



## Seasonal day-night variation of ozone (O<sub>3</sub>) in an eastern urban site, Bhubaneswar and its association with precursors (NO<sub>x</sub> and NMHC): More sensitivity of O<sub>3</sub> to NMHC than NO<sub>x</sub>

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### Abstract

The study reports on the relationships between surface ozone (O<sub>3</sub>) and its principal precursors, nitrogen oxides (NO<sub>x</sub>) and non-methane hydrocarbons (NMHCs), due to their scientific importance, in addition to the implementation of policies aimed at reducing O<sub>3</sub> pollution. Measurements and analyses of the mentioned trace gases revealed correlations between them in a city in Eastern India, Bhubaneswar, from October 2021 to May 2022. It demonstrates sensitivity to NMHCs when compared to NO<sub>x</sub> as a whole. The NMHC/NO<sub>x</sub> ratios confirm NMHC dominance in pre-winter and winter, while NO<sub>x</sub> dominance is confirmed in summer (post-winter). A multivariate regression approach was also utilised to develop a model to predict O<sub>3</sub> concentrations based on precursor concentrations, which offers significant information on the precursors (NO<sub>x</sub> and NMHCs) that contribute to O<sub>3</sub> synthesis. The slope and intercept derived from the prediction model indicate that, while these precursors can explain a significant portion of the concentration, there are other unidentified contributions from other precursors and some mysterious chemical activities going on (especially at night) that has remained unrevealed. Thus, the study emphasizes the necessity of identifying additional precursors via other methodologies such as remote sensing and limiting NMHC emissions, which could play a crucial role in both understanding and lowering ground-level ozone in the current scenario.

### 1. Introduction

In terms of trace gas concentrations, the atmospheric environment over an area is an open system that is influenced by both local and regional inputs. These contributions originate from the interaction of emission and transport mechanisms, which varies depending on the location of the atmosphere and the season of the year. Furthermore, chemical, photochemical, dynamical, and meteorological all have an impact on trace gas levels across a place. Quantification of local variability becomes critical in major urban areas; viz., megacities are key providers (and sometimes sinks) for a wide range of species,

influencing chemistry and the economy at regional to larger scales [1]. The characterization is improved when various chemical species are measured, which can act as substitutes for various sources and sinks. Some of these gases, such as ozone (O<sub>3</sub>), carbon monoxide (CO), nitrogen oxides (NO<sub>x</sub>), non-methane hydrocarbons (NMHCs), methane (CH<sub>4</sub>), Sulphur oxides such as Sulphur dioxide (SO<sub>2</sub>), and others, play vital roles in numerous atmospheric processes that impact our general health and ecosystem.

O<sub>3</sub> is a chemical that has piqued the scientific community's interest for many years. Ground-level O<sub>3</sub> is a significant secondary pollutant produced by the photochemical interaction of several main pollutants. Understanding the relationship between O<sub>3</sub> and its precursors (mostly NO<sub>x</sub> and NMHCs) is an important task in atmospheric chemistry because, while stratosphere-troposphere interchange is one source of tropospheric ozone, photochemistry is the dominating source. Several studies have been undertaken around the world to establish if O<sub>3</sub> developed in NO<sub>x</sub>- and NMHCs-limited settings, with regression analysis of O<sub>3</sub> being used to evaluate its restriction by either species. Regional air quality models were also utilized to explore O<sub>3</sub> sensitivities in the Pearl River Delta (PRD) region, and it was discovered that the formation regime was volatile organic compound (VOC)-limited in the morning and NO<sub>x</sub>-limited at peak O<sub>3</sub> hours [2]. Using a box model based on observations, [3] investigated the sensitivity of O<sub>3</sub> to its precursors during episodes of O<sub>3</sub> pollution. They found that O<sub>3</sub> sensitivity was between the transitional and VOC-limited regimes. [4] used the WRF-Chem model to study the seasonality of O<sub>3</sub> pollution and its sources in both India and China, and found that the sensitivity of O<sub>3</sub> in India was highest to transportation and biogenic sources and that addressing these might be substantial in reducing O<sub>3</sub> pollution. In the recent past, several observations of surface O<sub>3</sub> and its precursors were made over urban areas of Bhubaneswar [5]–[10]. This research has revealed the essential temporal interactions between surface O<sub>3</sub> and its precursor pollutants over the megacity. However, no research on NO<sub>x</sub> and NMHCs, as well as the sensitivity of O<sub>3</sub> to any of them, has been published in the Bhubaneswar atmosphere.

The role of NO<sub>x</sub> and NMHCs in influencing tropospheric O<sub>3</sub> is well established. These gases also have a significant impact on air quality and global climate change. It should be highlighted that continuous representative measurements of O<sub>3</sub>, NO<sub>x</sub>, and NMHCs are presently limited to only a few locations in India, and so knowledge regarding their prevalence and variability could have fundamental consequences for understanding and predicting tropical atmospheric chemistry. In this research, we describe a seasonal measurement of O<sub>3</sub>, NO<sub>x</sub>, and NMHCs (C<sub>2</sub>-C<sub>4</sub>) over the city of "Bhubaneswar," an urban location in eastern India. Because of its location in tropical South Asia, Bhubaneswar has a substantial impact on regional chemistry. Surface data shows that there are high levels of pollution over Bhubaneswar throughout the winter [8]. According to their findings of increased surface air pollution concentrations in Bhubaneswar during the winter months, an attempt has been made to analyse the distribution of C<sub>2</sub>-C<sub>4</sub> NMHCs, NO<sub>x</sub>, and O<sub>3</sub> as well as the photochemical behavior of the species from October 2021 to May 2022 for three seasons (post-winter, winter and summer). Despite the fact that winter is not a notable season in the area, the concentration of numerous airborne contaminants is noted to be more obvious due to a stable atmospheric state and long-range transmission from the Indo-Gangetic plains (IGP).

Furthermore, multivariate regression (MLR) analysis was utilised to better understand the relationship of precursors with O<sub>3</sub> and to develop a model for forecasting O<sub>3</sub> content based on its precursor concentrations. Multiple linear regression was used to assign the concentration of O<sub>3</sub> to the appropriate precursors. The current study's MLR model could be refined using the following formula:

$$y = \beta_1x_1 + \beta_2x_2 + \beta_3x_3 + \dots + \beta_ix_i + C \quad (1)$$

where y represents the O<sub>3</sub> concentration, 1-i represent distinct O<sub>3</sub> precursors' concentrations, x<sub>1</sub>-x<sub>i</sub> represent precursor contributions to O<sub>3</sub>, β<sub>1</sub>-β<sub>i</sub> indicate regression coefficients, and C is the constant. The acquired regression models were used to detect the responses (or contributions) of different precursors to O<sub>3</sub> concentration and to predict the O<sub>3</sub> concentrations based on different precursors' concentrations, respectively [11].

## 2. Site description

The sampling location is the Council of Scientific and Industrial Research-Institute of Mineral and Material Technology (CSIR-IMMT) laboratory (20°30'N, 85°83'E) in Bhubaneswar, the state capital of Odisha. Bhubaneswar is one of the most rapidly developing cities in the country's eastern region. The station experiences varied weather conditions throughout the year, including pre-monsoon or summer (March-May), monsoon (June-August), post-monsoon or pre-winter (Sept-Nov), and winter (Dec-Feb). The average temperature ranges from roughly 10°C to 30–35°C. During the winter, the region receives cold winds

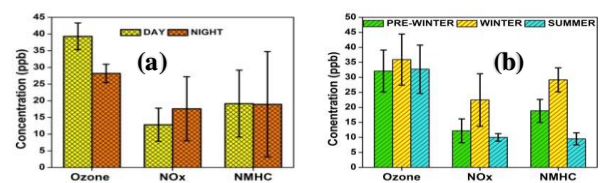
from the north and northeast at an average pace of 7 miles per hour, and temperatures drop to around 15°C. The city has an estimated population of 1.2 million people and is surrounded by tiny townships (within 150 kilometers) that are home to numerous industrial and production companies [8], [12].

## 3. Experimental details

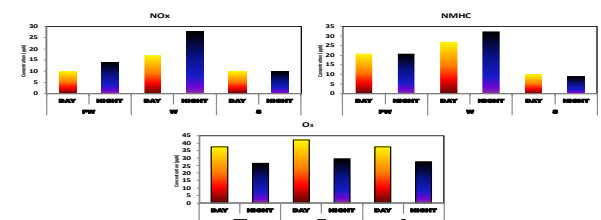
Ground-based analyzers were used to perform these measurements. Surface O<sub>3</sub> and CO levels are being measured continuously on the roof of the institute of CSIR-IMMT, Bhubaneswar. The O<sub>3</sub> analyzer (Thermo Scientific Model 49i) measures ambient O<sub>3</sub> using UV Photometric technology and is calibrated with an internal ozonator and zero gas. Simultaneous NO<sub>x</sub> measurement was performed using a chemiluminescence NO<sub>x</sub> analyser (Thermo Fisher Scientific, 42i). All instruments were calibrated on a regular basis with NIST traceable calibration standards and a multi-channel calibrator. More information is available from [8], [9], [12], [13]. The experimental equipment for NMHC analysis comprises a PerkinElmer Clarus 480 Gas Chromatograph (GC) with a flame ionization detector (FID) and a 30m x 0.53mm Elite-Alumina capillary column for NMHC analysis, whose working can be found in [8]. Sampling was done for 12 hours (8 am -8 pm) from ambient air, twice a week. Individual NMHCs were calibrated first using a standard gas mixture supplied by Alchemie Gases & Chemicals (certified by TUV India Private Limited) and then using National Institute of Standards and Technology (NIST) traceable standard gases supplied by the Linde group (USA).

## 4. Result and discussion

### 4.1. Day-night and seasonal variations in O<sub>3</sub>, NO<sub>x</sub>, and NMHCs



**Figure 1.** Average (a) day-night and (b) seasonal concentration of O<sub>3</sub> and its precursors (NO<sub>x</sub> and NMHCs)



**Figure 2.** Seasonal day-night variations of NO<sub>x</sub>, NMHCs and O<sub>3</sub> during the measurement period.

**Table 1.** Day-night in O<sub>3</sub>, NO<sub>x</sub>, and NMHCs

	DAY (06:00-17:00 h)	NIGHT (18:00-15:00 h)
O <sub>3</sub>	39.30 ± 4.01	28.19 ± 2.75
NO <sub>x</sub>	12.82 ± 4.96	17.59 ± 9.61
NMHCs	19.15 ± 10.03	18.95 ± 15.80

**Table 2.** Seasonal variations in O<sub>3</sub>, NO<sub>x</sub>, and NMHCs

	POST-WINTER	WINTER	SUMMER
O <sub>3</sub>	32.09 ± 6.99	35.91 ± 8.51	32.70 ± 8.06
NO <sub>x</sub>	12.18 ± 3.98	22.44 ± 8.77	9.99 ± 1.26
NMHCs	18.83 ± 3.83	29.16 ± 4.01	9.51 ± 2.02

**Table 3.** Average seasonal day-night variation of O<sub>3</sub> and its precursors (NO<sub>x</sub> and NMHCs)

	POST-WINTER		WINTER		SUMMER	
	DAY	NIGHT	DAY	NIGHT	DAY	NIGHT
O <sub>3</sub>	37.54 ± 6.01	26.64 ± 1.15	42.09 ± 8.15	29.73 ± 1.33	37.70 ± 8.88	27.69 ± 1.52
	10.22 ± 1.34	14.13 ± 9.05	17.21 ± 1.48	27.67 ± 5.21	10.17 ± 1.30	9.82 ± 6.26
	20.94 ± 11.08	20.83 ± 14.47	27.01 ± 7.05	32.38 ± 10.68	10.08 ± 4.88	8.65 ± 4.90
NO <sub>x</sub>						
NMHCs						

Maximum NO<sub>x</sub> mixing ratios were recorded at night, while minimum mixing ratios were observed during the day, particularly at noon (Fig. 1 (a)). After the sun goes down, NO<sub>x</sub> titration processes kick in, resulting in a reaction of NO with O<sub>3</sub> produced throughout the day to yield NO<sub>2</sub> [1] Furthermore, automotive pollutants, which are particularly strong during late evening and night traffic hours, accumulate in the falling nighttime boundary layer. During the day, however, photochemical destruction of NO<sub>2</sub> to produce O<sub>3</sub> could be the reason for the decreased mixing ratios. The daytime and nighttime NO<sub>x</sub> mixing ratios differ by roughly 40%, with the nighttime being higher during the study period (Table 1). The winter season had the highest NO<sub>x</sub> mixing ratios, followed by the pre-winter and summer seasons (Table 2). During the winter, the night-time NO<sub>x</sub> mixing ratios were consistently 60% more than the daytime mixing ratios, followed by pre-winter and summer (Table 3).

NMHCs consistently exhibit the highest mixing ratios during the winter months (Fig 1 (b)). The creation of a persistent nocturnal boundary layer (NBL) may be a beneficial element in the accumulation of NMHCs, resulting in enhanced mixing ratios in ambient air, which interferes with the commencement of photochemical reactions as the sun rises [21]. During the winter season, the nighttime NMHC mixing ratios were approximately 20% greater than the daytime NMHC mixing ratios (Table 3). Thus, the winter season had the highest NMHC mixing ratios, whereas the summer season had the lowest (Table 2). Seasonal fluctuations in NMHC mixing ratio are

primarily caused by differences in the categories and strengths of sources, air mass transfer, convection processes, and the seasonal abundance of OH radicals [22]. [23] also found seasonal dependence in NMVOCs, with a clear winter maximum and a significant decrease in mixing ratios from winter to monsoon. They discovered that local sources of emissions (anthropogenic, biogenic, and biomass burning) have a strong influence on the seasonal changes of NMVOCs. Photo-oxidation of NMHCs by OH radicals is another critical step. Because of increased chemical removal reaction rates produced by more sunlight and higher temperatures, the chemical elimination of these NMHCs by OH radicals is significantly faster in summers, or post-winter days. Furthermore, due to intense solar flux and water vapour, the largest concentration of OH radicals in the tropics is observed during the summer season [24]. This also implies that the higher the OH radical concentration, the greater the chemical elimination of NMHCs, and thus the lowest mixing ratios were recorded during this season.

During the study period, the seasonal average mixing ratio of O<sub>3</sub> was 32 ± 7 ppb (pre-winter), 36 ± 8 ppb (winter), and 33 ± 8 ppb (summer or post-winter) (Table 2). This is caused by the photo-oxidation of O<sub>3</sub> precursor gases during the day, which results in the creation of O<sub>3</sub>. This remains a prevalent property of any metropolitan site where precursor gas concentrations are suitable for O<sub>3</sub> generation activities. However, the lowest mixing ratios were found at night (Fig. 1 (a)). The absence of photochemical reactions, NO<sub>x</sub> titration mechanisms, and surface depositional procedures of O<sub>3</sub> in the low nocturnal boundary layer is most likely to blame [24], [25]. Throughout all seasons, the daytime O<sub>3</sub> mixing ratio has always exceeded the nighttime O<sub>3</sub> mixing ratio (Table 3). The average night-time values were almost 29% lower than the average daytime O<sub>3</sub> during pre-winter, and the differences observed were also nearly 29% lower during winter. During the summer season, however, the average daytime and nighttime O<sub>3</sub> mixing ratios differed by about 26%. This could occur because radiant heat disrupts the surface air, causing the stored contaminants to disperse. As a result, the gap is relatively smaller than in the pre-winter and winter seasons, when significant night-time cooling permits denser cold air to settle and suppresses the vertical movement of low-level air, resulting in a build-up of pollutants. Maximum ozone mixing ratios have been routinely reported throughout the winter season, which is about 10% greater than in other seasons (Table 2). Low winter temperatures and the use of low-quality fuels for domestic heating, along with inadequate dispersion of air pollutants due to low mixing heights, a lack of vertical dispersion of pollutants, and low wind speeds, In the winter, increased concentrations may be ascribed to fuel usage for heating and less production of OH radical (due to low solar radiation). Furthermore, winter maxima may be caused by post-harvesting activities, poor photochemical removal [26], and inadequate air mass circulation because of lower mixing heights, a paucity of vertical dispersion of pollutants [27], and low wind speeds [28].

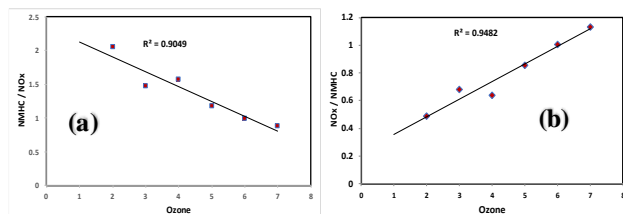
Long-distance transport of air pollutants could be a prominent option at this study location, especially during the summer days, when contaminated air masses travel and create O<sub>3</sub> via photo-oxidation processes in 1-3 days [29]. This could also explain why in summer O<sub>3</sub> mixing ratios are so high. Regional and local emission levels, photochemistry, boundary layer mechanics, convective and advective processes, solar flux fluctuations, prevailing meteorology, and air mass type all influence the day-night and seasonal pattern of trace gases [22], [29], [30].

## 4.2. Variations of the NMHC/NO<sub>x</sub> and NO<sub>x</sub>/NMHC ratio

For the study period, the overall seasonal NMHCs to NO<sub>x</sub> ratio and the NO<sub>x</sub> to NMHCs ratio were also estimated (Table 4). These ratios were found to vary between day and night. As a result, the NMHC/NO<sub>x</sub> and NO<sub>x</sub>/NMHC ratios were examined both during the day (06:00-17:00 h) and at night (18:00-05:00 h). The average daytime NMHC/NO<sub>x</sub> ratio was 1.49, and the NO<sub>x</sub>/NMHC ratio was 0.67, whereas the average nighttime ratios were 1.08 and 0.93, respectively. As a result, the results are consistent with an NMHC-sensitive regime. (Fig. 3) depicts differences in [NMHCs]/[NO<sub>x</sub>] and [O<sub>3</sub>] and [NO<sub>x</sub>]/[NMHC] and [O<sub>3</sub>]. The results revealed both a direct (R<sup>2</sup> = 0.95) and an inversely proportional (R<sup>2</sup> = 0.90), demonstrating that ozone grows with rising NO<sub>x</sub> and decreases with decreasing NMHCs. On a seasonal basis, the mean NMHCs/NO<sub>x</sub> ratio recorded was 1.55, 1.30, and 0.95 during pre-winter, winter, and post-winter days, respectively, while the NO<sub>x</sub>/NMHCs ratio observed was 0.65, 0.77, and 1.05. The NMHC/NO<sub>x</sub> ratios confirm NMHC dominance in pre-winter and winter, while NO<sub>x</sub> dominance is confirmed in summer (post-winter). This suggests that ozone formation conditions are expected to be more favorable during all three seasons, as they are all closer to the optimal ratio.

**Table 4.** Seasonal day-night NMHCs/NO<sub>x</sub> and NO<sub>x</sub>/NMHCs ratio

	PW		W		S	
	DAY	NIGHT	DAY	NIGHT	DAY	NIGHT
NO <sub>x</sub> /NMHC	0.49	0.68	0.64	0.85	1.01	1.13
NMHC/NO <sub>x</sub>	2.05	1.47	1.57	1.17	0.99	0.88



**Figure 3.** Variations between (a) NMHC/NO<sub>x</sub> vs O<sub>3</sub> and (b) NO<sub>x</sub>/NMHC vs O<sub>3</sub>

## 4.3. Correlation between Ozone (O<sub>3</sub>) and its precursors (NMHCs, NO<sub>x</sub>)

Regression analysis was used to study the relationship between ozone (O<sub>3</sub>) and its precursors [31]. The most significant contribution of the precursors to O<sub>3</sub> generation was determined using multivariate linear regression (MLR). The model used to determine the correlation produced the given equation for predicting O<sub>3</sub> based on precursor concentration.

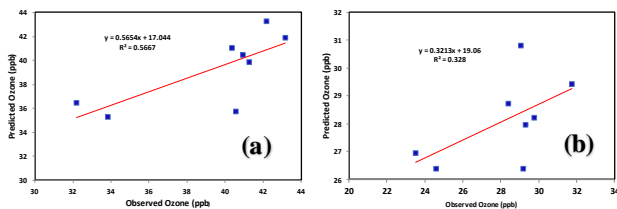
$$[O_3]\text{-day} = 31.57 + (-0.02) \times [NMHC] + 0.63 \times [NO_x] \quad (2)$$

$$[O_3]\text{-night} = 25.33 + (-0.02) \times [NMHC] + 0.18 \times [NO_x] \quad (3)$$

Table 5 summarizes the statistical results generated by the model individually for day and night. Individual precursor contribution to O<sub>3</sub> concentration was estimated using the prediction regression equation. The contribution of the precursors to each concentration could be determined using the prediction equation, and the yearly and seasonal averages of distinct precursors could also be calculated. In Fig. 4, the anticipated O<sub>3</sub> concentration according to the equation above is represented as a function of measured O<sub>3</sub> for both day and night. For the model, the coefficient of determination (R<sup>2</sup>) between observed and predicted concentration is 0.57 with a slope of 0.57 and an intercept of 17.04 during the day and 0.33 with a slope of 0.32 and an intercept of 19.06 during the night. As an ideal model, anticipated concentration as a function of observed concentration has a slope of one and a zero intercept. The slopes less than one and non-zero intercepts observed indicate that, while these precursors can explain a significant portion of the concentration, there are other unidentified contributions from other precursors and some unexplained chemical activities going on, particularly at night, that are not included. These unexplained sections are most likely due to several other precursors such as CO or any other volatile organic compounds (VOCs) that have yet to be identified but play a vital role in the creation of O<sub>3</sub>.

**Table 5.** Seasonal day-night Observed and Predicted O<sub>3</sub> concentration.

		OBSERVED O <sub>3</sub> -DAY	PREDICTED O <sub>3</sub> -DAY	OBSERVED O <sub>3</sub> -NIGHT	PREDICTED O <sub>3</sub> -NIGHT
PRE-WINTER	OCT	33.82	35.29	23.50	26.95
	NOV	41.25	39.89	29.78	28.22
WINTER	DEC	40.95	40.48	28.41	28.74
	JAN	42.17	43.25	29.04	30.82
	FEB	43.15	41.89	31.73	29.43
	MAR	40.34	41.06	29.31	27.97
SUMMER	APR	32.19	36.49	24.58	26.39
	MAY	40.55	35.77	29.19	26.40



**Figure 4.** Variation between (a) Observed O<sub>3</sub> - day vs Predicted O<sub>3</sub> - day (b) Observed O<sub>3</sub> - night vs Predicted O<sub>3</sub> - night

## 5. Conclusions

Photochemical oxidation activities of main pollutants such as NMHCs, CO, CH<sub>4</sub>, and others in the polluted urban environment of Bhubaneswar produce free radical species such as HO<sub>2</sub> and RO<sub>2</sub> radicals. These complex and many reactions are at the heart of the photochemical O<sub>3</sub> generation processes in the polluted urban environment, for which NO<sub>x</sub> and NMHCs are clearly the required raw ingredients, along with sunshine. NMHCs consistently exhibit the highest mixing ratios during the winter season. Maximum NO<sub>x</sub> mixing ratios were seen at night, whereas minimal mixing ratios were observed during the day, which is the inverse of O<sub>3</sub> mixing ratios. The results suggest that, in the present situation, surface O<sub>3</sub> at the research location during the period of study is more sensitive to NMHCs than NO<sub>x</sub> as a whole. The NMHC/NO<sub>x</sub> ratios support NMHC dominance in pre-winter and winter, while NO<sub>x</sub> dominance is confirmed in summer (post-winter). As a result of the obtained data, it is possible to deduce that reducing NMHCs emissions could lower the O<sub>3</sub> mixing ratios over the region. The majority of NMHC species have been shown to come from anthropogenic and biogenic sources, including burning fossil fuels, leaking natural gas, storing and transporting fuel, burning biomass, using gasoline as a solvent and evaporating it, using it as a solvent in other processes, emitting from plants, and geochemical processes. They also have a significant sink because of their reactivity with OH radicals. Because of the lack or reduction of NMHCs reactivity processes, it is possible that steps to reduce NMHCs will initially result in reduction of O<sub>3</sub> synthesis. A multivariate regression technique was also utilized to develop a model to predict O<sub>3</sub> concentrations based on precursor concentrations, which offers significant information on the precursors (NO<sub>x</sub> and NMHCs) that contribute to O<sub>3</sub> synthesis. The slope and intercept deduced from the prediction model indicate that, while these precursors can explain a significant portion of the concentration, there are other unidentified contributions from other precursors and some mysterious chemical activities going on (especially at night) that have remained unrevealed. These unaccounted-for components are most likely due to additional precursors like CO or any other volatile organic compounds (VOCs) that have yet to be identified but play a vital role in the creation of O<sub>3</sub>. Thus, the study emphasizes the importance of identifying other precursors via other methodologies such as remote sensing

as well as controlling and monitoring NMHC emissions, which could play a key role in both studying and reducing ground-level ozone in the current setup. Given the importance of O<sub>3</sub> and its numerous antecedents, additional in-depth research should be conducted at more sites across India.

## 6. Acknowledgements

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