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Review Article

Five-year Ground-based Observation Trend of $PM_{2.5}$ and PM_{10}, and Comparison with MERRA-2 Data over India

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ABSTRACT A comprehensive analysis of pollutant's trend and their measurement techniques are crucial for evaluating the air quality, and thereby helpful in formulating better control policies. In this report, we summarise ground based $PM_{2.5}$ and PM_{10} data report in India (2015–2019). The important points discussed here are: (i) review of the ground-based data of $PM_{2.5}$ and PM_{10} and the techniques used, (ii) mapping of the data over India with spatial and temporal distribution so that better understanding on PM pollution level can be made, (iii) identifying the technological gaps in measurement of PM concentration in India, and (iv) evaluation of MERRA-2's (Modern-Era Retrospective Analysis for Research and Applications, Version 2) simulation of $PM_{2.5}$ against surface measurements in India to better understand biases for spatial and seasonal distribution, and then (v) suggestions for better PM measurement protocols, policies and metrological aspects for both measurement and control policies. It is observed that the amount of ground data on ambient monitoring of fine PM is insufficient and has several inconsistencies which require adequate attention. In India, not much work has been done on developing certified reference materials, traceable standards and calibration facility for particulate matter measurement which is a crucial step to ensure quality checks. Further, the comparison of MERRA-2 and ground $PM_{2.5}$ concentrations revealed huge discrepancies with underestimating $PM_{2.5}$ measurements in highly polluted regions like the Indo-Gangetic plain, especially during winter when pollution load was high. Better $PM_{2.5}$ agreement was found in summer and monsoon season, based on performance statistics explained in this paper. Inconsistencies between MERRA-2 and ground $PM_{2.5}$ are partly due to few limitations in MERRA-2 reanalysis method which are discussed in this paper, apart from several issues in ground-based observation. The aim of this review and comparison is to highlight such issues and give more attention to the importance of data quality assurance for effective air quality management. The present study may be helpful for the researchers in evaluating and choosing appropriate reanalysis products for their future studies.

KEY WORDS MERRA-2, Ground-measurement, $PM_{2.5}$ and PM_{10}, Seasonal variability, Measurement techniques

1. INTRODUCTION

Atmospheric pollution is one of the major environmental concerns worldwide, especially in urban and sub-urban areas. Particulate Matter (PM) is known as one of

the significant contributors to poor air quality which is defined as solid, liquid and/or mixed particles suspended in air. They emit from both natural and anthropogenic sources. The particles having aerodynamic diameter $\leq$2.5 μm ($PM_{2.5}$) have adverse impact on human health since they can penetrate deep into the respiratory system causing cardiovascular and respiratory diseases. Thereby, $PM_{2.5}$ mass concentration was adopted as the primary parameter for ambient air particulate pollution and generally used as indicator of adverse health effects first in the United States in 1997, and later in different countries of the world. In India, CPCB (Central Pollution Control Board) notified National Ambient Air Quality Standards (NAAQS, revised 2009) stating standards of ambient air quality under the Air (prevention and control of pollution) Act 1981. As per the guidelines, cities are required to attain $PM_{2.5}$ concentrations below 40 μg/m^3 (annual average) and below 60 μg/m^3 (24-hour average). For PM_{10}, mass concentration standards below 60 μg/m^3 (annual average) and below 100 μg/m^3 (24-hour average) are permissible.

Despite being a key contributor to air quality, not much progress has been done to ensure the data quality of PM ambient mass measurements in India. Lack of uniform and continuous PM monitoring is one of the important issues hindering effective air quality management. These are the possible reasons for which several Indian cities are scaled as most polluted cities in Asia and world in the listing by WHO (Air quality database, WHO, April, 2022). To ensure accurate and precise PM measurements, regular quality checks and calibration of instruments/methods deployed are of utmost importance which in turn helps policy makers to effectively combat and control emissions. Various central and state regulatory bodies along with research institutes are involved in monitoring of PM at various stations throughout the country. However, the present literature shows inconsistent data and limited air monitoring network.

So, considering these issues, this paper aims to review and map ground-based PM studies published in recent years (2015–2019) across India and the techniques used for PM monitoring, highlight limitations and variations in measurements and to suggest better PM measurement strategy. Literature shows high loading of $PM_{2.5}$ in most of the Indian region, especially in the Ingo-Gangetic Plain (IGP), almost all year round with values exceeding national standards several folds. The IGP is regarded as the hotspot for aerosol source since it is densely populated, and meteorological as well as topographical conditions which favour the accumulation loading of PM with lower planetary boundary layer (PBL) height, especially in the winter season (Jain *et al.*, 2021). Likewise, trends in PM mass concentrations in different regions of India, their sources along with measurement techniques used are discussed in this paper. Furthermore, it is observed that although ground air quality monitoring stations exist in various regions in India, but due to their limited and non-uniform distribution with almost no station in rural areas, the air quality data is not sufficient to analyse spatial and temporal distribution of PM. Henceforth, to study better spatiotemporal $PM_{2.5}$ variation in India with the available data, satellite data is used to compare with ground-based PM monitoring and thereby calculate differences in $PM_{2.5}$ mass concentration on spatial and seasonal basis.

This review is organised into six sections. Section 2 reviews published studies on ground-based mass concentration of $PM_{2.5}$ and PM_{10} in five years (2015–2019) in India along with measurement techniques. Section 3 introduces satellite $PM_{2.5}$ data and reanalysis method. In Section 4 results and discussions are given, comparing ground $PM_{2.5}$ mass concentration with that of satellite assimilated data on seasonal and spatial basis using statistical parameters (correlation coefficient, mean bias, standard deviation of bias, mean fraction, FAC2) to study spatio-temporal PM distribution over India. In addition, factors resulting in underestimation of MERRA-2 $PM_{2.5}$ are discussed. In Section 5, the limitations in ground air quality monitoring network are discussed. Conclusion and suggestions for better and accurate PM measurement techniques are given in Section 6.

2. GROUND-BASED MEASUREMENT TRENDS

2.1 Ambient $PM_{2.5}$ Mass Concentrations in Various Cities during the Years 2015–2019

We have reviewed available literature on $PM_{2.5}$ mass concentration ground-based measurements in Indian cities during 2015–2019. Fig. 1 displays the average $PM_{2.5}$ concentration distribution in different cities in India. The concentration values for each region along with the monitoring techniques are given in Table 1. The Indian Himalaya Range (IHR) is a great mountain terrain which encompasses an area of about 2,500 km^2 and forms a bar-

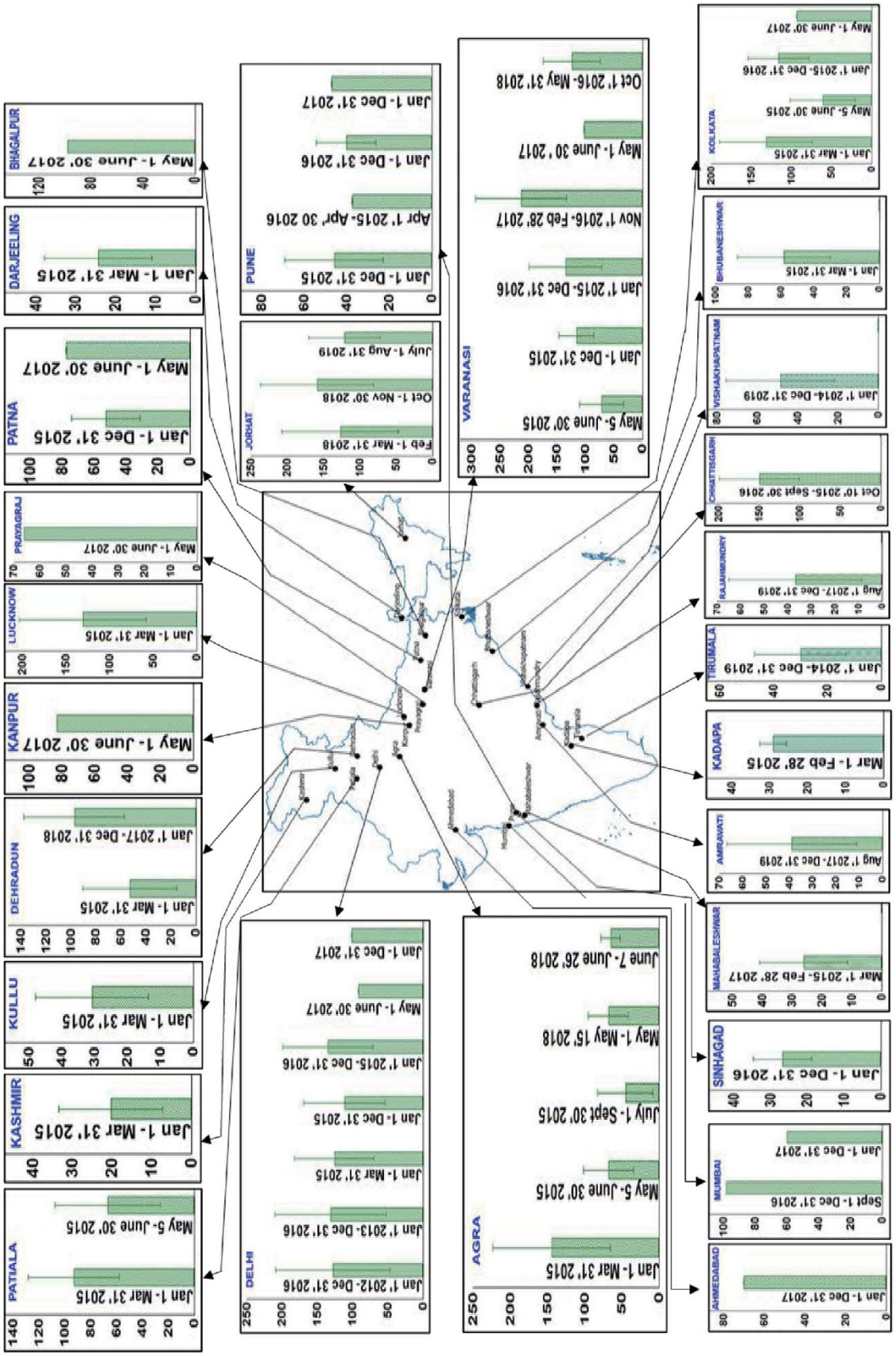

Fig. 1. Distribution of ground $PM_{2.5}$ mass concentration in India (2015–2019), where Y-axes represent mass concentration of $PM_{2.5}$ in $\mu g/m^3$ and date of sampling on X-axis.

Table 1. Summary of ground-based ambient average mass concentration measurements of PM from several cities across India in the past five years (2015–2019).

City	Sampling year	PM_{10} (μg/m^3)	Technique used	$PM_{2.5}$ (μg/m^3)	Technique used	Reference
			IHR			
Kashmir	January–March 2015	32 ± 27.6	Respirable dust sampler	20.3 ± 13.1	Fine dust HVS	Sen *et al.* (2017)
Kullu	January–March 2015	48.5 ± 16	Respirable dust sampler	30.8 ± 17.2	Fine dust HVS	Sen *et al.* (2017)
Nainital	January–March 2015	42 ± 24	Respirable dust sampler		Fine dust HVS	Sen *et al.* (2017)
Dehradun	November 2014–June 2015	90 ± 32 (winter) 87 ± 40 (summer) (Mean = 89 ± 36)	Partisol speciation sampler	63 ± 27 (winter) 39 ± 19 (summer) (Mean = 51 ± 23)	Partisol speciation sampler	Soni *et al.* (2020)
Dehradun	January–March 2015			52.9 ± 37.7	Fine dust HVS	Sen *et al.* (2017)
Dehradun	November 2015–May 2016	186.2 ± 16.7 (winter) 153.8 ± 17.9 (summer) (Mean = 170 ± 17.3)	HVS (Model: Spectro APM 460 BL, India)			Shridhar (2019)
Dehradun	2017–2018	134.21 ± 44.80 (Diwali sampling)	Portable aerosol spectrometer (Model-1371, GRIMM Aerosol Technik, Germany)	97.10 ± 40.36 (Diwali sampling)	Portable aerosol spectrometer (Model-1371, GRIMM Aerosol Technik, Germany)	Prabhu *et al.* (2019)
Darjeeling	January–March 2015	48 ± 18	Respirable dust sampler	24.3 ± 13.5	Fine dust HVS	Sen *et al.* (2017)
Darjeeling	December 2018–February 2019	48 ± 16	Particle sampler (1.2 m^3/min)			Sharma *et al.* (2020)
			IGP			
Delhi	January 2012–December 2016			128 ± 81	Fine particle sampler (APM 550, Envirotech, Delhi, IN)	Sharma *et al.* (2018b)
Delhi	January 2013–December 2016	238 ± 106	High volume respirable dust sampler (AAS-217 NL; Make: M/s. Ecotech, India)	131 ± 79	High volume respirable dust sampler (AAS-217 NL; Make: M/s. Ecotech, India)	Jain *et al.* (2020)
Delhi	January–March 2015	191.6 ± 58.1	Respirable HVS	125.7 ± 56.6	Fine dust HVS	Sen *et al.* (2017)
Delhi	January–December 2015	177.81 ± 95.50	BAM (SAFAR)	111.10 ± 59.90	BAM (SAFAR)	Krishna *et al.* (2018)

Table 1. Continued.

City	Sampling year	PM_{10} (μg/m^3)	Technique used	$PM_{2.5}$ (μg/m^3)	Technique used	Reference
Delhi	January 2015–December 2016	242 ± 95	High volume respirable dust sampler (AAS 217 NL, Ecotech, India)	135 ± 64	Fine particle sampler (APM 550, Envirotech, India)	Jain *et al.* (2021)
Delhi	January 2015–December 2017	222 ± 87	Respirable dust Sampler (Model: AAS-217 NL; Make: M/s. Ecotech, Delhi, India)			Sharma *et al.* (2018a)
Delhi	May–June 2017	162	HVS (APM 550M, Envirotech)	91.5	HVS (APM 550M, Envirotech)	Devi *et al.* (2020)
Delhi	January–December 2017	200	BAM	101	BAM	Anand *et al.* (2019)
Agra	January–March 2015	206.5 ± 90.9	Respirable dust sampler	144 ± 79	Fine dust HVS	Sen *et al.* (2017)
Agra	5 May–30 June 2015	156.2 ± 118.9	Respirable dust sampler	67.8 ± 33.6	Respirable dust sampler	Sen *et al.* (2017)
Agra	July–September 2015	137.09 ± 61 (urban) 270.14 ± 21 (semi-urban) (Mean = 203 ± 45)	Grimm portable aerosol spectrometer	41.45 ± 40 (urban) 48.88 ± 34 (semi-urban) (Mean = 45 ± 37)	Grimm portable aerosol spectrometer	Tiwari *et al.* (2020)
Agra	March 2016–March 2017	214.6 ± 60	HVS(APM 550, Envirotech)			Sah *et al.* (2019)
Agra	1–15th May 2018			68 ± 26.9	Envirotech sampler (APM 550)	Kumari *et al.* (2020)
Agra	7–26th June 2018	234 ± 84	BGI PQ200 particulate samplers (Mesa Labs, USA, USEPA)	65 ± 13	BGI PQ200 particulate samplers (Mesa Labs, USA, USEPA)	Nagar *et al.* (2020)
Patiala	January–March 2015		Respirable HVS	93 ± 35	Fine dust HVS	Sen *et al.* (2017)
Patiala	5 May–30 June 2015	175.4 ± 56	Respirable HVS	66.6 ± 40.7	Fine dust HVS	Sen *et al.* (2017)
Kanpur	May–June 2017	147	HVS (APM 550M, Envirotech)	83.2	HVS (APM 550M, Envirotech)	Devi *et al.* (2020)
Lucknow	January–March 2015	196 ± 90	Respirable dust HVS	130.5 ± 73.0	Fine dust HVS	Sen *et al.* (2017)

Table 1. Continued.

City	Sampling year	PM_{10} (μg/m^3)	Technique used	$PM_{2.5}$ (μg/m^3)	Technique used	Reference
Lucknow	October 2015–September 2016	161 ± 90	Respirable dust sampler (Model-460BL, Env)			Bharti *et al.* (2017)
Varanasi	January 2014–December 2017	178 ± 105	Respirable dust sampler (APM-460BL)			Murari *et al.* (2020)
Varanasi	January–March 2015	238 ± 82	Respirable dust HVS			Sen *et al.* (2017)
Varanasi	5 May–30 June 2015			71.7 ± 38.7		Sen *et al.* (2017)
Varanasi	January 2015–December 2016	257 ± 90	High volume respirable dust sampler (AAS 217 NL, Ecotech, India)	99 ± 33	Fine particle sampler (APM 550, Envirotech, India)	Jain *et al.* (2021)
Varanasi	November 2016–February 2017	213 ± 80	Respirable dust sampler (APM 460NL)	134 ± 48	Respirable dust sampler (APM 550)	Pratap *et al.* (2020)
Varanasi	May–June 2017	165	HVS (APM 550M, Envirotech)	102	HVS (APM 550M, Envirotech)	Devi *et al.* (2020)
Varanasi	October 2016–May 2018	207.992 ± 66.861 (winter, 2016) 177.344 ± 68.971 (winter, 2017) 173.784 ± 37.671 (post-monsoon, 2016) 153.577 ± 82.595 (post-monsoon, 2017) (Mean = 178 ± 66)	Respirable dust sampler (APM-460BL)	129.808 ± 36.348 (winter, 2016) 110.732 ± 42.581 (winter, 2017) 132.434 ± 51.752 (post-monsoon, 2016) 121.46 ± 65.374 (post-monsoon, 2017) (Mean = 124 ± 50)	APM 550 (Make: M/s Envirotech, India)	Kumar *et al.* (2020)
Mirzapur	December 2014–December 2017	131 ± 56	Respirable dust sampler (APM-460BL)			Murari *et al.* (2020)
Kolkata	January–March 2015			131.4 ± 58.1		Sen *et al.* (2017)
Kolkata	5 May–30 June 2015			60.6 ± 40.7		Sen *et al.* (2017)
Kolkata	January 2015–December 2016	179 ± 77	Respirable HVS (AAS 217NL, Ecotech)	116 ± 38	Fine particle sampler (APM-550, Envirotech)	Jain *et al.* (2021)

Table 1. Continued.

City	Sampling year	PM_{10} (μg/m^3)	Technique used	$PM_{2.5}$ (μg/m^3)	Technique used	Reference
Kolkata	May–June 2017	151	HVS (APM 550M, Envirotech)	93	HVS (APM 550M, Envirotech)	Devi *et al.* (2020)
			Western India			
Pune	January–December 2015	98 ± 45.5	BAM-1020 (SAFAR)	46 ± 23	BAM-1020 (SAFAR)	Krishna *et al.* (2018)
Pune	April 2015–April 2016			37.3	Fine dust sampler (NPM-FDS)	Gawhane *et al.* (2017)
Pune	January–December 2016	63 ± 25	Dust sampler (NETEL, India) at q = 16.7 lpm	40 ± 14	Dust sampler (NETEL, India) at q = 16.7lpm	Gawhane *et al.* (2019)
Pune	January–December 2017	87	BAM	47	BAM	Anand *et al.* (2019)
Mumbai	January–December 2017	99	BAM	60	BAM	Anand *et al.* (2019)
Mumbai	September 2016–December 2016			96.61 (urban) 100.30 (sub-urban) (Mean = 98.45)	MiniVol™ Tactical Air Sampler (TAS), USEPA	Masih *et al.* (2019)
Mahabaleshwar	March 2015–February 2017			26 ± 14	GRIMM Environmental Dust Monitor (EDM 180)	Buchende *et al.* (2019)
Sinhagad (Rural-high altitude site)	January–December 2016	46.8 ± 12	Dust sampler (NETEL India Pvt. Ltd.)	27 ± 8	Dust sampler (NETEL India Pvt. Ltd.)	Budhavant *et al.* (2020)
Ajmer	January–March 2015	80 ± 44	Respirable HVS			Sen *et al.* (2017)
Ahmedabad	May–June 2017	116 ± 36 (Bapunagar) 228 ± 43 (Narol) 133 ± 29 (Paldi) 101 ± 21 (Income tax) 70 ± 20 (Science City) (129.6/31.07) (Mean = 130 ± 31)			Respirable dust sampler (APM-460NL, Envirotech)	Patel *et al.* (2021)
Ahmedabad	January–December 2017	126	BAM (SAFAR)	70	BAM (SAFAR)	Anand *et al.* (2019)
			Eastern India			
Bhubaneshwar	January–March 2015	119.7 ± 36.9	Respirable HVS	58.1 ± 28.4	Fine dust HVS	Sen *et al.* (2017)
Patna	March 2013–December 2014	192.0 ± 132.8	BAM			Tiwari *et al.* (2016)

Table 1. Continued.

City	Sampling year	PM_{10} (μg/m^3)	Technique used	$PM_{2.5}$ (μg/m^3)	Technique used	Reference
Patna	January–December 2015	82 ± 27	Micro-aethelometer	52.62 ± 21.54	Micro-aethelometer	Arif *et al.* (2018)
Patna	May–June 2017	149	HVS (APM 550M, Envirotech)	77.4	HVS (APM 550M, Envirotech)	Devi *et al.* (2020)
Bhagalpur	May–June 2017	194	HVS (APM 550M, Envirotech)	98	HVS (APM 550M, Envirotech)	Devi *et al.* (2020)
Jorhat	February–March 2018	261 ± 89	Respirable dust sampler (APM-460NL, Envirotech)	126.76 ± 80.40	Fine particulate sampler (APM 550, Envirotech)	Islam *et al.* (2020a)
Jorhat	October–November 2018	274 ± 79	Respirable dust sampler (APM-460NL, Envirotech)	159 ± 77.6	Fine particulate sampler APM 550, Envirotech)	Islam *et al.* (2020a)
Jorhat	July–August 2019	153 ± 45	Fine particulate sampler (APM 550, Envirotech)	121 ± 49	Respirable dust sampler (APM 460NL)	Islam *et al.* (2020b)
Prayagraj	May–June 2017	118	HVS (APM 550M, Envirotech)	66	HVS (APM 550M, Envirotech)	Devi *et al.* (2020)
Chhattisgarh	October 2015–September 2016			71.8 ± 27 (rural) 133 ± 51 (urban) 244.5 ± 63.3 (industrial) (Mean = 150 ± 49)	Mini vol sampler	Sahu *et al.* (2018)
Southern India						
Vishakhapatnam (Urban industrial site)	January 2014–December 2019	110 ± 48	BAM	48.5 ± 27	BAM	Varaprasad *et al.* (2021)
Tirumala (Rural hilly site)		60 ± 25	BAM	30 ± 17	BAM	Varaprasad *et al.* (2021)
Amaravati (Residential)	August 2017–December 2019	79 ± 45	BAM	40 ± 28	BAM	Varaprasad *et al.* (2021)
Rajahmundry (Industrial site)		75 ± 42	BAM	37 ± 28	BAM	Varaprasad *et al.* (2021)
Kadapa (Andhra Pradesh)	March 2013–February 2015	61.4 ± 3.4	HVS	29.0 ± 3.5	HVS	Begam *et al.* (2017)

rier between the Plateau of Tibet to the north and alluvial Indo-Gangetic Plain (IGP) to the south (https://dst.gov.in). Out of all locations in IHR, Dehradun shows highest levels of $PM_{2.5}$, i.e. 53 ± 38 μg/m^3 (January–March 2015) (Sen *et al.*, 2017) and 97 ± 40 μg/m^3 (Diwali sampling months of 2017–2018) (Prabhu *et al.*, 2019). This is predominantly due to increased local sources such as vehicular emissions and also long-range transport of pollutants from IGP (Soni *et al.*, 2020; Sen *et al.*, 2017). While, low levels of mean $PM_{2.5}$ were reported in Kashmir and Darjeeling, i.e. 20 ± 13 μg/m^3 and 24 ± 13.5 μg/m^3, respectively during January–March 2015 (Sen *et al.*, 2017). Similarly, in Kullu the average $PM_{2.5}$ concentration levels have been reported as 31 ± 17 μg/m^3 (during January–March 2015) (Sen *et al.*, 2017). As per the available literature, it reveals that the average $PM_{2.5}$ concentration in IHR (32 μg/m^3) is around 4.5 times lower as compared to IGP urban sites (143 μg/m^3) such as Delhi, Agra and Varanasi. Low particulate concentration levels in the IHR may be attributed to the pristine conditions of Himalayas and its sparse population resulting in fewer anthropogenic emissions.

The Indo-Gangetic Plain (IGP) is a vast fertile flat land covering an area of about 700,000 km^2, situated in the foothills of Himalayas, flanked by Bay of Bengal on its east, the Thar desert on its west and semi-arid Deccan plateau to its south (Sen *et al.*, 2016; Beig and Ali, 2006). Based on the literature surveyed, we observed that in the IGP, the average $PM_{2.5}$ concentration levels are higher than other regions throughout the study period.

In Delhi, the mean $PM_{2.5}$ mass concentration ranged from 91.5 μg/m^3 (May–June 2017) (Devi *et al.*, 2020) to 135 ± 64 μg/m^3 (January 2015–December 2016) (Jain *et al.*, 2021) showing seasonal trend with high concentration in winter sampling months as compared to summer. The major reasons for high concentration are emissions from biomass burning, fossil fuel combustion and vehicular activities (Jain *et al.*, 2021). Wood/waste/biomass combustion is a significant contributor to $PM_{2.5}$ loading during winter while crustal material was found to be prominent source in summer (Pant *et al.*, 2015). Further, meteorological conditions like calm weather conditions and low inversion layer in winter favours higher loading of PM.

Patiala, located in the upper IGP, recorded an increase in $PM_{2.5}$ concentration, i.e. from 55 ± 13.5 μg/m^3 in May–June 2014 (Sen *et al.*, 2014) to 66.6 ± 41 μg/m^3 in May–June 2015 (Sen *et al.*, 2017), due to increase in open burning activities during 2015 compared to 2014. Relatively high PM concentration was observed during winter sampling months in Patiala i.e. 93 ± 35 μg/m^3 (January–March 2015) (Sen *et al.*, 2017). Due to fossil fuel emissions from industries located in the vicinity of Patiala, post-harvest stubble burning during October–November and wood/biomass burning in winters are the important sources of $PM_{2.5}$ in Patiala (Rastogi *et al.*, 2016).

As for Agra, the mean $PM_{2.5}$ concentration ranged from 45 ± 37 μg/m^3 (July–September 2015) (Tiwari *et al.*, 2020) to 144 ± 79 μg/m^3 (January–March 2015) (Sen *et al.*, 2017) showing a strong seasonal trend. Also, a slight increase in concentration can be observed in average PM mass concentration reported from previous study i.e. 121 ± 36 μg/m^3 (20 January–5 February 2014) (Sen *et al.*, 2014) to 144 ± 79 μg/m^3 (January–March 2015) (Sen *et al.*, 2017). It is a heavily polluted site in Northern India which is influenced by emissions from factories, vehicles, post-harvest stubble burning, brick kilns, etc. (Pipal *et al.*, 2014).

As for Varanasi, an increasing trend in the mean concentration of $PM_{2.5}$ can be observed from 99 ± 33 μg/m^3 (January 2015–December 2016) (Jain *et al.*, 2021) to 134 ± 48 μg/m^3 (November 2016–February 2017) (Pratap *et al.*, 2020) followed by a slight decline in mass concentration i.e. 124 ± 50 μg/m^3 (October 2016–May 2018) (Kumar *et al.*, 2020). Varanasi located in the middle IGP region observed very high pollution load due to combined effect of accumulation of particulates from upper IGP and emissions from local sources (Sen *et al.*, 2016). Kolkata, being the third most densely inhabited city in India (Census, 2011), recorded an increasing trend in $PM_{2.5}$ concentration from 48 ± 9.3 μg/m^3 (May–June 2014) (Sen *et al.*, 2016) to 93 μg/m^3 (May–June 2017) (Devi *et al.*, 2020). High population, vehicular and industrial emissions are predominant sources of $PM_{2.5}$ in addition to the emissions from biomass burning and anthropogenic activities predominately from upper and middle IGP cities (Jain *et al.*, 2020). Jamshedpur, an urban site in east India, also recorded a high $PM_{2.5}$ annual average (119 ± 43 μg/m^3) from December 2017 to November 2018 (Ambade *et al.*, 2021). Potential sources contributing to high PM concentration are the anthropogenic activities such as coal burning, fossil fuel burning wood burning etc.

Among the north-eastern Indian sites, Jorhat reported substantially high loading of $PM_{2.5}$ showing seasonal variability with highest average mass concentration

observed during post-monsoon season as 159 ± 77.6 μg/m^3 (October–November 2018) (Islam *et al.*, 2020a), followed by pre-monsoon, i.e. 127 ± 80.4 μg/m^3 (February–March 2018) and monsoon season i.e, 121 ± 49 μg/m^3 (July–August 2020) (Islam and Saikia, 2020b). Poor air quality in this region is predominantly due to emissions from small and medium scale industries involved in tea processing, oil and brick kilns and coal-based (Islam *et al.*, 2020a; Islam and Saikia, 2020b; Saika *et al.*, 2018). During wintertime in northeast India, due to festive biomass burning episode, traditionally known as *meji* burning, copious amounts of particulate matter is released into the atmosphere, worsening air quality in this region (Deka and Hoque, 2014). Some other important sources of $PM_{2.5}$ include coal combustion, crustal emissions induced by traffic, wood burning and vehicular emissions (Khare and Baruah, 2010).

Over western India, Mumbai recorded high mass concentration of $PM_{2.5}$. A study by Masih *et al.* (2019) estimated the average $PM_{2.5}$ concentration in Mumbai to be 98.45 μg/m^3 during September–December 2016 followed by a drop in mass concentration to 60 μg/m^3 observed during January–December 2017 (Anand *et al.*, 2019). The direction of wind flow majorly influences the air quality of Mumbai, being a coastal city. When clean coastal winds flow towards the city, it causes dispersion of pollutants thus improving the air quality, however the reversal of winds along with high humid conditions favour accumulation of pollutants in the region (Anand *et al.*, 2019).

Pune, on the other hand, observed quite low $PM_{2.5}$ concentration as it is located at a relatively high altitude and experiences moderate weather conditions in both summer and winter seasons. Annual $PM_{2.5}$ mass concentration in Pune fluctuated from 46 ± 23 μg/m^3 (January–December 2015) (Krishna and Beig, 2018) to drop in concentration to 37.3 μg/m^3 (during April 2015–April 2016) (Gawhane *et al.*, 2017), followed by slight increased concentration to 40 ± 14 (January–December 2016) (Gawhane *et al.*, 2019) which finally peaked at 47 μg/m^3 (January–December 2017) (Anand *et al.*, 2019). Pune observed a boom in housing industry since past few years along with the increased vehicular emissions and industrial setups in Pune (Pipal and Satsangi, 2015) which are probable sources of PM. High altitude sites in western India like Mahabaleshwar and Sinhagad recorded quite low $PM_{2.5}$ concentrations well within the NAAQS, i.e. 26 ± 14 μg/m^3 (March 2015–February 2017) (Buchende *et al.*, 2019) and 27 ± 8 μg/m^3 (January–December 2017) (Budhavant *et al.*, 2020), respectively.

Sampling locations in southern India, viz. Vishakhapatnam (48.5 ± 27 μg/m^3 during January 2014–December 2019), Tirumala (30 ± 17 μg/m^3 during January 2014–December 2019), Amravati (40 ± 28 μg/m^3 during August 2017–December 2019) and Rajahmundry (37 ± 28 μg/m^3 during August 2017–December 2019) (Varaprasad *et al.*, 2021) recorded substantially low $PM_{2.5}$ concentrations. Low concentration in Tirumala (viz a high-altitude site) as compared to Vishakhapatnam, Amravati and Rajahmundry is due to the absence of power plants and sugar industries in its near vicinity. Also, to meet electricity and energy demands, wind and solar energy is relied upon in Tirumala (Varaprasad *et al.*, 2021).

2.2 Ambient PM_{10} Mass Concentrations in Various Indian Cities during the Years 2015–2019

Majority of the cities located in IHR reported average PM_{10} mass concentrations well within NAAQS for PM_{10} (60 μg/m^3) prescribed by the Central Pollution Control Board, India. The IHR sampling locations, i.e. Kashmir (32 ± 27.6 μg/m^3), Kullu (48.5 ± 16 μg/m^3), Darjeeling (48 ± 18 μg/m^3) and Nainital (42 ± 24 μg/m^3) reported low average mass concentrations of PM_{10} during January–March 2015 (Sen *et al.*, 2017) as compared to that of other sub-urban cities. However, in Dehradun, PM_{10} levels were nearly doubled in one year period, i.e. from 89 ± 36 μg/m^3 (November 2014–June 2015) (Soni *et al.*, 2020) to 170 ± 17.3 μg/m^3 (November 2015–May 2016) (Shridhar, 2019). This is attributed to rapid urbanisation causing increased anthropogenic emissions such as vehicular emissions, biomass burning, road dust resuspension influx of air masses from IGP (Soni *et al.*, 2020; Shridhar, 2019).

The various mega-cities located in IGP, recorded remarkably high levels of average PM_{10} mass concentration, violating the NAAQS value of 60 μg/m^3. Amongst all sampling locations in IGP, Varanasi recorded the highest PM_{10} average mass concentration (ranged 165 to 257 ± 90 μg/m^3) followed by Delhi (ranged 162 to 242 μg/m^3), Agra (ranged 206.5 ± 91 to 234 ± 84 μg/m^3) and Kolkata (ranged 151 to 179 ± 77 μg/m^3) during 2015–2019.

As for Varanasi, an uptrend in annual average mass concentration is observed from 238 ± 82 (January–March 2015) (Sen *et al.*, 2017) to 257 ± 90 μg/m^3 (January 2015–December 2016) (Jain *et al.*, 2021) followed by a

decrease in concentration to 165 μg/m^3 observed during summer sampling months, i.e. May–June 2017 (Devi *et al.*, 2020). Major sources of high PM_{10} levels in Varanasi region are emissions from traffic, construction and development activities, resuspension of road dust and long-range transport of PM from upper IGP (Jain *et al.*, 2021).

In Delhi, PM_{10} annual mass concentration increased from 178 ± 96 μg/m^3 (January–December 2015) (Krishna and Beig, 2018) to 242 ± 95 μg/m^3 (January 2015–December 2016) (Jain *et al.*, 2021) followed by a slight downtrend to 222 ± 87 μg/m^3 (January 2015–December 2017) (Sharma *et al.*, 2018a) and subsequently to 200 μg/m^3 (January 2017–December 2017) (Anand *et al.*, 2019). Wind-blown dust in Delhi predominantly contributes to elevated PM_{10} levels (Anand *et al.*, 2019). Other sources of PM in Delhi include vehicular emissions, biomass burning, soil/road dust, fossil fuel combustion and industrial emissions (Jain *et al.*, 2021).

In Agra, a uniform trend of PM_{10} mass concentration can be observed which is nearly three times higher than the annual average standards of PM_{10} (60 μg/m^3) (NAAQS, 2009). Predominant sources of PM_{10} include emissions from factories and vehicles, construction and development activities, road and soil dust.

In Kolkata, Jain *et al.* (2021) calculated average PM_{10} mass concentration to be 179 ± 77 μg/m^3 (January 2015–December 2016) and Devi *et al.* (2020) reported mass concentration of about 151 μg/m^3 (May–June 2017). Kolkata being a coastal city, experiences high loading of coarse mode marine aerosols in addition to local anthropogenic emissions, resulting in elevated PM_{10} mass concentration levels especially in the winter sampling months (Sen *et al.*, 2014).

In western India, the annual average PM_{10} mass concentration for Pune slightly decreased from 98 ± 45.5 μg/m^3 (January–December 2015) (Krishna and Beig, 2018) to 63 ± 25 μg/m^3 (January–December 2016) (Gawhane *et al.*, 2019) and then increased to 87 μg/m^3 (January–December 2017) (Anand *et al.*, 2019). Anthropogenic activities, sea-salt and dust aerosols are the major sources contributing to PM_{10} concentration (Gawhane *et al.*, 2019). Ajmer, a semi-arid site, observed low PM_{10} concentration (80 ± 44 μg/m^3 during January–March 2015, Sen *et al.*, 2017) as it is surrounded by Aravalli range resulting in lower transport of windblown dust aerosols from Thar desert in its vicinity. Ahmedabad, which is large city in western India, recorded average PM_{10} concentration as 126 μg/m^3 during January–December 2017 (Anand *et al.*, 2019). Even in summer, the PM concentration remained equally high i.e., 130 ± 31 μg/m^3 (during May–June 2017) due to sea salt and mineral aerosols, frequent dust storms and heat wave conditions due to proximity to Thar Desert and Arabian Sea (Patel *et al.*, 2021).

Over north-eastern India, sampling of PM_{10} size fraction was carried out in Jorhat (Islam *et al.*, 2020a; Islam and Saikia, 2020b) for which the mean concentration values were 261 ± 89 μg/m^3, 274 ± 79 μg/m^3, and 153 ± 45 μg/m^3 during February–March 2018, October–November 2018 and July–August 2019, respectively exhibiting clear seasonal trend. High particulate concentrations at these sites could be due to the influx of air masses loaded with particulate matter from within IGP region into eastern region (BoB) due to favourable wind pattern during winter (Sen *et al.*, 2017). High PM level in Jorhat during winter/post monsoon is due to large number of forest fires in its near vicinity as reported by Sen *et al.* (2016) on the basis of open fire-count satellite data, in addition to festive biomass burning episode (*meji* burning) during this time (Deka and Hoque, 2014).

In southern India, at an urban coastal industrial sampling site Vishakhapatnam, PM_{10} average mass concentration was observed to be 110 ± 48 μg/m^3 (January 2014–December 2019) (Varaprasad *et al.*, 2021). Important sources of PM pollution at this site are emissions from industries, vehicles and port activities. Tirumala which is high-altitude rural region also famous as a pilgrim site, reported moderate PM levels i.e., 60 ± 25 μg/m^3 (January 2014–December 2019) (Varaprasad *et al.*, 2021) whereby, emissions from transport sector is the major source of pollution. During August 2017–December 2019, Varaprasad *et al.* (2021) reported PM_{10} levels in Amravati (residential site, capital of Andhra Pradesh) and Rajahmundry (industrial site) to be 79 ± 45 μg/m^3 and 75 ± 42 μg/m^3, respectively. In Amravati, the major sources of PM pollution are emissions from transport sector and construction activities, while in Rajahmundry, emissions from paper industries and powerplants located in its vicinity are the dominant sources.

3. MERRA-2 DATA AND METHODS

3.1 Data Analysis

NASA's (National Aeronautics and Space Administration) MERRA-2 dataset (Modern-Era Retrospective Analysis for Research and Applications, Version 2) is a

global atmospheric reanalysis tool developed by GMAO (Global Modelling and Assimilation Office) which is an advanced version of MERRA-1, Goddard Earth Observing System (GEOS) model. Atmospheric reanalysis integrating satellite observations is a helpful tool to provide the most complete picture of global climate change and spatial environment due to continual temporal and spatial resolution, high data quality and wide aerosol species (Randles *et al.*, 2017). MERRA-1 was available from 1979 to February 2016, and after its discontinuation the subsequent dataset is present with MERRA-2 (runs a few weeks prior real-time). Basic aspects of both the reanalyses such as resolution and data products are the same as given in Gelaro *et al.* (2017). MERRA-2 dataset can be accessed online through NASA Goddard Earth Sciences Data Information Services Center (GES DISC). The optical depth observations assimilated at 550 nm in MERRA-2, are obtained by reflectance from MODIS (Moderate Resolution Imaging Spectroradiometer) on board Aqua and Terra satellites (Gelaro *et al.*, 2017). The GOCART (Goddard Chemistry Aerosol Radiation and Transport) model coupled with GEOS provides information about the life cycle of predominant aerosol species, incorporating sea-salt (SS), dust (DS), organic carbon (OC), black carbon (BC) and sulfate (SO_4). At 550 nm, AOD is obtained by summation product of extinction coefficient of each species derived from cloud datasets, optical properties and mass concentrations of aerosols in the form of a column-species-integrated optical quantity.

Aerosol assimilation in MERRA-2 is done at a resolution of 0.5° latitude by 0.625° longitude and 73 vertical levels (from the surface to 80 km) all over the globe (Randles *et al.*, 2017; Chin *et al.*, 2002), thereby assessing the global distribution of $PM_{2.5}$ at high spatio-temporal resolutions. It is a very important tool to study particulate concentration and composition, especially in regions where ground monitoring is scarce or unreliable, or areas which are inaccessible. Having said that, there are also certain limitations in estimation methodologies used for simulation of MERRA-2 $PM_{2.5}$ mass concentrations, which must be carefully studied in order to better understand their merits and weaknesses. The purpose of this study is to assess MERRA-2's simulation of fine PM by comparison with ground-based measurements conducted at 27 stations across the Indian region between 2015 and 2019. The evaluation is based on seasonal and spatial variation, as the air quality of a region is predominantly influenced by life-style, meteorological and geographical factors.

To obtain ground-based $PM_{2.5}$ mass concentrations, a literature survey of available previous studies during 2015–2019 in India was done as shown in Table 1. Since $PM_{2.5}$ sampling durations varied in different studies, only those ground stations/cities are selected for which at least one month of continuous data is available, for better comparison with satellite data. It is also important to note that the MERRA-2 $PM_{2.5}$ mass concentration values at specific latitude and longitude coordinates were selected, corresponding to the coordinates of ground monitoring stations, in order to get spatially accurate data. Further, it maybe noted that there are many published studies which have used the data obtained from monitoring stations managed by the government, (for example pollution board monitoring stations) and then compared with satellite data (Navinya *et al.*, 2020; Mahesh *et al.*, 2019). However, to the best of our knowledge no study has used ground $PM_{2.5}$ mass concentration other than that of government managed stations for comparison with MERRA-2.

3.2 Evaluation method: Reconstruction of $PM_{2.5}$ Mass Concentration

Monthly MERRA-2 simulation data of five $PM_{2.5}$ species: SO_4, OC, BC, $DS_{2.5}$ and $SS_{2.5}$ has been evaluated from 2015 to 2019 to estimate the total concentration of $PM_{2.5}$. Using these individual aerosol species concentrations, the total mass concentration of $PM_{2.5}$ can be evaluated applying mass reconstruction method as shown in the equation below (Chow *et al.*, 2015):

$$(\text{MERRA-2 } PM_{2.5}) = DU_{2.5} + BC + SS_{2.5} + (OC \times 1.6) + (SO_4 \times 1.375) \quad (1)$$

where, $DU_{2.5}$, BC, $SS_{2.5}$, OC and SO_4 stand for dust (size $\leq$ 2.5 μm), black carbon, sea salt (size $\leq$ 2.5 μm), organic carbon and sulfate concentration, respectively. The factor of 1.375 is used to estimate absolute sulfate concentration as it is present in the form of neutralised $(NH_4)_2SO_4$ in MERRA-2 data (Buchard *et al.*, 2016). Similarly, a factor of 1.6 is used to calculate particulate organic carbon to account for organic matter contributions of Indian region (Aggarwal and Kawamura, 2009). It is important to note that eq. 1 lacks the concentration of nitrate particles predominantly anthropogenic in nature (Delmas *et al.*, 1997), which may result to biases when compared with ground data (He *et al.*, 2019; Mah-

sh *et al.*, 2019).

In this study, MERRA-2's simulated $PM_{2.5}$ concentration calculated by the equation above, is compared to the ground-based $PM_{2.5}$ concentration measured at 27 locations on seasonal as well as regional basis for a time period of January 2015–December 2019. In order to ensure the data reliability of the simulated and observed (ground-based) $PM_{2.5}$ data, a spatial consistency algorithm is used which is as follows: since trace concentrations are usually lognormally distributed, the bias between log-simulated concentration and log observed concentration ($B_{log} = \log(C_S) - \log(C_O)$; where C_S: simulated concentration, C_O: observed concentration) is calculated at all study locations. Thereby, reliability interval which justifies 95% of the normal distribution is calculated as: $\overline{Blog} \pm 2\sigma_{Blog}$; excluding all data pairs outside this interval.

Various statistical parameters were applied on monthly mean $PM_{2.5}$ concentrations for quantifying MERRA-2's accuracy. These are correlation coefficient (R), mean bias ($\overline{B} = \overline{C_S} - \overline{C_O}$), the standard deviation of bias (SD-B) mean fraction ($\overline{F} = \overline{C_S} / \overline{C_O}$), where C_O and C_S are observed and simulated mass concentration, respectively. Also, log transformed statistics were computed: B_{log}, and R_{log}. To evaluate air quality models, Chang and Hanna (2004) proposed Factor of Two (FAC2) index proposing that the proportion of data which satisfies $0.5 \leq C_O/C_S \leq 2.0$, should be equal to or more than 0.5 to be considered a good model performance. A detailed statistical comparison of ground and MERRA-2 $PM_{2.5}$ concentrations is given in section 4.1.

4. RESULTS AND DISCUSSION

4.1 Seasonal and Spatial Comparison of $PM_{2.5}$ Mass Concentration between MERRA-2 and Ground-based Data

The aerosol distribution at surface and in the atmospheric column is heterogeneous and shows strong seasonal trend owing to meteorological conditions and seasonally varying emission source strengths over India (Sreekanth *et al.*, 2007). Therefore, to evaluate the level of agreement between ground and MERRA-2 data, statistical metrics are applied on seasonal mean $PM_{2.5}$ concentrations shown in Table 2 (Navinya *et al.*, 2020; Mahesh *et al.*, 2019). We observed that the correlation coefficients are low during monsoon (0.22), indicating that $PM_{2.5}$ diurnal variability is not well captured by MERRA-2 in this season possibly due to restricted assimilation of MERRA-2 in presence of cloud cover (He *et al.*, 2019; Buchard *et al.*, 2016), while it is moderate during post monsoon (0.37), summer (0.38), and better during winter (0.59). However, Willmott (1982) discouraged the use of R to evaluate model performance since it is sensitive to extreme values and does not directly compare simulated with observed data. Therefore, to directly relate simulated and ground-based $PM_{2.5}$ concentrations, FAC2 index is evaluated given that this index is not disproportionately sensitive to extreme values. Based on FAC2 values, MERRA-2 and ground data performed well in all seasons ($FAC2 \geq 0.5$) i.e., 1.60, 1.52, 1.40 and 1.84 for winter, summer, monsoon and post-monsoon, respectively, suggesting good model performance and comparability between the two datasets. The mean bias was less during monsoon (−10.75 μg/m^3) and summer (−24.33 μg/m^3) as compared to post-monsoon (−35 μg/m^3) and winter (−32.76 μg/m^3). Also, the average observed $PM_{2.5}$ concentration reported at ground locations was higher in post-monsoon (103 μg/m^3) and winter (86 μg/m^3) as compared to summer (73 μg/m^3) and monsoon (45 μg/m^3). These results suggest MERRA-2's inability to simulate $PM_{2.5}$ mass concentration during high PM mass loading episodes. This is further supported by an underestimation of 34% and 38% during post-monsoon and winter respectively. Similar study by Navinya *et al.* (2020) reported the average observed $PM_{2.5}$ concentration over 20 Indian cities from 2015–2018 to be 80 μg/m^3, while 35 μg/m^3 simulated by MERRA-2, indicating MERRA $PM_{2.5}$ underestimation by 34%. The explanation for disagreement between the two datasets is to be discussed in section 4.2.

Table 2. Performance statistics of MERRA-2 $PM_{2.5}$ concentrations with regard to Ground $PM_{2.5}$ for the ensemble of locations in India in different seasons. AOC stands for "average observed concentration".

Statistical metrics	Winter	Summer	Monsoon	Post-monsoon
AOC (μg/m^3)	86	73	45	103
R	0.59	0.37	0.22	0.37
R_{log}	0.72	0.55	0.57	0.21
FAC2	1.60	1.52	1.40	1.84
$\overline{B}$ (μg/m^3)	−32.76	−24.33	−10.75	−35
SD-B (μg/m^3)	39	22	31	50
$\overline{Blog}$	−0.15	−0.16	−0.05	−0.17

Fig. 2 shows the spatial pattern of MERRA-2 simulated

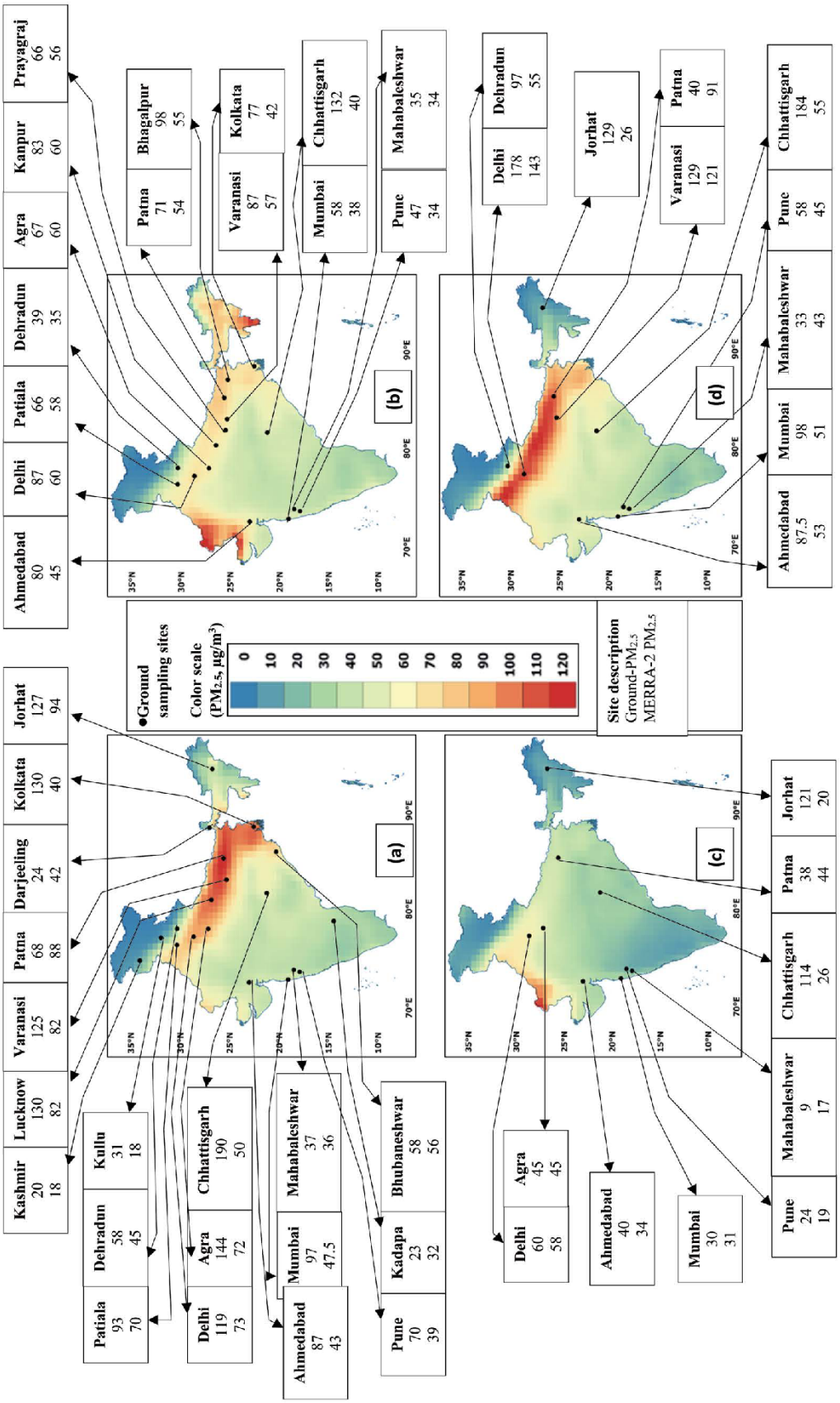

Fig. 2. Distribution of average $PM_{2.5}$ and location of measurement sites along with corresponding Ground $PM_{2.5}$ and MERRA-2 $PM_{2.5}$ concentration during the time period 2015–2019 in India, where (a) winter, (b) summer, (c) monsoon, and (d) post-monsoon.

$PM_{2.5}$ mass concentration throughout India for different seasons. The ground $PM_{2.5}$ mass concentration measured at 27 different sampling sites mentioned in this study are also depicted. A clear seasonal trend of $PM_{2.5}$ mass concentration is observed. Some key notable features are as follows: the air masses over IGP region are observed to have high dominance of $PM_{2.5}$ especially during the post-monsoon season (Fig. 2d) predominantly due to stubble burning over the western IGP which increases along the north eastern region during winter season (Fig. 2a), due to transport of aerosols from IGP (Sarkar *et al.*, 2018). Also, throughout the study period, relatively higher PM mass concentration is observed over the IGP region compared to other parts of the country indicated by both MERRA-2 as well as ground data. Rapid industrialisation, urbanisation and elevated prominent PM sources such as biomass burning, waste burning, vehicular emissions are some of the reasons due to which IGP has been regarded as one of the most polluted regions in India (Jain *et al.*, 2021; Ojha *et al.*, 2020). In contrast, less PM mass concentration is observed in IHR and south India due to pristine conditions and vegetation cover respectively (as explained in previous sections), which is indicated by both MERRA-2 and ground $PM_{2.5}$ concentrations. Therefore, we can say that $PM_{2.5}$ spatial trends observed from MERRA-2 data in different seasons reasonably conform to that of ground- based measurements.

However, it may be noted that MERRA-2 does not completely capture the size-resolved dust concentration in the IGP (Fig. 2b), since IGP is characterised by high dust loading during summer season (Goel *et al.*, 2015). The underestimation of fine dust particle concentration has been reported in previous study by Kramer *et al.* (2018). Over central and eastern regions, coal-based power and steel plants in addition to small and medium scale industries as well as mining activities significantly contribute to PM mass loading (Guttikunda *et al.*, 2019). It is important to highlight that the sampling site (located near the industrial site) at Chhattisgarh showed huge deviations (~3 times) between MERRA-2 and ground $PM_{2.5}$ concentrations throughout the study duration. This deviation could possibly be due to the inability of MERRA-2 to simulate nitrate concentrations from industries located in the vicinity of sampling location. Similar deviations between MERRA-2 and ground $PM_{2.5}$ (~4 times) was observed in Jorhat, where industrial emissions are the major factors for $PM_{2.5}$ emissions.

In terms of spatial distribution, when comparing the observed concentration (from ground data) and simulated concentration (from MERRA-2) an apparent deviation is evident in mass concentration of $PM_{2.5}$. We observed high mean bias between ground and simulated data over IGP (−28 μg/m^3) as compared to IHR (−7 μg/m^3) and southern region (−3 μg/m^3), and the corresponding underestimation by MERRA-2 to be 29%, 15% and 8%, respectively. Hence, it can be clearly observed that MERRA-2 underestimates mass concentration at regions showing very high pollution levels.

4.2 Factors Resulting in $PM_{2.5}$ Underestimation by MERRA-2

Fig. 3 shows the scatter between the MERRA-2 and ground-based data for monthly $PM_{2.5}$ mass concentrations. The solid line in the figure represents equiline (1 : 1 line) and $PM_{2.5}$ values are scattered below and above

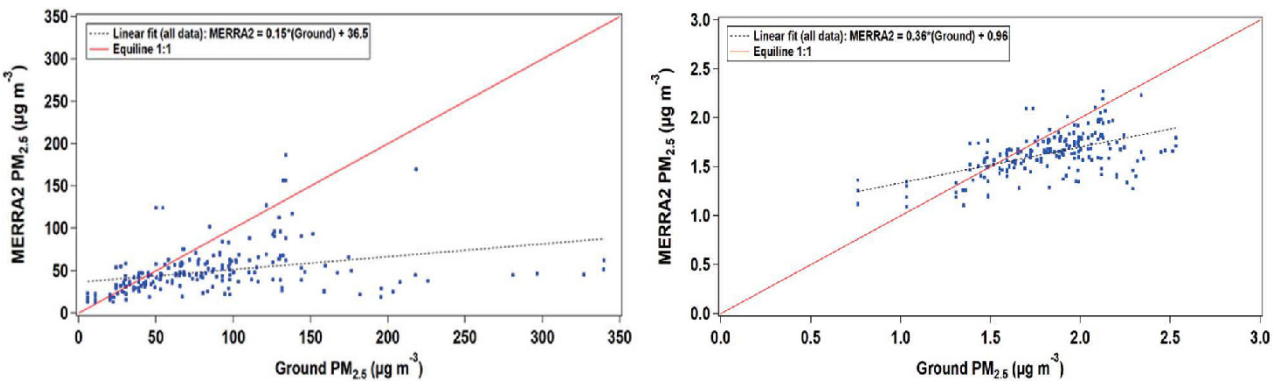

Fig. 3. Scatter plot for monthly mean (a) ground-based and MERRA-2 simulated $PM_{2.5}$ mass concentration and (b) log transformed ground-based and MERRA-2 simulated $PM_{2.5}$ mass concentration for the ensemble of locations in India.

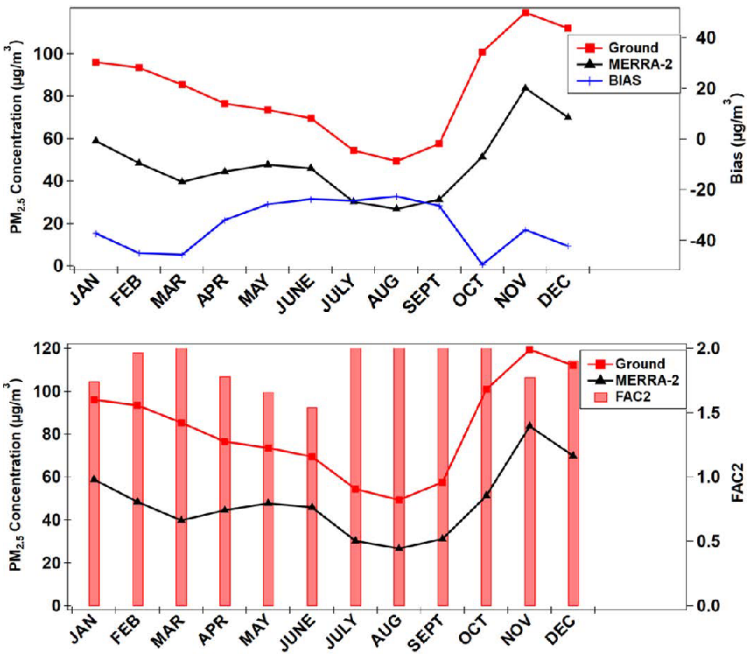

Fig. 4. Monthly average $PM_{2.5}$ mass concentration by ground-based and MERRA-2 simulation, mean bias and FAC2 values, for the ensemble of locations in India.

the equiline, indicating both under and overestimations by MERRA-2. The scatter becomes wider for higher ground-based concentrations. The plot reveals that for significantly high (low) loading conditions, MERRA-2 underestimates (overestimates) the mass concentrations. Similar analysis is supported by log-transformed data. To better understand agreement between ground-based and MERRA-2 simulated $PM_{2.5}$ concentrations, we compared monthly averaged observed and simulated data from 2015 to 2019 (Fig. 4). Similar trends in PM concentrations by both ground and MERRA-2 data is observed in all months. Moreover, the bias values indicate better model performance in summer and monsoon months (i.e., March to April) as compared to winter and post-monsoon (October to February), possibly attributing to the absence of nitrate in MERRA-2 $PM_{2.5}$ reanalysis which is more significant in India given that $PM_{2.5}$ is mostly composed of anthropogenic particles (He *et al.*, 2019; Mahesh *et al.*, 2019). Another reason for high deviations in winter and post-monsoon could be due to MERRA-2's inability to unresolve local sources of pollution since its resolution is too coarse to evaluate the urban core of cities (Provencal *et al.*, 2016).

In addition, MERRA-2 does not completely capture the size resolved dust concentration resulting in underestimation of fine dust particle concentration of size less than 2 μm (Kramer *et al.*, 2018). Further, there are missing AOD values in MERRA-2 reanalysis dataset due to cloud cover, orbital gaps, etc. (Mahesh *et al.*, 2019; Buchard *et al.*, 2016). Therefore, these datasets require extensive regional scale validations for complete data interpretation, especially over the highly diverse Indian region.

5. VARIATIONS AND LIMITATIONS IN MEASUREMENT OF GROUND-BASED PM MASS CONCENTRATION

Since PM monitoring plays an important role in improving air quality and health, therefore the accurate and precise measurement of PM mass concentration is of utmost concern. To monitor PM concentration across India, Central Pollution Control Board (CPCB) has notified three methods namely, Gravimetric Method (GMM), Tapered Element Oscillating Microbalance (TEOM) and Beta Attenuation Monitor (BAM). Gravimetric Method is the primary method to quantify PM mass concentration which is based on the difference in mass of the filter used before and after sampling, while

BAM and TEOM are the online/real-time measurement methods where the mass of PM collected on filter substrate is determined using beta attenuation, and change in frequency of oscillating element in TEOM, respectively. From the available ground-based data given in Table 1, we conclude that of all methods, Gravimetric Method is widely used in India since it is comparatively simpler, cost-effective and involves the primary measurement of PM mass.

For quality assured measurements, the periodic calibration of the instruments deployed for mass measurement is very important. As per Bureau of Indian Standards (BIS-IS 5182 (Part 24): 2019), the important parameters required during Gravimetric measurement include calibration of (1) impactor (2) flow rate of sampler, (3) micro balance, (4) temperature sensor, and (5) pressure sensor. However, there are many studies which have not mentioned the calibration and schedule of instruments, which is a significant measure to ensure QA/QC procedure (Murari *et al.*, 2020; Anand *et al.*, 2019; Yadav *et al.*, 2019; Gawhane *et al.*, 2019, 2017; Begam *et al.*, 2017; Bharti *et al.*, 2017; Leena *et al.*, 2017; Das *et al.*, 2015; Yadav *et al.*, 2015; Yadav and Satsangi, 2013).

In addition, some studies show inconsistency in mass concentration measurement even for the similar measurement technique used at same location for similar sampling duration. For example, Sen *et al.* (2014) reported $PM_{2.5}$ concentration in Varanasi to be 188 ± 36.5 μg/m^3 using IPM-FDS, Instrumex (flow rate of 1 m^3 h^{-1}, designed as per USEPA guidelines) during 20th January–5th February 2014, whereas another study by Kumar *et al.* (2015), reported $PM_{2.5}$ mass concentration as 138 ± 47.12 μg/m^3 in Varanasi during 1st January–31st March 2014 using the same instrument. Therefore, about 30% variation in PM mass concentration is observed despite similar conditions of measurement. A part of deviation in results could be attributed to the different local (site-specific) sources and measurement period (which was not exactly the same), but the inconsistency is primarily due to lack of standardisation of measurement method (Malik and Aggarwal, 2021). The calibration of instruments and the measurement methods deployed are very important, however in several studies most often this issue is not given adequate attention. The available literature shows that many studies lack the calibration details of instruments used, which is crucial for QA/QC purpose, i.e., Ezhilkumar *et al.*, 2021; Budhavant *et al.*, 2020; Islam *et al.*, 2020a; Islam and Saikia, 2020b; Begam *et al.*, 2017; Das *et al.*, 2015 etc.

Another issue of significant importance is variation in PM mass loading introduced due to different techniques. For comparison, $PM_{2.5}$ mass measured by MiniVol sampler during December 2013–January 2014 in Delhi was observed to be 277 ± 100 μg/m^3 (Pant *et al.*, 2015), while in another study PM mass measured using APM550 (high-volume sampler) in Delhi during January–February 2014 (Sen *et al.*, 2014) was reported to be 178 ± 56.5 μg/m^3, i.e. about 43% variation in mass measurement. Here both the techniques used are gravimetric mass measurement, however calibration of impactor and cyclone are the major issues to be addressed. Thus, the performance evaluation and limitations of different measurement techniques should be well understood as per country's ambient conditions before deploying an instrument which would be beneficial to effectively monitor air quality.

6. CONCLUSION AND SUGGESTIONS FOR BETTER AND ACCURATE GROUND-BASED PM MEASUREMENT TECHNIQUES, PROTOCOLS AND POLICIES

For better air quality management (AQM), extensive technical and scientific efforts of government regulatory bodies along with research institutions/local universities is crucial to generate reliable air monitoring data. In order to ensure data reliability following protocols should be followed; (1) periodic calibration of flow rate (2) comparison/intermediate check of data obtained with reference instrument (3) calibration certificate provided by the manufacturer (Aggarwal *et al.*, 2013). Some other important suggestions for improved air quality management are as follows:

1. For measurement of pollutants at air monitoring stations, the guidelines recommended by CPCB are followed. However, there are certain limitations resulting in incompliance in the target frequency monitoring (104 observations annually) due to (1) non-availability of continuous power supply (2) limited and non-uniform distribution of monitoring stations with almost no monitoring station in rural areas (3) many locations have manual stations which require two to three days to show measurement results (4) absence of calibration and laboratory inter-comparison details resulting in low credibility of data quality, henceforth the monitor-

ing data obtained is not absolute rather just indicative (CPCB, 2013). The regulatory bodies must ensure the following factors in order to ensure QA/QC procedure: (1) regular calibration of equipments such as blower, rotameter, microbalance, spectrophotometer etc must be carried out at regular intervals (2) continuous supply of electricity should be ensured for uninterrupted monitoring of pollutants (3) more uniform air monitoring network should be developed to obtain comparable data from various stations (CPCB, 2003).

2. Available literature shows that greater number of studies are carried out in larger cities when compared to small cities and rural areas. This issue should be given more attention. The air monitoring network should be reliable, accurate and uniformly available so that data from different stations is comparable to better understand PM concentration levels and trends in India.
3. Variations in PM concentration are observed for studies conducted in the same city. A part of this deviation could be due to different sampling locations and meteorological conditions, but to large extent variations are observed due to lack of standard operating procedures and limited availability of trained manpower. Therefore, all laboratories and stations involved in air quality monitoring should be accredited to ensure QA/QC procedures. Periodic workshops and capacity building training of all manpower, involved directly or indirectly in generating and reporting data, whether in fields and laboratories, should be arranged to familiarise them with appropriate sampling and analysis methods.

This review summarises ambient $PM_{2.5}$ and PM_{10} mass concentration across different sampling locations in India during 2015–2019, and also compare $PM_{2.5}$ mass concentration with MERRA-2 reanalysis data. The objective was to map this data with temporal and spatial distribution to better understand PM pollution and thereby help policy makers to formulate better control policies. Some important conclusions drawn from the study are:

1. Due to pristine conditions of IHR, low PM mass concentration is observed when compared to other locations however due to urbanisation and long-range transport of pollutants from IGP, slight increase in the average mass concentration of PM is reported in many locations in last few years. Amongst the IHR sites, lowest PM_{10} and $PM_{2.5}$ concentration were recorded in Kashmir in contrast to Dehradun which observed high PM mass concentration values showing uptrend/increasing concentrations in subsequent years, while Kullu recorded a slight drop in PM_{10} concentration from 2014 to 2015.
2. Throughout the study period, ground sampling locations in IGP like Delhi, Agra, Varanasi, Kolkata reported the highest fine and coarse PM mass loading, due to rapid urbanisation, industrialisation and increased anthropogenic emissions from biomass/waste burning activities. The combined effect of local sources and stable atmospheric conditions further deteriorated the air quality during winters in these regions. Especially in Delhi, air quality is worsened due its landlocked geographical location.
3. Most of the ground studies have been conducted in larger cities when compared to small cities and rural areas. This needs to be improved for better understanding of PM concentration levels and trends in order to comprehend the overall state of PM pollution in India.
4. Variations in ground PM concentration are observed for the studies conducted in the same city. This may be due to different sampling locations and meteorological conditions, but to large extent variations are observed due to lack of standard operating methods.
5. According to National Ambient Air Quality Standards, monitoring should be done for 24 h and at least 104 days in a year. However, the literature clearly indicates that most of the data are for limited days.
6. There are associated uncertainties in data from ground monitoring networks due to lack of quality control procedures. In the absence of QA/QC, data quality remains questionable.
7. MERRA-2 $PM_{2.5}$ exhibited seasonal variation with high mass loading in winter and low in monsoon. However, MERRA-2 mostly underestimated $PM_{2.5}$ mass concentration relative to corresponding ground measurement, which was even higher during pollution episodes indicating inability of MERRA-2's data assimilation when predicting higher mass concentration.

Effective air quality management has become one of the most important tools to be ensured in order to combat and control the negative impacts of atmospheric pollution on human health and environment. Hence, an accurate, continuous and precise air monitoring network is crucial for formulating effective abatement and control policies. The present study highlights better measurement techniques, protocols and policies to maintain

quality and reliability in the air monitoring data. The development of RM would prove to be beneficial for monitoring bodies working in this domain. Regular quality and calibration checks are mandatory to ensure quality of data generated. All resources and efforts need to be prioritized in improving air quality network and encouraging scientific community towards advancement of better techniques and standards.

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