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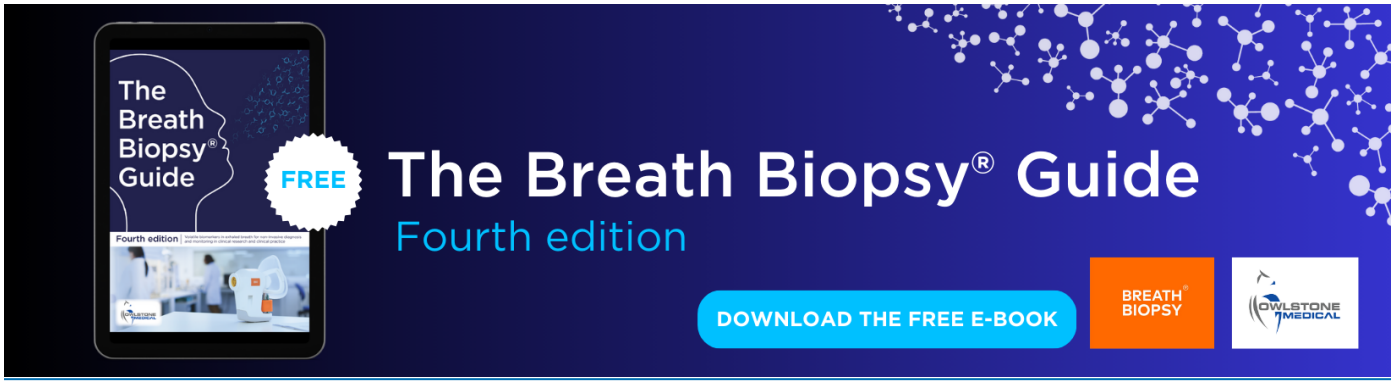

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LETTER

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

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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_2), carbon monoxide (CO), sulfur dioxide (SO_2), nitrogen oxides (NO_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_2, NO_x, and NH_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_2) from fossil fuel combustion and from industrial processes (production of cement, metals and chemicals) behind China and USA (Olivier *et al* 2015), after India surpassed Russia in 2010. Half of the top twenty most polluted cities in the world are in India (WHO 2016) and India is ranked the third worst of 180 countries for $PM_{2.5}$ (particulate matter with an aerodynamic diameter less than 2.5 μm) exposure (Hsu *et al* 2016). Several emissions inventories have been developed recently either specifically for India or for larger regions that include India (Streets *et al* 2003, Garg *et al* 2006, Ohara *et al* 2007, Klimont *et al* 2009, 2013, 2016, Zhang *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_2 and air pollutant precursors (carbon monoxide CO, sulfur dioxide SO_2, nitrogen oxides ($NO_x = NO + NO_2$), and particulate matter with an aerodynamic diameter less than 10 μm, PM_{10}) 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 CO_2, CO, SO_2 and NO_x emissions estimates by Garg *et al* (2006), total SO_2 emissions estimates by Smith *et al* (2011) and Klimont *et al* (2013), as well as the total NO_x emissions estimates by Ghude *et al* (2012).

2.2. Emissions comparison

We compared emissions of CO_2, CO, SO_2, NO_x, and PM_{10} 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 CO_2, seven inventories (EDGAR, REAS, INTEX-B, Nagpure–Gurjar, Sadavarte–Venkataraman, GAINS, and Garg) for CO and NO_x, seven inventories (EDGAR, REAS, INTEX-B, Nagpure–Gurjar, Sadavarte–Venkataraman, GAINS, and Lu) for SO_2, and four inventories (EDGAR, REAS, INTEX-B, and GAINS) for PM_{10}. 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; SO_2 transport and domestic; and NO_x 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

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	CO_2, N_2O, SO_2, CO, PM_{10}, NO_x	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	CO_2, N_2O, SO_2, CO, PM_{10}, $PM_{2.5}$, NO_x, BC, OC, NH_3, 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	SO_2, CO, PM_{10}, $PM_{2.5}$, NO_x, BC, OC, NMVOC	0.5° by 0.5°	spatial proxies at 1km × 1km resolution	22 Asian countries	Zhang *et al* 2009
Nagpure–Gurjar	2001–2011	residential and road-transport	CO_2, N_2O, SO_2, CO, PM_{10}, NO_x	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	CO_2, N_2O, SO_2, CO, $PM_{2.5}$, NO_x	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	CO_2	national level	NA	Global	IEA, 2014
GAINS	1990–2030 (5 year intervals, projection starting in 2015)	energy, domestic, industrial combustion and processes, road and non-road transportation and agriculture	CO, NO_x, PM_{10}, $PM_{2.5}$, CO_2, SO_2	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

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 O_3, it is similarand 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 (CBMZ) (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 SO_2 and NO_x

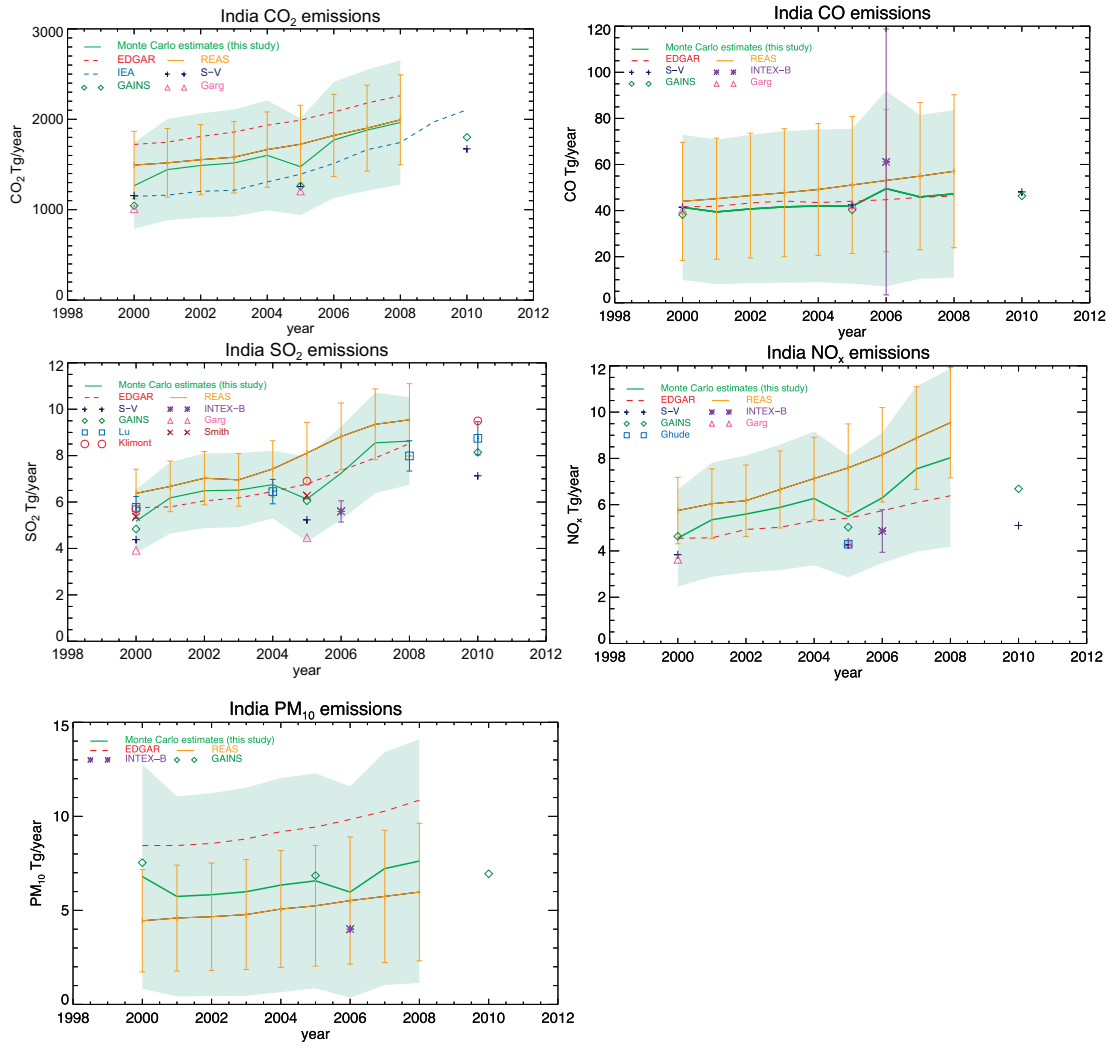

Figure 1. Time series of mean emissions and one standard deviation (green shade) from Monte Carlo results and mean and one standard deviation of different emission inventory estimates for Indian national CO_2, CO, SO_2, NO_x and PM_{10}. Different inventory is represented by a different style line or point. Sadavarte–Venkataraman emissions are represented by S–V and Nagpure–Gurjar emissions are represented by N–G in the legends.

concentrations (Maharashtra Pollution Control Board 2016), and monthly average CO and O_3 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. CO_2

Our Monte Carlo analysis illustrates that there is up to ±76% uncertainty in India's national total CO_2 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 CO_2 emissions are the lowest of all the inventories at 63 Tg CO_2 yr^{-1} in 2000, contributing to only 13% of REAS domestic CO_2 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 CO_2 emissions in India. EDGAR, REAS, GAINS, Sadavarte–Venkataraman, and IEA power sector CO_2 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

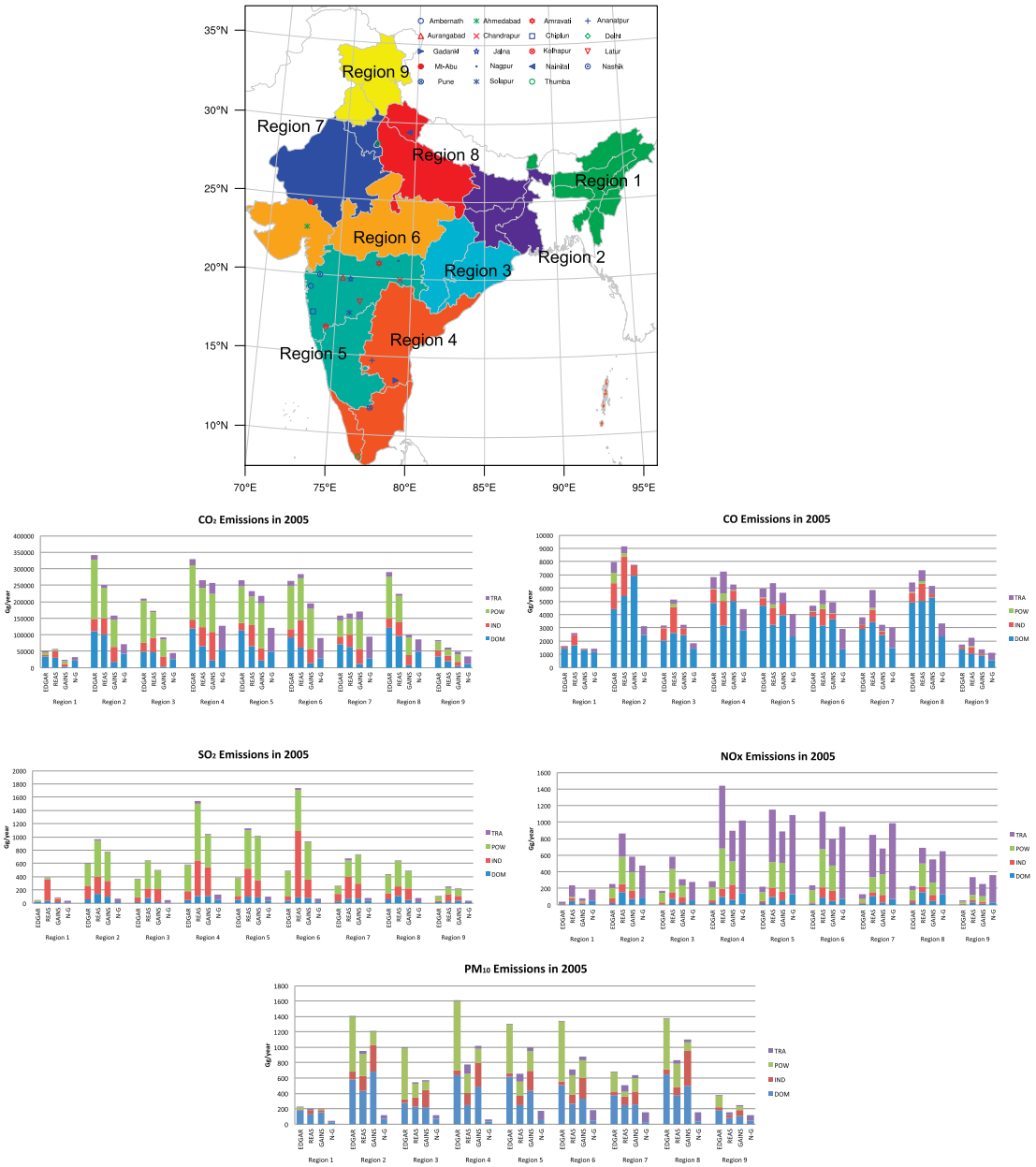

Figure 2. Map of India regions and 2005 emissions of CO_2, CO, SO_2, NO_x, and PM_{10} from EDGAR, REAS, GAINS, and Nagpure–Gurjar datasets from industrial (ind), transport (tra), power (pow), domestic (dom) sectors. Provinces included in each region are listed in supplementary table S4.

subnational regions in 2005 (figure 2). The largest difference of 75% SD in the domestic sector exists in region 8 on the Indo-Gangetic Plain (IGP). This is also the region with the largest difference for the regional total CO_2 (47% SD) and for transport (44%) among EDGAR, REAS, and GAINS. Another large difference of 73% SD in industry come from region 1, which is also on the IGP, and the largest difference for the power sector is found in region 3 (44%), where the greatest number of large (> 2000 MW) power plants exist.

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. 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.

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 Nagpure–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 Sadaverte-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 Sadaverte-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 Nagpure–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

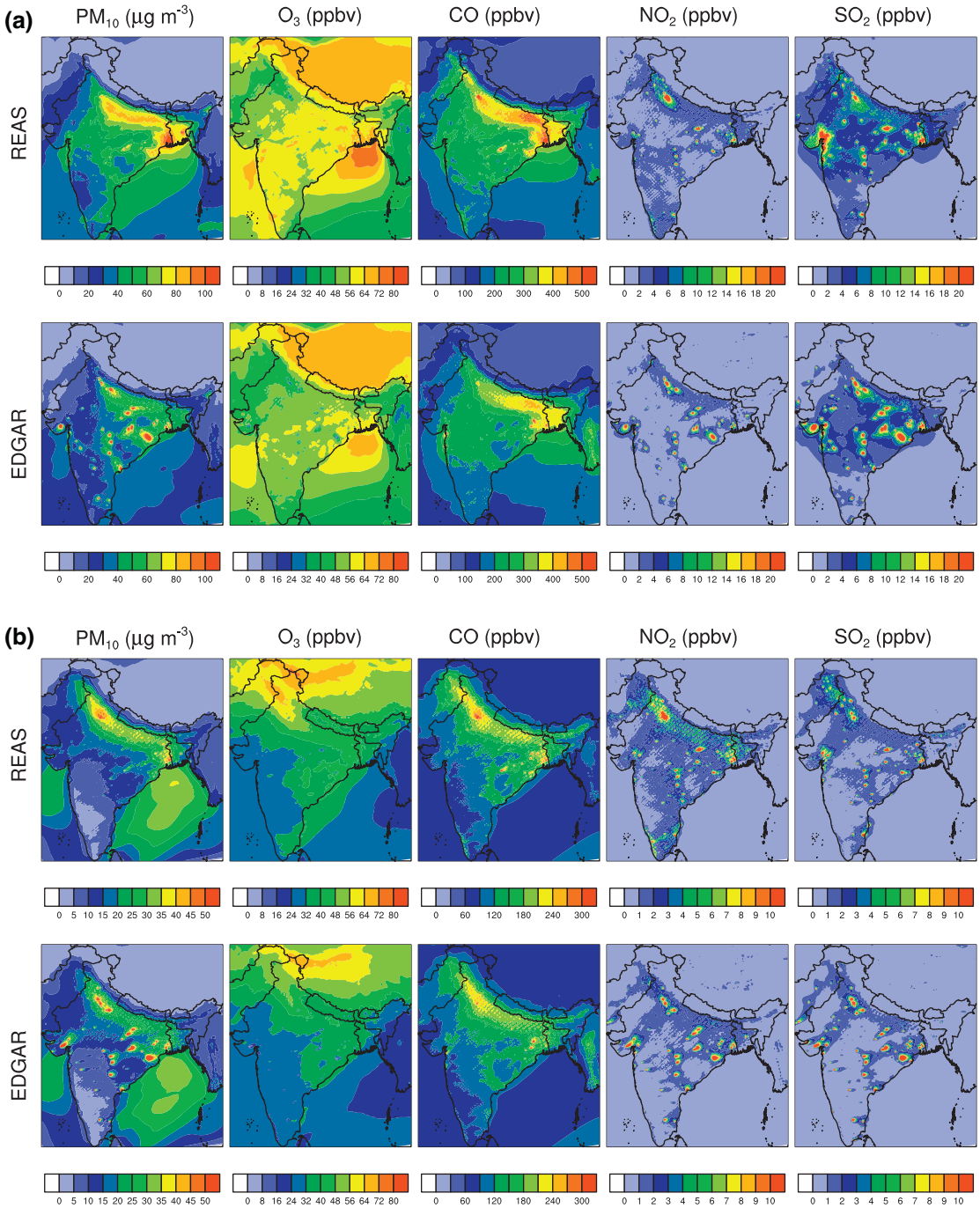

Figure 3. Monthly mean surface concentrations of PM_{10}, O_3, CO, NO_2, and SO_2 simulated using REAS and EDGAR as model inputs in (*a*) January and (*b*) July of 2008.

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 Sadavarte–Venkataraman and the third largest in EDGAR and REAS. The uncertainty of, on average, ±123% from

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 (REAS-EDGAR)/EDGAR (%)					2008 July (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	75	168	117

(b)

Regions	2008 January (REAS-EDGAR)/EDGAR (%)								2008 July (REAS-EDGAR)/EDGAR (%)							
	PM_{10}	NO_3	NH_4	SO_4	O_3	CO	SO_2	NO_2	PM_{10}	NO_3	NH_4	SO_4	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

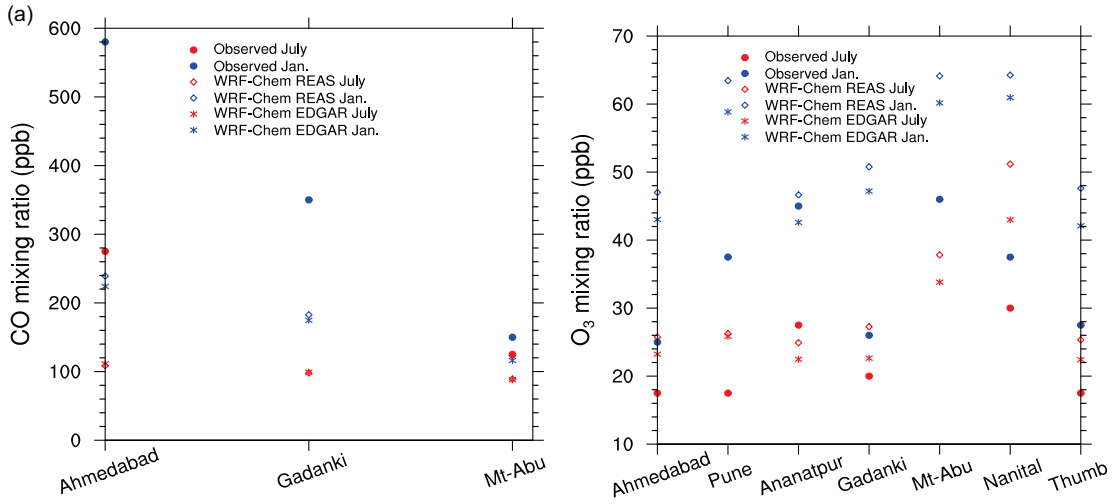

Figure 4. Model-observation comparison of monthly CO and O_3 mixing ratios (*a*); model-simulated daily mean comparison with instantaneous SO_2 and NO_x concentrations (*b*); and model-simulated monthly mean with MODIS AOD in January and July 2008 (*c*). For SO_2 and NO_x concentration comparison, the letters denote the following sites: A—Ambernath, B—Amravati, C—Aurangabad, D—Chandrapur, E—Chiplun, F—Delhi, G—Jalna, H—Kolhapur, I—Latur, J—Nagpur, K—Nashik, L—Pune, M—Solapur.

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

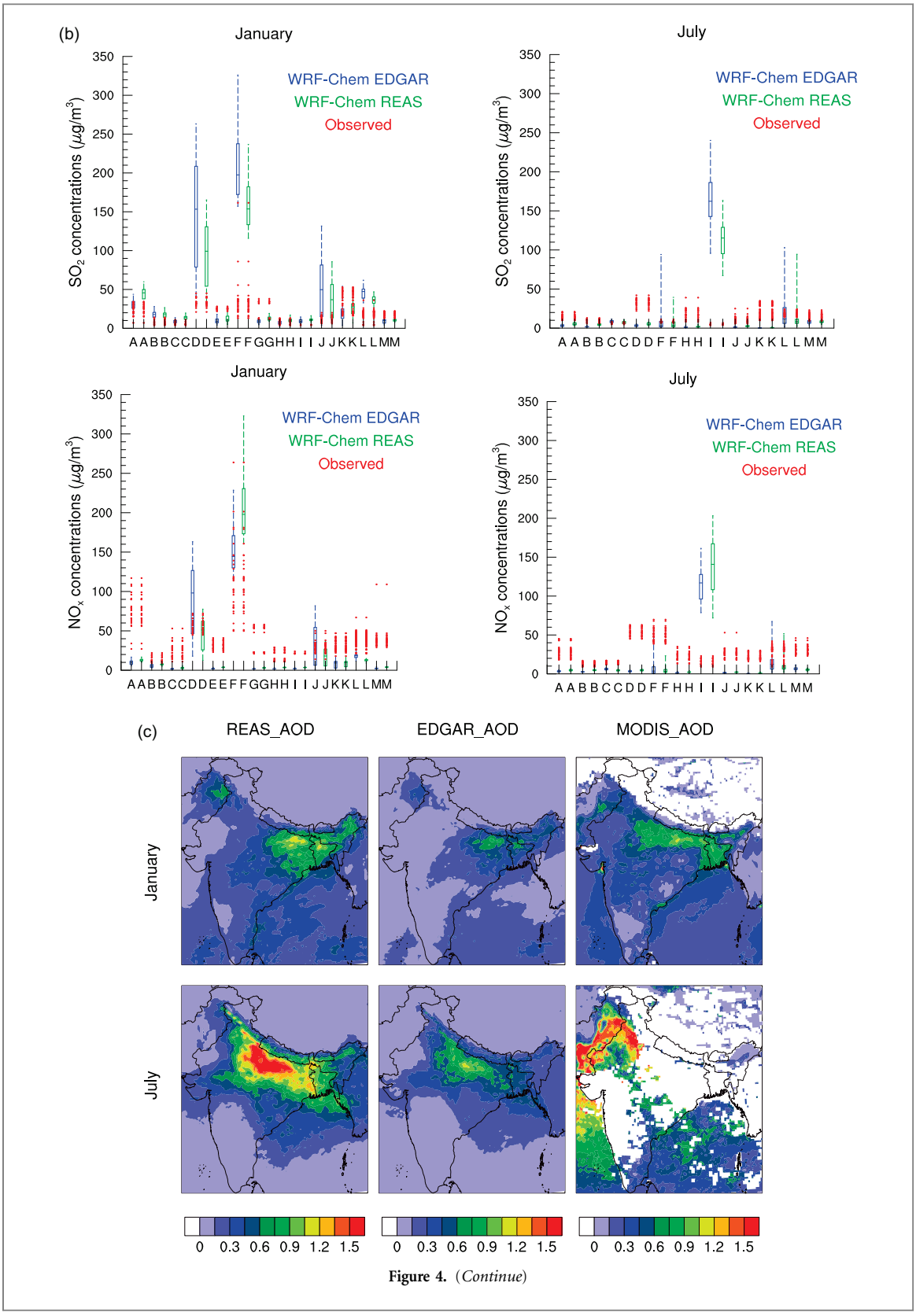

Figure 4. (*Continue*)

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, SO_2, NO_2, and O_3 mixing ratios, as well as PM_{10} 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 O_3 mixing ratios, SO_2 and NO_x 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 Ananatpur.

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_x 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 first 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 EMEP (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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