New Emission Inventory of Carbonaceous Aerosols from the On-road Transport Sector in India and its Implications for Direct Radiative Forcing over the Region

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ABSTRACT

In this study, we categorized detailed mass-based emission factors (EFs) by age, calculated new estimates of fuel use, and developed spatially resolved emission inventories of constituents (PM_{2.5}, black carbon [BC], and organic carbon [OC]) in the fine aerosol generated by the on-road transport sector in India. On a national level, this sector released an estimated 355 (104–607) Gg y⁻¹, 137 (47–227) Gg y⁻¹, and 106 (34–178) Gg y⁻¹ of PM_{2.5}, BC, and OC, respectively, for the base year 2013, contributing nearly 7%, 17%, and 6% of the total emissions for each constituent. Although super-emitter vehicles comprised only 24% of the total traffic volume, they were responsible for 67% and 47% of the national PM_{2.5} and BC emissions, respectively, which indicates that eliminating these vehicles may rapidly reduce emissions from the on-road transport sector in India. To predict the direct radiative forcing (DRF) from BC emissions in this sector, we then input emission estimates for the carbonaceous aerosols into the Community Atmosphere Model (CAM5) global climate model and found a positive DRF of up to 6 W m⁻² at the top of the atmosphere (TOA) and a negative DRF of up to 10 W m⁻² at the surface, suggesting that as much as 16 W m⁻² of energy remains trapped within the atmosphere. With the rapid economic growth and continued urbanization, the transport sector in India is likely to further expand in the future and hence requires immediate attention in order to reduce the BC burden and improve air quality in the nation.

Keywords: Emission factor; Black carbon; Organic carbon; Traffic volume; Climate forcing; BC burden.

INTRODUCTION

Development of a regionally representative EFs database is a prerequisite for preparing reliable emission estimates of anthropogenic aerosols and their precursor gases that are key input to climate models which are used as a tool for carrying out climate assessments (Reynolds *et al.*, 2011; Bond *et al.*, 2013; Pandey and Venkataraman, 2014). The net climate forcing by carbonaceous aerosols (BC and OC) can affect the temperature structure and winds in the atmosphere in a variety of ways at different scales (Wang, 2013). However, due to incorrect emission estimates of carbonaceous aerosols from South Asia, most climate models underestimate the aerosol burden over the region in their simulations and hence these models are generally associated with large uncertainty in terms of both magnitude and sign of climate forcing due to aerosols over South Asia (Bond *et al.*, 2013). Among

various combustion sources, the road transportation sector is already identified as one of the key emitters of carbonaceous aerosols in South Asia and especially in India (Pandey and Venkataraman, 2014). In fact, due to steadily increasing growth rate (9.1% per year) noted in this sector, the carbonaceous aerosol emissions from the on-road transport sector in India are likely to surpass emissions from all other sectors in the near future (SIAM, 2017).

The existing global (Ohara *et al.*, 2007; Borken *et al.*, 2008; Zhang *et al.*, 2009) and regional (Sahu *et al.*, 2008; Guttikunda and Mohan, 2014; Sadavarte and Venkataraman, 2014; Paliwal *et al.*, 2016) emission inventories of aerosols from South Asia often differ by almost a factor of 2–4. For road transport sector, most global emission inventories used the data reported by International Energy Outlook (U.S. EIA, 2013) for activity estimate, while the EFs (i.e., g of pollutant emitted (unit of fuel use)⁻¹) is either from AP-42 database (U.S. EPA, 2011) or emission model (MOBILE 5.0) or literature based on dynamometer studies conducted in developed countries (Yanowitz *et al.*, 2000; Zielinska *et al.*, 2004). On the other hand, recently refined regional emission inventories (Pandey and Venkataraman, 2014) used the Tier III approach (IPCC, 2013) in the inventory development

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process for South Asia. These regional emission inventories incorporated state-level activity data and EFs from emission models and dynamometer studies conducted for Indian vehicles by the Automotive Research Association of India (ARAI) and the Central Pollution Control Board (CPCB) (ARAI, 2008, 2009). This approach has undoubtedly improved the regional emission estimates; however, the use of EFs from dynamometer experiments and emission models are not representative of real-world emissions as shown by several researchers (Grieshop *et al.*, 2012; Goel and Guttikunda, 2015; Choudhary and Gokhale, 2016). These limited on-road emission measurement exercises in India have indicated that several factors including driving behavior, traffic congestion, road conditions, fuel tarnishing, engine capacity, load, and after-treatment techniques can strongly affect the emission profile from the on-road transportation sector in India. These variables are hard to simulate in a laboratory using a particular standard driving cycle. Hence, dynamometer studies are reasonable effort to study the engine performance against the compliance, but it fails to simulate real-world driving condition and therefore, emissions.

Therefore, this work presents new emission estimates of PM_{2.5}, EC (or BC), and OC from the on-road transport sector in India by overcoming some of the drawbacks of the earlier regional emission inventories. In the present work, EFs calculated from aerosol mass collected during on-road operation of light-duty passenger cars in Delhi. The new EFs, coupled with revised fuel consumption based on Tier III approach improved the emission estimates from the 4-wheeler passenger cars and considerably reduce the overall uncertainty associated with emission estimates. Emissions from other vehicle categories (HDV, LMV-commercial, tractor, and trailer) also estimated by multiplying the EFs reported in the literature with the fuel use determined in the present work. To examine the contribution of low- and high-emitting technology, we also quantified the on-road population of super-emitters and non-super-emitters vehicle and estimated their fuel consumptions and emissions. Further spatial distribution of emission fluxes (0.25° × 0.25° resolution) of BC for the year 2013 was used to estimate the BC atmospheric burden and direct radiative forcing (DRF) over the region using CAM5 global climate model.

MATERIAL AND METHODS

Total Emission Estimate

The present national emissions of PM_{2.5}, EC (onwards referred to as BC due to climate relevance), and OC are estimated for on-road transport sector of India for the base year 2013, which comprises new political states renowned after 2011. A schematic flow chart of emission estimation methodology has been presented in the supplementary information (SI; Fig. S1).

In brief, the national-level emissions of a given pollutant “p” from vehicle type “i” of various age group A–C and super-emitter (SE) using fuel type “k” was estimated by multiplying the consumption of mass of fuel of type “k” (in kg) with mass-based emission factor (in g kg^{−1}) of pollutant “p” for a given vehicle and fuel type (Eq. (1)). The age group

defined as A (2010–2013), B (2006–2009), C (2001–2005) and SE (super-emitter age group before the year 2000 and older and 17% from each category (A–C) (Fig. S1).

$$E_{p,i,k,A-C,SE} = \sum_{s=1}^{36} F_{s,i,k,A-C,SE} \times EF_{p,i,k,A-C,SE} \quad (1)$$

Emission Factors

In previous emission estimates, on-road vehicle EFs were used either from laboratory, modeled, or derived EFs (ARAI, 2008; Pandey and Venkatraman, 2014). Our motivation in the present work, the new distance-based EFs (g km^{−1}) of PM_{2.5}, OC, and BC for diesel-, gasoline-, and CNG-powered 4-wheeler (4W), 2-wheeler (2W; 4S), and LMV-passenger (3-wheeler, or 3W) vehicles of different age groups A (2010–2013), B (2006–2009), C (2001–2005) were adopted from our companion paper Jaiprakash and Habib (2017) and converted to mass-based EFs (g kg^{−1}) by multiplying it with fuel economy (listed in Table S1) and fuel density reported for Indian fuel (SIAM, 2010). In our companion paper, the tailpipe emissions are measured in 3 repetitive on-road experiments for diesel-, gasoline-, and CNG-powered 4-wheelers, which driven outside the IIT Delhi campus on a 10 km sampling route comprised of heterogeneous traffic (2W, 3W, 4W, and bicycles etc.) and 6 traffic signals. One should note that the EFs measured on 10 km stretch may not be representative of the entire country, however, due to the challenges in on-road measurement involving the continuous power supply to generate clean dilution air for diluting exhaust limited the measurement duration to 45–60 min. We still believe that the EFs are representative for congested urban roads of cities of India. The further improvement in emission factor database can be acquired by conducting the emission measurement in other cities of India using an identical systematic approach. The emissions from 2W (4S) and LMV-passenger (3W) CNG are measured at the stationary position by operating the vehicles with suspended wheels according to Modified Indian Driving Cycle (MIDC) method (ARAI, 2008).

The EFs of LMV-goods, HDV-trucks, buses (diesel and CNG) and others (such as tractor and trailer) as reported in published literature ARAI (2008, 2009) used in the present work. Since the EFs of Age Group A is not available in the literature (ARAI, 2008, 2009), hence the same EFs for Age Group B vehicles were applied in Age Group A for the emission calculation. In the absence of appropriate information, the average OC/PM_{2.5} and BC/PM_{2.5} ratios reported in ARAI (2009) and Jaiprakash and Habib (2017) for Indian vehicles are used to calculate OC and BC EFs in the present study. The ratios reported for vehicles from other countries (Alves *et al.*, 2011; May *et al.*, 2014) might be significantly different from values used in present work; however, we limited our assessment with emission factor database available for Indian vehicles to avoid the introduction of an unknown source of uncertainty. The PM_{2.5} EFs of “other vehicles” (tractors/trailers) are taken from Ramachandra *et al.* (2015), and the same ratio is used to calculate OC and BC EFs. The EFs for all categories of non-

super-emitter vehicles averaged for the age groups of A, B, C and listed in Table 1.

Generally, we assumed that old vehicle could be “super-emitter” vehicle, while few researchers explained that super-emitter is the fraction of vehicles which are poorly maintained or defective, and that could be from “old” or “new” vehicles on the fleet. Therefore, due to absence of their age, we averaged the EFs of vehicles older than the year 2000 or before from previous studies (Bond *et al.*, 2004; ARAI, 2008, 2009; Subramanian *et al.*, 2009; Sadavarte and Venkataraman, 2014; Goel and Guttikunda, 2015). Additionally, the EFs reported from any vehicle emitting two times or higher than the non-super-emitters also considered as super-emitters.

Fuel Use Estimate

Previous fuel estimation for the on-road transport sector, both approach “top” and “bottom” were used based on national or survey reports (Baidya and Borken-Kleefeld, 2009; Sahu *et al.*, 2014). In the present work, the fuel use estimate followed the Tier III approach for the on-road transport sector. In order to estimate fuel use in the present work, detailed on-road vehicle population using a survival model, annual distance travel, and fuel economy discussed in Section A1 in SI. The annual distance travel and fuel economy for different vehicle categories compiled in Tables S1 and S2. The uncertainty on fuel use and propagates to total emission

also discussed in Section A2.

Estimation of Super-emitters Vehicle

A small fraction of vehicles with poor maintenance or with outdated technology that contributes significantly towards the total emissions are termed as “super-emitters” (Bond *et al.*, 2004; Subramanian *et al.*, 2009; Yan *et al.*, 2014). The super-emitter fraction for Indian cities is not available. Nevertheless, Bond *et al.* (2004) assumed super-emitter fraction of vehicles to be in the range 5–60% for Asian countries and a recent study (Huo *et al.*, 2012) reported the super-emitter fraction in China as 23%. Therefore, the present study applied the geometric mean (17%) of the super-emitter fraction range reported by Bond *et al.* (2004) on vehicles population of Age Groups A, B, C and added all the vehicles of the model year 2000 and earlier. Finally, the super-emitter on-road population fraction estimated as 25% of the total, which is close to the value reported for China (Huo *et al.*, 2012).

Gridded Emission Inventory

The total emission of PM_{2.5}, BC, and OC from the on-road transportation sector in India for the base year 2013 spatially distributed into grids across the country. For preparing this gridded emission inventory, appropriate proxies are considered according to district-level (641 districts in India) activities of

Table 1. Fuel-based average emission factors (g kg⁻¹) of PM_{2.5}, BC, and OC for non-super-emitter vehicles.

	PM _{2.5}			BC			OC		
	A	B	C	A	B	C	A	B	C
Diesel									
LMV-Goods (3W)	2.9 ± 1.7	2.9 ± 1.7	9.9 ± 6.0	1.9 ± 0.8	1.9 ± 0.8	6.6 ± 2.6	0.9 ± 0.3	0.9 ± 0.3	3.0 ± 1.2
LMV-Goods (4W)	5.3 ± 1.1 ^a	6.5 ± 1.5 ^a	11.2 ± 7.0	3.4 ± 1.0 ^a	3.0 ± 1.0 ^a	7.8 ± 4.8	0.5 ± 0.1 ^a	1.1 ± 0.1 ^a	3.3 ± 2.0
Passenger car	5.3 ± 1.1 ^a	6.5 ± 1.5 ^a	1.8 ± 0.5 ^a	3.4 ± 1.0 ^a	3.0 ± 1.0 ^a	0.8 ± 0.2 ^a	0.5 ± 0.1 ^a	1.1 ± 0.1 ^a	0.4 ± 0.4 ^a
HDV	1.7 ± 0.4	1.7 ± 0.4	7.8 ± 0.8	1.1 ± 0.2	1.1 ± 0.2	5.5 ± 0.5	0.4 ± 0.1	0.4 ± 0.1	1.6 ± 0.2
Bus	1.7 ± 0.4	1.7 ± 0.4	5.5 ± 1.2	1.1 ± 0.2	1.1 ± 0.2	3.9 ± 0.8	0.4 ± 0.1	0.4 ± 0.1	1.1 ± 0.2
Other ^s	1.1 ± 0.2	1.1 ± 0.2	1.1 ± 0.2	0.7 ± 0.2	0.7 ± 0.2	0.8 ± 0.2	0.3 ± 0.1	0.3 ± 0.1	0.2 ± 0.1
Gasoline									
2-wheeler (2S)	2.2 ± 0.4	2.2 ± 0.4	3.0 ± 0.1	0.11 ± 0.04	0.11 ± 0.04	0.14 ± 0.06	1.3 ± 0.5	1.3 ± 0.5	1.7 ± 0.7
2-wheeler (4S)	0.5 ± 0.4 ^a	0.5 ± 0.2 ^a	1.5 ± 0.4 ^a	0.03 ± 0.0 ^a	0.04 ± 0.01 ^a	0.17 ± 0.04 ^a	0.3 ± 0.1 ^a	0.3 ± 0.1 ^a	0.9 ± 0.2 ^a
LMV-passenger (3W)	1.3 ± 0.3	1.3 ± 0.3	1.3 ± 0.3	0.07 ± 0.03	0.07 ± 0.03	0.06 ± 0.03	0.8 ± 0.4	0.8 ± 0.4	0.7 ± 0.4
Passenger car	0.7 ± 0.3 ^a	1.4 ± 0.4 ^a	1.4 ± 0.4 ^a	0.05 ± 0.01 ^a	0.08 ± 0.02 ^a	0.20 ± 0.04 ^a	0.7 ± 0.4 ^a	0.8 ± 0.4 ^a	0.7 ± 0.7 ^a
CNG									
LMV-passenger (3W)	0.4 ± 0.1 ^a	0.8 ± 0.3 ^a	3.3 ± 1.1 ^a	0.01 ± 0.01 ^a	0.09 ± 0.05 ^a	0.20 ± 0.05 ^a	0.3 ± 0.1 ^a	0.4 ± 0.1 ^a	1.8 ± 0.6 ^a
Car (4W)	0.4 ± 0.1 ^a	0.4 ± 0.1 ^a	1.1 ± 0.4 ^a	0.01 ± 0.002 ^a	0.02 ± 0.00 ^a	0.20 ± 0.13 ^a	0.3 ± 0.1 ^a	0.2 ± 0.1 ^a	0.2 ± 0.1 ^a
Bus	2.0 ± 1.1	2.1 ± 0.4	2.1 ± 0.7	0.10 ± 0.04	0.10 ± 0.01	0.10 ± 0.03	1.1 ± 0.4	1.0 ± 0.1	1.1 ± 0.4

Vehicle age groups classified as: A = 2010–2013; B = 2006–2009; C = 2001–2005.

LMV: Light motor vehicle: diesel-fueled 3-wheeler (3W) and 4-wheeler (4W) used as commercial vehicles.

^a Emission factors are taken from our companion paper (Jaiprakash and Habib, 2017) and converted to fuel-based emission factors of PM_{2.5}, BC, and OC.

The emission factors of LMV-goods, HDV-trucks, buses, LMV-passenger (3W-gasoline), 2-wheelers (2S), CNG buses were adopted from ARAI (2008, 2009) for PM_{2.5}, while average fraction of BC/PM_{2.5} and OC/PM_{2.5} were adopted from ARAI (2008, 2009) and Jaiprakash and Habib (2017) for deriving emission factors of BC and OC.

^s PM_{2.5} emission factors of other vehicles (tractor and trailers) were adopted from Ramachandra *et al.* (2015).

the road transport sector in India and illustrated in Table S3. The whole India divided into 16,384 grid cell and each cell has $0.25^\circ \times 0.25^\circ$ resolution. Total emissions from diesel-fueled vehicles and their super-emitters are distributed grid-wise following Pandey *et al.* (2014) and by assuming fixed fractions of road densities of “golden quadrilateral” roads (30%), national highways (40%), state highways (20%), and urban grids (10%) across the country. In the absence of exact information on road density for each grid, we followed the assumptions made by Pandey *et al.* (2014) for consistency with other recently published emission estimates. The uncertainties associated with assumed road density distribution are $\pm 7.5\%$, $\pm 10\%$, $\pm 5\%$, and $\pm 2.5\%$ for “golden quadrilateral” road, national highways, state highways, and urban grids, respectively (personal communication from Dr. Sarath Guthikunda). Furthermore, the proxy of gasoline vehicles was taken as urban population of districts grids following Pandey *et al.* (2014). Spatial distribution of $\text{PM}_{2.5}$, BC, and OC has been visualized using ArcGIS software (10.1).

Climate Model Description and Simulation Details

We use a variant of the three-dimensional global climate model called the Community Earth System Model (CESM1, released version 1.2) developed at the National Center of Atmospheric Research (NCAR) (Hurrell *et al.*, 2013). The direct radiative forcing over the Indian region due to emissions of BC from the on-road transportation sector in India estimated. The uncertainty in BC emission estimates is 33%; therefore, the radiative forcing calculation is expected to be well bounded. The earlier study also used BC emission from transport sector with 33% uncertainty for radiative forcing (Fuglestedt *et al.*, 2008).

The atmospheric components of CESM1 version used here are the Community Atmosphere Model (CAM5.3). The CAM5.3 has a hydrostatic, finite-volume (FV) dynamical core with a horizontal resolution of $0.9^\circ \times 1.25^\circ$, 56 hybrid sigma pressure levels and configured in the “specified dynamics” mode (Lin, 2004; Neale *et al.*, 2004). This resolution allowed us to constrain the model-simulated meteorology to match with the meteorological analysis from MERRA-2. The meteorology prescribed in the model include winds, temperature, surface heat, and moisture fluxes. The clouds and aerosols are allowed to evolve according to the model’s governing equations (Ma *et al.*, 2015; Tilmes *et al.*, 2015). The CAM5.3 uses a modal approach to represent the aerosol life cycle in a way such that different aerosol species are internally mixed within modes and externally mixed between modes (Ghan *et al.*, 2012; Liu *et al.*, 2012). The 3-mode version of the modal aerosol module (MAM3) used in this study provides internally mixed representations of number and mass size distributions for Aitken-, accumulation- and coarse-mode aerosols. Aitken-mode species include sulfate, secondary organic aerosol (SOA), and sea salt. Accumulation-mode species include sulfate, SOA, BC, primary organic matter (POM), sea salt, and dust, while coarse-mode species include sea salt, dust, and sulfate. The model consists of all the key processes influencing the aerosol life cycle, such as precursor gas and particle emissions, gas- and aqueous-phase chemistry, nucleation, condensation, coagulation, aging, precipitation

scavenging, and dry deposition (Liu *et al.*, 2012). In CAM5, the Rapid Radiative Transfer Method for GCMs (RRTMG) used to calculate the long and shortwave radiative transfer (Iacono *et al.*, 2008). The aerosol optical properties for each internally mixed modes of MAM defined according to Ghan and Zaveri (2007).

The model simulations are performed from 1 December 2009 to 31 December 2015, with the first one month of model output excluded from our analyses to allow the model to spin up following the initial conditions. We conduct two simulations with identical model configuration using prescribed greenhouse gas concentrations and anthropogenic emissions of aerosols and their precursors (Representative Concentration Pathway 8.5) as described by van Vuuren *et al.* (2011) and Lamarque *et al.* (2011), except, one using the BC emissions from on-road transportation sector within India as discussed in this work and the other one neglecting the BC emissions from on-road transportation sector in India. The default emissions of carbonaceous aerosols (BC and OC) from the road transportation sector in the emission inventories corresponding to RCP 8.5 scenario is replaced by the new local emission inventory developed as part of the present study for emissions within India and merged with the global emission inventory described by Lamarque *et al.* (2011) and van Vuuren *et al.* (2011). In this study, we have analyzed six years (1 January–31 December of 2010–2015) of monthly mean simulated data of aerosol and radiative fluxes to estimate the DRF due to BC emissions from the on-road transportation sector in India.

Calculation of Aerosol Radiative Forcing

We followed the IPCC AR5 (The Intergovernmental Panel on Climate Change Fifth Assessment Report) methodology for estimating the anthropogenic aerosol effects on the planetary energy balance in terms of effective radiative forcing (ERF) that allows clouds to respond to aerosol while surface temperature prescribed. This method allows us to quantify aerosol radiative effects in terms of effective radiative forcing from aerosol–radiation interactions (ARI) as well as from aerosol–cloud interactions (ACI). However, it is worth mentioning here that although constraining the model meteorology to match with the meteorological analysis from MERRA-2 has several advantages such as it facilitates direct comparisons between model simulations and observations at specific times and locations, one limitation of this approach is that it removes the aerosol-induced feedbacks to the atmospheric dynamics and the constrained temperature eliminates the semi-direct effects of aerosols (Ma *et al.*, 2015; Tilmes *et al.*, 2015). Hence, in this study, we restrict our discussion to effective radiative forcing from aerosol–radiation interactions alone, which is also known in conventional terms as the *aerosol direct radiative forcing*.

Our calculations of aerosol direct radiative forcing are based on the work of Ghan *et al.* (2012) and Ghan (2013). The BC aerosol DRF discussed here is defined as road-transportation-sector-induced changes in radiative fluxes at the TOA and at the surface, calculated as the difference of radiative fluxes at the desired level between simulations with and without BC emissions from on-road transportation

sector in India (denoted by Δ). In each simulation, aerosol forcing defined as the difference between all-sky and clean-sky radiative fluxes ($F - F_{\text{clean}}$), where F is the radiative flux under all-sky conditions and F_{clean} is the radiative flux calculated as a diagnostic from the same simulation, but neglecting the scattering and absorption of solar radiation by aerosols. Thus, in the present study, aerosol direct radiative forcing is estimated as $\Delta(F - F_{\text{clean}})$. The difference of the forcing values at the TOA and surface is further used to estimate the energy trapped within the atmosphere due to BC emissions from the road transportation sector.

As discussed later, in Section 3.3 that there is approximately 50% uncertainty in our emission estimates of BC from the road transportation sector in India. Hence, in order to understand how this uncertainty in the emissions of BC from the road transportation sector in India translates into uncertainty in DRF estimates, we carry out two more model sensitivity experiments in addition to the pair of standard simulations described earlier. These sensitivity experiments are identical to the simulation setup discussed earlier for the case which included the BC emissions from on-road transportation sector within India, except in one case we increased the gridded BC emissions from the road transportation sector in India by 50% and in the other case, we decreased the same emissions by 50% of the mean value used earlier, while the BC emissions outside India and across the world remained same in all our experiments.

RESULTS AND DISCUSSION

Emission Factors of $\text{PM}_{2.5}$ and Carbonaceous Aerosol

Emission Factors of Non-super-emitter Vehicles

Table 1 shows the EFs of $\text{PM}_{2.5}$ and carbonaceous aerosols (BC and OC) for non-super-emitter vehicles classified in terms of thirteen different vehicle categories, three different fuel types, and three different age groups namely, A (2010–2013), B (2006–2009) and C (2001–2005). The measured on-road EFs of 4W-diesel, 4W-gasoline, 4W-CNG, 2W-4S, and LMV-passenger emphasized as in bold are taken from our companion paper (Table 1). The other EFs factors mentioned in Table 1 compiled from the previous literature, which measured from controlled laboratory condition.

The average $\text{PM}_{2.5}$ EFs of non-super-emitter diesel-powered vehicles (Table 1 and Fig. S2) is found to be lowest ($1.1 \pm 0.2 \text{ g kg}^{-1}$) for “other” type (tractor and trailer) and highest for LMV-goods (4Ws; $11.2 \pm 7.0 \text{ g kg}^{-1}$) of Age Group C (2001–2005). As evident from Table 1 that $\text{PM}_{2.5}$ EFs increase by a factor of 2–5 with increasing vehicle age for most vehicle categories. In contrast the results also showed that the $\text{PM}_{2.5}$ EF for 4W-diesel of the model year 2001–2005 (C) is lower than the EFs for Age Groups A and B. Vehicles older than 10 years of age are not readily available in Delhi and hence, the measurements could not be repeated for more vehicles to confirm this trend. A previous study by Alves *et al.* (2015) also reported lower EFs from well-maintained old vehicles compared to poorly maintained and rigorously used newer vehicles. Besides, we also noted that the EF reported by ARAI (2008) for Indian vehicles of the same age group (C) was close to the values measured by

us. Therefore, we used the emission factor for 4-W diesel on-road from measurement even though it was contrary to the trend.

The EFs of super-emitter vehicles (Table 2 and Fig. S2) are a factor of 2–17 times higher than non-super-emitter category. Super-emitters characterized by their low contribution (~24%) to traffic volume but high contribution (over 50%) to emissions. The relationship between the traffic volume and emissions from non-super-emitter and super-emitter vehicles discussed in later section.

We compare the EFs used in previous global, national, and regional emission estimates with the values used in present work (Fig. 1). It is noteworthy that the emission factors used in present work are within the range of values used in previous emission estimates. In some case considerable difference in present and previous work (Sahu *et al.*, 2008; Lu *et al.*, 2011) arose because either earlier studies have compiled emission factors for few vehicle categories from dynamometer study or derived from emission models (Broken *et al.*, 2007; Ohara *et al.*, 2007; Klimont *et al.*, 2009; Zhang *et al.*, 2009). Therefore, in the present work, we used on-road EFs from the present study for 4W-passenger cars and did not average them with values reported in the literature from dynamometer study and emission models. The emission factors for other categories in which the on-road emission measurement not done in present work, the values from the literature averaged for emission calculation. Therefore, measured EFs is our motivation in this work to understand the impact on total emission and climate forcing.

Vehicle Traffic Volume and Fuel Use Estimate

The vehicle traffic volume (billion vehicles) which is the product of on-road vehicle population and annual distance travel was highest for 2W-gasoline (56% of 1550 billion vehicle km) followed by diesel-powered “other vehicles”, HDV, LMV-goods, passenger and buses, while CNG vehicles contribute the smallest fraction of total traffic volume (Figs. 2(a)–(c)). However, one can notice that fuel use showed the opposite trend as diesel fuel consumption by “other vehicles” and HDV-trucks was higher as compared to 2W-gasoline and CNG vehicles (Figs. 2(d)–2(f)), because of lower (4–10 times) fuel economy of diesel vehicles (Table S2) compared to 2W gasoline-powered vehicles.

Emissions Estimates of $\text{PM}_{2.5}$ and Carbonaceous Aerosol

The central value and 95% confidence interval (CI) of $\text{PM}_{2.5}$ emissions from on-road transport sector is estimated to be around 355 (104–607) Gg y^{-1} at the national level with maximum contribution from diesel vehicles (83.7%) followed by gasoline (15.6%), and an insignificant contribution from CNG (0.7%) (Fig. 3). Although, diesel vehicles comprise only 27% (416 billion vehicle km) of total traffic volume (1550 billion vehicle km) (Fig. 3), gasoline and CNG contributes rest 73%; however, low fuel economy and high EFs (Tables 1 and S2) of diesel vehicles and more than 2 times higher fuel consumption (52 MT y^{-1}) resulted in 5.3 times higher emissions (297 Gg y^{-1}) compared to gasoline vehicles. Interestingly, non-super-emitter and super-emitter vehicle comprise 76% and 24% of the total traffic volume,

Table 2. Emission factors of PM_{2.5}, BC, and OC for super-emitter vehicles.

Vehicle type	Aerosol and its constituents (g kg ⁻¹)		
	PM _{2.5}	BC	OC
Diesel			
LMV-goods (3W)	27.5 (22.4–33.8) ^{a,b}	(15.1 ± 14.8)*	(5.5 ± 5.4)*
LMV-goods (4W)	(26.2 ± 25.6) ^{a-d}	(14.4 ± 14.1)*	(5.1 ± 5.9) ^{a,c,d}
Passenger car	(11.1 ± 7.4) ^{a-e}	(5.0 ± 1.8) ^{c,d,e}	(2.0 ± 0.7) ^{a,c,d,e}
HDV	(18.4 ± 13.1) ^{a-d}	(6.6 ± 6.5) ^d	(3.9 ± 3.1) ^{a,c,d}
Bus	(17.9 ± 12.1) ^{a-d}	(4.9 ± 1.6) ^{a,c,d}	(3.9 ± 3.1) ^{a,c,d}
Other	(18.7 ± 13.6) ^{a-d}	(4.9 ± 1.6) ^{a,c,d}	(3.9 ± 3.1) ^{a,c,d}
Gasoline			
2-wheeler (2S)	(22.9 ± 6.2) ^{a,b,d}	1.2 (1.0–1.5) ^{a,b}	16.1 (11.0–23.7) ^{a,b}
2-wheeler (4S)	3.6 (3.2–4.0) ^{a,b}	(0.2 ± 0.2) ^a	(2.2 ± 1.1) ^a
LMV-passenger (3W)	(4.8 ± 1.0) ^{a,b,f}	0.7 (0.2–2.2) ^{a,f}	2.7 (2.4–2.9) ^{a,f}
Passenger car	(2.6 ± 1.7) ^{a,b,d,e}	(0.6 ± 0.5) ^{a,d,e}	0.9 (0.7–1.1) ^{a,f}
CNG			
LMV-passenger (3W)	8.6 (4.6–15.9) ^{a,f}	1.2 (0.9–1.6) ^{a,f}	4.7 (2.3–9.5) ^{a,f}
Passenger car	(2.5 ± 2.5) ^b	(0.8 ± 0.7)*	(0.9 ± 0.8)*
Bus	(2.0 ± 2.0) ^b	(0.3 ± 0.3)*	(0.9 ± 0.9)*

For three emission factor (EF) values averaged from following literature. Values in parentheses indicate standard deviation. If literature reported only two EF values, then we calculate geometric mean. Value in parentheses represent lower and upper range of EFs from literature.

* Average fraction of BC/PM_{2.5} and OC/PM_{2.5} were reported in Bond *et al.* (2004) and ARAI (2008, 2009).

^a Old-vintage (before 2000) EFs were compiled for PM_{2.5} from ARAI (2008, 2009).

^b Based on COPERT-modeled PM_{2.5} emission factors from Goel and Guttikunda (2015).

^c Based on dynamometer measurement for 33 light-duty vehicles from Subramanian *et al.* (2009).

^d Based on opacity-based measurement for “smoker” gasoline, diesel, and farm/non-farm vehicles from Bond *et al.* (2004).

^e Based on chassis dynamometer and opacity measurement for 4-wheeler gasoline and diesel vehicles from Yan *et al.* (2011).

^f From Grieshop *et al.* (2012): (29) 3-wheelers (10 petrol and 19 CNG) were measured on chassis dynamometer studies.

respectively but contribute 33% (119 [77–161] Gg y⁻¹) and 67% (236 [27–446] Gg y⁻¹) of total national-level PM_{2.5} emissions (Fig. 3). This implies that the vehicle growth in future and fraction of super-emitters can significantly intensify the PM_{2.5} emissions from the on-road transport sector. Therefore, a proper mitigation policy must not only focus on strengthening public transport as an emission reduction option but also should focus on controlling the fraction of super-emitters. It is worth noting that the uncertainty associated with emission estimates from the super-emitter vehicles (89%) is significantly higher than non-super-emitter vehicle category (36%) because the EFs for super-emitters not assessed yet and their exact fraction in terms of traffic volume is also not known.

It is important to note here that although the HDV-trucks and “other vehicles” (tractor/trailer) have a relatively smaller contribution (7–8%) towards the total traffic volume (Fig. 2(a)), these vehicles contribute significantly (55%) towards the national PM_{2.5} emissions from India with almost equal contributions from HDV-trucks (28% (99 Gg y⁻¹) and “other vehicles” (tractor/trailer; 27% (97 Gg y⁻¹)). On the other hand, the LMV-goods vehicles with 3 times higher EFs and much lower share towards the on-road traffic volume (5%) contribute nearly 12% of the national PM_{2.5} emissions from India. With economic growth and rapid urbanization happening across India, the on-road traffic volume of freight (i.e., “other vehicles”, LMV-goods, and HDV-trucks) is expected to increase rapidly in the coming

years and hence a mechanism of on-road measurement of emission profile for this category vehicles are crucial for quantifying their correct contribution towards the total national emission of PM_{2.5} and climate forcing agents like carbonaceous aerosols from India. Diesel-powered public transport (buses), and private cars (4Ws) contribute only 2% and 5% towards the total on-road traffic volume but contribute nearly 9% (44 Gg y⁻¹) and 7% (26 Gg y⁻¹) towards the national PM_{2.5} emissions (Fig. 3(a)). The contribution from non-super-emitters and super-emitter vehicles among the diesel-powered high-emitting technologies to the total PM_{2.5} emissions across India estimated as 34% (33.7 Gg y⁻¹) and 66% (65.3 Gg y⁻¹) respectively for HDVs, 80% (77.8 Gg y⁻¹) and 20% (19.4 Gg y⁻¹) respectively for “others” and 45% (19.3 Gg y⁻¹) and 55% (24 Gg y⁻¹) respectively for LMV-goods vehicles (Fig. 3(a)).

Our results further show that the gasoline-powered 2Ws, LMV-passengers, and 4W-passenger cars contribute nominally 10% (36.5 Gg y⁻¹), 3% (10.4 Gg y⁻¹) and 2% (8.7 Gg y⁻¹) respectively to national-level PM_{2.5} estimates (Fig. 3(b)). It is worth noting here that even though 2W vehicles have a maximum contribution towards the on-road traffic volume (56% of 1150 billion vehicle km) but contribute only 10% towards the national PM_{2.5} emissions and this is mainly due to low fuel consumption and low EFs reported for 2Ws. The contribution of gasoline-powered non-super-emitter and super-emitter vehicles in different categories towards the total PM_{2.5} emissions (56 Gg y⁻¹) estimated as 19.6% (11 Gg y⁻¹)

Fig. 1. Variation of EFs using emission estimate of previous and comparison with present study. Open square legends (□) correspond to estimates from previous studies which used category-wise vehicular EFs. Open circle (○) indicates the EFs adopted by earlier literature for few vehicle category and applied to national level emission estimate. Star (★) symbol corresponds to results from emission models and derived EFs. Open red circle indicates the EFs of super-emitter vehicles. The super-emitter vehicles were compiled from the previous studies (Bond *et al.*, 2004; ARAI, 2008, 2009; Subramanian *et al.*, 2009; Sadavarte and Venkataraman, 2014; Goel and Guttikunda, 2015).

and 46.4% (26 Gg y⁻¹) for 2Ws, 6.4% (3.6 Gg y⁻¹) and 12.1% (6.8 Gg y⁻¹) for LMV-passenger, and 8.7% (4.9 Gg y⁻¹) and 6.8% (3.8 Gg y⁻¹) for passenger cars respectively (Fig. 3(b)). The contributions from CNG-operated non-super-emitter and super-emitter vehicles towards the total PM_{2.5} emissions (2.4 Gg y⁻¹) estimated as 25% (0.6 Gg y⁻¹) and 50% (1.2 Gg y⁻¹) respectively for LMV-passenger (3W) vehicles (Fig. 3(c)) and the remaining 25% of PM_{2.5} emissions from

CNG-powered vehicles are contributed by CNG passenger cars and buses with higher contribution once again from super-emitter vehicles in this category. Thus overall, the super-emitter vehicles contribute more than 50% of PM_{2.5} emissions from all vehicle categories in India.

Results of present study suggests that the total BC emissions from on-road transport sector at the national level is around 137 (47–227) Gg y⁻¹ with maximum contribution

Fig. 2. Estimated total traffic volume (left panel) and fuel consumption (right panel) from on-road transport sector.

from diesel-powered vehicles (96%, 131 Gg y⁻¹) and much smaller contribution (4%, 6 Gg y⁻¹) from gasoline- and CNG-powered vehicles (Figs. 3(d)–3(f)). Overall super-emitters and non-super-emitters contribute nearly 53% (72 [5–139] Gg y⁻¹) and 47% (64 [42–88] Gg y⁻¹) respectively of total BC emissions from on-road transport sector. However, considerable uncertainty of nearly 92% and 36% are associated with our estimates of BC emissions from super-emitters and non-super-emitter vehicles, respectively. Most remarkably our estimates of BC emissions from on-road transport sector suggests that among the super-emitter vehicles, an overwhelming majority (94% (68 Gg y⁻¹)) of BC emissions contributed by diesel-powered vehicles and only a small fraction (6% (4 Gg y⁻¹)) comes from gasoline and CNG vehicles. Our results further suggest that while the overall contribution of non-super-emitter vehicles is around 47% towards the total national-level BC emissions from on-road transport sector, the diesel-,

gasoline-, and CNG-powered non-super-emitter vehicles contribute nearly 46.2% (63 Gg y⁻¹), 0.96% (1 Gg y⁻¹) and 0.03% (0.05 Gg y⁻¹), respectively. Among vehicle categories, HDV-trucks account for the highest 34% (46.3 Gg y⁻¹) contribution to the total BC, followed by “other vehicles” at 24% (33 Gg y⁻¹), LMV-goods at 18% (24.7 Gg y⁻¹), passenger cars at 11% (15 Gg y⁻¹), buses at 9% (12.3 Gg y⁻¹), 2-wheeler at 2% (2.7 Gg y⁻¹) and LMV-passenger at 1% (1.4 Gg y⁻¹). Overall, the contribution of commercial freight vehicles and passenger vehicles estimated around 76% and 24% respectively to the total national-level BC emissions from the on-road transport sector.

Furthermore, the total OC emissions from on-road transport sector at national level estimated as 106 (34–178) Gg y⁻¹ with leading contribution from diesel vehicles (58%, 61 [15–111] Gg y⁻¹) followed by gasoline (41%, 43 [22–64] Gg y⁻¹), and CNG-powered vehicles (1%, 1.3 Gg y⁻¹) (Figs. 3(g)–3(i)).

Fig. 3. Total emission estimate of $PM_{2.5}$ (left panel), BC (middle panel), and OC (right panel) from on-road transport sector.

In terms of vehicle category among diesel-powered vehicles, HDVs emit highest OC (20%, 22 Gg y⁻¹), followed by “other vehicles” (20%, 21 Gg y⁻¹), LMV-goods (7%, 7 Gg y⁻¹), buses (7%, 7 Gg y⁻¹), and passenger cars (4%, 4 Gg y⁻¹). Among gasoline vehicles, 2-wheelers are found to be the highest emitter of OC (75%, 32 Gg y⁻¹), while LMV-passenger and passenger cars contribute 14% (14.8 Gg y⁻¹) and 11% (11.7 Gg y⁻¹) respectively towards the total OC emissions. Overall passenger vehicles (i.e., 2Ws, LMV-passenger, 4Ws, and buses) and commercial freight vehicles (i.e., HDV-trucks, LMV-goods, “others”) contribute almost equally (52% and 48% respectively) towards the total OC emissions at the national level from on-road transport sector. Non-super-emitters and super-emitters contribute nearly 32% (34 Gg y⁻¹) and 68% (72 Gg y⁻¹) respectively of the total OC emissions from this sector. The uncertainty associated with the estimates of OC emission with 95% CI is calculated to be 79% and 43% for super-emitters and for non-super-emitter vehicles respectively.

Thus, our findings from the present study emphasize the need for emission control from the on-road transport sector in India. In fact, keeping in view the expected growth of road transport sector in India, our study suggests that the contribution of super-emitters and non-super-emitters needs to be ascertained more accurately by making the correct measurements of emissions from all kinds of vehicles under real-world driving pattern prevalent in India. Besides emission measurement, a compilation of activity data such as on-road vehicle population, vehicle lifetime, annual distance travel, and mileage is a necessity, should be collected across the country.

Comparison with Previous Emission Estimate

Fig. S3 presents the comparison of emissions of PM_{2.5}, BC, and OC for the base year 2013 as estimated in the present study with previous emission estimates reported for the base year anywhere between the period 2000–2010. For making this comparison, we scaled the previous emission estimates by multiplying them with the ratio of present-day fuel usage to previously reported fuel usage. Our comparison shows that the PM_{2.5} emission estimated in the present study is comparable to recently reported emission estimates by Guttikunda and Mohan (2014), and Pandey and Venkataraman (2014) from the on-road transport sector for the same base year 2013 (Fig. S3). Higher values of EFs used in previous studies by Broken *et al.* (2007), Baidya and Borken-Kleefeld (2009) and Zhang *et al.* (2009) for most vehicle categories (Fig. S3(a)) resulted in approximately 17%, 28% and 29% higher emission estimates respectively as compared to the present work. Lower values of EFs used by Reddy and Venkataraman (2002) and Ramachandra and Shewtmala (2009) for most vehicle categories as compared to our present work resulted in 23% and 37% lower emission estimates by these two studies respectively as compared to present work. The present estimates (355 Gg y⁻¹) of PM_{2.5} emissions from on-road transport is 1.6 times higher than the estimate (222 Gg y⁻¹) reported in Emissions Database for Global Atmospheric Research (EDGAR) (Crippa *et al.*, 2018). However, most importantly, the uncertainty associated with

the PM_{2.5} emission estimate from on-road transport sector with 95% CI reduced from 98% as reported in previous estimates by Pandey and Venkataraman (2014) to 70% in the present study.

We also compared our emission estimates with those reported for Delhi city by Goel and Guttikunda (2015) and Sharma and Dixshit (2016) by scaling the previous estimates for the base year 2013. Our estimate of PM_{2.5} emissions from 4-wheelers is 1.7 times higher than the values reported by Sharma and Dixshit (2016) and 1.8 times lower than values reported in Goel and Guttikunda (2015) for Delhi megacity. The difference is possibly because, Sharma and Dixshit (2016) used the EFs for all vehicle categories reported from chassis dynamometer measurements in ARAI (2008, 2009), which are lower than EFs determined from on-road measurement in the present work. On the other hand, the study by Goel and Guttikunda (2015) used fleet average (1999–2012) emission factor values derived from COPERT emission model, which could be higher than on-road measurement used in the present work.

BC emissions reported by Ohara *et al.* (2008) are 2.6–5.6 times higher than our estimates (Fig. S3(b)). Similarly, previous work by Street *et al.* (2003) and Sahu *et al.* (2008) have used higher BC EFs (10 g kg⁻¹) for all categories of vehicles as compared to our present study. In the present work, BC EFs vary from low values of 0.7 g kg⁻¹ for “other vehicles” to high values of 3.6 g kg⁻¹ for passenger cars, and LMV-goods vehicles among the non-super-emitters (Age Group A and B), which comprise nearly 10% of the total vehicle population. The uncertainty at 95% CI associated with BC emissions from on-road transport sector reduced from 105% as reported by Pandey and Venkataraman (2014) to 65% in the present work. The uncertainty in BC emissions as estimated in the present work is also consistent with the uncertainty reported by Paliwal *et al.* (2016) for the base year 2011.

OC emissions from on-road transport sector as estimated in the present work is mostly consistent (6–12% difference) with the previous estimates reported by Pandey and Venkataraman (2014) and Guttikunda and Mohan (2014), while the present estimates are found to be 32–42% lower than the emissions reported by Reddy and Venkataraman (2002) and Ohara *et al.* (2007). Whereas some other previous studies such as by Klimont *et al.* (2009) and Lu *et al.* (2011) reported lower OC emissions compared to present estimates by 33% and 58% respectively (Fig. S3(c)). Our OC estimates are significantly lower (by 1.8–4.4 times) than the scaled emissions derived from the studies by Ohara *et al.* (2007), Zhang *et al.* (2009) and Street *et al.* (2013) (Fig. S3(c)), this could be attributed to the use of high EFs of LMV, HDV and 2-wheeler in previous studies. The uncertainty at 95% CI associated with OC emissions from on-road transport sector reduced from 136% as reported by Pandey and Venkataraman (2014) to 95% in the present work.

Next, we compared the emission estimates of PM_{2.5}, BC and OC from diesel- and gasoline-based 4-wheelers derived using different approaches such as EFs measured in on-road experiments as used in the present work, previous dynamometer studies, and emission models. Fig. 4 shows

the analysis of national-level emission estimates of $\text{PM}_{2.5}$, BC and OC from 4Ws due to the application of EFs from different approaches. The Student's *t*-test analysis showed the use of EFs measured from the on-road operation of 4-wheelers resulted in significantly higher emissions ($p < 0.05$ at 95% CI) compared to those obtained using EFs from dynamometer study. For diesel-powered 4Ws the emission estimates of $\text{PM}_{2.5}$ and BC using the EFs derived from BC/ $\text{PM}_{2.5}$ ratios are slightly lower than the present estimates (Fig. 4), while for OC, model-derived EFs resulted in marginally higher emissions compared to present work. For gasoline-based 4-wheelers, the $\text{PM}_{2.5}$, BC, and OC emission estimates using model-derived emission factor are again marginally higher (7–18%) compared to the present estimate (Fig. 4).

Thus the emission estimates based on model-derived EFs

seem to be marginally different from the estimates of our present work, while estimates based on EFs from dynamometer are primarily lower than the present estimates. The EFs from on-road emission measurements used in the present work provide an intermediate well-bounded emission estimate.

Spatial Distribution of Emission Fluxes

The present emission estimates are then used to prepare a gridded emissions data ($0.25^\circ \times 0.25^\circ$ resolution) or spatial distribution of emission fluxes (tons $\text{grid}^{-1} \text{y}^{-1}$) of $\text{PM}_{2.5}$, BC, and OC from the Indian road transport sector for the base year 2013. Our gridded emissions data shows high emission fluxes with values greater than 1500 tons $\text{grid}^{-1} \text{y}^{-1}$ of $\text{PM}_{2.5}$ over the four metropolitan cities (Delhi, Mumbai, Kolkata, and Chennai) due to the high population, road density, and traffic volume (Fig. S4). The “golden quadrilateral” road network,

Fig. 4. Emission of $\text{PM}_{2.5}$, BC and OC from 4-wheeler diesel and gasoline using on-road measured emission factors from the present work, dynamometer studies, and emission model.

which connects the metro cities and industrial cluster showed a substantial magnitude of 500–1500 tons $\text{grid}^{-1} \text{y}^{-1}$ of $\text{PM}_{2.5}$ emission fluxes. Industrial clusters within this road network are found to be associated with a moderate level of emissions ranging from 100 to 500 tons $\text{grid}^{-1} \text{y}^{-1}$ over the states of Gujarat, Maharashtra, Tamil Nadu, West Bengal, and Delhi, even though these states accounted 50% of $\text{PM}_{2.5}$ emissions. The national and state highways showed $\text{PM}_{2.5}$ emissions fluxes varying in the range of 100–500 tons $\text{grid}^{-1} \text{y}^{-1}$ from diesel-powered vehicles, whereas the entire district and urban grid emissions are found to vary from 5 to 100 tons $\text{grid}^{-1} \text{y}^{-1}$ mainly due to gasoline and CNG vehicle emissions.

The gridded BC emissions showed a similar spatial variability as $\text{PM}_{2.5}$ because BC is a dominant component of $\text{PM}_{2.5}$ emissions from the on-road transport sector. Among vehicles, the primary source of BC is diesel-powered vehicles including LMV-goods, 4-wheeler diesel, HDVs, buses and tractor and trailers. The observed BC emission fluxes resemble road network density with higher values in Gujarat, Maharashtra, Tamil Nadu, West Bengal and Delhi ($> 1000 \text{ tons grid}^{-1} \text{y}^{-1}$) (Fig. S4, middle panel).

The clusters of OC emissions fluxes of magnitude $\geq 1500 \text{ tons grid}^{-1} \text{y}^{-1}$ noted over the Indo-Gangetic Plain (IGP) (Fig. S4, right panel), this is corroborated with dense population and demand of vehicles running on high-efficiency fuels such as gasoline and CNG, which emit more OC. The “golden quadrilateral” and national highways that are dominated by diesel vehicular traffic and related activities are also characterized by 50–1500 tons $\text{grid}^{-1} \text{y}^{-1}$ OC emission fluxes (Fig. S4, right panel). Interestingly, although the IGP region accounts for only 15% of the total Indian geographical area, this region accounts for nearly 42% (45 Gg y^{-1}) of the total OC emissions from the on-road transport sector in India. The remaining emissions (56%) of OC distributed over central, northwestern, northern–eastern, and eastern part of central India, this is due to rapidly increasing urbanization in these parts of India that also increases the fuel demand for on-road transport sector in these regions.

To compare our spatially resolved emissions with previous work, we scaled the previous estimates for the different base year using the vehicle population growth rate reported by MORTH (2013). Overall we noted a good agreement between the emission estimates of the present work with the estimates earlier reported by Sadavarte and Venkataraman (2014) both in terms of magnitude and spatial distribution pattern of $\text{PM}_{2.5}$, BC, and OC emissions. A root mean square error (RMSE) between the two inventories for $\text{PM}_{2.5}$, BC, and OC are calculated as 0.3, 6.0 and 3.5 tons $\text{grid}^{-1} \text{y}^{-1}$ and an index of agreement (Willmott, 1981; Jaiprakash *et al.*, 2010) between the two inventories found as 1.0, 0.99 and 0.99 for $\text{PM}_{2.5}$, BC, and OC respectively, indicating a good agreement between present work and previous work by Sadavarte and Venkataraman (2014).

Atmospheric BC Burden Due to Emissions from the Road Transport Sector

The emission estimates of carbonaceous aerosols are then used in CAM5 global climate model to estimate the atmospheric burden and DRF due to BC emissions from on-

road transport in India. As mentioned earlier the model simulations are performed from 1 December 2009 to 31 December 2015, with the first one month of model output excluded from our analyses to allow the model to spin up following the initial conditions. Fig. 5 shows the monthly mean climatology (2010–2015) of change in BC column burden (in kg m^{-2}) for all months from January to December over the South Asian region due to BC emissions from road transportation sector in India as estimated by the CAM5 model based on the difference of results from two simulations with and without BC emissions from road transportation sector in India. Interestingly, although there is no monthly variation in the emissions of aerosols from the road transport sector in our simulations, significant monthly variability is noted in the contribution of road transportation sector emissions from India towards the column burden of BC over South Asia. This monthly variability in the column burden of BC as noticeable in Fig. 5 despite using identical BC emissions for all months occurs primarily due to differences in the atmospheric transport pathways and the scavenging rate of aerosols from the atmosphere during different months. Fig. 6 shows the annual cycle of monthly mean climatology (2010–2015) of atmospheric BC burden (in kg m^{-2}) of India along with BC deposition rate (in $\text{kg m}^{-2} \text{s}^{-1}$), and precipitation rate (in mm day^{-1}) simulated by the CAM5 model with prescribed meteorology from MERRA-2. As light winds characterize the meteorological conditions over most parts of the Indian subcontinent during the months from December to March at the surface (Fig. 5) as well as at upper levels (not shown) and with insignificant precipitation (Fig. 6), it facilitates the stagnation of air and exacerbates BC and other pollutant levels in the atmosphere (Ganguly *et al.*, 2006). During December–March period, light winds at the surface along with stable conditions in the atmospheric boundary layer and limited convection hinders dilution of BC levels in the atmosphere due to weaker horizontal and vertical transport of aerosols, while insignificant precipitation during the same period prevents washout of aerosols and light upper-level winds associated with slow-moving synoptic systems allow accumulation and buildup of BC burden during these months. In fact, as the prevailing surface-level wind flow during the months from December to March is predominantly from the Indian subcontinent toward the Indian Ocean (Fig. 5), large amount of BC emitted from road transport sector in India is transported over the Bay of Bengal and the Arabian Sea, resulting in significantly increased column burden of BC not only over the continental regions but also over these otherwise pristine oceanic regions.

On the contrary, the meteorological conditions over the Indian subcontinent during the period from June to September characterized by strong southwesterly winds near the surface bringing relatively pristine marine air over the polluted continental region (Fig. 5), convectively unstable atmosphere allowing faster horizontal and vertical transport of aerosols thereby diluting the BC concentrations within the atmospheric boundary layer and facilitating cloud processing of BC aerosols mixed with other water-soluble species (Hoose *et al.*, 2006), and significant precipitation across the Indian region due to southwest monsoon resulting in efficient

Fig. 5. Monthly mean climatology (2010–2015) of change in BC column burden (kg m^{-2}) over the South Asian region due to BC emissions from road transportation sector in India as estimated by the CAM5 model and also shown are the wind vectors at 850 hPa from MERRA-2. Stipples in this figure represent areas where the anomalies are at or exceed the 95% confidence level based on the Student's *t*-test.

wet scavenging (both in-cloud and below-cloud removal) of BC aerosols from the atmosphere (Fig. 6). As mentioned previously by Ganguly *et al.* (2012a) that since BC is largely co-emitted with other soluble species, it is rapidly converted to an internally mixed hydrophilic aerosol in the CAM5 model thereby facilitating the uptake of these BC-containing internally mixed hydrophilic aerosols into cloud droplets which further undergoes rapid growth through collision-coalescence process into larger hydrometeors and eventually gets rapidly scavenged from the atmosphere during the monsoon season. Thus we show using the scavenging rates of aerosols available from our simulations that it is mainly due to efficient scavenging of BC aerosols and stronger winds during the southwest monsoon season (June–September) that the contribution of emissions from the road transportation sector in India towards the BC burden in the atmosphere is found to be lowest, while the lower scavenging rates of aerosols and calm wind conditions result in higher BC burden during the March–April period, despite considering no change in emission rates of BC from the road transportation sector

throughout the year in our simulations. In general, the column burden of BC is found to be higher over the Indo-Gangetic belt region compared to other parts of India due to higher emissions of BC from the road transportation sector from this major source region as shown in Fig. S4.

Radiative Forcing Due to BC Emissions from the Road Transport Sector

Fig. 7 shows the monthly mean climatology (2010–2015) of the spatial distributions of the shortwave direct radiative forcing at TOA for all months from January to December over the South Asian region due to BC emissions from the road transport sector in India. Since the magnitude of the shortwave DRF due to OC emissions from the road transportation sector is found to be more than an order of magnitude smaller than the DRF due to BC emissions from the same sector, we limit our discussions to the radiative effects of BC emissions alone in the present work. Spatial distributions of shortwave DRF at TOA for individual months from January to December over the South Asian

Fig. 6. Annual cycle of monthly mean climatology (2010–2015) of average atmospheric BC burden (kg m^{-2}) in India, BC deposition rate ($\text{kg m}^{-2} \text{s}^{-1}$), and precipitation rate (PRECT in mm day^{-1}) simulated by the CAM5 model. Decomposition of total BC deposition rate to wet and dry deposition rates is also shown in the figure.

Fig. 7. Annual mean climatology (2010–2015) of direct radiative forcing (W m^{-2}) (a) at surface, (b) at TOA, and (c) within the atmosphere over the South Asian region due to BC emissions from road transportation sector in India as estimated by the CAM5 model.

region due to BC emissions from the road transport sector in India is shown in Fig. S5. The positive DRF at TOA estimated during all months with values up to approximately 6 W m^{-2} indicates a warming effect over the South Asian region due to BC emissions from the road transportation sector in India. The spatial distributions of DRF at TOA (Fig. S5) for all months are similar but not identical to the pattern of change in BC column burden (Fig. 5), and these differences are likely due to the differences in clouds between the two simulations. It is worth emphasizing here that the radiative fluxes under both all-sky and clean-sky conditions are influenced somewhat differently not just due to differences in BC aerosols between the two simulations, but also due to differences in clouds, and the presence of BC

aerosols relative to (above or below) those clouds under all-sky conditions between the simulations (Ghan *et al.*, 2012). This happens because as discussed earlier in Section 2.7 that the methodology adopted in this study for the calculation of aerosol radiative forcing ensures that while our estimates of DRF includes the radiative warming enhancement by BC aerosols present above clouds or other bright surfaces, it avoids exaggeration of radiative cooling by scattering aerosols above clouds. However, unlike the positive DRF at TOA, BC emissions from road transportation sector in India results in a predominantly negative DRF at the surface across the South Asian region and thereby leading to a net loss of shortwave radiation at the surface by up to around 10 W m^{-2} over certain areas especially in the eastern part of

the Indo-Gangetic belt in some months (Figs. 7, S5, and S6). In fact, as noted earlier in the case of spatial distribution of atmospheric BC burden shown in Fig. 5, the extent of negative DRF at the surface extends well beyond the source regions of these aerosols over the Indian land mass to far-off areas over the Bay of Bengal and the Arabian Sea, particularly during the months from December to April when the prevailing low-level wind is predominantly from the Indian subcontinent toward the Indian Ocean and large amounts of BC emitted from the Indian land mass is transported over these oceanic regions (Fig. S6). Thus while a positive DRF at the TOA (Fig. 7) refers to a gain of additional energy for the earth-atmosphere system by up to 6 W m^{-2} , a negative DRF at the surface in spite of this gain of energy at the TOA means that there is a net loss of energy at the surface of the earth by up to 10 W m^{-2} , thereby indicating a trapping of energy within the atmosphere by up to 16 W m^{-2} due to the direct radiative effects of BC emissions from the road transportation sector in India and as estimated by taking the difference of the DRF values at the TOA and surface (Figs. 7 and S7). Fig. 8 shows annual cycle of monthly mean climatology (2010–2015) of BC direct radiative forcing averaged over the Indian region at surface, TOA, and within the atmosphere as estimated by the CAM5 model. The annual cycles of BC direct radiative forcing (Fig. 8) follows the same pattern as the BC column burden shown earlier in Fig. 6. We find the DRF at surface to be maximum (in terms of magnitude) during March when the BC column burden is also highest due to a buildup of BC associated with weaker horizontal and vertical transport of aerosols and less scavenging of aerosols due to insignificant precipitation during this time of the year, while DRF at surface to be lower (in terms of magnitude) during monsoon season due to lesser BC column burden during this season. We also find that the annual cycle of atmospheric absorption due to BC (Fig. 8) as estimated by taking the difference between DRF at the TOA and DRF at surface, closely follows the annual cycle of BC column burden (Fig. 6) thereby

showing that the atmospheric DRF due to BC is directly proportional to the BC column burden in the atmosphere.

As discussed earlier in Section 3.3 that the emission estimates of BC from the road transportation sector, as reported in this study are associated with an uncertainty of about 50%. Hence, with an aim to understand the uncertainty in our estimates of DRF due to BC aerosols arising due to the uncertainty in emissions of BC from the transportation sector in India, we perform two additional model sensitivity experiments where in one case we increased the BC emissions from the road transportation sector in India by 50% and in the other case, we decreased the same emissions by 50% of the mean value used previously in our standard simulation (see Section 2.6 for the simulation setup of these sensitivity experiments). In addition to providing us an idea about the uncertainty associated with our estimates of DRF, another usefulness of the results of our sensitivity simulations is that it also helps us to answer policy-relevant question such as the change in BC forcing that we can expect to get from changes in emissions caused by either future emission increases or implementation of deliberate mitigation measures involving cutting down emissions of BC. Fig. 9 shows the scatter plot of monthly mean DRF at TOA, surface, and atmosphere with increase in column burden of BC for individual model grid points within India as well as averaged over all grid points across India from the three sensitivity experiments. Our estimates of DRF due to BC from these sensitivity experiments reveal that the DRF remains uncertain up to approximately 35% due to uncertainties in the emission estimates of BC from transportation sector in India. Interestingly our results suggest that the DRF at TOA, surface, and atmosphere due to BC aerosols exhibit an approximately linear relationship with column mass burden of BC across the Indian region and for the range of BC emissions considered in this study. Since the estimates of BC emissions and hence the column mass burden of BC over the Indian region continue to remain uncertain, this linear relationship between the column burden

Fig. 8. Annual cycle of monthly mean climatology (2010–2015) of BC direct radiative forcing (W m^{-2}) averaged over the Indian region at surface, TOA, and within the atmosphere as estimated by the CAM5 model.

Fig. 9. Scatter plot of monthly mean direct radiative forcing (W m^{-2}) at TOA, surface, and atmosphere with increase in column burden of BC (kg m^{-2}) for individual model grid points within India (panels (a), (b), and (c) respectively) and averaged over entire India (panels (d), (e), and (f) respectively) from the control experiment and two additional sensitivity experiments for the period 2010–2015. Horizontal and vertical bars in panels (d), (e), and (f) represent the standard deviation in BC burden and radiative forcing values across all grid points within India about their mean values in the respective experiments. Annual mean climatology (2010–2015) of normalized DRF (in units of W mg^{-1}) at TOA, surface, and the atmosphere due to BC aerosols are shown in panels (g), (h), and (i) respectively.

of BC and the DRF due to BC aerosols allows us a more useful way of presenting the DRF due to BC as the normalized DRF or the DRF per unit column mass burden of BC in watts per milligram (W mg^{-1}). Fig. 9 also shows the annual mean climatology (2010–2015) of normalized DRF at TOA, surface, and the atmosphere due to BC aerosols. We find the

normalized DRF at TOA, surface, and atmosphere due to BC averaged over India to be around $+1.3$, -2.5 , and $+3.8 \text{ W mg}^{-1}$ respectively with an uncertainty of about 61% due to uncertainties in the DRF and column mass burden of BC. It is worth noting from Fig. 9 that unlike the spatial distributions of DRF due to BC (Fig. 7), the spatial distributions of

normalized DRF due to BC appears to be significantly different from the pattern of change in BC column burden (Fig. 5) and in fact the normalized DRF due to BC appears to be more spatially uniform in general and particularly for the normalized DRF values at the surface. The spatial homogeneity in normalized DRF due to BC at the surface (Fig. 9(h)) regardless of the actual column burden of BC exhibiting significant variability across the Indian region (Fig. S8) is also evident from the strong linear correlation between the increase in column burden of BC and the DRF due to BC at the surface (Fig. 9(b)). Thus, our estimates of normalized DRF due to BC is also useful for knowing the change in BC forcing that we can expect to achieve from either future emission increases or implementation of mitigation measures involving cutting down BC emissions from particular sectors. Interestingly, our results show that the spatial variability of the normalized DRF due to BC at TOA (Fig. 9(g)) is not only significantly different from the pattern of change in BC column burden, but also the normalized DRF due to BC at TOA gets amplified over areas characterized by the presence of these strongly absorbing BC aerosols above highly reflective surface including clouds (Figs. S9 and S10). For example, the normalized DRF at TOA and atmospheric absorption due to BC noted over Jammu and Kashmir in the north and western side of the peninsular India in the south (Figs. 9(g) and 9(i)) in spite of significantly lower column burden of BC existing over both these regions compared to highly polluted regions elsewhere such as the Indo-Gangetic belt (Figs. 5 and S8) are noted primarily due to higher surface albedo over Jammu and Kashmir in the north (Fig. S9) and persistent presence of highly reflecting low clouds over the western side of the peninsular India (Figs. S10 and S11) almost throughout the year resulting in an absorption amplification of shortwave radiation per unit column burden of BC over these regions. Consequences of such high values of normalized DRF at TOA and atmospheric warming due to BC over the Himalayan Karakoram region as well as over the peninsular India are not easy to speculate, but it is surely a subject of further investigation by the scientific community to understand the potential impact of such enhanced atmospheric warming on the melting of snow and glaciers over the Himalayan region as well as on the onset of the South Asian summer monsoon, regional climate and the hydrological cycle (Ramanathan *et al.*, 2005; Lau *et al.*, 2006; Meehl *et al.*, 2008; Randles *et al.*, 2008; Lau *et al.*, 2010; Ganguly *et al.*, 2012b; Yoon *et al.*, 2016).

Previous studies have shown that while the aerosol-induced negative forcing at the surface resulting in a cooling of the surface including the sea surface temperatures over the northern Indian Ocean during the pre-monsoon season (March–May) could lead to a weakening of the South Asian summer monsoon system (Ramanathan *et al.*, 2005; Chung *et al.*, 2006; Ganguly *et al.*, 2012a), the BC-induced trapping of energy within the atmosphere resulting in a warming up of the lower troposphere where the BC aerosols are mostly confined could also increase the near-surface atmospheric instability thereby intensifying the monsoon system (Lau *et al.*, 2006; Meehl *et al.*, 2008; Wang *et al.*, 2009; Ganguly *et al.*, 2012b). Hence, we wish to mention here that while the

ultimate impact of carbonaceous aerosol emissions on the South Asian monsoon system and the regional climate continues to be an important research question, which demands further investigation involving improved regional emissions from all sectors, but it is beyond the scope of the present study, and therefore it will be taken up as a separate study in the future.

SUMMARY AND CONCLUSIONS

Using detailed EFs for on-road vehicles categorized by age to decrease the uncertainty associated with the emission estimates, new emission inventories for the PM_{2.5}, BC, and OC for the on-road transport sector in India during 2013 were developed. The total national consumption of fuel (diesel, gasoline, and CNG) was estimated and then disaggregated at the state level. We discovered that the category of super-emitter vehicles contributed nearly 53–68% of the total PM_{2.5}, BC, and OC emissions from the on-road transport sector, with emissions from a super-emitter vehicle being significantly higher than those from a non-super-emitter. The continued growth of the GDP and urban population will likely result in a steep rise in the number of on-road vehicles across India in the coming years; therefore, increased gaseous and particulate matter emissions are expected. In addition, the current trends of poor vehicle maintenance and the absence of after-treatment technologies will likely elevate emissions of climate-relevant pollutants, which may severely affect the climate and local air quality, causing various health problems.

We substituted IPCC AR5's default estimates of carbonaceous aerosol emissions from the road transport sector with figures from our newly developed local emission inventory for India and merged our data with the global emission inventory. Using the CAM5 global climate model, we then estimated the BC column burden in the atmosphere and the aerosol direct radiative forcing due to BC emissions from the road transport sector in India. Our model simulations indicate that although the column burden of BC is generally higher over the Indo-Gangetic belt than other parts of the subcontinent regardless of the month, large quantities of BC emitted from this sector are transported across the Bay of Bengal and the Arabian Sea, resulting in a significantly increased column burden of BC over these oceanic regions. The BC emissions associated with this sector result in a positive DRF, with values of up to 6 W m⁻², at the TOA, suggesting that they cause a warming effect across the South Asian region. Despite the gain in energy at the TOA, a predominantly negative DRF, revealing a net energy loss of up to 10 W m⁻², was observed at the surface. The total estimated DRF values (16 W m⁻²) indicate that energy remains trapped within the atmosphere above the Indian subcontinent solely because of the road transportation sector. Furthermore, the normalized DRF (the DRF per unit column mass burden of BC) attributable to the total BC averaged across India at the TOA, at the surface, and in the atmosphere was estimated to be approximately +1.3, -2.5, and +3.8 W mg⁻¹, respectively, with an uncertainty of ~61% due to uncertainties in the DRF and the column mass burden of BC. In spite of column burdens of BC that were significantly lower than elsewhere in India,

such as the Indo-Gangetic belt, we found exceptionally high values for the normalized DRF at the TOA and for atmospheric absorption due to BC over the region of Jammu and Kashmir in the north as well as the western side of peninsular India due to higher surface albedo in the former and the persistent presence of highly reflective low clouds in the latter, which amplified the absorption of shortwave radiation per unit column burden of BC. Finally, although the overall effect of carbonaceous aerosol emissions on the South Asian monsoon system is undoubtedly a critical issue, which demands further investigation and a decrease in regional emissions from all sectors, it is beyond the scope of the present study.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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