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Seasonal Patterns and Trends of Air Pollution in the Upper Northern Thailand from 2004 to 2018

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ABSTRACT

The seasonal patterns and trends of air pollutants can be used in assessments of air quality, facilitating authorities' decisions on policies for monitoring and managing. The objective of this study was to investigate the seasonal patterns and trends of air pollutants in the upper northern Thailand (UNT) from 2004 to 2018. The hourly air pollutant concentration data including carbon monoxide (CO), nitrogen dioxide (NO_2), nitrogen oxide (NO_x), sulfur dioxide (SO_2), ozone (O_3), and particulate matter ≤ 10 µm (PM_{10}) recorded from 6 monitoring stations in the UNT were obtained from the Pollution Control Department, Ministry of Natural Resources and Environment of Thailand. Cubic splines were used to assess seasonal patterns and trends of the air pollutants. Linear regression was used to estimate the average increase in concentrations of air pollutants at each monitoring station. The results exhibited seasonal patterns for CO, NO_x, NO_2, O_3 and PM_{10}, in all stations while SO_2 exhibited seasonal patterns only in one station in Lampang and all stations in Chiangmai. The concentrations of these pollutants rose during August and September and reached peak levels in March. In the past 15 years, the levels of overall CO, O_3 and SO_2 in the UNT had significantly increased, on average by 0.015 ppm, 0.012 ppb and 0.015 ppb, respectively. In contrast, NO_2, NO_x and PM_{10} had significantly decreased on average of –0.010 ppb, –0.008 ppb and –0.011 µg m^{-3}, respectively. The results provide information to the authorities for setting up proper measures and policies to control and mitigate air quality impacts especially in the dry season.

Keywords: Air pollutants, Air quality, Particulate matter, Northern Thailand

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1 INTRODUCTION

The ambient air pollution level has received considerable attention in many countries, since air quality is a critical factor in climate change and also causes environmental and public health problems (Orru *et al.*, 2017). Air pollution is caused by various events or activities, including human activities, volcanic eruptions, forest fires, crop burning, fossil fuel combustion for transportation, and various factories and power plants (Li *et al.*, 2016). The main air pollutants contaminating in the atmosphere and affecting human health and environment consist of carbon monoxide (CO), ground-level ozone (O_3), nitrogen oxides (NO_x), sulfur dioxide (SO_2) and particulate matter (PM) (Mustafić *et al.*, 2012).

Investigating patterns and trends of air pollutant concentration is essential for authorities to set up proper air quality control policies and impact mitigation plans. The repeating patterns of air pollutants generally can exhibit variations by season, whereas a trend identifies tendency to increase, decrease or remain constant over time. Most air pollutants vary by season in the tropical countries such as Malaysia and Thailand, where CO, O_3, NO_2, SO_2 and PM_{10}, are comparatively high from January to March (Mohtar *et al.*, 2018). All of these air pollutants are commonly higher in the dry season and lower in the rainy season (Matsuda *et al.*, 2006; Kliengchuay *et al.*, 2018, Lalitaporn, 2018; Outapa and Ivanovitch, 2019; Lalitaporn and Boonmee, 2019). Air pollution trends differ by pollutant and by country. In Japan and Thailand, decreasing trends of CO, NO_x, SO_2 and PM were found, whereas O_3 was increasing (Aziz *et al.*, 2016; Ito, *et al.*, 2021). In Malaysia, CO and SO_2 had decreasing trends, and O_3 an increasing trend (Mohtar *et al.*, 2018).

The northern region of Thailand faces major smog problems yearly due to crop residue burning during the dry season. The burning causes various pollutants, including particulate matter, gases and biological molecules, whose levels tend to exceed the air quality standards (Wiwatanadate and Liwsrisakun, 2011). The upper northern Thailand (UNT) has vast mountains reaching high altitudes, causing variable seasonal temperatures. The weather in the North is hot and humid, alternating with the dry season. There are three predominant seasons: the rainy season (May–October), the cold season (October–February), and the hot season (February–May) (Chantara *et al.*, 2012). Every year between December and April, drought and forest fires occur together with crop residue burning by farmers to prepare for the next planting season at the onset of the rainy season. These activities cause haze over the whole UNT region (Chuang *et al.*, 2016). Additionally, the government has formed policies and measures to prevent and tackle northern haze and forest fires. The reduction in burning in agricultural areas depends on provincial policy measures, which operate as guidelines under the existing legal mechanisms such as the Public Health Act 1992 and the National Disaster Prevention and Mitigation Act 2007, to control areas or the sources that cause PM_{10} (Department of Disaster Prevention and Mitigation, 2015).

The Thai Pollution Control Department, Ministry of Natural Resources and Environment, has continuously collected hourly air quality data in UNT from six air monitoring stations since 1996 until present (Pollution Control Department, 2020). However, the utilization of these longitudinal air pollution data to assess the air pollution situation in this area has not been vigorous. Moreover, there are limited studies on air pollutants situation in this area using rigorous statistical methods. Therefore, this study aimed to investigate seasonal patterns and trends of air pollutants in the UNT.

2 METHODS

2.1 Data Source

The air quality data collected from 2004 to 2018 were obtained from the Pollution Control Department, Ministry of Natural Resources and Environment of Thailand. The hourly air pollutant concentration data of CO, O_3, NO_2, NO_x, SO_2 and PM_{10} were assessed from 6 air quality monitoring stations in UNT (3 stations in Lampang, two stations in Chiang Mai, and one station in Nan provinces) as shown in Table 1. The locations of the monitoring stations are shown in Fig. 1.

The national ambient air quality standards in Thailand and the ambient air concentration measurement methods for each pollutant are listed in Table 2. The average values of the measurements were transmitted hourly to a central computer from each station by a data logger and a modem.

The outcomes of this study are assessment of air pollutant concentrations of CO, O_3, NO_2, NO_x, SO_2 and PM_{10}. The hourly observations of these pollutants were aggregated into weekly averages, thus obtaining 52 observations per year. The determinant is time (52 weeks from 15 years each). There are 780 records for each station. Data transformation was performed by taking the natural logarithm, after adding one and dividing or multiplying constant value to all air quality pollutants.

2.2 Statistical Analysis

Descriptive analysis was carried out to summarize the data using measures of central tendency and dispersion. Boxplots and quantile-quantile (Q-Q) plots were used to assess the distribution of each pollutant. The trends and seasonal patterns of air pollutants were assessed from weekly average data. The seasonal patterns were presented by plotting the average weekly air pollutant concentration against the weeks of the year by station. The trends of air pollutants were shown by plotting the average weekly air pollutant concentrations against 15 years by station. The spline

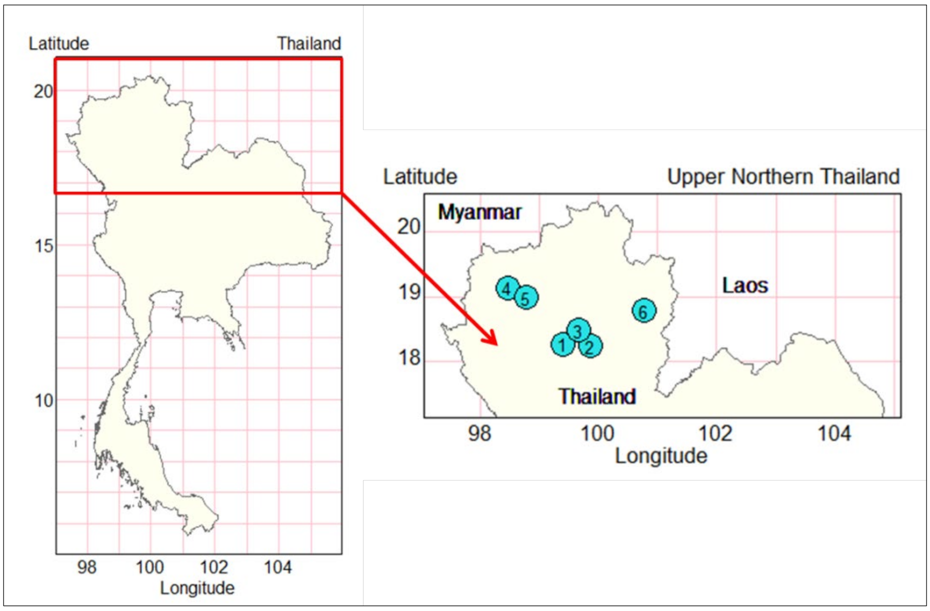

Fig. 1. Map of the study area in the UNT.

Table 1. The characteristic of six air quality monitoring stations in the UNT.

Province (code)	Monitoring station name	Air quality time interval	Latitude (°E)	Longitude (°N)	Characteristics
1. Lampang (LP1)	Meteorological Department	2004–2018	18.29	99.51	Sub-urban, 300 meters distance to the road, close to the airport, 269 meters above sea level
2. Lampang (LP2)	District Health Promoting Hospital, Ban Sop Pa	2004–2018	18.27	99.77	Rural, 5 meters distance to the quiet road, a park surrounding, 269 meters above sea level
3. Lampang (LP3)	Mae Moh Provincial Waterworks Authority	2004–2018	18.28	99.66	Rural, a park surrounding, 269 meters above sea level
4. Chiang Mai (CM1)	Chiang Mai Government Center	2004–2018	18.85	98.97	Sub-urban, 150 meters distance to the road, 310 meters above sea level
5. Chiang Mai (CM2)	Yupparaj Witthayalai School	2004–2018	18.80	98.99	Urban, close to a busy road, 310 meters above sea level
6. Nan (NN)	Nan Municipality Office	2009–2018	18.89	100.78	Sub-urban, 100 meters distance to the road, 2,112 meters above sea level

Table 2. National ambient air quality standards in Thailand: methods and techniques used in each air quality monitoring station for measuring ambient air pollutant concentrations.

Pollutant	Standard*				Methods/Techniques	Detection limit
	1 hour average	8 hours average	24 hours average	Annual average		
CO (ppm)	30	9	-	-	Non-dispersive infrared detection	0.05 ppm
SO_2 (ppm)	0.03	-	0.12	0.04	Pararosalinine/UV Fluorescence	1 ppb
NO_2 (ppm)	0.17	-	-	0.03	Chemiluminescence	0.5 ppb
NO_X (ppm)	-	-	-	-	Chemiluminescence	0.5 ppb
O_3 (ppm)	0.10	0.07	-	-	Chemiluminescence	0.6 ppb
PM_{10} (mg m^{-3})	-	-	0.12	0.05	Gravimetric high volume/Beta ray attenuation/Tapered Element Oscillating Microbalance (TEOM)/Dichotomous	1 µg m^{-3}

* Source: Pollution Control Department (2021).

function was used to display the trends and seasonal patterns of each air pollutant at each station. Time series plots were created for each air pollutant and station with an adequate number of knots, and their positions were fixed to smoothen the spline curve. The formula for a cubic spline function is as follows

$$s(t) = a + bt_i + \sum_{k=1}^{p} c_k (t_i - t_k)_+^3 \tag{1}$$

where $s(t)$ is the spline function, a, b, and c_k are parameters in the model, k indexes the location of each knot, i represents the observation from 1, 2, …, n, t_i denotes time in weeks, that is specified from 15 years, $t_1 < t_2 < \dots < t_p$ are specified knots and $(t_i - t_k)_+$ means the positive part of $(t_i - t_k)$ (i.e., set to zero if negative). Six knots were assigned to the data. Then, linear model was used to estimate the seasonality for the time series (Wongsai *et al.*, 2017). The model takes the following form

$$\hat{y}_i = a + b_0 t_i + b_1 c_{1i} + b_2 c_{2i} + \dots + b_k c_{ki} + e_i \tag{2}$$

where a is constant, b_0, b_2, …, b_k are the coefficients from the model, c_1, c_2, …, c_k are the coefficients derived from cubic spline function with $k = 3$ as we assigned six knots to the data, more details are explained elsewhere (Abdulmana *et al.*, 2021). Then, the air pollutant concentration data were seasonally adjusted by subtracting the fitted values from observed air pollutants in each time series observation and adding the mean of the seasonal component. The formula takes the form,

$$z_i = y_i - \hat{y}_i + \bar{y} \tag{3}$$

where, z_i is the seasonally adjusted air pollutant concentration at time i, y_i is the observed air pollution concentration, $\hat{y}_i$ is the fitted value from the linear model and $\bar{y}$ is the overall mean of observed air pollutant.

Time series models are based on stationarity with observed values having constant mean and variance over time. In this study, the autoregressive integrated moving average (ARIMA) model was used to analyze annual trends of each pollutant at each station. The parameters in this model are p, d, and q, where p is the number of autoregressive terms, d is the number of non-seasonal differences, and q is the number of lagged forecast errors in the prediction equation (Brockwell, 2010). The performance evaluations of the adopted models are carried out on the basis of correlation coefficient (r^2). In these studies, we used ARIMA (1, 0, 0), as the air pollutant concentration data were already seasonally adjusted. Therefore, differencing parameter (d = 0) was equal to zero and the parameter of moving average was also equal to zero. Therefore, an autoregressive (AR) model using cubic spline function (Eq. (4)) was used for fitting the annual trend of each pollutant at each station. The formula takes the form,

$$\hat{z}_i = \alpha + \phi z_{i-1} + e_i \tag{4}$$

where α is a constant, ϕ is autoregressive parameters, and e_i or error is white noise as the error value at time i. The filtered autocorrelation in seasonally adjusted air pollutant concentration (v_i) can be calculated using the following equation.

$$v_i = \left[s(\hat{z}_i) / s(e) \right] + \mu \hat{z}_i \tag{5}$$

where, e is the fitted residual from AR model, s is the standard deviation of each air pollutant and $\mu\hat{z}_i$ is constant term, which is the mean of the seasonally adjusted air pollutants.

A multivariate regression model incorporating the filtered autocorrelation in seasonally adjusted weekly average air pollutant concentrations was used to examine the average changes of air pollutants concentration over 15 years at six stations as follows.

$$f_{ij} = \alpha_i + \beta t_i + \varepsilon_{ij} \quad (6)$$

Here, f_{ij} is the filtered autocorrelation in seasonally adjusted air pollutant concentration at observation *i* in station *j*, α_i is the intercept, β is the regression coefficient, and ε_{ij} is the error term. After fitting the multivariate regression model, the normality assumption of residuals was evaluated for determining whether this model is appropriate. Confidence interval plots were created to illustrate the overall increase of air pollutant concentration at each station and the whole UNT area. All statistical analyses and graphical plots were performed using R statistical program version 3.6.3 (R Core Team, 2020).

3 RESULTS

The percent of missing data by each air pollutant, station and year ranged from zero to 76.9% as shown in Table 3. The means and standard deviations by pollutants, stations and years are shown in Supplementary Material. The distributions of the weekly average concentration of the transformed pollutants are shown in Fig. 2. All pollutants except SO_2 followed the normal distribution as most of data points are along the diagonal line. The percentages of missing values are also shown in the plot with the range from 10.1% for SO_2 to 18.4% for CO.

The distribution of the weekly average concentration of each air pollutant by station in the UNT is shown in the boxplots of Fig. 3. The percent of missing data for each pollutant by station is also depicted in the figure. The highest median CO, NO_2, NO_x and PM_{10} concentrations were found at CM2 station (0.77 ppm, 14.18 ppb, 22.77 ppb and 38.45 $\mu g\ m^{-3}$, respectively) while the highest median O_3 and SO_2 were found at CM1 station in Chiang Mai and at LP3 station in Lampang province (20.5 ppb and 1.11 ppb, respectively). The LP3 station in Lampang province had the lowest medians for CO and NO_2 (0.28 ppm and 2.68 ppb, respectively). On the other hand, the CM2 station in Chang Mai had the lowest median O_3 at 15.79 ppb. PM_{10} in NN station in Nan province had the lowest median at 29.87 $\mu g\ m^{-3}$.

3.1 Seasonal Patterns of Air Pollutants

Figs. 4 and 5 show seasonal patterns of weekly average air pollutant concentrations over 15 years at each station. The cubic spline was fitted to the seasonal pattern of an air pollutant and plotted against the 52 weeks of year for each station. Medians were fitted to assess the seasonal patterns of each air pollutant concentration and these are shown as blue lines. Besides, the six knots placed at week 1, 9, 19, 34, 44, 52 were selected to get smooth spline curves. The positions of the knots are shown with plus (+) symbols. The results showed that, overall, the air pollutant concentrations had seasonal patterns. The concentration gradually increased during the first week in December (cold) and peaked between March and April (summer season). Then it gradually decreased from May to September (rainy season), except for SO_2 which had no seasonal patterns in LP2, LP3 and NN stations.

3.2 Trends of Air Pollutants

Figs. 6 and 7 show plots of annual trends, or show the season-adjusted time series of weekly average air pollutant concentrations for 15 years at each station, with time (years) on x-axis and pollutant concentration on y-axis. The coefficients of autocorrelation and r-squared are shown on top of each plot. Four knots placed in 2004, 2009, 2014 and 2019 were selected for smooth spline curves, which divided the data equally into 5-year intervals. The positions of the knots are shown with plus (+) symbols. Annual trends of weekly average air pollutant concentrations for 15 years at 6 stations in the UNT revealed that overall, the air pollutant concentrations of NO_2, NO_x and PM_{10} in UNT had decreasing trends. In contrast, most CO, O_3, and SO_2 concentrations had increasing trends. Also, the trends at some stations had a fluctuating pattern, with some fluctuations increasing or decreasing with the average level of the time series. As an example, the CO concentration levels at LP1 and LP2 stations had decreasing trends from 2004 to 2008 and then the gradually increasing trends occurred from 2009 to 2013, with decreasing again from 2014 to 2018.

Table 3. Percent of missing values for each pollutant, by station and year.

Pollutant/ station	Year														
	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
CO															
LP1	5.8	0	0	0	7.7	3.8	0	0	46.2	3.8	0	0	0	0	0
LP2	0	0	0	0	0	0	3.8	3.8	0	1.9	0	0	0	11.5	0
LP3	0	0	0	3.8	1.9	0	1.9	0	0	76.9	NM	NM	NM	NM	NM
CM1	0	0	1.9	0	0	0	0	0	5.8	0	0	0	51.9	NM	NM
CM2	0	0	0	0	0	0	1.9	0	5.8	5.8	26.9	0	0	0	0
NN	NM	NM	NM	NM	NM	42.3	0	0	0	7.7	1.9	3.8	0	0	0
O_3															
LP1	0	0	0	0	7.7	3.8	0	0	69.2	3.8	0	0	0	0	0
LP2	0	0	0	1.9	7.7	0	0	3.8	0	1.9	0	0	0	0	0
LP3	0	0	0	0	1.9	0	3.8	0	1.9	48.1	0	0	0	0	0
CM1	0	0	0	0	0	0	0	0	0	1.9	0	0	3.8	0	0
CM2	0	0	0	0	0	0	0	1.9	0	0	42.3	NM	NM	NM	NM
NN	NM	NM	NM	NM	NM	42.3	0	13.5	7.7	1.9	7.7	3.8	0	0	0
NO_2															
LP1	0	0	0	1.9	7.7	3.8	0	0	46.2	3.8	0	0	0	0	0
LP2	0	0	0	0	0	0	0	32.7	0	1.9	0	36.5	1.9	0	0
LP3	0	0	1.9	5.8	0	0	1.9	0	1.9	48.1	0	3.8	0	0	0
CM1	0	0	0	0	0	0	0	0	1.9	0	3.8	1.9	5.8	0	0
CM2	0	0	0	0	0	0	0	0	0	0	3.8	0	0	0	0
NN	NM	NM	NM	NM	NM	42.3	0	0	0	3.8	1.9	11.5	0	0	0
NO_X															
LP1	0	0	0	1.9	7.7	3.8	0	0	46.2	3.8	0	0	0	0	0
LP2	0	0	0	0	0	0	0	32.7	0	1.9	0	36.5	1.9	0	0
LP3	0	0	1.9	5.8	0	0	1.9	0	1.9	48.1	0	3.8	0	0	0
CM1	0	0	0	0	0	0	0	0	1.9	0	3.8	1.9	5.8	0	0
CM2	0	0	0	0	0	0	0	0	0	0	3.8	0	0	0	0
NN	NM	NM	NM	NM	NM	42.3	0	0	0	3.8	1.9	11.5	0	0	0
SO_2															
LP1	0	0	0	0	7.7	3.8	0	0	46.2	3.8	0	0	0	0	0
LP2	0	0	0	0	0	0	0	3.8	0	1.9	0	0	0	0	0
LP3	0	1.9	0	0	0	0	0	0	0	65.4	0	0	0	0	0
CM1	0	0	0	0	0	0	0	0	0	0	0	0	3.8	0	0
CM2	0	0	0	0	0	0	0	0	13.5	0	7.7	0	0	0	0
NN	NM	NM	NM	NM	NM	42.3	0	0	3.8	13.5	7.7	0	3.8	0	0
PM_{10}															
LP1	0	0	0	1.9	7.7	3.8	0	0	53.8	3.8	0	0	0	1.9	0
LP2	0	0	0	3.8	1.9	0	0	7.7	0	1.9	0	1.9	0	0	0
LP3	1.9	9.6	0	1.9	0	0	1.9	1.9	0	40.4	0	0	0	0	0
CM1	1.9	0	0	0	0	0	0	0	0	0	0	0	51.9	NM	NM
CM2	0	0	0	0	0	0	0	0	0	0	3.8	0	0	0	0
NN	NM	ND	NM	NM	NM	42.3	0	0	0	1.9	1.9	0	0	0	0

NM is not measured.

Fig. 8 shows the increases in weekly average air pollutant concentrations with 95% confidence intervals for each station in UNT, from 2004 to 2018, using the linear model. The results show that overall trends of weekly average CO concentration levels at LP1, LP2, CM2 and NN stations had significantly increased means by 0.032 ppm (95% CI, 0.027 to 0.038 ppm), 0.073 ppm (95% CI, 0.068 to 0.078 ppm), 0.005 (95% CI, 0.001 to 0.009 ppm) and 0.026 ppm (95% CI, 0.017 to 0.036 ppm), respectively. In contrast, the stations LP3 had significantly decreased mean by –0.022 ppm (95% CI, –0.035 to –0.010 ppm) and CM1 had borderline significantly decreased mean by –0.006 ppm

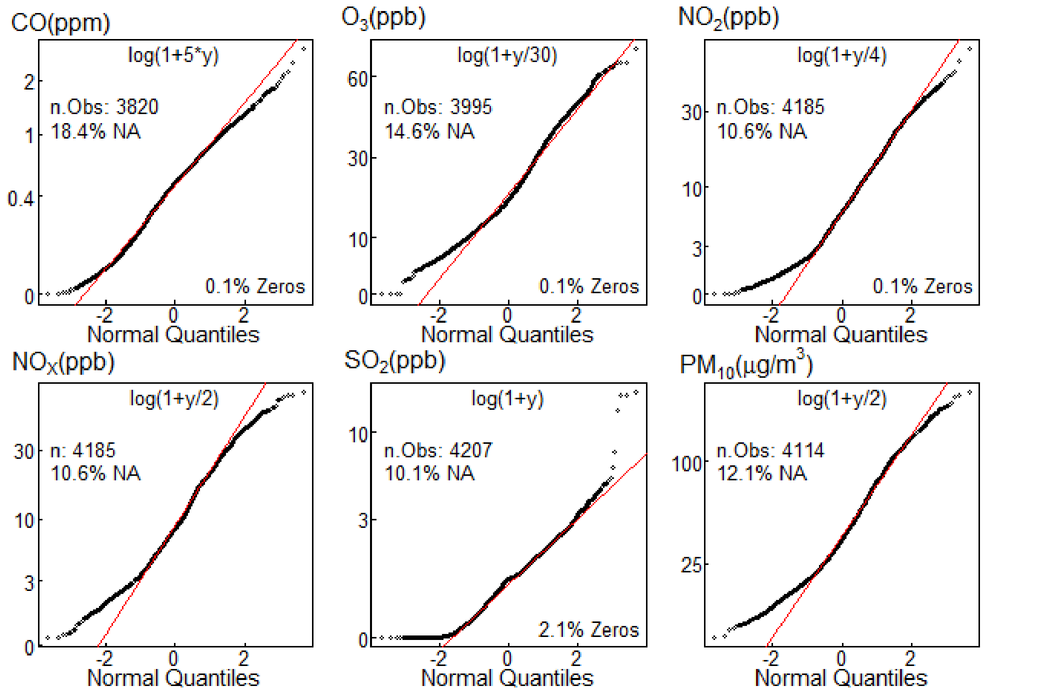

Fig. 2. Quantile-quantile (Q-Q) plots of weekly average air pollutant concentrations in UNT from 2004 to 2018.

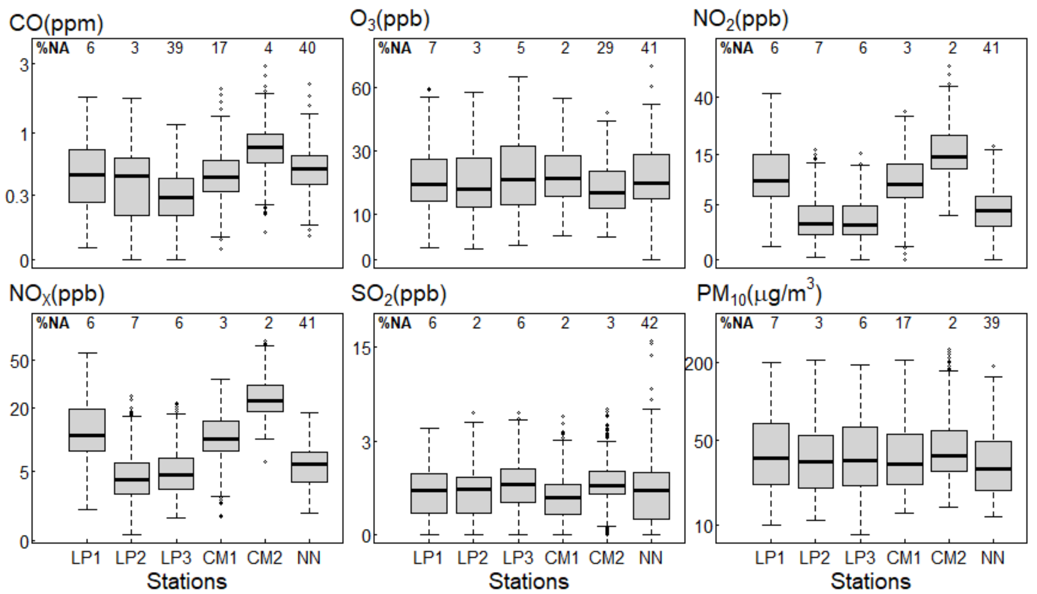

Fig. 3. Distribution of weekly average air pollutant concentrations at 6 stations in the UNT between 2004 and 2018.

(95% CI, –0.012 to 0.0001 ppm). Besides, the weekly average O_3 concentrations at LP1, LP2, LP3, CM1 and CM2 stations had significantly increased means by 0.016 ppb (95% CI, 0.014 to 0.018 ppb), 0.010 ppb (95% CI, 0.008 to 0.012 ppb), 0.022 ppb (95% CI, 0.020 to 0.025 ppb), 0.004 ppb (95% CI, 0.002 to 0.006 ppb) and 0.008 ppb (95% CI, 0.005 to 0.011 ppb). The overall trends of weekly average NO_2 concentrations at LP1 and LP2 stations had a significantly decreased means by –0.045 ppb (95% CI, –0.050 to –0.039 ppb) and –0.004 ppb (95% CI, –0.007 to –0.0004 ppb). In contrast, the stations LP3 had significantly increased mean by 0.008 ppb (95% CI, 0.004 to 0.012 ppb). Additionally, the weekly average of the NO_x concentration at LP1 and LP3 stations had significantly

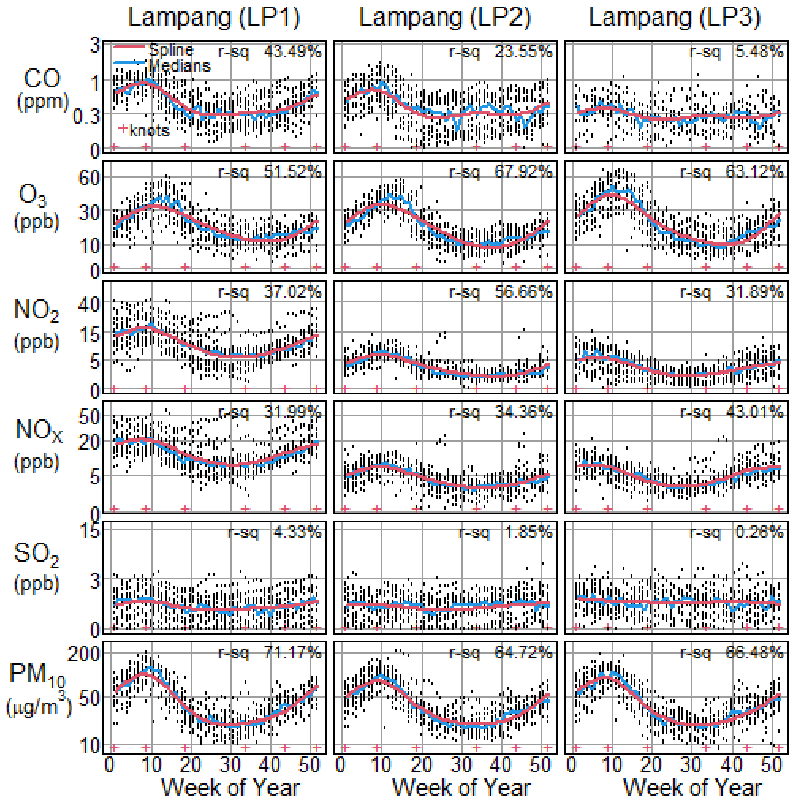

Fig. 4. Seasonal patterns of weekly average air pollutant concentrations at LP1, LP2 and LP3 stations.

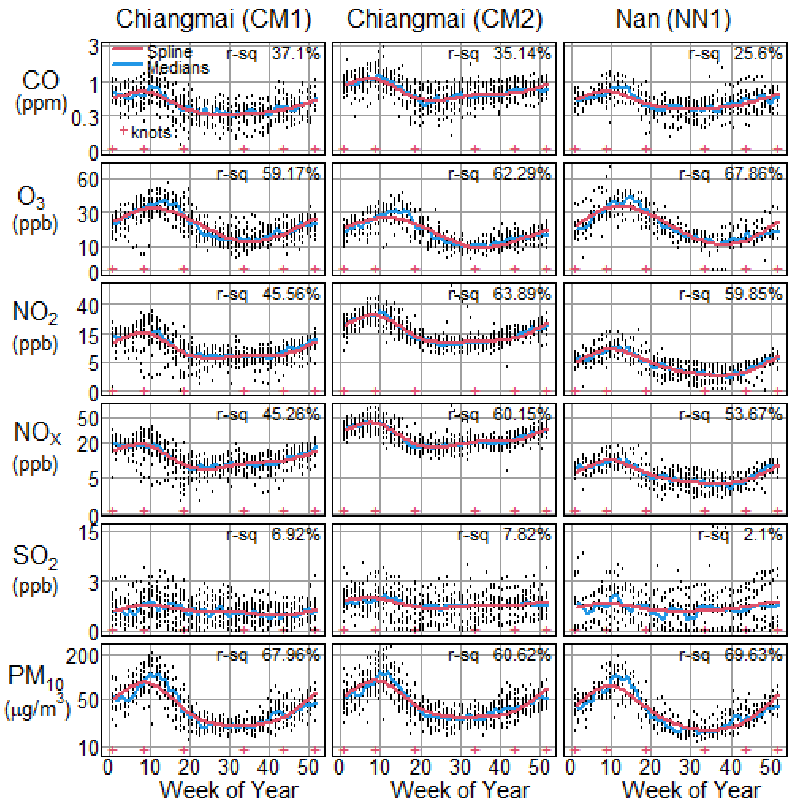

Fig. 5. Seasonal patterns of weekly average air pollutant concentrations at CM1, CM2 and NN1 stations.

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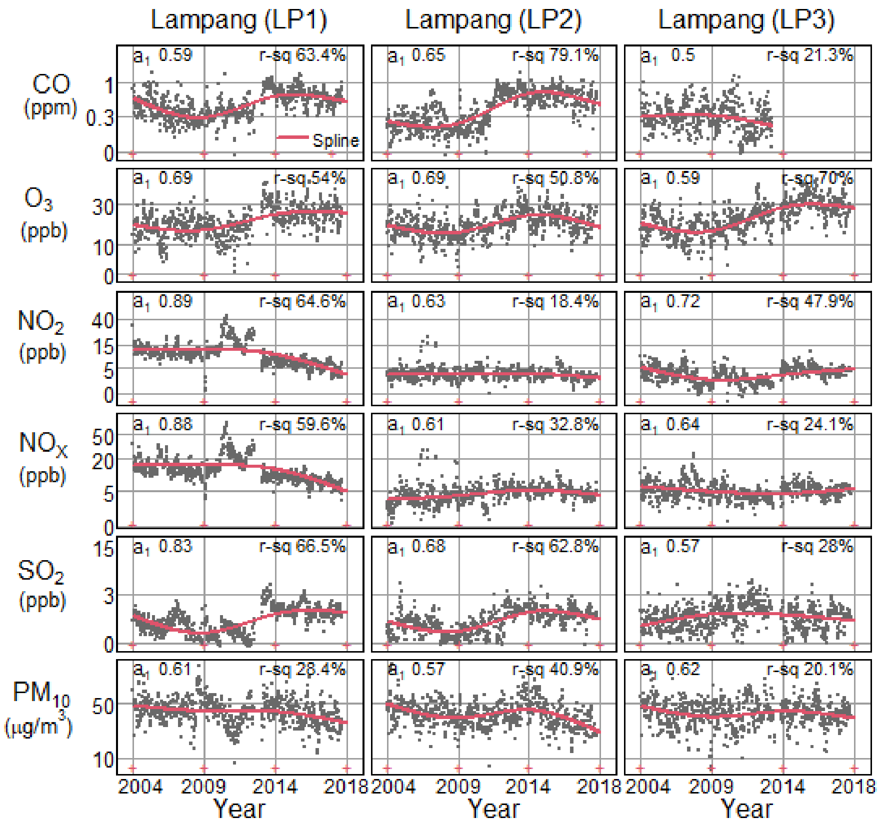

Fig. 6. Annual trends of weekly average air pollutant concentrations at LP1, LP2 and LP3 stations.

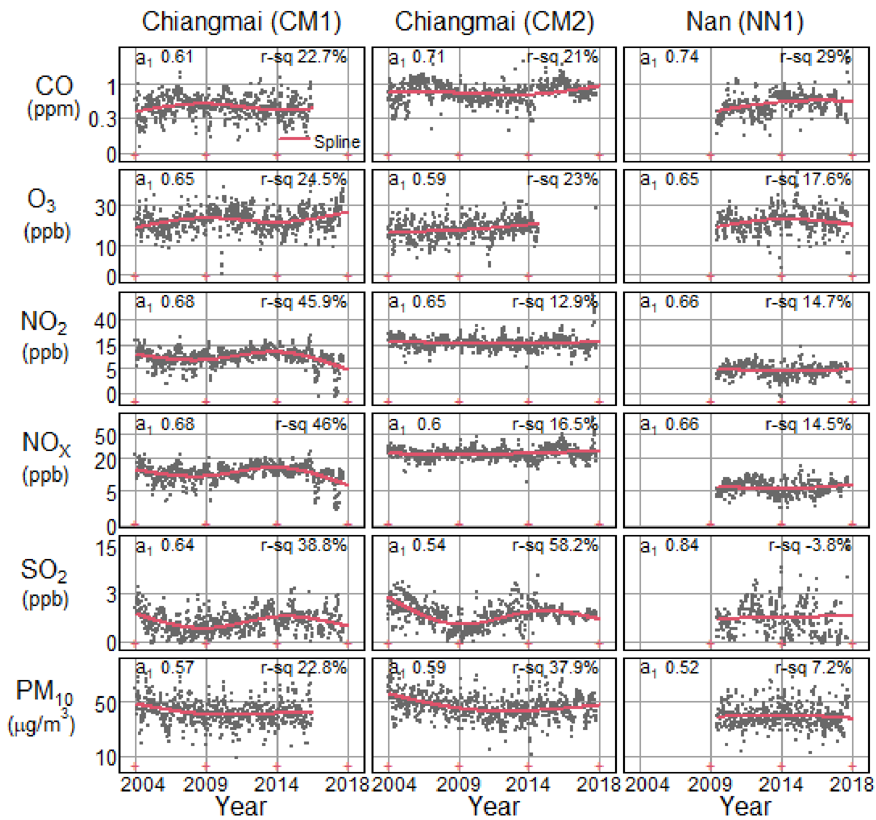

Fig. 7. Annual trends of weekly average air pollutant concentrations at CM1, CM2 and NN1 stations.

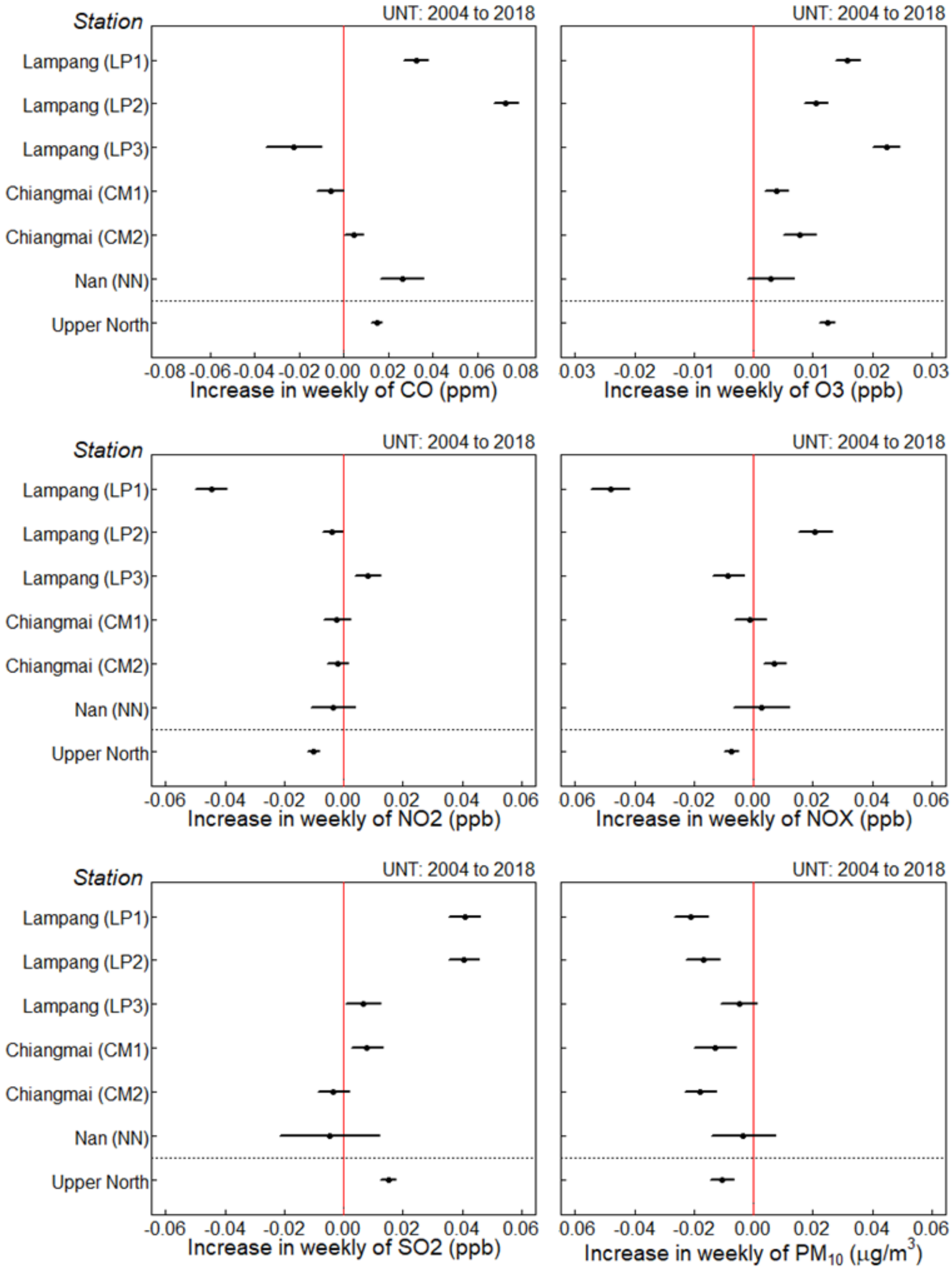

Fig. 8. Average weekly air pollutant concentrations increase from 2004 to 2018 with confidence intervals of each station in the UNT.

decreased means by –0.048 ppb (95% CI, –0.055 to –0.042 ppb) and –0.009 (95% CI, –0.014 to –0.003 ppb). However, LP2 and CM2 had significantly increased means by 0.021 ppb (95% CI, 0.015 to 0.026 ppb) and 0.007 ppb (95% CI, 0.003 to 0.011 ppb). The weekly average of SO_2 concentration at LP1, LP2, LP3 stations and CM1 station had significantly increased means by 0.041 ppb (95% CI, 0.035 to 0.046 ppb), 0.040 ppb (95% CI, 0.035 to 0.045 ppb), 0.007 ppb (95% CI, 0.001 to 0.013 ppb) and 0.008 ppb (95% CI, 0.002 to 0.013 ppb), respectively. The average weekly PM_{10} concentration level at LP1, LP2, CM1 and CM2 stations had significantly decreased means by –0.021 µg m^{-3} (95% CI, –0.026 to –0.015 µg m^{-3}), –0.017 µg m^{-3} (95% CI, –0.023 to –0.011 µg m^{-3}), –0.013 µg m^{-3} (95% CI, –0.020 to –0.006 µg m^{-3}) and –0.018 µg m^{-3} (95% CI, –0.023 to –0.012 µg m^{-3}), respectively. For the whole of UNT region, CO, O_3 and SO_2 had significantly increased, on average by 0.015 ppm (95% CI, 0.012 to 0.017 ppm), 0.012 ppb (95% CI, 0.011 to0.014 ppb) and 0.015 ppb (95% CI, 0.013 to 0.018 ppb), respectively. On the other hand, NO_2, NO_x and PM_{10}

had significantly decreased on average by –0.010 ppb (95% CI, –0.012 to –0.008 ppb), –0.008 ppb (95% CI, –0.010 to 0.005 ppb) and –0.011 µg m^{-3} (95% CI, –0.014 to –0.007 µg m^{-3}), respectively.

4 DISCUSSION

This study applied robust statistical techniques to investigate seasonal patterns and trends of air pollutant concentrations in the UNT during the years 2004 to 2018. Seasonal patterns of CO, O_3, NO_2, NO_x, SO_2 and PM_{10}, were found, with peak concentrations observed in March. CO, O_3 and SO_2 had significantly increasing trends whereas NO_2, NO_x and PM_{10} had significantly decreasing trends over time since 2004.

In our study, all air pollutants peaked in March. This finding is consistent with studies conducted by Lalitaporn (2018) and Lalitaporn and Boonmee (2019), which reported that seasonal emissions in UNT caused high levels of CO and NO_2 during the dry season, from anthropogenic and biomass burning activities, while the levels were lower during the rainy season. Matsuda *et al.* (2006) also reported that SO_2 and O_3 concentrations increased in the dry (January–April) season and decreased in the wet (May–August) season. Such trends are caused by the dry deposition above a forest in a tropical savanna climate in Mae Moh, Lampang Province. In addition, the O_3 formation caused by secondary reactions between Volatile Organic Compounds (VOCs) and NO_x, catalyzed by ultraviolet radiation, were also higher in the summer than in the winter. NO_2 and PM_{10} in northern Thailand were higher during the dry season between January and April (Wiriya *et al.*, 2013; Kliengchuay *et al.*, 2018; Outapa and Ivanovitch, 2019; Lalitaporn and Boonmee, 2019). An increase in PM_{10} is common in summers with highest level in March due to low precipitation and open burning in agricultural areas during harvesting season (Janta, 2020).

The overall CO concentration was found to have significantly increased. This finding supports a study conducted by Lalitaporn (2018), who found a significant increase in CO concentrations in the UNT from 2001–2015. This might be due to the intensive biomass burning and gasoline-powered vehicles.

A significant increase in the overall concentration of O_3 was also found in our study. This result is consistent with findings of Sonkaew and Macatangay (2015), which showed that the O_3 in the troposphere of northern regions in Thailand had a slightly increasing 8-year trend from 2005 to 2012, especially during the forest-clearing and agricultural burning season that usually occurs during February to April. NO_2 and NO_x in our study had been significantly declining in some station over time since 2004. Our finding in contrast to a study conducted by Lalitaporn and Boonmee (2019), which found a slight increase over 6 years in NO_2 concentration levels from satellite and ground measurements over northern Thailand for 2010 to 2016. NO_x and NO_2 concentrations both have the same sources from biomass combustion and fossil fuels (Dell'Antonia *et al.*, 2012). In addition, the O_3 formation is caused by secondary reactions between Volatile Organic Compounds (VOCs) and NO_x, catalyzed by ultraviolet radiation (Laothawornkitkul *et al.*, 2009; Fakkaew *et al.*, 2019). Han *et al.* (2011) reported an inverse relationship between O_3 and NO_x/NO_2 whereas NO_x and NO_2 have a positive linear relationship.

Significant increase overall in SO_2 concentration was observed in our study, especially at LP1 and LP2 stations, consistent with a study conducted by Thepanondh *et al.* (2002). This might be due to the emissions of SO_2 and sulphate from coal-fired power plants operated in Lampang province, Thailand (Thepanondh *et al.*, 2002).

Moreover, this study found that PM_{10} had significantly decreased over 15 years. The result is in line with the finding by Aziz *et al.* (2016) of decreasing trend for PM_{10} in Bangkok. The decrease of PM_{10} in UNT might due to the implementation of the zero-burning policy on open burning, for about a 3-month-long period (mid-February to mid-May) in the dry season of northern Thailand during 2017 to 2018 (Yabueng *et al.*, 2020). This might have reduced air pollutants, compared to 15 years prior.

The statistically significant increases in CO, O_3, and SO_2 in the past 15 years by 0.015 ppm, 0.012 ppb, and 0.015 ppb in this study are relatively small when compared to health-based air quality standards in Thailand (Pollution Control Department, 2021). On the other hand, the significant decreases in NO_2, NO_x, and PM_{10} by –0.010 ppb, –0.008 ppb, and –0.011 mg m^{-3} are also relatively small. These small changes could be due to very small changes in emissions or as a consequence of weather conditions that were not taken into account in this study. A limitation of this study is that climatological factors such as wind speed, wind direction, mixing height, rainfall, humidity, and temperature were not considered.

5 CONCLUSIONS

This study concludes that the seasonal patterns of most pollutant concentrations in the UNT were similar with the highest levels during the summer. The overall trends in most stations of CO, O_3 and SO_2 concentrations were steadily increasing, whereas PM_{10} concentration had been declining over the 15 years assessed in this study. Furthermore, NO_2 and NO_x concentrations had very slightly decreased. Despite regulations to control open crop residue burning, the air pollution in the UNT during the summer remains high every year. This indicates that the current burning control policies are not adequate, and government should implement more effective burning control policies to reduce the air pollution levels.

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SUPPLEMENTARY MATERIAL

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