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Reprint 1173

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29th Guangdong-Hong Kong-Macao Seminar on
Meteorological Science and Technology,
Macao, 20-22 January 2015

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Abstract

Ozone is a key constituent of the atmosphere in respect of climate change, oxidizing capacity and health impact. Tropospheric ozone is a particularly important greenhouse gas as it gives a positive radiative forcing to the Earth's surface. Long term trend of surface ozone is widely reported by different organizations or institutes. However, relatively little investigation has been done for upper-air ozone over tropical areas in terms of its long term behaviour and correlation to other environmental parameters like global solar radiation and moisture content in the atmosphere.

The Hong Kong Observatory started its ozonesonde operations at the King's Park Meteorological Station in October 1993. Up to the end of 2013, about 800 vertical ozone profiles were obtained. In this study, data of ozone profiles obtained between 1994 and 2013 are examined. The overall and monthly variation patterns in the troposphere and lower stratosphere are investigated. Trend analysis of the ozone mixing ratio using linear regression and decadal comparison is also carried out. Results of the analysis together with the correlation with other environmental parameters will be reported and discussed in this paper.

1. Introduction

The formation of ozone in the stratosphere absorbs most of the sun's ultra-violet radiation (UV-C ranges between 100 and 280 nm) and prevents the UV-C from reaching the Earth's surface. Stratospheric ozone depletion itself gives a slightly negative global mean radiative forcing (RF). In contrast, ozone in the troposphere is a direct greenhouse gas, which contributes 0.40($\pm$0.20) Wm^{-2} to the global mean RF [1]. Monitoring the variation or long term trend of ozone would be crucial in understanding the global warming issue. At the same time, the strong oxidizing power of ozone in the lower atmosphere can harm the health of human and plants, hindering photosynthesis which also exerts indirect positive feedback to the increase of global carbon dioxide [2]. All in all, the long term observations and analysis of atmospheric ozone are important in understanding the atmosphere in terms of climate change, oxidizing capacity and health impact.

The Hong Kong Observatory started to release ozonesonde once a month at the King's Park Meteorological Station (located at 22°18'43''N, 114°10'22''E) in Hong Kong since October 1993. The station was accepted into the Global Atmospheric Watch (GAW) programme by the World Meteorological Organization (WMO) in January 1996 for vertical ozone profile measurements with data continuously submitted to the World Ozone and Ultraviolet radiation Data Centre (WOUDC). The measurement frequency was once enhanced in support of the Pacific Exploratory Mission West-B (PEM-West B) project in 1993-1994 as well as the Transport and Chemical Evolution over the Pacific (TRACE-P) project in 2000-2001 [3]. The operations have been stepped up to weekly release since April 2003. Up to the end of 2013, close to 800 vertical ozone profiles were measured.

There were various studies of surface ozone in Hong Kong in the perspective of inter-seasonal and inter-annual behaviours. Effects of its precursors, temperature [4] and monsoon sensitivity [5] were also reported. However, most of the studies mainly focused on the ground level variation since more frequent data can be obtained from surface stations. Relatively fewer studies had been carried out for upper-air ozone. Among these studies, the Observatory [6] investigated the characteristics of ozone profiles above Hong Kong in early 2000s. Now after another decade, with 20 years of ozone profiles, the current study should give a better picture of the climatology

of upper-air ozone concentrations above Hong Kong.

In this study, we use the profiles obtained from 1994 to 2013 to summarize the upper-air ozone pattern above Hong Kong as well as to analyze its long term trend. Specifically, a comparison of global solar radiation data to ozone concentrations in the boundary layer (below 1 km) is also presented to assist in the discussions about the trend of ozone near the Earth's surface.

2. Data and methodologies

2.1 Instrumentation

The ozonesonde operated by the Observatory is of the electrochemical concentration cell type. It consists of an electrochemical cell ozone sensor (Science Pump Corporation Model ECC 6AB), a gas sampling pump, an electronic interface board and a radiosonde. Currently, model RS92-SGPD radiosonde of Vaisala [7] is used, while model RS80-15FE and RS80-15GE had been used in past operations. The ozone sensor itself contains two platinum electrodes immersed in potassium iodide (KI) solution of concentration 1% in both cathode and anode chambers. Ozone in air is continuously pumped into the cathode chamber and reacts with KI solution to produce current. More ozone in the air can induce larger current and hence ozone partial pressure can be deduced.

Each ozonesonde is attached to a 1.5 kg rubber balloon and will normally reach the stratosphere before the balloon bursts. The average ascent speed is about 5 ms^{-1} with a sampling rate of 2 seconds. Hence the vertical resolution of the profiles is about 10 meters. Apart from measuring ozone partial pressure, ambient pressure, air temperatures, relatively humidity and wind data are also measured and telemetered to the ground receiving system – DigiCORA system (MW15) from Vaisala.

2.2 Ozone profiles

Of the approximate 800 ozone profiles obtained between 1994 and 2013, only those 546 which reached 30 km or above are selected for the present study. This is to ensure that the ozone layer has been captured as much as possible for the computation of total ozone column as well as the monthly

mean of tropospheric/stratospheric ozone.

Total ozone column is computed by integrating the ozone profiles from the surface to the top of the atmosphere. This quantity is usually presented in Dobson Units (DU) (One DU is equal to 2.69 x 10^{20} molecules per m^2). Similarly, total tropospheric ozone column is calculated in the same way but integration from the surface to the tropopause.

In practice, if the balloon bursts in an altitude of around 30 km, the residual ozone column (total ozone after balloon bursts) has to be estimated. According to the ozone guidebook of WOUDC (2007) [8], the residual ozone column R can be corrected by extrapolating constant ozone mixing ratio for height after balloon bursts using the formula R(DU)=7.892 x O_{3p}, where O_{3p} is the partial pressure in mPa at burst level.

Surface ozone mixing ratio usually peaks in the afternoon under strong sunshine. The diurnal variation of it in Hong Kong can be significant and the peak-to-peak difference can be up to 100% [9]. The influence is also reported to have a significant correlation to upper-air ozone up to about 4 km above ground in Irene, South Africa [10], which is also a tropical station but in the Southern Hemisphere. For the Observatory's ozone sounding operation, the ozonesondes are scheduled to launch at 05 to 06 UTC (i.e 13 to 14H local time) and hence the diurnal effects in the boundary layer have not been taken into account in this study.

2.3 Meteorological data and trend analysis

Apart from the meteorological data measured by the ozonesondes, monthly reanalysis data of European Centre for Medium-Range Weather Forecasts (ECMWF) from 1994 to 2013 are also adopted in the present study to investigate the vertical motion and also the potential vorticity (PV) of the atmosphere above Hong Kong. Daily and monthly data of global solar radiation of the same period measured at King's Park Metrological Station, where the ozonesondes launch, are also used in this study.

In studying the changes of upper-air ozone and meteorological data over the 20-year period from 1994 to 2013, linear least square fitting is applied in different months at different levels. The slope and p-value (p) obtained from

fitting indicate the trend and the statistical significance respectively. The statistical significance defined in this paper is 95% ($p < 0.05$) unless otherwise specified.

3. Results and discussion

3.1 Overview of upper-air ozone pattern

The monthly variation of ozone profiles over Hong Kong is presented in Figure 1. Ozone mixing ratio increases drastically above the tropopause. The maximum stratospheric ozone mixing ratio is observed at a height of over 30 km above ground and it was about 100 times more concentrated than that in the troposphere. The stratospheric ozone also experiences inter-seasonal variation with a monthly maximum reaching 8800 ppbv (parts per billion by volume) or above in summer (JJA), closely related to the change of solar angle in the tropics.

The variation of the tropospheric ozone mixing ratio is more complicated. In general, a ridge of maximum intrudes down from the stratosphere in spring (MAM) and a trough of minimum extends from the surface in summer. This pattern agrees with the Brewer-Dobson circulation [11, 12], which describes the global transport of upper-air ozone between the stratosphere and the troposphere, accounting for the transport of ozone from the Equator towards the Poles as well as the seasonal variation of ozone in different latitudes.

Locally, for the springtime maximum, penetration of ozone from the stratosphere to the troposphere can be observed. Similar pattern was also reported in spring season in South Africa [10]. A case study of ozone penetration down to 8.5 km above ground was also reported in Linan over eastern China in spring [13]. However, in Figure 1, the ridge of the 60 ppbv line can go further down below 5 km above ground. The moist area below 5 km shown in the relative humidity profiles in Figure 2 indicates that the air masses in that level should not come from stratospheric origins. Thus, the relatively higher ozone below 5 km probably originates from anthropogenic source of ozone precursors (nitrogen oxide (NO_x), carbon monoxide (CO) and volatile organic compounds (VOC)) under the influence of the northeast monsoon with airflow mainly from inland.

In summer, with relatively clean and convective maritime airstream upwelling from the boundary layer, the tropospheric ozone content is basically lower. The monthly mean vertical velocity (positive and negative values mean downward and upward velocity respectively) of the atmosphere of the grid point (22.25^{o} N, 114.25^{o} E) nearest to Hong Kong from 1994 to 2013 is also plotted using ECMWF reanalysis data as shown in Figure 3. The vertical velocities align well with the upwelling motion in summer.

The near surface autumntime (SON) maximum up to about 50 ppbv around October (see Figure 1) is probably due to the onset of the northeast monsoon bringing air mass from mainland China together with ozone precursors [14]. This result is consistent with the surface ozone variation monitored by the Environmental Protection Department (EPD). The elevated ozone is also believed to be associated with fine and calm weather conditions in Hong Kong in autumn which favour the formation via photochemical reactions and accumulation of ozone [15].

On the other hand, the upper tropospheric wintertime (DJF) minimum (10 to 15 km height) as observed in Figure 1 is probably due to the transport of low ozone content maritime air masses to upper troposphere by convections from tropical origins following the East Asia Hadley circulation [16].

Table 1 summarizes the seasonal variation of ozone concentrations in term of ozone column. The total ozone column is the thickest in spring with a value of 280 DU. It decreases progressively and reaches a minimum of 237 DU in winter. Meanwhile, the total tropospheric ozone also possesses similar behaviour with a maximum value of 46 DU in spring but is around the same level for other seasons with values of about 36 DU. The ratio of total tropospheric ozone to total ozone is about 15% throughout the year with the highest value in spring (16%) and the lowest value in summer (13%).

3.2 Long term trend of upper-air ozone

The trend of upper-air ozone from 1994 to 2013 is shown in Figure 4(a). Linear least square fitting is applied to each month with vertical interval of 500 m. The change is presented in % per decade relative to the average mixing ratio over the 20-year period. The results show that the ozone mixing ratio decreases at around 20 km especially in winter and early spring. In contrast,

a statistically significant increase of over 50% per decade is observed in late summer and early autumn near surface. The trend varies seasonally in the middle and upper troposphere, generally positive in summer and autumn but negative in spring and winter.

The overall picture indicates that there were obvious changes of the ozone in the lower troposphere over the past decades. This tropospheric ozone is a product of photochemical reactions of its precursors NO_x, CO and VOC. They can be anthropogenic or natural. Also the precursors affecting one place can be produced both locally and from upwind areas. Hong Kong has been putting effort to reduce local emission of NO_x and CO over the past decades. Despite this, actual observations by the EPD show that the surface ozone level has been increasing. Certain literatures have reported that the rise is caused by the fact that increasing background ozone transported from upwind areas (southern and eastern China) negates the local effort resulting in an overall increase of surface ozone [17, 18].

Figure 4(b) is a zoom-in of the lower troposphere of 4(a) computed with a finer vertical resolution of 100 m. It can be seen that boundary layer (below 1 km) ozone is generally in an increasing trend throughout the year except in January and February at height above 500 m. In order to have a closer look of the change of ozone in this layer, a time series plot of the ozone column from surface to 1 km above ground is plotted in Figure 5. A statistically significant increasing trend with a slope of 0.6 ± 0.2 DU per decade is observed from 1994 to 2013. It is equivalent to about 19% rise per decade in reference of the 20-year average of 3.1 DU. Assisted by the upward motion in summer and early autumn as evidenced in Figure 3, the increased ozone is likely to be transported from the boundary layer and causes the rise of ozone in the middle or upper troposphere as observed in Figure 4(a). On the other hand, in spring and winter, the boundary layer ozone cannot be easily transported to upper atmosphere under stable environment due to downward transport motion as shown in Figure 3.

Besides, stratosphere troposphere exchange (STE) is also a possible source to transport the higher concentrated stratospheric ozone to the troposphere. The Observatory carried out a case study of increased ozone in the upper troposphere after the passage of a cold front system by investigating the change of potential vorticity (PV) above Hong Kong [6]. PV is employed as

a tracer as high PV values are characteristic of tropospheric air of stratospheric origin. In order to track whether there are any changes in frequency of STE events causing the increased ozone in troposphere over the past two decades, a time series of monthly mean PV from 1994 to 2013 of 350K obtained from ECMWF reanalysis is plotted in Figure 6. Grid point (22.25^{o} N, 114.25^{o} E) closest to Hong Kong is selected. Potential temperature of 350K is chosen because it is close to the transition layer between troposphere and stratosphere in tropical areas like Hong Kong (around 16 km, see Figure 1). No statistically significant trend can be found from 1994 to 2013, suggesting that the frequency of STE events due to upper air systems (upper air troughs, tropopause folding, etc) remained almost constant over the past 20 years. Therefore, we have no strong evidence to show that the variability of tropospheric ozone over the study period is due to the change of frequency of STE events.

Moisture in the atmosphere is one of the ozone sinks in decreasing ozone concentrations through the photolysis process to form free radicals and oxygen. To examine its effect to upper-air ozone, a plot of decadal trend of relative humidity (RH) from the ozonesondes selected in this study is shown in Figure 7. Focusing on the transition layer and the troposphere where most of the moisture is confined, the trend of upper-air RH exhibits somewhat similar pattern to the trend observed in Figure 4(a). The trend above 20 km is shown here but is not included in the discussion due to the extremely low RH of only a few percent. The uncertainty of the measurements above this level is very high as the accuracy of it is of order of unity. Over the 20-year period, moisture between 15 and 20 km increased throughout the year. The positive areas in Figure 7 also intrude from 15 km in spring and down to around 5 km in April. Other positive areas are also found in January and October in the middle troposphere. Comparing to the trend pattern in Figure 4(a), decreasing trends of ozone are mostly observed in these areas. However, from statistical point of view, the trend is not significant. Further observations and study have to be carried out for a better understanding of the local characteristics of the long term trend of upper-air ozone above Hong Kong.

3.3 Total ozone column

Global total ozone column was reported to be declining in 1980s and early

1990s due to the increasing release of Ozone Depleting Substances (ODS). The trend has levelled off since middle 1990s after the Montreal Protocol was introduced, restricting the production of ODS over the globe. The Antarctic ozone hole is also reported to have slow recovery since then [19,1]. While the global total ozone column shows a significant decreasing and then levelling off trend from 1980 to now, the tropical ozone column is found to be less affected. Eyring et al. (2013) [20] reported that only a small negative trend was seen based on satellite-borne observations in the tropics (25^{o}N to 25^{o}S) during the global declining period from 1980 to 2000.

Table 2 lists the seasonally and yearly mean ozone column above Hong Kong based on selected ozone profiles in this study. As visualized in Figure 4(a), the total tropospheric ozone column also gives different trends in different seasons. The trend is statistically significant in autumn. A generally negative trend is observed in the total ozone column but only significant in spring. In terms of yearly average, both total tropospheric ozone and total ozone column are not statistically significant with high uncertainty and a small change of only around 2% per decade with respect to their overall values shown in Table 1.

3.4 Correlation between global solar radiation and boundary layer ozone

It is well known that ozone can be easily produced on ground under stable atmosphere with abundant sunshine, especially in the cases of subsidence given by tropical cyclones. Global solar radiation measures the shortwave radiation reaching the Earth's surface from the Sun. Its variation depends on the amount of cloud cover, suspended aerosols (both scattering and absorbing) as well as the solar irradiance. As some of these contributors are involved in both warming and cooling forcings, changes in global solar radiation may not reflect fully the situation of global radiative balance. However, global solar radiation does represent the amount of sunshine reaching the Earth's surface and thus affects the rate of photochemistry of ozone in the boundary layer.

Figure 8 shows the correlation between daily global solar radiation and boundary layer ozone in different seasons. Positive correlations can be seen in all the seasons and the correlation is more prominent in winter. On the contrary, it is not statistically significant in summer ($p\sim0.13$). The main cluster of boundary layer ozone in summer stays in low values under maritime

airstream but there is a group of outliers which shows relatively high ozone content. The weather situations of them (not shown here) are studied and the high ozone content was caused by the presence of tropical cyclones to east of Hong Kong over the seas near Taiwan or Luzon, bringing continental airstream from the north as well as subsidence.

The long term trend of monthly global solar radiation is shown in Figure 9. A significant global brightening trend is observed in Hong Kong from 1994 to 2013. In a previous study of the trend of global solar radiation in Hong Kong for the period from 1961 to 2005, a declining trend was found in the 1960s and 1970s followed by a leveling off trend from early 1980s to 2005 [21]. The increasing trend from 1990s up to now is likely related to the decline in the emissions of sulphur and black carbon over southern China [22]. Besides the transport of ozone precursors from upwind areas, the global brightening trend may also be one of the causes to the rise of boundary layer ozone.

4. Conclusion

This study utilizes 20 years of ozone profiles to study the overall pattern and long term trend of upper-air ozone above Hong Kong. In the stratosphere, the ozone gives a monthly maximum above 8800 ppbv in summer. In the troposphere, a springtime maximum is observed to be transported from the stratosphere while a summertime minimum in the lower troposphere is mainly caused by advection of clean maritime airstream. With the onset of the northeast monsoon and the advection of upper tropospheric maritime air masses, there appears a near surface autumntime maximum and an upper tropospheric wintertime minimum respectively. The yearly mean ratio of tropospheric ozone to total ozone is found to be 15% over the study period with the highest value of 16% in spring and the lowest value of 13% in summer.

For the long term trend, the stratospheric ozone is found to be generally decreasing at around 20 km above ground. In contrast, the trend is seasonal dependent for the tropospheric ozone. It is positive in summer and autumn but negative in spring and winter. The trend is significant in autumn. Besides, the significant increase of surface ozone, which has been reported to be caused by upwind areas over southern or eastern China, is mainly concentrated in the boundary layer. Quantitatively, the boundary ozone

column below 1 km is found to be increasing with a rate of 0.6 ± 0.2 DU per decade by least square fitting over the 20 years of data. The increase of boundary layer ozone is believed to be transported to the middle and upper troposphere by upward motion in summer and early autumn.

Possibility of more frequent STE events above Hong Kong is also examined. There is no significant trend found in the 350K PV aloft Hong Kong based on the ECMWF reanalysis data. Therefore, we have no strong evidence to conclude any changes from ozone intrusions from the stratosphere caused by more frequent STE events. Comparison of the trend of atmospheric moisture to the trend of tropospheric ozone shows similar pattern, suggesting photolysis of ozone may have played a role here. But statistically, the trend of atmospheric moisture is not significant. Further study has to be carried out for better understanding.

Finally, correlations between global solar radiation and the boundary layer ozone in different seasons are investigated. The correlations are basically positive with statistically significance of 95%, except in summer. The strongest correlation is observed in winter. The trend of global solar radiation is found to be rising in the study period. The global brightening trend is believed to be one of the contributors to the rise of boundary layer ozone.

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Table 1 Seasonal variation of ozone column above Hong Kong from 1994 to 2013

	Spring	**Summer**	**Autumn**	**Winter**	**Overall**
Total tropospheric ozone column (DU)	46	36	37	36	39
Total ozone column (DU)	280	277	263	237	264
Ratio of total tropospheric ozone column to total ozone column (%)	16	13	14	15	15

Table 2 Decadal trends of seasonally and yearly mean ozone column from 1994 to 2013

	Total tropospheric ozone column		*Total ozone column*	
	Trend (DU per decade)	p value	Trend (DU per decade)	p value
Spring	-4.0 ± 2.5	0.13	-11.7 ± 5.4	< 0.05
Summer	+3.5 ± 1.9	0.09	-4.1 ± 3.9	0.30
Autumn	+3.9 ± 1.2	< 0.05	-2.8 ± 4.4	0.50
Winter	-0.7 ± 0.8	0.37	-5.7 ± 5.7	0.40
Yearly	+0.9 ± 0.9	0.33	-3.5 ± 3.6	0.33

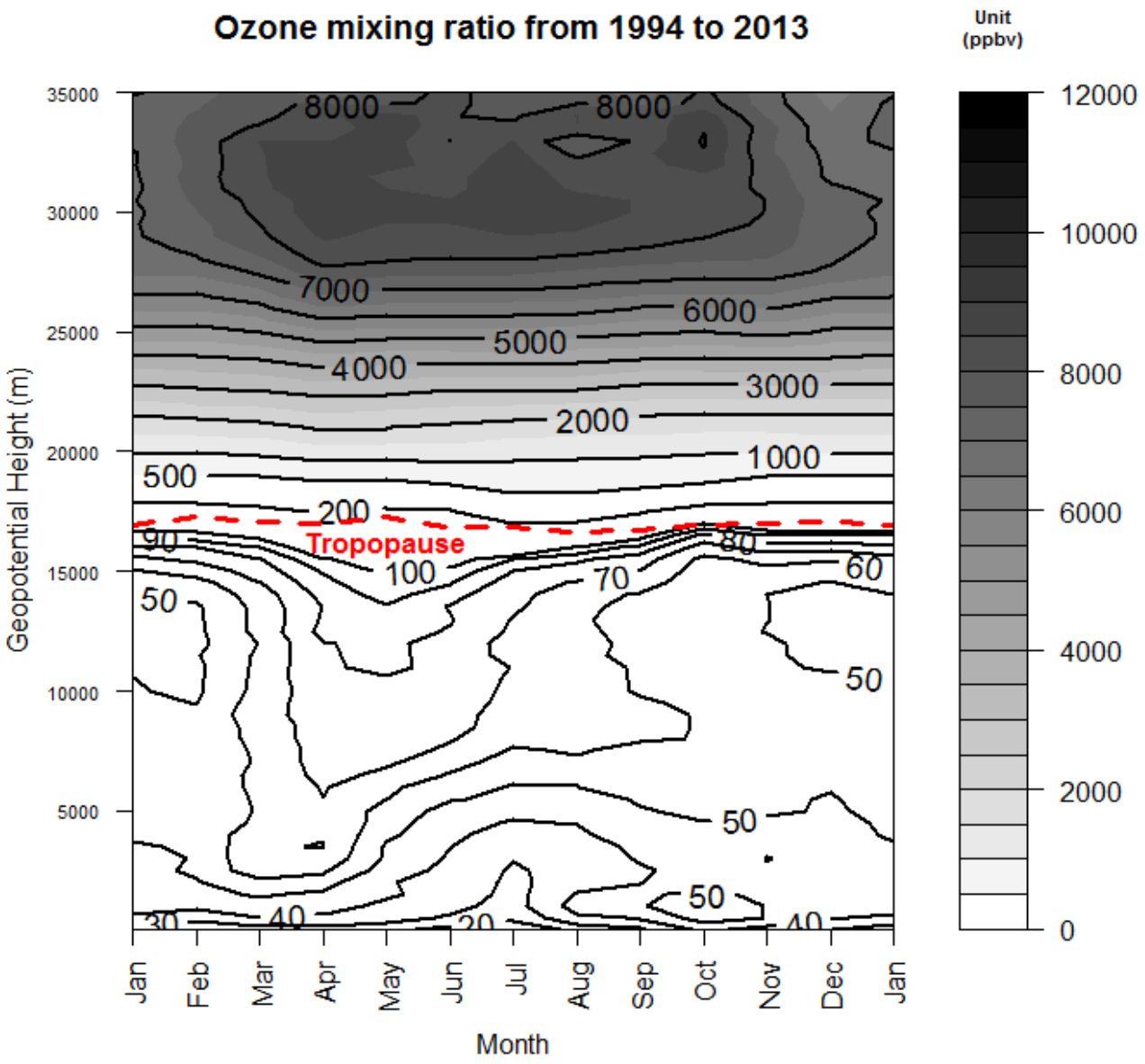

Figure 1 Ozone mixing ratio of the selected 546 ozonesondes launched between 1994 and 2013

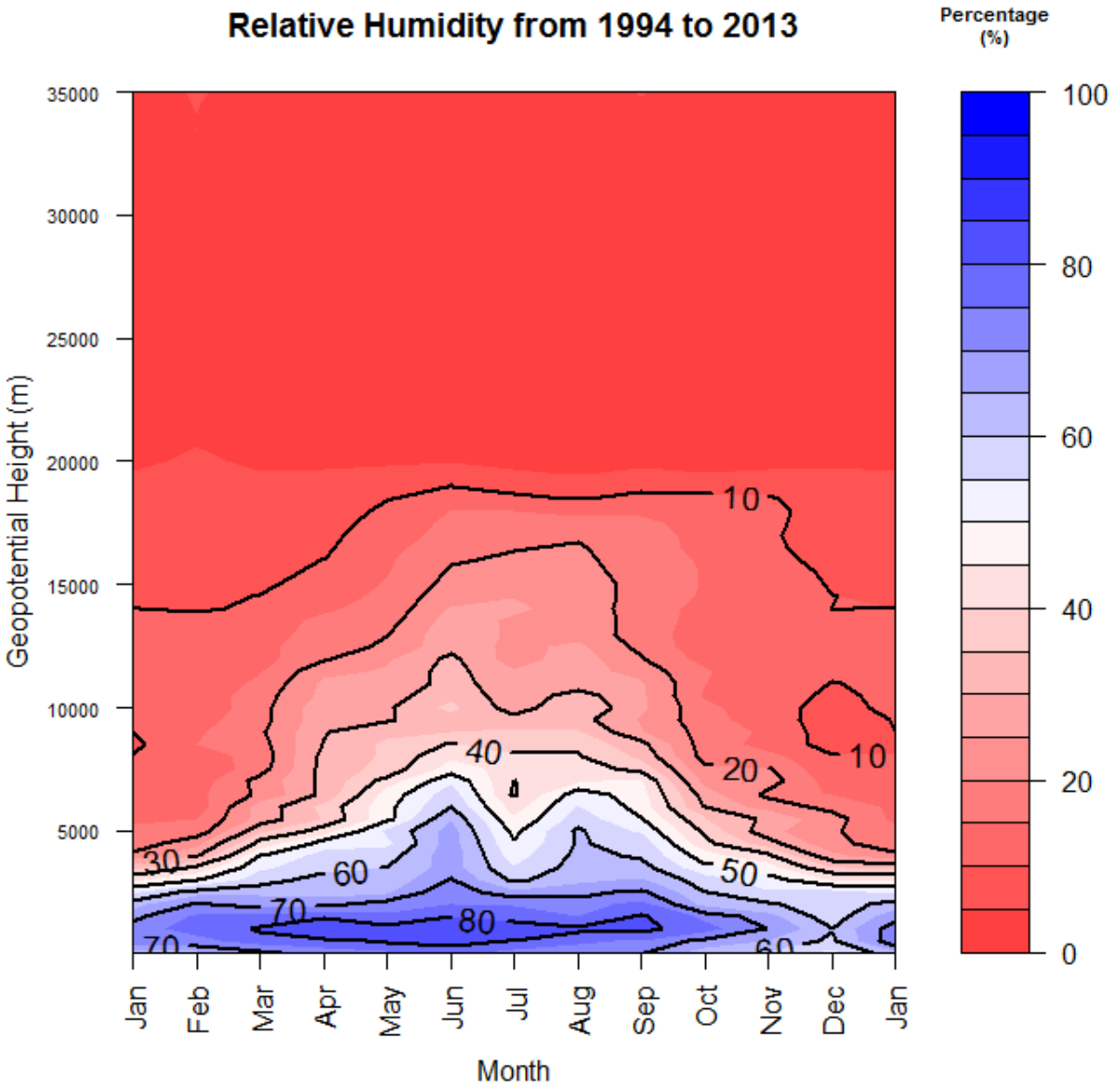

Figure 2 Relative Humidity profiles of the selected 546 ozonesondes launched between 1994 and 2013

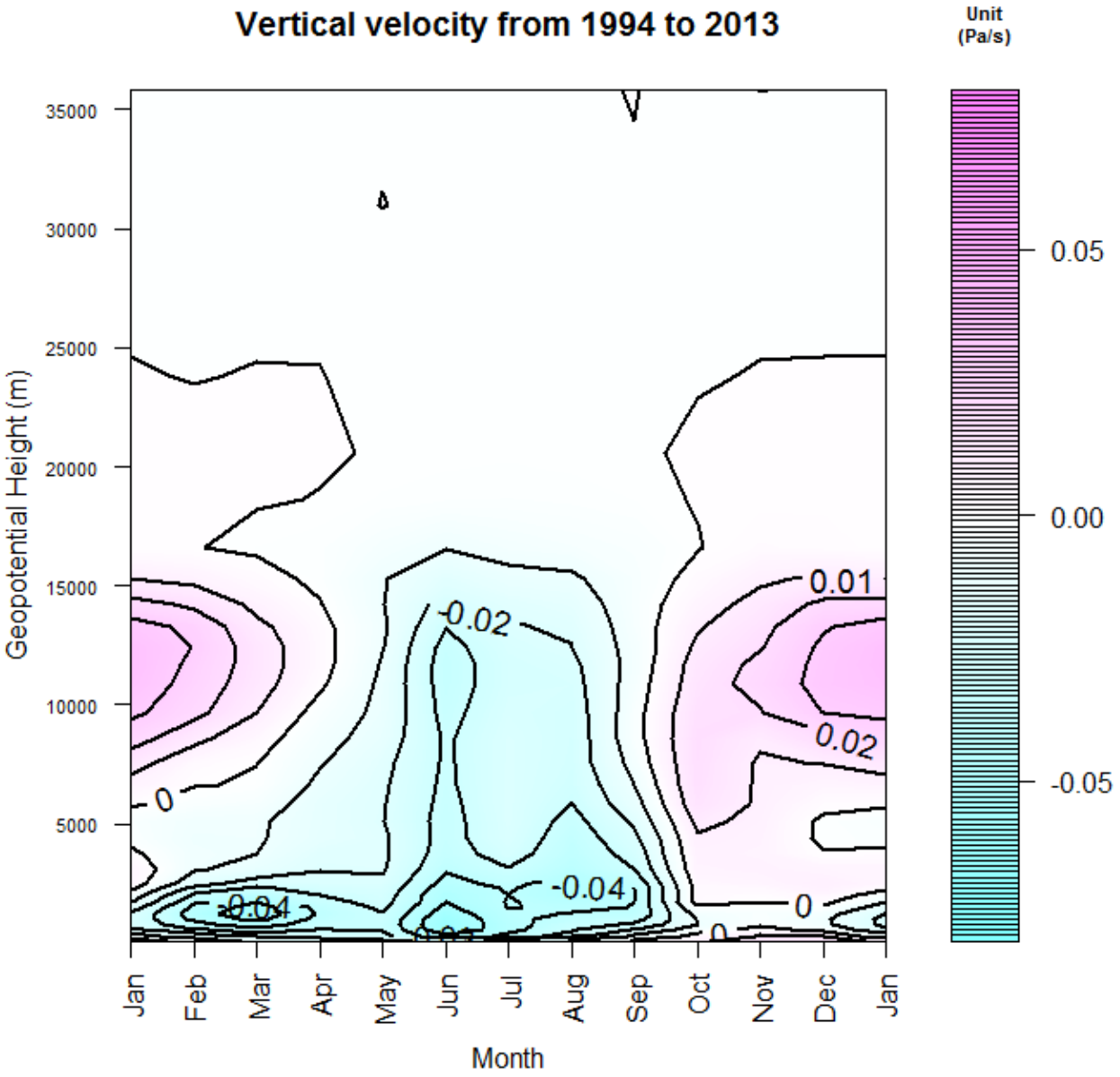

Figure 3 Monthly mean vertical velocity at 22.25°N, 114.25°E from 1994 to 2013 (positive and negative values mean downward and upward velocity respectively)

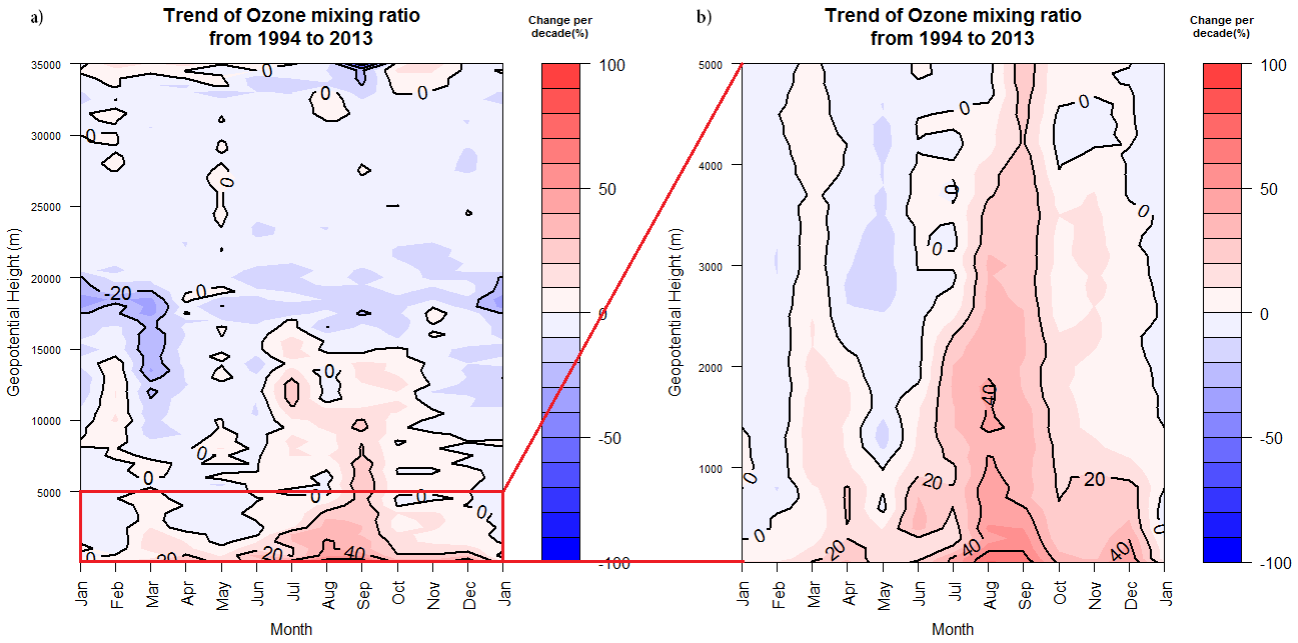

Figure 4 (a) Trend of upper-air ozone mixing ratio from 1994 to 2013, (b) Zoom-in of the lower troposphere

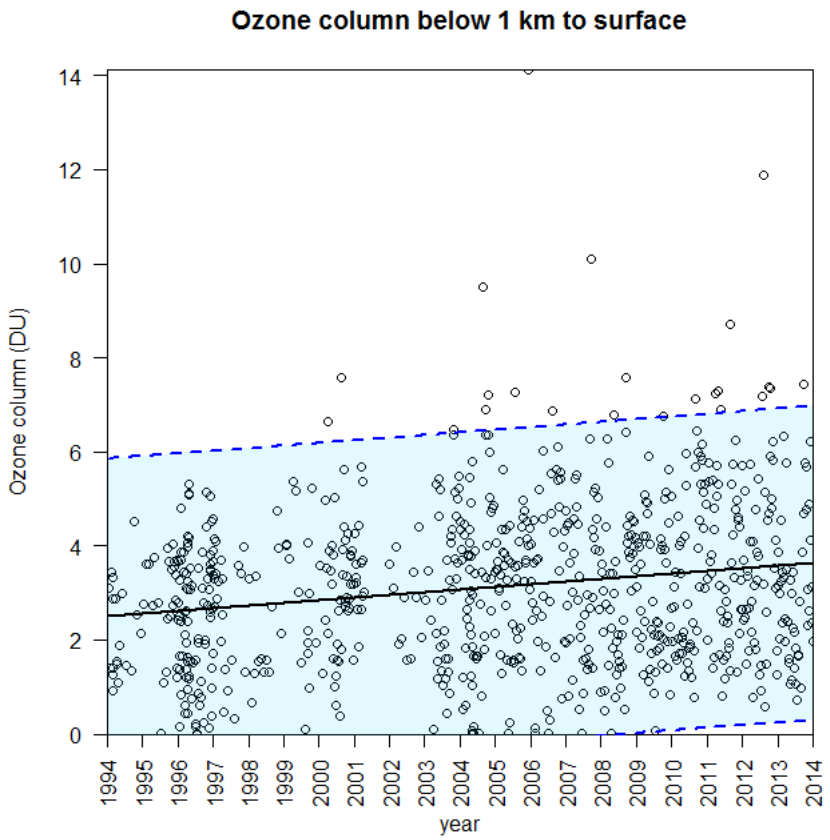

Figure 5 Time series of ozone column from surface to 1 km above ground. Shaded area represents 95% prediction level.

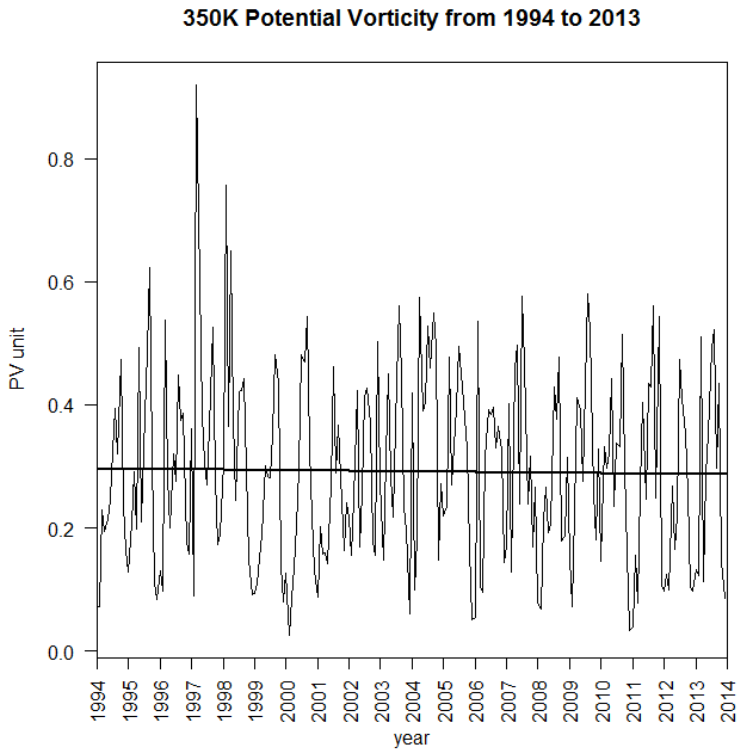

Figure 6 Time series of Potential Vorticity at 350K over 22.25°N 114.25°E from 1994 to 2013

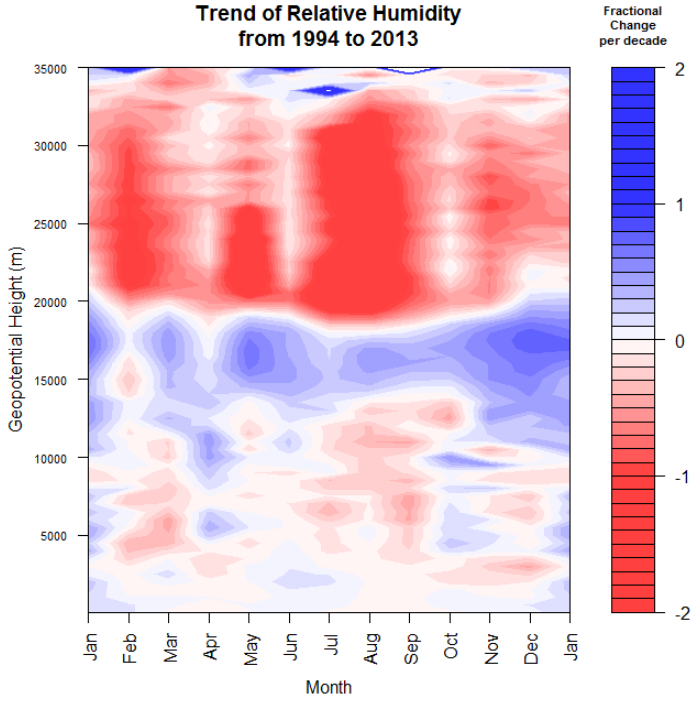

Figure 7 Trend of upper-air relative humidity from ozonesondes between 1994 and 2013

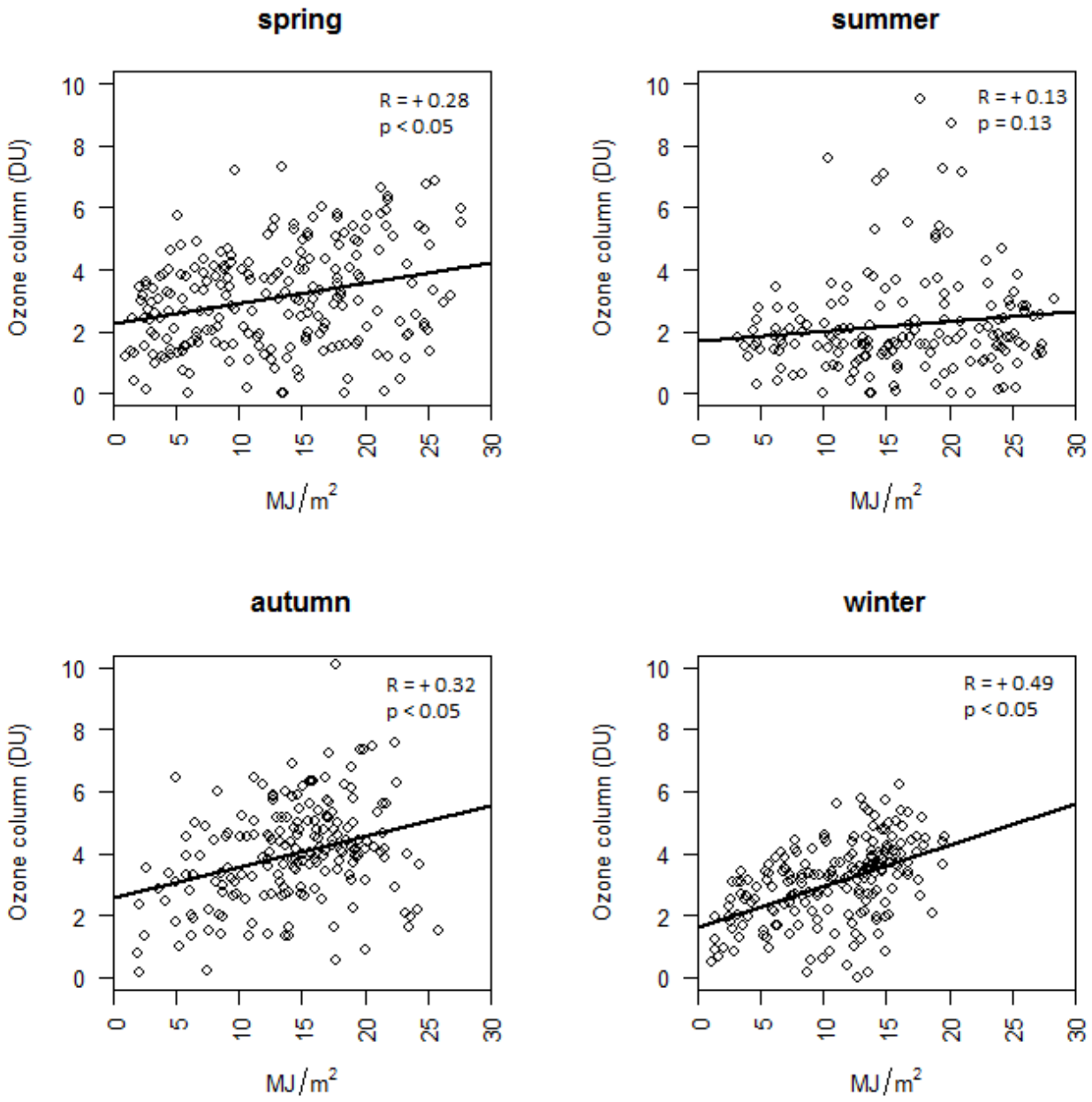

Figure 8 Correlation between boundary layer ozone and corresponding daily global radiation in four seasons for selected ozone profiles between 1994 and 2013 in the current study

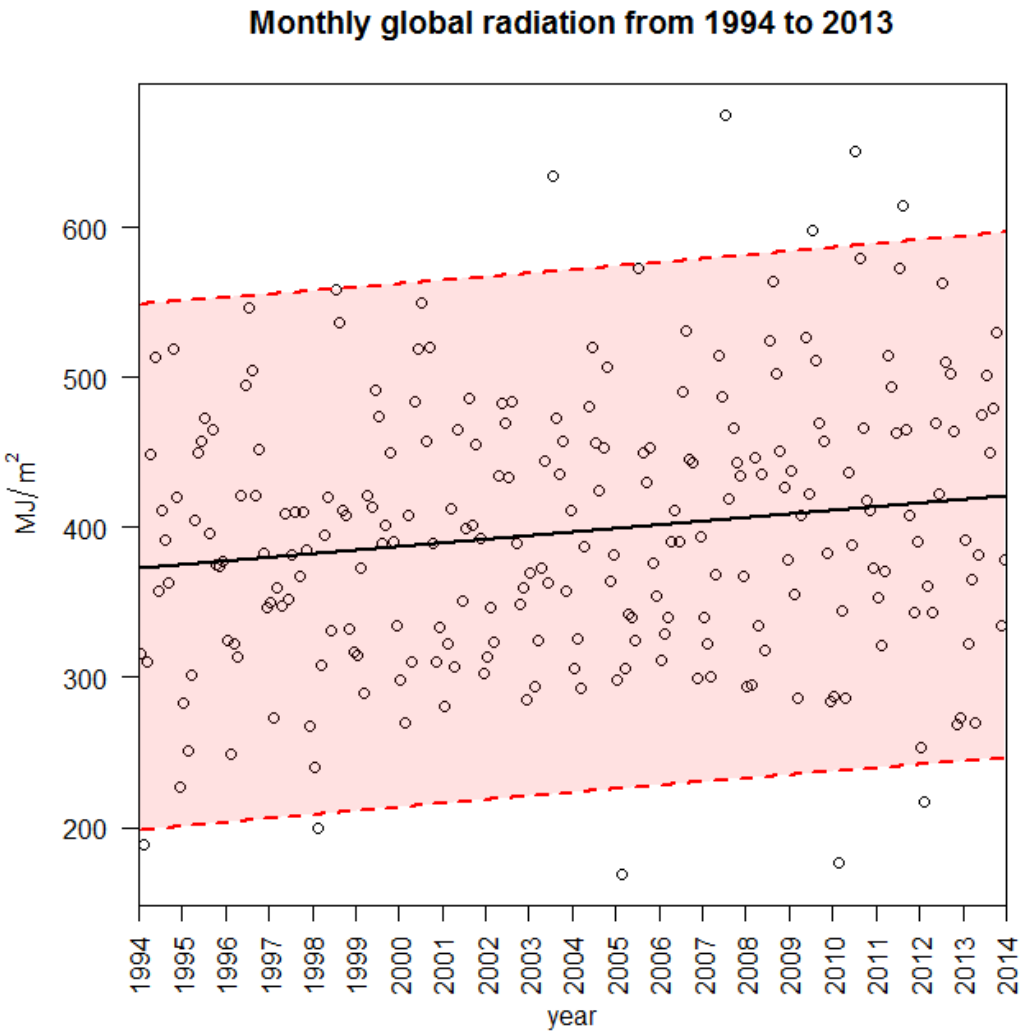

Figure 9 Time series of monthly global radiation. Shaded area represents 95% prediction level