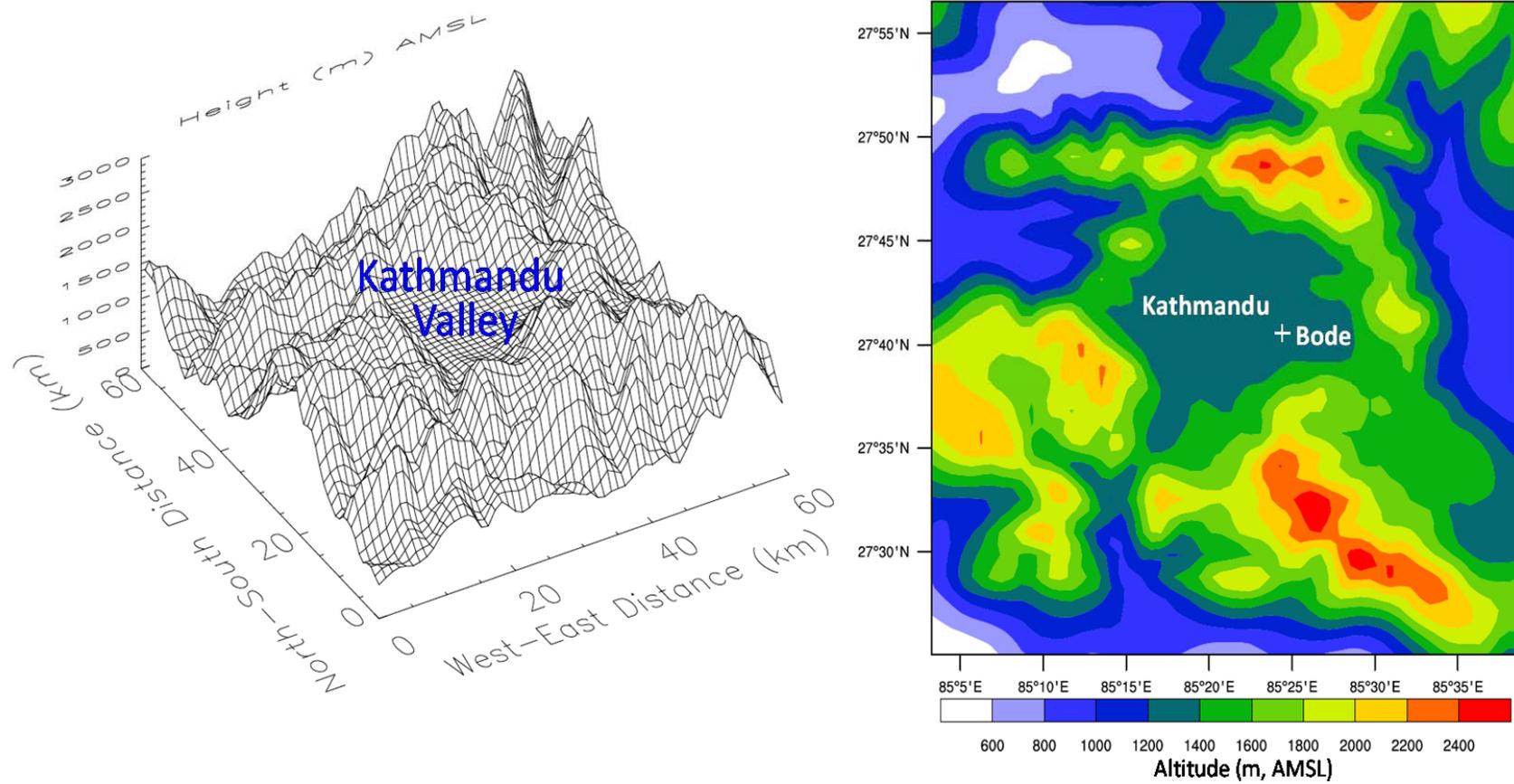


Figure 1. A topographic map of Kathmandu valley and its surroundings. The Bode station is located in the eastern part of the Kathmandu Valley.

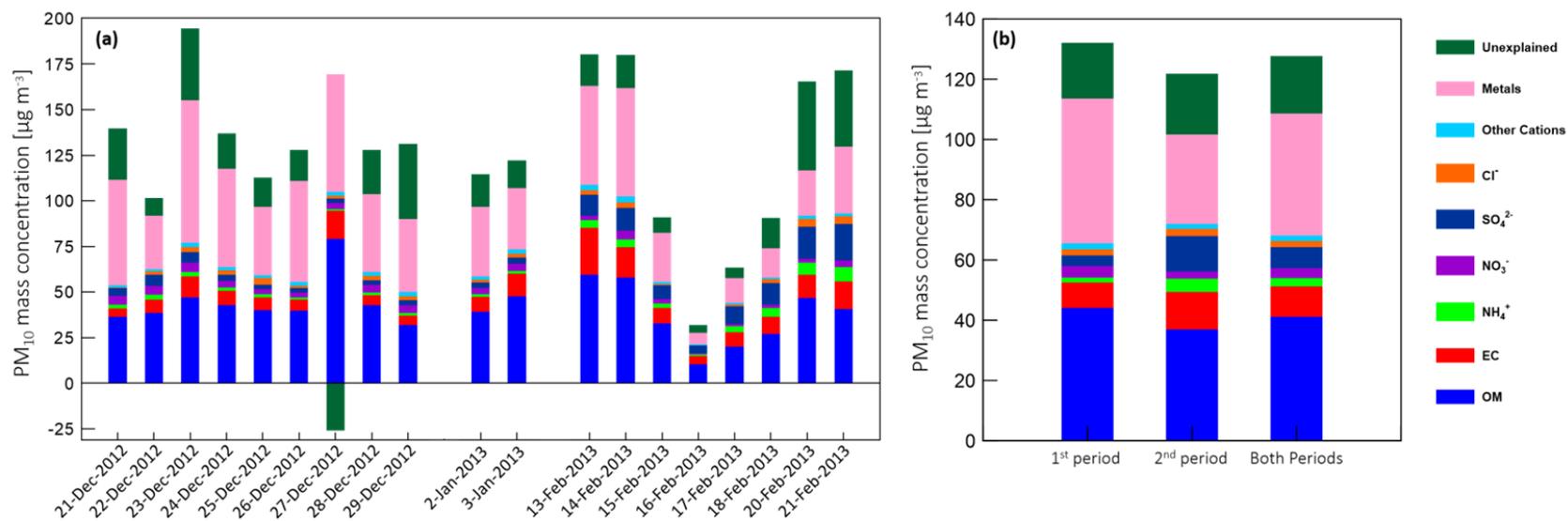


Figure 2. (a) Daily variation of PM₁₀ chemical species composition, and (b) average PM₁₀ chemical species composition measured in the Kathmandu Valley for each intensive measurement period and both periods combined.

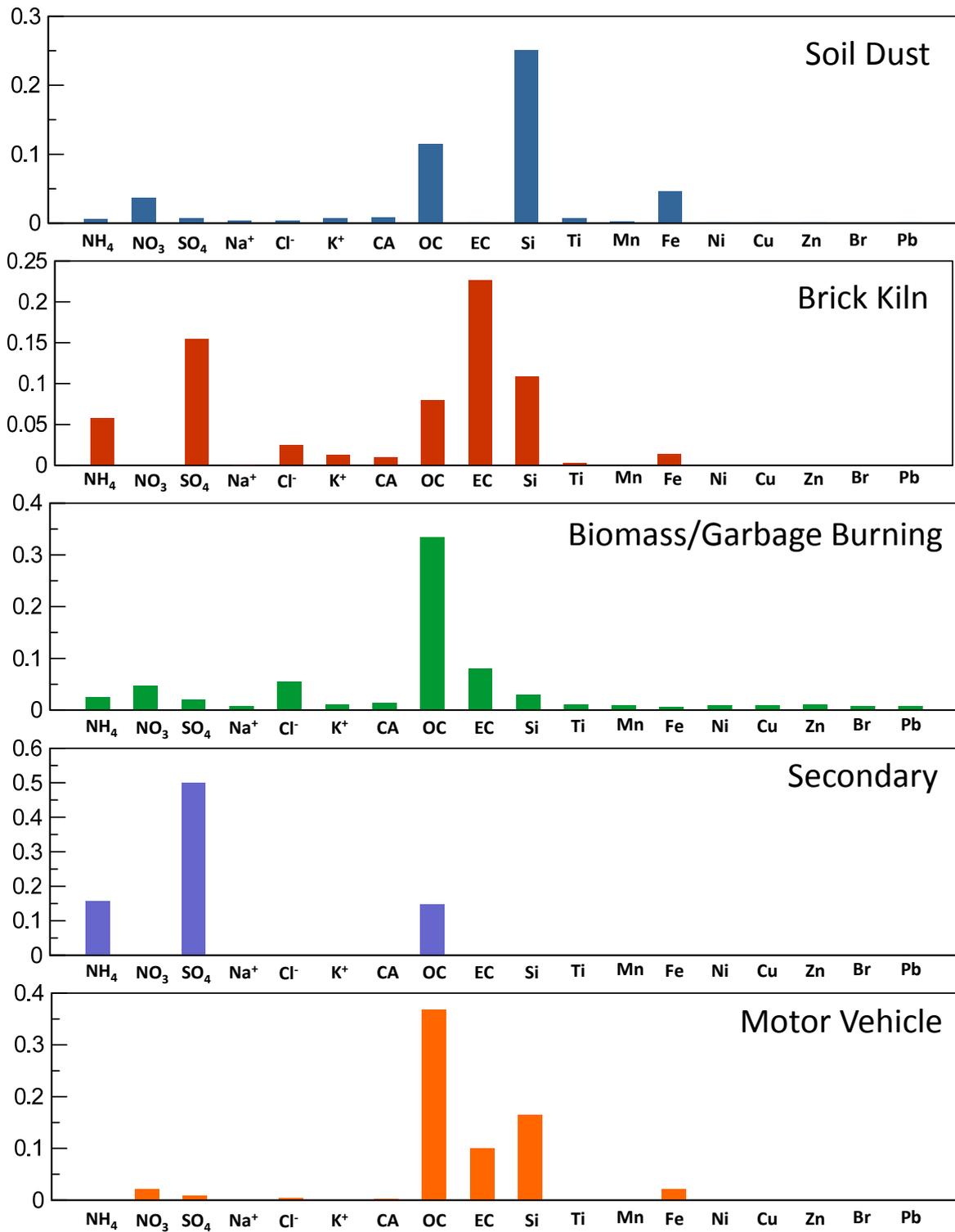


Figure 3. SMP model-estimated source compositions for each source.

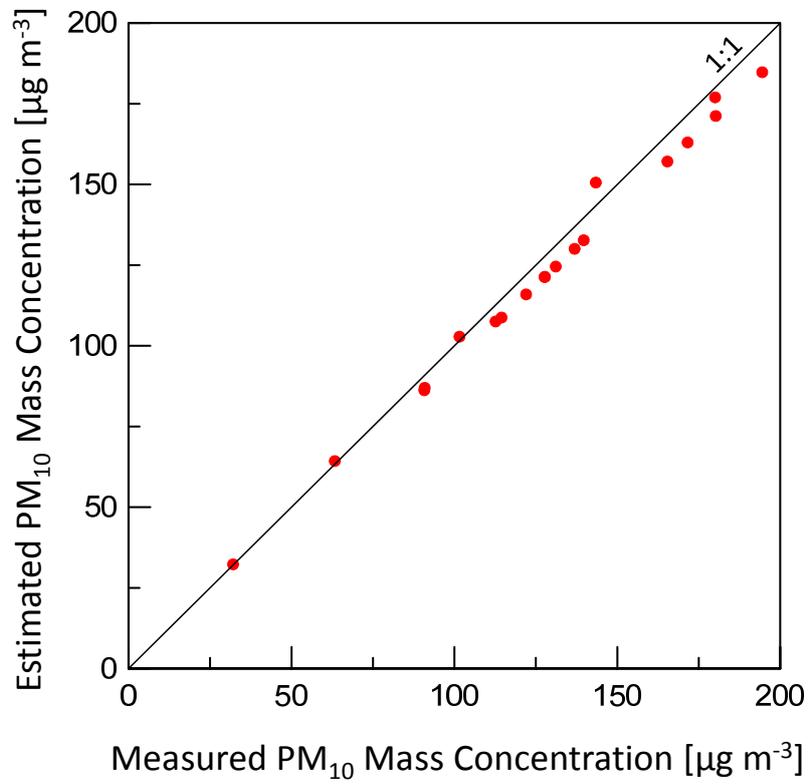


Figure 4. Intercomparison of measured and SMP model-estimated PM₁₀ mass concentrations (µg m⁻³).

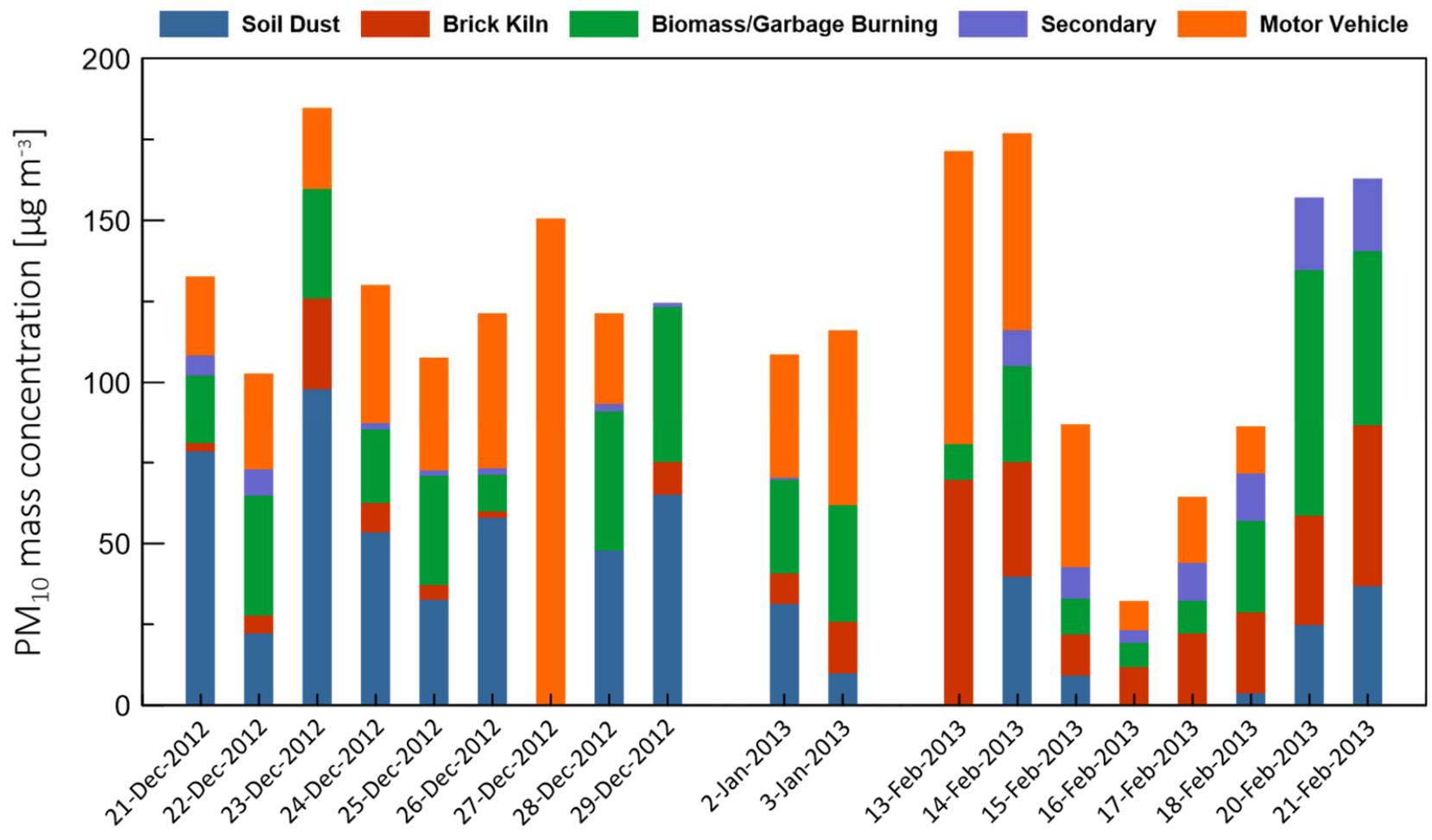


Figure 5. Daily variation of SMP model-estimated source contributions for each source.

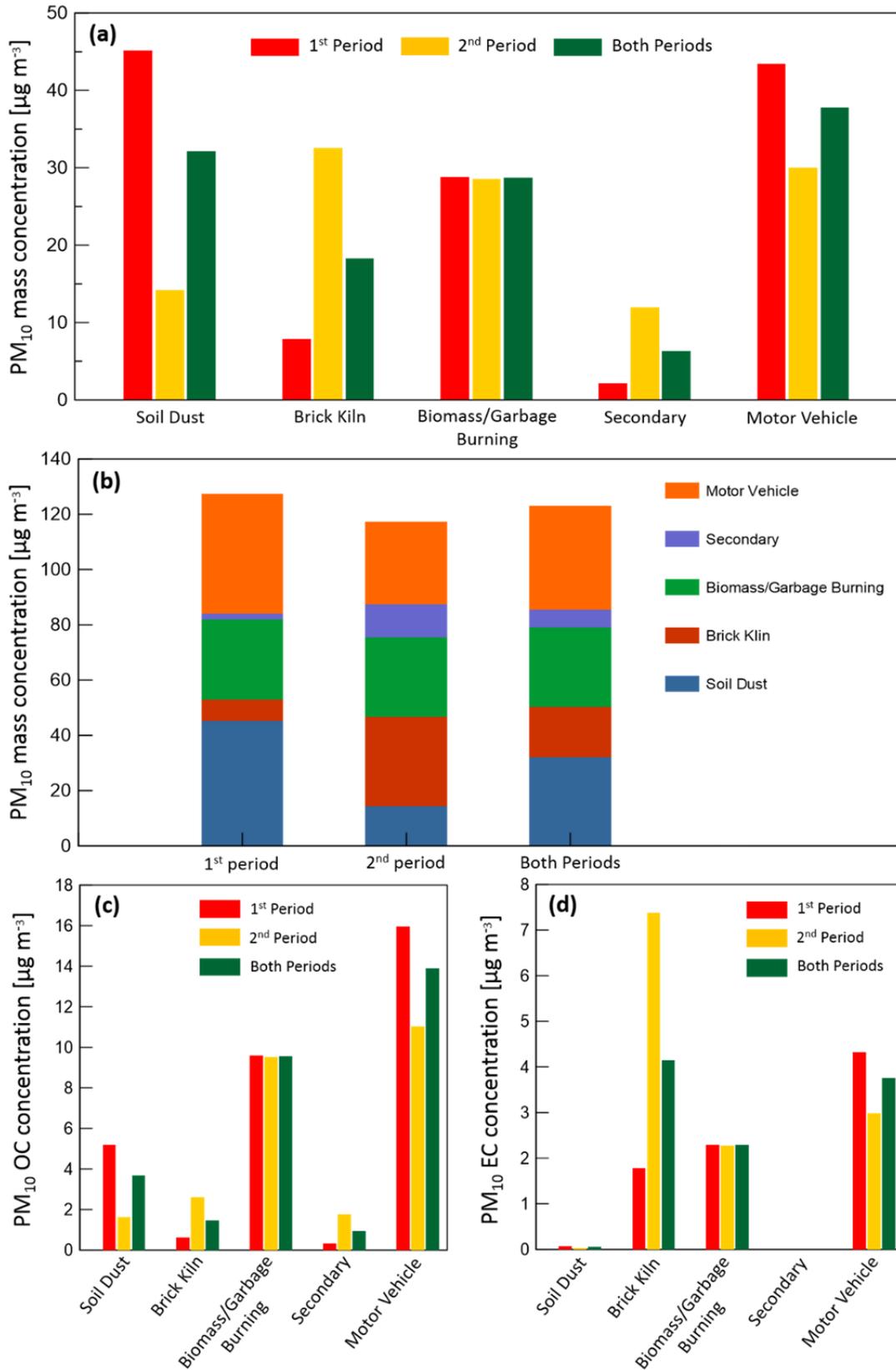


Figure 6. SMP model-estimated source contributions for average (a) PM₁₀ mass for each source, (b) PM₁₀ mass for each period, (c) organic carbon, and (d) elemental carbon.

Supplementary Information

“Source apportionment of PM₁₀ mass and particulate carbon in the Kathmandu Valley, Nepal” by Kim et al.

■ Multivariate receptor modeling

Ambient particulate concentrations measured on a filter can be expressed as a mixture of concentrations from an unknown number of unknown sources of particulate matters. As shown in matrix equation (1) below, measured ambient particulate concentrations are expressed as a linear sum of products of two unknown variables S (source apportionments, source contributions or source strengths) and A (source compositions, source profiles or source matrix).

$$C = SA \quad (1)$$

where C is a matrix of m rows of measured ambient concentrations of n columns of chemical species and typically measured in $\mu\text{g m}^{-3}$, S is a matrix of m rows of source contributions from sources of p columns in $\mu\text{g m}^{-3}$, and A is a matrix of p rows of sources of n columns of source compositions, which are the mass fractions of each chemical species for each source.

In equation (1), if the source compositions matrix A is known, a widely used regression-based receptor model, Chemical Mass Balance (CMB) model (Watson et al., 1984) can be used to determine source contributions matrix S. In contrast, if the source compositions matrix A is unknown, unavailable or uncertain, then a multivariate receptor model is generally applied to estimate S as well as A from the ambient particulate concentrations C only. Details of the multivariate receptor modeling can be found in Kim et al. (2015).

■ Source apportionment by new multivariate receptor model SMP

The new multivariate receptor model SMP (Kim, 2013) was recently developed based on primal-dual interior point nonlinear programming, and it was successfully applied to the source apportionment of roadside particulate polycyclic aromatic hydrocarbon (PAHs; Kim et al., 2015). As explained in the previous section, the multivariate receptor model attempts to estimate two unknown variables in equation (1) from one known variable C and is inherently ill-posed (Henry, 1987). In other words, many different but equally same pairs of S and A are

35 possible in the sense that the matrix multiplication of S and A will produce the measured data
36 C within the allowance of some measurement error. To restrict the feasible solution region
37 into a smaller one, known physical constraints are usually imposed in the model. In an
38 aerosol source apportionment study, Henry (1987) identified five fundamental natural
39 physical constraints (FNPCs), which are all minimum physical constraints that must be
40 imposed and satisfied in the model. If any one of the five FNPCs is not implemented, the
41 model results are questionable and untrustworthy. For a description of the five FNPCs, refer
42 to Henry (1987) and Kim (2013). The SMP model has implemented all five FNPCs; therefore,
43 the SMP-estimated source compositions and source contributions always satisfy all five
44 FNPCs and consequently, always guarantee physically sound results. Other multivariate
45 models such as the PMF model have neglected the implementation of all five FNPCs, which
46 can result in questionable results.

47

48

49 ■ **Measurement program**

50 The emissions that pollute the Kathmandu Valley's air have many different sources, such
51 as motor vehicle exhaust, smoke from brick kilns, dust from badly-maintained roads, and
52 garbage-burning, along with the transport of regional emissions into the valley. However,
53 their relative contribution to air quality degradation in the Kathmandu Valley and
54 surrounding area has not been well quantified yet. In order to understand the details of air
55 pollution in the Kathmandu Valley area and surrounding regions, the Institute for Advanced
56 Sustainability Studies (IASS) and the International Centre for Integrated Mountain
57 Development (ICIMOD) led the SusKat-ABC (Sustainable Atmosphere for the Kathmandu
58 Valley - project Atmospheric Brown Cloud) international field campaign and research
59 endeavor from December 2012 to June 2013 measuring air pollution and meteorological
60 parameters at several sites in the Kathmandu Valley and the surrounding Himalayan foothill
61 region, one of the most polluted but least sampled regions of the world. Over 20 local and
62 international research groups from 9 countries participated in the SusKat campaign, which
63 was endorsed by Project ABC of UNEP (United Nations Environment Programme), making
64 this the second-largest international air pollution measurement campaign ever undertaken in
65 South Asia. It has provided the most in depth air pollution data to date for the Kathmandu
66 Valley and surrounding region. During the SusKat-ABC campaign, the National Institute of
67 Environmental Research (NIER) of Korea collected daily PM₁₀ filter samples at Bode (the
68 supersite of the campaign) in the central part of the Kathmandu Valley. This site is in

69 residential-agricultural area, with about 10 brick kilns within a ca. 2 km radius of the site. It
70 receives polluted air outflows from three major cities in the valley: Kathmandu Metropolitan
71 city, Lalitpur Sub-metropolitan city, and Bhaktapur municipality. It is approximately 5 km to
72 the east of the Tribhuvan International Airport.

73

74

75 ■ Sampling and chemical analysis of PM₁₀

76 The National Institute of Environmental Research (NIER) of Korea deployed a PMS-103
77 sampling system from APM Engineering to collect particulate matter for an aerodynamic
78 diameter of less than or equal to 10 microns. Flow rate of the PM₁₀ sampler was maintained
79 at 16.7 $\ell \text{ min}^{-1}$ and 24-hour sampling was started daily at 9:00 a.m. local time. PM₁₀
80 sampling was conducted for two intensive measurement periods: (1) the first intensive
81 measurement period was between December 21, 2012 and January 3, 2013, and (2) the
82 second intensive measurement period was between February 13 and 21, 2013. PM₁₀ data was
83 then chemically analyzed for OC, EC, ions, and trace metals at the NIER laboratories.

84 Total mass and 33 species (Si, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Br, Rb,
85 Sr, Sn, Cs, Ba, Hg, Pb, organic carbon (OC), elemental carbon (EC), NO_3^- , SO_4^{2-} , NH_4^+ , Na^+ ,
86 Cl^- , K^+ , Mg^{2+} , and Ca^{2+}) were analyzed. Total mass was determined gravimetrically
87 following the USEPA CFR 58 Appendix L method as collected on Teflon filters (PT47-EP).
88 Filters were maintained at constant temperature and relative humidity of 20 °C and 35%,
89 respectively for 24 hours before weighing them by an automated filter weighing system
90 microbalance. The concentrations of 23 trace elements collected on Teflon filters (PT47-EP)
91 were determined by energy-dispersive X-ray fluorescence analyzer following the USEPA IO-
92 3.6 method. Three concentration levels on Aerosol membrane (Nucleipore) were used as
93 standard materials for 36 trace elements. Relative standard deviation (RSD) and method
94 detection limit (MDL) were calculated, and it was less than 10% and 0.2-5.9 ng m^{-3} ,
95 respectively.

96 Water-soluble ionic species (NO_3^- , SO_4^{2-} , NH_4^+ , Na^+ , Cl^- , K^+ , Mg^{2+} , and Ca^{2+}) were
97 extracted from ZefluorTM supported PTFE filters and analyzed by ion chromatography
98 following the USEPA IO-4.1 method. For QA/QC analysis, Multi-Component Anion Mix 18
99 Ion Chromatography Standard (IC-MAN-18-R1-1) and Multi-Component Cation Mix 2 Ion
100 Chromatography Standard (IC-MCA-02-1) from AccuStandard were used for anion and
101 cation standard, respectively. Three concentration levels of standard materials were

102 repeatedly analyzed for RSD and MDL calculation. RSD was less than 1% and MDL for
103 major ions of NO_3^- , SO_4^{2-} , and NH_4^+ are $0.014 \mu\text{g m}^{-3}$, $0.073 \mu\text{g m}^{-3}$, and $0.005 \mu\text{g m}^{-3}$,
104 respectively.

105 Quartz fiber filters were used to collect samples and analyzed for OC and EC. Quartz
106 filters were pretreated at 850°C for 4 hours before they are deployed for sampling. After the
107 24-hour sampling was completed, quartz filters were sealed and kept in a refrigerator until
108 filters were analyzed by a thermal optical transmittance carbon analyzer following the
109 USEPA NIOSH-5040 method. Four concentration levels of standard material injected on
110 quartz filters were analyzed repeatedly for three times and RSD and MDL were calculated.
111 RSD for total carbon was 3.6 and MDL for OC and EC were $0.258 \mu\text{g m}^{-3}$ and $0.028 \mu\text{g m}^{-3}$,
112 respectively.

113

114

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