

INVESTIGATING CONTRIBUTIONS OF GASES, METEOROLOGICAL PARAMETERS, AND AEROSOLS TOWARDS TROPOSPHERIC OZONE VARIABILITIES OVER MEGACITY LAHORE (PAKISTAN)

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Abstract. We investigated contributions of some tropospheric gases, meteorological parameters, and aerosol burden measured as aerosol optical depth (AOD) towards Tropospheric Ozone (TropoO₃) variabilities over Lahore by developing a stepwise multiple linear regression model. The tropospheric gases include; Methane (CH₄), Carbon Monoxide (CO), Carbon Dioxide (CO₂), and Sulphur Dioxide (SO₂), whereas the meteorological parameters include; cloud fraction (CLF), outgoing long-wave radiation (OLR), relative humidity (RH), temperature (Temp), and surface wind-speed (WS). Data products of these parameters have been retrieved from various satellite sensors during October 2004 – September 2015. Time series distributions of both observed and modeled TropoO₃ concentrations show similar increasing trends except for few months. Moreover, monthly-means of these observed and modeled TropoO₃ concentrations indicate similar decreasing trends with maximum during spring and summer (monsoon) seasons. Based upon weighted contributions of standardized regression coefficients, modeled TropoO₃ concentration was found dependent upon OLR[0.458], WS[0.262], CLF[0.256] TropoSO₂[-0.097], TropoCO[-0.231], and RH[-0.320] out of all parameters used in this study. Modeled TropoO₃ concentrations were validated against an independent source of TropoO₃ concentrations retrieved from original subtracted product of OMI-MLS over Lahore and the results were found in good agreement. The study is unique and would be helpful for further research investigations.

Keywords: *regression modeling, atmospheric outlook of Lahore, ozone contributors, ecology, atmospheric remote sensing*

Introduction

TropoO₃ is one of the short-lived but major atmospheric pollutants that plays a key role towards climate change (UNEP and WMO, 2011) by modifying the chemical composition of the troposphere and its oxidation capacity (Gauss et al., 2003). It is considered as a primary precursor for the production of hydroxyl radical (OH), that acts as an oxidizing agent severely disturbing the occurrence of tropospheric trace gases (Seinfeld and Pandis, 2016). TropoO₃ absorbs short-wave solar radiations and after attenuation re-radiates it in the form of long-wave radiations (Shan et al., 2008; Pal, 2010; Tan et al., 2014). TropoO₃ has harmful effects on human health, vegetation health & yield, and sensitive ecosystems (EPA, 2003; The Royal Society, 2008; Avnery et al., 2011; Burney and Ramanathan, 2014). Also, TropoO₃ is one of the major anthropogenic greenhouse gases, which is responsible for global warming (IPCC, 2013). Ozone in troposphere is either produced from chemical reactions of NO_x, CH₄, CO, and volatile organic compounds (VOCs) through non-linear, complex, and feedback-regulated processes in the presence of solar radiations (El-Fadel et al., 2002; Pulikesi et al., 2006;

Stevenson et al., 2006; Young, 2013; Seinfeld and Pandis, 2016) or transported directly from the stratosphere (Hsu and Prather, 2009). From an ecological point of view, both increased emission and destruction of VOCs by TropoO₃ may affect several of their multiple functions (Lerdau and Slobodkin, 2002; Holopainen, 2004; Van, 2009; Yuan et al., 2009). There are a number of anthropogenic and natural sources of these TropoO₃ precursors e.g.; crop-residue burning, biomass burning, fossil fuel combustion, enhancement of volatile organic compounds through convections of CO and aerosols, vehicular emissions, industrial processing, energy production activities, waste management, lightning NO_x, and soil emissions (Jaeglé et al., 2004; Satheesh and Moorthy, 2005; Stevenson et al., 2006; Aghedo et al., 2007; Wang et al., 2012; Ganesan et al., 2013; Ul-Haq et al., 2014). Anthropogenic activities pose numerous threats to the functioning, structural growth and diversity of natural and semi-natural ecosystems (Bobbink, 1989). Globally, an increase of 36% in TropoO₃ burden has been reported since 1850 (Cooper et al., 2014). TropoO₃ has strong effects on ecological interactions based on VOC signaling (Pinto et al., 2010).

Various natural, meteorological and anthropogenic factors are responsible for TropoO₃ concentration variability. Natural factors include; seasons, Earth-Sun distance, and location (altitude, latitude, and longitude), meteorological factors include; temperature, precipitation, humidity, soar flux, and wind speed, while anthropogenic factors include; emissions from precursor pollutants, biomass burning, vehicular emissions, and industrial processing (Ahmad and Aziz, 2013; Iqbal et al., 2014). Aerosols decrease TropoO₃ formation in polluted areas (Satheesh and Moorthy, 2005; Xing et al., 2017) and also decline its concentration after reacting with tropospheric gases (Sahoo et al., 2005). The primary objective of this study is to investigate the potential contributions of major tropospheric gases, meteorological parameters, and aerosol burden towards TropoO₃ variability over Lahore. TropoO₃ monitoring is being carried out either by onboard satellite sensors (Cooper et al., 2014; Ziemke et al., 2014) or via ground-based stations (Fioletov et al., 2008). Remote sensing techniques provide cost-effective, synoptic and repetitive coverages for spatio-temporal monitoring of tropospheric gases, meteorological parameters, and aerosol burden either at large or small scales. Whereas, data acquisition via ground-based stations is expensive, time-consuming, and almost impossible for inaccessible areas. The present study is carried out by retrieving datasets from onboard satellite sensors including; AIRS, AMSU-A, MODIS, MOPITT, OMI, and TRMM, as well as GLDAS model. To the best of our knowledge, no detailed study has so far been published for assessing TropoO₃ variability as functions of remotely sensed major tropospheric gases, meteorological parameters, and aerosol burden over Lahore.

Material and Methods

Location and description of the study area

Metropolitan corporation Lahore (31.5204°N, 74.3587°E) is the capital city of the Punjab province in Pakistan. As per 6th Population Census 2017 results, Lahore is the second-most populous city and financial hub of Pakistan after Karachi having population over 11 million and an area of 1,172 km² (Figure 1). Lahore is bounded by Sheikhpura District on the north and west bifurcated by Ravi River, Kasur District on its south, and Wagah Border along Indian province Punjab on the east. Lahore is the 16th largest and rapidly growing city in the world (CMF, 2018). Being financial hub, Lahore contains all types of Industrial, manufacturing, construction, business, and IT related activities

including but not limited to; pharmaceutical and chemical factories, steel mills, construction materials, power generation, manufacturing of motor vehicles, home appliance, electronics, leather goods, and paper products (Alam et al., 2012; Ali et al., 2014). Major air pollution sources in Lahore include automobile emissions, road dust, and biomass burning along with air pollutants transported from nearby regions (Alam et al., 2010, 2014). The Indian states of Punjab, Haryana, and Uttar Pradesh (shown in *Figure 1*) are reported to be the well-known air-pollutant emission sources from large scale rice and wheat crops generated residue burning and periodically transported to Lahore (Sidhu and Beri, 2008; Badarinath et al., 2009; Mishra and Shibata, 2012). Lahore falls under hot semi-arid and wet category based on the Köppen climate classification system with highly hot and wet summers (heavy monsoon rainfalls with frequent dust storms during June to September), while dry and cold winters (from December to February). Spring (March to May) and autumn (September to November with extensive crop residue burning in the surrounding regions) seasons are also observed in Lahore (Ali et al., 2014).

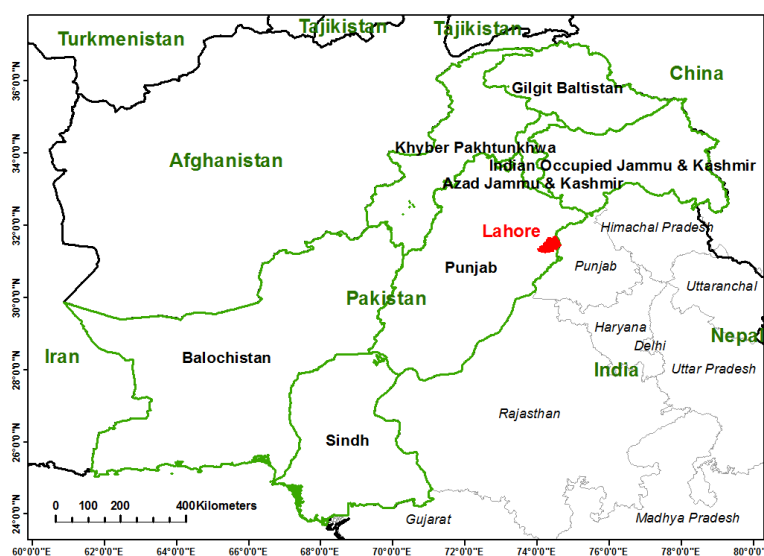


Figure 1. Map showing the geographical location of Lahore (31.5204°N , 74.3587°E).
(Source: <https://www.statsilk.com/maps/download-free-shapefile-maps>)

Datasets

Host of satellite borne sensors have been providing online data products for monitoring and analyzing atmospheric gases, meteorological parameters, and aerosol burden measured as aerosol optical depth (AOD). For current study, we retrieved monthly-mean data products of TropoCH₄, TropoCO₂, CLF, and OLR, while daily datasets of RH, and surface temperature over Lahore from Atmospheric Infrared Sounder (AIRS, Pagano et al., 2003; Abed, 2017) and the Advanced Microwave Sounding Unit-A (AMSU-A, Aumann et al., 2003) onboard AURA satellite operational since 2002. The combined use of AIRS and AMSU-A data products basically offers new as well as improved measurements (Susskind et al., 2003). Data products of TropoNO₂ (daily), TropoO₃ (monthly-mean), and planetary boundary layer SO₂ (monthly-mean) were acquired from NASA's EOS Ozone Monitoring Instrument (OMI) onboard Aura satellite launched in a sun-synchronous polar orbit on July 15, 2004 at an altitude of 705 km and inclination of

98.2° (Levelt et al., 2006a; Ziemke et al., 2011). OMI provides almost global coverage daily with a resolution of $13 \times 24 \text{ km}^2$ at nadir while $13 \times 100 \text{ km}^2$ at off-nadir (Boersma et al., 2006). It consists of hyperspectral imaging sensors to record radiances in the range of 270–500 nm (Levelt et al., 2006a,b). Additionally, monthly-mean data of TropoCO was retrieved from MOPITT onboard Terra, whereas, AOD from MODIS onboard Aqua deep blue. The datasets of surface level precipitation rate and near surface wind speed over Lahore were obtained from Tropical Rainfall Measuring Mission (TRMM, Liu et al., 2012) and Global Land Data Assimilation System (GLDAS, Fang et al., 2009), respectively. For validation of our resultant modeled, the original TropoO₃ data product of subtracted monthly-means of Total Column Ozone retrievable from OMI and Stratospheric Column Ozone retrievable from MSL onboard Aura satellite was utilized (Ziemke et al., 2006). Details about all the data products of major tropospheric gases, meteorological parameters, and aerosols used for this study are given in *Table 1*.

Methodology

We developed a bivariate correlation matrix representing tropospheric gases (CH₄, CO, CO₂, NO₂, O₃, and SO₂), meteorological parameters (CLF, OLR, PR, RH, Temp, and WS), and aerosol burden over Lahore using monthly-mean remotely sensed data during October 2004 – September 2015 (shown in *Table 2*). Each parameter mentioned in *Table 2* has either positive value (shown in blue colored bar) or negative value (shown in red colored bar), which indicates the type of correlation between the respective parameters. Longer the length of colored bars (either blue or red), stronger would be the correlation (either positive or inverse, respectively) between the corresponding parameters. Whereas, colored bars having smaller lengths (smaller values either positive or negative) indicate weaker correlations. In *Table 2*, correlation values written in bold text are significant at the 0.01 level (2-tailed), whereas, in italic text are significant at the 0.05 level (2-tailed). Respective statistical significance values, against each correlations of *Table 2*, are mentioned in *Table 3*. The highest statistical significance values at the 0.01 level (2-tailed) are highlighted in bold text, whereas, the statistical significance values at the 0.05 level (2-tailed) are mentioned in italic text in *Table 3*. The values written in regular text indicate least significance values. The highest correlation value [0.73] with 99% significance at 0.00 level (2-tailed) indicates that PR & CLF (meteorological parameters) are found to be positively highly correlated with each other in the study area during the observation period. Other higher positive correlations significant at 0.00 level (2-tailed) are; RH & CLF [0.70], RH & PR [0.69], Temp & AOD [0.69], and Temp & OLR [0.66]. Next equally higher but opposite correlations significant at 0.00 level (2-tailed) are; OLR & TropoO₃ [0.61], and TropoO₃ & TropoCO [-0.61]. Remaining relatively higher correlations significant at 0.00 level (2-tailed) are; WS & TropoO₃ [0.57], TropoCO₂ & TropoCH₄ [0.54], RH & TropoCH₄ [0.54], WS & TropoCO [-0.54], and RH & OLR [-0.52]. The parameters used in this study having weaker correlations with each other significant at 0.00 level (2-tailed) are (in descending order); OLR & CLF [-0.48], RH & TropoO₃ [-0.46], PR & AOD [0.45], PR & TropoCH₄ [0.45], RH & AOD [0.41], WS & TropoCO₂ [0.40], Temp & TropoCH₄ [0.39], AOD & TropoCH₄ [0.34], Temp & PR [0.33], OLR & TropoCO [-0.31], Temp & TropoO₃ [0.28], TropoNO₂ & TropoCO₂ [0.28], CLF & TropoCH₄ [0.27], WS & CLF [0.25], TropoCO₂ & TropoCO [-0.23], WS & TropoNO₂ [0.23], and TropoSO₂ & TropoO₃ [-0.23]. Out of all the tropospheric gases used in this study, TropoO₃ and TropoCO are found to be highly but inversely correlated with each other, whereas, TropoCO₂ and TropoCH₄ are next highly

and positively correlated gases significant at 0.01 level (2-tailed). Other tropospheric gases with weaker correlations including positive as well as inverse significant at 0.00 level (2-tailed) are; TropoNO₂ & TropoCO₂ [0.28], TropoCO₂ & TropoCO [-0.23], and TropoSO₂ & TropoO₃ [-0.23]. Only one correlation (least and positive) significant at 0.05 level (2-tailed) was found between TropoSO₂ & TropoNO₂ [0.18]. Positive as well as inverse correlations with 95% significance at 0.05 level (2-tailed) observed from this study are; OLR & AOD [0.22], CLF & TropoSO₂ [-0.22], AOD & TropoCO [-0.21], CLF & AOD [0.19], WS & RH [-0.19], and TropoSO₂ & TropoNO₂ [0.18]. All these correlations are also comparatively weaker ones.

Table 1. Metadata of data retrieved from different satellite sensors (level-3 products) and models used in this study

Product Name/Version	Sensor/Model	Units	Spatial Resolution	Temporal Resolution	Observation Period	Description
a) Tropospheric Gases						
TropoCH₄ – Tropospheric Methane (AIR3STM_VMR_A.700hPa v006)	AIRS/AMSU-A	ppbv	0.25°x0.25°	Daily	2002 – current	Tropospheric
TropoCO – Tropospheric Carbon Monoxide (MOP03JM v007)	MOPITT	ppbv	1° x 1°	Daily	2000 – current	Tropospheric
TropoCO₂ – Tropospheric Carbon Dioxide (AIR3C2M mole fraction v005)	AIRS/AMSU-A	ppm	1° x 1°	Daily	2002 – current	Tropospheric
TropoNO₂ – Tropospheric Nitrogen Dioxide (OMNO2d v003)	OMI	x10 ¹⁵ molecules cm ⁻²	0.25°x0.25°	Daily	2004 – current	Tropospheric
TropoO₃ – Tropospheric Ozone (AIR3STM_VMR_A.700hPa v006)	AIRS/AMSU-A	ppbv	0.25°x0.25°	Daily	2002 – current	Tropospheric
TropoSO₂ – Tropospheric Sulphur Dioxide (OMSO2e_PBL v003)	OMI	DU	0.25°x0.25°	Daily	2004 – current	Planetary boundary layer
b) Meteorological Parameters						
CLF – Cloud Fraction (AIRX3STM v006)	AIRS/AMSU-A	Unit less	1° x 1°	Daily	2002 – current	Total column
OLR – Outgoing Long-wave Radiation (AIRX3STM v006)	AIRS/AMSU-A	W m ⁻²	1° x 1°	Daily	2002 – current	Top of the atmosphere
PR – Precipitation Rate (TMPA/3B43 v007)	TRMM	mm month ⁻¹	0.25°x0.25°	Monthly	1998 – current	Surface level
RH – Relative Humidity (AIRX3STD v006)	AIRS/AMSU-A	%	1° x 1°	Daily	2002 – current	925 hPa
Temp – Air Temperature (AIRX3STD v006)	AIRS/AMSU-A	Kelvin	1° x 1°	Daily	2002 – current	925 hPa
WS – Wind Speed (GLDAS_NOAH025_M v2.1)	GLDAS Model	m s ⁻¹	0.25°x0.25°	Monthly	2000 – current	Near surface level
c) Aerosols						
AOD – Aerosol burden measured as Aerosol Optical Depth (MOD08_M3_550 v006)	MODIS	Unit less	1° x 1°	1–2 days	2002 – current	Total column
d) Independent data product of TropoO₃ for Model Validation						
TropoO₃ – Tropospheric Ozone OMI/MLS tropospheric ozone (original product)	OMI-MLS	ppbv	1° x 1.25°	Monthly-mean	2004 – current	Tropospheric

Table 2. Bivariate correlation matrix representing monthly-mean data of tropospheric gases including TropoCH₄ (ppbv), TropoCO (ppbv), TropoCO₂ (ppm), TropoNO₂ ($\times 10^{15}$ molecules cm⁻²) and TropoSO₂ (DU); major meteorological parameters including cloud fraction (CLF, unit less), outgoing long-wave radiation (OLR, W m⁻²), precipitation rate (PR, mm month⁻¹), relative humidity (RH, %), temperature at 925 hPa (Temp, K) and surface wind speed (WS, m s⁻¹); and aerosol burden measured as aerosol optical depth (AOD, unit less) over Lahore during October 2004 – September 2015

Parameters	TropoCH ₄	TropoCO	TropoCO ₂	TropoNO ₂	TropoO ₃	TropoSO ₂	AOD	CLF	OLR	PR	RH	Temp
TropoCO	-0.14											
TropoCO ₂	0.54	-0.23										
TropoNO ₂	-0.15	0.01	0.28									
TropoO ₃	-0.07	-0.61	0.14	-0.06								
TropoSO ₂	-0.02	0.13	0.08	0.18	-0.23							
AOD	0.34	0.21	-0.01	0.07	0.09	0.01						
CLF	0.27	-0.17	0.01	-0.09	-0.07	0.22	<i>0.19</i>					
OLR	0.07	-0.31	-0.06	-0.15	0.61	-0.05	<i>0.22</i>	-0.48				
PR	0.45	-0.16	0.02	-0.15	0.00	-0.12	0.45	0.73	-0.10			
RH	0.54	0.12	0.03	-0.15	-0.46	-0.03	0.41	0.70	-0.52	0.69		
Temp	0.39	-0.16	-0.11	-0.14	0.28	0.01	0.69	-0.09	0.66	0.33	0.11	
WS	-0.12	-0.54	0.40	0.23	0.57	-0.13	0.04	0.25	0.10	0.14	<i>0.19</i>	-0.11

Correlations in bold text are significant at the 0.01 level (2-tailed). Correlations in Italic are significant at the 0.05 level (2-tailed). Blue color indicates positive correlations, and red color indicates negative/inverse correlations. Longer the length of colored bars (either blue or red), stronger would be the correlation (either positive or inverse, respectively) between the corresponding parameters

Table 3. Based upon correlation matrix discussed in Table 2 above, matrix showing statistical significance values of tropospheric gases including TropoCH₄ (ppbv), TropoCO (ppbv), TropoCO₂ (ppm), TropoNO₂ ($\times 10^{15}$ molecules cm⁻²) and TropoSO₂ (DU); major meteorological parameters including cloud fraction (CLF, unit less), outgoing long-wave radiation (OLR, W m⁻²), precipitation rate (PR, mm month⁻¹), relative humidity (RH, %), temperature at 925 hPa (Temp, K) and surface wind speed (WS, m s⁻¹); and aerosol burden measured as aerosol optical depth (AOD, unit less) over Lahore during October 2004 – September 2015

Parameter	Tropo CH ₄	Tropo CO	Tropo CO ₂	Tropo NO ₂	Tropo O ₃	Tropo SO ₂	AOD	CLF	OLR	PR	RH	Temp
TropoCO	0.11											
TropoCO ₂	0.00	0.01										
TropoNO ₂	0.08	0.94	0.00									
TropoO ₃	0.39	0.00	0.12	0.50								
TropoSO ₂	0.77	0.13	0.36	<i>0.03</i>	0.01							
AOD	0.00	<i>0.02</i>	0.95	0.41	0.31	0.89						
CLF	0.00	0.06	0.91	0.30	0.44	<i>0.01</i>	<i>0.03</i>					
OLR	0.41	0.00	0.51	0.09	0.00	0.58	<i>0.01</i>	0.00				
PR	0.00	0.06	0.83	0.09	0.97	0.17	0.00	0.00	0.26			
RH	0.00	0.18	0.76	0.08	0.00	0.68	0.00	0.00	0.00	0.00		
Temp	0.00	0.07	0.22	0.11	0.00	0.97	0.00	0.29	0.00	0.00	0.22	
WS	0.15	0.00	0.00	0.01	0.00	0.13	0.63	0.00	0.27	0.12	<i>0.03</i>	0.22

The numbers written in bold text are statistical significance values for significant correlations at the 0.01 level (2-tailed). The numbers written in italic text are statistical significance values for significant correlations at the 0.05 level (2-tailed)

Annual trends (slopes and y-intercepts), overall changes (%), and R² of tropospheric gases, meteorological parameters, and aerosol optical depth over Lahore using their annual-means from October 2004 to September 2015 are tabulated in Table 4. All tropospheric gases except TropoCO indicate an increasing trend. Maximum positive

slope of 4.93 ± 0.18 with $R^2=0.99$ observed for TropoCH₄ (having high correlations [0.54] significant at 0.01 level with TropoCO₂ and RH) indicates a maximum increasing trend than other gases used for this study. Similarly, slopes of PR (3.21 ± 1.08) with maximum change of 49.61% (having strong correlation with CLF), and TropoCO₂ (1.97 ± 0.04 , having high correlation with TropoCH₄) show relatively increasing trends during the study period. However, a relatively large negative slope of TropoCO (-3.48 ± 0.54) shows a decreasing trend over Lahore, which may be due to an overall decrease in TropoCO (-23.8%) during the entire study period. Moreover, TropoCO has inverse but high correlations significant at 0.01 level with TropoO₃ and WS. Relatively higher positive slopes (~21%) have been noticed for TropoNO₂, CLF, and WS, whereas little negative slopes have been observed for OLR (-0.05 ± 0.21) and temperature (-0.10 ± 0.04). It is worth mentioning here that the monthly-mean temperature values over Lahore during October 2004 – September 2015 show a little increasing trend (*Figure 3* refers).

Table 4. Annual trends (linear equation showing slopes and y-intercepts), overall change (%), and R^2 based on annual-means of tropospheric gases including TropoCH₄, TropoCO, TropoCO₂, TropoNO₂ and TropoSO₂; major meteorological parameters including cloud fraction (CLF), outgoing long-wave radiation (OLR), precipitation rate (PR), relative humidity (RH), temperature (Temp) and surface wind speed (WS); and aerosol burden measured as aerosol optical depth (AOD) over Lahore during October 2004 – September 2015

Parameter	Annual Trend Line	Overall Change (%)	R^2
TropoCH ₄ (ppbv)	$y = (4.93 \pm 0.18)x + (1799.10 \pm 1.35)$	2.92	0.99
TropoCO (ppbv)	$y = -(3.48 \pm 0.54)x + (202.36 \pm 3.94)$	-23.80	0.81
TropoCO ₂ (ppm)	$y = (1.97 \pm 0.04)x + (377.07 \pm 0.27)$	5.40	1.00
TropoNO ₂ ($\times 10^{15}$ molecules cm ⁻²)	$y = (0.06 \pm 0.02)x + (2.23 \pm 0.17)$	21.62	0.40
TropoO ₃ (ppbv)	$y = (0.29 \pm 0.08)x + (47.91 \pm 0.62)$	6.18	0.54
TropoSO ₂ (DU)	$y = (0.00 \pm 0.00)x + (0.06 \pm 0.02)$	13.27	0.01
CLF (unit less)	$y = (0.00 \pm 0.00)x + (0.19 \pm 0.02)$	21.47	0.29
OLR (W m ⁻²)	$y = -(0.05 \pm 0.21)x + (302.66 \pm 1.52)$	-0.18	0.01
PR (mm month ⁻¹)	$y = (3.21 \pm 1.08)x + (32.62 \pm 7.97)$	49.61	0.47
RH (%)	$y = (0.41 \pm 0.15)x + (40.71 \pm 1.13)$	9.94	0.42
Temp (K at 925 hPa)	$y = -(0.10 \pm 0.04)x + (300.50 \pm 0.30)$	-0.37	0.38
WS (m s ⁻¹)	$y = (0.05 \pm 0.01)x + (1.86 \pm 0.08)$	21.43	0.66
AOD (unit less)	$y = (0.01 \pm 0.00)x + 0.62 \pm 0.02$	10.69	0.39

Results and Discussion

Parametric out-look of Lahore

Tropospheric gases

Concentrations of major tropospheric gases have been compared with those of TropoO₃ over Lahore during October 2004 – September 2015 and are shown in *Figure 2*. Large variations have been observed in TropoCH₄, TropoCO, and TropoNO₂, whereas, TropoCO₂ and TropoSO₂ show comparatively little variations. We also noticed that all tropospheric gases indicate increasing trends except TropoCO. Gradual increasing trend in TropoCH₄ indicates gradual increasing activities responsible for Methane production (Paddy crops, animals, industrial activities, and vehicular emissions etc.) in the study area

during the study period. Similarly, gradual increasing trend in TropoCO₂ is an indication of increased activities causing carbon dioxide to increase (Agricultural activities, population, animals, industrial activities, and vehicular emissions etc.) in Lahore. Whereas, slopes of TropoNO₂ and TropoSO₂ are quite gentle showing little increase similar to that of TropoO₃.

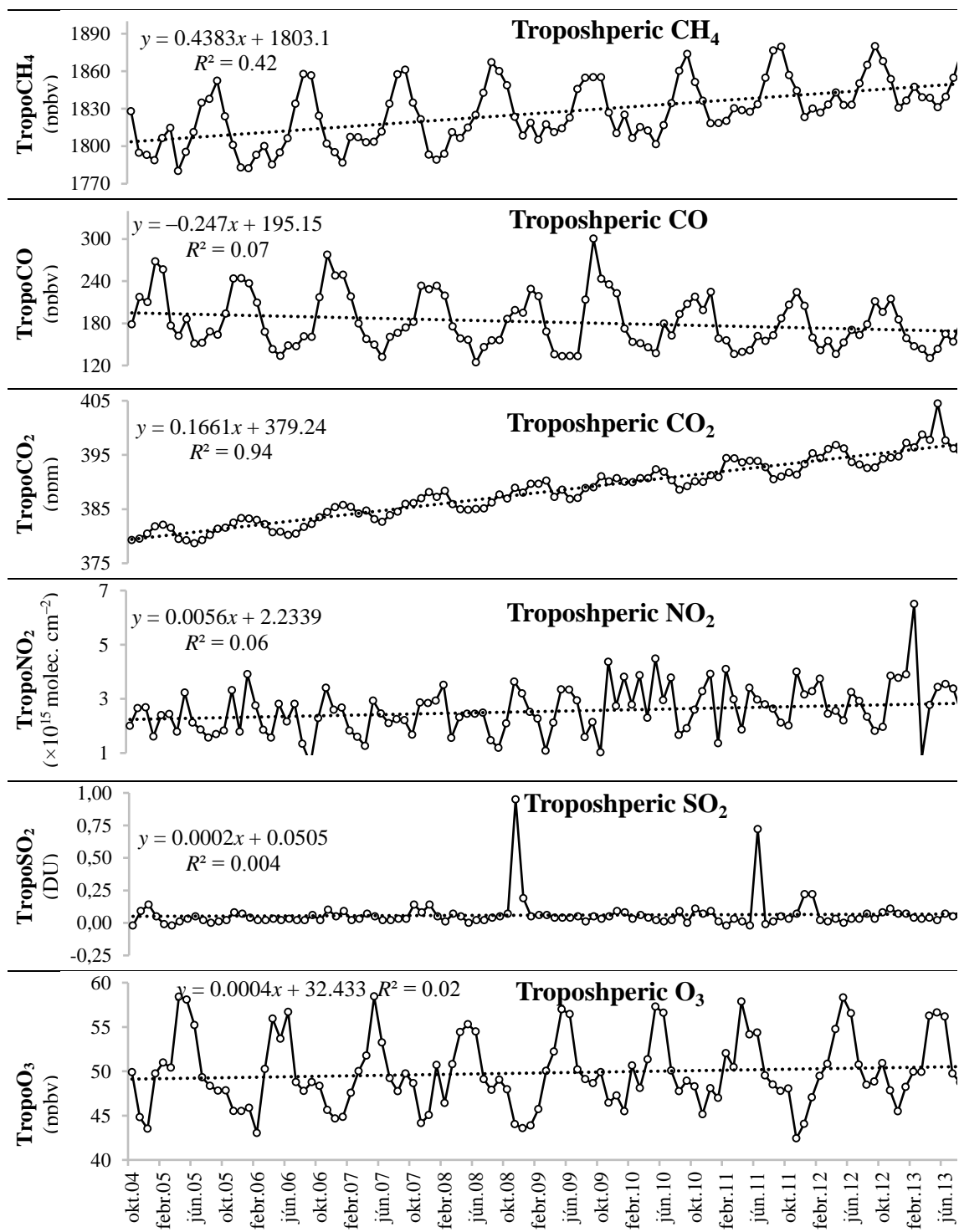


Figure 2. Time series monthly-mean variations of major tropospheric gases over Lahore during October 2004 – September 2015. Dotted lines correspond to respective trend lines

Meteorological parameters

Monthly-mean data products of major meteorological parameters as well as aerosol burden measured as Aerosol Optical Depth (AOD) have been compared with TropoO₃ concentrations over Lahore during October 2004 – September 2015 and are shown in *Figure 3*. Large variations have been observed in all parameters similar to that of TropoO₃ during the entire study period. Positive slopes of all parameters including TropoO₃, except OLR, indicate increasing trends during the study period. Comparatively larger slope value of PR indicates a gradual increase in precipitation in the study area.

TropoO₃

Maximum concentration of TropoO₃ has been reported during spring and summer seasons across the globe (e.g., Cooper et al., 2014). Accordingly, maximum concentrations have been observed in TropoO₃ over Lahore during spring and summer (monsoon) seasons (March to July) in Pakistan shown in *Figure 4*. Major causes of this increased TropoO₃ concentration over Lahore are mainly due to increased ozone precursor gas emissions, enhanced biomass burning activities, biogenic emissions, polluted air-masses migrated from nearby regions, removal of NO₂ concentration due to increased OH radicalization (Yienger and Levy, 1995), photolysis of water vapors which reduces NO₂ concentration during high rainfall in monsoon (Jacob, 2003; Ghude et al., 2009; Seinfeld and Pandis, 2016), and enhanced intrusion of O₃ from stratosphere (Noreen et al., 2018). An overall decreasing trend has been observed in the monthly-mean values of observed TropoO₃ concentration over Lahore (slope=-0.4303, y-intercept=52.786, and R²=0.1614) during the study period.

Modeling TropoO₃ concentration over Lahore

In order to investigate TropoO₃ dependencies, monthly-mean values of major tropospheric gases, meteorological parameters, and aerosol burden over Lahore during October 2004 – September 2015 have been incorporated in multiple linear regression model (e.g., Engel-Cox et al., 2004; Lin et al., 2012), mentioned in *Equation (1)*;

$$\text{Objective or Dependent Variable} = a_0 + \sum(b_n \times w_n) + \varepsilon \quad (\text{Eq.1})$$

where a_0 is an intercept, $b=1,2,3,\dots,n$ are regular (unstandardized) regression coefficients, $w=1,2,3,\dots,n$ are regressors or independent variables, and ε is an error term associated with the regression analysis. The resultant multiple linear regression model used in this study to model TropoO₃ (Objective/Dependent Variable) as functions of tropospheric gases, meteorological parameters, and aerosol burden (regressors or independent variables) is mentioned in *Equation (2)* below;

$$\begin{aligned} \text{TropoO}_3(\text{monthly mean}) &= a_0 + b_1 \times \text{TropoCH}_4 \\ &+ b_2 \times \text{TropoCO} \\ &+ b_3 \times \text{TropoCO}_2 + \\ &b_4 \times \text{TropoNO}_2 + b_5 \times \text{TropoSO}_2 + b_6 \times \text{CLF} \\ &+ b_7 \times \text{OLR} + \\ &b_8 \times \text{PR} + b_9 \times \text{RH} + b_{10} \times \text{Temp} + b_{11} \times \text{WS} \\ &+ b_{12} \times \text{AOD} + \varepsilon \end{aligned} \quad (\text{Eq.2})$$

where a_0 is an intercept, $b_1, b_2, b_3, \dots, b_{12}$ are regular (unstandardized) regression coefficients; TropoCH₄, TropoCO, TropoCO₂, TropoNO₂, TropoSO₂, CLF, OLR, PR, RH, Temp, WS, and AOD are the monthly-mean regressors or independent variables; and ε is an error term associated with the regression analysis. The multiple linear regression model mentioned in *Equation (2)* closely measures the intrinsic relationships of TropoO₃ with all regressors, separately. Therefore, this model offers better predictability of TropoO₃ based on relative impacts with all regressors than by simple linear model which is based only upon single regressor.

To improve the results of *Equation (2)*, stepwise multiple linear regression procedure (Lin et al., 2012) was adopted. Resultant stepwise multiple linear regression model shown in *Table 5* was generated. The model provides statistical information for modeling TropoO₃ concentrations based upon weighted contributions from its regressors. This procedure simultaneously regresses multiple variables (gases, meteorological parameters, and aerosol burden) and removes un-important parameter(s) from the model. *Table 5* shows that the modeled TropoO₃ is mainly dependent upon OLR, WS, CLF, TropoSO₂, TropoCO, and RH out of all regressors mentioned in *Equation (2)*. The standardized regression coefficients (β) obtained from the stepwise multiple linear regression model show weight-wise dependence of TropoO₃ upon OLR [$\beta=0.458$] with the highest contribution, followed by nearly equivalent contributions from WS [$\beta=0.262$] and CLF [$\beta=0.256$], whereas, the least contribution has been observed from RH [$\beta=-0.320$]. This model also reveals that TropoSO₂ [$\beta=-0.097$] and TropoCO [$\beta=-0.231$] are negatively contributing against TropoO₃ concentration over Lahore during the study period. The values of multiple error ($r=0.85$) and standard error (2.14) for this TropoO₃ model appear to be reasonable.

A subsequent bivariate correlation matrix representing observed TropoO₃, modeled TropoO₃, and the parameters which contributed in the modeled TropoO₃ concentration (OLR, WS, CLF, TropoSO₂, TropoCO, and RH) as mentioned in *Table 5* above was generated and is shown in *Table 6* below. Blue color indicates positive correlations, while red color indicates negative/inverse correlations. Longer the length of colored bars (either blue or red), stronger would be the correlation (either positive or inverse, respectively) between the corresponding parameters. In *Table 6*, correlation values written in bold text are significant at the 0.01 level (2-tailed), whereas, in italic text are significant at the 0.05 level (2-tailed). Respective statistical significance values, against each correlations of *Table 6*, are written in *Table 7*. The highest statistical significance values at the 0.01 level (2-tailed) are highlighted in bold text, whereas, the statistical significance values at the 0.05 level (2-tailed) are mentioned in italic text in *Table 7*. The values written in regular text indicate least significance values. All correlations shown in *Table 6* exhibit the same corresponding values as mentioned in *Table 2* above along with additive correlation values against modeled TropoO₃. All bivariate correlations with the modeled TropoO₃ concentration are significant at 0.01 level (2-tailed) except for CLF. The highest correlation value [0.85] with 99% significance at 0.00 level (2-tailed) between modeled and observed TropoO₃ concentrations over Lahore during the study period reveals the high suitability of the modeled developed in this study mentioned in *Table 5* above. Out of two tropospheric gases used in this model, modeled TropoO₃ is found to be highly but inversely correlated with TropoCO [-0.72], whereas, OLR [0.71] and WS [0.68] are the next highly and positively correlated meteorological parameters significant at 0.01 level (2-tailed).

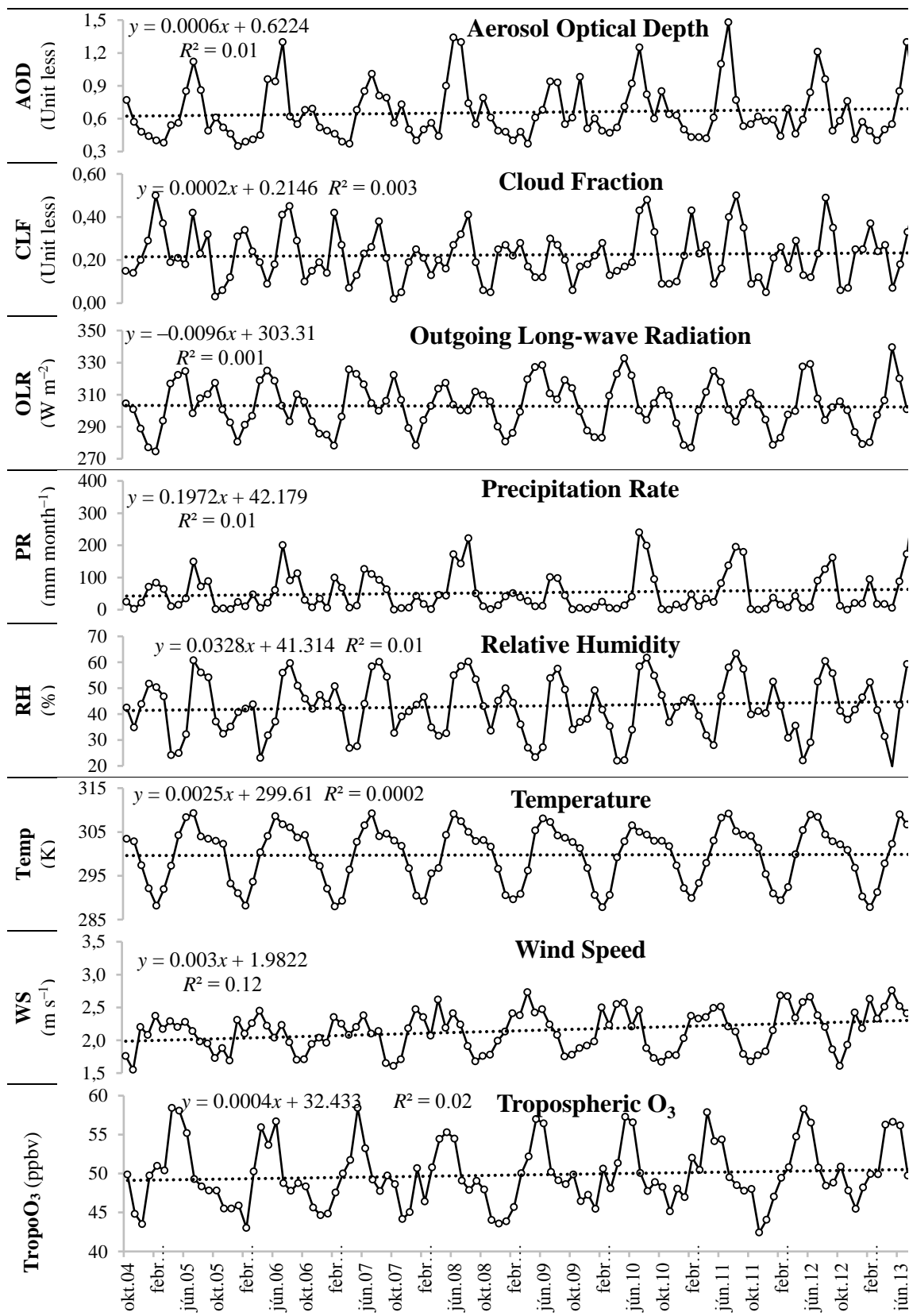


Figure 3. Time series monthly-mean variations of TropoO₃ and major meteorological parameters over Lahore during October 2004 – September 2015. Dotted lines correspond to respective trend lines

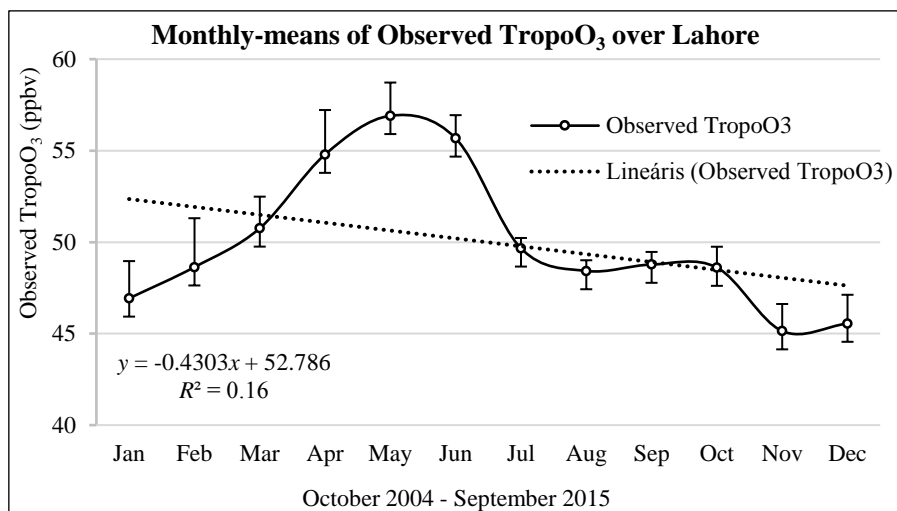


Figure 4. Monthly-means of Observed TropoO₃ concentration (ppbv) over Lahore during October 2004 – September 2015. Dotted line corresponds to the trend line

Table 5. Stepwise multiple linear regression model developed to model TropoO₃ (ppbv) variations based upon weighted contributions of monthly-mean data of OLR ($W m^{-2}$), WS ($m s^{-1}$), CLF (unit less), TropoSO₂ (DU), TropoCO (ppbv), and RH (%) over Lahore during October 2004 – September 2015

Stepwise multiple linear regression model equation	Multiple r	Standard Error
TropoO ₃ (monthly-mean) = 12.706 + 0.127OLR[0.458] + 3.213WS[0.262] + 9.078CLF[0.256] - 3.648TropoSO ₂ [-0.097] - 0.026TropoCO[-0.231] - 0.121RH[-0.320]	0.85	2.14

The values in parentheses are Standardized Regression Coefficients (Beta, β) representing regressor's relative weightage on modeled TropoO₃ concentrations in descending order, although these coefficients do not take part in calculations

Table 6. Bivariate correlation matrix representing monthly-mean data of observed TropoO₃, OLR ($W m^{-2}$), WS ($m s^{-1}$), CLF (unit less), TropoSO₂ (DU), TropoCO (ppbv), and RH (%) as well as modeled TropoO₃ concentrations over Lahore during October 2004 – September 2015

Parameters	Observed TropoO ₃	OLR	WS	CLF	TropoSO ₂	TropoCO	RH
OLR	0.61						
WS	0.57	0.10					
CLF	-0.07	-0.48	0.25				
TropoSO ₂	-0.23	-0.05	-0.13	0.22			
TropoCO	-0.61	-0.31	-0.54	-0.17	0.13		
RH	-0.46	-0.52	-0.19	0.70	-0.03	0.12	
Modeled TropoO ₃	0.85	0.71	0.68	-0.07	-0.27	-0.72	-0.53

Correlations in bold text are significant at the 0.01 level (2-tailed). Correlations in Italic are significant at the 0.05 level (2-tailed). Blue color indicates positive correlations, and red color indicates negative/inverse correlations. Longer the length of colored bars (either blue or red), stronger would be the correlation (either positive or inverse, respectively) between the corresponding parameters

Table 7. Based upon correlation matrix discussed in Table 6 above, matrix showing statistical significance values of observed TropoO₃, OLR (W m⁻²), WS (m s⁻¹), CLF (unit less), TropoSO₂ (DU), TropoCO (ppbv), and RH (%) as well as modeled TropoO₃ concentrations over Lahore during October 2004 – September 2015

Parameters	Observed TropoO ₃	OLR	WS	CLF	TropoSO ₂	TropoCO	RH
OLR	0.00						
WS	0.00	0.27					
CLF	0.44	0.00	0.00				
TropoSO ₂	0.01	0.58	0.13	<i>0.01</i>			
TropoCO	0.00	0.00	0.00	0.06	0.13		
RH	0.00	0.00	<i>0.03</i>	0.00	0.68	0.18	
Modeled TropoO ₃	0.00	0.00	0.00	0.42	0.00	0.00	0.00

The numbers written in bold text are statistical significance values for significant correlations at the 0.01 level (2-tailed). The numbers written in italic text are statistical significance values for significant correlations at the 0.05 level (2-tailed)

Other inverse correlations of the modeled TropoO₃ significant at 0.00 level (2-tailed) are noticed against RH [-0.53] and TropoSO₂ [-0.27]. The least and inverse correlation having 95% significance at 0.05 level (2-tailed) is against CLF [-0.07].

The observed TropoO₃ datasets retrieved directly from the AIRS/AMSU-A sensor over Lahore during October 2004 – September 2015 and the modeled TropoO₃ concentration values obtained from of the resultant step-wise multiple linear regression model (discussed in Table 5 above) have been plotted in Figure 5.

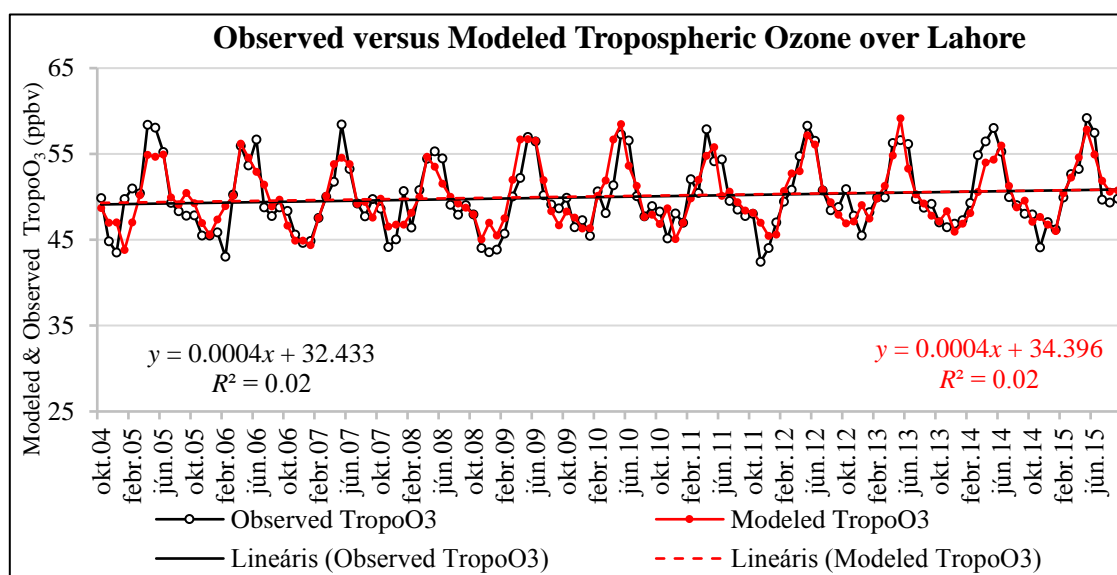


Figure 5. Comparison of observed versus modeled time-series TropoO₃ (ppbv) concentrations over Lahore during October 2004 – September 2015. Dotted lines correspond to respective trend lines

Time series distributions of these observed and modeled TropoO₃ concentrations show similar increasing and consistent trends except during the months of May 2005, Feb 2006, May 2007, Dec 2008, Nov 2011, Dec 2012, May 2014, and Nov 2014. The highest value in the modeled TropoO₃ concentration has been notice for the month of May 2013.

Monthly-mean data analysis of modeled TropoO₃ concentration indicate a similar pattern as that of the observed TropoO₃ concentration over Lahore during the study period as shown in *Figure 6(a)*. We find higher concentrations of both modeled and observed TropoO₃ during spring and summer (monsoon) seasons, whereas, minima during the winter season. A similar observation about TropoO₃ variation was reported by Ahmad and Aziz, 2013. Our modeled TropoO₃ variation attributes a similar decreasing trend (slope=-0.3254, y-intercept=52.192, and R²=0.12) over Lahore during the study period. Scatter plot drawn between observed and modeled TropoO₃ indicates a linear positive trend having a slope 0.732 and y-intercept 13.469 with R²=0.73 as shown in *Figure 6(b)*.

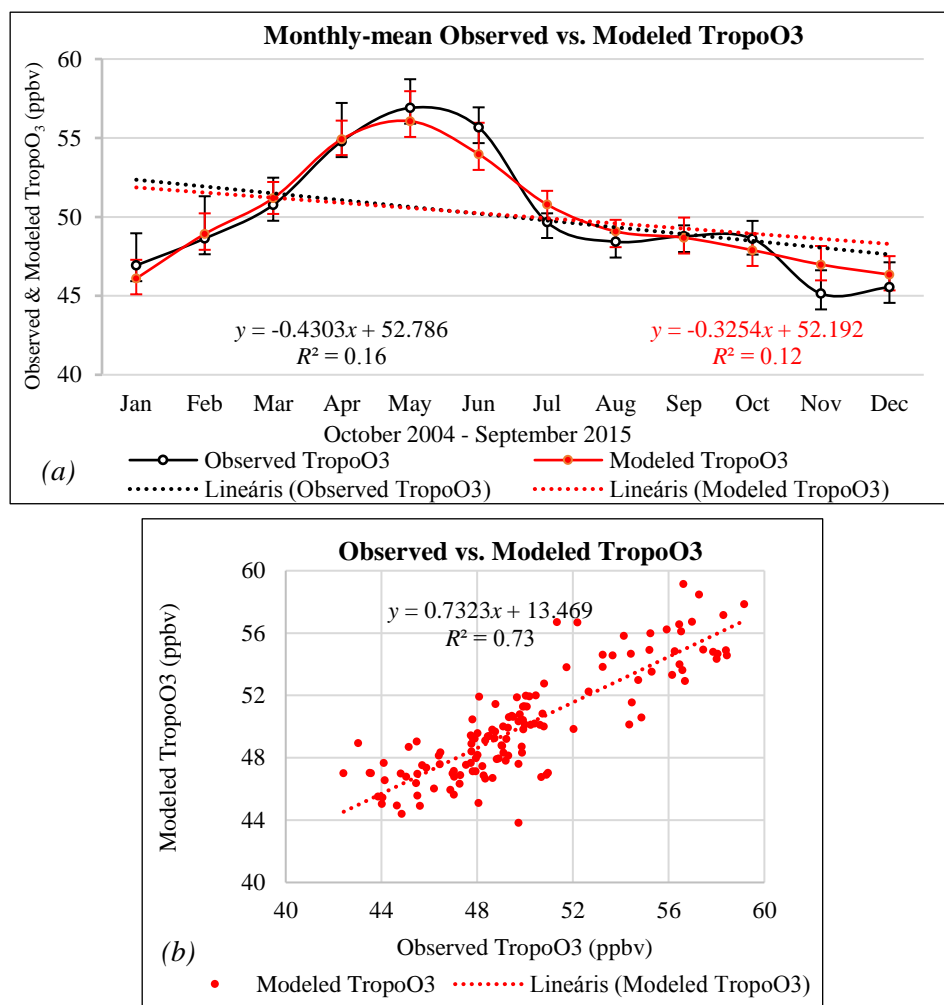


Figure 6. (a) Monthly-means of observed TropoO₃ (used as dependent variable) and modeled TropoO₃ concentrations (all data in ppbv) over Lahore during October 2004 – September 2015. (b) Scatter plot of observed versus modeled TropoO₃ concentrations (ppbv). Dotted lines correspond to respective trend lines

Annual trends, overall change (%), and R² based on annual-means of observed and modeled TropoO₃ concentrations over Lahore during October 2004 – September 2015 were calculated and are mentioned in *Table 8*.

Table 8. Annual trends (slopes and y-intercepts), overall change (%), and R^2 based on annual-means of observed and modeled TropoO₃ concentrations over Lahore during October 2004 – September 2015

Parameter	Annual Trend Line	Overall Change (%)	R^2
Observed TropoO ₃ (ppbv)	$y = (0.29 \pm 0.08)x + (47.91 \pm 0.62)$	6.18	0.54
Modeled TropoO ₃ (ppbv)	$y = (0.23 \pm 0.06)x + (48.48 \pm 0.42)$	4.89	0.62

Overall similar increasing concentrations of both observed (6.18%, $R^2 = 0.54$), and modeled (4.89%, $R^2 = 0.62$) TropoO₃ indicate good agreement of resultant model devised in this study. The slope of observed TropoO₃ [0.29 ± 0.08] is little larger than that of the modeled TropoO₃ [0.23 ± 0.06], however, the values of y-intercepts of both concentrations are nearly similar. The increase in TropoO₃ concentration observed in Lahore and adjoining areas during the study period is possibly due to increased anthropogenic activities including; population and traffic densities, industrial activities, infrastructure development activities, and biomass burning etc. These results are almost in agreement with those obtained by Noreen et al 2018 for ozone column over Lahore. Some other studies had also discussed about the TropoO₃ concentrations, and production of its precursors through different anthropogenic activities (e.g., Kulkarni et al., 2010; Lal et al., 2012; Ahmad and Aziz, 2013).

Model validation

Modeled TropoO₃ concentrations were crossed checked against an independent source of TropoO₃ concentrations. For this purpose, original TropoO₃ data product of subtracted monthly-means of Total Column Ozone retrievable from OMI and Stratospheric Column Ozone retrievable from MSL onboard Aura satellite was utilized (available at https://acd-ext.gsfc.nasa.gov/Data_services/cloud_slice/new_data.html). TropoO₃ product calculated by mean volume mixing ratio (in ppbv) formatted in 1-degree (latitude) by 1.25-degree (longitude) resolution was extracted for Lahore during October 2004 – September 2015. Our modeled as well as the observed TropoO₃ (used as dependent variable) concentrations have been plotted in *Figure 7* against these independently available product of OMI-MLS TropoO₃ concentrations (ppbv) over Lahore during the study period.

The results show similar seasonality patterns as well as trends in all three datasets with good agreements in their slopes and error values mentioned in *Figure 7*. These promising results validate our resultant model. OMI-MLS TropoO₃ concentrations have been plotted against secondary axis due to its wider data range. OMI/MLS data measurements have some anomalies especially during 2007 and 2009 (Ziemke et al., 2006).

To further investigate this validation process, monthly-mean data of OMI-MLS TropoO₃ (independent data) was compared with modeled as well as observed TropoO₃ concentrations over Lahore during October 2004 – September 2015 (shown in *Figure 8*). Monthly-mean graphical patterns, slopes, and trends in all three datasets are similar with higher concentrations during the spring and summer (monsoon) seasons, whereas, minima during the winter season shown in *Figure 8(a)*. Comparison of scatter plots drawn between OMI-MLS TropoO₃ (independent data), and observed TropoO₃ (used as

dependent variable) against modeled TropoO₃ concentrations shown in *Figure 8(b)* indicates similar linear positive trends.

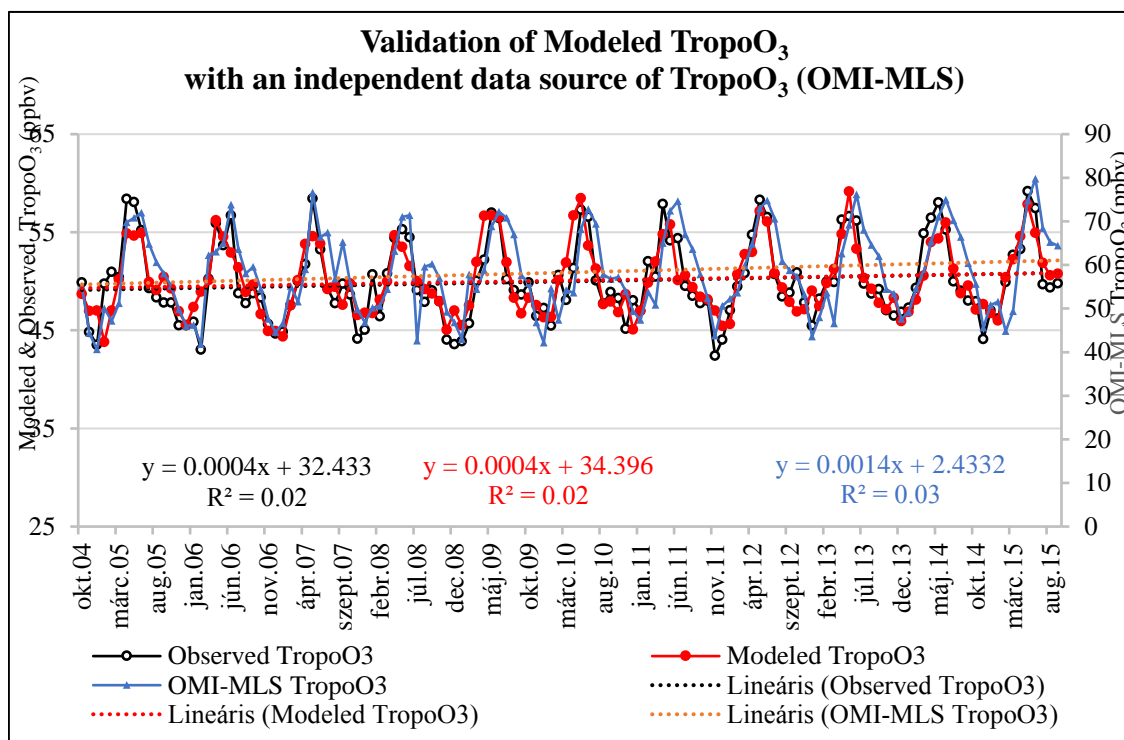


Figure 7. Validation of modeled and observed (used as dependent variable) TropoO₃ (ppbv) against an independent source of TropoO₃ concentrations (ppbv) retrieved from OMI-MLS data product over Lahore during October 2004 – September 2015. Dotted lines correspond to respective trend lines

Conclusions

The proposed step-wise multiple linear regression model reveals weighted contributions of major factors OLR, WS, and CLF, TropoSO₂, TropoCO, and RH (based on standardized regression coefficient, β) against TropoO₃ variability over Lahore during September 2004 – October 2015. Stepwise multiple linear regression modelling technique opted to model TropoO₃ concentrations over Lahore has shown highly consistent results as compared with the TropoO₃ observations retrieved directly from OMI satellite sensor. The trend-line of scatter plot drawn between observed and modeled TropoO₃ provides a steady linear increase analogous correlation with an overall increase of 6.18% in observed TropoO₃, while 4.89% in modeled TropoO₃ over Lahore during the study period. Based on the resultant value of multiple correlation coefficient ($r=0.854$) it can confidently be asserted that the stepwise multiple linear regression model opted to predict TropoO₃ over Lahore is quite reasonable. The modeled TropoO₃ concentrations were validated against an independent source of TropoO₃ concentrations retrieved from original subtracted product of OMI-MLS over Lahore for the study period and the results were found in good agreement. This study of modeling TropoO₃ variation as functions of satellite observations of major tropospheric gases, meteorological parameters, and aerosol burden over Lahore acquired during October 2004 – September 2015 using stepwise multiple

linear regression modelling technique is quite unique. The study would be helpful for further research investigations in this domain as well as provide useful information for policy/decision makers in Lahore, Pakistan.

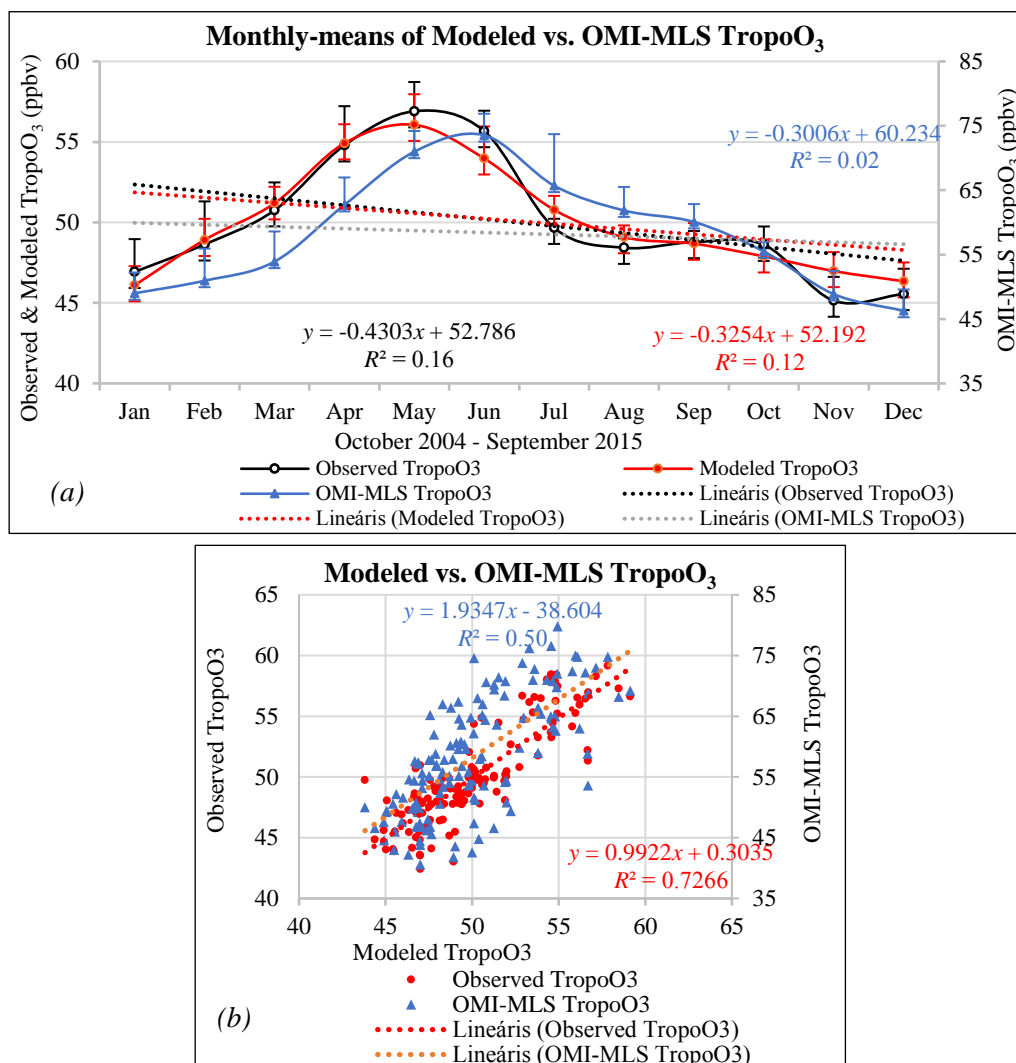


Figure 8. (a) Monthly-means of OMI-MLS TropoO₃ (independent data), observed TropoO₃ (used as dependent variable) and modeled TropoO₃ concentrations (all data in ppbv) over Lahore during October 2004 – September 2015. (b) Scatter plot of OMI-MLS TropoO₃ (independent data), and observed TropoO₃ (used as dependent variable) against modeled TropoO₃ concentrations (all data in ppbv). Dotted lines correspond to respective trend lines

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