Uncertainties in emissions estimates of greenhouse gases and air pollutants in India and their impacts on regional air quality

Eri Saikawa\textsuperscript{1}, Marcus Trail\textsuperscript{1,2}, Min Zhong\textsuperscript{1}, Qianru Wu\textsuperscript{1}, Cindy L Young\textsuperscript{1,3}, Greet Janssens-Maenhout\textsuperscript{4,5}, Zbigniew Klimont\textsuperscript{6}, Fabian Wagner\textsuperscript{6,7,8}, Jun-ichi Kurokawa\textsuperscript{9}, Ajay Singh Nagpure\textsuperscript{10} and Bhola Ram Gurjar\textsuperscript{11}

\textsuperscript{1} Department of Environmental Sciences, Emory University, 400 Dowman Drive, Atlanta, GA, 30319, United States of America
\textsuperscript{2} Georgia Environmental Protection Division, 2 Martin Luther King Jr. Drive, Atlanta, GA, 30334, United States of America
\textsuperscript{3} School of Earth and Atmospheric Sciences, Georgia Institute of Technology, 311 Ferst Drive, Atlanta, GA, 30332, United States of America
\textsuperscript{4} European Commission, Joint Research Centre, Institute for Environment and Sustainability, Via Fermi, 2749, 21027 Ispra VA, Italy
\textsuperscript{5} Ghent University, Campus Ardyna, Ghent-Zwijnaarde, Belgium
\textsuperscript{6} International Institute for Applied Systems Analysis, Schlossplatz 1, 2361 Laxenburg, Austria
\textsuperscript{7} Woodrow Wilson School of Public and International Affairs, Princeton University, Princeton, NJ, 08544, United States of America
\textsuperscript{8} Andlinger Center for Energy and the Environment, Princeton University, Princeton, NJ, 08542, United States of America
\textsuperscript{9} Asia Center for Air Pollution Research, 1182 Sowa, Nishi-ku, Niigata, Niigata, 950–2144, Japan
\textsuperscript{10} Humphrey School of Public Affairs, University of Minnesota, 301 19th Avenue S, Minneapolis, MN, 55455, United States of America
\textsuperscript{11} Department of Civil Engineering, Indian Institute of Technology Roorkee, Roorkee 247667, Uttarakhand, India

E-mail: eri.saikawa@emory.edu

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Abstract

Greenhouse gas and air pollutant precursor emissions have been increasing rapidly in India. Large uncertainties exist in emissions inventories and quantification of their uncertainties is essential for better understanding of the linkages among emissions and air quality, climate, and health. We use Monte Carlo methods to assess the uncertainties of the existing carbon dioxide (CO\textsubscript{2}), carbon monoxide (CO), sulfur dioxide (SO\textsubscript{2}), nitrogen oxides (NO\textsubscript{x}), and particulate matter (PM) emission estimates from four source sectors for India. We also assess differences in the existing emissions estimates within the nine subnational regions. We find large uncertainties, higher than the current estimates for all species other than CO, when all the existing emissions estimates are combined. We further assess the impact of these differences in emissions on air quality using a chemical transport model. More efforts are needed to constrain emissions, especially in the Indo-Gangetic Plain, where not only the emissions differences are high but also the simulated concentrations using different inventories. Our study highlights the importance of constraining SO\textsubscript{2}, NO\textsubscript{x}, and NH\textsubscript{3} emissions for secondary PM concentrations.

1. Introduction

Understanding the spatial and temporal distribution of greenhouse gases (GHGs) and air pollutant precursor emissions is vital to the implementation of appropriate climate and air quality mitigation measures. Obtaining accurate anthropogenic emissions estimates is especially critical in India, which is currently the world’s third largest emitter of carbon dioxide (CO\textsubscript{2}) from fossil fuel combustion and from industrial processes (production of cement, metals and chemicals) behind China and USA (Olivier \textit{et al 2015}), after India surpassed Russia in 2010. Half of the top twenty most polluted cities in the world are in India (WHO \textit{2016}) and India is ranked the third worst of 180 countries for PM\textsubscript{2.5} (particulate matter with an aerodynamic diameter less than 2.5 \textmu m) exposure (Hsu \textit{et al 2016}). Several emissions inventories have been developed recently either specifically for India or for larger regions that include India (Streets \textit{et al 2003}, Garg \textit{et al 2006}, Ohara \textit{et al 2007}, Klimont \textit{et al 2009, 2013, 2016}, Zhang \textit{et al 2009}, EC-JRC/PBL 2011,
Lu et al (2011), Smith et al (2011), Sahu et al (2012), Kurokawa et al (2013), Pandey et al (2014), Sadavarte and Venkataraman (2014), IEA (2014), Janssens-Maenhout et al (2015). These inventories are being used in the air quality and climate model simulations to better understand air pollution and climate change in India, in Asia, and globally.

Several studies have compared emissions inventories in India and other Asian countries (Garg et al. 2006, Granier et al. 2011, Kurokawa et al. 2013, Klimont et al. 2016). However, most studies focus their analysis on similarities and differences in national total emissions and do not analyze regional scale or source sector level emissions. These studies also do not include the recent national emissions inventories. Pandey et al. (2014) and Sadavarte and Venkataraman (2014) evaluated their Indian emissions inventory against several but not against many others, including the Regional Emissions inventory in Asia version 2.1 (REAS) and Emissions Database for Global Atmospheric Research version 4.2 (EDGAR), the two most commonly used datasets for Asian and global emissions, respectively.

In this study, we first compare emission inventories of anthropogenic, combustion-related surface emissions of CO, and air pollutant precursors (carbon monoxide CO, sulfur dioxide SO2, nitrogen oxides (NOx = NO + NO2), and particulate matter with an aerodynamic diameter less than 10 μm, PM10) for each of the four source sectors and compare existing estimates within subnational regions in India. We then use Monte Carlo sampling to assess the uncertainties of the existing emissions estimates per sector and per species. We further conduct a chemical transport model simulation with two different gridded emissions to validate them and assess their impacts on air quality.

2. Methodology

2.1. Datasets

This work relies on existing emissions inventories that provide estimates at provincial and/or national levels in India. We use the following eight inventories to compare and to assess uncertainties at the source sector level, using Monte Carlo sampling methods: 1) EDGAR v4.2 (EDGAR) (EC-JRC/PBL 2011); 2) REAS v2.1 (REAS) (Kurokawa et al 2013); 3) INTEX-B (Zhang et al 2009); 4) National emissions inventory for residential and road transportation emissions (Nagpure–Gurjar) (Nagpure and Gurjar 2012); 5) National emissions inventory for all sectors (Sadavarte–Venkataraman) (Pandey et al 2014, Sadavarte and Venkataraman 2014); 6) IEA (2014), 7) GAINS (Amann et al 2011, Klimont et al 2016); and 8) Lu et al (2011). The emissions inventories analyzed in this paper were developed using a similar bottom-up methodology, where emissions were calculated as the product of activity data, such as fuel consumption, and fuel- and technology-dependent emission factors. Table 1 describes the details of each inventory including years, source sectors, and species covered, its horizontal resolution, proxies used, as well as its coverage. In addition to these eight that provide sector-level emissions, we also use total CO2, CO, SO2 and NOx emissions estimates by Garg et al (2006), total SO2 emissions estimates by Smith et al (2011) and Klimont et al (2013), as well as the total NOx emissions estimates by Ghude et al (2012).

2.2. Emissions comparison

We compared emissions of CO2, CO, SO2, NOx, and PM10 from four source sectors, including power plants (power), industrial combustion and processes (industry), domestic combustion (domestic), and road and non-road transportation (transport) for the years between 2000 and 2010 at national and nine subnational regions. Table S1 available at stacks.iop.org/ERL/12/065002/mmedia describes how we categorize sources for each inventory into the four described above. For the national level analysis, we used seven inventories (EDGAR, REAS, Nagpure–Gurjar, Sadavarte–Venkataraman, IEA, GAINS, and Garg) for CO2, seven inventories (EDGAR, REAS, INTEX-B, Nagpure–Gurjar, Sadavarte–Venkataraman, GAINS, and Garg) for CO and NOx, seven inventories (EDGAR, REAS, INTEX-B, Nagpure–Gurjar, Sadavarte–Venkataraman, GAINS, and Lu) for SO2, and four inventories (EDGAR, REAS, INTEX-B, and GAINS) for PM10. For the subnational level analysis, we used five inventories (EDGAR, REAS, INTEX-B, Nagpure–Gurjar, and GAINS).

We also conducted one million Monte Carlo samplings, choosing an emissions inventory per sector per year randomly. We sampled a normal distribution for most but a few emission estimates in Sadavarte–Venkataraman inventory (CO all sectors; SO2 transport and domestic; and NOx all sectors), which assumed a log-normal distribution. We used the standard deviation (SD) values reported by each inventory if such information was available. For the inventories where this was not provided, the relative uncertainty estimates provided by REAS for each sector were used. The mean and the SD of the newly-composed emissions estimates were then calculated from one million samples. The uncertainty is reported as the 95% confidence interval, following Kurokawa et al (2013).

2.3. Air quality simulation

In order to assess how differences in emissions inputs affect air quality simulations, we conducted two simulations using the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) version 3.5 (Grell et al 2005), which has been validated previously in Asia (Zhong et al 2016). We chose two gridded emissions inventories, EDGAR
and REAS, and performed simulations in January and July of 2008. These two months were chosen to simulate the seasonality in air pollutant concentrations. January is the dry winter month with mostly high air pollutant concentrations, whereas July is in the middle of the monsoon season with relatively low concentrations (Gaur et al 2014). For O₃, it is similar and July tends to have higher mixing ratio than in January. The model covered the entire India, with a horizontal resolution of 20 × 20 km and 31 vertical levels. The initial and lateral chemical boundary conditions were taken from a present-day simulation of the NOAA Geophysical Fluid Dynamics Laboratory (GFDL) global chemistry-climate model AM3 (Naik et al 2013). The meteorological data were obtained from the National Center for Environmental Prediction (NCEP) Global Forecast System final gridded analysis datasets. We used Carbon-Bond Mechanism version Z (Zaveri and Peters 1999) for gas-phase chemistry and the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al 2008) for aerosol chemistry. The model simulation was spun-up for ten days before the beginning of each monthly simulation.

2.4. Surface observations
We compared our air quality simulations with existing instantaneous surface observations of SO₂ and NOₓ.

### Table 1. Description of emissions inventories used for this study.

| Years               | Source Sectors                                                                 | Species                  | Horizontal Resolution | Proxies for allocating emissions                                      | Coverage          | Reference                           |
|---------------------|--------------------------------------------------------------------------------|--------------------------|-----------------------|-------------------------------------------------------------------------|-------------------|-------------------------------------|
| EDGAR 1970–2008     | energy, industrial processes, product use, agriculture, large scale biomass burning, and other anthropogenic sources | CO₂, NOₓ, SO₂, CO, PM₁₀, NOₓ | 0.1° × 0.1°           | Rural, urban, and total populations, roads, railways, power plant maps, and selected industrial production | Global             | EC-JRC/PBL 2011                     |
| REAS v2 2000–2008   | power plants, combustible and non-combustible sources in industry, on-road and off-road sources in transportation, residential and agricultural sources | CO₂, NOₓ, SO₂, CO, PM₁₀, PM₂.₅, NOₓ, BC, OC, NH₃, NMVOC | 0.25° × 0.25°         | Rural, urban, and total populations, as well as road network             | 33 Asian countries | Kurokawa et al 2013                 |
| INTEX-B 2006        | power plants, industry, residential, and transportation                         | SO₂, CO, PM₁₀, PM₂.₅, NOₓ, BC, OC, NMVOC | 0.5° by 0.5°          | spatial proxies at 1 km × 1 km resolution                               | 22 Asian countries | Zhang et al 2009                    |
| Nagpure–Gurjar 2001–2011 | residential and road-transport                                                  | CO₂, NOₓ, SO₂, CO, PM₁₀, NOₓ | provincial level      | NA                                                                       | India             |                                    |
| Sadavarte–Venkataraman 1996–2015 (projected using 2010 data) | industry, transportation, residential, and ‘informal industries’ including brick production and processing operations for food and agricultural products | CO₂, NOₓ, SO₂, CO, PM₁₀, PM₂.₅, NOₓ | 25 × 25 km (0.25° × 0.25°) | Census data, urban population, road network, and district-level production data | India             | Pandey et al 2014, Sadavarte and Venkataraman 2014 |
| IEA 1960–2012       | fossil fuel combustion                                                         | CO₂                      | national level        | NA                                                                       | Global             | IEA, 2014                           |
| GAINS 1990–2030     | energy, domestic, industrial combustion and processes, road and non-road transportation and agriculture | CO₂, NOₓ, PM₁₀, PM₂.₅, CO₂, SO₂ | 0.5° × 0.5°           | RCP and GEA (Global Energy Assessment) sectoral proxies, population, and selected industrial plants location (e.g. smelters) | Global             | Amann et al 2011, Klimont et al 2016 | www.iiasa.ac.at/web/home/research/researchPrograms/air/Global_emissions.html |
concentrations (Maharashtra Pollution Control Board 2016), and monthly average CO and O3 mixing ratio (Kumar et al 2012) for January and July 2008. In addition, we used Aerosol Optical Depth (AOD) retrieved from the MODerate Resolution Imaging Spectrometer (MODIS) instrument aboard the Terra satellite and compared it with the simulated AOD from WRF-Chem. MODIS provides AOD retrievals at a resolution of 10/10 km. In this study, we use Level 2 and Collection 6 aerosol optical thickness at 550 nm. We calculated the monthly mean AOD values from daily observations of AOD over land and ocean.

3. Results

We present the mean and the SD of our Monte Carlo samplings for the national total emissions estimates for the five species in figure 1, as well as the estimates and SD provided by existing emissions inventories. The detailed means and SD of each sector per species from the Monte Carlo simulations are listed in table S2.

3.1. CO2

Our Monte Carlo analysis illustrates that there is up to ±76% uncertainty in India’s national total CO2 emissions estimates when we combine all the existing estimates equally. This is larger than the REAS’ existing uncertainty of ±49% (figure 1). The largest uncertainty comes from the domestic sector, with the maximum being ±149%. The IEA domestic CO2 emissions are the lowest of all the inventories at 63 Tg CO2 yr\(^{-1}\) in 2000, contributing to only 13% of REAS domestic CO2 emissions. This large difference can be explained in part because IEA does not include biofuel emissions in their estimates, similar to GAINS and Garg.

The smallest uncertainty stems from the power sector, which is the primary source of CO2 emissions in India. EDGAR, REAS, GAINS, Sadavarte–Venkataraman, and IEA power sector CO2 emissions continuously increase over time, with Sadavarte–Venkataraman emissions being the highest of all at all times.

Using EDGAR, REAS, GAINS, and Nagpure–Gurjar, we further compared the emissions in the nine...
The uncertainty of domestic sector CO emissions from the Monte Carlo simulations is on average ±195% and is the largest of all sectors. This value is equivalent to the ±192% uncertainty provided by the REAS inventory for the same sector. On a regional scale, we find the largest differences in domestic emissions within the region 2 on IGP in 2005 (figure 2) being 4.4 Tg yr\(^{-1}\) between GAINS and Nagpure–Gurjar.

The uncertainty of industrial CO emissions from the Monte Carlo samplings is the second largest among the four sectors with ±154% and is larger than the ±118% uncertainty by REAS. REAS industrial CO emissions are consistently high in all nine regions with the largest difference of 2.3 Tg yr\(^{-1}\) (55% and 310% larger than EDGAR and GAINS, respectively) occurring in region 2 on the IGP.

Figure 2. Map of India regions and 2005 emissions of CO\(_2\), CO, SO\(_2\), NO\(_X\), and PM\(_{10}\) from EDGAR, REAS, GAINS, and Nagpure–Gurjar datasets from industrial (ind), transport (tra), power (pow), domestic (dom) sectors. Provinces included in each region are listed in supplementary table S4.

3.2. CO
The majority of CO emitted in India stems from the domestic sector, making up 69%–70%, 49%–56%, 76%–79%, and 64%–77% of total EDGAR, REAS, GAINS, and Sadavarte–Venkataraman, respectively.
While transport is not as large of a CO source as the domestic and industry sectors, the difference between the REAS and EDGAR estimates has grown from 521 Gg yr$^{-1}$ (within 10%) in 2000 to 6278 Gg yr$^{-1}$ by 2008 with REAS emissions over three times larger than those of EDGAR. The difference among the four inventories in the transport sector is most apparent in regions 2, 4, and 7 with larger than 50% SD. It is most likely that Nagpur–Gurjar until 2008 and REAS assumed much larger emissions either from super-emitters or significantly lower penetration of vehicles equipped with catalytic converters; the comparison of NO$_x$ emissions in transport also support this.

3.3. SO$_2$
Fossil fuel combustion in the power sector contributes the largest portion of SO$_2$ emissions in India but has the least uncertainty of ±46%. Power plant SO$_2$ emissions make up 61%–63% of total SO$_2$ in EDGAR, 47%–50% of REAS, 54%–57% of GAINS, and 70%–75% of Sadavarte–Venkataraman. Although the relative contributions of the power sector to total India SO$_2$ emissions for EDGAR and REAS differ, power sector emissions agree well for the two inventories between 2000 and 2008. Power plant emissions are within 10% of each other and both increase by 46%–47% from 2000 to 2008.

For SO$_2$ emissions, our Monte Carlo analysis shows the largest ±99% uncertainty, on average, in the domestic sector. Industry and transport share ±69 and ±66% uncertainty, respectively, on average. Because of the large differences among the inventory estimates, the national SO$_2$ emissions have an average uncertainty of ±48% over time, which is higher than any of the individual inventory estimates (figure 1). These values are also higher than the calculated uncertainty of ±16% in Streets et al (2003) for Asian SO$_2$ emissions.

The difference is apparent also on a subnational scale in every region (figure 2). In region 6, where the REAS total SO$_2$ emissions are the highest, their industrial SO$_2$ are 14.5 times that of EDGAR. Although the total magnitude is not as large as in region 6, region 1 on the IGP has the highest difference (132% SD) in industrial SO$_2$ with REAS estimating 34 (six) times greater than EDGAR (GAINS). Region 1 also has the highest difference for the total SO$_2$ with 108% SD. Besides region 7, where the power sector difference is the largest with 63%, REAS total SO$_2$ emissions are always the highest, followed by GAINS and EDGAR.

3.4. NO$_x$
Transport NO$_x$ emissions differ substantially among inventories and while the transport sector is the largest NO$_x$ source in REAS and GAINS, it is the second largest in EDGAR and Sadavarte–Venkataraman. The average uncertainty from the Monte Carlo methods for the transport NO$_x$ emissions is the second highest of all sectors (118%). This large difference confirms that part of the difference is due to the assumptions about the fleet characteristics (i.e., share of super-emitting vehicles and penetration of vehicles with catalytic converters) assumed in each inventory. EDGAR, without emissions from super-emitting vehicles, has the second lowest transport emissions throughout the years. Excluding the emissions from super-emitting vehicles in GAINS, the difference between GAINS and EDGAR transport NO$_x$ emissions is reduced to 28% from 49% with super-emitting vehicle emissions. For CO, super-emitters are mainly gasoline vehicles but for NO$_x$, they can be either gasoline or diesel. We therefore find even larger uncertainties in transport NO$_x$ emissions than in CO. Within the same region, the ratio of the transport NO$_x$ emissions among the four inventories remaining the same as that of transport CO emissions provides further confirmation of this assumption. In all regions, REAS transport emissions are at least three times as large as those of EDGAR, confirming larger emissions either from super-emitters and/or lower penetration of control technology, similarly to CO.

The largest NO$_x$ source sector in India in EDGAR and Sadavarte-Venkataraman and the second largest in REAS and GAINS is power generation. The trend is the same in all inventories and the only noticeable difference is that GAINS and Sadavarte-Venkataraman (REAS) have approximately 1 (0.3) Tg/year difference from EDGAR. This sector has the second best agreement among the inventories and the average uncertainty is ±83%. The best agreement is seen in the industrial emissions, where the uncertainty from the Monte Carlo simulation is, on average, ±65%. Both of these are, however, still larger than the uncertainties in the existing inventories.

NO$_x$ domestic emissions show the largest uncertainty of ±196% from the Monte Carlo samplings, much higher than the ±37% in Streets et al (2003) for Asian NO$_x$ emissions (2003). In all regions, it is clear that EDGAR and GAINS have similarly low domestic emissions, with much higher REAS and Nagpur–Gurjar emissions. The average SD among the four inventories is the second highest with 31% and region 1 again has the largest difference for the domestic sector (80%) as well as for total emissions (89%).

3.5. PM$_{10}$
Primary PM emissions typically include black carbon (BC), organic carbon (OC), metals, and dust; composition varies strongly between sources. Ambient PM composition includes, beyond primary components, also a variety of secondary compounds, such as sulfates, nitrates, ammonium, metals and other organic and inorganic compounds. REAS and EDGAR emissions of PM$_{10}$ in India do not show good agreement in any of the sectors explored in this paper. Indeed, uncertainty in all sectors from the Monte Carlo simulation exceeds ±120%. PM$_{10}$ emissions from the power sector in EDGAR are over three times larger than those of REAS
and GAINS and six times larger than those of Sadaverte–Venkataraman estimates. It is not only the magnitude of emissions from power sector that is different among the inventories, but the inventories also have a varying emissions trend. While EDGAR, REAS, and Sadaverte–Venkataraman show a 44, 58, and 45% increase in power sector emissions over time, respectively, GAINS estimates an 18% decrease. Since power capacity has grown in the considered period, different trends are likely due to the assumptions on penetration and efficiency of control equipment (electrostatic precipitators, fabric filters) on newly build power plants.

Domestic sector is the largest source sector for PM$_{10}$ in REAS, GAINS, and Sadaverte–Venkataraman and is the second largest in EDGAR. This is also the sector with highest uncertainty of, on average, ±315% in the Monte Carlo samplings. EDGAR domestic sector emissions are more than twice (50%) as large as those of REAS (Sadaverte–Venkataraman). However, unlike the varying growth rate shown for the power sector emissions, the trend over time is consistent among all inventories. Industry sector is the second largest in GAINS and Sadaverte–Venkataraman and the third largest in EDGAR and REAS. The uncertainty of, on average, ±123% from

![Figure 3](https://example.com/figure3.png)

**Figure 3.** Monthly mean surface concentrations of PM$_{10}$, O$_3$, CO, NO$_2$, and SO$_2$ simulated using REAS and EDGAR as model inputs in (a) January and (b) July of 2008.
this sector in the Monte Carlo simulation is the smallest for PM$_{10}$.

On the subnational scale, REAS total PM$_{10}$ emissions are the lowest among the three inventories in all regions (figure 2). EDGAR PM$_{10}$ emissions from the industrial and transport sector are, however, constantly lower than REAS or GAINS in every region, with national average EDGAR just 6% of REAS transport PM$_{10}$ in 2008. Transport sector has more than 100% SD in all years, except 2000 and 2005. The main reason for this large difference is due to the lack of super-emitters in EDGAR as mentioned earlier and there is good agreement between REAS and GAINS transport PM$_{10}$ emissions. However, even after omitting super-emitter contribution, GAINS transport emissions are still 450% higher than EDGAR emissions, which indicates that there are also other differences in emission factors and Table 2.

### Table 2. Regional difference of monthly mean (a) emissions between EDGAR and REAS and (b) concentrations between simulations using EDGAR and REAS. Results are shown with deviations of REAS from EDGAR in January and July of 2008.

#### (a)

| Regions | 2008 January | | 2008 July | |
|---------|--------------|-------------------|-------------|
|        | (REAS-EDGAR)/EDGAR (%) | (REAS-EDGAR)/EDGAR (%) | |
| PM$_{10}$ | CO | SO$_2$ | NH$_3$ | NO$_x$ | PM$_{10}$ | CO | SO$_2$ | NH$_3$ | NO$_x$ |
| 1 | −14 | 28 | 238 | 96 | 94 | −16 | 26 | 242 | 95 | 123 |
| 2 | −7 | 16 | 19 | 185 | 52 | −10 | 15 | 12 | 185 | 70 |
| 3 | −70 | 47 | −53 | 77 | −36 | −70 | 47 | −56 | 77 | −35 |
| 4 | −50 | 4 | 29 | 131 | 95 | −52 | 4 | 19 | 131 | 99 |
| 5 | −52 | −4 | 33 | 101 | 63 | −54 | −5 | 27 | 101 | 64 |
| 6 | −62 | 3 | 13 | 85 | −7 | −64 | 2 | 1 | 85 | −2 |
| 7 | −38 | 19 | 15 | 70 | 76 | −40 | 16 | 1 | 70 | 105 |
| 8 | −24 | 4 | −5 | 163 | 17 | −29 | 0 | −11 | 162 | 53 |
| 9 | −29 | 44 | 86 | 175 | 100 | −46 | 9 | 73 | 168 | 117 |

#### (b)

| Regions | 2008 January | | 2008 July | |
|---------|--------------|-------------------|-------------|
|        | (REAS-EDGAR)/EDGAR (%) | (REAS-EDGAR)/EDGAR (%) | |
| PM$_{10}$ | NO$_3$ | NH$_3$ | SO$_2$ | O$_3$ | CO | SO$_2$ | NO$_2$ | PM$_{10}$ | NO$_3$ | NH$_3$ | SO$_2$ | O$_3$ | CO | SO$_2$ | NO$_2$ |
| 1 | 45 | 140 | 85 | 36 | 9 | 7 | 122 | 84 | 24 | 73 | 52 | 12 | 20 | 3 | 193 | 96 |
| 2 | 55 | 107 | 74 | 23 | 11 | 13 | 21 | 42 | 21 | 96 | 60 | 6 | 14 | 6 | 23 | 59 |
| 3 | 5 | 109 | 46 | 0.4 | 10 | 17 | −30 | −24 | −18 | 71 | 46 | 0 | 14 | 10 | −38 | −11 |
| 4 | 19 | 148 | 60 | 5 | 11 | 7 | 6 | 44 | −5 | 89 | 86 | 23 | 20 | 2 | 17 | 69 |
| 5 | 19 | 203 | 60 | 14 | 11 | 4 | 10 | 33 | −16 | 42 | 53 | 6 | 9 | −2 | 4 | 36 |
| 6 | 38 | 124 | 80 | 32 | 12 | 11 | 18 | −3 | −11 | 33 | 30 | 7 | 7 | −2 | −11 | −2 |
| 7 | 72 | 152 | 123 | 67 | 10 | 20 | 50 | 72 | 11 | 87 | 61 | 5 | 20 | 5 | 1 | 70 |
| 8 | 43 | 95 | 75 | 32 | 11 | 13 | −1 | 13 | 23 | 104 | 66 | 12 | 18 | 2 | −16 | 36 |
| 9 | 52 | 91 | 82 | 55 | 2 | 20 | 85 | 100 | 43 | 152 | 108 | 55 | 17 | 4 | 67 | 120 |

### Figure 4.

Model-observation comparison of monthly CO and O$_3$ mixing ratios (a); model-simulated daily mean comparison with instantaneous SO$_2$ and NO$_x$ concentrations (b); and model-simulated monthly mean with MODIS AOD in January and July 2008 (c). For SO$_2$ and NO$_x$ concentration comparison, the letters denote the following sites: A—Ambernath, B—Amravati, C—Aurangabad, D—Chandrapur, E—Chipun, F—Delhi, G—Jalna, H—Kolhapur, I—Latur, J—Nagpur, K—Nashik, L—Pune, M—Solapur.
technology trends among inventories and possibly the way non-exhaust emissions (road, tire, and break wear) are represented.

3.6. Emission impacts on air quality

Figure 3 compares the simulated monthly mean surface CO, SO₂, NO₂, and O₃ mixing ratios, as well as PM₁₀ concentrations in January and July 2008, using the REAS and EDGAR emissions inventories. Table 2 highlights the differences in emissions between the two inventories and in simulated concentrations by region. We compare these two simulations with existing observations of CO and O₃ mixing ratios, SO₂ and NOₓ concentrations, and AOD in figure 4. The simulation with REAS emissions leads to higher CO but the regional difference is less than 20% in most
regions. The model always underestimates but REAS does a better job in reproducing them. For $O_3$, the simulation using REAS produces a slightly higher mixing ratio than EDGAR in all regions, but the difference is often less than 12% (7 ppbv). The model usually overestimates and EDGAR thus reproduces better, except for one site in Anantapur.

Although the two simulations show similar spatial distributions of NO$_2$ and SO$_2$ surface concentrations, their magnitudes differ substantially. The difference is particularly apparent in regions 1 and 9, where the simulation using REAS emissions estimates 36%–55% higher SO$_2$ and 84%–100% higher NO$_2$ due to 86%–238% and 95%–100% higher emissions, respectively. The comparison of NO$_x$ and SO$_2$ concentrations with surface measurements indicate that the model overestimates in cities but usually underestimates NO$_x$ in rural areas.

The REAS model simulation reproduces SO$_2$ better in general, and NO$_2$ better in most non-urban regions, while the EDGAR model simulation reproduces $O_3$ better. There is also significant overestimation of NO$_x$ in urban areas and significant underestimation in rural areas using both inventories. These suggest that transport emissions are possibly overestimated in REAS, as transport sector emissions are mainly from the urban centers. We find that India’s $O_3$ is largely sensitive to NO$_x$ (table 2), as found in Sharma et al. (2016), and considering that REAS transport emissions were estimated high for all species affirms this hypothesis. Second, the spatial distribution of the different proxies (i.e. population) used by the two inventories may be the cause of the differences in gridded emissions, as reflected in the model simulations.

The simulation using REAS produces 19%–72% (6–24 $\mu$g m$^{-3}$) higher surface concentrations of PM$_{10}$ in most regions than those using EDGAR, even though the primary PM$_{10}$ emissions is lower in REAS. The highest difference occurs in region 7 (72%) and we find that this is due to over 150, 120, and 65% differences in nitrate, ammonium, and sulfate aerosols in the simulation using REAS emissions. SO$_2$, NO$_x$, and NH$_3$ emissions in most regions are much greater in REAS as compared to those in EDGAR (table 2). Larger emissions of these species lead to a greater production of secondary inorganic aerosols in the model, and consequently, to higher PM$_{10}$ concentrations despite the lower primary PM$_{10}$ emissions. Spatial and temporal emissions of SO$_2$, NO$_x$, and NH$_3$ play an important role in secondary PM formation, as found in EPME (2016). We further find that REAS reproduces the MODIS AOD better than when using EDGAR emissions in region 7, confirming the importance of these SO$_2$, NO$_x$, and NH$_3$ emissions on PM concentrations.

Differences in emissions estimates affect concentrations and mixing ratios of pollutants in varying ways, depending on a region. Emission differences are large in regions 1, 2, 3 and 9 but we find the largest difference in concentrations and mixing ratios of various pollutants in region 7 in January. In July, the largest differences in air quality and emissions for most species are both in regions 1, 3 and 9. We find that this is due to pollution transport from neighboring regions and to seasonal changes in prevailing wind patterns and precipitation. Constraining the emissions in IGP (regions 1, 2, 7, 8, and 9) through field emission measurement campaigns and enhanced surface measurement network, therefore, will be most useful for the development of air pollution mitigation strategies in India.

4. Conclusions

In this study, various inventories of anthropogenic CO$_2$ and air pollutant precursor emissions are compared in India on national, regional, and sectoral scales. For the global CO, SO$_2$, and NO$_x$ emissions, the difference between inventories is 28%, 42%, and 17% in 2000, respectively (Granier et al. 2011). In India, we find the differences of total CO, SO$_2$, and NO$_x$ emissions among existing inventories to be 15%, 62%, and 58% in 2000. However, our analysis of regional and sector-specific emissions revealed that much higher differences exist at the sector level than the national total emissions. Our Monte Carlo results also highlight higher uncertainties in existing emissions estimates both at the national total and source sector levels than currently considered for most species. Although we can infer whether some differences result from activity data or emission factors, in order to fully understand the reasons behind discrepancies among the multiple inventories, data needs to be transparent and available. Our model results on the impact of emissions uncertainties highlights the importance of constraining emissions through field emission measurement campaigns and enhanced surface measurement network in the IGP region for understanding the local and regional air quality. Our study further highlights the importance of constraining SO$_2$, NO$_x$, and NH$_3$ emissions for secondary PM formation.

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