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Source apportionment of PM₁₀ mass and particulate carbon in the Kathmandu Valley, Nepal

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HIGHLIGHTS

• High particulate carbon and low secondary inorganic ion levels are characteristic of PM10 data in the Kathmandu Valley.

• Motor vehicles and biomass/garbage burning sources are the main sources of primary organic carbon.

• Whereas brick kilns, motor vehicles and biomass/garbage burning sources are the major sources of elemental carbon.

A R T I C L E I N F O

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ABSTRACT

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

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1. Introduction

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The Kathmandu Valley in Nepal is a bowl-shaped basin surrounded by mountains that is home to seven UNESCO world







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heritage monuments, and the nation's capital city. According to the 2011 census, the population of the Valley doubled from 1.6 million in 2001 to the current population of 2.5 million residents, as well as around 1 million transient residents (CBS, 2001, 2013). Rapid but unplanned growth has led to urban sprawl, and even more rapid growth in economic activities and the vehicle fleet, including the use of small, mostly diesel-fired power generators. In addition, issues of power shortages have led to increased use of the power generators, as well as increased use of biomass burning and low-grade coal in over 110 traditional brick kilns in the Valley. This has resulted in highly elevated PM₁₀ concentrations, which threatens the health of local residents, deteriorates visibility, damages crops, and affects climate warming (Giri et al., 2006).

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

Therefore, mitigating elevated particulate carbon (both EC and OC) concentrations is critical to improving health, visibility, and climate impacts in the Kathmandu Valley. In order to develop the most effective mitigation strategy and control measures for the reduction of particulate carbon, it is essential to identify the sources of particulate carbon and to quantify their source contributions. While a general overview of different sources in the Kathmandu Valley is existent, the exact source contributions were not known or quantified (Aryal et al., 2008; Shakya et al., 2010). Typically, detailed air quality modeling is necessary to understand and evaluate the relationship between emission sources and ambient particulate carbon concentrations. Air quality models require extensive input of emissions and meteorological data. However, emissions inventory in the Kathmandu Valley is not well understood and the wind patterns and pollution transport pathways in the Kathmandu Valley can be quite complex (Panday et al., 2009; Regmi et al., 2003). In this case, source contributions can be calculated by a multivariate receptor model which does not require uncertain emissions and complex wind data, but only requires measured ambient data.

In this source apportionment study, recently developed multivariate receptor model SMP (Solver for Mixture Problem; Kim, 2013) is applied to PM_{10} filter samples collected in the Kathmandu Valley between December 2012 and February 2013. Four primary sources and one secondary source are identified and their contributions to the measured PM_{10} mass and particulate carbon concentrations are quantified. These findings are expected to provide an important scientific basis for developing and implementing effective air pollution control strategies and mitigation methods for the Kathmandu Valley. These findings may also be applicable to other South Asian countries with similar conditions. Furthermore,

this study can help reduce particulate concentrations if the findings are incorporated and utilized in policy decisions targeting the major particulate sources in the Kathmandu Valley and other South Asian countries. Therefore, the results of this study have the potential to benefit not only residents of the Kathmandu Valley, but also nearby South Asian countries and nations across the globe through a reduction in transported particulate carbons emanating from this region and the concomitant reduced impact on climate change.

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

2. Methods

2.1. Ambient measurements

2.1.1. Sampling site

Kathmandu Valley is surrounded by mountains and hilly areas, ranging from 0.5 to 1.5 km above the valley floor (Fig. 1). Diurnal variation of the wind in the Kathmandu Valley during winter dry season can be characterized as relatively calm during night and morning, with weak easterly or southeasterly winds drifting toward Bode bringing the plumes from brick kilns to the east, while relatively strong westerly winds blow urban emissions from Kathmandu city towards the sampling site in the afternoon and until the evening (Panday and Prinn, 2009; Regmi et al., 2003). Details of the measurement program can be found in the Supplementary Information section.

2.1.2. Sampling and chemical analysis of PM_{10}

Twenty four-hour PM_{10} filter samples were collected daily between December 2012 and February 2013 using a PM_{10} sequential sampler placed on the roof of a building (15 m above ground) at the supersite Bode, which is approximately 5 km east of the edge of the Kathmandu Valley (Fig. 1). Filters were changed daily at 09:00 local time. PM_{10} sampling was conducted for two intensive measurement periods: the first intensive sampling period was between December 21, 2012 and January 3, 2013, and the second sampling period was between February 13 and 21, 2013. PM_{10} data was chemically analyzed for OC, EC, ions, and trace metals at the NIER laboratories. Details of sampling and chemical analysis can be found in the Supplementary Information section.

2.2. Multivariate receptor modeling

Ambient particulate concentrations (C) measured on a filter can be expressed as a linear sum of products of two unknown variables; source contribution (S), and source composition (A). Estimating these two unknown variables from one known measured concentration is referred to as multivariate receptor modeling in aerosol source apportionment studies. Details of multivariate receptor modeling including the SMP model (Kim, 2013), can be found in the Supplementary Information section and Kim et al. (2016).

3. Results

3.1. Characteristics of PM₁₀ data

Samples of PM_{10} at Bode were collected as previously stated during the following two winter measurement periods: December 21, 2012 - January 3, 2013, and February 13 - 21, 2013. PM_{10} mass concentrations in the Kathmandu Valley often reach significantly high levels in winter dry season because of a relatively low wind



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

speed and low inversion layer height, and also the additional emission sources (such as brick kilns) which are operated in winter and pre-monsoon season only (January–April). Sharma et al. (2012) reported the highest BC concentrations, and the lowest average wind speed and precipitation in winter. For this reason, two intensive measurement periods in winter in the Kathmandu Valley were chosen to characterize high particulate matter and quantify its source contributions and thereby aid in the development of effective control strategies. The average PM₁₀ mass and chemical species concentrations for these two sampling periods are summarized in Table 1 and displayed in Fig. 2. Metals are assumed to be present as their major oxides (Table 1 and Fig. 2); the OC concentration is multiplied by 1.4 to account for hydrogen and oxygen present in the HCs and then converted to the concentrations of organic carbonaceous material (OM). Fig. 2 also shows the daily variation of chemical species compositions, where the unexplained portion of the measured concentration is the difference between the measured PM₁₀ concentration and the sum of the chemical species concentrations. In general, the mass closure shows that the sum of the chemical species compositions is less than the measured PM₁₀ mass concentration. There is a single observation

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

	1st period	2nd 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

on December 27, 2012, when the sum of the chemical species compositions is greater than measured PM₁₀ concentrations. As shown in Table 1, average PM_{10} mass concentration is 132.0 µg m⁻³ and 121.8 μ g m⁻³ for the first and second sampling period, respectively; it is 127.7 μ g m⁻³ for both periods combined. Fig. 2 shows that OM (32.2%) and trace metals (metals consisting mostly of crustal components; 31.7%) are the major chemical components that explain more than 60% of the average total PM₁₀ concentration for the entire measurement periods. EC (7.9%), sulfate (5.5%), nitrate (2.5%), ammonium (2.2%), chloride (1.6%), and other cations (sodium, potassium, magnesium, and calcium; 1.4%) consist of the remaining PM_{10} . However, 15% of the total PM_{10} mass concentration is unexplained. Meanwhile, the following factors can cause this discrepancy between the measured and the constructed PM₁₀ mass concentration: measurement errors in the total PM₁₀ mass and/or individual chemical species concentrations; use of a relatively small multiplying factor to convert OC to OM; evaporation of semi-volatile compounds during sampling and/or storage; and relatively high water content. There is only a small difference in the average PM₁₀ concentrations for two intensive measurement periods (10 μ g m⁻³) but there is a significant difference in compositions (shown in Fig. 2): OM and metals are higher in the first measurement period while EC and sulfate are higher in the second period. This difference in chemical species concentrations is reflected in the model-estimated source contributions and discussed in the source contributions section below.

In the Kathmandu Valley, secondary inorganic ion concentrations are relatively low (10%) compared to other high fine particulate pollution areas (Fine et al., 2008; Huang et al., 2014; Kim et al., 2000), whereas particulate carbon and trace metals concentrations are high. High particulate carbon concentrations of OM and EC, which are characteristic of the Kathmandu Valley, imply that primary combustion-related and/or secondary sources are dominant. The high level of metals implies a primary fugitive soil dust source. Although we can infer potential particulate carbon sources from the measured concentrations, it is unlikely to infer how much of the



Fig. 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.

measured concentrations are contributed from each particulate carbon source. It is also not feasible to distinguish between primary or secondary contributions. Therefore, to identify the PM₁₀ sources and quantify their contributions, the newly developed multivariate receptor model SMP is applied and its model-estimated source compositions and source contributions are discussed in the next two sections of this paper.

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

3.2. Identification of sources and their source characterization

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

The first source in Fig. 3 is soil dust because model-estimated compositions show typical characteristics of a primary fugitive soil dust, including relatively high compositions of Si, Fe, and OC, and some fractions of NO₃, Ca, Ti, and Mn. As shown in Table 2, Si composition of soil dust is 0.25, which is consistent with the typical range of the Si composition for the earth's crust (McDonough, 2001; Taylor, 1964). Characteristics of soil dust generated from paved roads, unpaved roads, construction activities and disturbed open areas are similar and generally undistinguishable in the

receptor modeling. In this study, therefore, these geological sources are treated as a single source category of soil dust.

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

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

The fourth source is characterized by NO₃, SO₄, NH₄ and OC, and this implies a secondary source. Secondary chemical species such as NH₄, NO₃, SO₄, and secondary organic aerosols (SOA) are not directly emitted, but rather formed in the atmosphere by chemical reactions from gaseous precursor compounds; these secondary species then get transported together to the receptor sampling site. Therefore, from the receptor point of view these secondary species appear to be coming from the same secondary source. The secondary source in the Kathmandu Valley seems to be dominated by ammonium sulfate while the SOA fraction is small and ammonium nitrate appears in a negligible amount. High ammonium sulfate fractions in the secondary source may be associated with the SO₂ compound emitted from the brick manufacturing that are scattered in the valley's agriculture fields, and NH₃ emitted from agricultural activities.

The last source shown in Fig. 3 is a motor vehicle source, which shows large fractions of OC, EC, Si and Fe. Si, Fe and Ca are the marker species of fugitive soil dust as explained above. In general, these soil components appear in the source compositions together with the motor vehicle source because fugitive soil

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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



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

dust on the road is re-suspended in the air through motor vehicle traffic. Then vehicle exhaust and fugitive soil dust are mixed in the air and reach the receptor site at the same time. Therefore, from the receptor perspective, mixed vehicle exhaust and fugitive soil dust are not differentiable and appears as a single motor vehicle source. A study of emissions from on-road traffic fleets of motorcycles and public transport vehicles (*e.g.*, buses, taxis, three-wheelers and vans) was conducted in 2010 in the Kathmandu Valley (Shrestha et al., 2013). The Shrestha study found that diesel-powered buses are a dominant contributor to PM, BC and OC emissions.

3.3. Source contributions

The comparison of measured and model-estimated PM_{10} mass is shown in Fig. 4. Model-estimated PM_{10} mass is a sum of the estimated source contributions from five sources; measured and model-estimated total PM_{10} mass shows a good correlation and satisfies the total mass constraint FNPC5, as expected (Fig. 4). The SMP model-estimated daily source contributions are summarized in Table 3 and Fig. 5. Average source contributions for each sampling period and both periods together are summarized in Table 4 and displayed in Fig. 6a and b.

As explained in the previous section (Table 3 and Fig. 5), the first and second sampling periods display a distinct variation in the total PM₁₀ mass and chemical species concentrations and consequently in their source contributions as well. OM and metals are observed to be high in the first period whereas EC and sulfate are high in the second period. This difference in chemical compositions of OM. EC. metals and sulfate between the two periods (Table 1 and Fig. 2) is reflected in the model-estimated source contributions. Motor vehicles and biomass/garbage burning sources are the main sources of primary OM whereas brick kilns, motor vehicles and biomass/ garbage burning sources are the major sources of EC. Sulfate is the major component of the secondary source and metals are the dominant fraction of fugitive soil dust. Table 3 and Fig. 5 show that the source contribution is solely from motor vehicles on December 27, 2012. As explained in Section 3.2, the motor vehicle source defined in this study is a mix of motor vehicle exhaust and fugitive soil dust. As a result, motor vehicle source contributions are not



Fig. 4. Intercomparison of measured and SMP model-estimated PM_{10} mass concentrations (µg $m^{-3}\text{)}.$

only from motor vehicles but also from soil dust. This is shown in the measured chemical species concentrations (Fig. 2a). OM, EC and metals in Fig. 2a explain a majority of the measured concentration, which is explained solely by the motor vehicle source with no room for contributions from other sources. Also, as explained in Section 3.1, the sum of the chemical species concentrations is greater than the measured PM₁₀ concentration on December 27, 2012, which could have resulted in overestimation of the motor vehicle contribution or underestimation of other source contributions. As shown in Fig. 6a, source contributions from motor vehicles and soil dust sources are higher in the first period. In particular, the soil dust contribution is almost three times higher in the first period. Fig. 6a also shows that the brick kilns and secondary source contribution is higher in the second period. These source contributions are consistent with the variation of measured species concentrations between the two periods and explain them well.

As shown in Fig. 6b, local primary source contributions of soil dust (45.12 μ g m⁻³, 35%), motor vehicles (43.39 μ g m⁻³, 34%), biomass/garbage burning (28.78 μ g m⁻³, 23%), and brick kilns (7.86 μ g m⁻³, 6%) explain almost all (98%) of the measured PM₁₀ concentration in the first period; only 2% is explained by secondary sources. In the second period, brick kilns (32.53 μ g m⁻³, 28%) are revealed as the largest primary source contributor to the measured PM_{10} concentration, with motor vehicles (29.98 µg m⁻³, 26%) as the second largest contributor, followed by biomass/garbage burning $(28.55 \ \mu g \ m^{-3}, 24\%)$, and soil dust $(14.22 \ \mu g \ m^{-3}, 12\%)$. Notably, the soil dust contribution was considerably less in the second sampling period. Factors that could have influenced the decrease of soil dust contribution are described as follows. Two precipitation events were observed on February 16 and 17, 2013 during the second sampling period. The rain effect appears to have lasted through February 18, 2013, which still shows a relatively low total mass concentration. Furthermore, wind speed and wind direction for the two sampling periods showed slightly different patterns. In the first period, higher wind speed and lower precipitation were observed, which are consistent with the findings of Sharma et al. (2012), and wind direction was more westerly which normally brings polluted air masses from urban areas of Kathmandu metropolitan city and Lalitpur sub-metropolitan city to the sampling site. Therefore, lower wind speed and more precipitation during the second sampling period resulted in a decreased soil dust contribution. In addition, winds blew with no dominant wind direction in the second sampling period and this could have resulted in a lower soil dust contribution. It is worth stating here that the sampling site is located in a mixed agricultural-residential setting. The primary and secondary source contributions in the second period explain 90% and 10%, respectively of the measured PM₁₀ concentration. Ninetyfive percent of the average PM₁₀ concentration during both periods is attributed to local primary sources: motor vehicles $(37.74 \ \mu g \ m^{-3})$, 31%), soil dust (32.11 μ g m⁻³, 26%), biomass/garbage burning (28.68 μ g m⁻³, 23%), and brick kilns (18.25 μ g m⁻³, 15%), while 5% is attributed to a secondary source.

The particulate carbon comprises 26–62% of the total measured PM_{10} concentration in the Kathmandu Valley. Therefore, it is important that the sources of particulate OC and EC are identified and their contributions quantified. Fig. 6c illustrates OC source contributions to the measured OC concentrations for the first sampling period, second period and also both periods combined. As shown in Fig. 6c, relative contributions of motor vehicles and soil dust sources to OC decreased in the second period while those of brick kilns, biomass/garbage burning and secondary contributions increased. Motor vehicles (15.96 µg m⁻³, 51%) and biomass/garbage burning (9.59 µg m⁻³, 30%) explain 81% of the measured OC and the rest is explained by soil dust (5.17 µg m⁻³, 16%), brick kilns (0.63 µg m⁻³, 2%), and secondary source (0.32 µg m⁻³, 1%) in the

Table 3

SMP model-estimated source contributions (ug 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



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

Table 4

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

	Soil dust	Brick kiln	Biomass/Garbage burning	Secondary	Motor vehicle
1st Period 2nd Period	45.12 14.22	7.86 32.53	28.78 28.55	2.16 11.98	43.39 29.98
Both Periods	32.11	18.25	28.68	6.29	37.74

first sampling period. In the second sampling period, motor vehicles (11.03 μ g m⁻³, 41%) and biomass/garbage burning (9.51 μ g m⁻³, 36%) sources contribute 77% to the OC mass concentration while the remaining OC concentration is explained by brick kilns (2.60 μ g m⁻³, 10%), secondary source (1.77 μ g m⁻³, 7%), and soil dust (1.63 μ g m⁻³, 6%). For both period combined, motor vehicles (13.88 μ g m⁻³, 47%) and biomass/garbage burning (9.56 μ g m⁻³, 32%) sources contribute almost 80% of the average OC mass concentration. The remaining OC is explained by soil dust (3.68 μ g m⁻³, 13%), brick kilns (1.46 μ g m⁻³, 5%), and secondary source (0.93 μ g m⁻³, 3%).

Fig. 6d shows EC source contributions to the measured EC concentration for the first period, second period and both

periods, respectively. Relative contributions of EC from motor vehicles and biomass/garbage burning sources decreased in the second period whereas those of brick kilns contribution increased by a factor of almost 3. In the first period, motor vehicles (4.31 µg m⁻³, 51%) and biomass/garbage burning (2.29 µg m⁻³, 27%) explain 78% of the measured EC while the remaining EC is explained by brick kilns (1.78 µg m⁻³, 21%) and soil dust (0.07 µg m⁻³, 1%), and there is no secondary contribution. In the second period, brick kilns (7.38 µg m⁻³, 58%), motor vehicles (2.98 µg m⁻³, 24%) and biomass/garbage burning (2.28 µg m⁻³, 18%) sources explain all of the EC mass concentration. For the average EC concentrations of both periods, three major sources for EC are brick kilns (4.14 µg m⁻³, 40%), motor



Fig. 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.

vehicles (3.75 μ g m⁻³, 37%), and biomass/garbage burning (2.29 μ g m⁻³, 22%). Soil dust contribution to EC is a negligible amount of only 1%. As explained in the previous paragraph, the first and second periods show large variations of particulate carbon contributions. In general, most of the brick kilns operate from January to April each year. However, during the sampling periods of this study, it was observed that firing of brick kilns increased suddenly from January 1, 2013. Firing of all 110 plus brick kilns in the valley was completed and all kilns became operational by January 20, 2013 (personal communication with the Chairperson of the Federation of Nepalese Brick Industries). High EC and sulfate concentrations observed in the second period (Table 1 and Fig. 2) match well with high brick kiln

contributions as shown in Fig. 6a and d.

4. Discussion

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

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

As brick kilns are furnaces used to bake bricks by burning coal and/or wood, these are one of the major sources of EC (and not just in the Kathmandu Valley, but over large parts of South Asia). Joshi and Dudani (2008) found emissions from brick kilns to adversely impact the health of children attending a nearby school. This demonstrates the importance of reducing the emission of EC, a toxic chemical and a primary indicator of adverse health effects, from brick kilns. Options to reduce emissions from the existing bull's trench brick kilns include the following: optimizing airflow and fuel in existing kilns to improve combustion efficiency such as adoption of zig-zag firing; switching to more capital intensive but cleaner brick making technologies; and switching to alternative building materials to reduce the dependence on baked clay bricks.

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

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

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

5. Summary and conclusions

The international SusKat-ABC air pollution measurement campaign took place in the Kathmandu Valley and surrounding regions in Nepal between December 2012 and June 2013. PM_{10} filter sampling was conducted at the Bode super site during the following two periods: December 21, 2012 to January 3, 2013, and

February 13, 2013 to February 21, 2013. For both sampling periods, high particulate carbon and low secondary inorganic ions in the Kathmandu Valley are found in the PM_{10} filter samples. The average PM_{10} mass concentrations for these two measurement periods are 132.0 µg m⁻³ and 121.8 µg m⁻³ for the first and second period, respectively, and 127.7 µg m⁻³ for both periods combined.

The two measurement periods show a distinct variation in PM_{10} chemical species compositions. Despite very similar PM_{10} mass concentrations, OC and trace metals are higher in the first sampling period while EC and sulfate are higher in the second sampling period. Large variation in the chemical compositions led to large variations in source contributions. Five sources (fugitive soil dust, brick kilns, biomass/garbage burning, secondary, and motor vehicles) are identified. Source contributions from motor vehicles and soil dust sources are higher in the first period and brick kilns and secondary source contributions are higher in the second period. For both measurement periods, 95% of the average PM_{10} concentration is attributed to local primary sources, motor vehicles (31%), soil dust (26%), biomass/garbage burning (23%), and brick kilns (15%), while only 5% is attributed to a secondary source.

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

In this study, it was not possible to differentiate the motor vehicles between gasoline and diesel vehicles. Similarly, the biomass/ garbage burning source was not able to be separated into biomass burning and garbage burning sources. In future studies, it would be crucial to measure individual organic compounds along with OC/ EC, ions and metals. Such individual organic compounds can be used as tracers for specific sources. This would likely enable us to separate contributions from gasoline and diesel vehicles. Such a study might also help elucidate the specific source contributions for biomass and garbage burning sources. Therefore, future study with more organic compounds as tracers would greatly help better quantification of relative contributions of these sources, and aid the establishment of effective strategies and actions to control particulate matters (particularly carbonaceous aerosols) in the Kathmandu Valley.

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Appendix A. Supplementary information

Supplementary information related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2015.10.082.

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