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# Mechanisms and Pathways for Coordinated Control of Fine Particulate Matter and Ozone

Narendra Ojha<sup>1</sup> · Meghna Soni<sup>1,2</sup> · Manish Kumar<sup>3</sup> · Sachin S. Gunthe<sup>4,5</sup> · Ying Chen<sup>6</sup> · Tabish U. Ansari<sup>7</sup>

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## Abstract

**Purpose of Review** Fine particulate matter (PM<sub>2.5</sub>) and ground-level ozone (O<sub>3</sub>) pose a significant risk to human health. The World Health Organization (WHO) has recently revised healthy thresholds for both pollutants. The formation and evolution of PM<sub>2.5</sub> and O<sub>3</sub> are however governed by complex physical and multiphase chemical processes, and therefore, it is extremely challenging to mitigate both pollutants simultaneously. Here, we review mechanisms and discuss the science-informed pathways for effective and simultaneous mitigation of PM<sub>2.5</sub> and O<sub>3</sub>.

**Recent Findings** Global warming has led to a general increase in biogenic emissions, which can enhance the formation of O<sub>3</sub> and secondary organic aerosols. Reductions in anthropogenic emissions during the COVID-19 lockdown reduced PM<sub>2.5</sub>; however, O<sub>3</sub> was enhanced in several polluted regions. This was attributed to more intense sunlight due to low aerosol loading and non-linear response of O<sub>3</sub> to NO<sub>x</sub>. Such contrasting physical and chemical interactions hinder the formulation of a clear roadmap for clean air over such regions.

**Summary** Atmospheric chemistry including the role of biogenic emissions, aerosol-radiation interactions, boundary layer, and regional-scale transport are the key aspects that need to be carefully considered in the formulation of mitigation pathways. Therefore, a thorough understanding of the chemical effects of the emission reductions, changes in photolytic rates and boundary layer due to perturbation of solar radiation, and the effect of meteorological/seasonal changes are needed on a regional basis. Statistical emulators and machine learning approaches can aid the cumbersome process of multi-sector multi-species source attribution.

**Keywords** PM2.5-O3 co-control · Global warming · Atmospheric chemistry · COVID-19 · Multi-pollutant Multi-effect · PBLH

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## Introduction

Fine particulate matter (PM<sub>2.5</sub>) and ground-level ozone (O<sub>3</sub>) adversely impact human health, leading to premature mortalities, especially in highly polluted regions [1, 2]. High levels of O<sub>3</sub> and PM<sub>2.5</sub> also have detrimental impacts on agriculture thereby incurring major economic losses [3–5]. Besides impacting human health and crop yields, PM<sub>2.5</sub> and O<sub>3</sub> affect the Earth's radiation budget thereby altering the climate. For example, particulate matter reflects (directly or via cloud formation) or absorbs incoming solar radiation depending on its chemical composition and physical properties [6], whereas ozone is an effective greenhouse gas (GHG) that traps outgoing terrestrial radiation and thereby increases surface temperatures [7–9]. Considering the impacts on air quality, health, economy, and climate, the coordinated control of both PM<sub>2.5</sub> and O<sub>3</sub> is highly desirable. Notably, achieving several of the sustainable development goals (SDGs) set by the United Nations [10] is not possible

without mitigating air pollution including both  $\text{PM}_{2.5}$  and  $\text{O}_3$ . Several regions of the world have already been experiencing pollution levels exceeding the air quality standards set by the World Health Organization (WHO). Recently revised standards [11, 12] for  $\text{PM}_{2.5}$  (24-h mean:  $15 \mu\text{g m}^{-3}$ ) and  $\text{O}_3$  (8-h mean:  $100 \mu\text{g m}^{-3}$ ) require even more stringent interventions to mitigate air pollution.

Anthropogenically sourced  $\text{PM}_{2.5}$ , besides having primary emissions from the combustion of fossil and biomass-based fuels, is also formed through reactions of inorganics (e.g.  $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{NH}_3$ , etc.) and volatile-organic precursors. Ozone ( $\text{O}_3$ ) on the other hand does not have direct emission and is formed in the atmosphere through a complex set of chemistry involving the oxidation of carbon monoxide (CO), methane ( $\text{CH}_4$ ), and volatile organic compounds (VOCs) and photolytic dissociation of  $\text{NO}_2$  to NO. In presence of  $\text{O}_3$ , higher concentrations of  $\text{NO}_x$  or  $\text{SO}_2$  can produce nitrate or sulphate and thereby enhance the  $\text{PM}_{2.5}$  concentrations in the atmosphere [13, 14].

The dependence of  $\text{O}_3$  on its precursors is highly non-linear and reductions in some precursors (which are also precursors for particulate matter) may enhance  $\text{O}_3$  pollution [15, 16].  $\text{PM}_{2.5}$ , with an atmospheric lifetime of about a week, can readily be transported over local to regional scales while surface  $\text{O}_3$  (with a lifetime of more than a month) can be transported over longer, inter-continental, and hemispheric scales depending on the prevailing meteorological conditions [17]. Historically, the problems of elevated levels of  $\text{PM}_{2.5}$  and  $\text{O}_3$  have been confined to specific seasons [18]. In general, wintertime stagnation and meteorological inversions cause severe haze contributed by  $\text{PM}_{2.5}$  [19–21], whereas, in contrast, during hot summer conditions, the intense solar radiation favours the photochemical formation of ozone. However, in addition to seasonal trends, several major cities around the globe experience higher levels of both  $\text{PM}_{2.5}$  and  $\text{O}_3$  in spring as well as summer [22]. Additionally, due to the aforementioned roles of chemistry and atmospheric dynamics, air pollution is no longer confined to highly urbanised regions but is being experienced in rural and remote locations as well [23, 24]. Higher  $\text{PM}_{2.5}$  levels are seen in parts of South Asia, East Asia, the Middle East, Southeast Asia, and Africa regions throughout the year [25]. However, the widespread enhancements are most pronounced typically during winter over the Indo-Gangetic Plain (IGP) and the North-China Plain (NCP) [21]. The studies show higher  $\text{NO}_2$  levels ( $\geq 16$  ppbv) generally near the source regions, i.e. over the IGP, eastern China and North America, whereas  $\text{O}_3$ -rich airmasses are seen to be distributed more uniformly also over remote mountains and oceanic regions [17, 24, 26, 27].

Dramatic reductions in aerosol loadings can allow more sunlight to reach near the Earth's surface resulting in more ozone production [28]. Heterogeneous losses of  $\text{O}_3$  and precursors are also lower due to less aerosol surface area

under lower  $\text{PM}_{2.5}$  conditions, which can also contribute to the enhanced  $\text{O}_3$  pollution [29, 30]. Such enhancements in ground-level  $\text{O}_3$  were profound during the recent socio-economic slowdown caused by the COVID-19 (coronavirus disease-2019) lockdown restrictions [31]. Furthermore, from an air quality perspective, both local/regional and inter-continental sources impose additional burden on pollution episodes during unfavourable meteorological conditions [21, 32, 33]. The winter-time stagnant conditions substantially increase the severity of haze episodes, whereas the heatwaves during summer lead to excess biogenic emissions of VOCs with escalated chemistry of tropospheric  $\text{O}_3$  production [34].

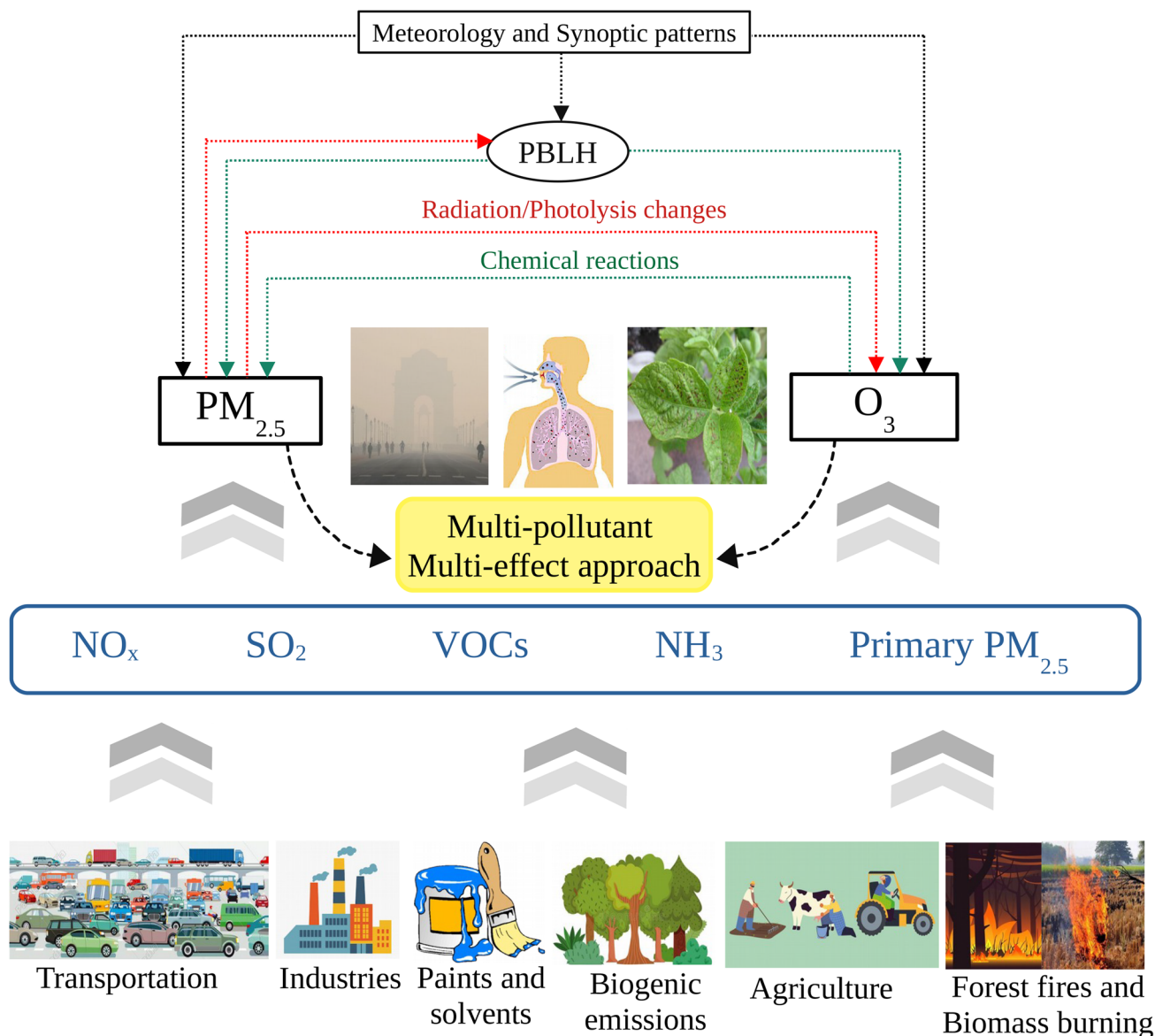
Figure 1 highlights the key connections of  $\text{PM}_{2.5}$  and  $\text{O}_3$  concentrations with emissions and meteorology. An increase in  $\text{PM}_{2.5}$  reduces incoming solar radiation and photolysis rates thereby reducing  $\text{O}_3$  production. This decrease in incoming solar radiation also reduces surface temperature leading to a general decrease in planetary boundary layer height (PBLH). Reduced PBLH means decreased turbulent mixing of all pollutants in the boundary layer which leads to increased surface concentrations of both  $\text{PM}_{2.5}$  and  $\text{O}_3$ . Daily, seasonal, and long-term variations in the PBLH, as well as levels of pollutants, are strongly governed by synoptic and seasonal weather patterns. For example, substantial build-up in  $\text{PM}_{2.5}$  levels corresponding to reduced BLH due to synoptic forcing is seen in several megacities in NCP, Sichuan Basin, and Central China [35]. On a seasonal basis, severe  $\text{PM}_{2.5}$  pollution is evident during winters when the average PBLH is lowest [25, 36].

Designing science-driven strategies to simultaneously reduce  $\text{PM}_{2.5}$  and  $\text{O}_3$  is a win–win solution toward clean air and climate change mitigation. Nevertheless, highly complex and non-linear chemical processes and atmospheric dynamics make it very challenging to pinpoint the driving sources/mechanisms over local-to-regional scales. In recent years, the availability of the state-of-the-art ground-based instrumentation [37, 38], more reliable satellite retrievals [39, 40], and models accounting for the detailed atmospheric chemistry [41, 42] have provided valuable insights into the atmospheric budgets of  $\text{PM}_{2.5}$  and  $\text{O}_3$ .

Here, we review the latest state-of-the-science of  $\text{PM}_{2.5}$ - $\text{O}_3$  interactions to inform strategies for their co-reduction, leading to improved air quality and mitigating the health impacts.

## Fine Particulates ( $\text{PM}_{2.5}$ ): Importance, Processes, and Reduction Challenges

The fine airborne particulate matter with an aerodynamic diameter of  $2.5 \mu\text{m}$  or less ( $\text{PM}_{2.5}$ ) is a leading climate and environmental health risk factor. These tiny solid or liquid airborne particles, ranging from a few nanometres



**Fig. 1** A summary of key processes governing the variations in O<sub>3</sub> and PM<sub>2.5</sub> in the atmosphere. Red lines denote decreasing effects with an increase in PM<sub>2.5</sub> while green lines denote increasing effects

to several micrometres in size, influence the energy budget of the Earth [43, 44] and boundary layer dynamics through scattering and absorption of solar radiation. Further, aerosols act as cloud condensation nuclei (CCN) and can increase or decrease rainfall thus modifying the hydrologic cycle [45, 46]. Being emitted from a variety of sources and with multiple societal impacts, PM<sub>2.5</sub> is a key target under the United Nations (UN) Agenda 2030. The motivation behind such expedited moves toward PM<sub>2.5</sub> reduction is considering its impacts on climate, health, and economy. Recent estimates of the World Health Organization (WHO) suggest that the PM<sub>2.5</sub>-induced annual global mortality is as high as 4.1 million in 2016. The major causes of mortalities from PM<sub>2.5</sub>

include stress on the cardiovascular and respiratory organ systems. The cases of morbidity and mortality are seen especially higher (>90%) in developing countries [47].

A recent synthesis from the State of Global Air [48] suggests an explicit geographical heterogeneity has been observed in the impacts of PM<sub>2.5</sub> on health and mortality patterns showing the highest burden over Asia and Africa. The combination of dense population and poor air quality has led to higher pre-mature mortalities in China and India. The episodic nature of air pollution in some regions of China additionally elevates the health risks associated with PM<sub>2.5</sub> [49]. Steep enhancement in PM<sub>2.5</sub>-induced mortality is also predicted for sub-Saharan, North Africa, and the Middle

East [48]. The burden of diseases has high economic costs associated with hospitalisation, loss of working days, treatment costs, and premature mortalities. The potential increase in  $PM_{2.5}$  and pollutants like tropospheric ozone is projected to raise the global healthcare costs from USD 21 billion in 2015 to USD 176 billion in 2060 [50].

The  $PM_{2.5}$  composition is chiefly constituted by inorganic ions, carbonaceous compounds, and some fractions of mineral dust. Besides direct emissions through,  $PM_{2.5}$  is also formed in the atmosphere via chemical reactions involving precursors through gas-to-particle conversion [51]. New particle formation (NPF) via gas-to-particle conversion can occur under diverse environmental conditions such as urban locations, forest areas, marine/coastal regions, and the remote or free troposphere [52]. A recent study has suggested a major role of NPF (~65% of the number concentration of haze particles from NPF) in causing present-day haze events in Beijing [53]. Among various sources, solid-fuel combustion (mostly coal) constitutes nearly 27% of the total global  $PM_{2.5}$  burden, eliminating which can help in avoiding 1.05 million deaths annually [54]. In the last decade, about 55% of the global population was exposed to higher levels of  $PM_{2.5}$  with varying spatial concentrations and impacts across the world. A decline in the population-weighted  $PM_{2.5}$  concentration from 12.4 to 9.8  $\mu\text{g m}^{-3}$  was observed over North America and Europe but a sharp increase from 54.8 to 61.5  $\mu\text{g m}^{-3}$  was noticed over Central and Southern Asia [55, 56]. Among major megacities in the world, Toronto (Canada), Miami and New York (USA), and Madrid (Spain) were identified among the least polluted with concentrations ranging between 7 and 10  $\mu\text{g m}^{-3}$ , while Delhi (India), Cairo (Egypt), Xi'an, Tianjin, and Chengdu (China) had experienced highest annual average  $PM_{2.5}$  (89–143  $\mu\text{g m}^{-3}$ ) [57]. A systematic time-series analysis [58] found population, urban ratio, and vegetation greenness as key socio-economic drivers of  $PM_{2.5}$ . Studies have revealed that the effects of different sources on human health can be different since all the components of  $PM_{2.5}$  are not equally toxic [59]. Out of all major sources such as biomass burning, diesel vehicles, and dust to  $PM_{2.5}$ , biomass burning was positively co-related with respiratory illness in the St. Louis metropolis [60]. Besides PM mass, nowadays,  $PM_{2.5}$  oxidative potential (OP) is considered a metric to understand PM toxicity [59]. The largest contribution to the cellular OP was from SOA (> 54%) in the urban region but from agricultural activities (62%) in the rural region of the mid-west US. However, these sources which were the largest contributors to the cellular OP were not the major contributors ( $\leq 21\%$ ) to the  $PM_{2.5}$  mass.

An econometric study of China suggests positive associations between  $PM_{2.5}$  concentrations and the size of the urban sprawl, population density, share of industry, and developmental indices like gross developmental product (GDP).

The urban sprawl and increased economic change have resulted in enhanced  $PM_{2.5}$  emissions over a large region of China [61]. An interesting observation was made that the expansion of urban set-ups in developed countries results in the reduction in  $PM_{2.5}$  while it increases the  $PM_{2.5}$  load in developing countries [58]. Such patterns in the developing world have many climatic implications, particularly over the tropical and Northern Hemisphere mid-latitude regions due to a significant drop in the wet deposition associated with less large-scale precipitation over land [62]. Although with increased awareness and stringent policy implementations, steep declines in  $PM_{2.5}$  are being recognised in some of the emission hotspots in Asia. An emerging dipole has been observed in the columnar aerosol loading with decreasing patterns over China while increasing over India [63, 64].

To tackle the impacts of  $PM_{2.5}$  on the climate, health, and socio-economic sustainability, worldwide efforts have been observed toward the reduction of ambient concentration as well as minimising human exposure. The U.S. Clean Air Act, the European Union Clean Air Policy, the Action Plan on Prevention and Control of Air Pollution (APPCAP) in China [15], and the Indian National Clean Air Program (NCAP) are some examples of major policy-oriented efforts. Even with the unequivocal global consensus on reducing  $PM_{2.5}$  reductions, the spatiotemporal heterogeneity in their sources and complex formation chemistry makes it a cumbersome task. The total mass concentration and chemical composition of  $PM_{2.5}$  depend on the local meteorological conditions and the nature and strength of emission sources. Moreover, air pollution is a transboundary phenomenon and does not adhere to geopolitical boundaries. Developing a precise understanding of the spatio-temporal trends of  $PM_{2.5}$  and its drivers and chemical and meteorological interactions of its precursors is hence of utmost importance before aiming to design effective mitigation measures.

## Ozone: Importance, Processes, and Control Pathways

Higher concentrations of  $O_3$  pose adverse impacts on human health including respiratory illnesses such as asthma exacerbation, chronic obstructive pulmonary disease (COPD), lung and cardiovascular diseases, etc [65]. Furthermore,  $O_3$  damages terrestrial vegetation by reducing photosynthesis, altering carbon allocation, and impairing stomatal function causing visible foliar injury and disturbing whole-plant level responses [66, 67]. Besides tropospheric ozone's role as a criteria pollutant, it is also a potent greenhouse gas which significantly contributes to global warming [11]. The photochemical reactions involving VOCs, CO, and methane ( $CH_4$ ) in presence of  $NO_x$  result in the production of  $O_3$  [6]. The efficiency of  $O_3$  production depends on the reactivity of VOC with hydroxyl



radical (OH) and the ratio of VOC/NO<sub>x</sub> concentration [6]. O<sub>3</sub> is lost through chemical pathways involving its reactions with water vapour (after photolysis), radicals (HO, HO<sub>2</sub>, NO), and through dry deposition at the surface. Besides gas-phase sinks, heterogeneous reactions of O<sub>3</sub> with aerosols contribute to O<sub>3</sub>-sink processes [68]. The global and regional scale meteorology (e.g. temperature, relative humidity (RH), wind speed and direction, and precipitation) affects the chemical conditions for O<sub>3</sub> production.

Recent investigations revealed a positive correlation between O<sub>3</sub> and temperature, driven by faster chemistry and enhanced natural emissions, e.g. isoprene [69, 70]. Such deterioration of air quality due to global warming—even without an increase in anthropogenic emissions—is considered the “climate penalty” [71]. Therefore, efforts to reduce anthropogenic emissions for controlling O<sub>3</sub> pollution could be offset by the projected rise in global surface temperatures in the future. In addition, enhanced biogenic VOCs due to higher temperatures can also increase the production of secondary organic aerosols [72]. This increase in aerosols may increase cloud condensation nuclei (CCN) and affect cloud albedo [72]. Such feedback, starting with climate warming (higher temperatures) either intensifies or reduces the warming depending on various atmospheric factors such as meteorology, location, topography, properties of aerosols, and concentration of reactive trace gases in the region. Stronger NO<sub>x</sub> emissions through soil microbes have been observed to contribute nearly half of the O<sub>3</sub> increase in a rural site in the USA with increasing temperature [73]. Additionally, meteorological variability could affect O<sub>3</sub> production by modulating NO<sub>x</sub> emissions by lightning [74], wildfire emissions [75], and methane emissions from wetlands [76] and shale gas [77].

The multiphase processes associated with the emissions of reactive halogen species also alter the ground-level O<sub>3</sub> chemistry [65]. Over the oceans, the halogen species (mainly involving iodine and bromine) catalytically react with O<sub>3</sub> resulting in lower O<sub>3</sub> levels [78, 79]. On the contrary, in rural-urban environments of as northern China and India, chlorine-related species are reported to accelerate the oxidation of several VOCs resulting in the enhancement of O<sub>3</sub> [80, 81]. Additionally, the oxidation of VOCs by chlorine radicals could be a potent source of secondary organic aerosol formation [82]. Therefore, it is suggested that reducing chlorine emissions along with reducing VOCs and NO<sub>x</sub> could be beneficial in reducing O<sub>3</sub> as well as SOA formation and thus reducing PM<sub>2.5</sub>.

## Challenges and Mitigation Pathways for Co-controlling PM<sub>2.5</sub> and O<sub>3</sub>

The effects of particulate-precursor chemistry must be carefully considered for designing an effective policy for PM<sub>2.5</sub> and O<sub>3</sub> control. The relative reduction of fine particles and

the reduced rate of hydroperoxyl radical removal help in the escalation of ozone formation [83]. Further, attempts to reduce oxides of nitrogen (NO<sub>x</sub>) can exacerbate the ambient O<sub>3</sub> pollution in urban regions, while equally controlling NO<sub>x</sub> and volatile organic compounds (VOCs) may have minimal effect on air quality improvement.

In view of highly complex atmospheric chemistry, Xiang et al. [84] suggested a stepwise strategy with focussing first on VOCs and then on NO<sub>x</sub>. Such attempts have resulted in the reduction of anthropogenic VOC emissions by ~60% and NO<sub>x</sub> emissions by ~20% in the first stage, while the removal of the rest of the VOCs and NO<sub>x</sub> emissions in the second stage in China. Several South Asian urban centres also experience the influence of local traffic emissions and regionally transported pollution affecting the ambient concentrations of PM<sub>2.5</sub> and O<sub>3</sub>. In such environments, only reducing the local particulate emissions will not be effective in controlling the overall air pollution. A study over Delhi [42] showed that reducing the regional emissions along with the local emissions can significantly reduce the local PM<sub>2.5</sub> by 25–30% without increasing O<sub>3</sub> over Delhi.

To address the PM<sub>2.5</sub> burden with O<sub>3</sub> in highly polluted regions, global efforts involving both developed and developing megacities are needed. Tropospheric Ozone Assessment Report (TOAR; <https://igacproject.org/activities/TOAR>) and Monitoring, Analysis, and Prediction of Air Quality (MAP-AQ; <https://igacproject.org/activities/map-aq>) are few important initiatives toward assessment and mitigation of air pollution. Further works also aiming at energy optimisation and application of advanced emission-reducing technologies globally are needed. Since PM<sub>2.5</sub> and O<sub>3</sub> have some common precursors and O<sub>3</sub> itself affects chemistry leading to PM<sub>2.5</sub> formation, a multi-pollutant multi-effect approach should be applied for co-control of O<sub>3</sub> and PM<sub>2.5</sub>. Akimoto et al. [85] recommended a stepwise approach in which first the anthropogenic emissions of NO<sub>x</sub> and SO<sub>2</sub> are to be controlled simultaneously. This step will help reduce nitrate (NO<sub>3</sub><sup>-</sup>) and sulphate (SO<sub>4</sub><sup>2-</sup>), the key constituents of inorganic PM<sub>2.5</sub>. As a precursor of O<sub>3</sub>, reducing NO<sub>x</sub> will also reduce O<sub>3</sub> in NO<sub>x</sub>-limited environments. Additionally, VOCs may get reduced when common sources of NO<sub>x</sub> and SO<sub>2</sub> (e.g. fossil fuel burning) are controlled. In the next step, NO<sub>x</sub> and VOC emissions should be reduced simultaneously to directly reduce O<sub>3</sub> significantly. This will also reduce secondary PM<sub>2.5</sub> since both NO<sub>x</sub> and VOCs are important sources of secondary aerosols. This strategy can reduce PM<sub>2.5</sub> and O<sub>3</sub> simultaneously as it focuses on preferential control of the secondary pollutants. Few studies reported that NO<sub>x</sub> reductions in the first step may exacerbate the O<sub>3</sub> pollution in polluted urban environments. For example, Xiang et al. [84] targeted the precursors contributing to both O<sub>3</sub> and PM<sub>2.5</sub> precursors, i.e. NO<sub>x</sub> and VOCs, over the Beijing-Tianjin-Hebei region and observed that

reducing  $\text{NO}_x$  enhances  $\text{O}_3$  due to VOC-limited chemistry [86, 87], especially where  $\text{O}_3$  pollution is severe. However, reducing  $\text{NO}_x$  and VOC emissions together led to small improvements in air quality. In this view, they proposed a “first VOC-focused, then  $\text{NO}_x$ -focused” strategy. In the first step, reducing anthropogenic VOC emissions by 60% and  $\text{NO}_x$  emissions by 20% and in the next step reducing more VOCs and  $\text{NO}_x$ . This policy was suggested to control both  $\text{PM}_{2.5}$  and  $\text{O}_3$  pollution to a greater extent.

An analysis over Delhi [42] showed that local traffic emissions from within the city and transport of pollution from the National Capital Region (NCR) surrounding Delhi were the dominant factors influencing  $\text{PM}_{2.5}$  and  $\text{O}_3$ . Reducing traffic emissions by 50% in Delhi alone reduces  $\text{PM}_{2.5}$  by 15–20% but increases  $\text{O}_3$  by 20–25%. Reducing domestic emissions can decrease  $\text{PM}_{2.5}$ , although lesser than the reduction achieved by reducing traffic emissions but without increasing  $\text{O}_3$ . This could be possible since domestic emissions might not be a major source of  $\text{NO}_x$  as traffic emissions. Moreover, VOCs are reduced more than  $\text{NO}_x$  when controlling domestic emissions i.e. VOC/ $\text{NO}_x$  ratio was 1.8 in contrast to a ratio of 0.4 for traffic emissions. Greater reductions of VOCs suppress the increase in  $\text{O}_3$  in Delhi, which is a VOC-limited environment [88–90]. However, the  $\text{O}_3$  formation regime may vary for different sites within Delhi depending on VOCs/ $\text{NO}_x$  [28, 91]. Reducing NCR regional emissions at the same time reducing traffic emissions in Delhi by 25–30% would further reduce the  $\text{PM}_{2.5}$  in Delhi by 5–10% and avoid an  $\text{O}_3$  increase [42].

Therefore, reducing VOC and  $\text{NO}_x$  simultaneously with a larger reduction in VOCs compared to  $\text{NO}_x$  can be an effective step to co-mitigate the  $\text{PM}_{2.5}$  and  $\text{O}_3$ . Conclusively, reducing common sources of  $\text{PM}_{2.5}$  and  $\text{O}_3$  will help reduce both together. Therefore, studies with more detailed information on specific emission sectors and their contribution to the polluted regions are required for effective mitigation policies. Since modelling of sector-specific and species-specific source contributions is computationally expensive, statistically trained reduced-form models as well as machine learning-based models can be used for exploring potentials of multi-sector and multi-species emission reductions [92, 93].

A holistic model evaluation approach is required to develop a correct understanding of the co-control of  $\text{PM}_{2.5}$  and  $\text{O}_3$ . Model simulations with reduced  $\text{NO}_x$  and VOC emissions for  $\text{O}_3$  control should evaluate not only the modelled  $\text{O}_3$  but also the modelled  $\text{NO}_x$  and VOC against observations. This highlights the need for continuous monitoring especially covering the periods when emissions were also reduced in reality. Since reducing  $\text{NO}_x$  and VOCs will also reduce secondary  $\text{PM}_{2.5}$ , therefore, in addition to validating the simulated  $\text{PM}_{2.5}$  against the observations, several individual SOA should also be validated—this requires

chemically-resolved monitoring of aerosol components. Additionally, quantification of the primary and secondary fractions of the  $\text{PM}_{2.5}$  burden for different regions is highly recommended. The reduction of primary aerosols is relatively straightforward and can ensure rapid improvement of air quality depending on its contribution to the total  $\text{PM}_{2.5}$  mass.

## COVID-19 Lockdown: a Reality Check

Despite large emission reductions during the COVID-19 lockdown, unexpected changes in  $\text{O}_3$  and  $\text{PM}_{2.5}$  were experienced in different regions of the world (Table 1). Mean  $\text{PM}_{2.5}$  levels generally showed reductions by about 30–40% but dust transport, biomass burning, and secondary PM formation caused enhancements in some parts of Europe and Asia [94, 95].  $\text{O}_3$  exhibited diverse trends with insignificant change or slight increase over Europe but 25–30% higher levels over East Asia and South America [94]. Enhanced relative humidity by 30–50% and reduced wind speed under lower PBL height during winter promoted the multiphase chemistry which enhanced SOA levels during lockdown in China [96]. Additionally, the reduced  $\text{NO}_x$  contributed to the enhancement in  $\text{O}_3$  levels due to non-linear chemistry. The higher  $\text{O}_3$  levels increased the atmospheric oxidising capacity and facilitated enhanced SOA and  $\text{SO}_4$  formation and further contributed to  $\text{PM}_{2.5}$  loading [96]. While there was a significant reduction in  $\text{PM}_{2.5}$  (13–29%) as well as  $\text{NO}_2$  (39–53%),  $\text{O}_3$  levels were found to be higher by a factor of 1.6–2 in northern China [97, 98]. In the USA, changes in  $\text{NO}_2$  from 5 to 49% are mainly due to lower transportation and utility demands [99]. Significant reductions (up to 45%) in  $\text{PM}_{2.5}$  were seen in Northeast and California/Nevada metropolises, where  $\text{NO}_2$  declined strongly. Minor changes (within  $\pm 20\%$ ) in  $\text{O}_3$  concentration were seen. The Sao Paulo region in Brazil experienced extreme reductions in  $\text{NO}_x$  (~54–77%), and  $\text{PM}_{2.5}$  (~29.8%) but ~30% increase in the  $\text{O}_3$  concentrations, compared to the 5-year monthly mean [100]. Here, lesser changes in the levels of pollutants were observed in the industrial areas, as industries were not restricted to shut down, but they were partially affected by the decreased demand.

Satellite-based observations revealed up to 50% reductions in boundary layer  $\text{NO}_2$  over urban areas of Europe [101]. The aircraft-based measurements under the BLUESKY campaign showed large variabilities in reactive nitrogen and aerosols (20–70%) over German cities during the lockdown as compared to multi-year average data [101]. In situ measurements across the UK also showed reductions in  $\text{NO}_2$  (up to 48%) but a slight increase in  $\text{O}_3$  by ~11% [102]. In some other European sites, the  $\text{NO}_x$  and  $\text{PM}_{2.5}$  concentrations were observed to be reduced by 50–71% and

**Table 1** A list of some studies from different regions of the world describing the changes in the levels of various pollutants during the COVID-19 lockdown period

Location	Pollutants		
	NO <sub>2</sub> (% reduction)	PM <sub>2.5</sub> (% change)	O <sub>3</sub> (% change)
Global [94, 95]	48–72	17–45	–2 to 10
North China [97, 110]	39–53	13–39	100–160
USA [99]	5–49	–45 to 50	±20
Sao Paulo, Brazil [100]	54–77	29.8	30
UK [102]	41–48	–	11
Spain [104]	50–69	13–50	2.4–50
Nice, France [103]	63	3	24
Delhi [105, 106]	40–60	40–60	20
Ahmedabad [107]	43–55	–	41
Kolkata [108]	–	20–66	11–91
Thiruvananthapuram [109]	40	–	–36

3–50%, respectively, while O<sub>3</sub> increased by ~2.4–50% [103, 104]. Such reductions were also attributed to the restrictions on population mobility leading to reduced road traffic, and industrial operations.

The reductions in some of the key air pollutants were also pronounced over India during the COVID-19 lockdown exhibiting a sharp decline in PM<sub>2.5</sub> (~25–60%) and mixed changes in O<sub>3</sub> across different regions [105, 106]. In Delhi, O<sub>3</sub> concentrations showed more or less site-specific trends with a mean increase of ~20%. This increase could be attributed to the significant reductions (~40–60%) in NO<sub>2</sub>, PM<sub>10</sub>, and PM<sub>2.5</sub> levels [106], which enhanced photochemical O<sub>3</sub> production via non-linear chemistry and more solar radiation in conditions of lower aerosol burden. Ahmedabad, an urban hotspot in western India experienced ~41% enhancement in O<sub>3</sub> levels in contrast to the reduction in NO<sub>x</sub> (~43–55%), which was attributed to chemistry (25%) and meteorology (16%) [107]. Here, enhancement in NO<sub>2</sub>/NO ratio during the lockdown (3.3) in comparison to the pre-lockdown period (2.6) results in the lesser titration of O<sub>3</sub> as during the lockdown period [107]. Kolkata over the eastern region of the Indo-Gangetic Plain showed a decrease in PM<sub>2.5</sub> levels by 20–66%, but an increase in O<sub>3</sub> levels by 11–91% during five different phases of lockdown [108]. In contrast, Thiruvananthapuram, a tropical coastal site in India experienced reduced daytime O<sub>3</sub> (~36%) accompanied by reduced daytime NO<sub>2</sub> (~40%) [109]. Similar reductions in the O<sub>3</sub> were also seen in the south and central regions of India [106]. Both PM and NO<sub>2</sub> show a strong reduction due to less fuel consumption in various economic sectors i.e. transport (50–60%), aviation (90%), industries (40%), and construction activities (70%), even with a 12% increase in household fuel consumption, while O<sub>3</sub> was increased in the populated areas of India [111].

Heterogeneities in the levels of pollutants were attributed to the complex air chemistry, meteorology, and episodic events such as dust, biomass burning, and crop fertilising [94, 95]. Such insights from the societal slowdown

and reduced anthropogenic activities during the COVID-19 restrictions, albeit not intended for co-mitigation of PM<sub>2.5</sub> and O<sub>3</sub> but for containing the pandemic, suggest the achievable improvements in the air quality by curbing major emissions, which were not economically and socially viable otherwise.

## Conclusion

The challenges to reducing air pollution greatly depend on the sources of specific pollutants, the effects of atmospheric dynamics and complex chemical interactions under various meteorological conditions. Exposure to both PM<sub>2.5</sub> and ground-level O<sub>3</sub> has been reported to have deleterious impacts on human and plant health and the built environment. Hence, a strong scientific consensus is building globally for co-controlling both PM<sub>2.5</sub> and ground-level O<sub>3</sub> for achieving several overarching air quality benefits. The complex interplay between the reduction in the hydroperoxyl radical removal helps in the escalation of ozone formation while efforts to reduce NO<sub>x</sub> exacerbate ambient O<sub>3</sub> (in several polluted urban environments). Further, equally controlling NO<sub>x</sub> and VOCs results in the marginal improvement of air quality. PM<sub>2.5</sub> and O<sub>3</sub> have some common precursors; therefore, a multi-pollutant multi-effect approach to co-controlling them is urgently needed including a stepwise approach for controlling NO<sub>x</sub> and SO<sub>2</sub> emissions simultaneously from the anthropogenic sources first, which will lower down NO<sub>3</sub><sup>–</sup> and SO<sub>4</sub><sup>2–</sup> while as a precursor of O<sub>3</sub>, reducing NO<sub>x</sub> will further reduce the levels of O<sub>3</sub>. The societal slowdown during COVID-19 with consequent lowered energy consumption provided a natural experiment and evidence for orienting future research on co-control of both pollutants. Further, it highlighted a strong prerequisite for further scientific investigation to understand the PM<sub>2.5</sub>-O<sub>3</sub> interactions over the polluted regions.



## Compliance with Ethical Standards

**Conflict of Interest** On behalf of all authors, the corresponding authors state that there is no conflict of interest.

**Human and Animal Rights and Informed Consent** This article does not contain any studies with human or animal subjects performed by any of the authors.

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