## **Response to Reviewers: "Monthly and Spatially Resolved Black Carbon Emission Inventory of India: Uncertainty Analysis"**

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Abstract. We have received and are grateful for the response provided by the two anonymous reviewers of our manuscript. The comments show a detailed and good understanding of the intention of the dataset and ambitions that we have for the scientific community. Further, they show a careful consideration of the data we used and amount of work that has gone into this 'bottom-up' analysis. Individual comments are addressed separately in the following sections. Original comments are italicized, with our responses following.

#### **1** Reviewer #1 comments

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AR-1 raised very good concerns, many of which are common with our own. We feel that the biggest challenge with providing accurate assessments of the climatologically significant black carbon aerosol arises from having an accurate estimate of the emissions. We have provided our emission inventory as an attempt to provide a detailed inventory that provides both temporal variability and a good statistical evaluation of the uncertainty with the different sources.

These are few raised concern by referee:

1) To build a scientifically viable BC emission inventory for developing country like India is very curial and sensitive for climate scientific point of view. I am happy that author has adopted some approach to build an inventory but same time I am not agreed with the methodology and approached they adopted. Moreover the spatial data used to build such

15 a fine resolution inventory is not up to mark and nearly impossible. To use the inventory in atmospheric model, it needs to be scientifically valid rather than just to adopt a statistical approach to come up an inventory.

We recognize that methodologically there are several different approaches that may be utilized for the development of an emission inventory. However, the reviewer provides no example of alternative methods they would recommend. It seems the primary concern of the approach employed in our work is related to the 40 km. resolution that we have used to prepare the

20 data. There are inverse methods that could be employed to address these concerns, but beyond the fact that such an analysis would be beyond the scope of this work, some of the methods require a good 'bottom-up' inventory as a starting point (e.g. Wang et al., 2016). We feel this is precisely where our work contributes, and are enthusiastic for inclusion in such an analysis. Further, we are aware of other recent work to assess the quality of emission inventories using inverse modeling methods (e.g.

Fang et al., 2014; Thompson and Stohl, 2014.), but note that these are generally limited to species for which "the atmospheric loss (if any) can be described as a linear process and can be used on continental to regional and even local scales with little or no modification." Thompson and Stohl (2014). For black carbon this assumption would be invalid.

- 2) The title of manuscript clams its spatially resolved BC emission inventory of India But in reality there is very little 5 or very poor spatial/geographical data are being used to build a 40x40 km resolution inventory. In fact there is no road network data is used or any discussion. If you input emission estimations are at such a coarse level i.e. district and state level then how can you generated spatially resolved inventory at such fine resolution? It is very easy to interpolate in GIS environment to any finer resolution. This kind of inventory will mislead in further application is in climate or chemistry model. It is likely to introduce further uncertainty in terms of spatial allocation.
- 10 We are glad that the reviewers raised this issue. The uniform distribution of emissions in a district from point sources would have definitely reduced the accuracy of our dataset. We have now changed the method and a considerable effort was undertaken to resolve this concern by using the precise location coordinates of industrial structures and locating them in a grid element. This way the emissions from a point source would only be highlighted in a particular grid as compared to uniform distribution in the complete district previously. We have done this analysis for all the sources in Industry category (Steel Plants, Cement
- 15 Plants, Coal & Diesel Power Plants and Sugar Mills) except Brick Industry for which little or no data is available regarding their location.

We also found the reviewers suggestion regarding the use of road network data constructive, and have now used this information in distributing emissions from districts to grids. Road network data from Open street maps has been used for this process. The emissions were distributed from districts to grids according the proportion of road length of the district contained

20 in the grid. In the case of a grid lying in multiple districts the emission in each segment of grid was estimated according to the aforementioned methodology and emissions from all these segments were added to get emission in a grid. Text indicating the changes in method have been included in Sections 2 & 3. We believe this change will definitely improve the quality of our dataset and thank reviewers for raising the concern.

3) I would like to suggest author to enlighten the detail of industrial units/ activity data used/its spatial pattern over
 India for present work. There is little discussion about activity data for each sector. I will suggest elaborating each dataset quantitatively.

We were reluctant to include substantial information regarding the activity data in order to keep the manuscript succinct. However, in our revisions we have tried to appropriately detail each activity dataset further in the new draft of the manuscript.

4) There is no road network data used to allocate transport emissions is another big gap in developing robust and reliable inventory.

Accepted. Kindly refer Point 2.

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#### 5) Some sectors like Mobile towers, etc. are calculated at very course resolution like state level.

The reviewer is correct that the initial data used in our inventory for some emissions is from state-level reports or available data sets. However, for none of the sources did we consider the emissions only at the state-level. In the case of mobile-towers or

35 municipal waste burning, we used district-level population data to distribute the emissions to a finer resolution. For agricultural

burning, we used the district-level reporting of net sown area. We are confident that this is a reasonable method to bring greater granularity to the data and distribute it to a higher resolution.

Lack of data was a serious challenge in the development of this inventory. For instance, India has around half a million mobile towers with information regarding their location only available at state level. We wanted to make the inventory as
comprehensive as possible by considering all the significant sources. In view of lack of data we had to use suitable proxies to get data as a finer resolution. Also note that, Mobile towers contributes less than 0.5% to the national emissions and so would not have significant impact on the overall magnitude of emissions.

6) An inventory has to build based appreciate emission factor which is scientifically correct and suitable to Indian condition rather than just emission factor/activity data generated based on statistical approach. An emission inventory

- 10 like climate agent BC has to be scientifically correct in terms of selection of emission factors data due to existing large uncertainty of the order of 2 to 4. We have to very careful in selecting the EFs as most of available EFs are generated for developed countries except in few cases. It is extremely important to select an EF suit to Indian condition rather than taking EF derived from simple statistical approach. This is a ongoing big issue and major reason of debate among scientific community across the globe. It has to be tackle scientifically instead of statistically.
- 15 We recognize that the lack of country specific emission factors is a big challenge in development of an accurate emission inventory and we share this concern. However, for a country like India with such a vast geography and variation in terms of technologies used for combustion processes even a country specific single emission factor would not do justice. For instance, considering firewood combustion as a domestic fuel, each community uses a different type of cooking stove and likely even different types of wood. With the difference in structure of the combustion technology, emission factors are bound to vary and
- 20 a single emission factor would not be sufficient to address the variation. This is exactly the issue we aimed to resolve through the statistical approach to emission inventories which enables us to better preserve and quantify the uncertainty. We wish to assess the uncertainty in the emissions in India because of these variation in technologies and the paucity of data associated to them.
- We absolutely agree that it would be incorrect to use the emission factors derived for developed countries for emission estimation in India. In view of this, most of our emission factors are from studies conducted in India , China or South Asia. In cases where we have derived factors from the global studies Bond (2004); Streets (2003); Akagi et al. (2011); Ito and Penner (2005) etc.) mostly we have used the emission factors explicitly given for India/South Asia/Developing country.

7) Emission inventory are never being interpolated with coarse level emission estimation i.e. state and district level data. I can agree for state/district level national estimation but can not with 40x40km surface emission date.

30 We have used strong and best available proxies to resolve data to a finer resolution(Population for domestic fuels, net sown area for agricultural residue burning) which should provide a fair estimate of emissions. Further, the changes that we have made in our methodology on the recommendation of both reviewers (inclusion of road network data, treating industrial units as point source) improves the accuracy of our 40x40 km2 spatially resolved dataset.

8) I am very surprised that NCT contributes just 6.7 Gg/yr of BC but in reality NCT account nearly between 5-10 %
 of transport and industrial load in India with high population density. This number appears to be very small and unre-

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alistic. I will recommend author to recalculation of emission especially sensitive regions like NCT and other industrial regions. These regions are very sensitive due to air quality issues in recent times. I pretty sure the BC estimation over NCT should have been much higher than author's calculation. Otherwise this kind of miss representation may be due to unappreciated emission factor and approach used for estimation.

5 We think reviewer mistakenly considered NCT (National Capital Territory) as NCR (National Capital Region). NCT is another name for Union Territory of Delhi and includes the seven districts of Delhi. On the other hand, NCR contains NCT and 19 other districts nearby to NCT. The generally quoted high emission figures represents the emissions from NCR rather than NCT which is 20 times the area and contains thrice the population of NCT.

In the last decade Govt. of Delhi has taken serious steps to reduce the emissions in Delhi. It has closed almost all of the industries in Delhi, made CNG compulsory for Govt sponsored public transport etc. which have reduced the emission in NCT significantly.

#### 2 Reviewer #2 Comments

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AR2 raises some similar concerns to that of AR1. Further, the reviewer has highlighted the quality of language usage and writing style in the article, which we appreciate. However, common with AR1 they raise concerns about the general approach and methodology employed to generate the emissions at the higher resolution.

I have only good words to say about this article. It is one of the easiest articles I have ever read and it flows very well for the reader. The science behind is novel and it would be a great tool for any modeller wants to include emission data from India to any transport, chemistry or climate model.

Thank you for the kind words, we are glad to hear that it reads well for the reviewers.

### 1) I am sceptic about the methodology used to create this spatial resolution, although the input was at a district level, hence, in course resolution. However, if we exclude inverse modelling approaches that would be able to create a fully resolved inventory, I admit this approach is the best we have.

Unfortunately, we agree. The district-level data is the finest available presently, and hence why we feel it is important to include the uncertainty analysis and several approaches for calculation of the EFs and multiple assessments of activity data. Our approach has been similar to that employed by Qin and Xie (2011) for China, and we feel it offers a first-of-kind inventory for India that can be evaluated / employed in other approaches.

Regarding the inverse methods, as discussed above, there have been a few approaches to employ such an approach to black carbon (e.g. Wang et al. (2016); Hakami et al. (2005). We feel however, that the level of uncertainty introduced in the meteorological modeling and furthermore chemical characterization and treatment of the black carbon aerosol in these

30 approaches is as significant a source of uncertainty as the sub-district level distribution. Loss processes associated with black carbon aerosol behave non-linearly in the atmosphere, therefore it is a challenge to use an inverse approach. If one employs a lagrangian approach (e.g. Thompson and Stohl (2014)) they would violate assumptions of linear loss. Therefore, it is perhaps an improvement to use a full chemical transport model such as GEOS-CHEM (e.g. Fu et al. (2012)) or STEM (Hakami et al. (2005)). The challenge with such an approach is that these models utilize Eulerian discretization and the grid resolution create numerical effects that misrepresent the true fillamental transport processes associated with an aerosol like black carbon Sodemann et al. (2011).

2) Another problem is the emission factors used in the study. I expect that the authors will justify more their choice of emission factor selection, since there are already-known and validated emission factors in the literature used for such purposes (showed also in Table 2). I am not sure if taking the mean emission factor would make sense since in some cases it varies a lot. In addition, I do not think that emission factors taken from Andrae and Merlet's paper (used in most models) can be given the same weight with other emission factor studies. Of course, since they do not validate their dataset, they do not need to evaluate the emission factors they used.

10 A similar concern was also raised by AR1. Our choice of emission factors was aimed to appropriately account for the variation in fuel combustion technology across the vast geography of India. Fuel combustion technologies vary significantly especially in domestic fuel category (Which accounts for almost 50% of BC emissions in India). This was one of the issues we wanted to address through the statistical approach in emission inventory development.

Most of our emission factors are derived from studies conducted in India , China or South Asia. In cases where we have 15 derived factors from the global studies (Bond (2004); Streets (2003); Akagi et al. (2011); Ito and Penner (2005) etc.) we have almost exclusively used the emission factors explicitly given for India/South Asia/Developing country.

3) I think that a missing point is evaluation of the database created from this work. I would highly recommend to the authors to try including this dataset to a global inventory and perform a few runs comparing the surface concentrations of BC from their model and the dataset with observations in Southeastern Asia. There are several measuring stations there and such a comparison would help all of us understand how valid this dataset is. At the moment, a poor discussion is performed for such an interesting topic. I believe that including the dataset to a model and performing the aforementioned analyses would expand the discussion a lot.

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Similar to (Qin and Xie, 2011) we do not include an evaluation. In fact, we find it interesting that the reviewer assumes we could simply conduct an analysis 'from their model'. We believe strongly such an analysis is not a casual affair and
should be conducted in a robust fashion, particularly given the aforementioned challenges associated with black carbon. To do such a study correctly would be beyond the scope of this study. A tremendous amount of effort goes into the collection and identification of valid sources of information for the inclusion in an emission inventory and is not necessarily work that would be conducted by the same scientists best suited to conduct transport simulations studies.

That being said, we obviously intend and hope that such an analysis be conducted. It is exactly toward this ambition that we 30 publish the inventory and we recognize that validation of the data set is an important process. We agree that the discussion on this topic needed improvement, and we have included some further discussion on the importance of this aspect.

4) My highest concern is if the present manuscript falls within the scope of the journal. According to the main page of the journal, it is "dedicated to the publication and public discussion of studies investigating the Earth's atmosphere and the underlying chemical and physical processes in an altitude range from the land and ocean surface up to the turbopause, including the troposphere, stratosphere, and mesosphere. The main subject areas comprise atmospheric

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modelling, field measurements, remote sensing, and laboratory studies of gases, aerosols, clouds and precipitation, isotopes, radiation, dynamics, biosphere interactions, and hydrosphere interactions. The journal scope is focused on studies with general implications for atmospheric science rather than investigations that are primarily of local or technical interest. In my opinion, the present study comprises a rather statistical methodology than a modelling, measurements,

5 lab-based methodology (perhaps a bit of GIS) that the journal requires. Furthermore, without the evaluation using model-simulated concentrations and comparison with measurements of the Southeastern Asia, the study is of local interest.

We are somewhat surprised by this comment. Starting at the end, we strongly feel that the development of a detailed and accurate emission inventory of black carbon for India has global significance. There are several examples of similar articles,

10 predominantly for China that provide novel approaches to developing higher resolution emission inventory data. For example similar manuscripts in ACP that deploy a bottom-up approach toward the development of emission inventories include those found in(Qin and Xie, 2011), (Kuenen et al., 2014) who use a novel proxy-based approach to distribute the data, and (Zhao et al., 2015) who have generated a city-scale high resolution inventory and compare with prior inventories.

A general comment is that the Editor should have probably rejected the present manuscript in its current form. I have seen several more subject-related manuscripts being rejected. However, I am glad he did not and the authors have the chance to provide a very useful database to the modelling community (after evaluation of their results, of course).

We are also glad that the editor has not rejected the manuscript and provided us the opportunity to address the good comments provided by the reviewers. In particular, we have included greater detail regarding the distribution of the transport-sector emissions through the inclusion of a road network – a concern raised by both reviewers. We also feel the valuable comments and reviews have contributed to the discussion, and we have made some additions that address the reviewers concerns.

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# Monthly and Spatially Resolved Black Carbon Emission Inventory of India: Uncertainty <u>AnalysesAnalysis</u>

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**Abstract.** Black Carbon (BC) emissions from India for the year 2011 is estimated to be  $892.12\pm 151.53$  Gg/yr based on a new ground-up, GIS-based inventory. The grid-based, spatially resolved emission inventory includes, in addition to conventional sources, emissions from kerosene lamps, forest fires, diesel-powered irrigation pumps and electricity generators at mobile towers. The emissions have been estimated at district level and were spatially distributed into grids at a resolution of 40x40 km2.

- 5 The uncertainty in emissions has been estimated using Monte Carlo simulation by considering the variability in activity data and emission factors. Monthly variation of BC emissions has also been estimated to account for the seasonal variability. To the total BC emissions, Domestic Fuels contributed most significantly (48%), followed by Industry (22%), Transport (17%), Open Burning (12%) and Others (1%). The spatial and seasonal resolution of the inventory will be useful for modeling BC transport in the atmosphere for air quality, global warming, and other process-level studies that require greater temporal resolution than
- 10 traditional inventories.

#### 1 Introduction

Carbonaceous aerosols, defined as Black Carbon (BC) also known as Elemental Carbon (EC) and Organic carbon (OC) (Pachauri et al., 2013), form a significant and highly variable component of atmospheric aerosols. Neither BC nor OC has a precise chemical definition. OC includes numerous organic compounds, some of which are found to be carcinogenic, such

- 15 as Poly Aromatic Hydrocarbons (PAHs) (Menzie et al., 1992; Pedersen et al., 2005). The Intergovernmental Panel on Climate Change (IPCC) defines BC as "Operationally defined aerosol species based on measurement of light absorption and chemical reactivity and/or thermal stability" (IPCC, 2013). BC is released from incomplete combustion of carbonaceous fuels such as agricultural and forest biomass, coal, diesel, etc. The type of combustion greatly affects the BC emission rates, notably; inefficient combustion emits more BC than efficient combustion for the same type of fuel. Aside from air quality and health effects,
- 20 there are a number of climate impacts of BC emissions including alterations to temperature through atmospheric adsorption, modifications to precipitation timing, increased melting of snow (Meehl et al., 2008; Flanner et al., 2007; Ramanathan and Carmichael, 2008; Quinn et al., 2007; Koch and Del Genio, 2010; Bond et al., 2013) all of which are consequential to global warming. BC has been implicated to be the second largest contributor to global warming after CO2 (Ramanathan and Carmichael, 2008).

Carmichael, 2008). There is a current debate that due to the short life span of BC, the BC atmospheric concentration will drop quickly if emissions are reduced, thereby potentially offering a rapid means to slow down global warming (Bond and Sun, 2005; Grieshop et al., 2009; Kopp and Mauzerall, 2010; Bowerman et al., 2013).

- India is a rapidly growing economy with massive future growth potential. The total energy and coal consumption has almost doubled from 2001 to 2011 (IEA, 2012). The emissions of particulate matter or aerosols have been rising over the last few decades and are expected to increase in the future as well, due to rapid industrial growth and slower emission control measures (Menon et al., 2010). Recent studies (Yasunari et al., 2013; Lau et al.) have shown that the deposition of BC in the Himalayan glaciers has accelerated their melting. While BC is a source of warming at global scale, at a regional scale, it has adverse effects on air quality and human health. BC is a major part of particulate matter with size less than 2.5 micron (PM2.5), and like other
- 10 PM2.5 particles, it is small enough to be inhaled. According to the World Health Organization (WHO), exposure to BC can lead to cardiopulmonary morbidity and mortality. WHO also suggests that BC may act as a universal carrier of chemicals of varying toxicity to lungs (Janssen et al., 2012). Understanding the sources of BC, their emissions and spatial distribution is important both for policy making and improving climate modeling. Preparation of an accurate emission inventory is the first step towards developing robust air pollution control strategies. Air quality measurement stations are installed at limited
- 15 locations and are unable to provide a measure of spatial variability. However, observations coupled with air quality models, can provide comprehensive information about the impact of various sources on ambient air quality and their spatial variability. The greatest benefit of these models is gained after preparing an accurate emission inventory, validating the models with observations, thereby enabling a tool for improved control measures.
- Although there have been several emission inventories developed for BC in the last decade, the estimates are variable without any knowledge of uncertainties. Model-predicted BC concentrations over India are 2 to 6 times lower than the observed concentrations (Ganguly et al., 2009; Nair et al., 2012; Bond et al., 2013; Moorthy et al., 2013). Further the current estimates vary considerably; Reanalysis of tropospheric chemical composition (RETRO) emission inventory (Schultz et al., 2007, 2008) estimated BC emissions in 2010 as 697 Gg/yr, the System of Air Quality Weather Forecasting and Research (SAFAR) emission inventory (Sahu et al., 2008) estimated it as 1119 Gg/yr for year 2011, Klimont et al. (2009) reports BC emissions as 1104
- 25 Gg/yr for year 2010 and Lu et al. (2011) reported it as 1015 Gg/yr for year 2010. Not only there is a need to get a meaningful total estimate but also to assess the uncertainty and spatial variability associated with these estimates. Most of the emission inventories provide yearly emissions and do not account for sub-annual temporal emission variability which leads to inaccurate impact assessments. To improve the nature of advanced numerical forecasts of impacts from aerosol pollution, we have developed an emission inventory at monthly resolution.
- The objective of this study is to prepare a sub-annual, high spatial resolution, comprehensive spatially gridded emission inventory of BC emissions for India for the base year 2011. The approach is a ground-up inventory based on activity data from various sectors, combined with emission factors. While results are provided for one year, the frequency and distribution should be general enough such that coupled with growth forecasts, multi-year use could be valid. In this study, we have prepared a district-wise emission inventory available on a  $40 \times 40 km^2$  grid. We have accounted for all the major sources of BC emissions
- 35 in India. For example, emissions from kerosene lamps (Lam et al., 2012) and forest fires which were previously unaccounted

for in many emission inventories have been included. Monthly variation of BC emissions has also been estimated to provide better input for air quality models. We employ a unique approach to quantify uncertainty in the emissions by considering variability in (i) activity data from various sources, and (ii) emission factors (EFs). Specifically, probabilistic distributions were assigned to both activity data and EFs. By employing Monte Carlo simulation, several activity level and EFs were generated

5 to arrive at emissions (by multiplying generated activity data and EF) which could be interpreted in terms of a mean value and associated uncertainty.

In Section 2 we present the methods used in our analysis, Section 3 describes the source sectors and activity data we considered, a description of the magnitude of emissions from each sector is presented in Section 4.

#### 2 Methods

10 Our approach may be divided into two parts. Figure 1 presents the methodology for developing national emissions and their uncertainty and Figure 2 presents the approach for extracting gridded emissions. For estimating national emissions, a thorough review of multiple national activity data and EFs for each source was conducted from available published and unpublished sources (Table 1 and Table 2).

We fit a probability distribution function (PDF) to both national activity data and EFs from a pool of distributions on the 15 basis of Kolomogorov-Smirnov test (KS statistic) using Mathwave Technologies EasyFit© software (Mathwave Technologies, 2015). Using the optimal probability distribution function (PDF) for both variables (EFs and activity data) for each source, we generated 1000 estimates of each variable from each of the two distributions. Further increasing the number of generations did not change the mean and the variance of the emissions.

For activity data that had only one source of information, a normal distribution with mean as the data point and standard
deviation of 20% of the data point was assumed based on the experience of other data sets (Table 1). Best fit distributions were only determined from KS statistic if the number of data points exceeded 5, in other cases, a uniform distribution was assumed. For preparing the gridded inventory, the emissions were first estimated within a Geographic Information System (GIS) using polygons at the district level. Data were subsequently interpolated to 40x40 Km2 grids . Polygons were subsequently dividing into 40 x 40 km<sup>2</sup> grid elements, and were proportionally assigned emissions based on area. The area for grid elements spanning

- 25 a district border was accounted for. Emissions from Industry (point data) were added directly to the overlying grid based on available location coordinates for the source. For the Road Transport (network) sector, the data from at the district level was distributed along the road network and then assigned to overlying grids proportionally to the length of road in the grid element. Interpolation of the data was not conducted, as this would lead to erroneous georeferencing of emissions, particularly in the case of point data. More details are found in the subsections below.
- 30 For the national level annual inventory, Monte Carlo simulations were undertaken to specifically estimate mean emissions and uncertainties, whereas, at the district level the mean of the EFs and district level activity data were used to arrive at average emission levels. An image of the political map of India (Census of India, 2011) was georeferenced using Google Earth and 640 districts were digitized as polygons to generate a national level shape file. This shape file had an attribute table containing all

the districts, and yearly emission quantities were recorded for each district. The shapefile and polygon data were resampled to a  $40 \times 40 km^2$  grid by calculating the area of each portion of the districts within a grid element, and attributing that portion of the emissions to the grid. As a grid cell may overlay over more than one district, the overall emission in each cell was calculated by summing up part of emissions from each contributing portion from the district, based on area of the district within the grid cell and emission density for the district:

$$E_{cell} = \sum_{i=1}^{n} (\rho_i \cdot A_i) \tag{1}$$

Where *n* is the total number of districts within each grid cell,  $\rho$  is the Emission Density  $(g/sm^2)$  for each district and *A* is the Area of the district  $(m^2)$  within the grid. Emission density (mass/time-area) was calculated by dividing the BC emission in the district with the total area of the district.

#### 10 3 Source Sectors and Activity Data

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The emissions sources considered in this study can be broadly categorized into five sectors: Open Burning, Industry, Transport, Domestic Fuel, and Others. In the following section we define the activity data and emission sources considered within each sector. All the emission sources identified by Reddy and Venkataraman (2002b) Reddy and Venkataraman (2002a, b) and Sonkar (2011) were included in this study. Also some of the highly emitting sources identified in the recent literature (Kerosene

15 lamps, Diesel generators) were also considered. Table 1 and 2 provide an overview of activity data and EFs for the sources considered.

#### 3.1 Open Burning

The open burning sector includes: forest fire emissions, open solid waste burning, and agriculture residue burning.

#### 3.1.1 Forest Fire

- 20 According to the 2013 Forest Survey of India (FSI), around 50% the forest area of India is prone to forest fires (FSI, 2013). There is a strong seasonality associated with forest fires in India with majority of fires occurring in the months from February to July. The causes of forest fire in India are both man-made and natural; natural causes being the high temperature and low humidity. Man-made causes include accidental fires and forest burnt for shifting cultivation. The forest fire burnt area in this study was determined using MODIS (Moderate Resolution Imaging Spectroradiometer) monthly burnt area product
- 25 MCD45A1 which has a resolution of 500m (Land Processes Distributed Active Archive Center (LP DAAC), 2000). MODIS product MCD12Q1 (500m resolution) was used to define forest cover. The burnt area and land cover products were retrieved from LP DAAC website (https://lpdaac.usgs.gov/).

The methodology used for emission estimation is presented in Figure 3. Burnt area (MCD45A1) and land cover product (MCD12Q1) are available in Hierarchical Data Format - Earth Observing System (HDF-EOS) format and have an Earth

gridded tile area of 1200 km x 1200 km. They were stitched to cover the whole geographical extent of India. The stitched products were converted to GeoTIFF image format and clipped to Indian domain using the ESRI shape file of the boundary of India. The same methodology was used for the burnt area product as well as the vegetation cover. Monthly burned area GeoTIFF images were overlayed on land cover image to determine the monthly forest burnt area pixels, and subsequently the

- 5 forest area burnt. Dry mass per unit area of forest burnt was taken to be 5.2 kg/m2 (Joshi, 1991). Emissions were distributed district-wise according to the number of incidents of forest fire occurring in that district in 2011. The data of district-wise incidents of forest fire were taken from the most recent forest survey (FSI, 2015). Figure 4 shows the land cover image and burnt area image used for estimating the forest fire burnt area in January 2011. It can be noted that the emissions from this subsector can easily be updated for the future years using the latest MODIS burnt area and land cover products and following
- 10 the aforementioned methodology.

#### 3.1.2 Municipal Solid Waste Open Burning

The dry weight content of Indian municipal solid waste (MSW) was estimated using the MSW composition in India (CPCB, 2007) and the dry matter content of MSW components per IPCC 2006 (2006). Indian MSW is primarily composed of vegetables (40%), stones (42%) and grass (4%) which has a dry matter content of 40%, 100% and 40% respectively. Dry matter content

15 was estimated to be 67.6%.

State-wise MSW generated and collected was derived from the Central Pollution Control Board (CPCB) Municipal Solid Waste Management Report 2012 (CPCB, 2012). The MSW generated was distributed among districts according to their urban population. For the states where MSW collected volume was not available, a value of 60% of the total MSW generated was assumed (Kumar, 2010). The total MSW openly burned was taken to be 10% of the collected waste and 2% of the

20 uncollected waste (National Environmental Engineering Research Institute (NEERI), 2010). To provide a second approach for the uncertainty analysis, per capita waste generation in India and fraction burnt was taken from IPCC 2006 (2006). The 2011 census population data were used to provide the urban population of the district. From this, the total MSW burned for each district was taken as the product of the IPCC guideline results and the urban population.

#### 3.1.3 Agricultural Residue burning

25 India generates a large amount of agricultural residues (e.g. waste biomass) every year after harvesting crops. These residues are used as domestic and industrial fuel, fodder for animals, etc. but a large amount remains unutilized in the fields. The quickest and easiest way for the farmers to manage this waste is to burn it. Figure 5 shows a flowchart for estimating emissions from crop residue burning.

The state-wise production of cotton, Jowar, Barley, Jute, Ragi, Rice, Maize, Bajra, Groundnut, Sugarcane, Wheat and 30 Rapseed and Mustard in 2011 was taken from the Ministry of Agriculture, 2012 (www.apy.dacnet.nic.in). The crop production was distributed among districts of that state according to the net sown area (Ministry of Agriculture, 2011) in that district. Emission from crop residue burning was calculated using the following equation as suggested by Jain (2014).

$$ECRB = \sum_{i=1}^{D} \sum_{j=1}^{C} (P \cdot Q \cdot R \cdot S \cdot T \cdot EF_{BC})$$
<sup>(2)</sup>

- where, ECRB is the Emissions from crop residue burning. The summation is done over the Districts, D and for each type
  of Crop, C. The emission is then calculated from the product of Crop production, (P), residue to crop ratio, (Q), dry matter fraction (R), the fraction burnt (S), the fraction actually oxidized, (T), and finally the EF for BC. Three estimates of crop residue burnt (P · Q · R · S · T) were obtained by varying Q, R, and S, while holding P, T constant for all the three estimates. In the first estimate, residue to crop ratio(Q), dry matter fraction (R) and fraction burnt (S) was taken from Jain (2014). In the second estimate, residue to crop ratio and dry matter fraction was kept same and fraction burnt was taken as 0.25 for all
  the crops (IPCC 2006, 2006). In the third estimate, residue to crop ratio and dry matter fraction and dry matter fraction was taken from Venkataraman
- et al. (2006), and fraction burnt was kept as 0.25 (IPCC 2006, 2006). This provided us with three estimates of the total crop residue burnt in the fields (Table 1).

#### 3.2 Industry

Industrial sector includes: brick industry, cement, steel plants, sugar mills and power plants. In general emissions and activity

- 15 data for these sectors are derived from available published reports and scientific literature. We then use location information from each of the facilities to develop district-wise emissions. For some sectors, the district-wise distribution is a function of population density if exact information regarding facility locations cannot be obtained directly. In order to construct the gridded inventory, industrial units were geolocated precisely using the provided GPS coordinates wherever available. In general, geolocated coordinate data is available for Iron & steel manufacturing, cement, and power production. However for
- 20 the two largest sources, Brick Kilns and Sugar Mills, specific geolocation data is not available, adding a source of uncertainty to the analysis.

#### 3.2.1 Brick Industry

The Indian Brick industry has more than 100,000 brick kilns producing 250 billion bricks and consuming about 25 million tons of coal annually (Gupta and Narayan, 2010; Maithel et al., 2012). Bricks in India are produced locally in small enterprises

- 25 at rural scale (Rajarathnam et al., 2014). It is a seasonal industry operating predominantly from months of October to June (Maithel et al., 2012). Brick kilns can be classified into two major categories based upon firing practice: intermittent and continuous kilns. Intermittent kilns include clamp, Scove, Scoth and Down Draft Kilns (DDK) in these kilns bricks are fired in batches. In Continuous kilns brick heating and cooling takes place simultaneously in different parts of the kiln. Several types of kilns including: Bull's Trench Kiln (BTK), Hoffman Kiln, ZigZag Kiln, Tunnel kiln and vertical shaft brick kiln (Heierli
- 30 and Maithel, 2015) operate continuously.

In India a majority of the bricks are produced from Fixed Chimney Bull's Trench Kiln (FCBTK) and clamps (Rajarathnam et al., 2014). There are around 60,000 small scale clamp kilns in India. Located all over India – mostly near or in villages and using biomass, coal, and lignite as fuel (Rajarathnam et al., 2014) these represent an important source of BC emissions. No account of their location, production, fuel consumption and Emission factors have been published. For this study, emissions

5 only from FCBTK's are used which account for 70% of the total bricks produced from India and use coal as the primary fuel (Weyant et al., 2014). The state-wise brick production (in Kg) through these kilns was compiled from consultation with industry experts. It was distributed district-wise according to the population of the districts in the state. The quantity produced was assumed to be normally distributed with 50% standard deviation (Maithel et al., 2012).

#### 3.2.2 Cement Manufacturing

10 The plant-wise cement production in 2011 was taken from Cement Manufacturers Association, Govt. of India (CMA). India had around 150 major cement plants in 2011, which produced 180 million tons of cement and consumed 28 million tons of coal. Cement being a transport-expensive product, plants are evenly distributed across India. Since the plant-wise coal consumption was not available, the national consumption by cement industry was taken from the same source. The fuel consumption was distributed district-wise according to their cement production in 2011.

#### 15 3.2.3 Iron & Steel Production

India produced 68.6 million tons of total finished steel in 2010-11 consuming 40 million tons of coal (Ministry of Steel, 2014). The plant-wise steel production was taken from Press Information Bureau (2011), Govt. of India. The coal consumption was distributed among plants according to their level of steel production. District-wise coal consumption in steel plants was subsequently determined from the location of these plants.

#### 20 3.2.4 Sugar Mills

India ranks second globally in terms of sugar production. Significant BC emissions result from sugar mills due to the usage of bagasse as a fuel. Bagasse is the fibrous residue obtained from sugarcane juice extraction and consists of cellulose (50%), hemicellulose(25%) and lignin(25%) (Ezhumalai and Thangavelu, 2010; Abhilash and Singh, 2008). India has a total of 529 around 550 sugar mills which produced 26.3 million tons of sugar by crushing 361 million tons of sugarcane (Indian Sugar

25 Mills Association (ISMA, 2012; DAC, 2013). Specific geolocated data is not available at higher resolution than the district level. The majority of sugar mills are located in Indo-Gangetic Plains (IGP) and Maharashtra, and population density was used to distribute the emissions. The mill-wise sugarcane crushing capacity was taken from the Department of Food & Public Distribution (DFPD, 2011). The total sugarcane crushed was distributed among mills according to their crushing capacity. The bagasse generated was taken as 30% of the total sugarcane crushed (Pessoa Júnior et al., 1997).

#### 3.2.5 Power Plants

The Indian Central Electricity Authority (CEA, 2012) reports the plant-wise fuel consumption for coal and diesel power plants in India. In 2011, India had a installed capacity of 112 GW of coal and 1.2 GW of diesel based thermal power plants. There are around 100 coal based and 14 diesel based major thermal power plants located across India with location data available from

5 government reports. District-wise fuel consumption was estimated by the location of these plants using the data contained in the report.

#### 3.3 Transport

From the transportation sector emissions from road vehicles, railways, shipping, and aviation have been accounted for individually. For road vehicles, to prepare gridded data from district level emissions, road network data from OpenstreetMap ©.

10 (OpenStreetMap, 2016) was utilized. The data provides a high resolution road network in vector format. The district shapefile, grid polygons and road network shapefile were resampled to a  $40 \times 40 km^2$  grid by calculating the total road length in each portion of the districts within a grid element, and attributing that portion of the emissions to the grid. For non-road vehicles, methodology as discussed in section 2 was used.

#### 3.3.1 Road Vehicles

15 Road vehicles have been divided into seven categories: two wheelers, cars, light motor vehicles (LMV), light commercial vehicles (LCV), taxies, trucks, buses, tractors and trailers.

State-wise number of registered vehicles in the aforementioned categories were taken from Ministry of Road Transport and Highways (2011). The vehicles were distributed among districts of that state according to the population of that district. In determining the emissions for 2011, we needed an estimate of the number of vehicles on road for that year. The reported number

- of registered vehicles represents the cumulative number of first registrations (Parikh and Radhakrishna, 2005). In India, vehicles are neither deregistered when they are no longer in use nor are double registrations deducted. The actual number of vehicles on the road is significantly smaller than the number of registered vehicles. Baidya and Borken-Kleefeld (2009), determined the rolling fleet in 2005 using survival functions. The category-wise number of vehicles on road as a fraction of registered vehicles was taken from Baidya and Borken-Kleefeld (2009). Emissions from road were estimated using the number of vehicles on zo and the annual distance traveled per vehicle type.
- 25 road and the annual distance traveled per vehicle type.

$$EV_{district} = \sum_{i=1}^{n} (N_i \cdot AKT_i \cdot EF_i)$$
(3)

Where EV is the total BC emissions from vehicles for a district (g/district/year), *i* is type of vehicle, *N* is the number of vehicles, AKT is the annual kilometer travelled for the vehicle type (km/year), and EF is the vehicle type emission factor (g/km).

Annual average distance traveled is difficult to quantify and is a source of uncertainty in the emissions. The annual distance traveled by various vehicle types was derived from seven different studies (Table 1). This provided us with multiple estimates of total distance travelled by a vehicles type in a year. For some vehicle types only few BC EFs were available, to compensate for lack of information, EFs were derived from PM2.5 emission factors using BC/PM2.5 fraction given by Chow et al. (2011).

#### 5 3.3.2 Railways

Railways in India are primarily powered by electricity and Diesel, the use of coal has decreased over the years and is negligible now. Annual report (2010-11) of Indian railways enumerates the consumption of diesel and coal (Ministry of Railways, 2012b). The state-wise allocation of fuel consumed was performed according to the railway track length in the state (Ministry of Railways, 2012a) and finally district wise according to the population of the district.

#### 10 3.3.3 Shipping

The Ministry of Petroleum and Natural Gas (MoPNG) reports the total consumption of Fuel Oil (FO), High-speed Diesel Oil (HSDO), and Light Diesel Oil (LDO) by the shipping subsector in 2011 (MoPNG, 2014). According to IPCC guidelines (IPCC 2006, 2006) the fuel used in international bunkers is not counted in the national emission inventory and their estimate is recorded separately. The proportion of shipping fuel used domestically was assumed from the European Environment Agency

15 (EEA, 2013). Due to the non-availability of a spatial proxy, the emissions from ships have not been distributed district-wise and have only been accounted for in the national emissions.

#### 3.3.4 Aviation

The total Aviation Turbine Fuel (ATF) consumption in India was taken from MoPNG (2014). Domestic operations accounts for 38% of the total fuel consumption (ICAO, 2010). Domestic fuel consumption was divided into that used for Landing and Takeoff (LTO) and for cruise operations. The Directorate General of Civil Aviation (DGCA) reports the total number of domestic scheduled and non-scheduled aircraft departures in 2011 (DGCA, 2013). The fuel consumption per LTO was taken from IPCC 2006 (2006). The LTO ATF consumption was distributed district-wise according to the number of flights landing and taking off from the airports in that district. The cruise emission was not distributed and was only counted in national emissions.

#### 3.4 Domestic Fuel

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25 The Domestic Fuel sector includes: emissions from firewood, agricultural residue, coal, liquid petroleum gas (LPG), kerosene (cooking and lighting) and dung cake.

India faces a crucial challenge of providing clean and affordable energy sources to its rural households, especially in the cooking sector. Eighty-five percent of the rural households are still dependent upon traditional biomass fuel for their cooking needs (MoSPI, 2014). Figure 6 shows the distribution of rural households on the basis of energy source used for cooking (MoSPI, 2014).

The stoves used for cooking are inefficient causing incomplete combustion hence releasing more BC than would result from efficient combustion. In the year 2000, domestic fuels contributed 64% to the total BC emissions in Asia (Streets, 2003). State-wise per capita consumption (rural and urban) of firewood, LPG, coal was taken from National Sample Survey (MoSPI, 2014) which releases a report of household consumption of various commodities using a large sample of households every 5

- 5 years. Apart from this, Yevich (2003) report the state-wise total consumption of firewood, agriculture residue and dung cake in 1985. We extrapolated the fuel consumption data to 2011 by using the growth rate of rural population from 1985-1991 and change in number of households using these fuels for cooking from 1991 to 2011. Smith et al. (2000) also report the state-wise consumption of firewood, dung cake and agricultural residue in 1991. We extrapolated the data to 2011 using the change in number of households using these fuels for cooking from 1991 to 2011. Using data from MoSPI (2014), Yevich (2003), and
- 10 Smith et al. (2000), three estimates of domestic fuels consumed in 2011 were prepared and used within the uncertainty analysis. According to the World Bank (2010), 25% of the Indian population does not have access to electricity. As a result Kerosene fueled lamps are the only source of lighting after sunset for a large part of the population. In 2011, over a billion liters of kerosene was consumed to fuel these lamps (MoSPI, 2014). The information on kerosene consumed was available from two sources: MoSPI (2014) and Lam et al. (2012). The National Sample Survey (MoSPI, 2014) reports the state-wise per capita
- 15 (rural and urban) kerosene consumption. The proportion of kerosene used for cooking versus lighting in India was taken from Lam et al. (2012). Another estimate of kerosene consumed in lamps was derived following the methodology described in Lam et al. (2012).

#### 3.5 Other

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Others sector incorporates emissions from use of diesel in power generation for mobile towers and irrigation pumps.

#### 20 3.5.1 Mobile Towers

In 2011 India had more than half a million cell towers (Press Information Bureau, 2011). Most of these towers are located in villages where grid-connected electricity is not available. They use small generators fueled by diesel for their power needs. The total diesel consumption by cell towers was taken from MoPNG (2013). The fuel consumed was distributed state-wise according to the number of mobile towers in that state. It was then distributed district-wise according to the population of the district.

#### 3.5.2 Irrigation Pumps

Agriculture is a core economic activity of India with about 60% of the population involved in the activity. In 2011 India used around 2.4 billion liters of diesel for irrigation pumps (MoPNG, 2013). The use of dug wells and tube wells is very common for irrigation purposes in India. Diesel generators are used to pump water up from mini irrigation schemes. The diesel consumed

30 was distributed district-wise according to the net sown area in that district (Ministry of Agriculture, 2011).

#### 4 Results and Discussion

Tables 1 and 2 present the probabilistic best fit distributions, mean and standard deviation for activity data and EFs for sources considered in this study. The mean district level activity data and EFs were used to estimate the district-wise emissions. It may be noted that kerosene lamps have the highest EF among all sources considered in this study; these lamps convert 8.5% of the

5 fuel directly into BC. In the Open Burning sector, forest fires have the highest EF. In the Industry sector, EF is highest for sugar industry, as the industry uses bagasse as a fuel in a very inefficient combustion process. For Transport sector, EF for diesel operated vehicles (railways, ships, bus, truck, tractor & trailer, LCV) are higher than that for gasoline operated vehicles (two wheeler, LMV, car).

Total BC emissions for year 2011 have been estimated to be 892 ± 152 Gg (Table 3) of which 48% (425 Gg) originated
from Domestic Fuel consumption, 22% (198 Gg) from Industry, 17% (154 Gg) from the Transport sector and 12% (103 Gg) from Open burning. Diesel use in mobile towers and irrigation pumps contributed 1% (11 Gg) to total BC emissions (Figure 7). Firewood with emissions of 177 Gg is the single most emitting source. It emits more than transportation (154 Gg) and open burning (103 Gg) categories. As shown in Figure 6, 76.3% of the 140 million rural households MoSPI (2014) use firewood as the primary source of energy for cooking.

- The spatial distribution of national emissions is presented in Figure 8. From the map it can be easily concluded the Indo Gangetic Plain (IGP) IGP is the main contributor to national BC emissions. This can be attributed to the very high population density and presence of major BC emitting industries like sugar and brick production in this region. With annual emissions of 140 Gg, the state of Uttar Pradesh emits the most in IGP followed by West Bengal (57.67 Gg), Bihar (47.8 Gg), Punjab (34.01 Gg), Haryana (26.82 Gg), and the National Capital Territory (NCT) of Delhi (6.74 Gg). High emissions from IGP and
- 20 its vicinity to the Himalayas potentially pose a serious threat to water security in the region resulting from impacts to the cryosphere from BC deposition and atmospheric heating. The state of Uttar Pradesh is also the highest emitting state in India and contributes 15.7% to the national BC emissions.

As discussed in Section 2, best fit probabilistic distributions were obtained for EF and activity data (for each source) using the KS statistic. A sample of 1000 numbers were generated from each of the two distributions (EF and activity data), the

- 25 product of which provides over one million emission points. Mean and standard deviation was determined for each source using the obtained emission points. The emission points were added up for all the sources to get overall national thousand emission points and subsequently the national mean emission and standard deviation. A best fit probabilistic distribution curve was obtained for the national emission points on the basis of KS statistic. The probabilistic distribution for overall national emissions was found to be General extreme value distribution with KS statistics of 0.01 (Figure 9). Figure 10 presents the
- 30 sector-wise optimally fit distributions for the BC emissions.

#### 4.1 Open Burning

The national level emissions from this sector contribute 12% (103 Gg) to the total emissions. Burning of crop residue has been the major contributor (62%) followed by forest fires (36%). MSW burning contributed only 2% to the open burning emissions.

The source-wise emission contribution and spatially distributed open burning emissions are presented in Figures 7 & 8. The emissions from open burning are highest from the north-west states of Punjab, Haryana (crop residue burning) and north eastern states of Nagaland, Manipur, Mizoram and Tripura (forest fires). Punjab and Haryana are the main food producing states of India, in the months of April, May, October and November, the crop residue is burned for clearing the land for next crop. In

5 the north-east, open burning emissions arise primarily from forest fires, however, some tribal communities also practice slash and burn agriculture in this region as well.

#### 4.2 Industry

National level industry sector emissions account for 22% (198 Gg) of the total emissions. In this sector, brick and sugar production contribute the maximum emissions (37% each), followed by steel production (11%), cement (8%) and power plants

- 10 (7%) (Figure 7). Spatially distributed emissions from the Industry sector are presented in Figure 8. The hotspots of industrial emissions are the states in IGP as most of the brick and sugar industries lie in this area. It is also evident and expected from Figure 8 that metropolitan cities contribute significantly to the sector as they have major industrial belts at the periphery. High emissions from the brick and sugar industry result from the use of low-grade fuels and from dated and inefficient systems and processes. While power plants accounts for 75% of the coal consumption, their BC emissions are just 7% of the total industrial
- 15 emissions, due to the higher efficiency of combustion in these systems. An acknowledged source of uncertainty in our approach is the lack of specific geolocated coordinate data for the two largest emission sources, brick and sugar.

#### 4.3 Transport

Transportation sector emissions account for 17% (154 Gg) of the national BC emissions in 2011. In the transport sector trucks have been found to be most emitting (24%) followed by tractor & trailers (15%). Emissions from bus, car, LCV, LMV and
two wheelers contributed 12%, 10%, 13%, 11% and 13% to national transport sector BC emissions respectively. Railways contributed 0.2% to BC emissions in 2011, shipping and aviation combined emitted less than 0.05% (Figure 7). The spatial distribution of Transportation emissions are presented in Figure 8. The main contributors are the metropolitan cities, NCT of Delhi, Mumbai and Bangalore. The results also indicated the majority of the emissions from the transport sector originate from diesel road vehicles (truck, tractors & trailers, bus, LCV and LMV).

#### 25 4.4 Domestic Fuel

Domestic fuels account for almost half of the national BC emissions (48%, 425 Gg). Within the sector, firewood contributes most significantly, (42%), followed by kerosene lamps (26%). Agricultural residue, dung cake, and coal used for cooking contributed 17%, 13% and 2% respectively (Figure 7). Figure 8 shows the spatially distributed emissions of Domestic Fuel usage. Here also, the majority of emissions arise from the IGP due to the high population density in this area. Also the poverty

30 levels are high in this region, so a larger proportion of the population tends to use, cheaper, biofuels for cooking. The biofuel used in hand made stoves has low combustion temperatures leading to inefficient combustion process and consequently the

domestic fuel sector has higher BC emissions. Also these are uncontrolled emission sources. Kerosene lamps (109 Gg) are the second highest emitting source as a result of the very high EF of kerosene lamps. While the emissions from kerosene lamps is more than the entire open burning sector combined, studies must be conducted to evaluate the potential impact and transport of this source of BC. It likely has extremely significant health impacts due to the emissions being contained within homes, but the climate impact is likely as large as for Open Burning.

#### 4.5 Other

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Emissions from this category account for slightly more than 1% (11 Gg) of the national BC emissions. Within this category emissions from use of diesel in irrigation pumps contribute 7 Gg and its use in generating electricity for mobile towers contribute 4 Gg.

#### 10 4.6 Uncertainty Analysis

Figure 11 shows the mean and standard deviations based on best fit probabilistic distributions of emissions from the major sectors. Based on the Monte-Carlo simulations using the multiple emissions estimates and available information on uncertainty, the Probability Distribution Functions (PDF) for each of the sectors is calculated as shown in Figure 10. The best fit distribution for the Domestic Fuels sector was found to be Burr distribution with KS statistic of 0.01, for Industrial emissions, Gamma

15 distribution with KS statistics of 0.02, for open burning emissions, Johnson SU distribution with KS statistic of 0.02 and Log-Logistic (3P) for transport sector with KS statistic of 0.03. The uncertainty is highest for emissions from domestic fuels sector. The EFs and activity data for the sources in domestic fuel sector show a large variation leading to high uncertainty in the BC emissions as there is no accurate database of the population using cook stoves, quantity of fuel consumed, and their efficiency.

#### 4.7 Comparison with prior estimates

- 20 Emissions in this study have been determined using Monte-Carlo simulation of multiple activity data and emission factors. As previous studies have used point estimates for these highly uncertain quantities, the results are bound to differ. Figure 12 presents the comparison of the results of this study (Table 3) with emission inventories developed in the past. For the base year 2011 the estimate is about 80% than that reported in SAFAR emission inventory (1119 Gg/yr). For inventories with base year 2010, total national emissions estimated in this study are a factor of 1.3 higher than RETRO (697 Gg/yr), factor of 0.8 from
- than estimated in Klimont et al. (2009) (1104 Gg/yr), factor of 0.9 from that estimated in Lu et al. (2011) (1015 Gg/yr) and were in agreement with emissions determined by Ohara et al. (2007) (862 Gg/yr). All prior national emission estimates lie within two standard deviations of our mean estimate.

Emissions estimates from the Domestic Fuels sector (425±112 Gg/yr) is lower by a factor of 0.7-0.9 with Pandey et al. (2014) (488 Gg/yr), Klimont et al. (2009) (628 Gg/yr) and Lu et al. (2011) (579 Gg/yr). For the Transport sector our emission

30 estimate  $(154 \pm 56$ Gg/yr) is almost identical to that presented in Sadavarte and Venkataraman (2014) (144 Gg/yr) and a factor of 1.1-1.3 higher than the emissions determined by Lu et al. (2011) (111 Gg/yr), Baidya and Borken-Kleefeld (2009)(123 Gg/yr) and Klimont et al. (2009)(136 Gg/yr). In the Industry sector our emissions (198± 83Gg) are 10-20% lower in view of the inclusion of only higher emitting industries in this study. The combined industrial emission estimate of Sadavarte and Venkataraman (2014) (formal industry) and Pandey et al. (2014) (informal industry) (212 Gg/yr) is in good agreement with our emission estimate (factor 0.94). Estimated industrial emissions are a factor of 0.8-0.9 lower than Lu et al. (2011) (227 Gg/yr)

5 and Klimont et al. (2009) (261 Gg/yr). Emissions from open crop residue burning (64 ± 17Gg/yr) are in close agreement (factor 0.8-1) with Jain (2014)(68 Gg/yr), Lu et al. (2011) (74 Gg/yr) and Pandey et al. (2014) (80 Gg/yr). Forest fire emissions (37 ± 13Gg) are almost identical to those determined in Reddy and Venkataraman (2002a) (39 Gg/yr). As with the national emission estimate, for all sectors prior emission estimates are within one or two standard deviations from our mean emission estimate.

#### 4.8 Seasonality of Emissions

- 10 There is a strong seasonality associated with BC emissions in India. Crop residue burning, forest fires, brick and sugar industry have a seasonal dependence in emissions. Forest fires are predominant from February to July, monthly BC emissions from forest fires were estimated using MODIS burnt area data. Brick industry becomes active after the monsoon season from months of October to June (Maithel et al., 2012), the sugar industry operates from November to June (Tyagi, 1995) and the emissions are equally distributed among the months of operation. Burning of crop residues generally occurrs in the harvesting months
- 15 which are October-November for Kharif crop and April-May for Rabi Crop. Emissions of agricultural open burning are equally distributed among the months of April, May, October and November. For all the other sources, emission rates are assumed to be uniform throughout the year. Using this data monthly variation of BC emissions has been estimated and is shown in Figure 13.

The emissions in April are highest due to the burning of crop residues. Despite the absence of crop residue burning, emissions in March are also high because of the emissions from forest fires. As we have shown a considerably amount of the emissions to come from the IGP in close proximity to the Himalaya, this causes further concern to the potential cryospheric impact of these aerosols as they are strongest during the period when the seasonal snow melt period is beginning and could be incorporated into the snow pack.

#### 5 Conclusions

- A spatially resolved Black Carbon (BC) emission inventory for 2011 has been developed considering major sectors with careful consideration for sub-sector sources. The sources were classified into five major sectors: i) Open Burning, including: forest fire emissions, open solid waste burning, and agriculture residue burning; ii) Industry, including: brick industry, cement, steel plants, sugar mills and power plants; iii) Transport, including: two wheelers, cars, light motor vehicles passenger, light commercial vehicles, taxies, trucks, buses, tractors & trailers, railways, shipping and airways; iv) Domestic Fuel, including:
- 30 firewood burning, agricultural residue, coal, liquid petroleum gas, kerosene (cooking and lighting) and dung cake; and v) Other, including: use of diesel in power generation for mobile towers and irrigation pumps.

This is a first-of-kind comprehensive study which included sources such as kerosene lamps and forest fires that were not part of earlier emission inventories. Furthermore, for each sector, source uncertainties in emissions have been estimated based on variability in available activity data and emission factors. Lastly, and significantly, we provide our estimate of emissions at monthly temporal resolution on a spatially distributed 40 x 40 km. grid.

- The national BC emissions for India in 2011 are estimated to be  $892 \pm 151$  Gg/yr with domestic fuels contributing maximum (48%) followed by industries (22%), transport (17%), open burning (12%) and others (1%). Large emission in domestic fuels sector stems from the extensive use of biomass for cooking in India. Firewood is the single largest emitter with 177 Gg (19.9%) of BC emissions in 2011. The emissions from firewood are more than the entire transportation sector combined. Kerosene lamps surprisingly contribute 12% to the national BC emissions. The emissions have been found to be have a significant seasonality,
- 10 varying from 54 Gg in July to 89 Gg in April 2011.

The results of the study could be used to assess the contribution of different sources to national and regional emissions. The spatial resolution of the inventory should be useful for modelling the Black Carbon processes in atmosphere through air quality models. Monthly gridded emission datasets can also be prepared for finer temporal resolution input. To improve the future BC emission estimates, local emission factors and activity data should be improved, especially for domestic fuels and

15 brick industry. Emission inventory can be improved nationally, regionally and temporally by comparing the modeled emission estimates (providing the inventory as input to air quality models) with the observed data.

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Figure 1. Methodology for National Emissions.



Figure 2. Methodology for preparing gridded emissions.



Figure 3. Flow chart for the calculation of Forest Fire emissions based on MODIS products.



Figure 4. Land Cover and Burnt Area for March 2011.



Figure 5. Flow chart for the calculation of agricultural waste burning.



Figure 6. Energy sources used for cooking in Rural India 2009-2010.



Figure 7. (a-d) Proportion of sub-categories to the major sector emissions and e) contribution of major sector emissions to the national emissions total.



Figure 8. (a-d) Maps of major sector emissions and e) spatial variability of national emissions total for BC.



Figure 9. Gen. Extreme Value Distribution fit for the National BC Emissions.



Figure 10. Sectorial emission histograms and associated best-fit PDFs.



Figure 11. Mean and Standard Deviation for each of the major sectors of emissions for India, 2011.



Figure 12. Comparison of current BC emissions estimate with prior published results for India. A: Streets et. al. 2003 (Base year 2000); B: Reddy and Venkataraman 2002a & 2002b (Base year 1997); C: Sahu, et. al. 2008 (Base year2011); D: Schultz et. al. 2008 (Base year 2010); E: Lu et. al.(2011) (Base year 2010); F: Klimont et. al.(2009) (Base year 2010); G: Ohara et. al.(2007) (Base year 2010); H:This study (Base year 2011))



**Figure 13.** Timeseries of monthly emissions for India, 2011. Note the strong seaonality of the Open burning and Industry sector. In the latter case, the seasonality results predominately from the sugar cane production.

Table 1. Mean activity data, standard deviation, and best fit probabilistic distribution.

Subsector	Activity Level	Mean ± SD	Distribution
Open $Burning(MT/year)$			
Crop Residue Burning	99.93 <sup>1,2</sup> , 89.79 <sup>1,3</sup> , 90.94 <sup>1,4</sup>	93.56±4.96	Uniform
Forest Fire*	47.83 <sup>5</sup>	47.83±9.56	Normal
Garbage Burning	3.90 <sup>2,6</sup> , 2.51 <sup>2,6,7,8,9</sup>	3.2±0.76	Uniform
Industry ( $MT/yr$ )			
Brick*	474 <sup>10</sup>	474±237	Normal
Steel*	40.05 <sup>11</sup>	40.05±8.01	Normal
Sugar*	77.1 <sup>12,13</sup>	77.1±15.42	Normal
Cement*	28.06 <sup>14</sup>	28.06±5.61	Normal
Power Coal*	380.91 <sup>15</sup>	380.91±76.18	Normal
Power Diesel*	0.7115	0.71±0.01	Normal
<b>Transport</b> (BillionKm/yr)			
Bus	38.77 <sup>16,17</sup> , 39.3916 <sup>16,18</sup> , 32.8316 <sup>16,19</sup>	35.46±3.94	Uniform
Car	$128^{16,17}, 196.27^{16,18}, 130.85^{16,19}, 167.87^{16,21}$	155.06±29.76	Uniform
LMV	$104.22^{16,17}, 131.51^{16,18}, 65.75^{16,19}$	105.32±22.74	Uniform
LCV	$78.55^{16,17}, 99.12^{16,18}, 74.34^{16,19}$	111.37±38.70	Uniform
Truck	$122.99^{16,17}, 87.30^{16,20}, 125.50^{16,21}$	109.60±16.72	Uniform
Taxi	$38.25^{16,17}, 40.26^{16,18}, 48.32^{16,19}, 16.91^{16,21}, 60.44^{16,22}, 51.01^{16,23}$	42.52±14.86	Gumbel
Two Wheeler	$450.8^{16,17}, 764.07^{16,18}, 481.36^{16,21}, 2062.89^{16,22}, 313.26^{16,23}$	814.50±716.18	Uniform
Tractor & Trailer	11.08 <sup>16,17</sup> , 26.38 <sup>16,18</sup>	18.73±8.38	Uniform
Railway Coal (KT/yr)*	1 <sup>24</sup>	1±0.02	Normal
Railway Diesel (KT/yr)*	21.06 <sup>24</sup>	21.06±42.10	Normal
Shipping HSDO (KT/yr)*	0.11 <sup>25,26</sup>	0.11±0.02	Normal
Shipping Fuel Oil (KT/yr)*	80 <sup>25,26</sup>	80±16	Normal
Shipping LDO (KT/yr)*	$0.36^{25,26}$	$0.36 \pm 0.07$	Normal
Aviation LTO (KT/yr)*	514.16 <sup>2,25,27,28</sup>	514.16±102.83	Normal
Aviation Cruise (KT/yr)*	1505.83 <sup>2,25,27,28</sup>	1505.83±301.16	Normal
<b>Domestic Fuel</b> $(MT/yr)$			
Dung Cake	144.84 <sup>29</sup> , 75.62 <sup>30</sup>	$110.23 \pm 37.91$	Uniform
Agriculture Residue	125.34 <sup>29</sup> , 81.25 <sup>30</sup>	103.30±24.14	Uniform
Firewood	$209.99^{31}, 281.99^{29}, 193.87^{30}$	228.62±41.96	Uniform
Coal*	4.77 <sup>31</sup>	4.77±0.95	Normal
Kerosene Cooking*	4.57 <sup>31,32</sup>	4.57±0.91	Normal
LPG*	12.37 <sup>31</sup>	12.37±2.47	Normal
Kerosene Lamps	$1.68^{32}, 1.21^{31,32}$	1.45±0.25	Uniform

Table 2. Mean EFs, standard deviation and best fit probabilistic distribution.

Subsector	EFs Used	Mean EF ± SD	Best Fit Distribution
<b>Open Burning</b> (g/kg)			
Crop Residue Burning	$0.69^1, 0.78^2, 0.73^3, 0.47^4, 0.75^2$	0.69±0.19	Dagum
Forest Fire	$0.56^1, 0.98^4, 0.99^5, 0.56^6$	0.76±0.21	Error
Garbage Burning	$0.65^7, 0.37^8$	0.51±0.15	Uniform
<b>Industry</b> (g/kg)			
Brick	0.11 <sup>9</sup> , 0.27 <sup>9</sup> , 0.09 <sup>9</sup>	0.16±0.09	Uniform
Steel	$0.32^3, 1.1-1.58^{10}, 0.224^{11}, 0.23-0.13^{12}, 0.06^5, 0.0095^{13}$	0.45±0.51	Log Pearson 3
Sugar	$1.2^{14}, 0.7^{15}$	0.95±0.27	Uniform
Cement	$0.32^3$ , $1.1$ - $1.58^{10}$ , $0.224^{11}$ , $0.23$ - $0.13^{12}$ , $0.06^5$ , $0.0095^{13}$	0.45±0.51	Log Pearson 3
Power Coal	$0.003 - 0.032^{16}, 0.077^{13}, 0.0029^{11}, 0.002^5, 0.06^5$	0.03±0.03	Gamma (3P)
Power Diesel	$0.25^{11}, 0.15^8, 0.06^{13}$	0.15±0.08	Uniform
<b>Transport</b> (g/km)			
Bus	$0.35^{17,18}, 0.8^{18,19}, 0.225^{18,20}, 0.61^{18,21}$	$0.49 \pm 0.24$	Uniform
Car	$0.16^{22}, 0.17^{17,18}, 0.05^{18,19}, 0.07^{18,20}, 0.16^{18,21}$	0.09±0.06	Uniform
LMV	$0.16^{22}, 0.138^{17,18}, 0.17^{18,21}$	0.15±0.01	Uniform
LCV	$0.27^{17,18}, 0.13^{18,19}, 0.16^{18,21}$	$0.19 \pm 0.07$	Uniform
Truck	$0.61^{17,18}, 0.26^{18,19}, 0.19^{18,20}, 0.31^{18,21}$	0.34±0.17	Uniform
Taxi	$0.01^{22}, 0.06^{17,18}, 0.076^{18,20}, 0.057^{18,21}$	0.05±0.03	Uniform
Two Wheeler	$0.013^{23}, 0.012^{17,18}, 0.038^{18,19}, 0.023^{18,20}$	0.02±0.01	Uniform
Tractor & Trailer*	$1.24^{23}$	$1.24 \pm 0.25$	Normal
Railway Coal (g/Kg)	1.83 <sup>13</sup> , 3 <sup>8</sup>	2.415±0.33	Uniform
Railway Diesel (g/Kg)	$1.53^{24}, 0.51^8, 0.29^{13}$	0.78±0.59	Uniform
Shipping HSDO (g/Kg)	$0.85^{25}, 1.19^8, 1.32^{26}, 0.36^{25}$	0.78±0.49	Gen. Extreme Value
Shipping Fuel Oil (g/Kg)	$0.38^{25}, 0.36^{25}, 0.97^{25}, 0.85^{25}, 1.19^8, 1.32^{26}$	0.72±0.40	Wakeby
Shipping LDO (g/Kg)	$0.85^{25}, 1.19^8, 1.32^{26}$	0.89±0.46	Uniform
Aviation LTO (g/Kg)	$0.08 - 0.1^{27}$	0.09±0.01	Uniform
Aviation Cruise (g/Kg)	$0.02 - 0.1^{27}$	$0.06 \pm 0.02$	Uniform
<b>Domestic Fuel</b> $(g/kg)$			
Dung Cake	$\overline{0.53^8, 1^{14}, 0.8^{28}, 0.25^4, 0.49^{29}, 0.18^{30}, 0.12^{31}, 0.4^{15}}$	$0.47 \pm 0.31$	Gen. Extreme Value
Agriculture Residue	$0.43^{32}, 0.66^{11}, 0.75^2, 0.47^4, 0.37^{29}, 1^8, 1.3^{33}, 0.24^{30}, 1.38^{34}, 0.6^{31}, 0.9^{31}$	$0.74 \pm 0.37$	Gen. Extreme Value
Firewood	$1^{3}$ , $0.59^{1}$ , $0.41^{4}$ , $0.7^{32}$ , $1.2^{14}$ , $1^{28}$ , $0.85^{8}$ , $0.6^{31}$ , $0.35^{29}$ , $1.1^{33}$ , $0.25^{30}$ , $0.83^{35}$ , $1.33^{6}$ , $0.7^{36}$	$0.78 \pm 0.32$	Gen. Extreme Value
Coal	1.91 <sup>3</sup> , 2.84 <sup>10</sup> , 1.83 <sup>4</sup> , 5 <sup>8</sup> , 0.28 <sup>37</sup> , 2.295 <sup>24</sup> , 0.8 <sup>11</sup> , 0.3 <sup>11</sup> , 0.69 <sup>11</sup> , 0.79 <sup>11</sup> , 0.32 <sup>11</sup> , 0.497 <sup>11</sup> , 0.07 <sup>36</sup> , 5.4 <sup>15</sup> <b>32</b>	$1.64 \pm 1.73$	Pearson 6 (4P)
Kerosene Cooking	$0.16^4, 0.02^{15}$	$0.18 \pm 0.02$	Uniform
LPG	$0.067^{11}, 0.01^{15}$	$0.04 \pm 0.03$	Uniform

Sector/Subsector	Emissions (Gg/yr)	
Open Burning	102.84 ± 27.56	
Crop Residue Burning	64.31 ± 17.19	
Forest Fire	36.90±12.85	
Garbage Burning	$1.63 \pm 0.62$	
Industry	198.5 ± 83.391	
Brick	74.11 ± 61.38	
Steel	$21.09 \pm 32.18$	
Sugar	$72.76 \pm 25.05$	
Cement	$15.45 \pm 22.26$	
Power	15.09±23.88	
Transport	154.34 ± 56.14	
Bus	$17.64 \pm 8.72$	
Car	$14.69 \pm 10.54$	
LMV	$17.01 \pm 25.03$	
LCV	$20.62 \pm 10.51$	
Truck	$37.46 \pm 20.49$	
Taxi	$2.13 \pm 1.44$	
Two Wheeler	$20.11 \pm 39.50$	
Tractor & Trailer	$22.79 \pm 11.41$	
Railway	$1.60 \pm 1.32$	
Shipping	$0.15 \pm 0.07$	
Aviation	$0.14 \pm 0.04$	
Domestic Fuel	425.36 ± 111.97	
Dung Cake	54.79 ± 48.15	
Agriculture Residue	$74.38 \pm 44.17$	
Firewood	$177.34 \pm 83.88$	
Coal	$9.02 \pm 14.622$	
Kerosene Cooking	$0.83 \pm 0.19352$	
LPG	$0.47 \pm 0.39$	
Kerosene Lamps	$108.53 \pm 27.10$	
Others	11.08 ± 1.94	
Mobile Towers	$4.00 \pm 0.93$	
Irrigation Pumps 33	$7.08 \pm 1.70$	
Total	892.12±151.53	