

# Sensitivity of photochemical surface ozone formation regimes to emissions and meteorology in India

Gopalakrishna Pillai Gopikrishnan<sup>1,2</sup>, Daniel M. Westervelt<sup>2</sup> and Jayanarayanan Kuttippurath<sup>1</sup>

<sup>1</sup>CORAL, Indian Institute of Technology Kharagpur, Kharagpur–721302, India.

<sup>2</sup>Lamont-Doherty Earth Observatory, Columbia University, New York - 10964, NY, USA

Correspondence to:

Daniel M. Westervelt ([danielmw@ldeo.columbia.edu](mailto:danielmw@ldeo.columbia.edu))

Jayanarayanan Kuttippurath ([jayan@coral.iitkgp.ac.in](mailto:jayan@coral.iitkgp.ac.in))

**Abstract.** Atmospheric aerosols significantly contribute to air pollution and influence atmospheric chemistry, impacting air quality and public health. Decrease in aerosols can hinder the radical uptake sink of HO<sub>2</sub>, and thus increase NO<sub>x</sub> and OH, and subsequently increase ozone levels. This study investigates the seasonal variations of PM<sub>10</sub> and aerosol surface area and their effect on surface ozone levels in India, using the GEOS-Chem Chemical Transport Model for the years 2018 and 2022, two years with high and low simulated PM<sub>10</sub> concentrations, respectively. The results reveal substantial seasonal variations in PM<sub>10</sub> and aerosol surface area. In winter (DJF), higher PM<sub>10</sub> and aerosol surface area in the Indo-Gangetic Plain (IGP) and western Central India (CI) result from biomass burning and industrial activity, while coastal regions show lower aerosol surface area. A decrease in aerosol surface area is seen during the pre-monsoon (March-April-May; MAM) and monsoon (June-July-August-September; JJAS), followed by an increase in the post-monsoon (ON) season. As a result, aerosol-induced HO<sub>2</sub> uptake during winter and post-monsoon suppress surface ozone by approximately 5–10 μg/m<sup>3</sup> in 2022 when compared to that of 2018. In contrast, during monsoon in 2022, the decrease in aerosol surface area caused an ozone increase of 5–7.5 μg/m<sup>3</sup> when compared to that of 2018. On average, this increase in surface ozone due to the decrease in aerosols can be mitigated by reducing anthropogenic NO<sub>x</sub> emissions by about 25–50%. Thus, we recommend integrated strategies addressing aerosols, precursor emissions and regional meteorology to combat ozone pollution.

## 1. Introduction

In recent decades, surface ozone (O<sub>3</sub>) has gained significant research attention due to its role as a transient (ranging from a few hours to a few weeks) secondary pollutant and its detrimental impacts on human health and agricultural yield (e.g., Mills et al. 2007; Avnery et al. 2011; Rathore et al., 2023; Gopikrishnan and Kuttippurath, 2024). It is an important trace gas in tropospheric chemistry, facilitating oxidation processes as the principal source of hydroxyl radicals (OH) (Logan, 1985) while also involved in chemical reactions with various organic molecules (Anderson, 2007). Ozone in the troposphere is a secondary pollutant generated via photochemical reactions that involve carbon monoxide (CO) and volatile organic compounds (VOCs) together with nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>), recognised as ozone precursors (Crutzen 1995; Gopikrishnan et al., 2022). However, these processes are dependent on solar radiation, with the reaction rate often peaking during summer months.

36 The formation dynamics of surface ozone is greatly affected by the presence of precursors such as NO<sub>x</sub> and VOCs,  
37 resulting in NO<sub>x</sub>-limited and VOC-limited regimes (Fiore et al., 2002; Lu et al., 2019). In the NO<sub>x</sub>-limited regime,  
38 the concentration of NO<sub>x</sub> is low compared to VOCs, implying that increasing NO<sub>x</sub> levels can promote further  
39 ozone generation due to inadequate NO<sub>x</sub> to completely react with VOCs. In a VOC-limited regime, the availability  
40 of VOCs is restricted, and further VOC emissions can significantly enhance ozone formation, whereas additional  
41 NO<sub>x</sub> may have less effect on ozone formation. Nevertheless, these regimes are significantly affected by local  
42 emission sources, atmospheric chemistry and climatic conditions. For instance, aerosols can influence these  
43 regimes by modifying the concentration of hydroxyl (OH) and hydroperoxyl (HO<sub>2</sub>) radicals, affecting the  
44 equilibrium between NO<sub>x</sub> and VOCs and eventually determining the rate and extent of ozone generation in  
45 different locations (Jacob et al., 2005; Feng et al., 2016; Wang et al., 2018; Li et al., 2019; Ivatt et al., 2022;  
46 Gopikrishnan et al., 2025).

47 Previous studies based on ground-based measurements from urban centers and industrial sites have shown  
48 substantial decrease in the particulate matter in the Indian cities post the implementation of National Clean Air  
49 Programme (NCAP), which primarily targets reducing the city-level PM pollution by 20–30% in 131 cities when  
50 compared to that of 2017–18. Gopikrishnan and Kuttippurath (2024) studied the Daily maximum 8-hour average  
51 (MDA-8) ozone changes and observed that cities such as Visakhapatnam and Tirupati reported zero days of MDA-  
52 8 ozone surpassing 100 ppb post the implementation of NCAP in 2022 when compared to that of 2018, the base  
53 year, during which Visakhapatnam and Tirupati experienced about 60 and 10 such days, respectively.  
54 Nevertheless, some cities in the Indo-Gangetic Plains (IGP) of India, for instance, Agra, Singrauli, Ghaziabad,  
55 with decreasing aerosol loading and particulate matter pollution (Gopikrishnan and Kuttippurath, 2025), show an  
56 increase in the surface ozone levels. This trade-off between particulate matter (PM) and surface ozone was also  
57 mentioned in other highly populated regions of the world. For example, Wang et al. (2020) also observed that the  
58 opposite changes in surface ozone and PM<sub>2.5</sub> emerged as an unforeseen consequence of China's Clean Air Action  
59 Plan, aimed at reducing air pollution. Following the implementation of the plan, ozone levels increased over the  
60 summer in the North China Plain, due to reduced NO<sub>x</sub> emissions and steady or rising VOC emissions, alongside  
61 significant decreases in PM<sub>2.5</sub> concentrations. This indicates that addressing ozone pollution requires deeper  
62 knowledge than broadly categorising it as NO<sub>x</sub>-limited or VOC-limited, as the influence of PM and aerosols is  
63 significant (Zhao et al., 2023). Nevertheless, this variability in PM can also arise substantially due to changes in  
64 meteorology (Ivatt et al., 2022). Therefore, future air quality management systems worldwide must include the  
65 intricate relationship between ozone, particulate matter, precursor emissions and the regionally prevailing  
66 meteorology to effectively reduce both pollutants simultaneously.

67 Rural pollution is also a critical, yet often overlooked aspect of air quality management, especially in a country  
68 like India, where a significant portion of the population resides in rural areas (Pathak and Kuttippurath, 2022;  
69 2024). Rural regions are not immune to air pollution; they face unique challenges such as solid fuel combustion  
70 for cooking, agricultural residue burning and increasing vehicular emissions due to expanding road networks  
71 (Bhuvaneshwari et al., 2019; Chanana et al., 2023). These activities contribute to elevated levels of PM and ozone  
72 precursors, which can drift into urban areas, exacerbating pollution in cities through regional transport  
73 mechanisms. Furthermore, rural air pollution has profound implications for public health, as rural populations  
74 often lack access to healthcare infrastructure to address respiratory and cardiovascular diseases caused by poor air

75 quality (Coker and Kizito, 2018; Manisalidis et al., 2020). It also impacts agricultural productivity, with rising  
76 ozone levels reducing crop yields, thereby threatening food security in agrarian economies like India (Pandya et  
77 al., 2022; Anagha et al., 2023). Addressing rural pollution is essential for achieving holistic improvements in air  
78 quality and ensuring equitable environmental health benefits across urban and rural populations.

79 Studies on the impact of aerosols on near-surface ozone formation have predominantly focused on the significance  
80 of Aerosol Optical Depth (AOD) extinction in photochemical processes that generate ozone (Bian et al., 2007;  
81 Kim et al., 2013; Wang et al., 2019). AOD quantifies the degree to which aerosols scatter and absorb sunlight,  
82 hence influencing the quantity of solar radiation that reaches the Earth's surface. This subsequently affects the  
83 photochemical mechanisms that facilitate ozone synthesis in the lower atmosphere. Nevertheless, in areas with  
84 high aerosol concentrations, the reduction of surface ozone is not only governed by AOD extinction, but also the  
85 aerosol surface area by offering reactive sites for chemical reactions with molecules such as HO<sub>2</sub> (Macintyre and  
86 Evans, 2011; Song et al., 2021). The interaction between aerosols and these species leads to the elimination of  
87 ozone precursors, including OH radicals, which are vital for ozone production.

88 In this study, we aim to evaluate the impact of PM<sub>10</sub> and aerosol surface area on surface ozone pollution in India.  
89 Since several studies are available on the air pollution concentrated in the urban regions of India, our analysis  
90 therefore explores the ozone formation regimes across entire India, using a high-resolution GEOS-Chem Chemical  
91 Transport Model. This allows us to examine the impact of regional and larger scale dynamics of ozone production,  
92 which are often influenced by agricultural practices, biomass burning and long-range pollutant transport. By  
93 simulating two distinct years—2018 (a high PM<sub>10</sub> year and low rainfall year) and 2022 (a low PM<sub>10</sub> year and  
94 normal rainfall year) [with and without fixed meteorology for the two years](#)—and incorporating/excluding the  
95 reactive absorption of HO<sub>2</sub> onto aerosols (via the  $\gamma$ HO<sub>2</sub> parameter), we aim to understand how reductions in PM<sub>10</sub>  
96 levels influence aerosol surface area and the secondary ozone chemistry (**Figures S1 and S2**). Furthermore, we  
97 categorize ozone generation regimes into NO<sub>x</sub>-limited, VOC-limited and Aerosol Inhibited Regime (AIR) regions  
98 based on the dominant termination reactions in surface ozone formation for each model grid ([Ivatt et al., 2022](#)).  
99 Additionally, by scaling down NO<sub>x</sub> emissions by 25 and 50%, we assess the efficacy of national and local emission  
100 control measures in mitigating NO<sub>x</sub> emissions and their subsequent impact on surface ozone levels. This approach  
101 provides important insights into the chemistry between PM, precursor emissions and surface ozone across diverse  
102 geographical regions in India, highlighting the need for integrated air quality management strategies beyond urban  
103 boundaries.

## 104 **2. Data and Methods**

### 105 **2.1 Region of Study**

106 India can be delineated into six regions based on topographical characteristics, demographic factors, and pollution  
107 levels: Peninsular India (PI), Northwest (NW), Northeast (NE), Central India (CI), Indo-Gangetic Plain (IGP),  
108 and Hilly Regions (HR), as shown in **Figure 1**. Peninsular India, which encompasses Kerala, Tamil Nadu,  
109 Karnataka, Goa, Andhra Pradesh, and Telangana, has three of the ten most populous states—Andhra Pradesh,  
110 Tamil Nadu, and Karnataka—and features substantial forest cover in the Western Ghats. Sources of pollution in  
111 this region are automobile emissions, industrial operations and biomass combustion, especially in urban areas like

112 Bengaluru and Chennai. Central India, comprising Maharashtra, Madhya Pradesh, Chhattisgarh, Jharkhand and  
113 Odisha, is marked by low population density yet is linked to thermal power plants, coal mines and steel factories.  
114 Cities like Mumbai and Pune have higher pollution levels attributable to industrialisation and trash incineration.  
115 Northwest India includes Rajasthan and Gujarat, characterised by dry landscapes like the Thar Desert; pollution  
116 is attributed to dust from unpaved roads and construction activity, along with industrial emissions from sectors  
117 such as textiles. Northeast India comprises Tripura, Mizoram, Manipur, Nagaland, Meghalaya and Assam; despite  
118 its low population density and extensive vegetation, this region is affected by automobile pollution and slash-and-  
119 burn agriculture methods. The Indo-Gangetic Plain encompasses West Bengal, Bihar, Uttar Pradesh, Haryana and  
120 Punjab; it is characterised by a high population density exceeding 1000 individuals per square kilometre and has  
121 substantial pollution resulting from heavy traffic, industrial effluents and extensive crop residue burning during  
122 harvest periods. The Hilly Regions encompass Jammu and Kashmir, Himachal Pradesh, Uttarakhand, Sikkim and  
123 Arunachal Pradesh. These areas are less industrialised and ecologically sensitive, with economies dependent on  
124 agriculture and forestry. They experience localised pollution from vehicular emissions and biomass combustion,  
125 which is intensified by winter temperature inversions that confine pollutants near the surface (Kuttippurath et al.,  
126 2020; Gopikrishnan et al., 2022).

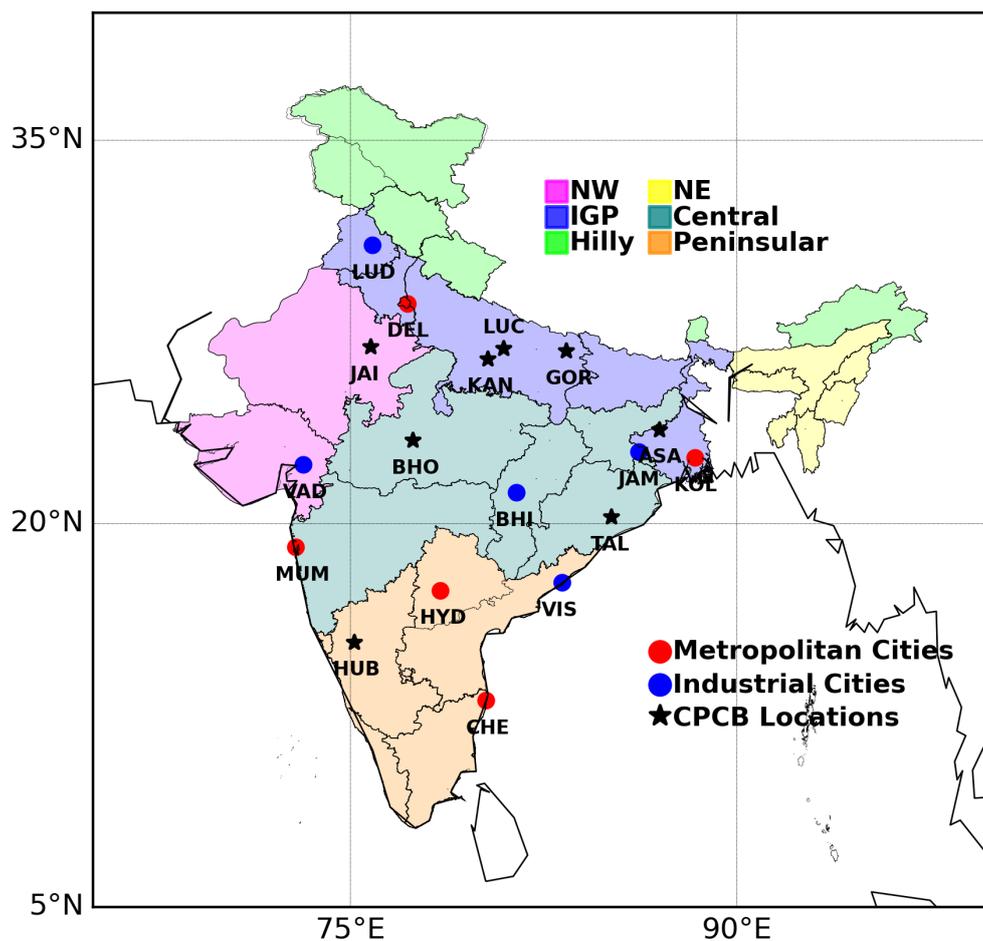
## 127 2.2 GEOS-Chem

128 GEOS-Chem is an extensive model developed for modelling complex oxidant-aerosol chemistry in the  
129 troposphere and stratosphere (Zhang et al., 2011; Kim et al., 2015). [We have used GEOS-Chem version 14.4.3 in  
130 this study \(https://doi.org/10.5281/zenodo.13314490\)](https://doi.org/10.5281/zenodo.13314490). The model complies with the most recent JPL/IUPAC  
131 guidelines for chemical mechanisms, with substantial modifications that improve the depiction of diverse  
132 chemical processes, including those related to isoprene, aromatics and nitrates (Bates et al., 2024). Recent  
133 advancements have enhanced the treatment of complex chemical reactions, including methanol synthesis and  
134 mercury redox chemistry, facilitating more precise atmospheric forecasts (Horowitz et al., 2017; Guil-López et  
135 al., 2019). Additionally, the model incorporates the reactive absorption of NO<sub>x</sub> by aerosols and computes aerosol  
136 hygroscopicity, which is essential for comprehending aerosol-cloud interactions. GEOS-Chem offers a  
137 comprehensive framework for investigating atmospheric chemistry and its effects on air quality and climate  
138 change, accommodating various chemical species and reactions, including halogens and  
139 hydroxymethanesulfonate. This model provides thorough understanding into ozone generation mechanisms and  
140 pollutant interactions, serving as an essential resource for addressing air quality challenges and comprehending  
141 the wider implications of atmospheric dynamics.

142 GEOS-Chem has been rigorously validated by researchers globally. For instance, Travis and Jacob, (2016)  
143 showed that, despite the successful simulation of ozone and its precursors in the SEAC4RS aircraft data below  
144 1 km of altitude, MDA8 surface ozone was biased high in the model by +6 µg/m<sup>3</sup> on average. David et al. (2019)  
145 used the GEOS-Chem transport model in India and observed that the model reasonably simulated the tropospheric  
146 O<sub>3</sub> abundances and vertical profiles, with a mean bias of 1–3 DU compared to observations for the period 2000–  
147 2015. Christiansen et al. (2022) found that the GEOS-Chem model, when validated against ozonesonde, aircraft,  
148 and satellite observations globally, showed reasonable agreement with a mean bias of 1–3 DU for the period from  
149 1990 to 2017. Mao et al. (2024) reported that the GEOS-Chem model, validated against MDA8 O<sub>3</sub> concentration

150 observations in central and eastern China from May to July 2017, showed a strong correlation of 0.77 (95%  
151 confidence level). In Nanjing, the simulated MDA8 O<sub>3</sub> concentrations converged with the observed trend, with a  
152 correlation coefficient of 0.65, a normalised mean bias (NMB) of 5%, and a normalised mean error (NME) of  
153 21%. Karambelas et al. (2022) used the GEOS-Chem model to simulate PM<sub>2.5</sub> concentrations during high PM  
154 episodes in the fall of 2015 and 2017, finding that the model underestimated observed concentrations by 28%,  
155 with an average observed PM<sub>2.5</sub> concentration of 142 µg/m<sup>3</sup> compared to 125 µg/m<sup>3</sup> from the model. [Pai et al.  
156 \(2022\) validated the GEOS-Chem simulation against speciated airborne aerosol measurements over India and  
157 reported significantly reduced normalized mean biases: NMBModified = 0.19 compared to NMBBase = 0.61  
158 overall—with nitrate improving from 1.64 to 0.08 and ammonium from 0.90 to 0.49.](#) Despite this underestimation,  
159 the model demonstrated strong spatial correlations with observed concentrations ( $r^2 = 0.67$ ), accurately capturing  
160 the spatial distribution of PM<sub>2.5</sub> across India. David and Ravishankara (2019) used the GEOS-Chem model to  
161 estimate boundary layer ozone (BLO<sub>3</sub>) contributions across eight regions of the Indian subcontinent, finding that  
162 BLO<sub>3</sub> in northern India, Pakistan, and Sri Lanka is largely (about 65–70%) influenced by regions outside the  
163 subcontinent. They also highlighted the growing importance of central India in contributing to ozone pollution,  
164 with regional meteorology playing a key role in the redistribution of BLO<sub>3</sub> and its precursors including Chlorine  
165 (Cl).

166 In this study, GEOS-Chem is simulated in a 2.0°x2.5° latitude by longitude grid resolution globally for the  
167 boundary conditions for the years 2018 and 2022. One year spin up simulations are used to generate the model  
168 restart files and the boundary conditions. The nested model has a grid resolution of 0.25°x0.3125° latitude by  
169 longitude and a spin up period of 1 month. We use the data from the lowest level of the ETA coordinate system  
170 from GEOS-Chem model simulations to analyse the correlation between modelled and observed PM<sub>10</sub> and ozone  
171 data. [The model is driven by the GEOS-FP assimilated meteorological data provided by the Goddard Earth  
172 Observing System \(GEOS\) of the Earth Modelling and Assimilation Office \(GMAO\) and with a temporal  
173 resolution of 1 hour \(Lucchesi, 2013\). Emissions in the GEOS-Chem model are handled using the Harvard-NASA  
174 Emission Component \(HEMCO; Keller et al., 2014\). Anthropogenic emissions are derived from the Community  
175 Emissions Data System \(CEDS\), which provides global coverage at a native resolution of 0.1° × 0.1°, with data  
176 aggregated to the model resolution \(Hoesly et al., 2018\). Sea salt emissions are calculated offline following the  
177 approach of Zhu et al. \(2019\), while dust emissions are calculated offline following the method of Ginoux et al.  
178 \(2010\). Emissions of biogenic origin are simulated using the Model of Emissions of Gases and Aerosols from  
179 Nature \(MEGAN\), which provides global emissions at 0.01° latitude × 0.01° longitude resolution \(Guenther et  
180 al., 2012\) and Soil NO<sub>x</sub> emissions follow the approach outlined by Hudman et al. \(2012\). Biomass burning  
181 emissions are sourced from the Global Fire Emissions Database version 4 \(GFED4\), which provides a global  
182 coverage at 0.25° latitude × 0.25° longitude \(Giglio et al., 2013\).](#)



183

184 **Figure 1: The region of study. The regions considered are also shown in the respective colours. Here, NW is Northwest,**  
 185 **NE is Northeast India and IGP is Indo Gangetic Plain. The star marks indicate the respective ground-based stations.**  
 186 **The red and blue dots indicate the metropolitan and Industrial cities in India. The abbreviations represent the first**  
 187 **three letters of each city considered (e.g. DEL is Delhi), and their full names are given in Figure S3.**

188 GEOS-Chem uses a set of chemical mechanisms implemented with the Kinetic PreProcessor (KPP) (Damian et  
 189 al., 2002). The standard chemical mechanism in GEOS-Chem has undergone continuous development since the  
 190 original tropospheric gas-phase scheme of Bey et al. (2001), with subsequent extensions to include aerosol  
 191 chemistry (Park, 2004), stratospheric chemistry (Eastham et al., 2014), and a detailed tropospheric–stratospheric  
 192 halogen chemistry module (Wang et al., 2019), which include 299 chemical species (Fritz et al., 2022). Sulfate–  
 193 nitrate–ammonium aerosols are simulated with a bulk thermodynamic approach (Park, 2004), where gas–particle  
 194 partitioning is calculated using ISORROPIA II (Fountoukis and Nenes, 2007). Sea salt is represented with two  
 195 modes distinguishing fine and coarse particles (Jaeglé et al., 2011), while mineral dust is described using four  
 196 particle size bins (Fairlie et al., 2007). Furthermore, the simple SOA scheme is employed for secondary organic  
 197 aerosols, based on reversible partitioning of semivolatile products of VOC oxidation (Pye et al., 2010).  
 198 Furthermore, in GEOS-Chem, PM<sub>10</sub> is diagnosed as the sum of PM<sub>2.5</sub> plus size-resolved dust (70% of DST2, all  
 199 of DST3, 90% of DST4) and coarse sea-salt (SALC scaled by the hygroscopic growth factor, e.g., 1.86 at 35–  
 200 50% RH), with the final values archived at standard temperature and pressure (STP) using the ideal gas law  
 201 (GEOS-Chem Support Team, 2023).

202 Furthermore, we use ground-based measurements from Central Pollution Control Board (CPCB) monitoring sites  
203 situated in different regions of India, as shown in **Figure 1** and supplementary **Figure S3**, reflecting a wide array  
204 of meteorological conditions. The CPCB monitors ambient ozone using UV photometric methods (based on the  
205 absorption of UV light by ozone at 254 nm), chemiluminescence (where ozone reacts with a reagent to produce  
206 light that is quantified), and chemical methods (colorimetric reactions providing concentration estimates). PM<sub>10</sub>  
207 is measured using gravimetric analysis (filter-based mass determination after sampling), Tapered Element  
208 Oscillating Microbalance or TEOM (continuous real-time mass measurement through frequency change of a  
209 vibrating element), and beta attenuation techniques (attenuation of beta radiation as it passes through particle-  
210 laden filters to infer mass concentration) and these data are available at a resolution of 1 hour (NAAQS, 2013).

211 The altitude of the lowest model level may not consistently align with the actual altitude of ground-based  
212 monitoring locations, thereby causing inconsistencies between modelled and observed data. Therefore, we  
213 calculate the correlation coefficient for the time series of ozone and PM<sub>10</sub> values at each station to evaluate the  
214 strength of the association between these variables, as shown in **Figure S3**. The correlation coefficient between  
215 modelled and observed values of PM<sub>10</sub> and ozone is above 0.6 for most of the stations, except in Jaipur in the  
216 northwest, where the correlations are 0.5 and 0.4 for PM<sub>10</sub> and ozone, respectively. Although these moderate to  
217 strong correlations suggest a satisfactory alignment between the model and the facts, certain systematic  
218 differences remain. The GEOS-Chem model generally underestimates PM<sub>10</sub> concentration, possibly attributable  
219 to its 0.25°x0.3125° resolution, which fails to account for fine-scale local variations, including point sources of  
220 pollution. In contrast, the model often overestimates surface ozone concentrations for most of the stations  
221 considered here. Despite the model's higher bias caused by uncertainties in emissions of anthropogenic and  
222 biogenic precursors, uncertainties in ozone sinks such as dry deposition, and the coarse resolution of the model  
223 limits its ability to capture ozone titration and other localised chemical processes (Westervelt et al., 2019). Our  
224 analysis focuses on the differences between simulations for 2018 and 2022, with model performance evaluated  
225 using ground-based CPCB observations of surface ozone and PM<sub>10</sub> (**Figure 2, Figure S4 and Figure S5**). GEOS  
226 Chem broadly captures regional patterns of annually averaged PM<sub>10</sub> and ozone, but ground-based stations show  
227 higher PM<sub>10</sub> in NW and IGP (140–160 µg/m<sup>3</sup> vs. model 120–140 µg/m<sup>3</sup>, which is about 15–20% low bias) and  
228 lower values in CI and PI (about 10–15% high bias). For ozone, the model underestimates HR and IGP peaks by  
229 about 10–15% (observed >150 µg/m<sup>3</sup>) while overestimating coastal PI levels by about 10–20% (observed 70–90  
230 µg/m<sup>3</sup>). The model reproduces the temporal patterns reasonably well, with annually averaged differences between  
231 modelled and observed values remaining within 10–20% across most stations. Despite a few regionally specific  
232 biases (higher relative differences in PM<sub>10</sub> in cities like Talcher, Noida, Jodhpur during the monsoon season), the  
233 consistent performance across two meteorologically different years lends confidence to the overall robustness of  
234 the high-resolution GEOS-Chem simulations for India.

### 235 **2.3 Methods**

236 The aerosol uptake coefficient, which quantifies the efficacy of particles in absorbing reactive gases, substantially  
237 impacts surface ozone concentrations (Jacob, 2000). In areas with elevated aerosol concentrations, aerosols  
238 possessing a high uptake coefficient can more efficiently eliminate ozone precursors, including HO<sub>2</sub> and other  
239 reactive radicals, from the atmosphere (Li et al., 2018). These aerosols interact with atmospheric chemical species,

240 reducing the levels of OH and HO<sub>2</sub> radicals, which are essential for ozone synthesis. The aerosol uptake coefficient  
241 of HO<sub>2</sub> ( $\gamma_{HO_2}$ ), however, is affected by several factors, including the physical and chemical characteristics of the  
242 aerosol particles, such as size, composition and moisture content (George and Abbatt, 2010). These parameters  
243 show the efficacy of aerosol interactions with atmospheric radicals, thereby affecting ozone production.

244 Termination rates of chain reactions were determined using archived species concentrations and physical  
245 parameters such as temperature, pressure and humidity, in addition to aerosol properties, as in Ivatt et al. (2022).  
246 Although we do not classify radical product generation in peroxy-radical self-reactions as termination stages, we  
247 regard non-radical products as ongoing termination processes. The heterogeneous loss rate of HO<sub>2</sub> was assessed  
248 by evaluating the radius and surface area of different aerosol forms from the archived model outputs. For our  
249 simulations, we employed a default HO<sub>2</sub> reactive uptake coefficient ( $\gamma_{HO_2}$ ) of 0.2. Laboratory studies of pure  
250 synthetic aerosols indicate lower uptake coefficients ( $\gamma_{HO_2} < 0.2$ ), whereas real-world aerosol studies reveal  
251 values between 0.08 to 0.40, implying that elements such as transition metals may increase aerosol uptake (Kolb  
252 et al., 2010; Christian et al., 2018). We have also simulated the model with  $\gamma_{HO_2}$  to be zero to evaluate its impact  
253 on ozone formation mechanism. While we presumed H<sub>2</sub>O to be the exclusive product of HO<sub>2</sub> absorption, our  
254 results remain valid when H<sub>2</sub>O<sub>2</sub> is considered. Employing a singular  $\gamma_{HO_2}$  value likely oversimplifies the  
255 variability, as existing models fail to account for its temporal oscillations (Stavrakou et al., 2013; Sheehy et al.,  
256 2010).

257 The heterogeneous radical loss rate of HO<sub>2</sub> is determined by iterating through the radius and surface area of  
258 various aerosol types, which includes sulfate–nitrate–ammonium thermodynamics, secondary organic aerosol,  
259 dust, sea salt, black carbon, and primary organic carbon (Ivatt et al., 2022). These factors, along with temperature  
260 and air density, are used to calculate the first-order loss rate of HO<sub>2</sub>. Furthermore, emissions and photochemical  
261 reactions are strongly affected by seasonal variations in India. The seasons are defined as Winter (December–  
262 January–February; DJF), Pre-monsoon (March–April–May; MAM), Monsoon (June–July August–September;  
263 JJAS) and Post-monsoon (October–November; ON).

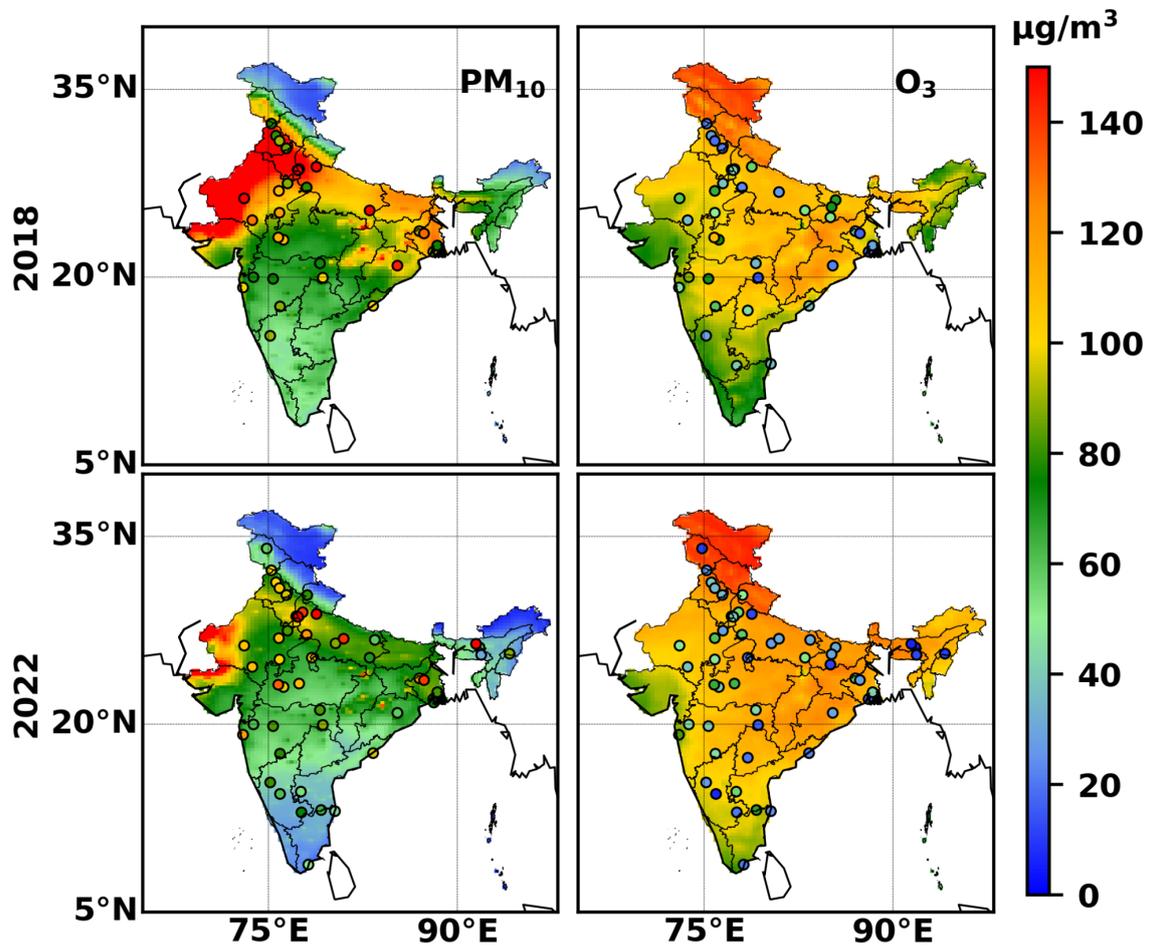
## 264 3. Results

### 265 3.1 Distribution of Surface Ozone and PM for the years 2018 and 2022

#### 266 3.1.1 Annual Distribution

267 **Figure 2** shows the annual average PM<sub>10</sub> and ozone in India for the years 2018 and 2022. The highest PM<sub>10</sub>  
268 concentrations (130–150  $\mu\text{g}/\text{m}^3$ ) occur in the NW dry regions due to dust mobilisation from the Thar Desert  
269 (Santra et al., 2018; Kashyap et al., 2024), consistent with studies showing dry and semi-arid areas as key mineral  
270 dust sources (Maji and Sonwani, 2022). Ground-based observations here often exceed 160  $\mu\text{g}/\text{m}^3$ , indicating that  
271 the model underestimates by about 10–20%. IGP also shows higher PM<sub>10</sub> (120–140  $\mu\text{g}/\text{m}^3$ ) from industrial,  
272 vehicular and agricultural emissions including biomass and crop-residue burning (Devi et al., 2020; Hassan et al.,  
273 2023). Station data shows average concentrations of about 140–160  $\mu\text{g}/\text{m}^3$ , indicating that model is biased low  
274 by about 15%. Despite higher emissions, greater humidity and rainfall promote wet deposition and particle  
275 scavenging, lowering levels relative to NW (Sen et al., 2017; Singh et al., 2023). CI records 60–100  $\mu\text{g}/\text{m}^3$  with

276 industrial hotspots up to 130–150  $\mu\text{g}/\text{m}^3$  (Lokhande and Khan, 2021; Saini et al., 2023), but observations show  
 277 about 50–90  $\mu\text{g}/\text{m}^3$ , suggesting the model slightly overestimates by about 10–15%. NE shows lower levels (40–  
 278 70  $\mu\text{g}/\text{m}^3$ ) due to sparse population and vegetation sinks (Guttikunda and Nishadh, 2022; Kumari et al., 2025).



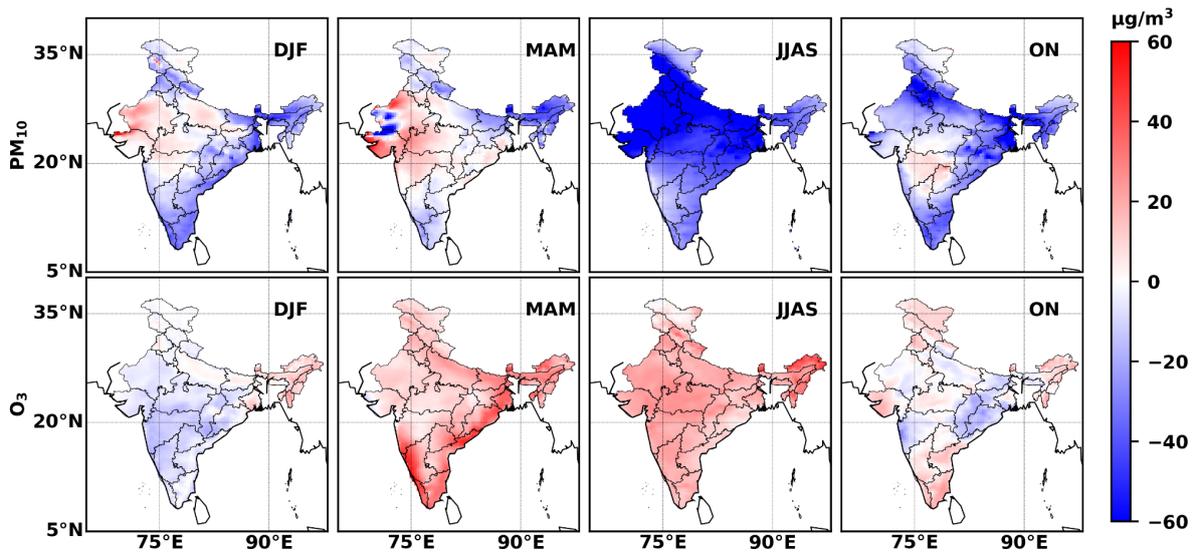
279  
 280 **Figure 2: Annual average distribution of  $\text{PM}_{10}$  and ozone for the years (Top) 2018 and (Bottom) 2022 from GEOS-**  
 281 **Chem simulations. The dots show the annually averaged mean values of  $\text{PM}_{10}$  and ozone at the respective ground-based**  
 282 **CPCB stations for the years 2018 and 2022.**

283 Surface ozone peaks in the HR (120–140  $\mu\text{g}/\text{m}^3$ ) due to high stratospheric-to-troposphere transport, Ground-based  
 284 data here often reach 150–160  $\mu\text{g}/\text{m}^3$ , showing that model underpredicts by about 15%. (Phanikumar et al., 2017;  
 285 Wang et al., 2024). High values also occur in IGP and eastern CI (100–120  $\mu\text{g}/\text{m}^3$ ) driven by secondary  
 286 photochemistry from  $\text{NO}_x$  and VOCs (Payra et al., 2022; Sinha and Sinha, 2019; Mahilang et al., 2021;  
 287 Kuttippurath et al., 2022) under stagnant conditions (Gopikrishnan and Kuttippurath, 2024). Observations in these  
 288 regions are typically 110–130  $\mu\text{g}/\text{m}^3$ , reflecting a model underestimation of about 10–15%. PI shows the lowest  
 289 ozone (80–100  $\mu\text{g}/\text{m}^3$ ) due to coastal ventilation, humidity, precipitation and oceanic loss, with observed levels  
 290 closer to 70–90  $\mu\text{g}/\text{m}^3$ , showing that the model overestimates by about 10–20% (Li et al., 2019; Lakshmi et al.,  
 291 2024; Galbally and Roy, 1980; Lavanyaa et al., 2023; Nilaya et al., 2024). A detailed discussion of seasonal  
 292 changes in surface ozone is provided in the supplementary material, **section 1.1**.

293 Seasonally, PM<sub>10</sub> is highest in IGP during winter and post-monsoon (100–140 µg/m<sup>3</sup>) under stable, inversion  
294 conditions and intensified biomass and crop-residue burning (Mhawish et al., 2020; Paulot et al., 2022;  
295 Jayachandran and Rao, 2024; Payra et al., 2022; Mogno et al., 2021). The NW records its peak during the monsoon  
296 from dust storms (Yadav et al., 2022; Yu et al., 2023), while pre-monsoon sees widespread elevated PM<sub>10</sub> (60–  
297 100 µg/m<sup>3</sup>) with NW reaching 140–160 µg/m<sup>3</sup> due to dust, traffic, industrial and construction sources (Patil et al.,  
298 2013; Garg and Gupta, 2020). These high levels pose severe health risks, particularly for vulnerable groups  
299 (Adhikary et al., 2024; Pathak et al., 2024; Gopikrishnan and Kuttippurath, 2025). Ozone peaks in pre-monsoon,  
300 especially over CI, IGP and NE, where it exceeds 140–160 µg/m<sup>3</sup>, driven by strong solar radiation and abundant  
301 precursors (Kutal et al., 2022; Sinha et al., 2014). Winter ozone is also high in CI (100–120 µg/m<sup>3</sup>), followed by  
302 IGP and NE (80–100 µg/m<sup>3</sup>) under stagnant meteorology and local emissions (David and Nair, 2011; Gao et al.,  
303 2020). In contrast, monsoon levels are lowest, with the west coast at 60–80 µg/m<sup>3</sup>, NW at 80–100 µg/m<sup>3</sup> and NE  
304 and IGP at 120–140 µg/m<sup>3</sup>, owing to cloud cover, precipitation and circulation that suppress ozone production  
305 and enhance removal (Lal et al., 2014; Lu et al., 2018). Post-monsoon levels are moderate (100–120 µg/m<sup>3</sup>) across  
306 CI, NW and IGP, with PI and NE slightly lower (60–90 µg/m<sup>3</sup>) (Lu et al., 2018). A detailed discussion of seasonal  
307 changes in surface ozone is provided in the supplementary material, **section 1.2**.

### 308 **3.1.3 Comparison of PM<sub>10</sub> and Ozone levels**

309 The relationship between PM<sub>10</sub> and ozone is often inverse, attributable to their distinct formation mechanisms and  
310 atmospheric interactions, but varies substantially depending on meteorological conditions and precursor  
311 availability (Jia et al., 2017). Higher PM levels, frequent during winter in areas such as IGP, often correlate with  
312 lower ozone concentrations (Song et al., 2022). PM disperses and absorbs sunlight, reducing the solar radiation  
313 essential for photochemical ozone formation, which depends on NO<sub>x</sub> and VOCs concentration in the presence of  
314 intense sunlight. Black carbon (BC), a constituent of PM from combustion, can directly scavenge ozone by acting  
315 as a deposition surface (Gao et al., 2018). However, this direct BC–ozone scavenging pathway is not explicitly  
316 represented in most chemical transport models (Koch et al., 2009), but its overall contribution is considered  
317 modest compared to dominant photochemical production and deposition processes, it may still have localised  
318 importance in high-BC environments such as South Asia (Kumar et al., 2015). Furthermore, stable atmospheric  
319 conditions including temperature inversions that confine PM at the surface also impede vertical mixing, limiting  
320 ozone transport from the upper atmosphere, hence facilitating the accumulation of ozone near the surface  
321 (Gopikrishnan and Kuttippurath, 2025). In such cases, the co-accumulation of PM and ozone near the surface does  
322 not imply a simple inverse relationship, as aerosol–radiation interactions can further suppress the planetary  
323 boundary layer (PBL) height (Li et al., 2017; Zou et al., 2017), thereby enhancing pollutant trapping. Under  
324 suppressed PBL conditions and high NO<sub>x</sub> emissions, ozone titration can also occur, leading to additional  
325 complexity in the surface ozone response (Lin et al., 2008; Li et al., 2024). Conversely, cleaner environments with  
326 reduced PM concentrations enhance ozone generation due to the lack of light scattering and scavenging, which  
327 fosters photochemical activity (Rathore et al., 2023; Sicard et al., 2023).



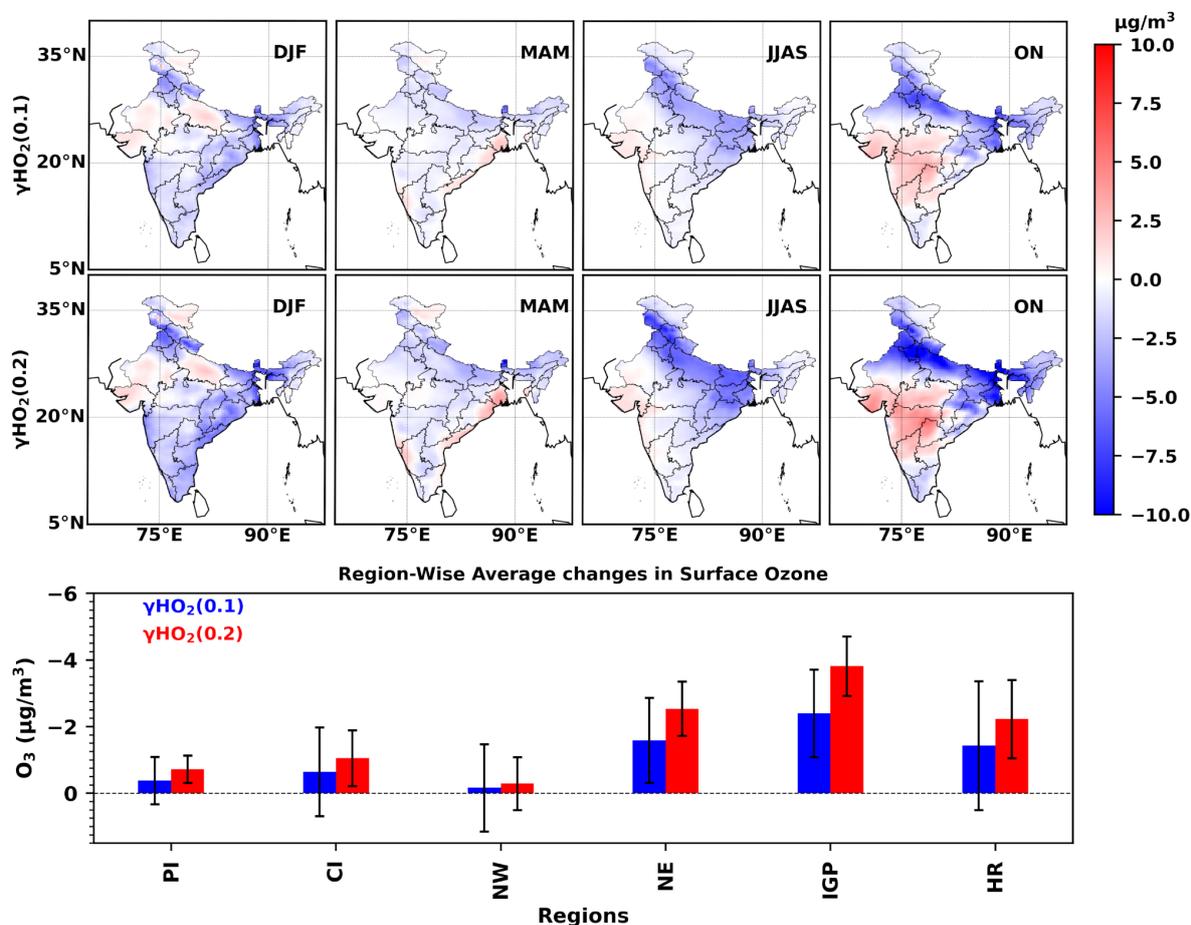
328

329 **Figure 3: The difference in (Top) PM<sub>10</sub> and (Bottom) Ozone in 2022 when compared to that of 2018.**

330 Figure 3 shows the change in PM<sub>10</sub> and ozone in India for the year 2022 when compared to that of 2018. In the  
 331 monsoon and post-monsoon seasons, PM concentrations drop by 40–60 µg/m<sup>3</sup> in the NW, IGP and eastern CI,  
 332 while reductions of 10–30 µg/m<sup>3</sup> occur in other regions, due to changes in the spatial distribution of precipitation  
 333 as shown in **Figures S6 and S7** (Singh et al., 2023). **Figure S8** presents the changes in aerosol surface area  
 334 between 2018 and 2022, whereas **Figure S9** illustrates the contributions from different aerosol sources in these  
 335 two years. The reduction in PM facilitates increased solar radiation penetration, hence promoting photochemical  
 336 ozone production, resulting in a substantial rise in ozone concentrations (20–30 µg/m<sup>3</sup> in most of India during  
 337 monsoon and 20–30 µg/m<sup>3</sup> in PI and western CI during post-monsoon. The decrease in PM<sub>10</sub> reduces the surface  
 338 area accessible for heterogeneous processes that might otherwise reduce ozone levels (Gopikrishnan et al., 2025).  
 339 Conversely, in pre-monsoon and winter seasons, PM concentrations increase by 10–30 µg/m<sup>3</sup> regionally,  
 340 particularly in NW and central IGP, inhibiting ozone formation due to reduced solar radiation and enhanced  
 341 heterogeneous chemistry that depletes reactive species such as HO<sub>2</sub> (Wang et al., 2024). The stagnant  
 342 meteorological conditions associated with winter, including temperature inversions, elevated humidity, and  
 343 minimal wind speeds, intensify PM buildup, particularly in IGP, resulting in further reductions in ozone levels  
 344 (Gopikrishnan and Kuttippurath, 2024). Nonetheless, geographical disparities remain, exemplified by a 20 µg/m<sup>3</sup>  
 345 reduction in PM in the Upper Northeast of IGP during pre-monsoon, which causes a 20–30 µg/m<sup>3</sup> rise in ozone  
 346 levels, and a 10–20 µg/m<sup>3</sup> decline in PM near the west coast, resulting in a 40–60 µg/m<sup>3</sup> increase in ozone. In  
 347 winter, ozone variations are negligible (within 10 µg/m<sup>3</sup>) over much of India; however, a reduction in PM (10–30  
 348 µg/m<sup>3</sup>) in the northeast correlates with a 20–30 µg/m<sup>3</sup> rise in ozone levels. The seasonal variability is additionally  
 349 affected by the aerosol-photolysis feedback, wherein PM influences ozone by absorbing important species such  
 350 as H<sub>x</sub>O<sub>y</sub> and NO<sub>x</sub>, hence reducing surface photolysis rates and inhibiting ozone production (Ivatt et al., 2022;  
 351 Gopikrishnan et al., 2025).

352

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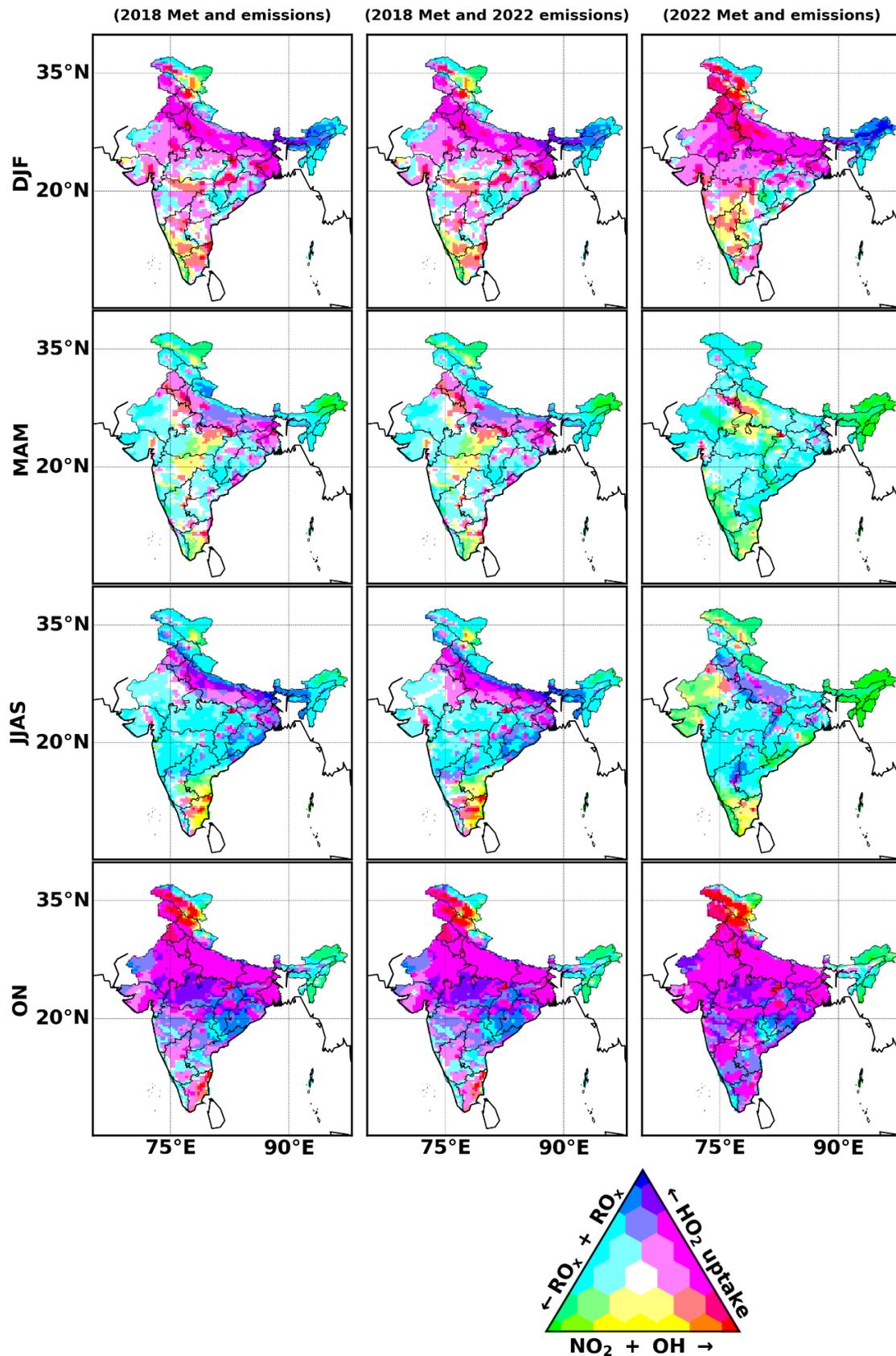
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357 **Figure 4: Difference in surface ozone response to aerosol uptake between 2022 and 2018, expressed relative to the no-**  
 358 **uptake case ( $\gamma\text{HO}_2 = 0$ ). The top panel shows  $[(\text{O}_3 \text{ at } \gamma\text{HO}_2 = 0.0 \text{ in } 2022 - \text{O}_3 \text{ at } \gamma\text{HO}_2 = 0.1 \text{ in } 2022) - (\text{O}_3 \text{ at } \gamma\text{HO}_2 = 0.0$**   
 359 **in 2018 -  $\text{O}_3 \text{ at } \gamma\text{HO}_2 = 0.1 \text{ in } 2018)]$ , while the middle panel shows the analogous calculation for  $\gamma\text{HO}_2 = 0.2$ . The bottom**  
 360 **panel shows the change in surface ozone averaged for the entire year when  $\gamma\text{HO}_2 = 0.1$  and  $\gamma\text{HO}_2 = 0.2$ . The bottom**  
 361 **panel presents a bar chart of regionally averaged annual surface ozone changes for  $\gamma\text{HO}_2 = 0.1$  and  $\gamma\text{HO}_2 = 0.2$ , derived**  
 362 **from the top and middle panels. Here, negative values indicate stronger ozone suppression in 2018, while positive values**  
 363 **indicate stronger suppression in 2022.**

364 The GEOS-Chem model simulations for 2018 and 2022 were performed without and with aerosol uptake of HO<sub>2</sub>  
 365 to study the impact of  $\gamma\text{HO}_2$  on surface ozone concentrations in India in the absence of this mechanism. The results  
 366 show that the aerosol absorption of HO<sub>2</sub> substantially influences ozone concentrations, as shown in **Figure 4**,  
 367 especially in areas like the IGP and eastern CI. The increase in ozone concentrations in these regions without  
 368 aerosols (**Figure S10**), attributed to the absorption of HO<sub>2</sub> by aerosols, varies from 5 to 10  $\mu\text{g}/\text{m}^3$ , with the most  
 369 significant changes during winter and post monsoon season. Winter season is defined by higher aerosol surface  
 370 area, which promotes heterogeneous processes that enhance the removal of HO<sub>2</sub>, hence restricting its availability  
 371 for ozone formation (Dyson et al., 2022). In 2022, aerosol concentrations rose substantially in winter when

372 compared to 2018, especially in the middle IGP and western CI. This augmentation of aerosol surface area in  
373 2022 inhibited ozone generation by about 5–7.5  $\mu\text{g}/\text{m}^3$  in these areas. The most notable changes in ozone  
374 concentrations were during post-monsoon, with an increase of about 5–10  $\mu\text{g}/\text{m}^3$  when no aerosol absorption is  
375 considered, especially in IGP and western CI. In 2022, there was a rise in aerosol surface area in western CI  
376 compared to 2018, resulting in an additional suppression of surface ozone. The pre-monsoon season period had  
377 similar changes in ozone concentration, with an increase of about 2.5–5  $\mu\text{g}/\text{m}^3$ , whereas the monsoon period  
378 observed minimal variation, with ozone concentration changes between 0 and 5  $\mu\text{g}/\text{m}^3$ . Thus, the changes in ozone  
379 concentrations are directly influenced by aerosol surface area, which amplifies the heterogeneous elimination of  
380  $\text{HO}_2$ , thereby restricting its availability for ozone synthesis (Anurose et al., 2024). Seasonal fluctuations in aerosol  
381 load, influenced by factors like monsoon dynamics, atmospheric transport, and emissions, greatly influence the  
382 effects of aerosol- $\text{HO}_2$  interactions on ozone levels. The rise in aerosol concentrations in 2022, especially during  
383 winter and post-monsoon, can be attributed to higher anthropogenic activities increasing sulphate, BC and OC,  
384 which enhance the suppression of ozone production in these seasons (Austin et al., 2015; Zhang et al., 2023).

385 In addition to the simulation with a  $\gamma_{\text{HO}_2}$  of 0.2, a sensitivity analysis using  $\gamma_{\text{HO}_2} = 0.1$  was conducted for the  
386 years 2018 and 2022, as shown in **Figure 4** and **Figure S11**, respectively. This lower value was chosen, as  
387 previous studies suggest that  $\gamma_{\text{HO}_2} = 0.2$  may overestimate heterogeneous uptake, particularly over East Asia,  
388 where values closer to 0.1 have been reported under typical ambient conditions (Yang et al., 2023). Given the  
389 large uncertainty and variability in  $\gamma_{\text{HO}_2}$  arising from factors such as aerosol composition, relative humidity, and  
390 particle acidity (Lakey et al., 2024), it is critical to assess the sensitivity of modeled ozone to a range of uptake  
391 efficiencies. This uncertainty can lead to substantial differences in simulated ozone, as even a halving of  $\gamma_{\text{HO}_2}$   
392 (from 0.2 to 0.1) significantly reduces the magnitude and spatial extent of ozone suppression. The comparison  
393 between the two scenarios indicates that the impact on surface ozone concentrations is both spatially and  
394 seasonally dependent, with  $\gamma_{\text{HO}_2} = 0.2$  yielding a more pronounced and widespread increase when compared to a  
395 no aerosol uptake scenario. During the DJF and ON seasons, the increase in ozone is up to 10  $\mu\text{g}/\text{m}^3$  across IGP,  
396 eastern, and central India, while the  $\gamma_{\text{HO}_2} = 0.1$  simulation shows more modest reductions in the range of 5–10  
397  $\mu\text{g}/\text{m}^3$  over similar regions. In the MAM and JJAS seasons, the differences in ozone concentrations are  
398 comparatively weaker in both scenarios, generally within 0 to 5  $\mu\text{g}/\text{m}^3$ , although localized reductions persist over  
399 northern and eastern parts of India. The interannual differences (2022–2018) shows a consistent spatial pattern,  
400 particularly when  $\gamma_{\text{HO}_2}$  is 0.1, where negative anomalies of up to 7.5  $\mu\text{g}/\text{m}^3$  are observed across northern and  
401 central India. Additionally, small positive anomalies (up to 3  $\mu\text{g}/\text{m}^3$ ) are observed in isolated regions such as parts  
402 of western Rajasthan, Gujarat and southern peninsular India.



403

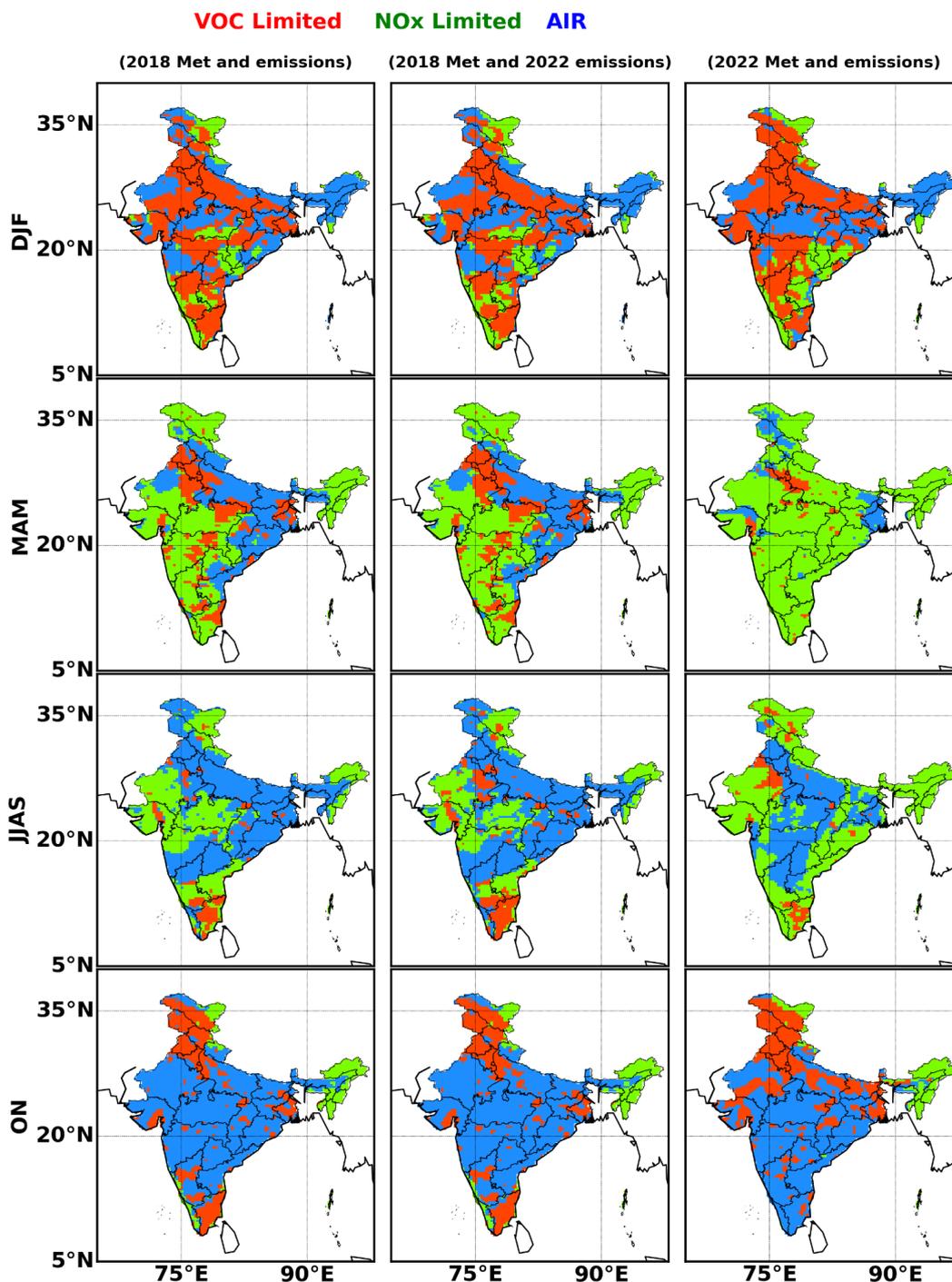
404 Figure 5: Mean fraction of radical termination at the surface that occurs through OH + NO<sub>2</sub> (red), peroxy-radical self-  
 405 reactions (green) and aerosol uptake of HO<sub>2</sub> (blue) for the year (left) 2018 and (right) 2022.

## 406 4.2 Surface Ozone Formation Regimes

407 **Figure 5** shows the surface distribution of the relative chemical fluxes associated with the termination steps of  
408 ozone formation in India for the years 2018 and 2022. During monsoon in 2022, peroxy-radical self-reactions  
409 (green) dominate most of PI, NE and NW arid regions, while in 2018, ozone termination was equally influenced  
410 by  $\text{NO}_2 + \text{OH}$  reactions and peroxy-radical self-reactions in PI, whereas NW and NE by  $\text{HO}_2$  uptake onto aerosols  
411 (blue). When emissions are updated to 2022 levels but meteorology is fixed at 2018 (middle column), the chemical  
412 regimes remain almost similar, indicating that meteorology contributes substantially to the changing chemical  
413 regimes, as found in the 2022 meteorology and chemistry coupled simulations (third column). The peroxy-radical  
414 self-reactions are more widespread across major cities in India (for e.g., Visakhapatnam, Delhi and Hyderabad),  
415 reflecting the reduced availability of  $\text{NO}_x$  as a radical sink. This weakening of  $\text{NO}_2 + \text{OH}$  termination is particularly  
416 evident outside IGP, where declining  $\text{NO}_x$  emissions shift the balance toward radical–radical termination  
417 pathways. Thus, the dominance of peroxy-radical self-reactions indicates that  $\text{NO}_x$  emissions in these cities limit  
418 ozone formation, promoting pathways where peroxy radicals neutralize ozone, through the VOC limited ozone  
419 formation regime (Romer et al., 2018).

420 During winter,  $\text{NO}_2 + \text{OH}$  termination and the uptake of  $\text{HO}_2$  on aerosols became prominent over much of India,  
421 particularly in IGP. The increased aerosol surface area during this period enhances the uptake of  $\text{HO}_2$ , thereby  
422 inhibiting ozone production (**Figure S8** and **Figure S9**). The presence of aerosols, often exacerbated by biomass  
423 burning and dust transport, amplifies this effect in areas where aerosols are abundant (Ivatt et al., 2022; Singh et  
424 al., 2018). Under fixed 2018 meteorology with 2022 emissions, the chemical termination mechanisms remain  
425 similar in most regions. Nevertheless, we observe the dominance of  $\text{HO}_2$  uptake chemical termination in the PI  
426 region in 2022 when model simulations are coupled with the 2022 meteorology and emissions. In post-monsoon,  
427 the termination of ozone formation shifts predominantly to  $\text{HO}_2$  uptake across many regions, particularly in IGP,  
428 CI, and parts of PI. The NE, however, is an exception, where peroxy-radical self-reactions continue to dominate  
429 the termination process. This variation in termination pathways can be attributed to regional differences in aerosol  
430 loading and the influence of seasonal emissions, including biomass burning during the post-monsoon season,  
431 which contributes to higher aerosol concentrations in IGP and CI. The fixed meteorology simulations show that  
432 the spatial extent of chemical termination mechanisms closely resembles the results of the simulations with 2022  
433 emissions and 2018 meteorology in ON.

434 During the pre-monsoon period in 2022, the southern parts of northern IGP are predominantly influenced by  $\text{HO}_2$   
435 uptake, whereas the rest of IGP and much of India find dominant termination by peroxy-radical self-reactions.  
436 The increased aerosol surface area and the presence of fire events, particularly in the dry regions of NW and NE  
437 regions, enhance the uptake of  $\text{HO}_2$ , suppressing ozone formation (Kumari et al., 2025). Fire events and desert  
438 dust also contribute to higher aerosol concentrations, reinforcing the impact of heterogeneous reactions on ozone  
439 chemistry (Dyson et al., 2023; Dhanurkar et al., 2024). With 2018 and 2022 emissions under 2018 meteorology,  
440 the chemical termination in MAM is clearly through peroxy-radical reactions and  $\text{NO}_2 + \text{OH}$  regimes, except in  
441 IGP where  $\text{HO}_2$  uptake dominated the chemical termination mechanism, which can be attributed to high fire  
442 activity and dust loading (Kuttippurath et al., 2022).



443

444 **Figure 6: The regional distribution of various ozone generation photochemical regimes modelled with  $\gamma_{HO_2}$  of 0.2 in**  
 445 **(left) 2018 and (right) 2022. Here, AIR is Aerosol Inhibited Regions.**

446 Furthermore, **Figure 6** splits the domain based on the local largest termination step. In regions characterized as  
 447 'NO<sub>x</sub>-limited,' the peroxy self-reactions dominate, making reductions in nitrogen oxides (NO<sub>x</sub>) the most effective  
 448 strategy for mitigating ozone pollution; this contrasts with 'VOC limited' regimes, where VOC reductions are  
 449 more beneficial, with the transition between these regimes dictated by the atmospheric concentrations of NO<sub>x</sub> and

450 VOCs. Aerosol uptake of hydroperoxyl radicals ( $\text{HO}_2$ ) on aerosol surfaces can lead to an 'aerosol inhibited'  
451 environment, diminishing ozone formation in areas with elevated aerosol concentrations.

452 In 2018, much of IGP and CI were in an AIR during summer and monsoon, where the presence of aerosols,  
453 particularly in regions with high anthropogenic emissions and biomass burning, suppressed ozone formation by  
454 promoting the heterogeneous uptake of  $\text{HO}_2$ . However, a substantial reduction in aerosol surface area in 2022,  
455 shifted some of these regions into a  $\text{NO}_x$ -limited regime (for e.g. eastern CI, eastern IGP), where the availability  
456 of  $\text{NO}_x$  becomes the limiting factor for ozone formation. This transition to a  $\text{NO}_x$ -limited regime is associated  
457 with a subsequent increase in ozone concentrations in 2022 compared to 2018, as the reduction in aerosols resulted  
458 in more ozone to form via the photochemical pathways involving  $\text{NO}_x$  and VOCs. The decrease in aerosol surface  
459 area, coupled with reduced aerosol-induced  $\text{HO}_2$  removal, enhanced the ozone production efficiency, leading to  
460 higher ozone levels in these regions. Nevertheless, when emissions are updated to 2022 but meteorology is fixed  
461 for 2018, large parts of CI, PI, and NE transit out of AIR into  $\text{NO}_x$ -limited regimes. This shows that emission  
462 reductions, particularly in  $\text{NO}_x$ , are sufficient to drive regime shifts, but the meteorological changes also play a  
463 major role in these regions, with IGP remaining the strongest VOC-limited hotspot among the urban centers.

464 In contrast, during winter and post-monsoon, there is less to no difference in the dominant regimes between 2022  
465 and 2018. In winter, most of India remains in a VOC-limited regime, where VOCs (such as those from biogenic  
466 sources and fossil fuel emissions) limit ozone formation. CI and NE continue to be in AIR during this period, with  
467 high aerosol concentrations suppressing ozone production through  $\text{HO}_2$  uptake. During post-monsoon, the  
468 majority of India remains in AIR, except for northern IGP, which is in a VOC-limited regime, and NE, where the  
469 region is predominantly  $\text{NO}_x$ -limited. The fixed meteorology simulations show that, during the post-monsoon  
470 season, the western coast turns out to be in an AIR, whereas this region is  $\text{NO}_x$ -limited in the 2022 meteorology  
471 simulations. However, IGP remains strongly VOC-limited due to persistent high  $\text{NO}_x$  emissions and stagnant  
472 meteorological conditions, mostly during DJF and partly in ON. The seasonal consistency in winter and post-  
473 monsoon suggests that the aerosol levels together with the overall seasonal and regional variations in emissions,  
474 meteorology and chemistry play a larger role in initiating the ozone formation regime in these periods. Therefore,  
475 while aerosol changes in 2022 had a notable impact on ozone production during monsoon, they had a lesser  
476 influence on the seasonal regimes in winter and post-monsoon. This enhanced sensitivity during the monsoon is  
477 attributable to elevated humidity and aerosol liquid water content, stronger photochemical radical production, and  
478 deeper boundary layer dynamics, which collectively amplify the influence of aerosol uptake on ozone formation  
479 relative to winter or post-monsoon.

#### 480 **4.3 Impact of meteorology on surface ozone formation regimes**

481 The interannual comparison shows that surface ozone variability in India is governed primarily by meteorological  
482 drivers, with a secondary contribution from emission-induced changes in PM and associated  $\text{HO}_2$  uptake. The  
483 difference between 2018 and 2022 (**Figure 3**) shows large and spatially coherent ozone anomalies, often  
484 exceeding  $\pm 20\text{--}60 \mu\text{g m}^{-3}$  across northern and central India. These differences are most pronounced during MAM  
485 and JJAS, when enhanced solar radiation, elevated temperatures, and changes in circulation patterns strongly  
486 modulate precursor availability and photochemical activity (Rathore et al., 2023). For instance, ozone increases  
487 of about  $30\text{--}40 \mu\text{g m}^{-3}$  are evident across northern India during JJAS, coinciding with large PM reductions ( $>40$

488  $\mu\text{g m}^{-3}$ ), while the signal is weaker in southern India (about 10–15  $\mu\text{g m}^{-3}$ ), reflecting the combined influence of  
489 regional circulation and boundary layer dynamics (Gopikrishnan and Kuttippurath, 2024). Such large magnitudes  
490 strongly suggest that the direct impact of meteorology on surface ozone pathways, through direct modulation of  
491 precursor lifetimes, photolysis rates and vertical mixing also exert dominant influence on ozone variability.

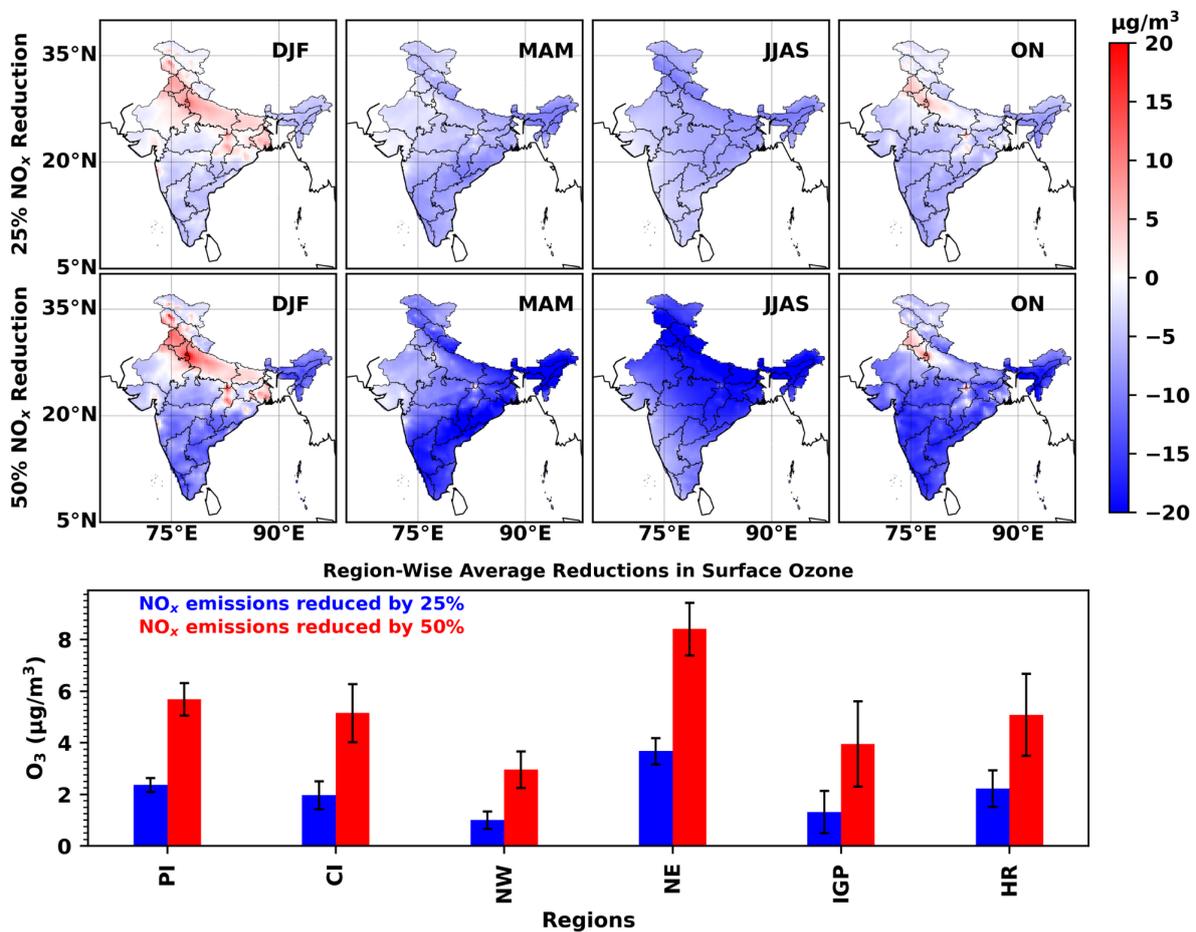
492 In contrast, the isolated influence of aerosol HO<sub>2</sub> uptake, as represented in the  $\gamma\text{HO}_2$  sensitivity simulations  
493 (Figure 4), yields substantially smaller ozone changes, typically limited to about 5–10  $\mu\text{g/m}^3$ . This is most visible  
494 in IGP and eastern coasts of India, where high PM levels enhance HO<sub>2</sub> uptake, thereby suppressing ozone  
495 formation under polluted conditions. However, even in these regions, the HO<sub>2</sub> uptake effect is an order of  
496 magnitude smaller than the direct meteorological impact (20–40  $\mu\text{g/m}^3$ ). The radical termination budget (Figure  
497 5) provides mechanistic clarity: across most of India, ozone termination is dominated by OH+NO<sub>2</sub> reactions  
498 (accounting for about 60–70%), whereas the contribution of HO<sub>2</sub> uptake is rarely about 20–30%, and is substantial  
499 only in persistently polluted environments (cities/urban regions). This means that while aerosol uptake of HO<sub>2</sub>  
500 can influence local ozone production regimes by shifting the balance between propagation and termination, its  
501 influence on interannual ozone variability is minor compared to the large-scale and seasonally varying imprint of  
502 meteorology.

503 However, in future, the relative balance between these pathways is expected to evolve. As PM concentrations  
504 decline due to emission controls, the buffering capacity of aerosol HO<sub>2</sub> uptake will weaken, leaving ozone  
505 formation increasingly sensitive to direct meteorological perturbations. Under present conditions, the PM–HO<sub>2</sub>  
506 uptake pathway contributes only about 10–20% of the ozone variability seen in Figure 5. However, in a future  
507 low-PM environment, even normal fluctuations in rainfall and atmospheric circulation could lead to  
508 disproportionately higher ozone. For example, during dry years with reduced wet scavenging and enhanced  
509 precursor accumulation, the absence of strong PM sinks may amplify ozone increases relative to normal or wet  
510 years, particularly in NO<sub>x</sub>-rich urban regions of northern India. This suggests that while emissions have already  
511 reduced the potential for PM-driven radical termination, the dominant role of meteorology in setting ozone levels  
512 will likely strengthen, leading to more frequent and intense ozone enhancements during adverse meteorological  
513 episodes. Therefore, to counteract the enhanced ozone production expected under declining PM conditions,  
514 additional NO<sub>x</sub> controls will be essential.

#### 515 **4.4 Impact of the national efforts in reducing NO<sub>x</sub> and its effect on surface ozone pollution**

516 National and local efforts to reduce NO<sub>x</sub> emissions in India have had a noticeable impact on surface ozone  
517 pollution, particularly in urban and industrialized regions (Chen et al., 2021; Misra et al., 2021; Gopikrishnan et  
518 al., 2022). NO<sub>x</sub>, primarily emitted by vehicular traffic, power plants and industrial processes, plays a critical role  
519 in the formation of surface ozone through photochemical reactions. By reducing NO<sub>x</sub> emissions, several regions  
520 in India have observed a significant decrease in ozone formation. National initiatives such as stricter emission  
521 standards for vehicles, the promotion of cleaner fuels, and the adoption of advanced technologies in industrial  
522 operations have contributed to the reduction in NO<sub>x</sub> emissions (UNESCAP, 2022). These efforts are particularly  
523 impactful in regions like IGP, which has been a hotspot for high NO<sub>x</sub> concentrations and consequent ozone  
524 pollution. Furthermore, policies aimed at reducing vehicular traffic, such as improved public transport systems  
525 and the promotion of electric vehicles, have shown promise in lowering NO<sub>x</sub> levels and mitigating surface ozone

526 formation (Saikia, 2025). These measures help disrupt the photochemical cycle that leads to high ozone levels, as  
 527 lower NO<sub>x</sub> concentrations slow down ozone generation. However, the success of these efforts is often offset by  
 528 increasing emissions from other sectors, such as agriculture, where practices like biomass burning continue to  
 529 release NO<sub>x</sub> (Usmani et al., 2020). Despite these challenges, sustained efforts to reduce NO<sub>x</sub> emissions have  
 530 contributed to a gradual improvement in air quality, particularly in urban centers, leading to a secondary reduction  
 531 in surface ozone pollution in time. Nevertheless, more stringent emission reductions are required to tackle the  
 532 increasing ozone problem in India. Therefore, we scaled down the emissions of NO<sub>x</sub> in the model simulations to  
 533 see how this decrease in NO<sub>x</sub> would facilitate the ozone generation or destruction in different regions of India.  
 534 **Figure S12** shows the impact of 25% and 50% NO<sub>x</sub> emission cuts on aerosol surface area. Eastern India and the  
 535 IGP region experience the most substantial reduction in aerosol surface area due to NO<sub>x</sub> reductions, especially  
 536 during the post-monsoon (ON) and winter (DJF) periods. In the 50% reduction scenario, the decrease in these  
 537 areas reaches values as low as  $-4 \times 10^{-7} \text{ cm}^2 \text{ cm}^{-3}$ .



538

539 **Figure 7:** The change in ozone levels when NO<sub>x</sub> emissions are scaled down by (top) 25% and (middle) 50% when  
 540 compared to that of base simulation in 2022 with no reduction of emissions. (bottom) Region-wise averaged surface  
 541 ozone reduction when NO<sub>x</sub> emissions are scaled down by 25 and 50% for the year 2022. The whiskers represent the  
 542 standard deviations. Here, PI is Peninsular India, CI is Central India, NW is NorthWest, NE is North East India, IGP  
 543 is Indo-Gangetic Plains and HR is Hilly Regions.

544 **Figure 7** shows the difference in surface ozone in 2022 with 25% and 50% reduction in NO<sub>x</sub> emissions when  
545 compared to that of 2022 with no reduction in emissions. A decrease of approximately 5–10 µg/m<sup>3</sup> in ozone levels  
546 is found with a 25% reduction in NO<sub>x</sub> emissions, while a 10–15 µg/m<sup>3</sup> decrease occurs with a 50% reduction in  
547 NO<sub>x</sub> emissions in India. The most substantial reduction occurs during the monsoon season, characterised by a  
548 NO<sub>x</sub>-limited regime prevalent throughout much of India, succeeded by the post-monsoon and pre-monsoon  
549 periods. During winter, particularly in IGP and eastern CI, ozone levels increase, as these regions are limited by  
550 VOCs during this period. The increase is about 5–10 µg/m<sup>3</sup> with a 25% decrease in NO<sub>x</sub> emissions, and about  
551 10–15 µg/m<sup>3</sup> with a 50% reduction. Additionally, regional variations in ozone reduction are more pronounced in  
552 the 50% decrease scenario, while the 25% reduction shows a more uniform decrease across India (5–10 µg/m<sup>3</sup>).  
553 The reduction of ozone is most noticeable in PI during pre-monsoon and post monsoon (10–15 µg/m<sup>3</sup>), but, the  
554 ozone reduction occurs primarily during monsoon in IGP (10–15 µg/m<sup>3</sup>), with the smallest levels (5–10 µg/m<sup>3</sup>)  
555 in PI. The seasonal change can be due to decreased NO<sub>x</sub> levels in the PI during the monsoon, as rainfall  
556 significantly removes precursors in the atmosphere during this season. The findings indicate that roughly 75–80%  
557 of the ozone increase attributed to the decrease in PM and aerosols in India could possibly be alleviated through  
558 enhanced regulation of NO<sub>x</sub> emissions. Also, the findings suggest that winter strategies must focus on reducing  
559 VOCs, whereas stricter NO<sub>x</sub> control efforts are essential in other seasons to mitigate ozone pollution.

## 560 **5. Conclusions**

561 This study examines the interactions between PM<sub>10</sub>, aerosols and surface ozone concentrations in India, focusing  
562 on the changes modelled between 2018, a higher PM<sub>10</sub> year, and 2022, a lower PM<sub>10</sub> year. The findings highlight  
563 that the seasonal and regional variability of PM<sub>10</sub> and aerosol surface area play a major role in modulating surface  
564 ozone formation. During winter and post-monsoon seasons, elevated PM<sub>10</sub> and aerosols, particularly in IGP and  
565 CI, led to higher uptake of HO<sub>2</sub>, reducing its availability for ozone production and hence suppressing ozone levels  
566 by about 30–40% when compared to the surface ozone levels in 2018 and 2022. However, during monsoon and  
567 pre-monsoon seasons, reduced aerosol surface area and PM concentrations enhance photochemical ozone  
568 formation, as the decrease in aerosol surface area allows more HO<sub>2</sub> to participate in ozone production. The shift  
569 from an aerosol-inhibited regime to a NO<sub>x</sub>-limited regime in 2022, due to lower aerosol pollution compared to  
570 2018, resulted in an increase in ozone levels in several regions, especially in the IGP contributing to an increase  
571 in its levels by about 20–30 µg/m<sup>3</sup>. This suggests that the balance between aerosols, NO<sub>x</sub> and VOCs dictates ozone  
572 formation, with aerosol concentrations continuing to significantly influence ozone levels during these seasons.  
573 Nevertheless, local emissions from vehicular traffic, regional transportation, industrial operations, household  
574 sources and agricultural residue combustion substantially control PM<sub>10</sub> and ozone levels, highlighting the  
575 necessity for thorough, region-specific emission control measures. **Furthermore, scaling down the NO<sub>x</sub> emissions  
576 in the model simulations by 25 and 50% shows a spatial variability in the reduction of surface ozone, with  
577 decreases of about 5–10 and 10–15 µg/m<sup>3</sup> across India, respectively.** Thus, this unintended increase in surface  
578 ozone due to the decrease in PM and changing meteorological conditions can be reduced by additional efforts in  
579 reducing anthropogenic NO<sub>x</sub> emissions. Therefore, this study recommends the need for integrated air quality  
580 management strategies that consider both aerosol and precursor emissions, along with regional meteorological  
581 patterns, to address ozone pollution in India effectively.

582 *Data availability.* GEOS-Chem model is available via <https://geos-chem.readthedocs.io/en/stable/> and CPCB  
583 data is available via <https://app.cpcbcr.com/ccr/>

584 *Authorship Contributions.* GSG: Writing – review & editing, Writing – original draft, Visualization, Validation,  
585 Software, Methodology, Investigation, Formal analysis, Data curation. DMW: Writing – review & editing,  
586 Writing – original draft, Supervision, Visualization, Validation, Software, Methodology, Investigation,  
587 Conceptualisation. JK: Writing – review & editing, Writing – original draft, Supervision, Visualization,  
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