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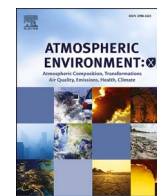
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Air pollution scenario analyses of fleet replacement strategies to accomplish reductions in criteria air pollutants and 74 VOCs over India

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ABSTRACT

Traffic emissions are a major source of air pollution and associated damage to human health in India. Many of the Indian metro cities urgently require cleaner transportation technologies to ensure cleaner air. Here, using newly compiled spatially disaggregated, gridded, high-resolution ($0.1^\circ \times 0.1^\circ$) road transport emission inventory for India for 2030 (RTEII) of 74 speciated VOCs, CO, SO₂, NO_x, NH₃, CH₄, CO₂, BC, OC and PM_{2.5} from varied fuels and vehicle technologies that are currently in use in India, we investigated changes in emission in response to substitution of the existing vehicular fleet by cleaner alternatives. Three “what-if” intervention scenarios were considered to assess the extent in improvement of air quality due to the reduction in the primary emission of air pollutants. The results show that significant reductions in direct emission of pollutants (Non-Methane VOCs, –91%; CO, –80%; PM_{2.5}, 44%) including toxic VOCs (e.g., isocyanic acid, –76%; BTEX, –93%; as well as individual VOC classes (e.g., sum of OVOCs, –61% and sum of alkenes, –80%) can likely be achieved in 2030 by shifting from highly polluting Internal Combustion Engine (ICE) based 2 and 3-wheeled vehicles to Electric Vehicles (EVs) under scenario 1. The amount of secondary pollutants such as SOA and O₃ that can potentially be formed from traffic also showed significant reduction of 94% and 84%, respectively, under scenario 1. Conversion of diesel fuelled vehicles to CNG under scenario 2 can lead to a larger reduction in black carbon emissions (–50%). Scenario 3, in which the benefits of scenarios 1 and 2 are combined, represents the best long-term strategy moving forward, which can result in massive emission reductions of pollutants through existing technologies of greener transport fleets over India. Large scale conversion of the vehicle fleets as explored here can lead to a substantial reduction of air pollution and fewer lives lost.

1. Introduction

Air pollution poses the largest environmental health threat globally, accounting for 7 million deaths around the world every year (WHO, 2014). Almost three fourth of cities in India exceed the prescribed standards of ambient air quality (CPCB, 2020; David et al., 2019). For several air pollutants, including fine particles (PM_{2.5}), emissions due to the road transport sector present the most rapidly growing urban emissions sources (Goel et al., 2015). The total number of registered motor vehicles in India was 230 million as of 31.03.2016, an increase by more than 300% when compared to the year 2001 (MORTH, 2018) with a Compound Annual Growth Rate (CAGR) of 9.6% CAGR between 2001 and 2018 against a population CAGR of less than 2% during the same period (SIAM, 2019; World Bank, 2020). The number of vehicles is expected to further grow to 649 million by 2030 under a business-as-usual

approach to the transport sector, which is best represented by the shared socioeconomic pathway 5 (SSP5) of fossil fuel based economic growth (Kriegler et al., 2017) in the SSP database. The SSP database contains other narratives for potential futures in which the growth in the future vehicle number is suppressed by extreme poverty (SSP3, Fujimori et al., 2017), income inequality (SSP4, Calvin et al., 2017) or sustainable development aspirations that result in a less individual centric transport system (SSP1, van Vuuren et al., 2017). It also includes a scenario based on past trends of the decade from the mid 1990s till the early 2000s (SSP2, Fricko et al., 2017). In this study, we focus on the SSP5 narrative, which appears to best capture current trends in the Indian transport sector.

The scientific literature estimates large numbers, ranging between 483,000 and 1,267,000 cases of premature deaths annually from outdoor pollution in India (WHO, 2016; Purohit et al., 2019), which cost the

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Indian economy US\$28.8 billion (21.4–37.4) and \$8 billion (5.9–10.3), on account of premature deaths and morbidity, respectively in 2019 (Pandey et al., 2020). This amounts to 1.36% of India's gross domestic product. These assessments are based on direct exposure to just two air pollutants namely particulate matter (PM_{2.5}) and ozone. Several VOCs are also known to be hazardous to human health (van Zelm et al., 2008; Laurent and Hauschild, 2014; Huang et al., 2011; Weng et al., 2009). For example, carbonyls can irritate human eyes and lungs (Ho et al., 2006), isocyanic acid can increase risk of cardiovascular disease and eye cataracts (Roberts et al., 2011), nitromethane and benzene are carcinogens (Durmugoglu et al., 2010; Espenship et al., 2019; WHO, 2019). Recent studies have reported that policies that target vehicle emission reductions are critical to reduce the high levels of pollutants including VOCs (Stewart et al., 2020). Current health studies and economic damage assessments fail to include direct effects of VOCs, primarily because of lack of observational data and research on dose-exposure relationships on many emerging contaminant VOCs.

To curb the pollution generated from steep rise in transport activity, in the past decade, India has promulgated several measures, such as implementation of more stringent Euro III (2005 13 cities, 2010 Nationwide) IV (2010 13 cities, 2017 Nationwide) and Euro VI (2020 Nationwide) emission norms for new vehicles over the years. Older vehicles are also being phased out in cities. Metro Rapid Transport Service (MRTS) is being expanded in cities to improve public transport and fuel portfolios are being diversified. Several cities in India have experimented with substituting diesel vehicles with alternatives that have lower air pollutant emissions such as compressed natural gas (CNG) (Wadud and Khan, 2013; Chong et al., 2014; Yao et al., 2014; Ravindra et al., 2006). These build on examples from megacities (Rio de Janeiro, Mexico City, Delhi, Patna, Mumbai, Karachi) that have successfully introduced CNG vehicles in their vehicle fleet (Wadud and Khan, 2013). In India, Delhi and Mumbai have now made CNG mandatory for 3-wheelers and buses (Kathuria, 2004).

Thus, conducting clean air intervention scenario analyses to evaluate the impacts and benefits of proposed air quality intervention measures is meaningful and urgently needed to assess the potential impacts of such measures on reducing emissions. The use of electric vehicles (EVs) can decrease concentrations and exposure to air pollutants (Tobollik et al., 2016), particularly in congested inner-cities (Jochem et al., 2016), and also decrease emissions of greenhouse gases (Electric Power Research Institute, 2007; Stephan and Sullivan, 2008; Becker et al., 2009). In Madrid and Barcelona 40% EV conversion accomplished more than 10% reduction in NO_x, while in Taiwan, 100% EV penetration along with all additional power coming from thermal power plants could reduce CO, VOCs, NO_x, O₃ and PM_{2.5} pollution by 85%, 79%, 7% (net), 39% and 7.2% respectively (Soret et al., 2014; Li et al., 2016). For India, the country with the world's third largest road transport network, no such intervention analyses have been conducted till date.

Hakkim et al., 2021 recently compiled a new (0.1° × 0.1°) road transport emission inventory over India (<https://doi.org/10.17632/n6db9gynn.1>) for 74 speciated VOCs, CO, NO_x, NH₃, CH₄, CO₂, PM_{2.5}, BC, OC, SO₂ for the year 2015. The study showed that the most polluting vehicle types in India were petrol-2 and 3-wheelers and diesel fuelled vehicles. In the current study, we present a what-if scenario analysis for the year 2030, which allows to assess how much lower the emissions will be for the same transport activity executed by a more diversified fleet when compared to a business-as-usual approach in which the existing fleet is gradually replaced with vehicles that use the same fuels but comply with newer emission norms. We studied three scenarios and provide gridded (0.1 degree × 0.1 degree) emissions for 74 VOCs and criteria air pollutants for each of these:

- 1) Scenario 1- Conversion of all petrol, diesel, LPG and CNG fuelled 2- and 3-wheel vehicles to electric vehicles
- 2) Scenario 2- Conversion of all diesel fuelled vehicles to CNG fuelled vehicles

- 3) Scenario 3- Conversion of all petrol, diesel, LPG and CNG fuelled 2 and 3-wheeled vehicles to electric vehicles and additionally conversion of diesel 4-wheelers and heavy-duty vehicles (HDV) to CNG fuelled vehicles.

We evaluate the air pollution impact of these potential fleet substitution strategies relative to business-as-usual annual emission fluxes from the road transport sector under the projected transport demand for the SSP5 fossil fuel based economic growth (Kriegler et al., 2017) for the same year, 2030, to allow well informed policy formulation.

2. Materials and methods

2.1. Compilation of emission inventory

Details of the new transport emission inventory RTEII used as a starting point in this work are described in Hakkim et al. (2021) and the data in gridded form has been provided at (<https://data.mendeley.com/datasets/n6db9gynn/draft?a=afae596d-bd9d-4e1c-aff3-eb2d41ca17d1>). Hence, the same is described only briefly here stressing on aspects relevant for the analyses presented in this work. RTEII is the first emission inventory for the road transport sector over India with detailed VOC speciation and provides spatially disaggregated, gridded, high-resolution (0.1° × 0.1°) emissions data for the year 2015. It accounts for emissions of key air pollutants including primary particulate matter (e.g., PM₁₀, PM_{2.5}, BC and OC) and gaseous precursors of secondary particulate matter (e.g., SO₂, NO_x, CO, NH₃, and VOCs) as well as greenhouse gases (CO₂ and CH₄). Annual emission for air pollutants in RTEII was calculated following the IPCC 2006 guidelines using emission factors of 74 volatile organic compounds measured from tailpipe-emissions (Hakkim et al., 2021, Table S5), all-India state-wise fuel consumption data for the year 2015–16 (MNPG, 2016), and the fraction of fuel consumed by different vehicle categories within the transport sector (Nielson, 2013). A detailed list of fuel specific emission factors and standard error of selected NMVOCs, along with trace gases used in the current study, are provided in Tables S1–S5. The fuel consumption data was then disaggregated on a gridded map of India at a resolution of 0.1° × 0.1° using population density (CESIN, 2015) and traffic density along with the road network as proxies. A grid wise digitized map of the national highway road network in India was developed using ArcGIS version 10.2 (ESRI Inc., Redlands, USA). For a particular pollutant, emission contribution due to each vehicle type was calculated per grid and summed to derive the emission distribution maps over India. In this study, using similar approach as Hakkim et al. (2021) we first compile future road transport emissions under a SSP5 shared socioeconomic pathway trajectory. SSP5 represents a rapid fossil fuel based economic growth scenario (Kriegler et al., 2017). Population forecasts for the year 2030 were taken from a report by the National Commission on Population, Ministry of Health and Family Welfare, Government of India (National Commission on Population, 2019). Spatial disaggregation of population data was accomplished by scaling the downloaded and regridded Global One-Eighth Degree Population Base Year and Projection Grids Based for the SSP 5, v1.01 (2000–2100) (Jones and O'Neill, 2016) such that the projected rural and urban population of each Indian state agreed with the projections of National Commission on Population, 2019. The latter projection is based on recently observed fertility trends and projects a marginally higher 2030 population (1465 million) than the SSP5 population projections (1457 million) in the SSP database. We use the economic projections of the IIASA model (Crespo Cuaresma, 2017), which uses age structure and educational attainments from the IIASA human core database (Samir and Lutz, 2017) to forecast 2030 per capita income. Per capita income is used to project future vehicle ownership and fuel consumption with the help of the Gompertz function. The Gompertz function (equation (1)) is widely applied to establish the relationship between vehicle ownership and per capita income (Dargay et al., 2007; Huo and Wang, 2012; Mittal

et al., 2015).

$$V_{r,t} = V^* \times e^{\alpha e^{\beta GDP_{r,t}}} \quad (1)$$

where r denotes the region and t denotes different years; $V_{r,t}$ represents the vehicle ownership/fuel consumption (vehicles/fuel consumed per 1000 people) of region r in year t ; α and β are fit parameters; $GDP_{r,t}$ is the Gross Domestic Product of region r in year t ; V^* represents the ultimate saturation level of vehicle ownership/fuel consumption (vehicles/fuel consumed per 1000 people). We calibrate the Gompertz function with state wise vehicle and fuel sales for the years 2009–2019 and use it to establish a relationship between fuel consumption and income as described in greater detail in Fig. S1 and the accompanying text. The fit parameters derived are given in Tables S6 and S7. The data so organized enabled us to calculate the emissions for each of the intervention scenarios, which were constructed under the assumption that the number of vehicle km travelled by each vehicle class in each location stays the same but the activity is carried out using a different fuel/technology type.

2.2. Calculation of secondary pollutant formation potentials and CO₂ equivalent emissions for different scenarios

Using the maximum incremental reactivity method (Carter, 1994) and secondary organic aerosol formation yields of 38 VOCs compiled from the literature, ozone formation potential (OFP) and SOA production factor (PF) for fuel and vehicle categories were calculated using equations (2) and (3) respectively.

$$OFP = \sum_i EF_i \times MIR_i \quad (2)$$

where EF_i is the emission factor and MIR_i is the ozone formation coefficient for VOC species i in the maximum increment reactions of ozone

(Carter, 1994, 2009). The MIR scale has been widely used by the US Environmental Protection Agency (EPA) and the California Air Resources Board for many decades to devise a reactivity-based ranking of VOCs for implementing regulations geared towards reduction of photochemically formed ozone from its VOC precursors and is very meaningful for inter-comparison of ozone formation tendencies.

$$PF_{SOA} = \sum_i EF_i \times Y_i \quad (3)$$

where PF_{SOA} is the secondary organic aerosol production factor, EF_i is the emission factor and Y_i are the yield of VOC species i to form SOA. Table S8 lists the SOA production factor values for individual VOCs and the original studies from which the values were taken.

CO₂ equivalent emissions were calculated using relative Global Warming Potentials (GWP₁₀₀) (Myhre et al., 2011) per equation (4).

$$Total\ CO_2eq = (28 \times [CH_4]) + [CO_2] \quad (4)$$

where $[CH_4]$ and $[CO_2]$ are the total annual emission of methane and carbon dioxide respectively for each fuel type.

2.3. Intervention scenarios

Fig. 1 shows the different what-if intervention scenarios explored in the current study. The default scenario (Fig. 1 a) represents the projected road transport emissions of the year 2030 under a SSP 5 projection. Fig. 1b–d illustrate the three intervention scenarios considered in this work:

Scenario 1: In this scenario, we consider the conversion of all petrol, diesel, LPG and CNG fuelled 2 and 3-wheeler vehicles to electric vehicles (EVs). The large population of 2 and 3-wheeled vehicles (63% in 2030), which are the top two highest polluting fleet of vehicles, accounted for a

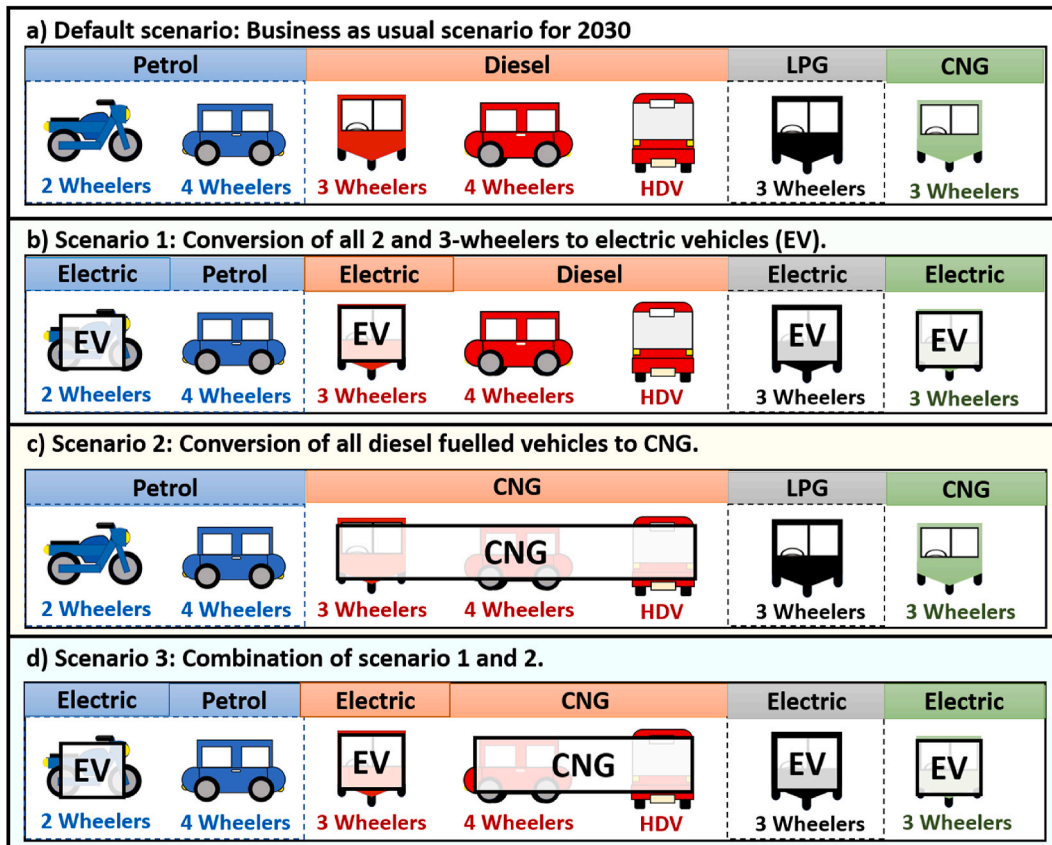


Fig. 1. The schematic illustrating the three intervention scenarios for potential emission reductions for the year 2030.

large chunk (>70%) of the annual VOCs, carbon monoxide (CO), and NO_x emissions in 2015 (Hakkim et al., 2021). Two-wheeler and three-wheeler vehicle production has been expanding rapidly over the past decade, especially in the urbanized areas of India. They offer superior fuel efficiency and play an important role in fulfilling both personal and commercial transportation needs in most Indian cities. In recent years, economically competitive emission free alternative technologies in the form of electric vehicles are becoming available to replace both the vehicle classes. For example, India has specifically targeted electric two- and three-wheelers as a segment eligible for subsidy support under its Faster Adoption and Manufacture of (Hybrid and) Electric Vehicles (FAME) scheme which began in 2015 (MHPE, 2020). As a result, electric two-wheelers are currently a particularly large and growing market segment in India with sales expanding by 138% (54800) in the fiscal year 2017-18 from the year prior (23000) (SMEV, 2018).

This shift from conventional vehicles based on internal combustion engine (ICE) can bring substantial emission reduction benefits from the transport sector and was hence chosen as potential intervention scenario. Scenario 1 also presents a good case study for assessing the reduction in combustion of fossil fuels such as petrol, diesel, CNG and LPG in congested urban environments where exposure to road traffic air pollution is the highest and 2- and 3-wheelers are a popular means of transport for daily commuting.

Scenario 2: Scenario 2 assumes the conversion of all diesel fuelled vehicles including 3-wheelers (3W), 4-wheelers (4W) and heavy-duty vehicles (HDV) by CNG fuelled vehicles. Successful implementation of electric vehicles is a long-term process that requires substantial time and a large investment. While electric two- and three-wheelers have led the first wave of adoption of electric vehicles (EVs) in India, passenger cars and heavy-duty vehicles are still not affordable due to their high total

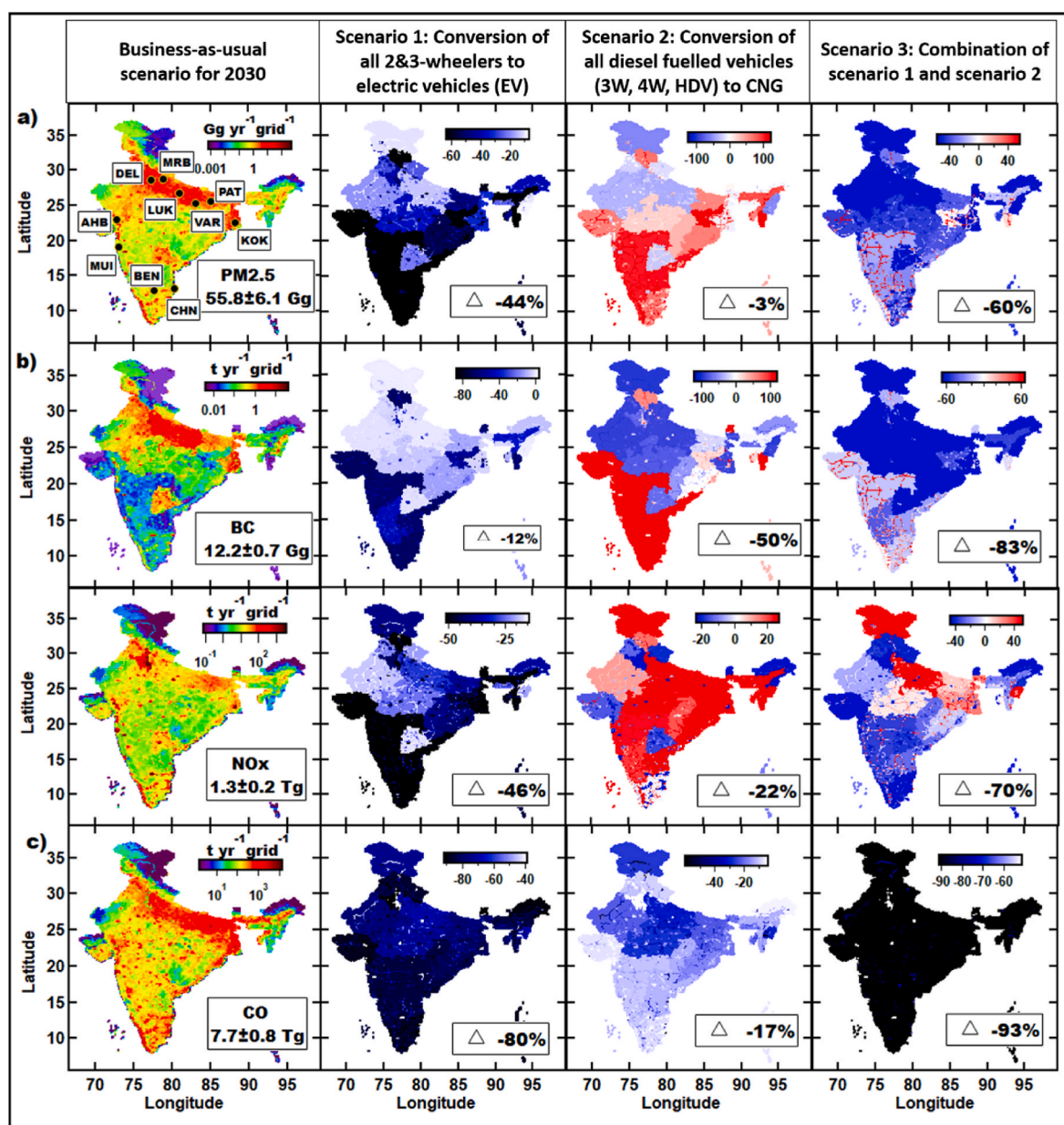


Fig. 2. Annual gridded ($0.1^\circ \times 0.1^\circ$) emissions of PM_{2.5} (panel a), BC (panel b), NO_x (panel c) and CO (panel d) for SSP5 projected transport demand in year 2030 (first column) under different intervention scenarios. The second, third and fourth column in each panel shows the percentage differences for scenarios 1, 2 and 3 relative to the first column, respectively. Markers indicate the location of major polluted cities identified by central pollution control board of India (CPCB) in which the prescribed national ambient air quality standard (NAAQS) limits are often violated (non-attainment cities). DEL – Delhi, LUK – Lucknow, PAT – Patna, VAR – Varanasi, MUI – Mumbai, KOK – Kolkata, MRB – Moradabad, AHB – Ahmedabad, BEN – Bengaluru and CHN – Chennai.

cost of ownership (TCO) (Lee et al., 2013; Lebeau et al., 2019; Kumar and Chakrabarty, 2020). This includes the cost of acquisition, running, and maintenance, which is the most important factor determining the viability of the vehicle (Feng and Figliozzi, 2013). Alternative fuels such as CNG and bio-CNG could extend sustainable transport to heavier vehicles and those seeking longer ranges, while using the existing refueling infrastructure and vehicle fleet through retrofitting. Therefore, emission projected under scenario 2 represents a short-term intervention strategy that promotes a shift from diesel and petrol fuels to an already popular alternative fuel CNG.

Scenario 3: Intervention scenario 3 considers a hybrid approach combining elements of Scenario 1 and 2 to achieve maximum emission reductions. It assumes the electrification of all petrol, diesel, LPG and CNG fuelled 2 and 3-wheeled vehicles and conversion of all the remaining diesel fuelled vehicles (4W, HDV) to CNG.

In summary, the choice of these intervention scenarios represents some of the feasible short-term, medium-term and long-term emission possibilities for the reduction of road sector air pollutant emissions over India.

3. Results and discussion

3.1. The impact of intervention scenarios on reduction of the criteria air pollutants PM_{2.5}, Black Carbon, NOx and CO

Fig. 2 shows the annual gridded ($0.1^\circ \times 0.1^\circ$) emissions of major criteria pollutants in the 2030 Road Transport Emission Inventory of India (RTEII) for SSP5 and under different intervention scenarios for potential emission reductions. It should be noted that the transport sector is considered a major source of PM, CO and NOx in all the Indian megacities contributing approximately 50–70% of total emissions (Gurjar et al., 2016). Hence a >80% emission reduction by transport sector intervention has the potential to ameliorate ambient air pollution significantly. The introduction of electric vehicles under scenario 1 can bring about a 44% and 12% reduction in PM_{2.5} (Fig. 2a) and BC (Fig. 2b) emissions, respectively indicating that electrification of 2 and 3-wheelers has the potential to reduce primary particulate emissions, primarily through the substitution of diesel 3-wheelers, which are the highest emitter of PM_{2.5} in terms of emission per litre of fuel burned (0.7 gL^{-1} ; See Table S5) under BSVI norms. In contrast, results show that PM_{2.5} emissions reductions under scenario 2, when all the diesel fuelled vehicles are replaced by their CNG variants are much lower; only 3% while BC emission reductions are higher (50%). This is not surprising because diesel 4-wheelers (4.5 gL^{-1} ; Table S3) and heavy-duty vehicles (1.5 gL^{-1} ; Table S3), which together consume 61% of the road transport fuels are currently the main contributors to PM_{2.5} (84%) emissions (Hakkim et al., 2021). They have shorter permitted vehicle lifetimes and will mainly be replaced by BSVI vehicles that contain particle filters and have lower emissions ($<0.08 \text{ gL}^{-1}$; Table S5) by 2030. Since diesel HDVs with particle filters have lower PM_{2.5} emission factors than their CNG counterparts following BSVI norms (0.14 gL^{-1} ; Table S5) the air quality gains that can be made by replacing them will be mostly restricted to BC emission reductions. Several epidemiological studies have associated diseases such as Ischemic Heart Disease (IHD), Cerebrovascular Disease (Stroke), Chronic Obstructive Pulmonary Disease (COPD), Lower Respiratory Infection (LRI), and Lung Cancer (LNC) to long-term PM_{2.5} exposure resulting in premature mortality (Huang et al., 2017; Shi et al., 2020). Roberts and Liu (2019) have investigated potential associations between estimated exposure to ambient air pollution in late childhood and mental health problems prospectively in late adolescence and reported that the cumulative effect of chronic exposure to higher estimated pollution levels of NOx and PM_{2.5} at a younger age can increase the likelihood of depression symptoms at a later age. Janssen et al. (2011) showed that a $1 \mu\text{gm}^{-3}$ decrease in PM_{2.5} exposure would lead to an increase in life expectancy of 21 days per person, whereas the same reduction in BC concentration would yield an increase between 3.1 and

4.5 months. The introduction of the BSVI emission norms, which primarily targeted diesel particulate matter emissions, will ensure that by 2030 transport sector PM_{2.5} emissions will drop by 69% from 2015 levels ($181 \pm 20 \text{ Ggy}^{-1}$; Hakkim et al., 2021) to ($56 \pm 6 \text{ Ggy}^{-1}$) despite an increase in the transport demand. This air quality improvement is much larger than what was projected by Guttikunda and Mohan (2014) for a BSV introduction in 2020. Results obtained under intervention scenario 3 thus illustrate the magnitude of the additional potential health and economic benefits to society that can be achieved by reducing particulate pollution emission and exposure through substitution of diesel vehicles by CNG or compressed biogas vehicles and the substitution of 2- and 3-wheelers with electric vehicles.

2030 transport sector NOx emission under business-usual projections ($1.3 \pm 0.2 \text{ Tgy}^{-1}$) are comparable to 2015 emissions ($1.2 \pm 0.3 \text{ Tgy}^{-1}$) as the lower NOx emissions from petrol vehicles are compensated by the increase in the vehicle fleet. Limited reductions from the baseline value (22%) was seen for the emissions projected under scenario 2 for NOx (Fig. 2c), as the NOx emissions of some vehicle types increase when diesel engines are converted to CNG (Hallquist et al., 2013; Huang et al., 2016; Suarez-Bertoa et al., 2020). By 2030 a shift of 2- and 3-wheelers to EVs can result in larger reductions in NOx emissions (46%).

Unlike the case of PM_{2.5} and BC, 2-wheeler engines are the single largest contributor to CO emissions (71%) due to the known issue of inefficient and incomplete combustion, as reported by Liu et al. (2008) and Platt et al. (2014). Carbon monoxide has 210 times greater affinity for haemoglobin than oxygen (Blumenthal, 2001; Levy, 2015). Therefore, an exposure to unsafe concentration can cause toxic levels of carboxyhaemoglobin and reduces the capacity of the blood to carry oxygen. The diesel vehicle's engine combustion process is more efficient, adding up to higher fuel efficiency and lower CO emissions (21%) than petrol. Consequently, the reduction in CO emissions was higher under scenario 1 (80%) when compared to scenario 2 (17%).

To evaluate the extent in the improvement of air quality in urban areas due to the reduction in the primary emission of criteria pollutants due to our intervention scenarios, we have selected few of the major polluted cities identified by the central pollution control board of India (CPCB) in which the prescribed national ambient air quality standard (NAAQS) limits are often violated (non-attainment cities) for illustration. This includes Delhi, Mumbai, Chennai, Kolkata, Kanpur, Lucknow, Patna, Varanasi, Bengaluru, Ahmedabad and Moradabad. In each of these cities, the BC concentrations were reduced by 50% under scenario 2, while CO emissions were reduced by 80% under scenario 1. A combination of scenarios 1 and 2 as presented by scenario 3 can reduce the annual concentrations of both particulate matter, BC as well as CO by 60%, 83% and 93%, respectively, thereby achieving maximum reduction compared to a business-as-usual scenario. Given the fact that the Indo-Gangetic Plain is responsible for nearly half (46%) of the premature mortality over India linked to air pollution (David et al., 2019) the reduction in criteria pollutants would thus lead to substantial benefits for both the health of the population and the economy over the region.

3.2. The impact of intervention scenarios on reduction of toxic and reactive VOCs

Fig. 3 shows annual gridded ($0.1^\circ \times 0.1^\circ$) emissions of isocyanic acid, BTEX, sum of oxygenated VOCs (OVOCs) and sum of the alkenes for the business-as-usual year 2030 emission inventory and under different intervention scenarios. In the base case, more than 93% of the total annual emission of BTEX compounds ($577 \pm 17 \text{ Gg y}^{-1}$) from road transport sector in India were contributed by 2-wheelers and 3-wheelers. Upon replacing them with electric vehicles (EVs) under scenario 1, the sum of BTEX compounds was reduced by 93%. A maximum reduction in the range of 93–96% was observed in toluene, xylenes and ethyl benzene annual emissions, while benzene showed a reduction of 79% in comparison to the default scenario. Replacing the diesel fuelled vehicles (3W, 4W and HDV) with CNG alternatives under scenario 2 resulted in

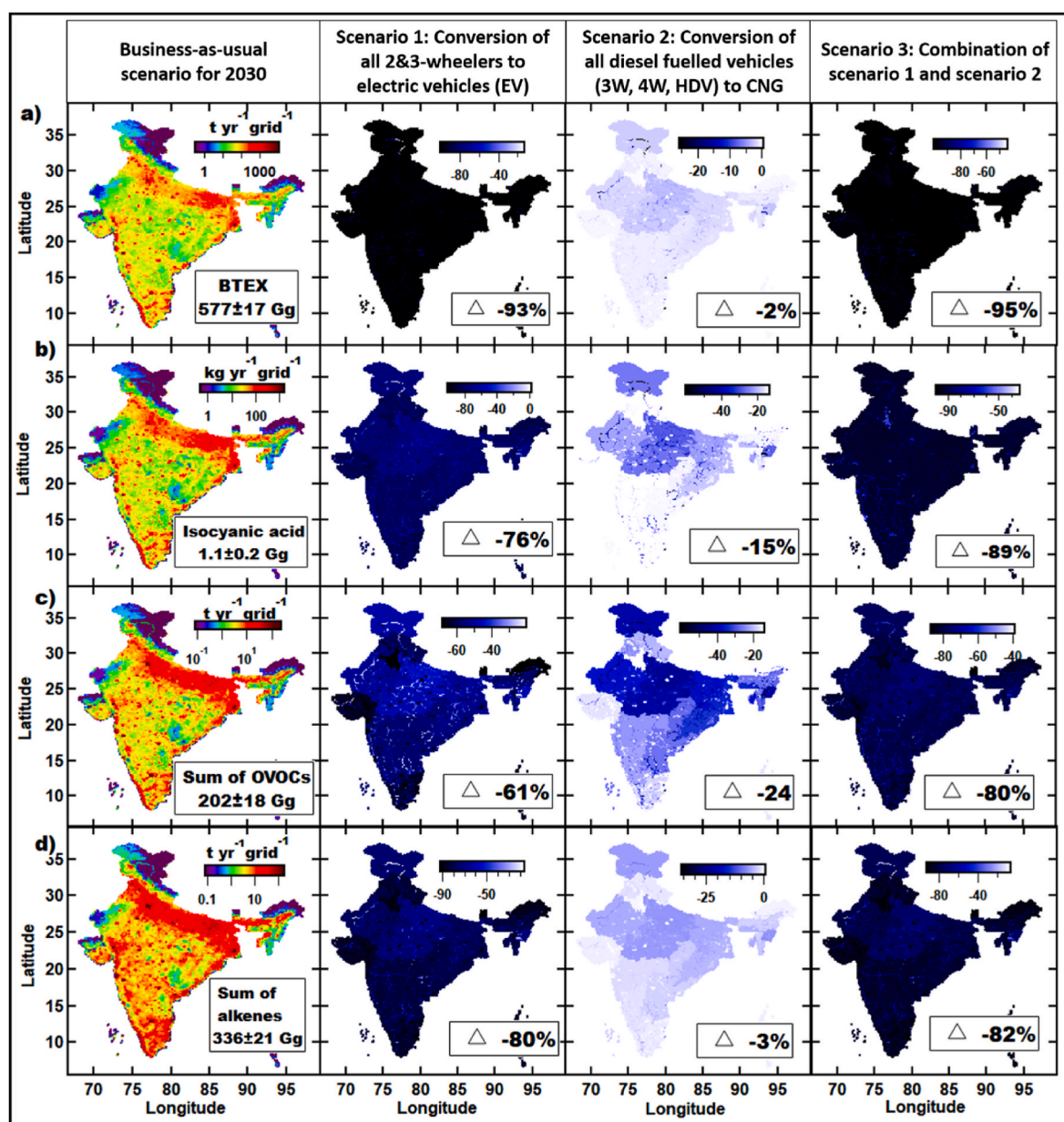


Fig. 3. Annual gridded ($0.1^\circ \times 0.1^\circ$) emissions of isocyanic acid (panel a), BTEX (panel b), sum of oxygenated VOCs (OVOCs; panel c) and sum of alkenes (panel d) in the 2030 Road Transport Emission Inventory of India (RTEII) (first column) under different intervention scenarios for potential emission reductions. The second, third and fourth column in each panel shows the percentage differences for scenarios 1, 2 and 3 relative to column 1, respectively.

much lower reductions in BTEX (2%). Unlike in the case of scenario 1, benzene emissions showed the maximum reduction (−11%) while rest of the BTEX compounds hardly showed any difference (−3%) under scenario 2. Benzene is categorized as a carcinogen (USEPA) while ethylbenzene is recognized as a potential carcinogen (IARC, 2000). Toluene and xylene are non-carcinogenic, but they impact the reproductive and central nervous systems, especially when exposures are chronic at low to high concentrations (Chen et al., 2011; Correa et al., 2012; Duarte-Davidson et al., 2001; Kalenge et al., 2013; Moolla et al., 2015; Tunsar-Ingkarn et al., 2012). Our results indicate that substantial reductions in the exposure to carcinogenic benzene can be accomplished by measures targeted at promoting electric 2- and 3-wheelers.

Recently it has been reported that isocyanic acid (Wentzell et al., 2013; Brady et al., 2014; Link et al., 2016; Suarez-Bertoa and Astorga, 2016; Jathar et al., 2017) can be emitted by on road vehicles as a by-product of the catalytic conversion of CO and NO in the presence of H_2 over a catalyst in petrol and diesel vehicles or as primary emissions

due to fuel combustion inside the diesel engine (Jathar et al., 2017). The total annual emission of isocyanic acid from the road transport sector in India from all vehicle categories plying in India was 0.7 Gg y^{-1} in 2015. This will almost double to 1.1 Gg y^{-1} by 2030 (Fig. 3a; column 1). For isocyanic acid, switching to EVs instead of 2 and 3-wheelers under scenario 1 results in emission reduction of 76%. Considering the fact that CNG vehicles have relatively lower emission potential for isocyanic acid when compared to diesel fuelled vehicles, a conversion from diesel to CNG brings about a further reduction of 15%. Isocyanic acid is a highly toxic gaseous acid that dissociates at physiological pH to form cyanate anions (NCO^-), a biochemical intermediate, which in turn participate in protein damaging carbamylation reactions, thereby leading to adverse health outcomes such as cataracts, atherosclerosis, rheumatoid arthritis, cardiovascular disease, and renal failure (Roberts et al., 2019; Leslie et al., 2019). Roberts et al. (2011) used the physical properties of ICA to estimate that ambient mixing ratios as low as 1 ppbv could be harmful to humans (Wang et al., 2007). Despite having such active emission

sources and potential for toxicity, no ambient air quality standards or exposure limits for isocyanic acid currently exist.

Alkenes and oxygenated organic compounds (OVOCs) typically have relatively high reactivity to the main atmospheric oxidants, namely hydroxyl radicals (OH) and play an active role in the sequence of chemical reactions responsible for tropospheric ozone formation chemistry. OVOCs for example, are important intermediates in the oxidation of many primary pollutants and precursors for peroxyacetylnitrates (PANs) (Fischer et al., 2014). Under scenario 1, both alkenes and OVOCs were observed to reduce by 80 and 61%, respectively. All the individual VOCs under the alkene chemical class showed significant reduction (−49 to −97%) upon conversion of 2 and 3-wheelers to EVs under scenario 1. In scenario 2, we see a decrease (−29% to −32%) in the emissions of higher alkenes (>C5), 1-butene (−23%), propene (−18%), acetylene (−15%), and an increase in ethene emissions (21%) with negligible change in the emission of other alkenes (−3%) upon shifting from diesel to CNG fuel. This is not surprising as the average ethene emission factor for CNG fuelled vehicles (0.62 gL^{-1} ; Table S1) are nearly 2 times higher than that of diesel fuelled vehicles (0.33 gL^{-1} ; Table S1). At the same time, the conversion of diesel fuelled vehicles to CNG could accomplish significant reduction of OVOC emissions (−24%). Even though OVOCs showed a larger reduction under scenario 1 compared to 2 with a relative decrease of 61 and 24% in emission respectively, all the top emitted OVOCs including acetaldehyde (−50% and −40%), acetone (−44% and −48%), and nitromethane (−52% and −26%) showed significant reductions under both scenario 1 and 2, respectively. This reduction in top OVOCs under scenario 2 is partially negated by the increase in methanol emissions (+41%) caused by the higher emission factor of CNG vehicles (0.26 gL^{-1} ; Table S1) compared to diesel vehicles ($0.05\text{--}0.15 \text{ gL}^{-1}$; Table S1).

The results observed for various scenarios indicate that fleet

electrification of certain vehicle classes can synergistically deliver greater air quality and health benefits by effectively lowering the direct exposure to toxic VOCs such as isocyanic acid, benzene, nitromethane and by offering a potential for emission abatement of precursor compounds that form secondary pollutants such as ozone (O_3) and secondary organic aerosol (SOA). Substitution of diesel by CNG doesn't have an impact on all VOC classes, but the combination as shown by scenario 3, can further reduce the pollutant emissions.

3.3. The impact of intervention scenarios on total volatile organic compound emissions and majorly emitted compounds

In the default scenario, the total annual emission of Non-Methane VOCs from the road transport sector increase from $1.2 \pm 0.3 \text{ Tg y}^{-1}$ in 2015 to $2.9 \pm 0.8 \text{ Tg y}^{-1}$ in 2030, primarily because a much larger section of the Indian population will be earning enough to afford a 2-wheeler by 2030. Among all the fuel types, petrol powered vehicles dominate with the highest relative contribution to the Non-Methane VOC emissions with ~63% (1.8 Tg y^{-1}) followed by CNG 30% (0.87 Tg y^{-1}) diesel 7% (0.19 Tg y^{-1}). In terms of pollutant emission potential, LPG 3-wheelers were the most dominant vehicle category in 2015 with a total NMVOC emission factor of $62.3 \pm 18.2 \text{ gL}^{-1}$ followed by petrol 2-wheelers ($52.0 \pm 15.4 \text{ gL}^{-1}$), CNG ($9.3 \pm 4.4 \text{ gL}^{-1}$) and diesel 3-wheelers ($6.7 \pm 1.3 \text{ gL}^{-1}$). However, LPG 3-wheelers will be almost phased out by 2030, as new sales will likely become increasingly dominated by the more economical electric or CNG 3-wheelers. Fig. 4a shows the annual gridded ($0.1^\circ \times 0.1^\circ$) emissions NMVOC emissions from road transport sector for 2030 in RTEII under different intervention scenarios. The emissions obtained under scenario 1 suggests that conversion of 2 and 3-wheelers vehicle fleet to EVs will lead to the reduction of total NMVOC emission by 91%. Given the fact that 2 and 3-wheelers

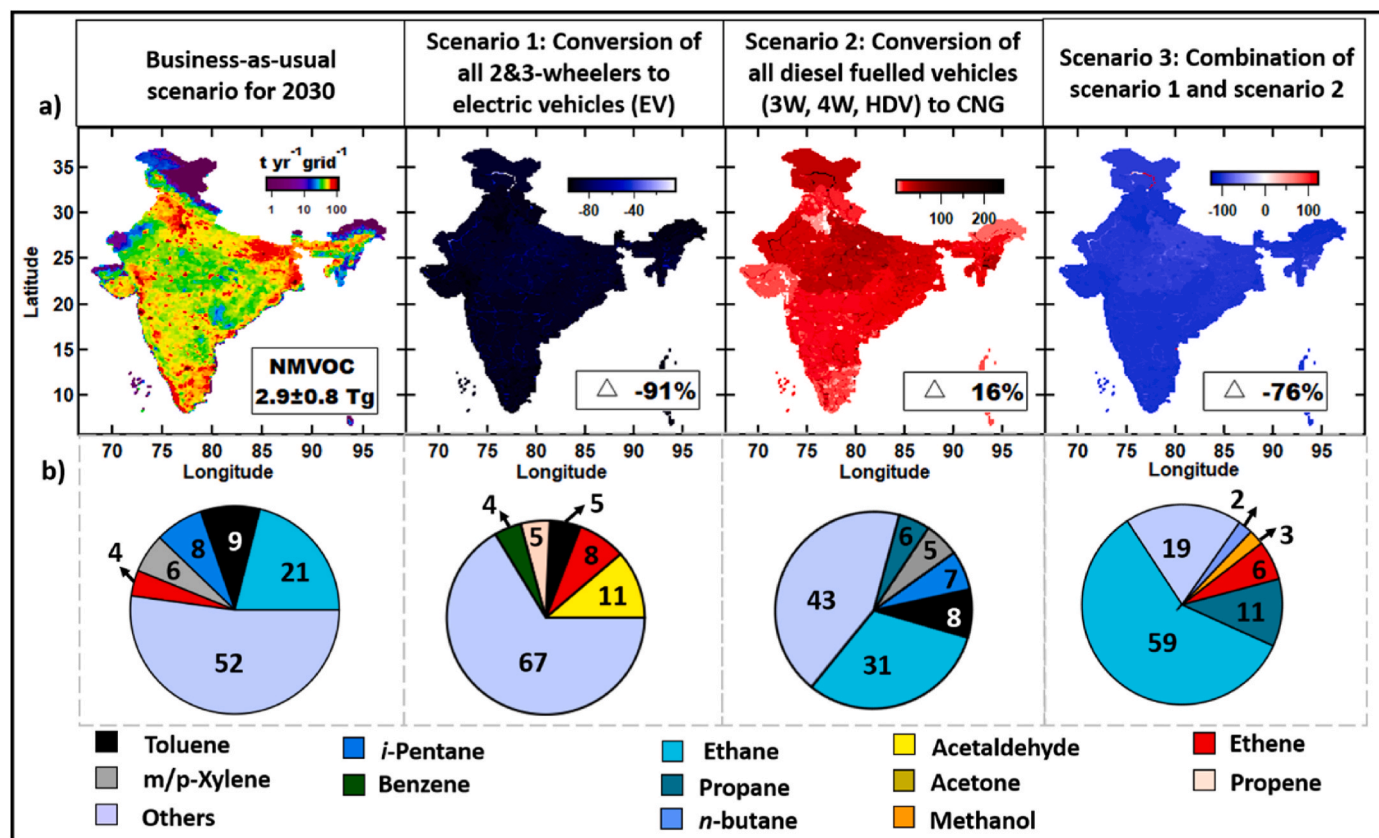


Fig. 4. Panel a) shows the annual gridded ($0.1^\circ \times 0.1^\circ$) emissions of total Non-Methane VOCs from road transport sector in the 2030 Road Transport Emission Inventory of India (RTEII) (first column) under different intervention scenarios for potential emission reductions. The second, third and fourth column in each panel shows the percentage differences relative to scenarios 1, 2 and 3 respectively. Panel b) shows the percentage contribution of top 5 emitted VOCs for each scenario.

will still be contributing more than 60% of all registered vehicles in India, a shift to electric variants with zero tailpipe emission is expected to significantly reduce the pollutant emissions within congested urban conglomerates.

Most of the 2-wheelers in the city are owned by individual owners, used for non-commercial purposes and their increase is driven by population growth in the region (DSH, 2016). India's per capita mobility is highly correlated with per capita income and is expected to grow even further till 2050 (Dhar et al., 2017). The benefit of replacing internal combustion engine vehicles by electric vehicles (EVs) is not only limited to emission reduction and improved energy efficiency in the transportation sector but extends to future improvements in the carbon footprint with increasing penetration of renewable power sources within the grid throughout the vehicles' lifetime. In this context, it should be noted that if all vehicle km travelled by 2- and 3-wheelers in 2015 had been travelled by EVs instead, the total power consumption in the Indian electricity grid would have increased by a mere 2.7%. Given the low-capacity utilization factor of many power plants within the grid, this measure will hardly impact total GHG emissions from the Indian power sector. However, it must be stressed that conversion of heavier vehicles such as cars, SUVs and HDVs to EV would have significant ramifications for the energy sector. Hence, EV adoption across all vehicle classes may not be favourable in the Indian context. The analysis of scenario 2 suggests that converting all diesel fuelled vehicles to CNG variants will further increase the NMVOC emissions by 16%. The increase in total NMVOC emission under scenario 2 is attributable to the high emission

potential of lower alkanes by CNG vehicle when compared to diesel vehicles. The total emission of ethane and propane increased by 71% and 72%, respectively, under scenario 2. However, alkanes have much lower OH reactivity than some of the more reactive OVOCs and alkenes, indicating that the substitution may be beneficial despite the increase in the total NMVOC emissions. Under scenario 3 which is a combination of scenario 1 and 2, the total NMVOC emission reduced by 76%. The annual emissions of highly reactive VOCs including toluene, xylene, acetaldehyde, propene decreased by 85% or more in scenario 3 (Fig. 4b).

3.4. The impact of policy intervention scenarios on reducing formation potentials of secondary pollutants and climate active emissions

Two of the major secondary pollutants having an impact on air quality and health effects are ozone (O_3) and secondary organic aerosol (Fann et al., 2012; Sujith and Sehgal, 2017; Lelieveld et al., 2015a,b). Chemical composition studies across polluted cities in the Indo-Gangetic plain have shown that secondary organic aerosols (SOA) originating from gas phase precursors contribute about more than 50% mass to the fine-mode particulate matter (Behera and Sharma, 2010; Gani et al., 2019). O_3 is known to be associated with respiratory morbidity and mortality (Jerrett et al., 2009; Orru et al., 2013) and long-term exposure to high concentrations of surface O_3 can also damage vegetation with substantial reductions in crop yields and crop quality (Mills et al., 2011; Sinha et al., 2015). Therefore, a significant reduction in the secondary pollutant formation has a large potential to reduce $PM_{2.5}$ pollution and

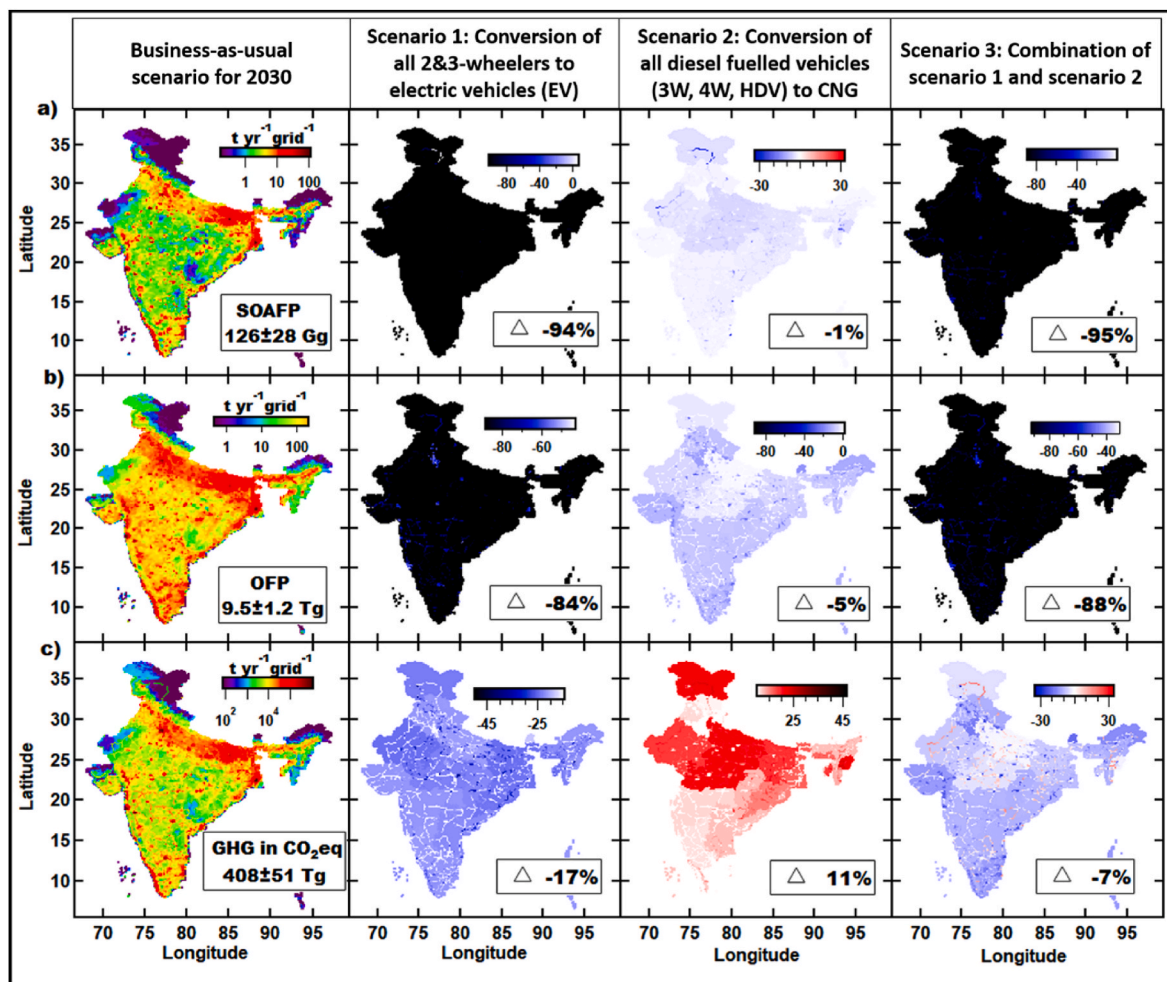


Fig. 5. The total amount of secondary organic aerosol, SOA (panel a) and ozone (panel b) that can potentially be formed from traffic emissions in India under different intervention scenarios for potential emission reductions in 2030.

improve the food security.

Fig. 5a and b shows the total amount of secondary organic aerosol (SOA) and ozone that can potentially be formed from traffic emissions in India under different intervention scenarios for emission reductions. The amount of SOA that can potentially be formed from traffic showed a significant reduction of 94% from the deployment of electric 2 and 3-wheeled vehicles having zero tailpipe emissions under scenario 1. Tailpipe emissions from petrol 2-wheelers are the important source of secondary organic aerosol in urban environments with a SOA production potential value of 3.53 gL^{-1} (Hakkim et al., 2021). 2 and 3-wheelers together contributed more than 92% of the total aromatic VOC emission. The conversion of all diesel fuelled vehicles to CNG under scenario 2 reduced the primary emission of BC but resulted in only a small decrease of 1% in SOA formation potential. This is mostly due to the fact that diesel exhaust contributes only $\sim 7\%$ of the total aromatic VOC emissions from the road transport sector.

The highly reactive ozone precursor compounds under different chemical classes, including aromatics, alkenes and OVOCs showed a significant decrease under scenario 1 (See Fig. 3), leading to a reduction in ozone formation potential by 84% (Fig. 5 b). CNG did not have such a significant impact in Scenario 2 ($\sim 5\%$ reduction). Among the 74 VOCs measured, m/p-xylene (1.7 Tg; 18%), toluene (1.1 Tg; 11%), propene (0.6 Tg; 6%), ethene (1.0 Tg; 10%) and acetaldehyde (0.4 Tg; 4%) were top 5 VOCs with a combined contribution of 49% to the total ozone formation. About $\sim 95\%$ of the top 2 contributors in m/p-xylene and toluene was due to petrol 2-wheelers, the most dominant vehicle type in terms of ozone formation with a contribution of 71% (6.8 Tg) to the total. Upon converting all the diesel-fuelled vehicles into CNG under scenario 2, the contribution of both propene and acetaldehyde reduced by 18 and 40% respectively while the contribution of ethene to ozone formation increased by 21%. Similarly, ethane, one of the tracer compounds for CNG exhaust emissions, showed an increase of $\sim 71\%$ in contribution under scenario 2. This is largely due to the higher ozone production potential of both ethane (1.8 gL^{-1} ; $\text{gO}_3/\text{Litre of fuel burnt}$) and ethene (5.6 gL^{-1}) from CNG exhaust emissions when compared to their average ozone production from diesel exhaust (0.01 and 3.4 gL^{-1} respectively). At the same time, the contribution of aromatic VOCs such as m/p-xylene and toluene remains relatively unchanged ($<2\%$) as the petrol-fuelled vehicles are unaffected under scenario 2. As a result, the overall impact of conversion from diesel-fuelled vehicles into CNG was observed to be low as it could fetch a reduction of only 5% in the total amount of potential ozone formation.

In addition to the reduction in secondary pollutant formation, a major co-benefit related to increasing the number of EVs is that it reduced direct emissions of greenhouse gases. Conversion of 2 and 3-wheelers to EVs under scenario 1 reduces the overall CO_2eq emissions by 17% (Fig. 5c). On the other hand, diesel to CNG conversion results in an increase in the CO_2eq emissions by 11%. When compared to other vehicle types, diesel engines are highly efficient which allows relatively more complete burning of fuel and high emission of CO_2 (2604 gL^{-1}). Considering that they were also the dominant consumers of automotive fuel (68%), diesel fuelled vehicles were responsible for the largest share of CO_2eq emissions (74%) in 2015. By 2030 their contribution to the total CO_2eq emissions will decrease to 25%, because diesel subsidies have been slashed in 2014 and prices are now linked to the international oil market. The share of CNG will increase to 43% of the total CO_2eq emissions by 2030 under a business-as-usual trajectory, as the fuel is currently favoured by lower taxation. Conversion to CNG increased CH_4 emissions ($50 \pm 26 \text{ gL}^{-1}$; Table S1) when compared to diesel ($0.34 \pm 0.05 \text{ gL}^{-1}$; Table S1) and hence increased the total CO_2eq emissions. However, the substitution of CNG with domestically produced Compressed Bio-Gas (CBG) under the Sustainable Alternative towards Affordable Transportation (SATAT) scheme, targeted at producing CH_4 from manure and biodegradable waste, could offset the effect and could make the conversion carbon negative. Overall, the combined scenario 3 shows only a marginal decrease of GHG emissions (-7%), if the long-term

intervention strategies are adapted.

3.5. Electric vehicles in India: opportunity, challenges and impact on power grid

The results obtained under different intervention scenarios suggest that the use of electric vehicles could be an important option for reducing exhaust emissions of many of the major air pollutants. Electric vehicles not only reduce the dependency on fossil fuel but also diminish the formation of secondary pollutants. Through different schemes such as the National E-Mobility Programme and NEMMP, the Government of India is focusing on creating charging infrastructure and a policy framework to ensure that by 2030 all public transport and a significant fraction of private vehicles are electric. While the urban air-quality benefits of electric vehicles under scenario 1 in our study are unquestionable, the fleet electrification poses different challenges in terms of implementation and life cycle emissions. Even though EVs have zero carbon emissions on a tank-to-wheel basis, aspects of their production can induce similar, less, or alternative environmental impacts, most prominently due to electricity generation and vehicle manufacturing (Hawkins et al., 2013; Onat et al., 2014). The extent of the impact depends on the power source used to manufacture and drive the EVs. However, studies involving the comparative life cycle assessment of conventional fuelled vehicles and EVs conducted from manufacturing to final disposal, including operation of the vehicles, have reported that the carbon footprint of an EV ($116\text{--}146 \text{ gCO}_2/\text{km}$) is lower than an Internal Combustion Vehicle (ICE) ($262\text{--}363 \text{ gCO}_2/\text{km}$) (Bieker, 2021; Wolfram et al., 2021). According to the recent report by the International Council on Clean Transportation (ICCT), comparing the life cycle assessment of motorcycles and scooters registered in India in 2021, electric motorcycles and scooters emit 33–45% and 38–50% less CO_2eq over their combustion-powered counterparts, respectively (ICCT, 2021). Abhyankar et al. (2017), reported that the fleet electrification would still reduce per-kilometre CO_2eq emissions by about 30% for two-wheelers, even if the electrical grid in India 2030 remains as coal-heavy as it was in 2015–16. As the grid adds renewable energy capacity and becomes less carbon-intensive, the carbon emissions associated with EVs could be even lower and could also be beneficial for the large-scale adoption of EVs. This is especially important for India as it has the cheapest solar power globally (IRENA, 2018).

India currently has an installed capacity of 382 GW with the peak demand of 170 GW and coal remains the primary source of electricity generation (55%) (NPP, 2021). Using the performance parameters of a range of electric 2- and 3-wheelers currently sold in India, we calculate that under scenario 1 (complete conversion of 2-and 3-wheelers to EV) the projected power consumption in 2030 would increase by 76 TWh which amounts to 4% of the total electricity consumption in the same year. When substitution is not focused on light vehicles, but includes 4-wheelers and HDVs, 33 percent penetration of EVs in sales by 2030, may increase total electricity demand by an additional 37 to 97 TWh, which will be less than 5% of the total demand. It is unlikely that the conversion would have a significant impact on the power system (Ali and Tongia, 2018). Overall, pursuing policies in line with scenario 1 promises significant reductions in the population exposure to carcinogenic benzene, secondary PM and tropospheric ozone, a reduced need to import petroleum products, and reduced carbon emission. It would also enable India to honour its obligation under the Paris Climate Agreement to reduce the emissions intensity of GDP by 33–35% from 2005 levels by 2030. A study released by the Council on Energy, Environment and Water (CEEW) reports that India could save on crude oil imports worth over rupees one trillion (Rs. 1 lakh crore) annually if EVs were to garner 30 percent share of India's new vehicle sales by 2030 thus providing energy and economic security with improved air quality (CEEW, 2020).

4. Conclusion

We have compared the annual emissions of criteria air pollutants and 74 VOCs from the road transport sector in India considering three intervention scenarios with respect to emissions that would occur under a business-as-usual developmental approach for the year of 2030. The results suggest that a shift from petrol to electric vehicles (scenario 1) could lead to a decrease in total NMVOC emissions as well as in individual NMVOC classes which can result in prompt and substantial improvements in air quality and health gains. Gaseous pollutants including the toxic VOCs such as isocyanic acid (−76%) and BTEX (−93%), reactive VOC classes including sum of OVOs (−61%) and sum of alkenes (−80%) and criteria pollutants such as CO (−80%), PM_{2.5} (−44%) and NO_x (−46%) all show tendency for significant reductions upon replacing the highly polluting 2 and 3-wheeled vehicles by electric vehicles. Highly polluted cities may benefit from aggressively promoting electric 2 and 3-wheelers by laying out the charging infrastructure. Large reduction BC (−50%) emissions under scenario 2 provide an important decision in favour of the introduction of CNG. The increased GHG emissions due to such a conversion could potentially be offset by a switch to CBG rather than CNG. Particulate Matter in the recent past has been considered one of the most potent pollutants with regard to its impact on human health and therefore, the reduction obtained thus has the potential to increase life expectancy and decrease the mortality rate. As suggested by our results, both scenarios 1 and 2 complement each other in reducing the annual emission of all pollutant species. While scenario 1 helps in improving air quality by reducing direct emission of toxic VOCs and criteria air pollutants as well as precursor compounds for secondary pollutant formation, scenario 2 reduces the BC emission. Scenario 3 in which benefits of scenario 1 and 2 are obtained by combination, represent the best long-term strategy moving forward to not only improve human health and the health of ecosystems and agriculture, but also place reasonable constraints from climate relevant emission perspective.

CRediT authorship contribution statement

Haseeb Hakkim: performed the measurements and analyses as part of his PhD thesis work, wrote first draft of paper which was revised by. **Ashish Kumar:** contributed to the primary data. **Baerbel Sinha:** designed the study and supervised analyses of the data and all aspects of the study. **Vinayak Sinha:** designed the study and supervised analyses of the data and all aspects of the study.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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