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17-year trend of tropospheric columnar NO_2 over India's northeast region observed by OMI: investigating probable anthropogenic and natural sources

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सार — यह अध्ययन भारत के पूर्वोत्तर क्षेत्र (NERI) में 17 वर्षों (2005-2022) में फैले क्षोभमंडलीय स्तंभाकार NO2 (TCN) सांद्रता का व्यापक विश्लेषण प्रस्तुत करता है। औरा उपग्रह पर लगे ओजोन मॉनिटरिंग इंस्ट्रूमेंट (OMI) से सुदूर संवेदन डेटा का उपयोग क्षेत्र में नाइट्रोजन डाइऑक्साइड (NO2) सांद्रता के स्थानिक-कालिक पैटर्न का विश्लेषण करने में किया गया। NO2 एक प्रमुख वायुमंडलीय प्रदूषक है जो औद्योगिक उत्सर्जन, वाहन दहन, जैवभार दहन और प्राकृतिक प्रक्रियाओं जैसे तड़ित और मृदा उत्सर्जन जैसे विविध स्रोतों से निकलता है। NERI में NO2 प्रदूषण के विभिन्न स्तरों के लिए इसकी विशिष्ट स्थलाकृति और मौसम संबंधी कारकों के साथ-साथ शहरीकरण, जनसंख्या वृद्धि और ऊर्जा उपयोग के लिए जिम्मेदार ठहराया जा सकता है। मॉनसून पूर्व और शीत ऋतु के दौरान TCN सांद्रता बायोमास दहन और मानवजनित गतिविधियों जैसे कारकों के कारण चरम पर होती है। दीर्घकालिक आंकड़ों से पता चलता है कि समग्र TCN में वृद्धि हुई है, जो बढ़ते वाहनों, औद्योगिक विस्तार और जनसंख्या घनत्व के बढ़ते प्रभावों को दर्शाती है। मासिक परिवर्तन मॉनसून पूर्व ऋतु के महत्व को इंगित करते हैं, जिसमें तड़ित और परिवहनित NO2 के कारण NO2 का स्तर बढ़ जाता है। वनों की आग, बायोमास जलाना और दहन इंजन प्राकृतिक और मानवजनित NO2 के प्रमुख स्रोत आवृत्ति वितरण विश्लेषण के परिणाम एनईआरआई राज्यों में वायु गुणवत्ता की अलग-अलग स्थिति दर्शाते हैं, तथा लगातार उच्च टीसीएन स्तर का अनुभव करने वाले क्षेत्रों में लक्षित हस्तक्षेप की आवश्यकता पर बल देते हैं। इसके अलावा इस अध्ययन में कोविड-19 महामारी के प्रभाव का आकलन किया गया तथा मॉनसून पूर्व ऋतु में लॉकडाउन के दौरान नाइट्रोजन डाइऑक्साइड (NO2) की सांद्रता में उतार-चढ़ाव की देखी गई। यह शोध NERI में बढ़ते नाइट्रोजन डाइऑक्साइड प्रदूषण से निपटने के लिए निगरानी और शमन रणनीतियों की आवश्यकता पर जोर देता है, वायु गुणवत्ता और व्यापक पर्यावरणीय स्वास्थ्य मुद्दों को संबोधित करता है तथा स्वस्थ जीवन परिस्थितियों के लिए सुविचारित उपायों की आवश्यकता बताता है।

ABSTRACT. This study presents a comprehensive analysis of Tropospheric Columnar NO_2 (TCN) concentrations spanning 17 years (2005-2022) in the Northeastern Region of India (NERI). Remote sensing data from the Ozone Monitoring Instrument (OMI) aboard the Aura satellite was utilized in analyzing the spatiotemporal patterns of nitrogen dioxide (NO_2) concentrations within the region. NO_2 is a prominent atmospheric pollutant that emerges from diverse sources like industrial emissions, vehicle combustion, biomass burning, and natural processes such as lightning and soil emissions. The varying levels of NO_2 pollution in the NERI, with its distinctive topography and meteorological behaviors, may be attributed to urbanization, population growth, and energy utilization. TCN concentrations peak during pre-monsoon and winter months, driven primarily by factors like biomass burning and anthropogenic activities. Long-term data reveals an overall TCN increase, reflecting growing influences from rising vehicles, industrial expansion, and population density. Monthly variations indicate the significance of the pre-monsoon season, characterized by elevated NO_2 levels influenced by lightning and transported NO_2. Forest fires, biomass burning, and combustion engines contribute as major sources of both natural and anthropogenic NO_2. Frequency distribution analysis results exhibit varying air quality statuses across NERI states, emphasizing the need for targeted interventions in regions consistently experiencing high TCN levels. Furthermore, the study assesses the impact of the COVID-19 pandemic, identifying fluctuations in NO_2 concentrations during lockdowns in pre-monsoon seasons. This research emphasizes the requirement for strong monitoring and mitigation strategies to combat increasing NO_2 pollution in the NERI, addressing air quality and broader environmental health issues, necessitating well-informed measures for healthier living conditions.

Key words– Nitrogen dioxide, North Eastern Indian Region, Anthropogenic source, Lightning.

1. Introduction

Nitrogen dioxide (NO_2) significantly influences the Earth's atmospheric radiative balance, altering oxidation capacity and chemistry and impacting the lifetime of greenhouse gases. It primarily emanates from industrial processes, vehicle combustion, biomass fuel and crop residue burning, and soil emissions (Richter & Burrows, 2002; Cheng *et al.*, 2012). Natural sources include lightning and soil emissions. NO_2 contributes to degraded air quality, adversely affecting human health (Molina and Molina, 2004). It is a key pollutant with regulated national ambient air quality standards worldwide. High tropospheric NO_2 concentrations pose a threat due to spatiotemporal variability influenced by local emissions, seasons, land use land cover (LULC) changes, and meteorological conditions. Satellite remote sensing is a viable approach. Studies in coal mining areas indicated positive correlations between thermal power plant output and NO_2 concentrations (Prasad *et al.*, 2012). Thunderstorm lightning has been considered a major source of nitrogen oxides (NOx, *i.e*., NO (nitric oxide) and NO_2) since von Liebig (1827) proposed it as a natural mechanism for the fixation of atmospheric nitrogen (Hutchinson, 1954). Lightning-induced nitrogen oxides (LNOx) have several important implications for atmospheric chemistry and climate (WMO, 1999; IPCC, 2001). Tropospheric (or ground-level) ozone also plays a role in the production of NO_2 through photochemical processes (Bradshaw *et al.*, 1999) The global LNOx source is one of the largest natural sources of NOx in the atmosphere (Galloway *et al.*, 2004) and certainly the largest source of NOx in the upper troposphere, in particular, in the tropics (WMO, 1999)

Noteworthy, NO_2 is also an important contributor to air pollution as a primary pollutant and as a precursor to ozone and fine particulate matter production. Human exposure to elevated NO_2 concentrations is associated with a range of adverse outcomes such as respiratory infections (Pannullo *et al.*, 2017; Tao *et al.*, 2014; Zeng *et al.*, 2020), increases in asthma incidence (Anenberg *et al.*, 2018; Achakulwisut *et al.*, 2019), lung cancer (Hamra *et al.*, 2015) and overall mortality (Brook *et al.*, 2007; Crouse *et al.*, 2015). NO_2 observations indicate air quality relationships with combustion sources of pollution such as transportation (Achakulwisut *et al.*, 2019). Initial investigations found substantial decreases in the atmospheric NO_2 column from satellite observations (Goldberg *et al.*, 2020; Biswal *et al.*, 2021; Koukouli *et al.*, 2021; Field *et al.*, 2020; Bauwens *et al.*, 2020; Liu *et al.*, 2020; Prunet *et al.*, 2020) and in ambient NO_2 concentrations from ground-based monitoring (Shi *et al.*, 2021; Ropkins *et al.*, 2021; Fu *et al.*, 2020; Venter *et al.*, 2020) during lockdowns enacted to reduce the spread of COVID-19. However, questions remain about the relationship of atmospheric columns with health- and policy-relevant ambient ground-level concentrations, and about the representativeness of sparse ground-based monitoring for broad assessment. Thus, there is a need to relate satellite observations of NO_2 columns to ground-level concentrations. It is also important to consider the effect of meteorology on recent NO_2 changes (Shi *et al.*, 2021) and to quantify NO_2 changes due to COVID-19 interventions in the context of longer-term trends (Liu *et al.*, 2021). Furthermore, air quality monitoring sites tend to be preferentially located in higher-income regions, raising questions about how NO_2 changed in lower-income regions where larger numbers of potentially susceptible people reside. Estimates of changes in ground-level NO_2 concentrations derived from satellite remote sensing would fill gaps between ground-based monitors, offer valuable information in regions with sparse monitoring, and more clearly connect satellite observations with ground-level ambient air quality.

North Eastern Region (NER) of India, Bangladesh, Bhutan, Tibetan Plateau, and Indo-Gangetic Plane (IGP) have a complex topography. These areas are affected by dry, cold westerly converge with hot moist southerly and help to develop thunderstorms associated with lightning (Kumar and Kamra 2012; Qie *et al.*, 2022). Lightning flashes are significantly correlated with convective rain, total column water vapor (TCWV), or surface relative humidity over both land and sea regions, according to previous studies (Price and Federmesser, 2006; Siingh *et al.*, 2011; Shi *et al.*, 2018). In this study, we utilized the tropospheric NO_2 and the lighting count over the North Eastern Region of India (NERI) to investigate the evolution and concentration trends across different states

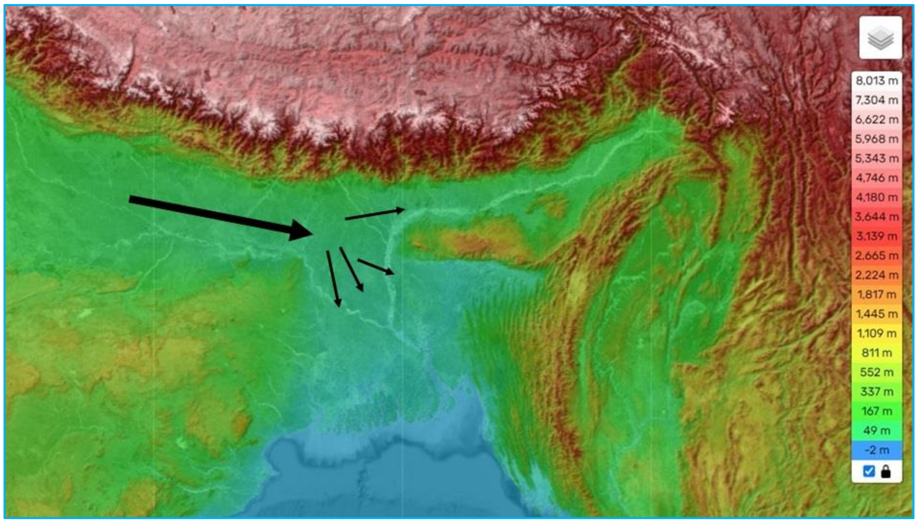

Fig. 1. Topographical map of NERI adopted from topographic-map.com, the black lines represent the transportation of pollutants and the arrowhead indicates the direction

of the region. Specifically, we aim to quantify the increasing/decreasing trends of troposphere column NO_2 and examine seasonal variations. Besides, the lightning count is also considered to connect with the NO_2 for its contribution. The NERI faces increasing NO_2 production due to population growth, urbanization, industrialization, agriculture demands, and energy consumption. Additionally, changes in LULC, such as deforestation and agricultural expansion, play a significant role in altering NO_2 levels by influencing local emissions and atmospheric dynamics. Assessing NO_2's spatiotemporal distribution and identifying emission sources are crucial to formulating effective reduction strategies.

2. Data, Methodology and Study Area

2.1. *Study domain and synoptic meteorology*

The study area encompasses latitude 20° N - 30° N and longitude 88° E - 98° E, with a primary focus on the NERI. The NERI comprises of eight states: Assam (AS), Arunachal Pradesh (AR), Manipur (MN), Meghalaya (ML), Mizoram (MZ), Nagaland (NG), Tripura (TR) and Sikkim (SK) (Fig. 1). Notably, approximately 23.75% of India's forest area is found in the NERI (FSI, 2021). The region experiences a sub-tropical climate due to its distinctive topography. The Himalayan Mountains lie to the north, the Indo-Myanmar ranges of foothills and hills are in the east, and Mizoram and Tripura are to the south (Fig. 1). During the winter (Dec-Feb) and pre-monsoon (Mar-April) seasons, westerly winds significantly influence the atmospheric processes in the NER, as the region opens westward towards the IGP. The southwest monsoon serves as the primary source of rainfall in the area, and Cherrapunjee receives the highest rainfall globally, owing to orographic lifting by the Meghalaya plateau. The mean temperatures vary from 15 °C (January) to 28 °C (August) in the plain areas and from 9 °C to 21 °C in the hilly regions during the same periods. Relative humidity averages between 30-50% in the pre-monsoon season and 80-95% during the monsoon. To understand the wind patterns, we analyzed the averaged wind vector at 850 hPa from the European Centre for Medium-Range Weather Forecasts (ECMWF) Reanalysis-version 5 (ERA-5) over 15 years (2006-2020) from January to April, covering the area between 20° N - 30° N and 84° E-98° E. The analysis revealed dominant westerly winds during these months, with wind speeds ranging from 2 ms^{-1} to 3 ms^{-1} in January and February and 4 ms^{-1} to 5 ms^{-1} in March and April. Notably, during January and February, the westerly wind shifts to southwesterly over the Brahmaputra valley and further converts to southeasterly over the eastern part of the NER. Additionally, it converts to southwesterly/southeasterly over Nagaland, parts of Manipur, and other eastern borders.

2.2. *Data Sources*

Tropospheric NO_2 concentrations were obtained from the Ozone Monitoring Instrument (OMI) on the Aura satellite (Schoeberl *et al.*, 2004). Daily daytime global NO_2 datasets were generated by OMI aboard the polar-orbiting, sun-synchronous Earth Observing System Aura satellite, with an equator crossing time of 13:45 LT. OMI measures the Earth's backscattered radiation within the spectral range of 264-504 nm (Streets *et al.*, 2013). To derive tropospheric column concentrations of NO_2, the

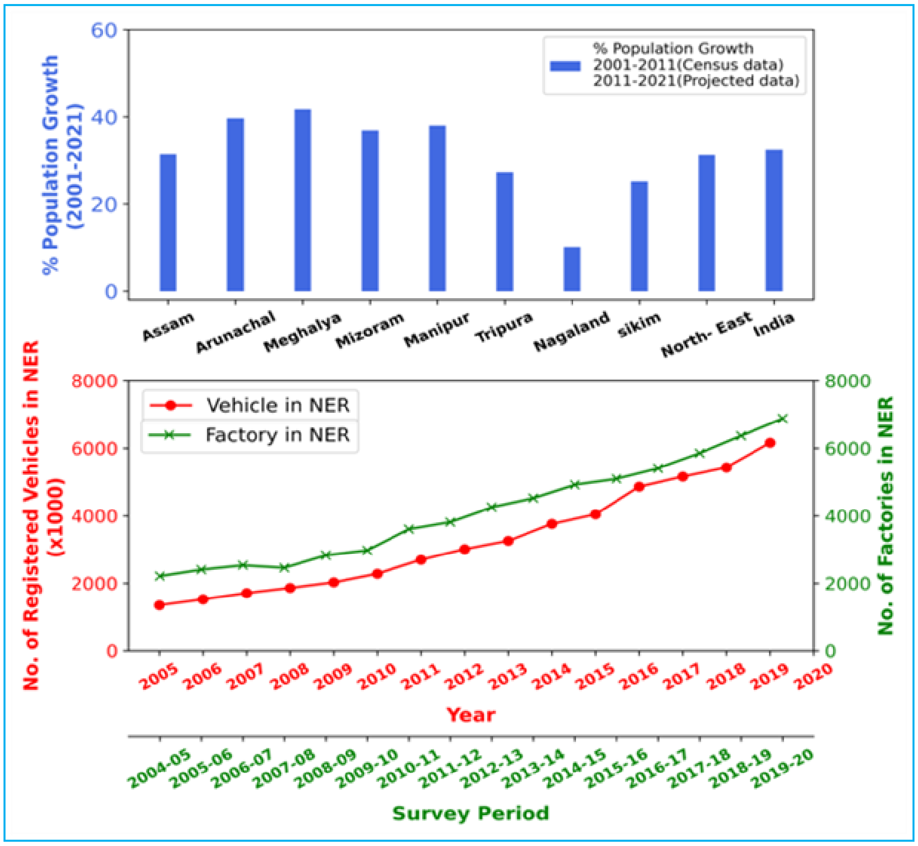

Fig. 2. Population, factories and vehicle growth over NERI

attenuation measurements provide slant column densities, converted to vertical column densities using appropriate air mass factors. Retrieval errors for NO_2 in high-emission areas are up to 30% (Streets *et al.*, 2013). The retrieval process considers clear and cloudy conditions, incorporating air mass factors derived from simulated NO_2 profiles (Bucsela*et al.*, 2006). Tropospheric NO_2 has an uncertainty of 0.1×10^{15} mol.cm^{-2} and is underestimated by 15-30% (Celarier*et al.*, 2008). Land Use/Land Cover (LULC) data were obtained from ISRO's Bhuvan geoportal (https://bhuvan.nrsc.gov.in) collected using multi-temporal satellite imagery from the Resourcesat-1 LISS III sensor. Tropospheric ozone (O_3) concentrations were acquired from AURA OMI/MLS, computed by subtracting MLS-derived stratospheric ozone from OMI total column ozone (Ziemke *et al.*, 2006). NO_2 data was utilized which had been monitored as part of the National Air Quality Monitoring Programme (NAMP), which measures air pollutants including SO_2, NO_2 and PM_{10} at 931 stations across 398 cities/towns in India. The monitoring is being carried out by Central Pollution Control Board (CPCB) in collaboration with other state and national agencies (https://cpcb.nic.in). Fire count datasets were utilized from a recent study (Borgohain *et al.*, 2023) of the impact of biomass burning over the region.

In this study, the Mann-Kendall test and Sens Slope are utilized for the trend analysis. The details about the technique are elaborated elsewhere (Theil 1950; Sen 1968; Gilbert 1987; Sirois 1998; Wang and Swail 2001; Yue *et al.* 2002; Wang *et al.* 2015).

2.3. *Anthropogenic forcing in NERI*

Based on data published by the Ministry of Statistics and Project Implementation, Government of India (www.mospi.gov.in) and projected population data from the Ministry of Health and Family Welfare, Government of India (GOI; https://nhm.gov.in), along with industrial data from the Annual Survey of Industries (http://microdata.gov.in) and vehicle data from the Ministry of Road Transport and Highways (https://morth.nic.in), Fig. 2 illustrates population growth, year-wise vehicle registrations, and factories over NERI from 2005 to 2019. Population data is available up to 2011 (As per GOI standard, there is a census every ten years, but since the 2021 census is still incomplete, projected

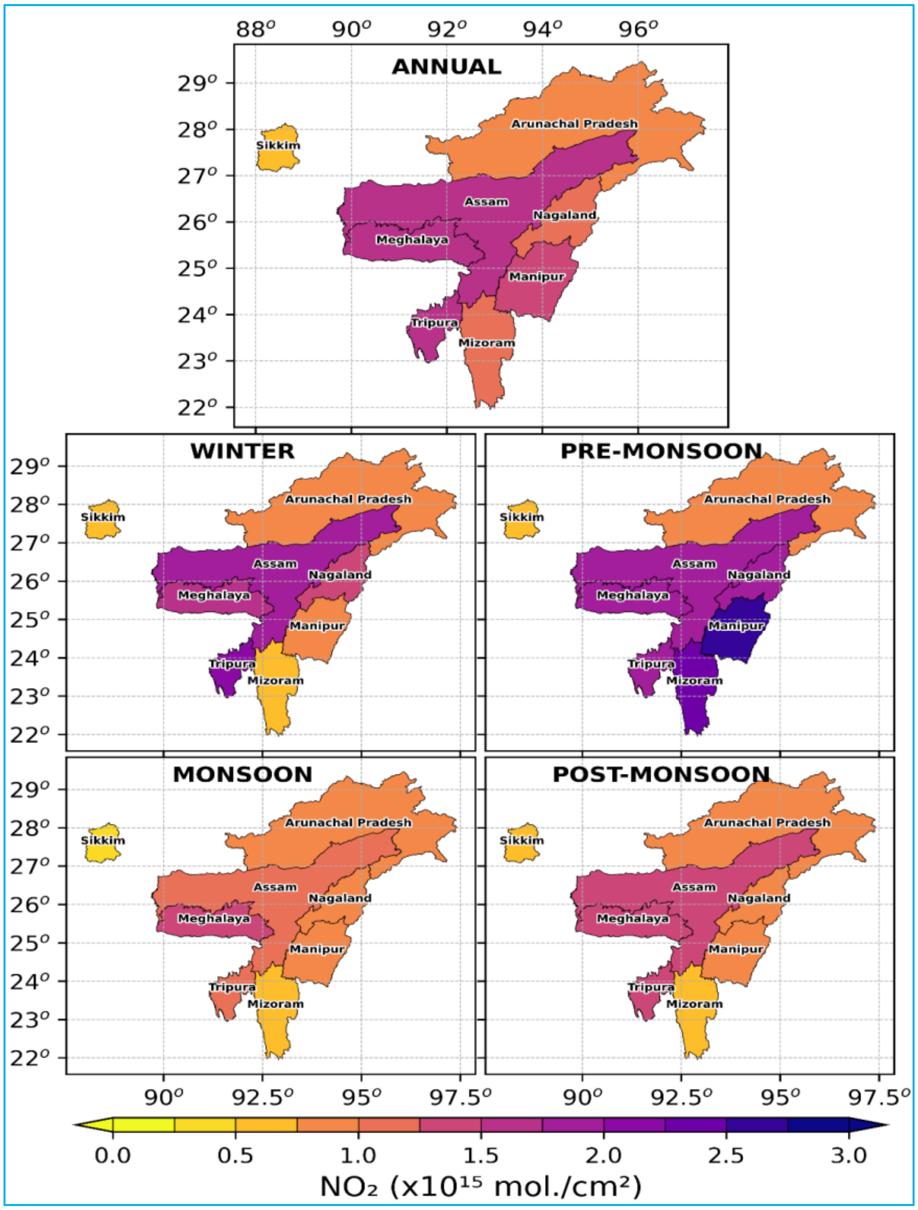

Fig. 3. Spatial distribution of mean TCN over the NERI (2005-2022)

data was only published). According to the combined census and projected data, Meghalaya experienced the largest population growth (~41%) in NERI, while Nagaland had the lowest (~10%). Overall population growth in NERI is around 31%, slightly below the national rate of about 32%. The number of vehicles and factories in NERI significantly increased, with data available up to 2019 and 2020, respectively. Vehicle numbers rose from 1.3 million to 6 million between 2005 and 2019, at 0.33 million per year. Factories increased from roughly 2200 to 6800 from 2005 to 2020, at 306 factories per year across NERI.

3. Results and discussion

3.1. *Intra-annual variation over NERI*

This study utilizes Tropospheric Columnar NO_2 (TCN) concentration data obtained from the Ozone Monitoring Instrument (OMI) spanning 17 years, from 2005 to 2022. Fig. 3 illustrates the temporal and spatial mean seasonal TCN over the eight states of NERI. The four seasons considered are pre-monsoon (March, April, May), monsoon (June, July, August, September), post-monsoon (October, November), and winter (December, January, February). The annual mean TCN exhibits the highest concentrations in Assam (1.57×10^{15} mol cm^{-2}), Meghalaya (1.60×10^{15}mol cm^{-2}), and Tripura (1.62×10^{15}mol cm^{-2}), while Sikkim records the lowest (5.28×10^{14} mol cm^{-2}). TCN levels are generally elevated during pre-monsoon and winter across all states, followed by post-monsoon and monsoon seasons. In the pre-monsoon period, Manipur shows the highest TCN (2.58×10^{15}mol cm^{-2}), followed by Mizoram and other states, while Arunachal Pradesh and Sikkim exhibit the lowest TCN (8.74×10^{14} mol cm^{-2} and 5.27×10^{14} mol cm^{-2}), respectively. This observed pattern of high TCN during the pre-monsoon months is attributed to significant biomass burning in these states (Borgohain *et al.*, 2023).

The spatial and temporal distribution of TCN depends on meteorological factors, such as rainfall, wind direction, and Atmospheric Boundary Layer (ABL) height. TCN concentrations peak during the pre-monsoon months, followed by winter in all states, with significant contributions from biomass burning. The northeast part of India experiences elevated TCN levels during the pre-monsoon season due to extensive biomass burning emissions. Manipur (2.58×10^{15}mol cm^{-2}) and Mizoram (2.37×10^{15}mol cm^{-2}) exhibit the highest TCN, while Sikkim and Arunachal Pradesh consistently record the lowest in all seasons. The pre-monsoon pattern of maximum NO_2 concentrations can be attributed to low humidity and mild temperatures, reducing the photolysis removal process of NO_2, thus stabilizing its concentration. Lower concentrations of OH (due to low humidity) also limit NO_2 photolysis and the formation of HNO_3, a principal sink for NO_2 (Jacob, 1999). In winter, high TCN is due to extensive biomass fuel usage for home heating and reduced availability of UV radiation to initiate photolysis reactions that break down NO_2 (Richter *et al.*, 2005; Tariq *et al.*, 2014; Uno *et al.*, 2007; Nishanth and Kumar, 2011).

Additionally, a shallower ABL in winter results in lower vertical dispersion, reducing dilution and removal rates of NO_2, contributes to its enhancement (Barnes *et al.*, 2013). Elevated NO_2 concentrations during winter may also result from NO_2 transportation from the IGP to NERI due to westerly winds. Haridas *et al.* (2019) reported that the IGP shows the highest levels of NO_2 concentrations during winter.

Low TCN concentrations during monsoon result from wet removal processes. The presence of a moist clean air mass, increased actinic fluxes enhancing NO_2 photo dissociation, elevated OH radical levels aiding NO_2 removal via HNO_3, and potentially reduced traffic due to decreased social and educational activities during the hot summer contribute to the decrease in TCN (Ghude *et al.*, 2009; Tariq *et al.*, 2014; Ravindra *et al.*, 2003; Yoo *et al.*, 2014). During the rainy season, the impact of lightning on increasing NO_2 concentrations is not observed due to the opposing influence of rain washout (Yoo *et al.*, 2014).

3.2. *Long-term TCN over NERI*

Fig.4 illustrates the annual variations of TCN from 2005-2022 for four seasons as observed by the satellite over the eight northeastern states of India. Across all seasons, Assam consistently displayed the highest TCN value (1.57×10^{15}mol cm^{-2}), along with Meghalaya, Tripura, and Nagaland. On the other hand, Sikkim and Arunachal Pradesh recorded the lowest TCN values (5.28×10^{14}mol cm^{-2}). Notably, all states experienced an increase in TCN over the seventeen years (2005 to 2022) (Fig. 7). This long-term TCN increase is influenced by anthropogenic factors, such as rising vehicle numbers, industries, and population density.

Mann Kendall test and Sen Slope have been utilized for the TCN trend analysis and rate of change of TCN over the states. Arunachal Pradesh, Manipur, Nagaland, and Meghalaya show no trend in any season. Positive trends are detected in Assam during pre-monsoon and monsoon with Sen Slopes of $2.57\times10^{13}$and 1.65×10^{13}, respectively. Pre-monsoon TCN increase is higher than during monsoon. Assam's valley region, developed cities, and national highways contribute to continuous anthropogenic emissions. Pre-monsoon TCN includes local and transported NO_2, while monsoon is dominated by locally emitted NO_2 due to the absence of transported NO_2. Meghalaya exhibits positive trends in winter and monsoon with Sen Slopes of 1.33×10^{13} and 1.66×10^{13}, respectively. Mizoram shows positive trends in monsoon and post-monsoon with Sen's Slopes of $1.23\times10^{13}$and 9.96×10^{12}. Tripura displays positive trends in winter, monsoon and post-monsoon with Sen's Slopes of 1.99×10^{13}, 1.76×10^{13} and 3.38×10^{13}, respectively. The post-monsoon TCN increase is higher than in winter and monsoon. In winter and pre-monsoon, an aerosol river from North-west India reaches the NER corridor and bifurcates into two streams: one flows along the Brahmaputra valley (Assam), and the other flows along West Bengal and Bangladesh, finally reaching Tripura and Mizoram (south-east region). Kundu *et al.* (2018) reported that the Brahmaputra River valley in NERI is significantly affected by the prolonged dispersion of pollutants from the IGP during local winter and is burdened with a lot of natural and anthropogenic aerosol. Short-term trends for 2005-2008 showed increased NO_2 levels in the IGP, with some cities exhibiting positive NO_2-O_3 correlations (Streets *et al.*, 2013). The IGP experienced an increasing NO_2 trend of 3×10^{13} mol/cm^2/yr during 2005-2014 (Haridas *et al.*, 2019). Ul - Haq *et al.* (2015) reported an

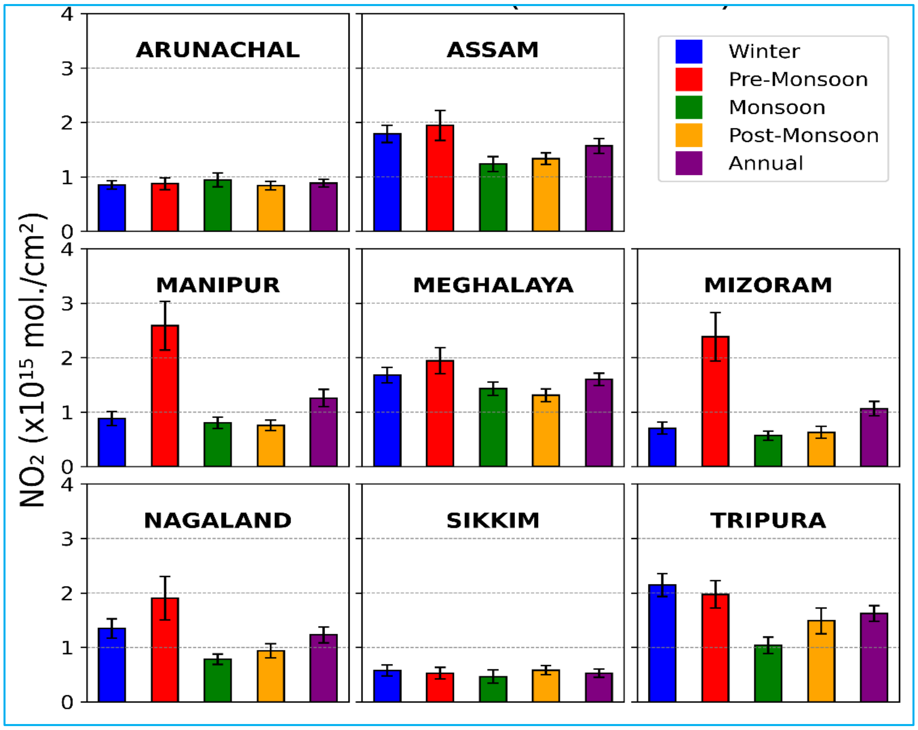

Fig. 4. State-wise seasonal TCN over NERI (2005-2022)

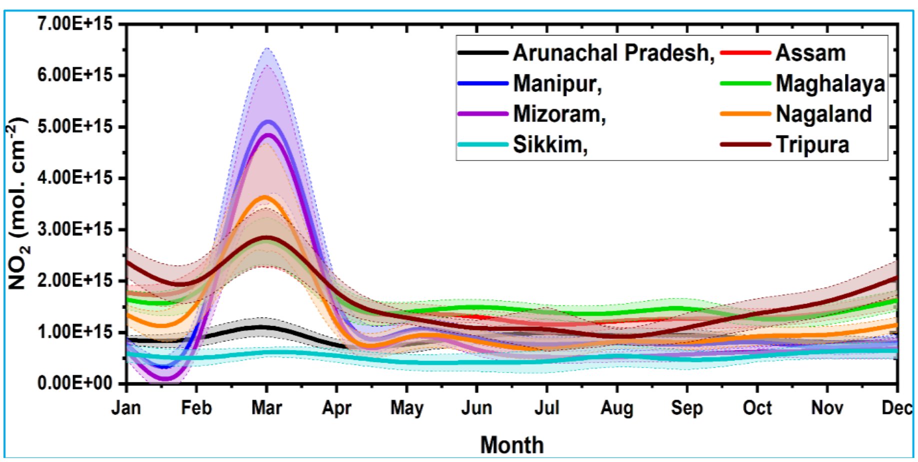

Fig. 5. Average monthly variation of NO_2 over different states of NER

average of 1.0 ± 0.05 × 15 mol. cm^{-2} with a 14% decadal increase in South Asia, linked to anthropogenic emissions from power generation, urbanization, vehicles, and industries.

High concentrations of NO_2 observed during winter may result from the transportation of NO_2 from the IGP to NERI due to westerly winds. Haridas *et al.* (2018) also reported that the IGP shows the highest NO_2 concentrations during winter. Road transport remains the dominant source of NO_2 emissions, with the number of registered vehicles in NERI increasing from 1.4 million in 2001 to approximately 6.1 million by 2019 (https://morth.nic.in). Another significant source of NO_2 emissions is industries, with the number of industries growing from 2200 to 6800 between 2005 and 2020. Population growth is a key factor contributing to the increase of TCN. About three in four rural households in India rely on traditional energy sources for cooking, heating, etc. Census 2011 shows that over 65 percent of

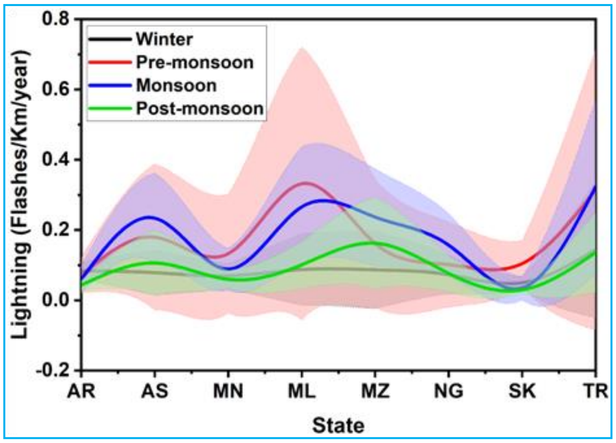

Fig. 6. Average seasonal variation of lightning flashes over different states of NERI

the population resides in rural India and consumes around 45 percent of the total domestic energy. While the use of modern energy sources is increasing, rural India continues to be predominantly dependent on traditional sources (Ranjan and Singh, 2017). Despite this, NERI generates significant agricultural waste, leading to NO_2 generation. In India, the annual crop residue generated for 2008-2009 was about 620 Mt/year, with 15% being burnt on farms, emitting substantial amounts of NO_x (Jain *et al.*, 2014).

3.3. Monthly variation of NO_2

The monthly variation of NO_2 from 2005 to 2022 is depicted in Fig.5 The NO_2 concentration remains relatively flat during the monsoon and post-monsoon seasons. However, a higher peak is observed in the pre-monsoon season, particularly over Manipur, Mizoram, and Nagaland. During this time, westerly air masses carry NO_2 from the North-west direction, flowing along the Brahmaputra valley and encountering the Arbi-Allong hills in Nagaland [Fig. S1(b)]. Lightning also contributes to enhancing the NO_2 concentration in the region. Additionally, air from Tripura and Bangladesh carries NO_2, leading to higher concentrations over Manipur and Mizoram [Fig. S1(b)]. Tripura exhibits higher NO_2 levels than other states due to local burning and transported NO_2 from Bangladesh [Fig. S1(a)]. In Assam, NO_2 concentration is higher than during the Monsoon and post-monsoon seasons, attributed to local and transported NO_2 from the hilly areas [Fig. S1(a)]. NO_2 is primarily anthropogenic and tends to be highest in highly urbanized areas (Lin et al., 2019). Over the ocean, NO_2 levels are generally low. In China, NO_2 exhibits a seasonal variation, with the highest levels in winter and the lowest in summer, influenced by emission sources and meteorological conditions (Lin *et al.*, 2019). The evolution of aerosol particles in the area is influenced by both anthropogenic and natural events (Barman *et al.*, 2018). The aerosols in NERI come from various sources, including open agricultural fields, burning vegetation, combustion releases, brick kilns, coal mines, and oil wells (Pathak *et al.*, 2015; Kundu *et al.*, 2018; Barman *et al.*, 2018). The rugged topography in NERI causes aerosols to be mainly confined to the Brahmaputra valley, where air convergence supports an ideal environment for the accumulation of transported and local pollutants (Pathak *et al.*, 2015).

3.4. *Natural and anthropogenic sources of TCN*

3.4.1. *Natural source*

This is because higher humidity levels lead to stronger hydrometeor concentration and updraft velocities, both of which contribute to intense lightning. Lightning stands as one of the largest natural sources of atmospheric NOx (von Liebig, 1827; Galloway *et al*., 2004; Hutchinson, 1954). The high temperature during lightning causes oxygen and nitrogen to combine and form nitric oxide. NO and NO_2 are collectively referred to as NOx because NO rapidly reacts with O_3 in the atmosphere, producing NO_2, and achieves equilibrium concerning the photo-dissociation of NO_2 within a few minutes, maintaining the sum of both species essentially unchanged (Bradshaw *et al.*, 1999). LNOx have significant implications for atmospheric chemistry and climate (WMO, 1999; IPCC, 2001). In NERI, lightning activity exhibits bimodal variation, with the first prominent peak (13 flashes/pass) in April and a second lower peak spread between September and October (5.5 flashes/pass), resulting in an average value of 8.4 flashes/pass (Kandalgaonkar, 2010). During the monsoon season, lightning activity is minimal, ranging from 4 to 6 flashes/pass. April and May are considered the most lightning-producing months in the sub-Himalayan region of Nepal (Saha *et al.*, 2012). These observations align with previous studies (Kumar and Kamra, 2012). Seasonal lightning activity over NERI remains one of the major natural sources of NO_2. Fig. 6 illustrates the average seasonal variation of lightning flashes across different states in the NERI. The pre-monsoon and monsoon seasons exhibit the highest lightning activity. During the pre-monsoon season, Meghalaya and Tripura record the highest number of lightning flashes, while Arunachal Pradesh experiences the lowest. In the monsoon season, Meghalaya, Tripura, and Mizoram witness the highest lightning frequencies. In the comparison of lightning flashes and $NO_{2,}$ it is observed that the highest lightning states have not shown the highest concentration of NO_2. It is noteworthy that the NO_2 existence over NER indicates anthropogenic emissions e.g. biomass burning, vehicle emission, and long-range transported $NO_{2.}$

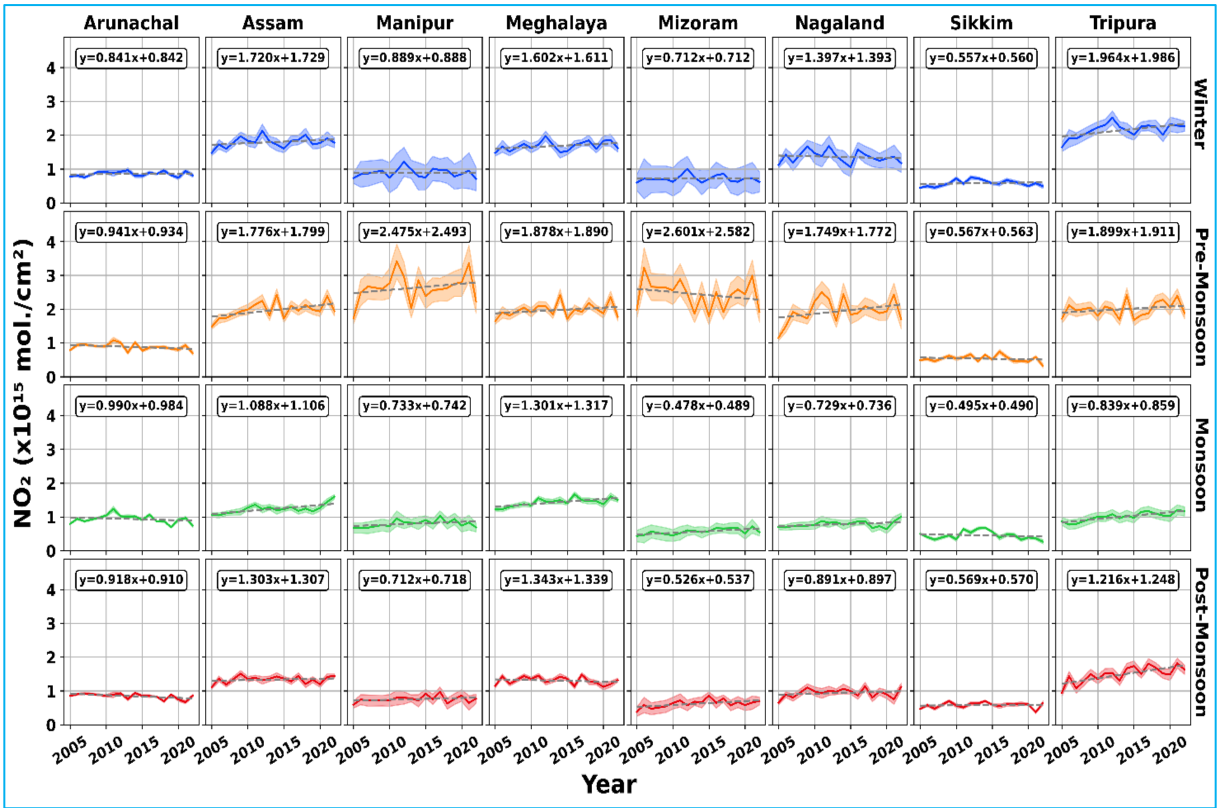

Fig. 7. Sidewise annual variation of TCN from 2005 to 2022

3.4.2. *Anthropogenic source*

Anthropogenic NO_2 primarily originates from combustion engines that burn fossil fuels, such as oil, natural gas, and coal, to produce energy. NOx formation during combustion processes involves three gas phase reaction mechanisms (García *et al.*, 2014): thermal NOx mechanism (resulting from atmospheric nitrogen oxidation at high temperatures >1300 °C), fuel NOx mechanism (involving oxidation of fuel-bound nitrogen), and prompt NOx mechanism (caused by the reaction of CHi-radicals with atmospheric nitrogen in the flame front). For fossil fuel combustion, thermal and prompt NOx mechanisms play a significant role. However, in biomass combustion, the combustion chamber's temperature typically remains below 1300 °C, leading to the dominance of the fuel-N mechanism in NOx formation. Biomass burning can occur naturally or be initiated by human activities such as agriculture (crop residue burning), cooking, and heating. People often utilize solid biomass as a renewable and carbon-neutral energy source, replacing fossil fuels. Despite its environmental benefits, there are concerns regarding biomass combustion in small-scale domestic appliances. In the NERI region, the principal sources of NO_2 are attributed to an increasing number of vehicles and factories, biomass mass burning in household appliances, and the burning of crop residues (Fig. 2).

3.4.3. *LULC changes and forest fire*

India's North Eastern Region (NER) has some of the largest tropical and sub-tropical forest reserves of wet evergreen, semi-evergreen, moist deciduous, coniferous forests, mixed forests, and shrubs (Roy and Joshi, 2002), covering almost 64.66% of its geographic area. Every year forest cover of NERI undergoes significant changes due to shifting (called "Jhum") cultivation. Some people burn forest areas to create new agricultural fields, leading to fires from those fields (Puri *et al.*, 2011) and causing forest fires. Historical data analysis of forest fire counts in NERI shows an average of 100 thousand fire events annually (Sarma *et al.*, 2017). In the FSI, 2021; a decrease of 1020 sq. km of forest area in the region has also been reported due to shifting cultivation, natural calamities, development activities, etc. Spatial variation in peak fire months or seasons was observed over the Indian subcontinent in the study by Chand *et al.* (2006) which was attributed to the varying forest types and climatic conditions throughout the region.

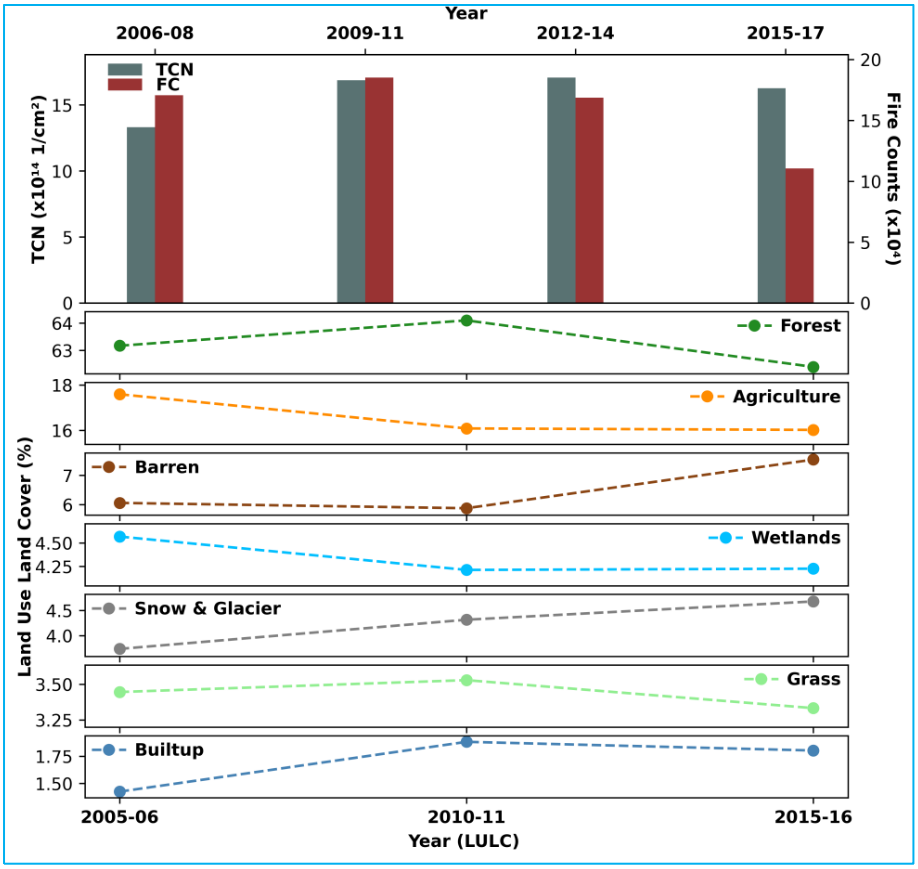

Fig. 8. Variations in TCN, fire counts and LULC types over the NER

For example, the same study stated that they reported intense fire episodes over Himalayan zone forests during May and June every year due to moist conditions in April and prevailing dry weather conditions in May. Whereas, high forest fires were recorded in the tropical evergreen forests of North East India during February, March, and April with a maximum number of ~1200 fires per day. Kharol *et al.* (2008) also recorded high forest fire incidents over NER of India due to the burning of agricultural lands in March and April. In the same study, the author also mentions high forest fire incidents in Mizoram due to forest fires. Fire counts vary from 64,092 to 1, 07,506 in March, 4,972 to 6,204 in January, and 12,268 to 28,393 in February. From 2009 to 2011 there was a maximum fire count in winter and the third maximum fire count in pre-monsoon, related to the drought episodes (NRAA, 2009, 2013). It is known that during drought, less availability of moisture in vegetation makes it more flammable, thereby increasing the probability of forest fire (Littell *et al.*, 2016).

The LULC changes over the north-eastern region of India (Fig. 8) showed an increase in forest cover from 2005-2006 to 2010-2011, primarily due to regeneration in abandoned shifting cultivation areas and afforestation activities (ISFI-FSI 2007). This trend was followed by a decline in forest cover towards 2015-2016 (ISFI-FSI 2013), attributed to factors such as biotic pressure, soil erosion, and ongoing shifting cultivation practices (ISFR-FSI 2021). Agricultural land showed a slight decrease during this period, likely due to land degradation resulting from unsustainable agricultural practices. This degradation may have contributed to an increase in barren land, potentially caused by soil erosion or the abandonment of fields (Singh & Chaudhary, 2023).

The increase in forest density during this period, coupled with episodes of drought (NRAA, 2009, 2013), likely contributed to the high fire counts observed in the winter and pre-monsoon seasons of 2009 - 2011 (Borgohain *et al.*, 2023). Reduced moisture availability in vegetation, combined with the accumulation of dry biomass from denser forests, likely provided ample fuel for forest fires, thereby elevating the risk of fire outbreaks in the region (Littel *et al.*, 2016).

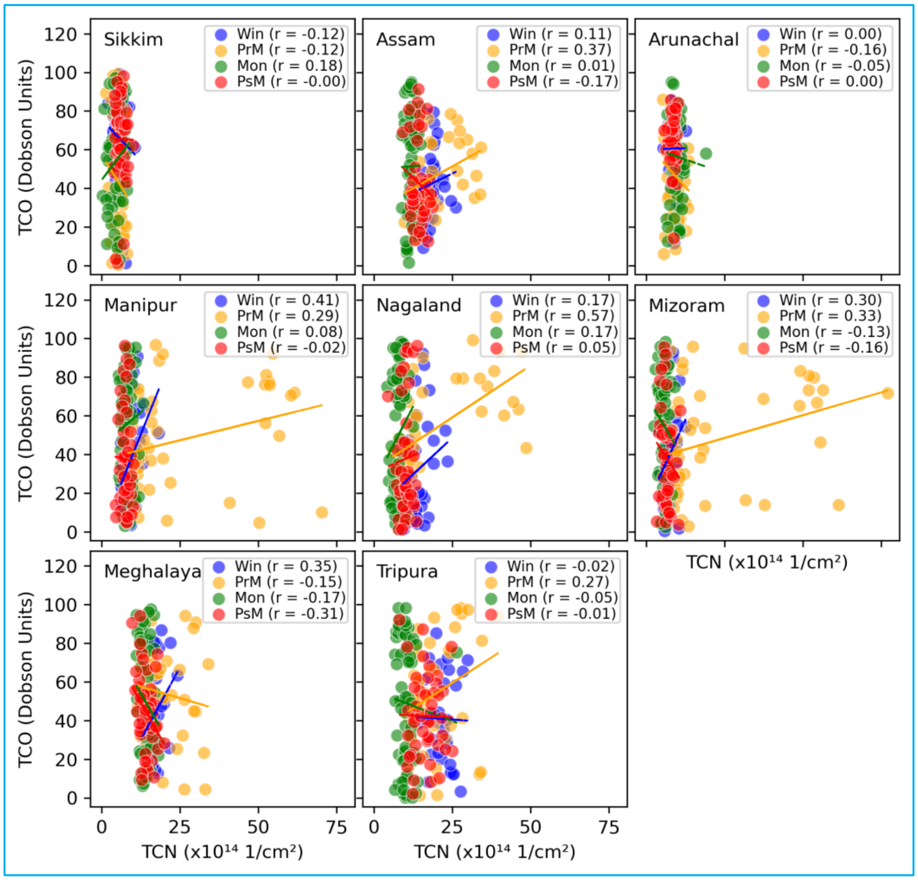

Fig. 9. TCN and TCO over NEI states from 2005-2019

Simultaneously, the rise in built-up areas points to increased urbanization and anthropogenic activities, which are associated with elevated NO_2 levels, a pollutant primarily originating from human activities (Lin *et al.*, 2019). The decline in vegetation cover, indicated by the increase in barren land, may have reduced the region's capacity to act as a sink for pollutants like NO2, leading to its accumulation (Gourdji, 2018).

3.4.4. *TCN and Tropospheric Ozone*

Tropospheric ozone (O_3) is a short-lived climate pollutant formed through the interaction of sunlight with volatile organic compounds (VOCs) and nitrogen oxides (NO_x), which are mainly produced by human activities. O_3 and NO_x, consisting primarily of nitric oxide (NO) and nitrogen dioxide (NO_2), are dynamically interconnected. Nitric oxide reacts with ozone to produce NO_2, establishing a near-equilibrium state within minutes through the photodissociation of NO_2, which stabilizes the combined concentrations of NO and NO_2 (Bradshaw *et al.*, 1999). This equilibrium is why NO and NO_2 are collectively referred to as NO_x.

Sources of NO_x include soil emissions, fossil fuel combustion, biomass burning, and lightning, all of which significantly contribute to atmospheric NO_2 levels (Richter *et al.*, 2005; Lin, 2012). The chemical relationship between O_3 and NO_x is central to atmospheric chemistry, as their concentrations are interrelated (Clapp & Jenkins, 2001). This interplay underscores the role of NO_2 as an indicator of ozone levels and the complexity of tropospheric composition.

The correlation between monthly averaged TCN and troposphere column O_3 (TCO) over the different seasons in the 2005-2019 periods varies across north-eastern Indian states due to differences in emission sources, meteorological conditions, and topography (Fig. 9). In general, winter (Win) and pre-monsoon (PrM) seasons exhibit positive correlations, with the strongest seen in Manipur (r = 0.41) and Nagaland (r = 0.57) during these periods, indicating that higher NO_2 corresponds with increased ozone. Negative or near-zero correlations are more common during monsoon (Mon) and post-monsoon (PsM), as seen in Meghalaya (r = -0.31, PsM) and Mizoram (r = -0.16, PsM), likely due to reduced ozone

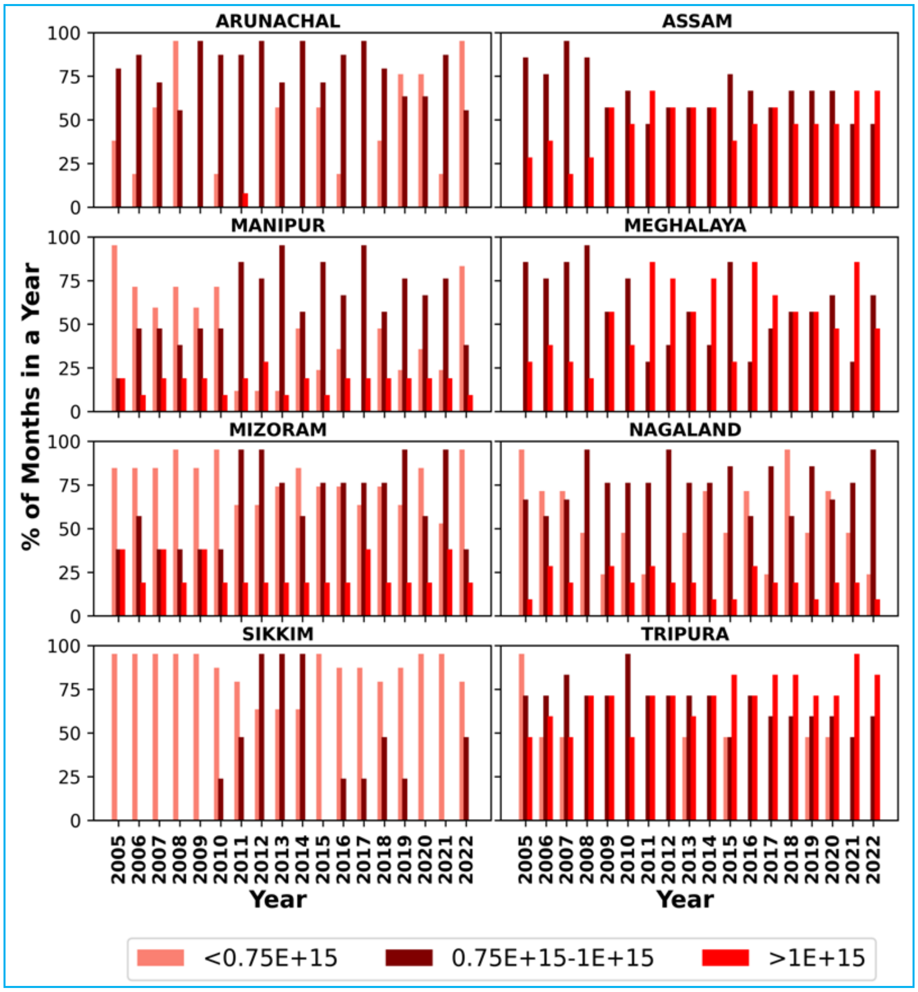

Fig. 10. Frequency analysis of NO_2 Concentrations in NERI

formation from wet deposition and cloud cover. Sikkim and Arunachal in the mountainous terrains show consistently weak correlations across all seasons, while others like Assam and Mizoram have mixed results, with both positive and negative associations depending on the season.

3.5. *Frequency distribution and air quality*

The frequency distribution of the number of days with different levels of TCN concentrations throughout the year is depicted in the figure for all states in the NERI (Fig 10). TCN refers to the amount of nitrogen present in the entire atmospheric column above a specific location, measured in units of mol cm^{-2}. This parameter is crucial for understanding air quality and pollution levels, as nitrogen compounds play a significant role in various atmospheric processes and can have both natural and anthropogenic sources. The figure shows that Tripura and Assam consistently experience more than 50 percent of days each year with TCN values exceeding 1×10^{15} mol cm^{-2}. This indicates that these states often have high concentrations of nitrogen compounds in the atmosphere, which can be attributed to various sources, such as industrial activities, vehicular emissions, and agricultural practices. On the other hand, Meghalaya, Mizoram, and Nagaland have more than 25 percent of days with TCN values ranging between 0.75×10^{15} and 1×10^{15} mol cm^{-2}. Manipur, on the other hand, has approximately 10 percent of days with a similar TCN concentration range. These states exhibit moderately elevated TCN levels, suggesting a moderate impact on air quality, likely due to a combination of both local emissions and atmospheric transport of pollutants from neighboring regions. The findings from this frequency distribution analysis provide valuable insights into the air quality status of the NERI states. The higher TCN concentrations in Tripura and Assam imply that these areas might require specific attention and intervention to address air pollution concerns Similarly, the moderate.

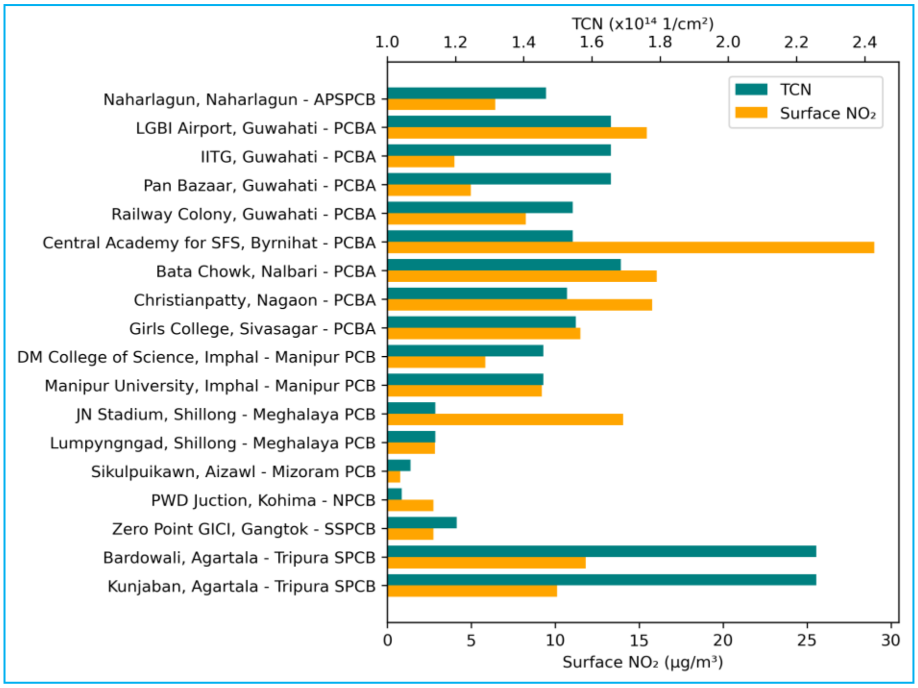

Fig. 11. Surface NO_2 and TCN over CPCB stations

TCN concentrations observed in Meghalaya, Mizoram, Nagaland and Manipur call for continued monitoring and mitigation efforts to maintain and improve air quality in these regions. Overall, this scientific analysis highlights the importance of understanding and managing nitrogen-related air pollution to safeguard the environmental health and well-being of the NERI states.

Surface NO_2 over the years 2019-2023 were collected from the 18 CPCB monitoring stations over the 8 NE states and its average is compared with average TCN data over the same locations and over the same time period. This data is represented in Fig. 11, showing the variation of these concentrations over the different stations. In Assam, the oil refinery, paper, and cement industries are major contributors to air pollution (Pandey & Ghosh, 2000), showing relatively higher emissions of ground level NO_2. Byrnihat exhibits significantly higher surface NO_2 levels compared to other stations, despite having TCN values similar to nearby locations in Assam, potentially indicating substantial local emissions with subsequent dispersion into surrounding areas. The Byrnihat Industrial Area, located near the Assam-Meghalaya border, has been recognized as a critically polluted region by the CPCB over recent years (Borgohain *et al.*, 2024; CPCB, 2020). Conversely, stations in Tripura report the highest TCN values among all monitored locations but show moderate surface NO_2 levels, likely due to emissions from non-ground sources or the transport of pollutants from nearby regions (Borgohain *et al.*, 2023).

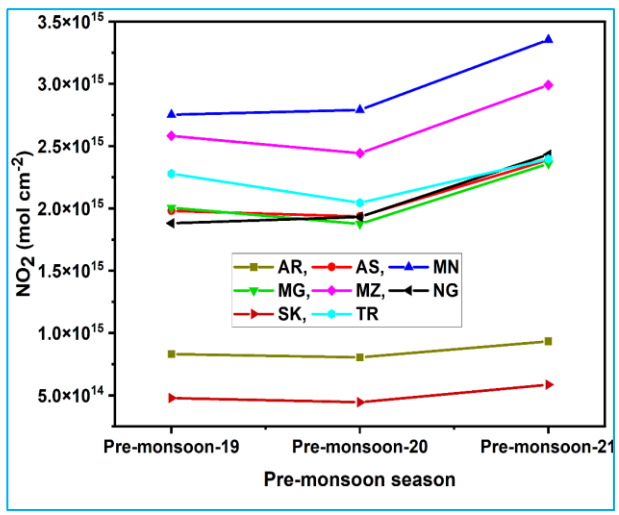

Fig. 12. NO_2 concentration over different states of NERI during and pre and post-lockdown pre-monsoon seasons

3.6. *NO_2 rise after COVID-19*

In Fig. 12 NO_2 concentration is shown over the various states of NERI during and pre and post-lockdown years of pre-monsoon season. In pre-monsoon-20, AR, AS, ML, MZ, TR, and SK show a lower NO_2 concentration compared to pre-monsoon-19 (Table1). At the same time, MN and NG showed a higher NO_2 during pre-monsoon-20 compared to pre-monsoon-19.

TABLE 1

NO_2 concentration over different states of NERI during and pre and post-lockdown pre-monsoon seasons

State	Pre-monsoon-19	Pre-monsoon-20	Pre-monsoon-21
	NO_2 (mol cm^{-2})		
AR	8.30×10^{14}	8.04×10^{14}	9.33×10^{14}
AS	1.98×10^{14}	1.94×10^{15}	2.39×10^{15}
MN	2.75×10^{15}	2.79×10^{15}	3.35×10^{15}
MG	2.01×10^{15}	1.88×10^{15}	2.36×10^{15}
MZ	2.58×10^{15}	2.44×10^{15}	2.99×10^{15}
NG	1.88×10^{15}	1.93×10^{15}	2.43×10^{15}
SK	4.77×10^{15}	4.45×10^{14}	5.86×10^{14}
TR	2.28×10^{15}	2.04×10^{15}	2.40×10^{15}

In pre-monsoon-21, NO_2 concentration increased in all states. In contrast, the rise of NO_2 in AS, ML, MN, NG, and TR are significant. A decrease in NO_2 concentrations has been reported as a result of lockdown measures to reduce the spread of COVID-19 (Koukouli *et al.*, 2021; Field *et al.*, 2020; Bauwens *et al.*, 2020; Liu *et al.*, 2020; Prunet *et al.*, 2020). Questions remain, however, regarding the relationship of satellite-derived atmospheric column NO_2 data with health-relevant ambient ground-level concentrations, and the representativeness of limited ground-based monitoring data for global assessment. Cooper *et al.* (2022) reported with the help of TROPOMI satellite data (during 2019-2020) that NO_2 changes in more than 200 cities, including 65 cities without available ground monitoring, largely in lower-income regions. Mean country - level population - weighted NO_2 concentrations are $29\% \pm 3\%$ lower in countries with strict lockdown conditions than in those without (Cooper *et al.*, 2022). Relative to long-term trends, NO_2 decreases during COVID-19 lockdowns exceed recent Ozone Monitoring Instrument (OMI)-derived year-to-year decreases from emission controls, comparable to 15 ± 4 years of reductions globally (Cooper *et al.*, 2022).

4. Conclusions

In summary, this comprehensive 17-year analysis of TCN concentrations in the NERI has revealed seasonal and long-term trends, shedding light on the dynamics of air quality in this region. TCN levels exhibited distinct variations across the four seasons, with pre-monsoon and winter months consistently displaying the highest concentrations, primarily due to biomass burning and increased anthropogenic activities. The long-term trends in TCN demonstrated an overall increase, reflecting the growing impact of factors like rising vehicle numbers, industrial expansion and population density.

Monthly variation of NO_2 concentrations highlights the significance of the pre-monsoon season, where various factors, including lightning and transported NO_2, contribute to elevated NO_2 levels. Both natural and anthropogenic sources, such as forest fires, biomass burning, and combustion engines, have been identified as key contributors to TCN concentrations. LULC changes in relation to TCN concentrations across different states highlight the effect land cover effects on air quality. Troposphere ozone, as a climate pollutant, was compared with TCN data to explore the interactions between atmospheric pollutants. Frequency distribution analysis of air quality status of NERI states emphasized the need for targeted interventions in areas where elevated TCN levels are observed.

The study also explored the impact of the COVID-19 pandemic, revealing fluctuations in NO_2 concentrations, with some states experiencing lower levels during lockdowns in pre-monsoon-20, followed by increased concentrations in pre-monsoon-21.

This study highlighted the various factors influencing the air quality in NERI and calls for informed measures to address evolving air quality challenges in the face of increasing anthropogenic influences and natural variations. The study offers valuable insights into tropospheric column nitrogen dioxide in the North Eastern Region of India (NERI), with a comprehensive 17-year analysis highlighting seasonal and long-term trends. The study effectively integrates data on both natural and anthropogenic sources and examines the impact of the

COVID-19 pandemic on NO_2 levels. Its focus on the region's specific environmental conditions and the combination of satellite and ground-based data strengthen the findings.

Future research should focus on several key areas, including expanding ground-based monitoring to improve the validation of satellite data, assessing the health impacts associated with variability in NO_2 levels, conducting detailed source apportionment studies, exploring how climatic variables affect TCN and NO_2, and evaluating the impact of air quality regulations. Addressing these areas will enhance our understanding and inform more effective environmental management strategies.

Authors' Contributions

Arup Borgohain: Conceptualization, methodology, manuscript writing, data curation, supervision.
Arban S. Youroi: Data collection and analysis, formal analysis, manuscript writing, visualization.
Rohit Gautam: Writing manuscript and editing, formal analysis.
Manasi Gogoi: Data collection, formal analysis, visualization.
Ribanda Marbaniang: Data collection, formal analysis, visualization
Nilamoni Barman: Data analysis, visualization.
Arundhati Kundu: Data collection, visualization.
Abhay Srivastava: Data collection, visualization.
Shyam S. Kundu: Review and supervision.
S.P. Aggarwal: Review and supervision.

Disclaimer: The contents and views presented in this research article/paper are the views of the authors and do not necessarily reflect the views of the organizations they belongs to.

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