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Road transport impact on $PM_{2.5}$ pollution over Delhi during the post-monsoon season

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ABSTRACT

We use the WRF-Chem atmospheric chemical transport model, driven by local emission inventories, to quantify the contribution of on-road transport emissions to surface $PM_{2.5}$ over Delhi during the post-monsoon season. We compare this contribution to other local (within Delhi) and regional (within the broader National Capital Region, NCR) anthropogenic sectors during the post-monsoon period when seasonal burning and stagnating meteorological conditions exacerbate baseline pollution levels. We find that local on-road transport contributes approximately 10% to daily mean $PM_{2.5}$ over Delhi, rising to 17% if regional on-road transport sources in the NCR are included. The largest individual contributions to Delhi daily mean $PM_{2.5}$ are from regional power and industry (14%) and domestic (11%) sectors, dominating nighttime and almost all daytime concentrations. Long range transport contribution from sources beyond the NCR is found to account for approximately 40%. The contribution from the local on-road transport sector to diurnal mean $PM_{2.5}$ is largest (18%) during the evening traffic peak. It is dominated by contributions from two- and three-wheelers (50%) followed by heavy-duty vehicles (30%), which also collectively represent 60–70% of the total on-road transport sector at any hour of the day. The combined contribution from passenger cars and light duty vehicles and from resuspended road dust to daily mean $PM_{2.5}$ is small (20%). Our work highlights two important factors which need to be considered in developing effective policies to meet $PM_{2.5}$ air quality standards in Delhi during post-monsoon. First, a multi-sector and multi-scale approach is needed, which prioritise the reduction in local transport emissions within Delhi, and, in the order, regional industries, domestic and transport emissions from NCR. Second, two-and three-wheelers and heavy-duty vehicles dominate on-road transport impact to $PM_{2.5}$, thus reductions from these vehicles should be given priority, both within Delhi and in the NCR.

1. Introduction

Delhi, India, is one of the most density populated megacities on the planet with 30 million inhabitants in 2020 living in an area of 1484 km^2. It also suffers from some of the most unhealthy air on Earth (WHO, 2016) with particulate matter ($PM_{2.5}$, particles with aerodynamic diameters ≤2.5 μm) pollution often exceeding average 24h mean World Health Organisation (WHO) Global Air Quality Guidelines of 15 μg m^{-3} and the Indian National Ambient Air Quality Standard (NAAQS) of 60 μg m^{-3} (WHO, 2021; CPCB, 2020). In 2019, elevated ambient PM pollution was responsible for an estimated 16,600 deaths in Delhi (Pandey et al., 2021). Mitigating this human health crisis is challenging, with PM pollution levels driven by seasonal local and regional emissions and seasonal meteorology, exacerbated by the geography of the Indo-Gangetic Plain (Mogno et al., 2021). Here, we use the WRF-Chem regional atmospheric chemistry model to explore the drivers of air pollution during the post-monsoon season (October–December) when PM is particularly high (Mogno et al., 2021; Patel et al., 2021; Chen

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et al., 2020a).

During the post-monsoon season, Delhi air quality is worsened by seasonal biomass burning from post-harvest crop in the neighbouring states of Haryana and Punjab (Liu et al., 2018; Kulkarni et al., 2020; Bikkina et al., 2019; Kumari et al., 2021; Sembhi et al., 2020). Deteriorating air quality due to crop residue burning has been a major focus for mitigation strategies but these emissions occur when there are already hazardous background levels of $PM_{2.5}$ (Ojha et al., 2020; Kulkarni et al., 2020; Bikkina et al., 2019; Liu et al., 2018). Thus there is the need to investigate more *in situ* control of anthropogenic emissions. In this work we focus in particular on on-road transport emissions, which is one of the main targets of emission control strategies for Delhi.

Generally, the main sources of air pollution in Delhi are local anthropogenic sources from within the megacity Delhi and from nearby states within the National Capital Region (NCR) (Chen et al., 2020b; Guo et al., 2019; Amann et al., 2017; Marrapu et al., 2014). This highlights the need for a coordinated emissions reduction strategy to mitigate Delhi $PM_{2.5}$ pollution. The local on-road transport sector is a substantial source of $PM_{2.5}$ pollution in Delhi. Source apportionment studies based on $PM_{2.5}$ measurements at sites in Delhi found that vehicular emissions contribute seasonally between 17 and 30% for $PM_{2.5}$ pollution, with the highest share during the post-monsoon and winter seasons (Jain et al., 2020; TERI & ARAI, 2018).

The on-road transportation fleet in Delhi includes two- and three-wheelers, passenger cars, and freight vehicles. Two- and three-wheelers such as auto-rickshaws and motorized two-wheelers represented almost 65% of the total number of registered motor vehicles in Delhi in 2019 (MoRTH, 2021), and are used for personal and commercial transportation. Passenger cars accounted for 30% of the total number of registered motor vehicles in Delhi in 2019 (MoRTH, 2021). Freight vehicles (light-duty and heavy-duty vehicles) are a smaller fraction of the total number of registered motor vehicles, but share a high fraction of the total on-road transport sector emissions because they generally travel further each day into and within the city (Malik et al., 2019; Jain et al., 2016; Goel and Guttikunda, 2015).

The on-road transport fleet in Delhi has grown from 7.5 million vehicles in 2012 to 11.4 million in 2019, the highest number of registered vehicles of any state in India (MoRTH, 2021; Goel and Guttikunda, 2015). Passenger cars and freight activity in Delhi is forecasted to more than double from 2020 to 2050 (He et al., 2021). The Indian government has in the past few decades promoted measures to curb the impact of the growing on-road transport sector on air pollution. These include imposing stringent emission regulations on all new vehicles from Bharat Stage III (corresponding to EURO III European vehicle emission standards) nationwide in 2010 to Bharat Stage VI (corresponding to EURO VI European vehicle emission standards) in 2020 (Hakkim et al., 2022). Since the beginning of the 2000s, Delhi has been converting public transport vehicles, including buses, three-wheelers and taxis, to operate on compressed natural gas (CNG) following a Supreme Court judgment in 1998 (Kathuria, 2004). Additionally, recent measures to control traffic emissions include the introduction of a alternating "odd–even" licence plate pilot policy for passenger cars during the winter and the pre-monsoon seasons. However, the odd–even strategy only marginally reduced the $PM_{2.5}$ concentrations in Delhi (Sharma et al., 2017; Chowdhury et al., 2017; Kumar et al., 2017). Despite these interventions, air quality levels in Delhi remain hazardous to human health, and thus there is an urgent need for a more systemic analysis of the traffic contribution to Delhi's pollution aiming at assessing and improving current intervention strategies. In this study we use the WRF-Chem regional atmospheric chemistry transport model to investigate the contributions from on-road transport sectors and fleet segments (two- and three-wheelers, cars and light duty vehicles, heavy duty vehicles and resuspended road dust) to surface $PM_{2.5}$ levels over Delhi during the post-monsoon season. We drive WRF-Chem with local-scale emissions inventories at a spatial resolution of 4 km, and through a series of sensitivity studies we investigate the contribution to $PM_{2.5}$ of on-road transport and its subsectors compared to other anthropogenic sectors. We consider contributions from local (within Delhi) and regional (within the NCR) anthropogenic sectors and how they impact the 24-h mean and diurnal cycle of $PM_{2.5}$. In the next section we describe the data and methods, including the WRF-Chem model, emission inventories, and methods we use to explore individual section contributions to surface $PM_{2.5}$. The results are presented in section 3, and conclusions are discussed in section 4.

2. Data and methods

2.1. WRF-chem atmospheric chemistry and transport model

We use the WRF-Chem Weather Research and Forecasting model coupled with Chemistry v3.9.1. WRF-Chem has been widely used for modeling air quality over India and over Delhi (Mogno et al., 2021; Jena et al., 2021; Ojha et al., 2020; Kulkarni et al., 2020; Chen et al., 2020b; Gupta and Mohan, 2015). Our set-up has two one-way nested domains. The parent domain encompasses north India at 12 km horizontal resolution, while the nested domain covers the entire NCR at 4 km horizontal resolution (Fig. 1). Both model domains have 33 levels from the surface to 50 hPa. We run simulations for the post-monsoon season from 6th October to 16th October 2019. Each simulation has a spin-up period of 5 days (1–5 October 2019) to minimize the influence from the established initial conditions at the start of the model run. The period to simulate has been chosen as a compromise between three main factors: 1) representativeness of the chemical and meteorological environment which characterises the post-monsoon season in Delhi, 2) minimisation of the influence of the biomass burning on air quality over Delhi, in order to reduce the degrees of freedom in our simulations and better isolate the contribution of the anthropogenic sectors, and 3) limit the computational burden of the simulation experiments. For condition 1), we make sure that the 10 days considered are representative of a longer post-monsoon period by comparing the hourly frequency distribution of modeled $PM_{2.5}$ between 6–16 October over Delhi with hourly distribution of observations in Delhi (listed in Table S1 and Fig. S1). The frequency distributions of modeled $PM_{2.5}$ are similar to the observations (Fig. S2). Even if high concentration spikes are not fully captured by the model, the difference in the mean $PM_{2.5}$ concentrations is 10% and in the median concentrations is 11%, making the selected period acceptable as representative for a longer post-monsoon period. For condition 2), we check the fire activity in the north-west part of India for the whole post-monsoon season, using satellite-derived fire counts. The period chosen between 6–16 October 2019 is before the full onset of the biomass burning emissions, which peak later in October and November 2019 (Fig. S3). This allows minimisation of the impact of the biomass burning and enables us to focus on the influence of anthropogenic emissions. For condition 3), we run some test simulation, and established that a period of 10 days (in addition to 5 days of spin up) is for us manageable when considering computational burden of simulations. The chosen 10 days simulation period doesn't take into account monthly and yearly variability of the chemical environment that characterise the post-monsoon period, and we acknowledge that is a limitation of our set-up. However, the chosen period is a good compromise for the scope of this study. Our choice of a reduced simulation period is also in line with similar studies requiring a large number of chemical transport model simulations (Chen et al., 2020b; Conibear et al., 2021)

The main physical and chemical parametrisations in this study are the same as in our previous study of atmospheric chemistry over the Indo-Gangetic Plain (Mogno et al., 2021). Briefly, the Model for OZone And Related chemical Tracers, version 4 (MOZART-4) is used for describing gas chemistry (Emmons et al., 2010) and the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) for describing aerosol chemistry, which includes aqueous phase chemistry with 1-D Volatility Basis Set (VBS) treatment for secondary organic aerosol (SOA) formation (Knote et al., 2014, 2015). We modified the code to

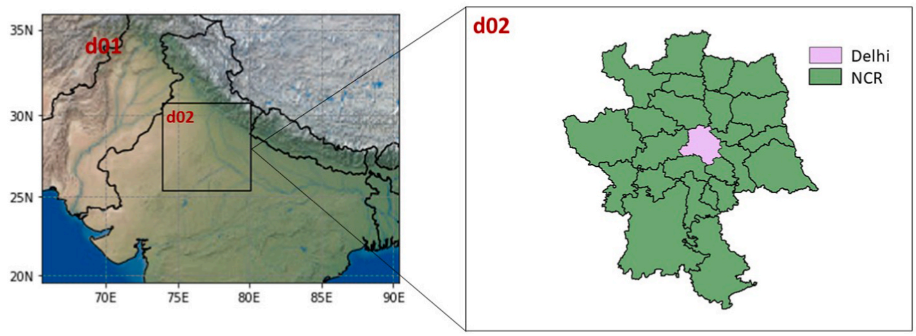

Fig. 1. WRF-Chem model domains used in this study: d01 refers to parent domain described with a 12 km horizontal resolution, and d02 refers to the nested domain, described with a 4 km resolution, that covers Delhi and the broader NCR.

increase the minimum value of the vertical turbulent diffusion coefficient within the planetary boundary layer (PBL), following (Du et al., 2020) who previously identified that this modification was key in successfully reducing model bias for diurnal variations of urban $PM_{2.5}$, particularly during nighttime. Through sensitivity runs, we found that a lower limit for the PBL vertical turbulent diffusion coefficient of 5 $m^2 s^{-1}$ led to the best comparison with ground-based observations over Delhi (Fig. S4). This is the same value found by Du et al. (2020) for cities in eastern China. Additional information on our model set up can be found in Mogno et al. (2021).

For the meteorological initial and boundary conditions, we use meteorological reanalyses from NCEP FNL Operational Model Global Tropospheric Analyses Data (National Centers for Environmental Prediction, National Weather Service, NOAA, U.S. Department of Commerce, 2015) at a spatial resolution of $0.25° \times 0.25°$ and at a temporal resolution of 6 h. The model meteorology is nudged to the observations to prevent the model from deviating too far from observed meteorology. Chemical initial conditions and lateral boundary conditions for each month are provided by six-hourly CAM-CHEM global model data (Buchholz et al., 2019).

We include anthropogenic, biomass burning, and biogenic emissions. For the parent domain (d01), we use the EDGARv5.0 monthly anthropogenic emission inventory for 2015 at $0.1° \times 0.1°$ resolution ($\sim$11 km) (Crippa et al., 2019a,b,c). For the nested domain (d02) we use the anthropogenic emission inventory from Energy and Resource Institute and The Automotive Research Association of India (TERI/ARAI) that covers the NCR region (TERI & ARAI, 2018); more details are provided in Section 2.2. Emissions for the fraction of the nested grid that lies outside the NCR are described by the EDGARv5.0 inventory. We used the TERI/ARAI emissions inventory over Delhi instead of the EDGARv5.0 because it relies on detailed information gathered at the local level, with spatial resolution higher than EDGARv5.0, and subsectoral transport information needed for this study. In general bottom-up urban inventories, when available, should be preferred to the corresponding top-down (Vedrenne et al., 2016). We can anticipate though that the EDGARv5.0 and the TERI/ARAI would give different estimates of pollutants emission over Delhi, with discrepancies mainly driven by the different methodologies and the spatial resolution used for estimating emissions, as found in out in previous studies (Saikawa et al., 2017; Trombetti et al., 2018).

For both inventories, we map the original speciated non-methane volatile organic compounds (NMVOC) emissions to the MOZART chemical mechanism used by WRF-Chem. Details of this mapping for the EDGARv5.0 inventory are summarised in Mogno and Marvin (2022) and details of the mapping for the TERI/ARAI inventory are described in Table S2. We describe biomass burning using the FINNv1.5 emission inventory (Wiedinmyer et al., 2011). Biogenic emissions are calculated online using MEGAN (Guenther et al., 2006).

2.2. TERI/ARAI anthropogenic emission inventory for the NCR

The TERI/ARAI anthropogenic emission inventory (TERI & ARAI, 2018) describes emissions of nitrogen oxides ($NOx = NO + NO_2$), sulfur dioxide (SO_2), carbon monoxide (CO), particulate matter (PM_{10}), and NMVOCs for all the main anthropogenic sectors and subsectors (listed below) for NCR at a spatial resolution of 4 × 4 km for 2016. The sectors include transport, domestic (use of biomass fuel, kerosene and LPG for cooking and heating), power, industry, and other (including refuse burning, crematoria, landfill fires, incinerators, refinery, airport, restaurant, construction). The inventory also includes factors that describe seasonal (monthly) and diurnal (hourly) emission variations. Fig. 2 shows sector and on-road transport subsector contributions to key pollutant emissions in the NCR and Delhi. Fig. 2c shows the corresponding annual mean distribution for on-road transport, domestic, and industry, power and other sectors.

The on-road transport sector of the TERI/ARAI inventory includes emissions for eight vehicles types: 1) two wheelers, 2) three wheelers, 3) passenger cars, 4) multi utility vehicles, 5) buses, 6) light duty vehicles, 7) heavy duty vehicles, and 8) tractors. The inventory also includes PM emission estimates from road dust resuspension due to vehicles. Except resuspended dust, other non-exhaust emissions (tyre or brake wear) from transport are not included in the current TERI/ARAI emissions inventory. In contrast to the situation regarding exhaust emissions, in general no policies are in place to reduce tyre or brake wear, and usually countries do not report these emissions in national and local inventories. However, because they are mainly emitted through mechanical processes, they can be an additional source for direct emissions of PM_{10} and $PM_{2.5}$ (Gietl et al., 2010; Denier van der Gon et al., 2013). As a consequence, primary emission of PM from transport in the TERI/ARAI inventory could be underestimated. Fig. 2b shows the contribution from individual on-road transport subsectors to total emissions by pollutant species within the NCR and Delhi. Emissions from different vehicle types are assumed to have the same spatial distribution at 4 km and to follow the same diurnal cycle. The seasonal change in road dust re-suspension was customized for local conditions using the information on sampling silt loading on the roads, and the weight of vehicular fleet (taken from traffic counts). On-road transport sector emissions in Delhi and the NCR have been estimated using a bottom-up approach, through activity data estimated from primary traffic count surveys in Delhi and surroundings. The inventory takes into account high-emitting vehicles, which are not taken into account in the development of standard emissions factors for representative vehicles in the fleet. (TERI & ARAI, 2018).

2.3. Observations for model evaluation

The model ability to reproduce observed surface $PM_{2.5}$ over Delhi is evaluated using ground based observations. We use the Central Pollution Control Board (CPCB) hourly observations of $PM_{2.5}$ for the period 6–16 October 2019 which are available to download at the portal

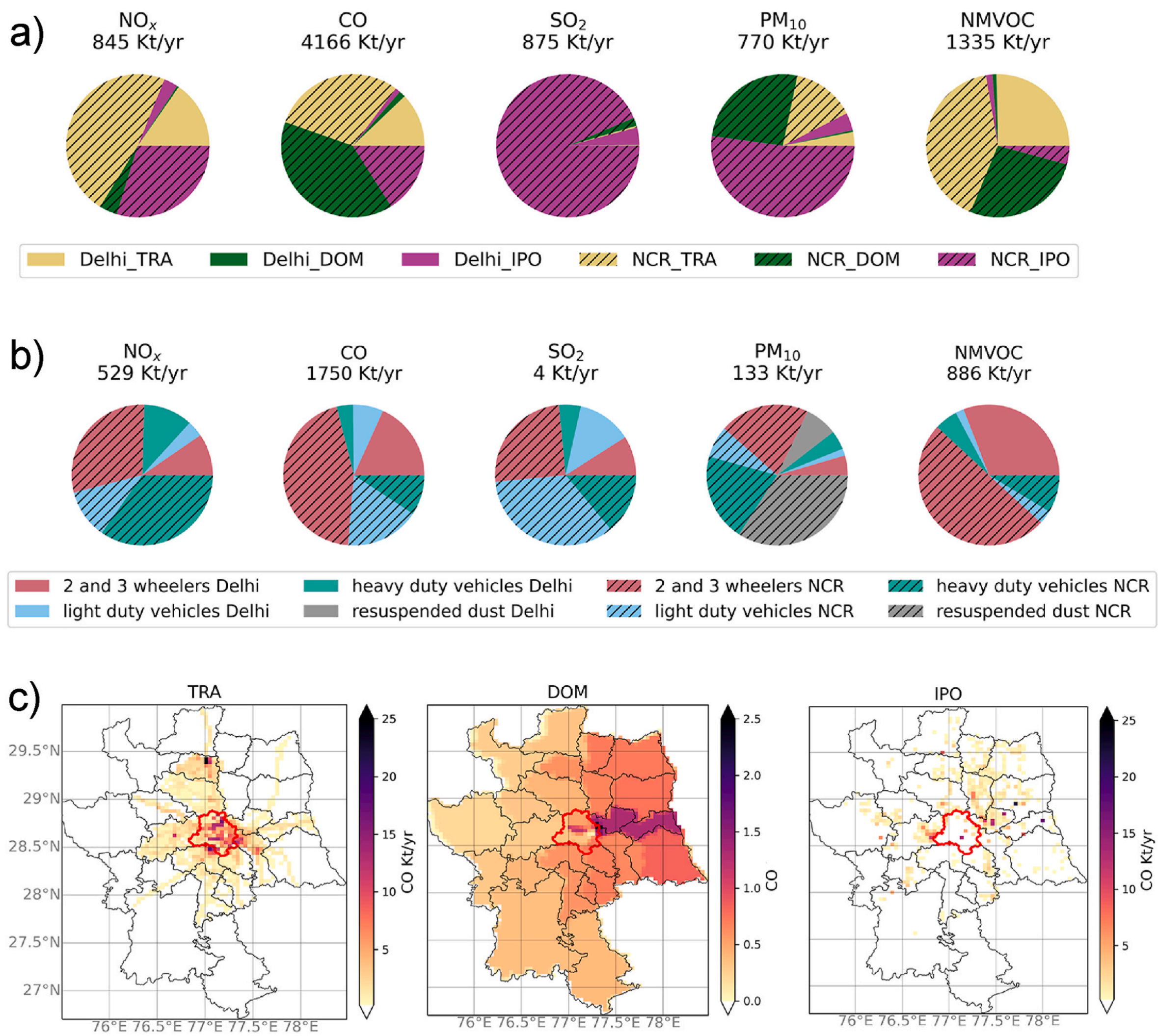

Fig. 2. Annual sector contributions and distributions of the TERI/ARAI anthropogenic emissions inventory for the regional NCR (areas outlined in black) and Delhi (central area outlined in red). Panel a) shows sector contributions to total annual emissions of nitrogen oxides (NOx), sulfur dioxide (SO_2), carbon monoxide (CO), particulate matter (PM), and non methane volatile organic compounds (NMVOCs). Sectors include on-road transport (TRA), domestic (DOM), industry power and others (IPO). Emissions within Delhi only and within the broader NCR but excluding Delhi are denoted by "Delhi" and "NCR" respectively, determined by using relevant administrative boundaries. Panel b) is the same as Panel a) but for on-road transport subsectors. Panel c) shows the spatial distribution of CO emissions from the TRA, DOM and IPO sectors.

(CPCB). Data that have negative values, outliers, and stations with poor temporal coverage have been discarded following Mogno et al. (2021). The resulting 34 selected stations are shown in Fig. S1, and listed in Table S1.

2.4. Emission reduction impact (ERI) method

We use the emission reduction impact (ERI) method to quantify the role of the on-road transport sector and its subsectors compared to other main sectors on surface $PM_{2.5}$ over Delhi. The ERI method determines the role of a given source to air pollution levels by taking the difference of two air quality model simulations performed with the full emission source (control) and a reduced emission source (perturbed). This method is also referred to as the brute-force method, or a one at time sensitivity analysis (Clappier et al., 2017; Thunis et al., 2019). The ERI method has been widely used for air quality management studies (Huang et al., 2018; Conibear et al., 2018; TERI & ARAI, 2018; Huszar et al., 2016), and it is usually preferred to other methods for its suitability to support air quality interventions planning (Thunis et al., 2019). Care is taken to ensure the perturbation is sufficiently small that the resulting change in the non-linear chemistry is approximately linear. Formally, the relative impact of an emission source s on a receptor pollutant P is given by:

$$I^{P}_{[s,\alpha]\%} = \frac{P_{s,\alpha} - P}{\alpha P} = \frac{\Delta P_{s,\alpha}}{\alpha P}, \quad (1)$$

where the P is the pollutant concentration from the control model and $P_{s,\alpha}$ is the modeled pollutant concentration from the perturbed model for which the source s reduced by percentage α. The value of the relative impact $I^{P}_{[s,\alpha]\%}$ is expressed in percentage (%). Negative values of $I^{P}_{[s,\alpha]\%}$ indicates that the reduction of the source s reduces the pollutant concentration, while positive values indicates that it increases the pollutant concentration. Where $\alpha = 100\%$ the source is completely switched off (zero-out approach). To reduce the computational burden of the ERI method, we group together the main anthropogenic emissions sectors by source type: on-road transport, TRA (linear sources), domestic, DOM (area sources) and industry, power and others, IPO (point sources). For the on-road transport sector, we also group the emissions in four subcategories that reflect the Indian emissions regulation categories for vehicles (Bansal and Bandivadekar, 2013): light commercial vehicles (TRL), heavy duty vehicles (TRH), two- and three-wheelers (TRW). We also consider resuspended dust (DST). For each source we consider both emissions within Delhi only and emissions from NCR (excluding Delhi). Table 1 summarises the anthropogenic sources parameters used for the ERI application.

The key assumption associated with using the ERI method is that emission sources are changed by an amount that maintains linearity between emissions and concentration changes, as mentioned above. Here, we perform a series of simulations to establish the threshold value of α_t beyond which the perturbed chemical regime is inconsistent with the control calculation. In practice, this means identifying the value of α for which 1) there remains a linear relationship between a change in emissions and $PM_{2.5}$ concentrations and 2) the interaction terms associated with perturbed multiple related parameters are negligible. The second criterion ensures that the individual impacts are additive, i.e., the sum of the impacts are equal to the impact of the sum where $\alpha \leq \alpha_t$.

3. Results

3.1. Model performance

To evaluate our model performance in describing surface observations of $PM_{2.5}$ over Delhi, we consider the city scale mean value for $PM_{2.5}$. Model gridcells are selected corresponding to the observation sites and compare the mean across the stations (Fig. 3). Modeled $PM_{2.5}$ has a positive mean bias (MB = 0.14), particularly during the middle of the day. The model is not able to capture the observed morning peak at 0800–0900 local time. This is possibly due to inaccuracy in the inventory $PM_{2.5}$ primary precursors emissions rates in the morning, rather than the timing of PBL increase, since we checked that the model is able to capture the morning peak for other pollutants, such as NOx. However the broader observed diurnal pattern is well reproduced by the model (r = 0.89). These discrepancies likely reflect limitations of using meteorology and emissions at 4 × 4 km that cannot capture finer scale processes. On a city-scale resolution we argue this is acceptable performance. Other sources of error may also be responsible for the overestimation of individual $PM_{2.5}$ components. The inorganic fraction (sulfate, nitrate, ammonium) of model $PM_{2.5}$ represents ~37% of the total $PM_{2.5}$ mass (Fig. S5),the organic fraction (primary and secondary organic aerosols) represents ~22%, and black carbon represents ~6%. Previous measurement studies over Delhi have reported an inorganic fraction of 23–27%, an organic fraction (OA, usually calculated at 1.4 times organic carbon, OC) of 20–32%, and an elemental carbon fraction (equivalent to our model black carbon) of 5–15% (Bawase et al., 2021; Jain et al., 2020; Dumka et al., 2017). These observed values are broadly consistent with our model values except for the inorganic fraction for which the model has a bias of 10%. We attribute this to the model overestimating the sulfate component, which is a common model deficiency linked with overestimating its SO_2 precursor over the Indian region (Mogno et al., 2021; Conibear et al., 2018; Kota et al., 2018). It could lead to overestimating the impact of the IPO sector that is the main emitter of SO_2 in the NCR (Fig. 2).

Table 1
Groupings of anthropogenic emissions used in the ERI study. For each parameter we consider both emissions within Delhi only (Delhi_) and within the National Capital Region only, excluding Delhi (NCR_).

TERI/ARAI sectors	ID
industry + power + others	IPO
domestic	DOM
on-road transport	TRA
bus + trucks + heavy duty vehicles	TRH
light duty vehicles + multi utility vehicles + passenger cars	TRL
2wheelers+ 3wheelers	TRW
resuspended dust	DST

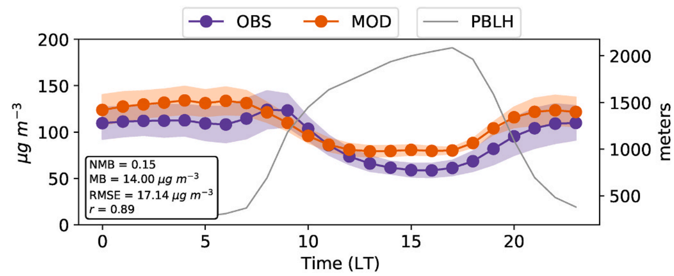

Fig. 3. Model evaluation of $PM_{2.5}$ over Delhi using ground-based observations during 6–16 October 2019. The plot shows the city-scale mean comparison of model and observed diurnal cycle of $PM_{2.5}$($\mu g\ m^{-3}$) as function of local time (LT). Shaded envelopes denote the standard deviation of the spatial variation of $PM_{2.5}$ at a particular time of observation. The grey line is the model boundary layer height (PBLH). Inset statistics are the normalised mean bias (NMB), mean bias (MB), root mean square error (RMSE), and Pearson correlation coefficient (r).

3.2. Emission reduction impact (ERI) range of applicability

To establish the range of applicability of the emissions reduction impact method in our study, we consider the response of emissions changes from each source in Table 1 to the 24-h average $PM_{2.5}$ (WHO metric) and the hourly value of $PM_{2.5}$ over Delhi for the whole period of the simulation. We consider symmetrical emission decreases and increases in the scaling range from 0% to 200% from the baseline, with a 25% step.

Fig. 4a shows a near-linear relationship between changes for emissions from individual sectors and 24-h mean $PM_{2.5}$ over Delhi over our range of considered perturbations. This is true also for each of the individual on-road transport subsectors (Fig. 4b). Simultaneous changes in emissions from all the main sectors results in interactions between pollutants that cannot be captured when considering one source at the time. These interactions account for an additional impact that increases with larger emission changes (Fig. 4c). We find this nonlinearity result in additional reductions of up to 16% in $PM_{2.5}$ when all the sectors are turned off but only a reduction of 2% when all sector emissions are doubled. Moderate changes in emissions (<25%) result in non-linearities of <5% in $PM_{2.5}$. Non-linearities are smaller when considering the on-road transport subsectors, where they account for ±5% additional impact in $PM_{2.5}$ over the whole range of emissions changes (Fig. 4d), likely because of their total smaller magnitude. We found similar results for hourly values of $PM_{2.5}$ over Delhi, which show an almost linear relation to changes in emissions over the range of ±75% (Fig. S6-S9). The sign and magnitude of non-linear terms vary across the range of emission changes, especially when considering the symmetric reduction/increase in emissions, suggesting that different chemical regimes may be reached. Based on our analysis, we establish an upper limit for linearity as $\alpha_t = 75\%$ within which non-linearities account for maximum ~±5% on top of the total impact on $PM_{2.5}$ given by summing the individual sectors impacts. This magnitude of non-linear impact is small in the context of other uncertainties associated with air quality modelling, and still provides practical information for policy makers.

Other studies have also identified linearity in the response of surface $PM_{2.5}$ to changes in main sector emissions (Conibear et al., 2021; Chen et al., 2020b; Thunis et al., 2015). Non-linearities are expected to be more important in particular for the formation of secondary pollutants at higher spatial and temporal resolution (e.g. local pollution episodes) (Thunis et al., 2015). In this study, we choose to calculate the impacts with $\alpha = 75\%$. We note that our choice of α doesn't affect the results obtained for the relative impact $I^{P}_{[s,\alpha]\%}$ of each sector s on $PM_{2.5}$. Indeed, because of the linear response of $PM_{2.5}$ to emissions reductions, any choice of the scaling factor α between zero and 75%, would have given the same impact results.

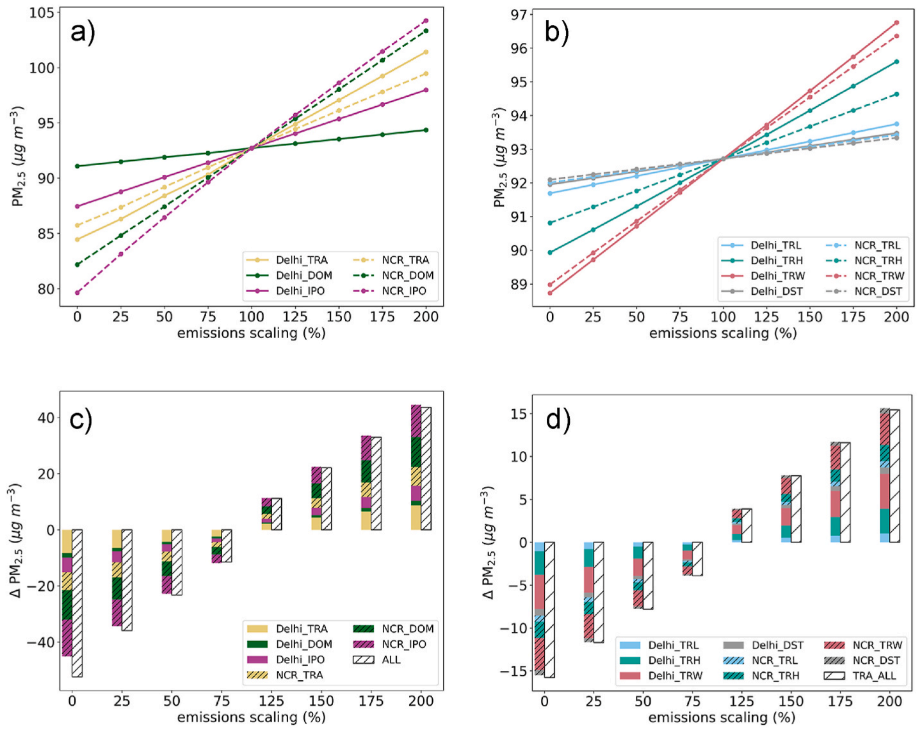

Fig. 4. Linearity test for average 24-h $PM_{2.5}$ response over Delhi due to fractional emission changes from different sectors. Panel a): response to change in emissions from individual main sectors (DOM, TRA, IPO) within Delhi (Delhi_) and the surrounding region (NCR_). Panel b): same as panel a) but for the on-road transport subsectors. Sector names are defined in Table 1. Panel c): comparison between the response obtained by summing the responses of individual change in emissions from the main sectors and the response obtained by changing emissions from the main sectors all simultaneously. 'ALL' refers to the simultaneous change of emissions for all the sectors together. Panel d): same as panel c) but for the on-road transport subsectors.

The contribution from all anthropogenic sectors considered in this study, including any non-linear terms, accounts for ~60% of the 24-h average $PM_{2.5}$ mass over Delhi. This suggests there is a large contribution originating outside of Delhi and the broader NCR region. To quantify this pollution contribution, we perform a simulation for which the boundary conditions for the nested domain (Fig. 1) are switched off. We find that the boundary conditions account for ~40% of changes in the 24-h average $PM_{2.5}$, ignoring the additional non-linear impacts. This highlights that long range transport makes a substantial contribution to surface $PM_{2.5}$ in Delhi in the post-monsoon season even before the full onset of the agricultural biomass burning.

3.3. *Impacts on 24-h average $PM_{2.5}$*

Fig. 5a shows the relative impact of each anthropogenic sector on the 24-h average $PM_{2.5}$ over Delhi, calculated from equation (1) with $\alpha = 75\%$. We find that NCR anthropogenic sources from industries and power (NCR_IPO) and domestic (NCR_DOM) have the highest impact on Delhi $PM_{2.5}$, with potential reductions from baseline $PM_{2.5}$ concentrations of 14% and 11%, respectively. The same two sectors (Delhi_IPO and Delhi_DOM) within Delhi have the lowest impacts, with potential reductions of <6%. This contrast between the local and regional sources is broadly consistent with the emission distribution from the domestic (DOM) and the industry and power (IPO) sectors. Almost all power plants, industries, and other point sources are located outside Delhi and they account for most of the IPO sector emissions. As shown in Fig. 2c, the rate of emissions from the domestic sector is similar inside and outside Delhi. However, because the area outside Delhi is much larger, domestic sector emissions from outside Delhi make a much larger contribution to regional emission totals (see Fig. 2a).

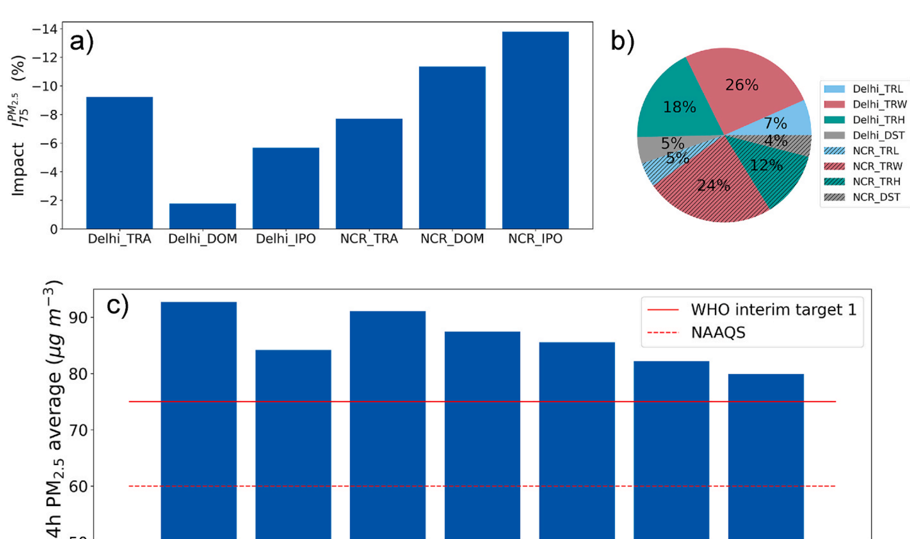

Fig. 5. Impacts of anthropogenic sectors on 24-h mean $PM_{2.5}$ over Delhi. Panel a) Relative impact $I_{[s,\alpha]\%}^{PM_{2.5}}$ for each main anthropogenic sector, calculated for each sector s as defined in Eq. (1) with $\alpha = 75\%$. Panel b) Share of subsectors to the total on-road transport (Delhi_TRA + NCR_TRA) impact reported in panel a). Panel c) Potential mitigation of $PM_{2.5}$ concentrations achievable when reducing each sector emissions by 75%, compared to the baseline $PM_{2.5}$ concentrations (BASE). The potential mitigation is benchmarked against the WHO interim target 1 (red line) and the Indian National Ambient Air Quality Standard (NAAQS) (red dotted line).

As seen in Fig. 5a, the on-road transport sector within Delhi (Delhi_TRA) has the highest impact on $PM_{2.5}$ among the local sources, with a potential reduction up to 9.2%, which is almost a third higher than the local industry and power sector and almost five times the local domestic sector. The impact from the local on-road transport sector is slightly higher than for regional sources (NCR_TRA, up to 7.7%). While emissions from local on-road transport represent almost half of emissions from regional on-road transport, traffic in Delhi remains the main hotspot for on-road transport emissions in the region, with spatially homogeneous distribution and with higher emissions rates (Fig. 2a and c), which might help explain its higher impact on Delhi $PM_{2.5}$. Two- and three-wheelers from local and regional traffic account for almost 50% of the impact of the on-road transport sector on $PM_{2.5}$, with an approximately equal contribution of local (Delhi_TRW) and regional (NCR_TRW) two- and three-wheelers (Fig. 5 b). Local heavy duty vehicle traffic (Delhi_TRH) accounts for an additional 18% of the total on-road transport impact, followed by 12% from regional heavy duty vehicle traffic (NCR_TRH). Traffic from passenger and light duty vehicles (TRL) and resuspended dust (DST) have a relatively small impact on Delhi $PM_{2.5}$, each accounting for around 10% of the total traffic impact, equally split between local and regional sources. In summary, we find that two- and three-wheelers and heavy duty vehicles collectively account for almost 80% of the total traffic impact. This is consistent with these subsectors accounting for 50% of PM and SO_2, for more than 80% of NO_x emissions, and for more than 90% of NMVOCs emissions from total on-road transport sector (Fig. 2b).

In terms of absolute $PM_{2.5}$ concentrations, we find that no one sector alone is impactful enough that its removal would improve air quality to meet the WHO interim target 1 (Fig. 5c). If the emissions from local on-road transport were eliminated completely, $PM_{2.5}$ in Delhi would be reduced to 84 $\mu g\ m^{-3}$ from a baseline concentration of 93 $\mu g m^{-3}$. If we then also removed the impact from regional on-road transport, the level of $PM_{2.5}$ in Delhi would decline to 77 $\mu g\ m^{-3}$, still above the Indian NAAQS (60 $\mu g\ m^{-3}$) and the WHO interim target 1 (75 $\mu g m^{-3}$) (WHO, 2021). We find that only a multi-sectoral, multi-scale approach targeting regional and local emissions is sufficient to reduce mean Delhi $PM_{2.5}$ values to target values. For example, if we consider the linear regime and the impacts in Fig. 4a for each sector, reducing regional domestic emissions by 75%, regional industry and power by 50% and local traffic by 50% would achieve the WHO interim target 1. If also local and regional on-road transport emissions were reduced by 75%, mean Delhi $PM_{2.5}$ would reach the Indian NAAQS threshold.

3.4. Impacts on $PM_{2.5}$ diurnal cycle

The diurnal cycle of $PM_{2.5}$ is determined by the diurnal cycle of emissions, pollutant transport, boundary layer dynamics, and atmospheric chemistry. Fig. 6 shows the percentage impact of local sectors (panel a) and regional sectors (panel b) on the mean diurnal cycle of $PM_{2.5}$ over Delhi, determined from equation (1) with $\alpha = 75\%$.

Nighttime concentrations (00:00–06:00 local time) are impacted most strongly by regional sources, with contributions of ~15–25% from industry and power, ~15% from the domestic sector, and ~10% from on-road transport. Contributions from local emission sectors are below 10%, with the largest impacts from industry and power followed by the on-road transport sector. The contribution from local domestic emissions is negligible, reflecting the large reduction of emissions from this sector during nighttime.

Continuous emissions from the energy and industry sector combined with the shallow nighttime boundary layer, which attenuates pollutant dispersion, results in the contribution from this sector peaking during the nighttime. Concentrations of $PM_{2.5}$ during daytime hours (06:00–18:00) are still mainly impacted by non-local sources from the industry, power, and domestic sectors although their contributions almost halve (reaching below 10%) towards late afternoon when the boundary layer continues to expand and mixing is over longer vertical scales. The contribution from local on-road transport increases to ~10% during peak traffic activity at 08:00 and then plateaus around 7% between 11:00–17:00. The contribution from non-local on-road transport decreases during daytime, reaching an impact of <5%, comparable to local industry and power sector. The contribution from the local domestic sector increases during the daytime to <5%. Local on-road transport makes the highest anthropogenic contribution to $PM_{2.5}$ during evening hours (18:00–00:00), reaching up to 18% at around 20:00. This is due to the evening traffic peak combined with the collapse of the boundary layer. Regional sources progressively contribute more to $PM_{2.5}$ during the evening, peaking during nighttime hours.

The impact of local on-road transport on $PM_{2.5}$ reflects typical diurnal patterns of on-road transport emissions activity, with morning and evening peaks in concentrations reflecting the corresponding traffic peak hours. The local transport contribution has on average a bigger impact than the local domestic and industry and power sectors, mainly linked to the interplay between emissions totals and spatial patterns of local sectors, as already highlighted in Sec 3.3. This result is consistent with what found in a similar modeling study in Delhi during the pre-monsoon season (Chen et al., 2020b). The contribution to $PM_{2.5}$ from regional on-road transport emissions shows a similar trend to local emissions but smoothed, reflecting the influence of PBL mixing and atmospheric transport timescales. Overall, the sum of the local and regional on-road transport sectors contributes up to 20% to $PM_{2.5}$ during nighttime hours, ~12% during the central hours of the day, and peaking at almost 30% during the evening traffic peak (Fig. 6c). Two- and

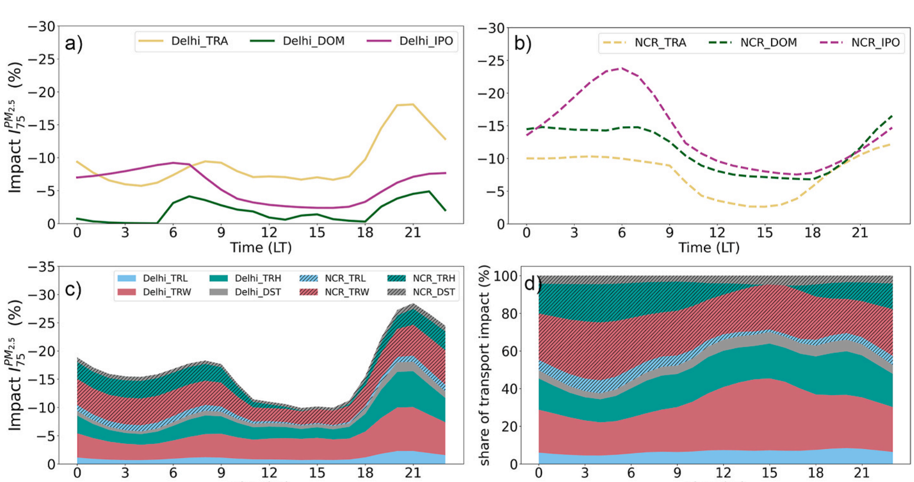

Fig. 6. Impacts of anthropogenic sectors on average hourly mean $PM_{2.5}$ over Delhi. Panel a) Relative impact $I^{PM_{2.5}}_{[s,\alpha]\%}$ for each main local anthropogenic sector, calculated for each sector s as defined in Eq. (1) with $\alpha = 75\%$. Panel b): same as panel a) but for regional anthropogenic sectors. Panel (c) Relative impact $I^{PM_{2.5}}_{[s,75]\%}$ for each of on-road transport subsector. Panel (d) Percentage hourly share among transport subsectors of the total on-road transport impact (Delhi_TRA + NCR_TRA) shown in panel c).

three-wheelers and heavy duty vehicles dominate the impact of the on-road transport sector on the diurnal cycle of $PM_{2.5}$, accounting for 60%–70% of the total impact at any hour of the day. Local two- and three-wheelers (30–40%) and local heavy duty vehicles (15–23%) have the highest share of the total on-road transport contribution during the central hours of the day and during the evening. Regional two- and three-wheelers (up to 30%) and regional heavy duty vehicles (up to 20%) dominate during the nighttime hours (Fig. 6c and d). We find minimal contributions from passenger cars and light commercial vehicles and from resuspended road dust, each accounting for <10% at any time during the day for the local and regional fleets.

3.5. Limitation and uncertainties

The methods we used to conduct our impact analysis come with some limitations. Here we summarize the main uncertainties associated with these methods and their impact on our results. Despite relying on detailed local information, the TERI/ARAI inventory has some limitations. In particular, for the transport sector, the exhaust emission factors have been developed by ARAI through lab-based testing (ARAI, 2008), which are known to underestimate emission factors from real driving conditions (Fontaras et al., 2017; Mogno et al., 2022). Although the inventory accounts for high-emitting vehicles, it assumes a 25% homogeneous increase in the transport emissions based on previous studies, lacking thus more detailed disaggregation of high-emitters across different vehicle types. Using the same diurnal profile for different vehicle categories is also an assumption in the current inventory that might underestimate the impact of heavy duty vehicles emissions, which have high activity also during nighttime. Delhi and the surrounding NCR is a fast-growing and changing area, and using emissions estimates of year 2016 in the inventory might not be fully representative of the emissions for 2019 and of subsequent years. For example, in 2020 there has been a major update of vehicle emissions standards (from Bharat Stage III to Bharat Stage VI) and there is the plan to phase out coal power plants in the NCR starting from 2023 (CAQM). These interventions can be expected to make significant changes in the emissions from the transport and the industry sectors in the coming years, and as a consequence the sector contribution and mitigation strategies presented in this study need to be reassessed in the future. At this regard, the current inventory for the NCR could be improved in the future by updating emissions factors and activity data using estimates from more recent studies (Singh et al., 2018; Conibear et al., 2020; Hakkim et al., 2021). Moreover, there is a need to develop chemical speciation of profiles of NMVOC emissions from different sources (Sharma and Khare, 2017). This recently started to be addressed for different sectors in India (Sarkar et al., 2016; Stewart et al., 2021; Hakkim et al., 2022). Including similar NMVOC speciation profiles in the TERI/ARAI inventory would be an important update, especially for better quantifying the anthropogenic impact on secondary particulate matter. Our results are reported for city-scale mean, so they might not be representative of the sub-city scale, where the different source attribution and thus potential impact reductions may be different. Indeed, socio-economical factors also play an important role in determining high air pollution inequalities within the city (Lane et al., 2022; Nguyen and Marshall, 2018; Fecht et al., 2015). In addition, mean values tend to drive the responsibility towards regional action that might be less effective in solving the pollution issue in different parts of the city (Thunis et al., 2021).

4. Discussion and conclusions

We studied the contribution of the on-road transport sector to surface $PM_{2.5}$ over Delhi during the post-monsoon season using the WRF-Chem model. By using the WRF-Chem model we were able to find that the response between changes in emissions from different emissions sectors and $PM_{2.5}$ concentrations is linear at the city level scale during post-monsoon. We defined the range of validity of the linear response, which holds for reduction in emissions within 75%. We calculated the impacts of the transport sector and its subsectors compared to other local (within Delhi) and regional (within NCR) sources on both mean 24-h and diurnal cycle of $PM_{2.5}$ in Delhi, and quantified mitigation strategies to achieve $PM_{2.5}$ air quality standards in the city during the post-monsoon season. Although *a posteriori* the response of $PM_{2.5}$ to emissions changes has proven to be linear, our results cannot be captured by the soley analysis of the emissions inventory, highlighting the added value of using a chemical transport model for emission reduction and mitigation studies. We found that local on-road transport sectors contribute less than 10% to 24-h mean $PM_{2.5}$ values over Delhi during the post-monsoon season, and if we add the regional contribution, the total impact of the on-road transport sector increases to 17%. The highest impact from individual regional sectors is from power and industry (14%) and domestic (11%), while the local power and industry and domestic sectors have minimal impact (6% each). This is consistent with previous studies that showed that on a city-scale regional sources from the NCR can have a significant impact on annual mean $PM_{2.5}$ values over Delhi (Amann et al., 2017) and during the pre-monsoon season (Chen et al., 2020b). Long range transport from outside the NCR also contributes substantially to $PM_{2.5}$ over Delhi for the period considered in our simulations (~40%), which is comparable to the ~30% annual average long range transport contribution found by Amann et al. (2017). Within the on-road transport sector, we found that two- and three-wheelers dominate the on-road transport contribution to ambient $PM_{2.5}$ in Delhi, with around 50% share of the total on-road transport impact for the joint regional and local fleet. Heavy duty vehicles contribute 18% for the local and 12% for the regional fleet. Light commercial vehicles, including passenger cars and light duty vehicles, and resuspended dust from on-road transport individually contribute about 10% of the total impact. We showed that targeting emissions reductions from only the on-road transport sector would fail to bring $PM_{2.5}$ down to meet relevant ambient air quality standards (namely WHO interim target 1 or Indian NAAQS). Similar results are found for all the other individual sectors we studied. Only regional cooperation with stringent emissions reductions (50–75%) of the NCR industrial and energy and domestic sectors, jointly with the on-road transport sector (local and regional) has the potential to meet ambient air quality standards. The recent order from the Commission for Air Quality management (CAQM) to phase out of coal from across Delhi and the NCR from January 2023 is a significant step in this direction (CAQM). When considering the impact of each sector on the average diurnal cycle of $PM_{2.5}$ over Delhi, we found that the industry and power sectors from NCR dominated nighttime and almost all daytime concentrations (10–25% day/night), with significant contributions from the NCR domestic sector (7–15% day/night). The local on-road transport sectors have the highest absolute impact (18%) during the evening traffic peak (18:00–21:00). If the contribution from the regional on-road transport sector is also included the total impact from the traffic sector reaches almost 30%. We found that within the on-road transport sector, two- and three-wheelers dominate the on-road transport contribution to daily mean $PM_{2.5}$ values, followed by heavy duty vehicles. Two- and three-wheelers and heavy duty vehicles combined account for 60–70% of the total on-road transport impact on $PM_{2.5}$ throughout the day, with local two- and three-wheelers accounting between 30 and 40% of the total on-road transport hourly impact on $PM_{2.5}$ during daytime hours. These findings suggest that prioritising reduction of emissions from two- and three-wheelers (e.g., converting to electric vehicles), and heavy duty vehicles (e.g., converting to cleaner fuel such as compressed natural gas), could be an efficient way to reduce the impact of the on-road transport sector on $PM_{2.5}$, particularly during the evening traffic peak. Our results also help to explain why the even-odd pilot mitigation strategies have yet to deliver the expected benefit on surface air quality. We find that the targeted on-road transport subsector of passenger cars play only a minor role in on-road transport sector emissions during the post-monsoon season (and this will likely hold true for other seasons). Our findings can therefore help inform the development of more effective mitigation strategies for $PM_{2.5}$ over Delhi during the post-monsoon season. However, if mitigation strategies focus exclusively on minimising $PM_{2.5}$ concentrations, elevated levels of surface ozone (O_3) could become more of a significant health concern. Because ozone forms through non-linear pathways from its precursors in the troposphere, reductions in some ozone precursors emissions which are also precursors for $PM_{2.5}$, may reduce $PM_{2.5}$ concentrations but enhance O_3 pollution. In particular, reductions in NOx emissions, emitted mainly from the transport sector, may lead to large increases in ozone concentrations in a VOC-limited regime such as in Delhi if VOC emissions are not sufficiently reduced (Ojha et al., 2022; Chen et al., 2021; Nelson et al., 2021; Tiwari et al., 2015). We also report that decreasing emissions from the transport sector increases surface ozone concentrations while reducing $PM_{2.5}$ concentrations. Reduction in emissions from other sectors led to similar (but smaller) increase in ozone (Fig. S10). Through a simplified linear model, we found that for minimising the total health impacts when considering both $PM_{2.5}$ and O_3 during the post-monsoon season, emission reductions from domestic and industry and power sectors should be prioritised over reducing emissions from the on-road transport sectors (details in the Supplementary Material). This is because in our calculation we assume that on-road transport emissions are reduced uniformly (i.e., all pollutants by the same amount), and the negative health impact resulting from the increase in ozone might be higher than the benefit of reducing $PM_{2.5}$ for the transport sector. Although our estimate is a simple calculation, it highlights the importance of the coordinated control of $PM_{2.5}$ and ozone. This is important in particular in VOC-limited urban environments such as Delhi, where reduction of emissions, especially from transport sector, could increase the adverse health impact from rising of ozone levels. Future work is needed to characterise $PM_{2.5}$ at finer spatial and temporal scales over Delhi with up-to-date high resolution emissions inventories, in order to establish which controls can be effective at reducing pollution at the neighbourhood level. In addition, sensitivity studies within the city will need to consider the location-dependent photochemical environment that will also help determine how the balance of emission changes will influence surface ozone production, to inform effective and impactful emissions control strategies that maximise health benefits for Delhi residents.

Code and data availability

The WRF-Chem model code is available from https://www2.acom.ucar.edu/wrf-chem. NCEP FNL global tropospheric meteorological analyses were taken from https://rda.ucar.edu/datasets/ds083.3/. CAM-CHEM global model results were downloaded from https://www.acom.ucar.edu/cam-chem/cam-chem.shtml. The FINN biomass burning emissions dataset was downloaded from https://www.acom.ucar.edu/Data/fire/. The original EDGAR-v5.0 anthropogenic emissions dataset is available at https://data.jrc.ec.europa.eu/dataset/377801af-b094-4943-8fdc-f79a7c0c2d19. The gridded TERI/ARAI emission inventory for the NCR is available upon request to TERI. Model setup files and code scripts for the analysis described in the paper are available in the Zenodo repository https://doi.org/10.5281/zenodo.7498969.

CRediT authorship contribution statement

Caterina Mogno: Conceptualization, Methodology, Investigation, Validation, Formal analysis, Software, Visualization, Data curation, Writing – original draft, Writing – review & editing. **Paul I. Palmer:** Conceptualization, Supervision, Funding acquisition, Writing – review & editing. **Margaret R. Marvin:** Methodology, Data curation. **Sumit Sharma:** Resources, Methodology, Writing – review & editing. **Ying Chen:** Conceptualization, Methodology, Writing – review & editing. **Oliver Wild:** Conceptualization, Methodology, Writing – review & editing.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Caterina Mogno reports financial support was provided by Ford Motor Company. Paul I. Palmer is part of the Advisory Editorial Board of Atmospheric Environment: X.

Data availability

Code and Data availability Section is included in the manuscript.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.aeaoa.2022.100200.

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