Quantifying effects of long-range transport of NO₂ over Delhi using back-trajectories and satellite data

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Abstract.

Exposure to air pollution is a leading public health risk factor in India, especially over densely populated Delhi and the surrounding Indo-Gangetic Plain. During the post-monsoon months, the prevailing north-westerly winds are known to influence aerosol pollution events in Delhi, by advecting pollutants from agricultural fires as well as from local sources. Here we investigate the year-round impact of meteorology on gaseous nitrogen oxides (NOₓ = NO + NO₂), a hazardous primary air pollutant for health, which can lead to the formation of secondary aerosols and ozone. We use bottom-up NOₓ emission inventories (anthropogenic and fire) and high-resolution satellite measurement based tropospheric column NO₂ (TCNO₂) data, from S5P on-board TROPOMI, alongside a back-trajectory model (ROTRAJ) to investigate the balance of local and external sources influencing air pollution changes in Delhi, with a focus on different emission sectors. Our analysis shows that accumulated emissions (i.e. integrated along the trajectory path, allowing for chemical loss) are highest under westerly, north-westerly and northerly flow during pre- (February - March) and post- (October - January) monsoon periods. During the pre-monsoon period, the residential and transport sectors together account for more than 50% of the total accumulated emissions, which are dominated by local sources (90%) under easterly winds and by non-local sources (>70%) under north-westerly winds. The high accumulated emissions estimated during the pre-monsoon season under north-westerly wind directions are likely to be driven by high NOₓ emissions locally and in nearby regions (since NOₓ lifetime is reduced and the boundary layer is relatively deeper in this period). During the post-monsoon period non-local (60%) transport emissions are the largest contributor to the total accumulated emissions as high emissions, coupled with a relatively long NOₓ atmospheric lifetime and shallow boundary-layer aid the build-up of emissions along the trajectory path. Analysis of surface daily NO₂ observations indicates that high pollution episodes (> 90th percentile) occur predominantly in the post-monsoon and more than 75% of high pollution events are primarily caused by non-local sources. Overall, we find that in the post-monsoon period, there is a substantial import of NOₓ pollution into Delhi with a large contribution from the transport sector. This work indicates that the advection of highly polluted air originating from outside Delhi is of concern for the population and air quality mitigation strategies need to be adopted not only in Delhi but in the surrounding regions to successfully control this issue. In addition, our analysis suggests that the largest benefits to Delhi NOₓ air
quality would be seen with targeted reductions in emissions from the transport sector, particularly during post-monsoon months.

1 Introduction

The Indo-Gangetic Plain (IGP) covers ~20% of the Indian subcontinent geographical area and houses almost 40% of the population, producing >40% of the total food grain (Badarinath et al., 2009). Within the region, much of the agricultural land is used for rice and wheat production, with ~12 million ha in a wheat-rice crop pattern (Badarinath et al., 2009). Each year there are two major growing seasons; rice is grown from May-September, with harvesting in October-November, and wheat is grown from November-April, with harvesting in April-May. Since the 1980s there has been a shift towards machine harvesting of crops (Badarinath et al., 2009; Jethva et al., 2019). However, this method leaves a large portion of the crop stem root-bound and difficult to remove quickly before sowing the next crop. Wheat stubble is valued for animal feed and a considerable fraction is removed for this purpose before burning. However, rice stubble is generally burned to clear the land quickly as burning is more economically viable than manual removal (Ahmed et al., 2015). Recently Kumar et al. (2021) estimated that a total of ~17,000 Tg of rice stubble is burnt each year in India. As a result, burning of rice stubble in the north-west IGP alone accounts for almost 85% of biomass burning in India seasonally and almost 20% annually (Jethva et al., 2019).

The timing of agricultural waste burning has shifted over the past decade due to the introduction of the Subsoil Water Act (SSWA). The SSWA was passed by the government of Punjab in 2009 and requires farmers to delay sowing and transplantation of rice until mid-May and mid-June, respectively (Singh, 2009). The law was introduced to reduce ground water demand by bringing the rice-growing season into line with the arrival of the summer monsoon rainfall (Singh, 2009). However, the SSWA shortened the time period between rice harvest and wheat sowing, resulting in an increased reliance on rice stubble burning to clear land more quickly (Jethva et al., 2019). This has been observed through shifts in the timing of burning from early to late October and an increase in the prevalence of fires (Sembhi et al., 2020; Kant et al., 2022; Liu et al. (2021)).

During the post-monsoon months of October and November synoptic meteorology is favourable for the advection of air pollution from agricultural burning towards Delhi and surrounding areas (e.g. Mor et al., 2022; Jethva et al., 2018). Bikkina et al. (2019) performed cluster analysis of back-trajectories released from Delhi and found that north-west IGP emissions have a large (80-100%) influence on air quality in Delhi during autumn (October–November) and winter months (January–February). Additionally, a shallow boundary layer over Delhi during autumn and winter months, low wind-speeds (1-3 ms⁻¹) and poor ventilation and mixing means air pollutants from the IGP advected towards Delhi remain close to the surface, aiding the build-up of pollutants. Previous studies have found that ~40% of black carbon loadings in Delhi originate from crop residue and other biomass burning sources in October and November (e.g. Bikkina et al., 2019; Kanawade et al., 2020).

Delhi is one of the world’s largest cities, with a population exceeding 11 million (Census of India, 2011), experiencing some of the poorest air quality in the world (Pandey et al., 2021). A rapid population growth coinciding with the ever-expanding transportation and city infrastructure has made Delhi one of the most polluted megacities in India (Singh et al., 2021), and indeed the world (Kumar et al., 2017; Hama et al., 2020), suffering ~10k cases of premature mortality per year caused by air pollution (Chen et al., 2020). Winter air pollutant concentrations are particularly high, with daily mean particulate matter with a diameter less than 2.5 micrometres (PM$_{2.5}$) concentrations of 100-200 µg m$^{-3}$ (Singh et al., 2021), peaking at over 1000 µg m$^{-3}$ (Bikkina et al., 2019). As a result of the high pollution levels in Delhi several policies have been implemented over the past few decades (Guttkunda et al. (2023)). For example, an odd-even traffic intervention was brought in during winter and summer in 2016, allowing only odd and even number plates to be used on alternate days. However, analysis of surface observational data did not show any clear concentration reduction in PM$_{2.5}$ (Kumar et al., 2017) highlighting one of the key challenges in this
region: a lack of understanding surrounding the impacts of emissions from the wider region on air quality in Delhi, which prevents effective policy implementation.

Several previous studies have used back-trajectories and chemical transport models to link high pollutant concentrations in selected cities to the arrival of air masses which had passed over high emission regions (Reddington et al., 2014; dos Santos et al., 2021; Wehner et al., 2008). Reddington et al. (2014) found that during high pollution episodes, linked with biomass burning, increased concentrations of related tracer species (e.g. levoglucosan) were observed (Atwood et al., 2013; Engling et al., 2014). Similarly, back-trajectory methods and weather typing have also been applied to assess the contribution of local emissions and long-range transport to regional and city-level air pollution in many European regions (Graham et al., 2020; Pope et al., 2016, 2014; Stirling et al., 2020). For example, Pope et al. (2014) sampled satellite NO\textsubscript{2} under different synoptic weather patterns (Lamb weather types, LWTs) to show that UK NO\textsubscript{2} is generally increased under winter-time anti-cycloonic conditions through pollutant accumulation. NO\textsubscript{2} was also enhanced under south-easterly flow due to long-range transport of pollutants from continental Europe. The wintertime increase was attributed to the combined effect of increased emissions, more stable meteorological conditions and decreased photolysis, allowing accumulation over emission sources. Subsequently Stirling et al. (2020) and Graham et al. (2020) extended this methodology to analyse changes in NO\textsubscript{2} and PM\textsubscript{2.5} concentrations. They found that long-range transport of NO\textsubscript{2} and PM\textsubscript{2.5} played a larger role in cities in the south of the UK than the north, with the highest contribution from long-range transport under easterly, south-easterly, and southerly flows.

Here our approach is similar to Stirling et al. (2020) and Graham et al. (2020). We combine satellite tropospheric column NO\textsubscript{2} (TCNO\textsubscript{2}) (top-down) and anthropogenic and fire emissions of NO\textsubscript{x} (bottom-up) with back-trajectories released from Delhi during 2017 and 2018. We aim to quantify the contribution of local and non-local emissions sources under different seasons and wind directions. Studies such as this have proved useful elsewhere in the world (e.g. Grundström et al., 2015, 2017; Pope et al., 2014, 2016; Tang et al., 2009), identifying source sectors and air masses contributing to air pollution and allowing for targeted emissions reductions. However, there have been very few source contribution studies in India. We then go on to identify key source sectors contributing to poor NO\textsubscript{x} AQ in Delhi by individually excluding sectors from the bottom-up anthropogenic emissions dataset. Section 2 describes the datasets and method used, Section 3 presents our results and Section 4 summarises the implications of our findings.

2. Methods

2.1. Anthropogenic NO\textsubscript{x} emissions

2.1.1. Delhi Emissions

We created a merged emissions dataset containing both anthropogenic and daily fire emissions (Figure 1f). Anthropogenic NO\textsubscript{x} emissions for Delhi are taken from the 2018 System of Air Quality and Weather Forecasting And Research (SAFAR) dataset (Beig, 2018) (Figure 1a). Data for the SAFAR emission inventory were collected during a 37,500-hour campaign and is provided at very high (400 m) resolution over a region covering 70 km x 65 km. Emissions are provided for 26 different source sectors. Emissions are very detailed, for example emissions for transport were calculated using traffic volume collected by click counters across the region. Collected data was then converted into emissions using a Geographical Information System (GIS)-based statistical model. Since SAFAR provides a much more detailed, and up-to-date, inventory of emissions in Delhi we replace EDGAR-HTAP2 Delhi emissions (see 2.1.2 for more details) with SAFAR emissions, which we regrid from 400 m to 10 km resolution.

2.1.2. Global Emissions
We combine Delhi-specific NOx emissions from SAFAR with global NOx emissions from the Emission Database for Global Atmospheric Research with Task Force on Hemispheric Transport of Air Pollution (EDGAR-HTAP2) 2010 dataset (Janssens-Maenhout et al., 2015), which are at 10 km resolution (Figure 1c). A comparison between emissions over Delhi from the SAFAR and EDGAR-HTAP2 emissions datasets is also shown in Figure 1a and 1b. This clearly demonstrates the SAFAR dataset provides more spatial detail than EDGAR-HTAP2. EDGAR-HTAP2 is a global, gridded, air pollution emission inventory compiled using officially reported, national gridded inventories; if national emissions datasets for specific sectors were not available EDGAR v4.3 grid maps are used. The resulting EDGAR-HTAP2 dataset provides monthly and annual emission distribution along with emission factors that are fuel-, technology-, process- and human activity-dependent. Emissions include all anthropogenic emissions except large-scale biomass burning (e.g. wildfires).

2.1.3. Fire Emissions

Wildfire NOx emissions for 2018 are taken from the Fire Inventory from NCAR (FINNv2.5) dataset (Wiedinmyer et al., 2011) at 10 km resolution (Figure 1e) and merged with the anthropogenic emissions dataset (to generate Figure 1f). FINNv2.5 combines data from the Moderate Resolution Imaging Spectroradiometer (MODIS) sensors on NASA’s Terra and Aqua satellites with Visible Infrared Imaging Radiometer Suite (VIIRS) on Suomi NPP. We chose to use FINNv2.5 because of its improved ability to detect small, low temperature fires which are likely to be important in India (e.g. agricultural burning). Both MODIS and VIIRS use a Thermal Anomalies Product to provide detections of active fires. MODIS detections are provided at 1 km resolution, while VIIRS has a resolution of 375 m. In FINN, fire hotspot detections from MODIS and VIIRS are combined with land cover, biomass consumption estimates and emissions factors to calculate daily fire emissions globally at 1 km resolution. Burned area is assumed to be 1 km² or 0.14 km² for each fire identified by MODIS or VIIRS, respectively, and scaled back based on the density of vegetation from the MODIS Vegetation Continuous Fields (VCF) (i.e. if 50% bare = 0.5 km² or 0.07 km² burned area). The type of vegetation burned during a detected fire is determined using the MODIS Collection 5 Land Cover Type (LCT). Each fire pixel is assigned to one of 16 possible land cover/land use. The 16 land cover types are then aggregated into eight generic categories to which fuel loadings are applied (Wiedinmyer et al., 2011). Fuel loadings are from Hoelzemann et al. (2004) and emissions factors are from Akagi et al. (2011), McMeeking (2008) and Andrae and Merlet (2001). FINN includes all emissions from above-ground vegetation but not from the combustion of peat (Kiely et al., 2019). Fire types included are wildfires, prescribed and agricultural burning. However, trash-burning or biofuel use are not included.

The merged emission dataset described above provides the control scenario in which all sectors are included (Figure 1f). Additionally, a further 6 emissions files were generated where individual sectors are excluded (transport, industry, power, residential, other and fires) to quantify their contribution to accumulated NOx along the trajectory path.

2.2. Satellite data

We use tropospheric column nitrogen dioxide (TCNO₂) measurements for February 2018 to January 2020 from the TROPOMI instrument on-board ESA’s Sentinel-5 Precursor (S5P) satellite that was launched on 13th October 2017. TROPOMI is a hyper-spectral nadir-viewing imager with an equator overpass time at ascending node of 13:30. The NO₂ columns are derived using TROPOMI’s UVIS spectrometer backscattered solar radiation measurements in the 405–465 nm wavelength range (van Geffen et al., 2015, 2019). The swath is divided into 450 individual measurement pixels, which results in a near-nadir resolution of 7.0 km × 3.5 km. The total NO₂ slant column density is retrieved from the Level 1b UVIS radiance and solar irradiance spectra using Differential Optical Absorption Spectroscopy (van Geffen et al. 2019). Tropospheric and stratospheric slant column densities are separated from the total slant column using a data assimilation system based on the TM5-MP chemical transport model, after which they are converted into vertical column densities using a look-up table of altitude-dependent air-mass factors. As
data was available from February 2018 onwards, we selected the two years (2018-2019) closest to the back-trajectories (2017-2018) and anthropogenic (2010/2018) and fire emissions (2018) for our analysis. We follow the approach of Pope et al. (2018) to map TROPOMI TCNO₂ data onto a 0.05° × 0.05° grid over India.

2.3. Back Trajectories

Our approach is to combine back trajectories with either top-down satellite TCNO₂ or bottom-up NO₃ emission estimates to investigate the influence of long-range transport (advection) of NO₃ on ambient concentrations under different wind-directions in Delhi. In the following section we will refer to bottom-up NO₃ emissions, however the method is the same when using satellite TCNO₂.

We use primary NO₃ emissions integrated over air mass back-trajectories to determine the relative influence of direct NO₃ emissions on those air masses. Back-trajectories are calculated using the Reading Offline TRAJectory (ROTRAJ) Lagrangian transport model (Methven, 2003). The model uses dynamical fields from ERA-Interim reanalysis from the European Centre for Medium-Range Weather Forecasts (ECMWF) to calculate trajectories at 1.125° horizontal resolution. After a trajectory parcel is released, its location is calculated every 6 hours; for vertical interpolation the model uses cubic Lagrangian interpolation and horizontal fields are calculated using bilinear interpolation. This approach primarily focuses on large-scale advection and does not resolve small-scale sub-grid turbulent transport or convection.

In this study, ROTRAJ back trajectories were released from just above the surface (0.99 sigma level; a terrain following coordinate system where 1 is the surface) in central Delhi at 06:00 UTC (11:30 local time) for the years 2017 and 2018, extending back 4 days in 6-hour time steps. We choose 06:00 UTC (11:30 local time) as this is close to mid-day local time and so matches the time of the satellite overpass time (13:30 local time) most closely. We are restricted to using ROTRAJ back-trajectories for 2017 and 2018, while using satellite data for 2018 and 2019, as the ECMWF ERA-Interim reanalysis dataset was terminated in August 2019 and satellite data is not available until 2018. NO₃ emissions were accumulated along each trajectory over 4 days at 15-minute time intervals (interpolated linearly from 6-hourly position output) (Figure 2).

NO₃ emissions were only accumulated only if the trajectory path was within the boundary layer (based on ERA-Interim). At each location, we accumulate the entire emission within an emission grid box (TCNO₂: ∼5 km, emissions: ∼10 km) over which the trajectory passes. The surface area of each grid box that the trajectory points passed over is also accumulated over time.

The along-trajectory emission accumulation can be represented by Equation (1):

\[ E_i = [E_{i-1} + \alpha \Delta t] e^{-\Delta t/\tau_i} \]  

\[ \text{where } i=1, N (=384) \text{ and } E_0=0.0 \]

\[ E_i \text{ is accumulated NO}_3 \text{ (kg) at any given point } i \text{ along the trajectory, } \phi_i \text{ is the emissions flux of NO}_3 \text{ (kg m}^{-2} \text{ s}^{-1}) \text{ at point } i, \Delta t \text{ is the 15-minute time step, } \alpha \text{ is the surface area of the grid box (m²) at point } i \text{ and } \tau_i \text{ is the specified NO}_3 \text{ lifetime (t) at point } i. \text{ Therefore, } E_N \text{ is total accumulated NO}_3 \text{ mass (kg) and } N \text{ is the number of 15-minute time steps within the 4-day trajectory (384).} \]
To account for chemical loss of NO\textsubscript{x} along the trajectory path, the lifetime of NO\textsubscript{2} was calculated at each timestep from TOMCAT (Chipperfield, 2006; Monks et al., 2017) 3-D hourly hydroxyl radical (OH), pressure and temperature fields, assuming the main loss pathway in Equation (2), where NO\textsubscript{2} is oxidised by OH to form nitric acid (HNO\textsubscript{3}) (which dominates over photolysis within the boundary layer). The NO\textsubscript{2} lifetime ($\tau$) (Equation (3)) was calculated using a temperature and pressure-dependent rate constant ($k$) (IUPAC, 2022). The calculated lifetime ($\tau$) was then applied to the total NO\textsubscript{x} accumulated emission in the air parcel in Equation (1).

\[
\text{OH + NO}_2 + M \rightarrow \text{HNO}_3 + M \quad (2)
\]

\[
\tau = \frac{1}{k[\text{OH}]} \quad (3)
\]

To remove the dependence of the accumulated emissions calculated in Equation (1) on emission grid resolution (since we assumed the air mass has the same width as the emission grid box), the total accumulated NO\textsubscript{x} mass ($E$) was divided by accumulated surface area ($S$) and then scaled by $10^9$ to give $E$ units of $\mu$g m\textsuperscript{-2}. $S$ is given by Equation (4):

\[
S = \sum_{i=1}^{N} a_i \quad (4)
\]

Emissions accumulated within Delhi ($E_{Delhi}$) were also determined using the same approach, but only implemented when the trajectories enter the Delhi region, which we define using a bounding box (76.8 - 77.5\textdegree E, 28.1 - 28.9\textdegree N). To derive $E_{Delhi}$ in units of $\mu$g m\textsuperscript{-2}, the accumulated NO\textsubscript{x} mass from Delhi was divided by the accumulated surface area ($S$) over the full trajectory path. The ratio between $E_{Delhi}/E$ represents the fractional contribution of Delhi sources towards the total accumulated NO\textsubscript{x} emissions.

Finally, the daily (06:00 UTC) total accumulated emission and $E_{Delhi}/E$ ratios from all sites were binned by the wind direction. This methodology provides a powerful tool to identify which flow directions are the most polluted and to derive the proportion of pollutant emissions from long-range transport versus local sources.

2.4 Observational Data

The hourly averaged surface NO\textsubscript{2} concentration at 36 sites over Delhi for 2018 and 2019 (Figure 3) were obtained from the CAAQMS (Continuous Ambient Air Quality Monitoring Stations) portal (https://app.cpcbccr.com/ccr/#/caaqm-dashboard-all/CAAQM-landing) of the Central Pollution Control Board (CPCB) of India. The ambient NO\textsubscript{2} concentration was measured based on the Gas Phase Chemiluminescence methodology; the technical details of monitoring can be found in CPCB (2019). The data was further quality controlled to remove outliers and missing values (Singh et al., 2020). As TROPOMI has a local overpass time of approximately 13:30, the average NO\textsubscript{2} between 13-14 hours was calculated for each site to represent the daily NO\textsubscript{2} corresponding to the overpass of TROPOMI. From this, the daily median NO\textsubscript{2} concentration across all sites was calculated to represent daily NO\textsubscript{2} levels over Delhi in 2018 and 2019.

3. Results

The contribution of local and non-local sources to NO\textsubscript{2} levels in Delhi are controlled by primary emissions, the chemical lifetime of NO\textsubscript{2} and advection by meteorology (i.e. accumulation/advection of
pollutants under stable conditions or ventilation of source emissions downwind). Figure 4b, c and d show the annual average TCNO$_2$, surface NO$_3$ lifetime and boundary layer height (BLH) over India along with the anomalies for the pre-monsoon (February - April), monsoon (May - September) and post-monsoon (October - January) seasons.

Peak annual TCNO$_2$ ranges between 4000 and 5000 $\mu$g m$^{-2}$ over the source regions (e.g. Delhi, Kolkata, Nagpur and industrial sources in the east (approximately 80-85°E, 20-25°N)) (Figure 4b). Smaller urban NO$_2$ hotspots range between 3000 and 4000 $\mu$g m$^{-2}$. In the seasonal anomaly, the pre-monsoon TCNO$_2$ values increase by approximately 500-1000 $\mu$g m$^{-2}$ around Delhi but with similar decreases to the northwest. However, the majority of the country experiences TCNO$_2$ increases greater than 2000 $\mu$g m$^{-2}$. During the monsoon season there is a decrease in TCNO$_2$ by 1000-1500 $\mu$g m$^{-2}$ over Delhi, which is reflected in the majority of other urban centres and industrial regions. The post-monsoon season experiences the largest degradation in NO$_2$-related air quality as all hotspots increase by $>$2000 $\mu$g m$^{-2}$ (e.g. for Delhi) with peak enhancements over the industrialised region in the east of India.

The calculated NO$_2$ lifetime (based on modelled OH, temperature and pressure) typically ranges between 1.5 to 15 hours over India (between 20 and 24 hours over Bangladesh and Myanmar) and 8 to 12 hours over Delhi (Figure 4c). During the pre-monsoon, there is little change from the annual average, decreasing by 1 to 2 hours over Delhi. Along the Indian east coastline, there is a general decrease of 3-5 hours. During the monsoon season, there are large reductions in the NO$_2$ lifetime over the northern segment of the domain, decreasing by 5 to 10 hours, propagating southwards to Delhi with a decrease of approximately 3-5 hours. Central India remains similar to the annual average while the east coastline now experiences increases in the NO$_2$ lifetime of 3 to 5 hours. In the post-monsoon, while southern India experiences little change, Delhi and the north of India see an increase in the lifetime by 3-7 hours.

Western India has an annual average BLH ranging between 800 and 1100 m (400-800 m over eastern India), while Delhi BLH is between 400 and 600 m (Figure 4d). In the pre-monsoon, the Indian boundary layer is well ventilated with an increase in BLH by $-$200 to 400 m (50-100 m for Delhi). In the monsoon season, the boundary layer remains well ventilated with enhancements of typically 100 to 200 m (although with some regions of reduction in the BLH), relative to the annual average. In the post-monsoon season, colder temperatures cause country-wide shallowing of the boundary layer by 200 to 400 m, including Delhi, peaking at over 500 m in central India.

The pre-monsoon and, especially, post-monsoon season conditions are favourable for the degradation in NO$_2$ air quality given the typically larger primary NO$_3$ emissions (e.g. from increased domestic heating and power demands), the longer NO$_2$ lifetime (i.e. less chemical loss) and shallower boundary layer trapping emissions over Delhi and the wider IGP. In contrast, the lower emissions, shorter NO$_2$ lifetime and atmospheric ventilation of the boundary layer yield lower pollution levels during the monsoon season.

We have thus demonstrated the seasonal influences on NO$_2$ levels in Delhi and surrounding region but now exploit the back-trajectory integrated emissions methodology (see Section 2) to determine the balance of local versus non-local sources of NO$_2$ to Delhi air quality. Here, we accumulate TCNO$_2$ from TROPOMI under the back trajectories to investigate the seasonal influence of wind direction on the advection of TCNO$_2$ into Delhi. As discussed above, there is a clear seasonal influence of the monsoon circulation on the back-trajectories integrated emission totals.

On average, the back-trajectories arrive into Delhi at the surface from the south-west, east and north-west (Figure 4a). Trajectories from the south-west have the lowest accumulated TCNO$_2$ levels (0-200 $\mu$g m$^{-2}$), largely originating over the Arabian Sea (i.e. fewer NO$_3$ sources) before arriving in Delhi. Trajectories originating from eastern India pass over several large urban centres and industrial regions yielding integrated TCNO$_2$ trajectories with TCNO$_2$ levels that range between 400 to 700 $\mu$g m$^{-2}$ before arriving in Delhi. The trajectories from the north-west are the most polluted, with peak TCNO$_2$ levels of $>$1000 $\mu$g m$^{-2}$. When split seasonally, the pre-monsoon shows trajectories approaching Delhi from the north-west (200-400 $\mu$g m$^{-2}$) and east (600-900 $\mu$g m$^{-2}$). In the monsoon, cleaner airmasses (0-200 $\mu$g m$^{-2}$) originate from the south-west while eastern airmasses remain similarly polluted as the pre-monsoon (600-
800 µg m⁻²). During the post-monsoon, most trajectories are from the north-west of India and show the largest seasonal pollution levels (i.e. integrated TCNO₂ back-trajectories of typically 800 to >1000 µg m⁻²).

We repeated this approach using the bottom-up emissions inventory (Figure 5) with close agreement to the TCNO₂ results. In the pre-monsoon season, the integrated emission back-trajectories range between 80 and 120 µg m⁻² from both the east and north-west. Note the emission trajectory values are lower than the equivalent TCNO₂ values as they are emission fluxes and not the tropospheric integrated column. In terms of altitude, the trajectories range from the near-surface to 400 hPa. At t=96 hours before arrival at Delhi, the median trajectory position is near 800 hPa before descending below 950 hPa at approximately 30 hours from Delhi. In the vertical distribution, there is no clear link between the integrated emission trajectory values and altitude. During the monsoon season, there are two limbs of integrated emission trajectories with TCNO₂ ranging between 0 and 40 µg m⁻² from the south-west and 40-80 µg m⁻² from the east. The average trajectory position is lower in altitude, starting at approximately 950 hPa and remaining below this pressure level. There appears to be a split in the trajectory origins and altitudes with south-western trajectories coming from the Arabian Sea near the surface with few emissions sources (airmasses will be influenced to some extent by shipping emissions but the short NO₂ lifetime will yield limited impact in Delhi), while more polluted trajectories (i.e. substantial upwind sources) from the east are typically 900-600 hPa in altitude. During the post-monsoon season, the median trajectory altitude is approximately 875 hPa at 96 hours upwind of Delhi. For the final 24 hours or so, the trajectories converge on Delhi below 950 hPa. As a result, while all seasons experience trajectories 1-day out from Delhi below 950 hPa, the post-monsoon trajectories are closer to the surface than the pre-monsoon equivalent throughout the 4 days and are exposed to larger emission fluxes than those during the monsoon. Secondly, as these post-monsoon trajectories originate from the north-west and are trapped in the IGP by the Himalayas, there is the opportunity for re-circulation of the trajectory over up-wind NO₂ sources. Overall, the meteorological, chemical and emission factors yield, all trapped against the Himalayas, in the post-monsoon season, are key for the substantial degradation in the air quality.

The integrated emission trajectories also provide the opportunity to gain better insight about the proportion of local vs. non-local sources (i.e. integrated trajectories over the full 4 days vs. integrated trajectories just over Delhi). Figure 6 shows this but note we only plot wind directions where the sample size is greater than or equal to 10. We find that during the pre-monsoon season, the grouped integrated emissions trajectories are moderately polluted (south-west, west, north-west and north range between 10 and 50 µg m⁻²) with a peak average easterly value of approximately 65 µg m⁻². In terms of local contributions, east, north and south-westerly flows are dominated by local sources (i.e. >60%). Trajectories from the west and north-west are dominated by non-local sources (i.e. <40% local) but the absolute values of the integrated back trajectories are relatively smaller with limited impact on Delhi AQ. During the monsoon, all the wind directions (easterly, south-easterly, south-westerly, westerly and north-westerly) have lower integrated emission back-trajectories (integrated NOₓ emissions peaking at approximately 40 µg m⁻²) with over 80% of emissions coming from local sources. For poor air quality, as discussed above, the post-monsoon season is the most important as integrated emission trajectory values from the north (sample size = 17), north-west (179), north-east (10) and east (21) range between 60 and >100 µg m⁻² on average. Despite the larger integrated emissions trajectory median values, the key factor is the local contribution only ranging between 30 and 65%. The north-westerly trajectory value is the largest and most frequent (i.e. 73% of trajectories originate from the north-west) during the post-monsoon season with approximately 70% of emission contributions coming from outside of Delhi. Thus, we find that the dominating advection of highly polluted air originating from outside Delhi is the primary contributor to the Delhi population and air quality mitigation strategies need to be adopted not only in Delhi but also in the surrounding regions to successfully control this issue.

Next, we quantify the influence of individual sectoral emissions to the total accumulated emissions by running the back trajectory analysis without individual emissions source sectors and analysing accumulated emissions (Figure 7, note that only wind-directions with a sample size greater than or equal to 10 are shown). During the pre-monsoon season, accumulated emissions from transport are the dominant
contributor (>50%) and non-local sources are most important (60-80%) under the most frequent north-westerly and westerly wind directions. Trajectories travel close to the surface, within the boundary layer, in the final 24 hours and the NO\textsubscript{2} lifetime is short. This indicates that a large fraction of the total integrated emissions reaching Delhi are likely to have originated in the region surrounding the city, where the NO\textsubscript{2} burden from transport is high. Additionally, the contribution of non-local industrial and residential emissions is evident. This is likely because there are few large coal-fired power stations located to the north-west of Delhi in Punjab and Haryana. There is also a strong reliance on open stoves and fires for cooking and heating in nearby states such as Uttar Pradesh.

During the monsoon season, easterly (easterly and south-easterly) and westerly (south-westerly and north-westerly) winds dominate. Under all directions local accumulated emissions show nearly similar contribution (50-90%) for all sectors. Although many trajectories travel from distant regions, remaining close to the surface, there are few sources over the ocean (apart from shipping) and the NO\textsubscript{2} lifetime is short, so the advection of accumulated emissions concentration is much lower. Therefore, the small contribution of non-local sources is likely from nearby regions within the polluted IGP. Overall, transport emissions have the highest contribution to overall accumulated emissions (>40%), which is likely driven by transport emissions dominating the NO\textsubscript{2} burden in Delhi. In addition, contributions from local power and industrial emissions are also important under easterly and south-easterly (and north-westerly) wind directions with only a small non-local contribution to the overall emissions (10-20%) and are likely to originate from the large industrial region to the east of Delhi and from power generation to the north-west.

During the post-monsoon winds are primarily north-westerly and are associated with high contributions from transport emissions (70% non-local emissions), with non-negligible contributions from industry, other, power and residential emissions (and negligible contributions from fires). Again, non-local sources generally dominate for these sectors (up to 100%) with the largest probable contribution from within the IGP region surrounding Delhi where residential emissions are high. The large contribution of non-local emissions in all sectors is likely driven by the combination of several factors. First, a longer NO\textsubscript{2} lifetime, arising from OH being less abundant (to react with NO\textsubscript{2}) that enables accumulated emissions to be advected over long (and short) distances from high emission regions north-west of Delhi. Second, a shallow boundary layer that traps emissions close to the surface, allowing them to accumulate. Third, an increase in emissions during winter months due to increased demand for heating (residential sector) and power (power sector).

Finally, we apply the same back-trajectory emissions method to the high pollution days in Delhi to investigate the drivers of high-pollution events in the city. We use daily median ground-based NO\textsubscript{2} observations from sites in Delhi to identify high pollution days, defined as days where daily median NO\textsubscript{2} concentrations are above the 90\textsuperscript{th} percentile of daily median NO\textsubscript{2} concentrations (>37 \(\mu\)g m\textsuperscript{-3}) between 1\textsuperscript{st} January 2017 and 31\textsuperscript{st} December 2018 (Figure 8a). We then subsample the trajectories using these high pollution days and attribute the accumulated emissions to specific emissions sectors.

There is a clear seasonal cycle in NO\textsubscript{2} concentrations in Delhi, with maximum NO\textsubscript{2} values occurring during pre- and post-monsoon months that peak in December (up to ~60 \(\mu\)g m\textsuperscript{-3}), and minima during monsoon months (20 to ~30 \(\mu\)g m\textsuperscript{-3}) (Figure 8a). We also find that BLH is inversely related with median NO\textsubscript{2} concentrations, indicating a shallow boundary layer (<600 m) during the pre- and post-monsoon and a higher boundary layer (600-1500 m) during the monsoon (Figure 8b). Additionally, the atmospheric lifetime of NO\textsubscript{2} is highest in pre- and post-monsoon months (>10-70 hours) and is lowest during the monsoon season (~10 hours (Figure 8c). Finally, NO\textsubscript{2} emissions are also highest during the pre- and post-monsoon (~0.05-0.07 kg m\textsuperscript{-2} s\textsuperscript{-1}) and lowest during the monsoon season (<0.055 kg m\textsuperscript{-2} s\textsuperscript{-1} (Figure 8d). The combination of these factors aids the accumulation of increased local emissions during the pre- and post-monsoon months and the dispersal of the decreased emissions during the monsoon. Thus, the non-monsoon season, especially in the post-monsoon, experience a substantial degradation in air quality.
The back trajectory emissions analysis indicates that high-pollution days are associated with trajectories from the north-west of Delhi (Figure 9a) in both the pre- (n=12 days) and post-monsoon (n=60 days). Note that we only include wind-directions with a sample size greater than or equal to 5 (easterly and north-westerly). Trajectories gradually descend towards the surface between 90 and 20 hours out from Delhi, remaining close to the surface (>950 hPa) until they reach Delhi (Figure 9b). However, during the post-monsoon trajectories are much closer to the surface from hour 90 (910-810 hPa) compared with the pre-monsoon (840-770 hPa), likely allowing increased accumulation of emissions. During post-monsoon high-pollution days non-local (>80% non-local) transport emissions dominate (>75%) the total accumulated emissions (Figure 10). This is likely due to the trajectories remaining close to the surface for the final 24 hours, within a shallow boundary layer, at a period when the NOx atmospheric lifetime and emissions are increased. The contribution of other sectors to the remaining accumulated emissions (residential, industrial, other and power) is smaller but non-local emissions remain important (25-100%). There is also a negligible contribution (<2 µg m⁻³) from non-local (100%) fires under north-westerly winds. Overall, this suggests that non-local emissions from long-range transport (advection) are important during episodes of very poor AQ in Delhi.

4. Discussion and Conclusions

4.1 Seasonal wind flow regimes

In this study we have used high-resolution satellite tropospheric column NO2 (TCNO2) data from TROPOMI and bottom-up NOx emission inventories alongside a back-trajectory model to investigate the influence of local and the advection of pollutants to Delhi throughout the year. We also performed sensitivity simulations to separate the influence of various sectoral emissions on the accumulated pollution, by removing individual emission source sectors. We summarise our findings in Table 1, which gives the dominant wind-direction(s) in each season as a percentage of days in the season, the source sectors with the largest contribution to the accumulated emissions under that wind direction and finally, the contribution of local (Delhi) or non-local (rest of India) accumulated emissions to the total accumulated emissions. Our analysis indicates that emissions accumulated over 4 days are highest under easterly, northerly and north-westerly directions during the pre- (<60-90 µg m⁻³) and post- (<70-110 µg m⁻³) monsoon months. During the pre-monsoon season, north-westerly wind directions dominate (54%) and transport emissions account for 80% of the total accumulated emissions, with non-local emissions contributing ~70% of the total transport accumulated emissions. The high accumulated emissions observed under these wind directions are likely driven by high local NOx emissions during the pre-monsoon (since NOx lifetime is relatively short and the boundary layer is relatively deep). During the post-monsoon, north-westerly wind dominates (73%) and transport emissions (73%), which are non-local (70%), are most important. During this period high emissions are accompanied by a longer NOx atmospheric lifetime and shallow boundary-layer, aiding the build-up of emissions along the trajectory path and leading to increased contribution from the advection of NOx into the city under north-westerly flow. In addition, non-local (50-100%) residential, power and industrial emissions also contribute to the total accumulated emissions under north-westerly winds during the post-monsoon. Emissions are likely due to increased demand for heating and power during the colder post-monsoon months, particularly in Punjab and Haryana, where there are few large coal-fired power stations, and nearby states, such as Uttar Pradesh, where there is a strong reliance on open stoves and fires for cooking and heating.

4.2 Source attribution during high pollution episodes

We also applied the back-trajectory method to high-pollution days, defined as days where median NO2 concentrations (from ground-based observations) are above the 90th percentile. The ground-based observations indicate that high-pollution episodes are most common in the post-monsoon (>80% of occurrences) and are dominated by days where winds were from the north-west (>75%). On these days (north-westerly winds during the post-monsoon) non-local (>80%) transport emissions dominate (>75%). The large contribution of non-local transport emissions is likely driven by trajectories (air masses) that
travel towards Delhi remaining close to the surface, accumulating increased emissions that are not quickly lost due to decreased sunlight and therefore less abundant OH during winter months. The small contribution (20%) of local transport emissions is likely driven by high NOx emissions in a shallow boundary layer, which acts to trap pollutants at the surface.

The results of this study are in line with previous work by Sembhi et al. (2020) who used a model to simulate air quality in Delhi during a poor AQ episode in 2016 with and without the implementation of the SSWA. They found that timing shift in agricultural burning in north-west India caused by the introduction of the SSWA contributed only around 3% to the poor AQ observed, indicating that this was largely driven by other factors. We also find that trajectories originating from the north-west during post-monsoon months have a polluted footprint in both the satellite and emissions analysis. However, within our work we do not see an impact from the advection of NOx fire emissions, which could be for a number of reasons. Firstly, Sembhi et al (2020) focussed on PM2.5, which has a much longer atmospheric-lifetime than NOx (days to weeks compared with hours to days). This leads to a smaller contribution in the advection of NOx from fires occurring in north-west India during the post-monsoon. In addition, fire emissions are generated using polar orbiting satellites which have a single daytime overpass and thus may miss fires which have a short burn time; fire emissions inventories currently struggle to detect agricultural waste burning fires due to their small size and often short burn times (Zhang et al., 2020; Liu et al., 2020). Although we have used VIIRS in this study (which is able to detect smaller, lower temperature fires than MODIS) the total emissions from agricultural waste burning may still be underestimated (Zhang et al., 2020; Liu et al., 2020). With the introduction of geostationary satellites and sensors which can continuously detect smaller fires (e.g. Himawari) it should be easier to constrain the emissions from agricultural waste burning in the future. However, this work provides further evidence that the advection of non-local pollutants to Delhi plays an important role in both ambient and high pollution conditions, in line with Sembhi et al. (2020).

4.3 Implications for policy to control air pollution over Delhi

Transport emissions dominate under all wind-directions and seasons indicating that emissions reductions in this sector would lead to the largest benefits. However, to improve local AQ in Delhi transport emissions would need to be reduced both locally and regionally. This is particularly important for high pollution events where non-local transport emissions dominate.

Code Availability

Code used to generate the figures in this paper is available on request.

Data Availability

Data used in this paper is available on request.

Author Contributions

AMG processed emissions datasets, adapted and ran back-trajectory code, previously written by RJP, and created all figures. RJP downloaded and processed satellite data. SD downloaded and processed ERA-Interim data. AMG wrote the manuscript with input from RJP, SD and MPC. MP provided TOMCAT 3D OH fields from model simulations which WF helped set-up. VS provided ground-based NO2 observations from Delhi. YC and OW helped with NO2 lifetime calculations. RS provided guidance and feedback on the work. GB provided SAFAR NOx emissions.

Competing Interests

The authors declare that they have no conflict of interest.
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DOI: https://doi.org/10.5194/acp-20-10687-2020.
Figures:

Figure 1. (a) NO\textsubscript{x} emissions (kg m\textsuperscript{-2} s\textsuperscript{-1}) within Delhi from the System of Air Quality and Weather Forecasting And Research (SAFAR) (Beig, 2018) for 2018 regridded from 400 m to 10 km resolution. (b) NO\textsubscript{x} emissions within Delhi from the Emission Database for Global Atmospheric Research with Task Force on Hemispheric Transport of Air Pollution version 2.2 (EDGAR-HTAP2) (Janssens-Maenhout et al., 2015) emissions for 2010 at 10 km resolution. (c) Global NO\textsubscript{x} emissions from EDGAR-HTAP2 for 2010 at 10 km resolution. (d) Merged EDGAR-HTAP2 and SAFAR NO\textsubscript{x} emissions, where EDGAR-HTAP2 NO\textsubscript{x} emissions within Delhi have been replaced with NO\textsubscript{x} emissions from SAFAR. (e) Global fire emissions provided by the FINNv2.5 dataset for 2018 at 10 km resolution. (f) Merged anthropogenic and fire NO\textsubscript{x} emissions dataset, where SAFAR, EDGAR-HTAP2 and FINNv2.5 emissions have been combined.
Figure 2. Back-trajectory method used in this study. Gridded emissions from a bottom-up inventory (Figure 1e) are summed every 15-minutes (15 mins) along the back-trajectory path (arrows) when the air parcel is within the boundary layer (indicated by boxes). NO\textsubscript{x} accumulates along the trajectory path towards Delhi (indicated by line shading). Satellite tropospheric column NO\textsubscript{2} (TCNO\textsubscript{2}) is also summed in a similar way.

Figure 3. Map showing the location of NO\textsubscript{2} monitoring sites over Delhi. In this study we use the daily 07:30-08:30 UTC (13:00-14:00 local time) median NO\textsubscript{2} concentration (calculated using all sites shown) to determine the timing of high pollution days in Delhi. We define high pollution days as days where the NO\textsubscript{2} concentration exceeds the 90\textsuperscript{th} percentile of NO\textsubscript{2} concentrations between 2018 and 2019.
Figure 4. (a) Total annual (and seasonal) accumulated TROPOMI TCNO$_2$ (µg m$^{-2}$) arriving in Delhi along 4-day back trajectories. Trajectories are coloured by their total accumulated TCNO$_2$, with darker trajectories indicating higher levels of accumulated TCNO$_2$. (b) Mean annual (and seasonal) TCNO$_2$ anomaly (µg m$^{-2}$), (c) mean annual (and seasonal) NO$_2$ lifetime (anomaly) (hours) and (d) mean annual (and seasonal) boundary layer height (BLH) (anomaly) (hPa) compared to all seasons. All panels show the time period of 2017 and 2018 annually and seasonally - during the pre-monsoon (Feb_Mar_Apr), monsoon (May_Jun_Jul_Aug_Sept) and post-monsoon (Oct_Nov_Dec_Jan) seasons, respectively.
Figure 5. (a) Total accumulated NO\textsubscript{x} emissions (µg m\textsuperscript{-2}) arriving in Delhi in 2017 and 2018 and (b) pressure level of air parcels as they converge on Delhi, just above the surface (~1000 hPa) at hour 0 in pre-monsoon (Feb_Mar_Apr), monsoon (May_Jun_Jul_Aug_Sep) and post-monsoon (Oct_Nov_Dec_Jan) seasons. Median (black/white line), 25\textsuperscript{th} and 75\textsuperscript{th} percentiles (black/white dashed lines) of the trajectory pressure are also indicated. Trajectories are coloured by their total accumulated emissions, with darker trajectories indicating higher levels of accumulated NO\textsubscript{x}.

Figure 6. Wind rose of median accumulated NO\textsubscript{x} emissions (µg m\textsuperscript{-2}) from 4-day back trajectories with 6-hr time steps arriving at Delhi in 2017 and 2018 for pre-monsoon (Feb_Mar_Apr), monsoon (May_Jun_Jul_Aug_Sep) and post-monsoon (Oct_Nov_Dec_Jan) seasons. Total accumulated emissions are indicated by the shading of the segments. The area of the segment indicates the non-local contribution to the total integrated emissions. The percentage of local emissions is indicated on the circles. The number of days on which each wind direction occurs in each season is also indicated in brackets. For example, NW in the post-monsoon is very polluted (~100 µg m\textsuperscript{-2}), 70% of total integrated emissions are non-local and this wind direction occurs on 179 days.
Figure 7. Sector-specific accumulated NOx emissions (µg m\(^{-2}\)) from 4-day back trajectories with 6-hr time steps arriving in Delhi in 2017 and 2018 in pre-monsoon (pre) (Feb_Mar_Apr), monsoon (mon) (May_Jun_Jul_Aug_Sept) and post-monsoon (post) (Oct_Nov_Dec_Jan). Light shading indicates accumulated NOx emissions originating within Delhi and darker shading indicates accumulated NOx emissions originating from outside of Delhi. Sectors included are fire (FIRES) (red), industrial (IND) (green), other (OTHER) (black), power generation (POW) (gold), residential (RES) (magenta) and transportation (TRANS) (cyan) emissions. The number of days for which each wind direction occurs in each season is indicated in brackets. For example, north-westerly winds occur on 179 days during the post-monsoon and transport emissions dominate (~80 µg m\(^{-2}\)). The contribution from transport is split into ~55 µg m\(^{-2}\) that are non-local (India) and ~25 µg m\(^{-2}\) that are local (Delhi).

Figure 8. (a) Daily median NO\(_2\) concentrations (µg m\(^{-3}\)) in Delhi (blue). Concentrations above the 90th percentile of observations are indicated in red. (b) Dually mean boundary layer height (BLH) (m) in Delhi from ERA5 re-analysis. (c) Daily mean NO\(_x\) lifetime (hours) in Delhi and (d) Monthly mean NO\(_x\) emissions (kg m\(^{-2}\) s\(^{-1}\)) from the SAFAR emissions inventory. All panels show the time-period between 1\(^{st}\) January 2017 and 31\(^{st}\) December 2018.
Figure 9. (a) Total accumulated NO\textsubscript{x} emissions (µg m\textsuperscript{-2}) arriving at Delhi during high pollution events in 2017 and 2018, (b) pressure level of air parcels as they converge on Delhi just above the surface (~1000 hPa) at hour 0 in pre-monsoon (Feb_Mar_Apr), monsoon (May_Jun_Jul_Aug_Sept) and post-monsoon (Oct_Nov_Dec_Jan). Median (black line), 25\textsuperscript{th} and 75\textsuperscript{th} percentiles (black dashed lines) of the trajectory pressure are also indicated. Trajectories are coloured by their total accumulated emissions, with darker trajectories indicating higher levels of accumulated NO\textsubscript{x}.

Figure 10. Sector-specific accumulated NO\textsubscript{x} emissions (µg m\textsuperscript{-2}) from 4-day back trajectories with 6-hr time steps arriving at Delhi during high pollution events in 2017 and 2018 in the pre-monsoon (pre) (Feb_Mar_Apr), monsoon (mon) (May_Jun_Jul_Aug_Sept) and post-monsoon (post) (Oct_Nov_Dec_Jan). Light shading indicates accumulated NO\textsubscript{x} emissions originating within Delhi and darker shading accumulated NO\textsubscript{x} emissions from outside of Delhi. Sectors included are fire (FIRES) (red), industrial (IND) (green), other (OTHER) (black), power generation (POW) (gold), residential (RES) (magenta) and transportation (TRANS) (cyan) emissions. The number of days for which each wind direction occurs in each season is indicated in brackets. For example, north-westerly winds occur on 45 days during high pollution days in the post-monsoon and transport emissions dominate (>100 µg m\textsuperscript{-2}). The contribution from transport is split into ~80 µg m\textsuperscript{-2} that are non-local (trans India) and ~25 µg m\textsuperscript{-2} that are local (trans Delhi).
<table>
<thead>
<tr>
<th>Season</th>
<th>Dominant wind direction (% of days)</th>
<th>Key Source Sectors (% contribution)</th>
<th>Local/non-local contribution to accumulated emissions for Key Source Sector (% contribution)</th>
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<td>Transport (80%)</td>
<td>Local (32%)/Non-local (68%)</td>
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<td>Post-monsoon</td>
<td>NW (73%)</td>
<td>Transport (73%)</td>
<td>Local (30%)/Non-Local (70%)</td>
</tr>
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</table>

Table 1s. Summary of key wind-directions in the pre-monsoon (Feb-Apr), monsoon (May-Sept) and post-monsoon (Oct-Jan) monsoon (as % of total days in each season). The key source sectors with the largest contribution to the overall total accumulated emissions are also indicated (as % contribution) alongside the contribution of local (Delhi) and non-local (rest of India) accumulated emissions to overall accumulated emissions for that sector. For example, in the post-monsoon NW winds occur 73% of days, transport emissions contribute 72% of the overall integrated emissions and 30% of transport emissions are local, while 70% are non-local. Seasonal sector-specific accumulated NOx emissions (µg m⁻²) are calculated from 4-day back trajectories with 6-hr time steps arriving in Delhi in 2017 and 2018 in pre-monsoon, monsoon and post-monsoon periods.