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

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Research Article

Keywords: Air pollutants, air quality, patterns and trends, particulate matter, sulfur dioxide, nitrogen oxide, upper Northern Thailand

Posted Date: September 21st, 2021

DOI: https://doi.org/10.21203/rs.3.rs-914607/v1

Version of Record: A version of this preprint was published at Aerosol and Air Quality Research on January 1st, 2022. See the published version at https://doi.org/10.4209/aaqr.210318.

Title page

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Abstract

The objective of this study was to investigate the seasonal patterns and trends of air pollutants in upper northern Thailand (UNT) from 2004 to 2018. The hourly air pollutant concentration data 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 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 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 between March and April. In the past 15 years, the levels of overall CO, O_3, and SO_2 in the UNT had significantly increased, on average of 0.032 ppm, 0.012 ppb, and 0.017 ppb, respectively. In contrast, NO_2, NO_X, and PM_{10} had significantly decreased on average of 0.012 ppb, 0.011 ppb, and 0.016 mg/m^3, respectively. In conclusion, it should be of concern for such activities that related to air pollutants variation accordingly.

Keywords: Air pollutants, air quality, patterns and trends, particulate matter, sulfur dioxide, nitrogen oxide, upper Northern Thailand

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). Carbon monoxide (CO) is a gas emitted by incomplete combustion of a fuel, mainly from transportation and industrial production activities (Zhong et al. 2017). NO_X and NO_2 are pollutants mainly attributed to vehicles, industrial combustors, and electric utility power plants (Liard et al. 1999; Gilbert et al. 2003). SO_2 is generally emitted from burning of a fossil fuel, especially coal (Zhang et al. 2017). PM10 is mainly from the open burning of agricultural biomass and from forest fires (Tsai et al. 2013). These events and activities produce different kinds of air pollutants and there are changes in the air pollutant levels over time.

Most air pollutants vary by season. In countries near the Arctic, the concentrations of $PM_{2.5}$, PM_{10}, NO_2, O_3, SO_2 and CO peak in the winter, whereas O_3 is higher in the summer; in China, Romania, and Poland among others (Chen et al. 2015; Bodor et al. 2020; Cichowicz et al. 2017). However, in tropical countries such as Malaysia and Thailand, CO, NO_2, SO_2, PM_{10}, O_3 are comparatively high from January to March (Mohtar et al. 2018). A decreasing trend of air pollution is commonly documented for the developed countries. For example, decreasing trends of PM_{10} and NO_2 in Europe in 1998-2008 were found (Colette et al. 2011), while increasing trends of air pollutants such as CO, NO_2 and SO_2 were reported in most developing countries (Jaiswal et al. 2018). Thailand is one of the Southeast Asian countries currently facing air pollution problems. Increasing trends of CO, O_3 and NO_2 have been reported (Aziz et al. 2016; Lalitaporn 2018). Seasonal patterns have also been found for PM_{10}, CO, SO_2, NO_2 and O_3 that are 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). The northern region of Thailand faces major smog problems yearly due to crop burns 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 et al. 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 had formed the policies and measures to prevent and tackle northern haze and forest fires. The reducing burning in agricultural areas, especially during dust accumulation over 7-14 days or 1-2 months, was dependent on each provincial policy measure, which adheres to operate as guidelines under 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 (Thai 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.

Materials and method

Data source

The air quality data collected from 2004 to 2018 were obtained from the Pollution Control Department, Ministry of Natural Resources and Environment. 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.

[Table 1 is about here.]

[Fig. 1 is about here.]

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

[Table 2 is about here.]

The outcomes of this study are 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 adding one with diving or multiplying constant value for all air quality pollutants.

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 52 weeks of 15 years by station. The trends of an air pollutant were shown by plotting the average weekly air pollutant concentrations against 15 years by station. The spline 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

$$\hat{y}_i = a + bt_i + \Sigma_{k=1}^{p} c_k (t_i - t_k)_+^3 \quad (1)$$

where $\hat{y}_i$ is the spline function, a, b and c_k are parameters in the model, k is the location of knots, t_i denotes time in weeks, that is specified from 15 years and $t_1 < t_2 < \ldots < t_p$ are specified knots and $(t_i - t_k)_+$ means the positive part of $(t_i - t_k)$ (i.e. set to zero if negative).

Then, the air pollutant concentration data were seasonally adjusted by subtracting the fitted values from observed air pollutants. The formula took the form,

$$z_i = y_i - \hat{y}_i + \bar{y} \quad (2)$$

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

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). The formula for an ARIMA model ($\Delta_d g_i$) is as follows

$$\Delta_d g_i = \delta + \emptyset \Delta_d z_{i-1} + \emptyset \Delta_d z_{i-2} + \ldots + \emptyset \Delta_d z_{i-p} + \varepsilon_i - \theta_1 \varepsilon_{i-1} - \ldots - \theta_q \varepsilon_{i-p} \quad (3)$$

where d is the number of differences for stationarity, p is autoregressive order, q is moving average order, δ is a constant, Δ_d is the differencing operator of order d, $\emptyset, .., \emptyset_p$ are autoregressive parameters, $\emptyset, .., \emptyset_q$ moving average parameters, and ε_i or error is white noise as the error value at time *i*. In these studies, we use 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 (Equation 4) was used for fitting the annual trend of each pollutant at each station.

$$g_i = \delta + \emptyset_1 z_{i-1} + \Sigma_{k=1}^{p} c_k (t_i - t_k)_+^3 + \varepsilon_i \quad (4)$$

The filtered autocorrelation in seasonally adjusted air pollutant concentration (v_i) can be calculated using the following equation. Hence,

$$v_i = [s(z_i)/s(e) \times e] + \mu z_i \quad (5)$$

where, e is the fitted residual from AR model, s is the standard deviation of each air pollutant and μz_i is constant term, which is the mean of the seasonally adjusted air pollutants.

A linear 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.

$$f_i = \alpha + \beta_i v_i + \varepsilon_i \quad (6)$$

Here, f_i is the average increase of air pollution concentration, α is the intercept, β is the regression coefficient, v_i is the filtered autocorrelation in seasonally adjusted air pollutant concentration at observation i, and ε_i is the error term. After fitting the linear 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. All statistical analyses and graphical plots were done using the R statistical program (R Core Team 2020).

Results

The weekly average concentration of CO ranged between 0 and 2.92 ppm, with an average of 0.54 ppm. There were 18.40% missing values and 0.10% zero values. For O_3, NO_2, NO_X, SO_2 and PM_{10}, the weekly average concentrations were in the ranges 0 to 73.55 ppb, 0 to 66.12 ppb, 0 to 70.29 ppb, 0 to 16.60 ppb and 8.31 to 248.80 μg/m^3, respectively. Figure 2 presents the distributions of the weekly average concentration of each pollutant (CO, O_3, NO_2, NO_X, SO_2 and PM_{10}) after applying logarithm transformation plus one and multiplying or dividing pollutants by different constant values.

[Fig. 2 is about here.]

The median values of the weekly average concentrations of CO, NO_2 and NO_X were highest in CM2 station. The median values of these pollutants were 0.77 ppm, 14.18 ppb and 22.77 ppb, respectively. The Lampang station, LP3 had the lowest medians for CO, NO_2 and NO_X (0.28 ppm, 2.68 ppb and 4.59 ppb, respectively). On the other hand, the O_3 in CM2 station had the lowest median of 15.79 ppb, while the median concentration of SO_2 in CM2 station was the highest (1.05 ppb). In addition, the weekly average concentration of PM_{10} in CM2 station had the highest median of 38.45 μg/m^3 while at the NN1 station it had the lowest median of 29.87 μg/m^3, as shown in Fig. 3. Overall, the highest concentrations of CO, NO_2, NO_X, SO_2 and PM_{10} were found at a station in Chiang Mai province (CM2).

[Fig.3 is about here.]

Seasonal patterns of air pollutants

Fig. 4 and 5 show seasonal patterns of weekly average air pollutant concentrations over 15 years at each station. Week of year is on the x-axis while air pollutant concentration at each station is on y-axis. The cubic spline was fitted to the seasonal pattern of an air pollutant and plotted against the 52 weeks of year for each station. The cubic splines are shown as red lines. Medians were fitted to assess the seasonal patterns of each air pollutant concentration and these are shown as blue lines. Besides, six knots placed at 1, 9, 19, 34, 44, 52 were selected to fitted to smoothen the spline curve. 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 NN1 stations.

[Fig.4 is about here.]

[Fig.5 is about here.]

Trends of air pollutants

Fig. 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 red lines represent the trends from the cubic spline functions. 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 to smoothen the spline curve, 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 concentration of NO_2, NO_X and PM_{10} in UNT had a decreasing trend. In contrast, most CO, O_3, and SO_2 concentrations had an increasing trend. Also, 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 level at LP1 and LP2 stations had decreasing trend from 2004 to 2008 and then the gradually increasing trend occurred from 2009 to 2013, with decrease again from 2014 to 2018.

[Fig.7 is about here.]

[Fig.8 is about here.]

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 and NN1 stations had a significantly increased means of 0.009 ppm (95% CI, 0.005 to 0.012 ppm), 0.010 ppm (95% CI, 0.007 to 0.014 ppm) and 0.008 ppm (95% CI, 0.002 to 0.014 ppm), respectively. While the station in LP3 had a significantly decreased mean of -0.012 ppm (95% CI, -0.022 to -0.001 ppm). Besides, the weekly average O_3 concentrations had a significantly increased mean at all stations, except at NN1 station had a very slightly decreased trend. The overall trends of weekly average NO_2 concentrations at LP1 station had a significantly decreased mean of -0.004 ppb (95% CI, -0.006 to -0.001 ppb). Furthermore, at LP2, CM1, CM2 and NN1 stations the means very slightly decreased trends. Additionally, the weekly average of the NO_X concentration at LP1 station had significantly decreased mean of -0.005 ppb (95% CI, -0.008 to -0.002 ppb). However, LP2 had significantly increased mean of 0.008 ppb (95% CI, 0.003 to 0.012 ppb). Besides, the weekly average of SO_2 concentration at stations in Lampang LP1 and LP2 had significantly increased means of 0.004 ppb (95% CI, 0.002 to 0.007 ppb) and 0.008 ppb (95% CI, 0.005 to 0.011 ppb), respectively. Furthermore, at LP3, CM1 and NN1 stations the means very slightly increased trends. Moreover, the average weekly PM_{10} concentration level had significantly decreased in almost all stations, except at LP3 and NN1 stations had a very slightly decreased trend.

[Fig.8 is about here.]

Discussions

This study applied robust statistical techniques to investigate seasonal patterns and trends of air pollutant concentrations in the UNT during the year 2004 to 2018. Seasonal patterns of CO, O_3, NO_2, NO_X and PM_{10}, SO_2 in LP1, LP2 and NN1 stations were found, with peak concentrations observed between March and April. Most the CO, O_3 and SO_2 had been significantly increasing in some stations whereas most the NO_2, NO_X and PM_{10} had been significantly decreasing in some stations over time since 2004.

In our study, most air pollutants peaked in March to April. This finding is consistent with a study conducted by Lalitaporn (2018) which reported that seasonal emissions in UNT caused high levels of CO during dry season from anthropogenic and biomass burning activities, while the levels were lower during rainy season. Matsuda et al. (2006) also reported that SO_2 and O_3 concentrations increased in the dry (Jan-Apr) season and decreased in the wet (May-Aug) 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 VOCs and NO_X, catalyzed by ultraviolet were also higher in summer compare to winter. Therefore, a higher concentration of NO_X resulted in a higher concentration of O_3 (Fakkaew et al. 2019). 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 due to low precipitation and open burning in agricultural areas during harvesting season.

CO concentration was found to have significantly increased overall in LP1, LP2 and NN1 stations. This finding supports a study conducted by Lalitaporn (2018), who found a significant increase of CO concentration in the upper part of Thailand 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. In addition, sunlight splits NO_2 into NO and an oxygen atom (O). A single oxygen atom then combines with an oxygen molecule to produce O_3 (Finlayson-Pitts and Pitts 1999). NO_2 and NO_x in our study had been significantly declining in some station over time since 2004. Our finding is in line with a study conducted by Lalitaporn and Boonmee (2019), which found a slight increase over 6 years of 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). The significant increase in overall SO_2 concentration was observed in our study especially at LP1 and LP2 stations, consistently 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 supports the finding of Yabueng (2020) who found that 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, has an impact on the patterns of ambient air pollutants. This might lead to the reduction of air pollutants, compared to 15 years ago.

Future studies should focus on the effects of climate factors such as wind speed, wind direction, mixing height, rainfalls, humidity and temperature on the air pollutant concentrations.

5. Conclusion

This study concludes that the seasonal patterns of most pollutant concentrations in the UNT had similar seasonal patterns with highest levels during summer (Mar-Apr). The overall trends in most stations of CO, O_3 and SO_2 concentrations has been steadily increasing, whereas PM_{10}

concentration had been declining over 15 years of the study. Furthermore, NO_2 and NO_X concentrations had been very slightly decreased trends. Despite regulations to controls open crop burning, the air pollution in the UNT during March to April remains high every year. These indicated that the current burning control policies were not adequate, governments should implement more effective burning control policies to reduce the air pollution levels.

Acknowledgements

This research was supported by National Science, Research and Innovation Fund (NSRF) and Prince of Songkla University (Grant No SAT64050885), the Air Pollution and Health Effect Research Center, Faculty of Engineering and the Graduate School, Prince of Songkla University. This research is also partially supported by the Centre of Excellence in Mathematics, Commission on Higher Education (CHE), Ministry of Education, Ratchathewi, Bangkok, 10400 Thailand. The authors extend our profound appreciation to the Pollution Control Department, Ministry of Natural Resources and Environment of Thailand, for providing the data for this study. We are gratitude to Emeritus Prof. Don McNeil for his comments and suggestions throughout this research.

Author contributions

Sarawut Sukkhum: investigation, methodology, validation, data curation, formal analysis, writing – original draft, visualization

Apiradee Lim: conceptualization, project administration, funding acquisition, supervision, writing – review & editing

Rattikan Saelim: supervision, writing – review & editing

Thammasin Ingviya: supervision, writing – review & editing

Competing interests

The authors declare that they have no competing interests.

Data availability

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Ethics declarations

Not applicable

Consent for publication

Not applicable

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Figures

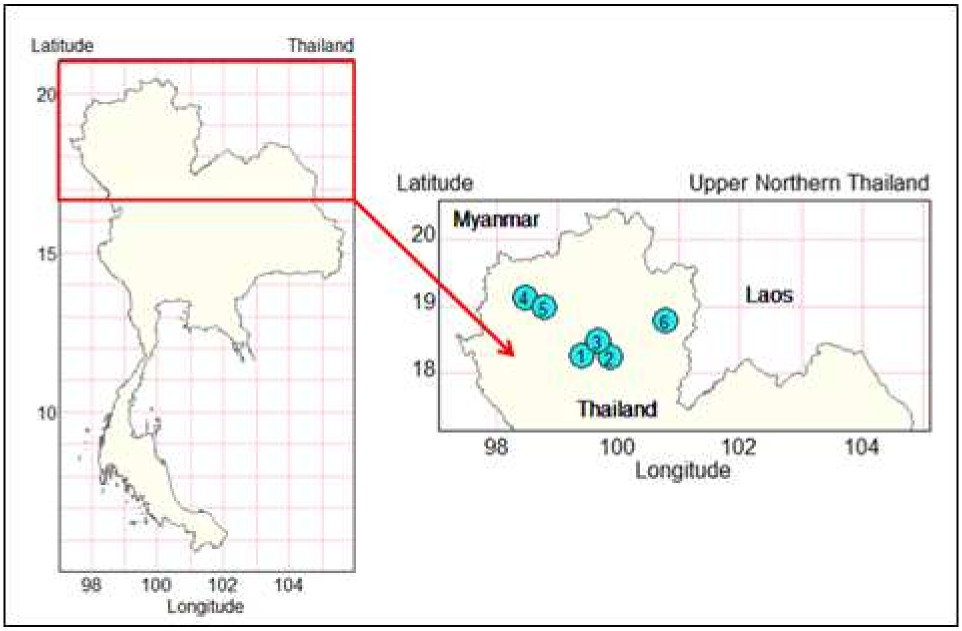

Figure 1

Map of the study area in the UNT

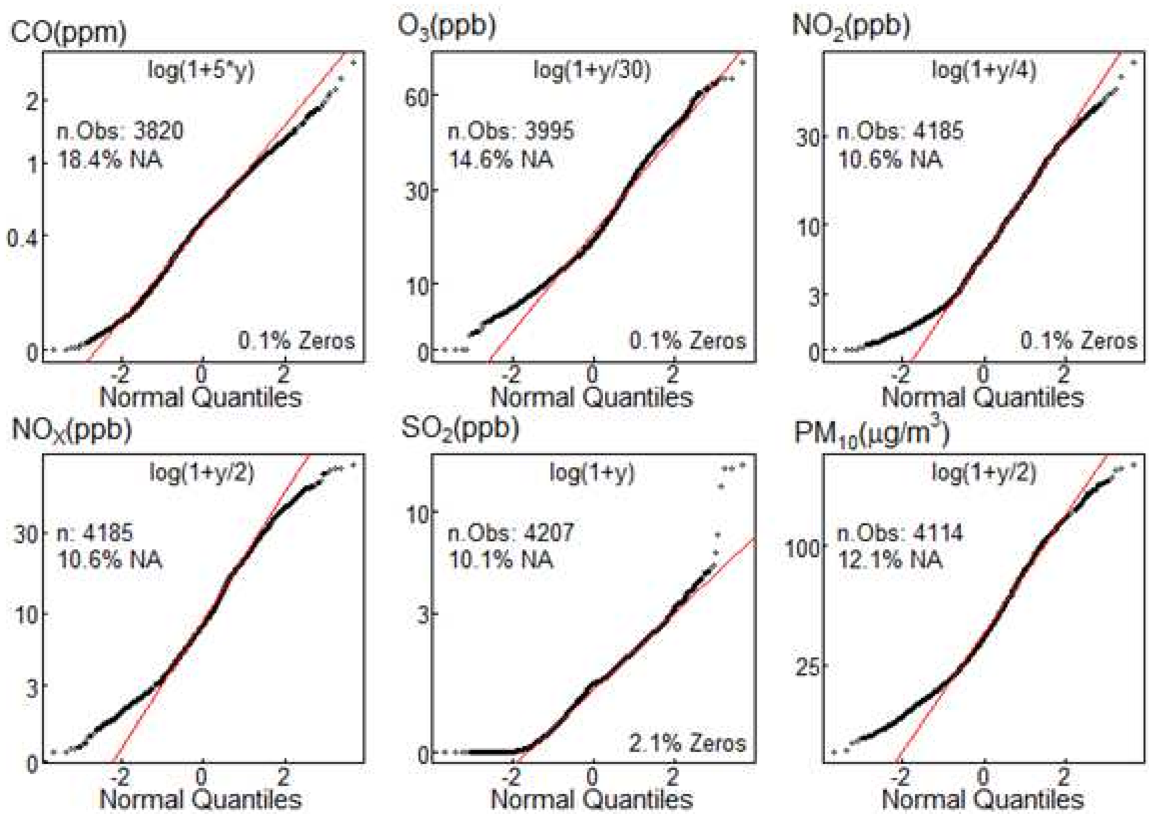

Figure 2

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

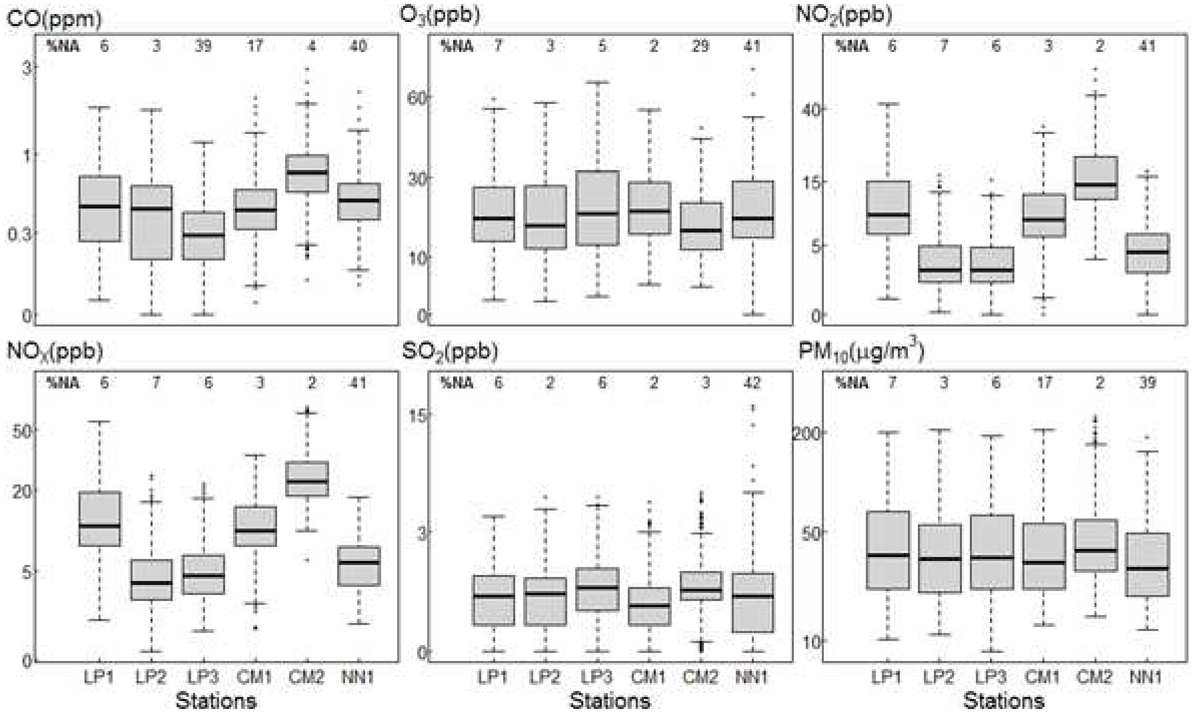

Figure 3

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

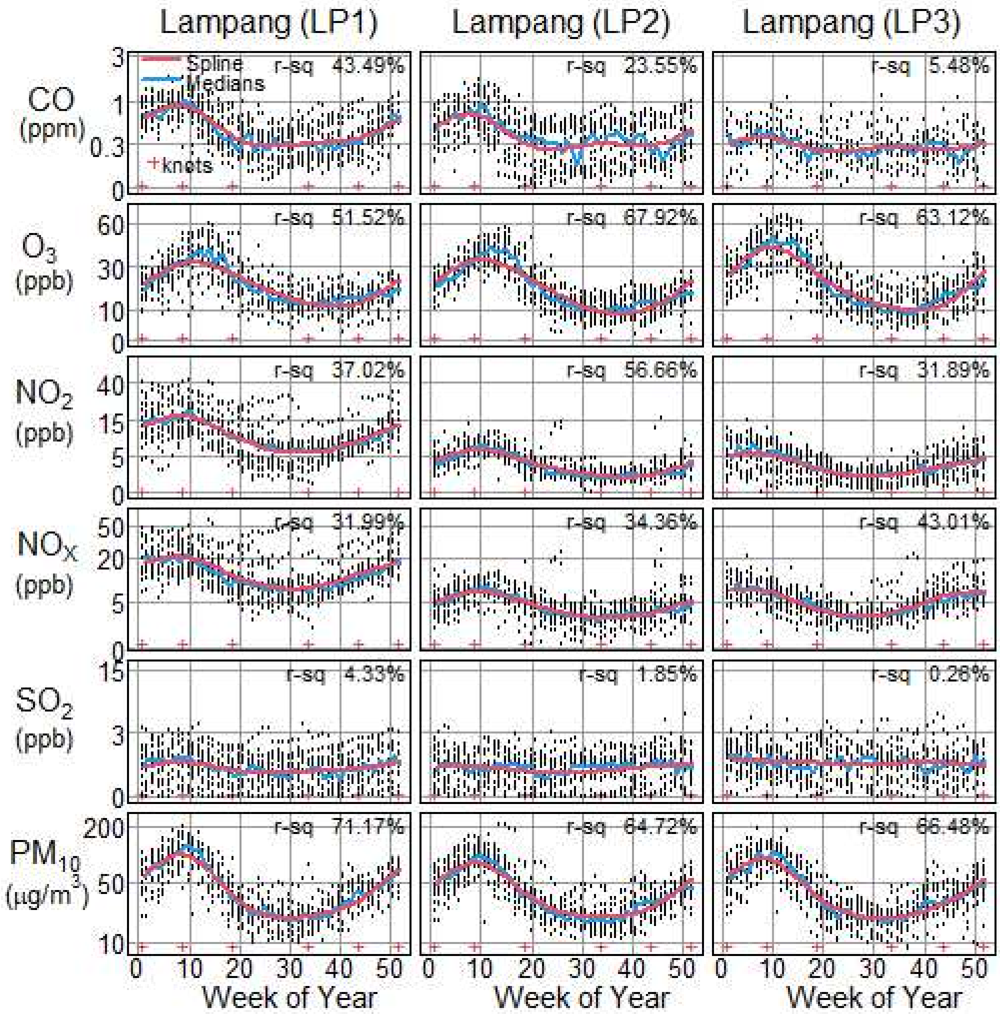

Figure 4

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

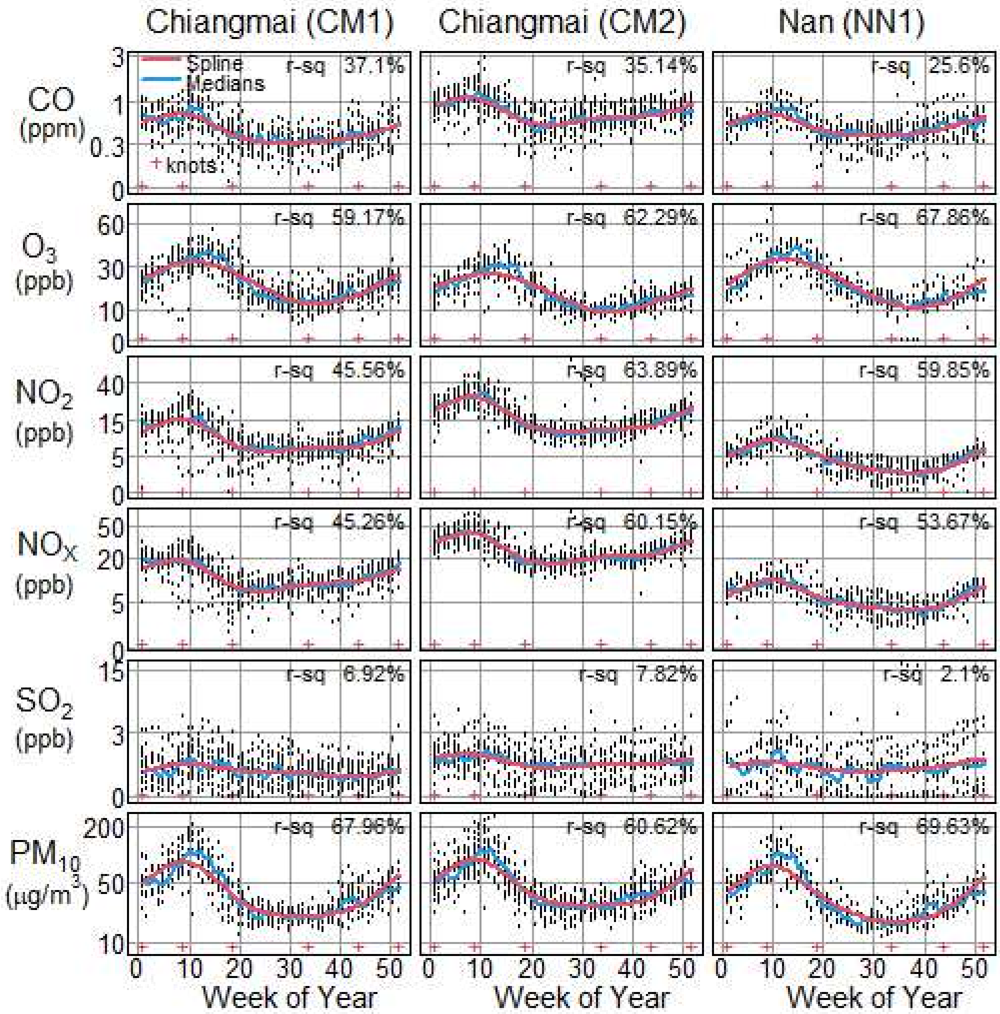

Figure 5

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

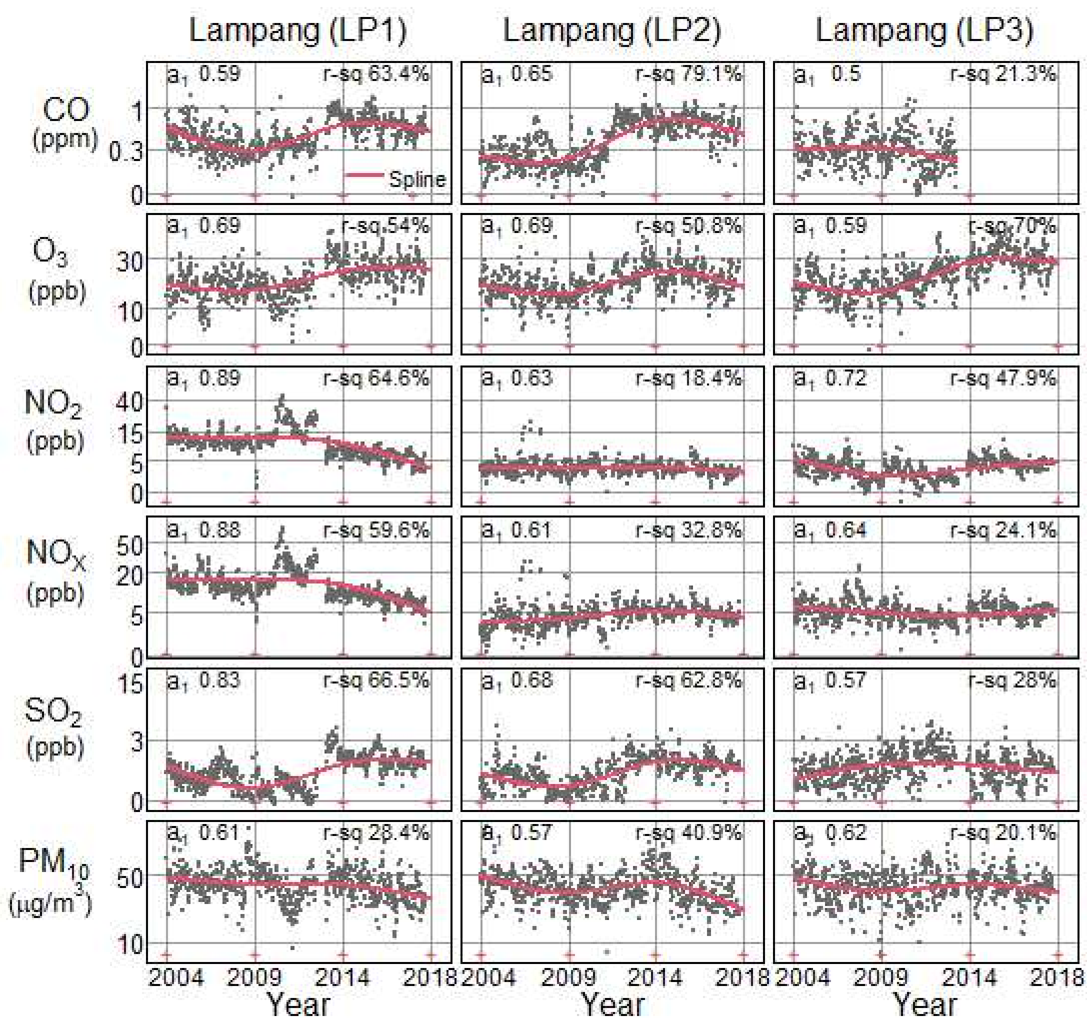

Figure 6

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

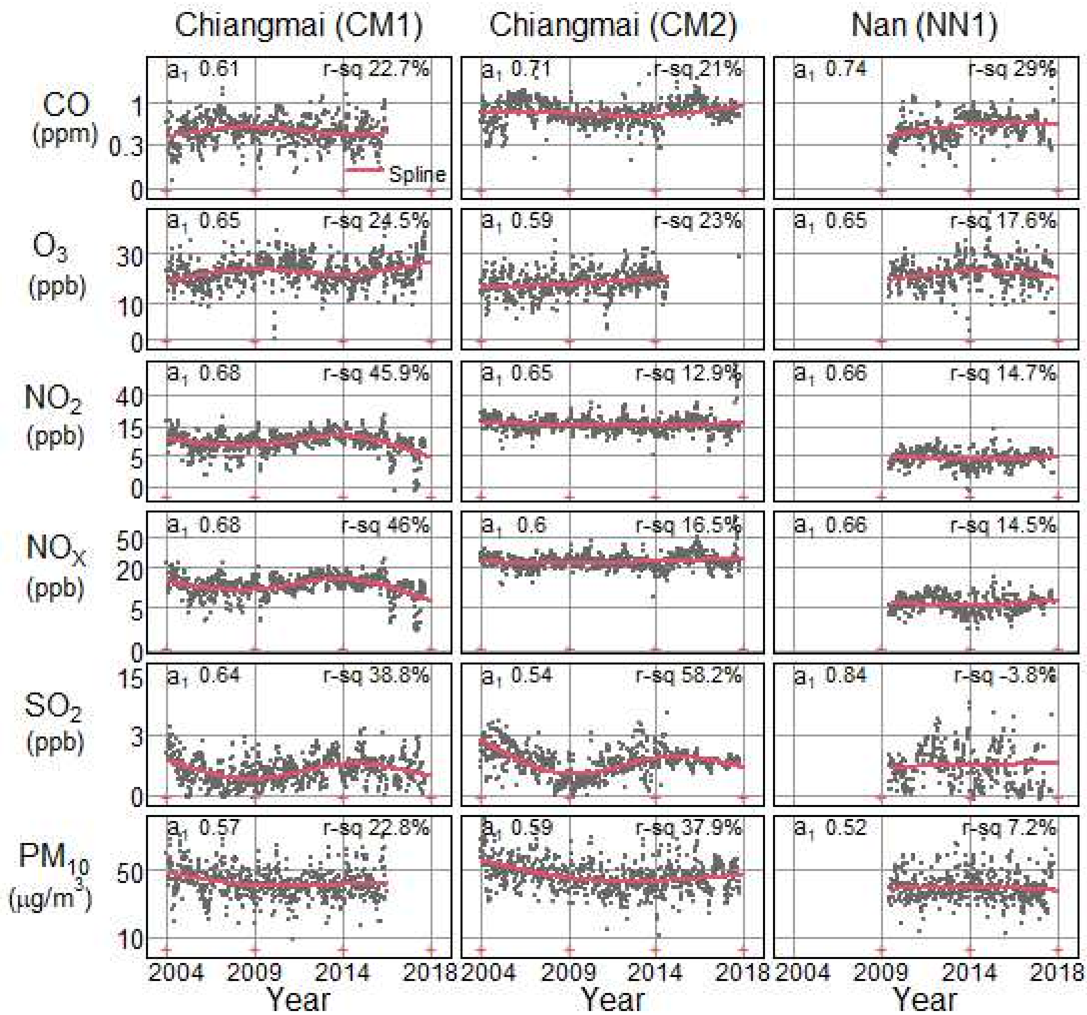

Figure 7

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

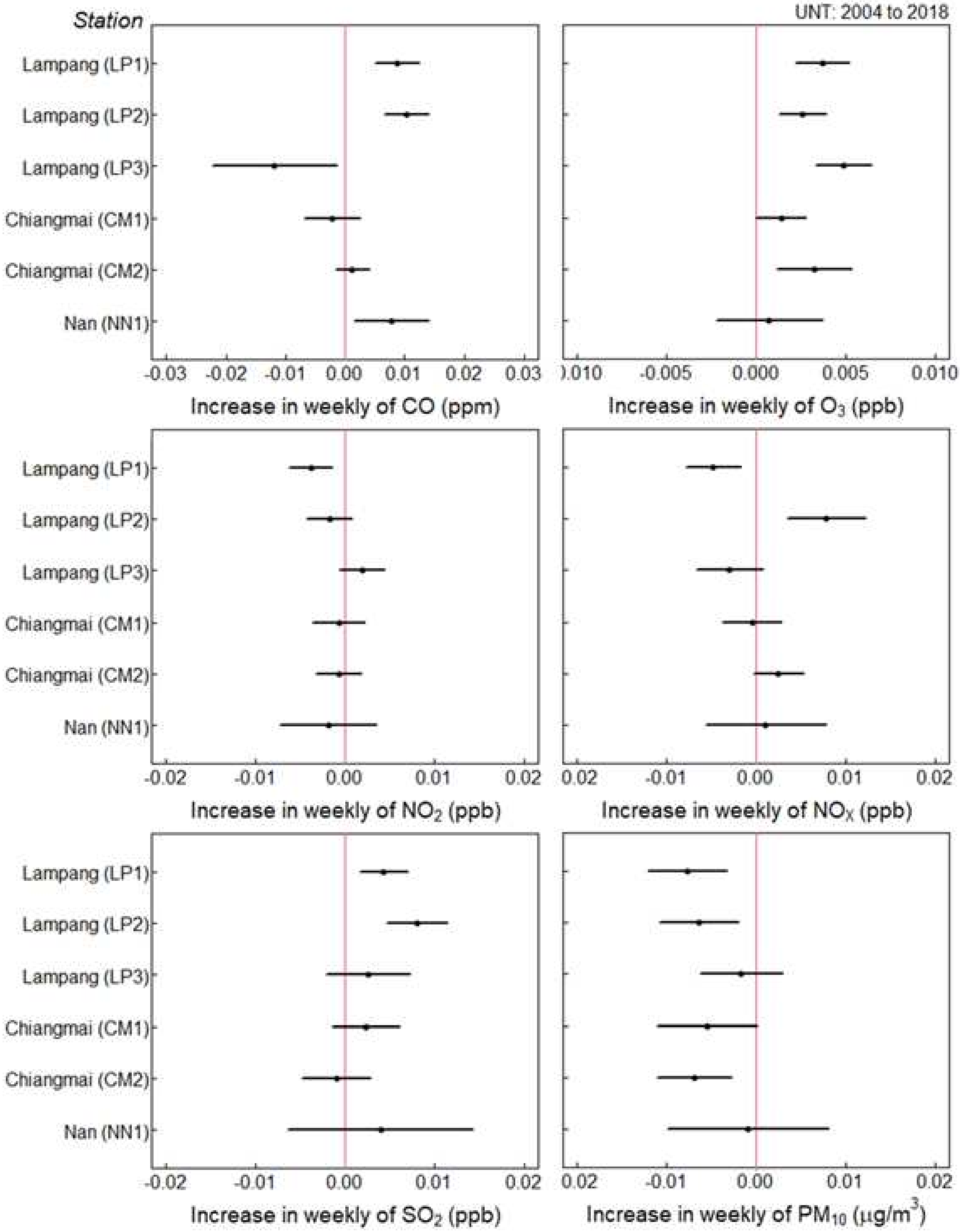

Figure 8

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