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Research Paper

Influence of Solar Radiation, Temperature and Relative Humidity on Seasonal Variability of Ambient Air Quality at Kannur- A Tropical Site in India

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Abstract: *This study analyses the influence of solar radiation, temperature and relative humidity on tropospheric ozone concentration at Kannur, a tropical site in India, from November 2009 to December 2011 using regression analysis. The paper presents the observations of continuous measurements of surface ozone and various meteorological parameters over the site. It has been noticed that the monthly mean temperature and relative humidity were gradually increasing from 2009 to 2011. The increase in solar radiation and temperature is favorable for the photochemical generation of ozone. Ozone shown a positive correlation with both solar radiation and temperature, and negative correlation with relative humidity. The effect of these parameters on ozone concentration is critically analyzed during winter, summer, monsoon and post-monsoon seasons. Seasonally, ozone exhibits a good positive correlation with solar radiation and temperature, except in winter season and negatively correlated with relative humidity. The study also examines the diurnal effect of these parameters at two locations of different characteristics: at Kannur University Campus (KUC) at a rural location in Mangattuparamba, and at Kannur Town (KT), an urban location.*

Keywords: Air quality, Meteorological parameters, Relative humidity, Solar radiation, Surface ozone, Temperature.

Introduction

Tropospheric ozone (O_3) is an important trace gas considering its role in the oxidative capacity of the global atmosphere, climate and its impact on air quality[1]. There are various chemical and physical aspects of ozone formation. It produces photo chemically in the troposphere from reactions involving a variety of volatile organic compounds (VOCs) composed mainly of non-methane hydrocarbons (NMHCs) and nitrogen oxides (NOx) composed of nitric oxide (NO) and nitrogen dioxide (NO_2) in the presence of sufficient UV radiation from the sun[2]. The production of ozone in the troposphere occurs through the conversion of NO to NO_2 by peroxy radicals ($HO_2\cdot$ and $RO_2\cdot$), followed by photolysis of NO_2. The NOx are emitted from a variety of natural and anthropogenic sources, and through the reactions R1 to R4 produce ozone [1]:

$$HO_2\cdot + NO \rightarrow OH\cdot + NO_2 \quad \text{R1}$$
$$CH_3O_2\cdot + NO \rightarrow CH_3O\cdot + NO_2 \quad \text{R2}$$
$$NO_2 + h\gamma\ (\lambda < 420\ nm) \rightarrow NO + O\ (^3P) \quad \text{R3}$$
$$O\ (^3P) + O_2 + M \rightarrow O_3 + M \quad \text{R4}$$

where N_2 or O_2 represents M, a third body molecule required for simultaneous conservation of energy and momentum and it stabilizes the excited intermediate before it dissociates back into the reactants. In addition to precursors' levels, ozone formation is also influenced by various meteorological parameters as they are helpful in ozone formation, dispersion and dilution through transport. Meteorological processes play important role in the accumulation of ozone in the troposphere, causing large day-to-night, day-to-day, season-to-season and year-to-year variations[3].

The concentration of air pollutants in ambient air is governed by the meteorological parameters such as solar radiation, relative humidity, temperature, rainfall, wind speed, wind direction and planetary boundary layer height. Extensive studies have been done around the world on the relationship of air pollutants with meteorological parameters[4-8]. Ozone concentrations are influenced by meteorology in many ways. A large number of observations have shown that ozone concentrations increase with radiation and temperature elevation on clear days[9]. Besides, variations of ozone concentration are more intense on sunny days[10]. Water vapor has competing effects on ozone levels. It begins with the photolysis of ozone, which produces an excited oxygen atom, and an oxygen molecule (O_2). The oxygen atom can then react with water vapor to produce a hydroxyl radical, which undergoes further reactions, eventually leading to ozone formation[9].

The formation of ground level ozone depends also on the absolute concentrations of NOx and VOCs, and the ratio of NOx and VOCs[11]. An increase in temperature accelerates photochemical reaction rates[12]. Studies find a strong correlation between higher ozone levels and warmer days[12]. Prominent air pollutants are sulphur compounds, nitrogen compounds, carbon monoxide and organic compounds. These trace gas species in the atmosphere produce other secondary pollutants via thermal, chemical and photochemical pathways[13,14]. Trace gas emission from anthropogenic sources have been increasing during the last few decades and these emissions leads to local and regional air quality issues.

The influence of temperature, relative humidity and seasonal variability of ambient air quality in a coastal urban area with respect to meteorological parameters was studied by Jayamurugan et al [15]. The influence of temperature on gaseous pollutants was more effective in the summer than other seasons, negative correlations were found with humidity. Lal et al has studied on the seasonal variation in surface ozone and its precursors over an urban site in India[16]. They found in winter season, the ozone concentration is higher due to the elevated amount of precursor gases and low solar radiation.

Reddy et al has analyzed the diurnal and seasonal behavior of surface ozone and its precursors (NOx) at a semi-arid rural site in Southern India[17]. Diurnal variation of surface ozone with meteorological parameters at Kannur, India has been studied by Nishanth and Satheesh Kumar[18]. The result shows that surface ozone was higher at rural area than at urban area. Surface ozone variation in Bhubaneswar and intra co-relationship study with various parameters has been undertaken by Mahapatra et al.[19].

Increases in tropospheric ozone even at levels well below established air quality standards have implication on a more global scale for the chemistry of the lower atmosphere because ozone is a major source of the hydroxyl radical (OH·). Thus, photolysis of ozone by short wavelength UV radiation produces electronically excited oxygen atom, O (^{1}D), which rapidly react with water vapor, forming OH radicals.

$$O_3 + h\gamma\ (\lambda < 320\ \text{nm}) \rightarrow O(^1D) + O_2 \qquad \text{(R5)}$$
$$O(^1D) + H_2O \rightarrow 2OH\cdot \qquad \text{(R6)}$$

It is the hydroxyl radical which initiates the majority of organic oxidations in the troposphere, and, thus is a controlling element in atmospheric chemistry. Increased global tropospheric ozone levels will lead to increased OH production and decreased tropospheric life times of species such as methane (CH_4) and the Hydrochlorofluoro carbons (HCFCs). Thus, increased tropospheric ozone has the potential to impact stratospheric chemistry as well, via the indirect control of how much of the trace species survives to reach the stratosphere. In short, changes in ozone levels are of concern, for a variety of reasons, and hence understanding the chemistry of converting VOCs and NOx to ozone and the implications for control strategy development is critical.

Observation site and general meteorology

The location of the study is Kannur University Campus (KUC; 12.26°N, 75.39°E) at Mangattuparamba, 15 km north of Kannur town (KT), a location along the coastal belt of Arabian Sea in the west coast region of the Indian subcontinent. The location of the sampling site is shown in Figure 1. It is a rural location with no major industrial activities except a few small-scale industries including plywood and mattress manufacturing units. The air distance to the seashore is 4 km and that to the Western Ghats is 50 km. In Kannur, the four major seasons are winter (December-February), summer (March- May), monsoon (June-August) and post-monsoon (September-November). This site is located very close to the Arabian Sea and lies at a height of 5m above mean sea level. This region experiences easterly winds during the winter months and westerly winds during the summer months from March to May. The north-easterly wind starts in November. This region is strongly influenced by the marine as well as mountain environmental boosts containing high amounts of vegetation. Kannur Town (KT) (11.86°N, 75.35°E) is another site located in the city which is by the side of a national highway where the vehicular traffic is very high. Kannur town is the administrative headquarters of the Kannur district and has had industrial importance from its early days.

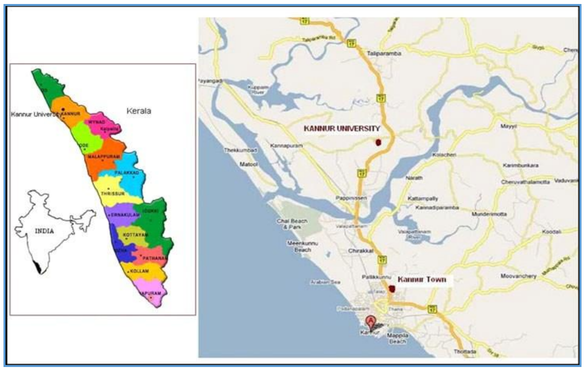


Figure 1: Location of observation sites

Data sets – solar radiation, temperature, relative humidity and surface ozone

The meteorological parameters such as temperature and relative humidity in the observational site were retrieved from the local automatic weather station operated by the Meteorological and Oceanographic Satellite Data Archival Centre (MOSDAC) established by the Indian Space Research Organization (ISRO). The data on solar radiation and concentrations of O_3 were obtained from the Kannur University Campus and Kannur Town. The total solar radiation was measured using a pyranometer installed at the local weather station of MOSDAC established by the ISRO and the data were taken at 30 minute time intervals. Continuous measurements of surface ozone are made by UV-absorption based analyser and is obtained routinely at 15-minute intervals. An analyser from Environment S.A., France (Model O3 42M) has been used for this study. The details of the analyser and its principle have been given elsewhere [20-22].

Results and Discussion

Figure 2 shows the monthly average variations of meteorological parameters like solar radiation, temperature and relative humidity and also ozone, in Kannur. Solar radiation is low during monsoon season and is most intense during late winter and summer season.

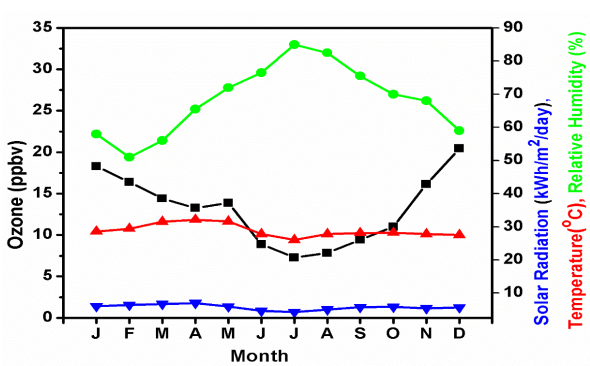


Figure 2: Monthly variation of surface ozone and meteorological parameters at KUC during the period of study

The temperature is high in the months March to May and is low during June through August. The maximum and minimum relative humidity was observed during monsoon months and winter months respectively. It is evident from the figure that ozone concentration is inversely related to humidity. That is, the maximum and minimum ozone concentration was observed during the winter and monsoon months respectively. The observations found that temperature and relative humidity show an opposite relation to each other.

Correlation of O_3 with solar radiation, temperature and relative humidity

The temporal variations of O_3 are greatly affected by the changes in local meteorological conditions such as solar radiation, temperature, relative humidity, etc. A linear correlation between monthly mean ozone and meteorological parameters like solar radiation, temperature and relative humidity during the period of observation has been made at KUC.

Solar radiation

Figure 3 depicts the scattered diagram of variation of monthly mean ozone values against corresponding monthly mean solar radiation during the period of study at KUC. Ozone exhibits a good positive correlation with solar radiation. It clearly depicts that concentrations of ozone increased with an increase in solar radiation, which also supports the fast increasing trend of the ozone concentration with high solar radiation over KUC for the said period. The correlation between ozone and solar radiation shown in the figure yields a linear relationship with a value of 0.54. The rate of increase of O_3 was 2.83149 ppbv per unit increase in solar radiation in kWh/m^2/day.

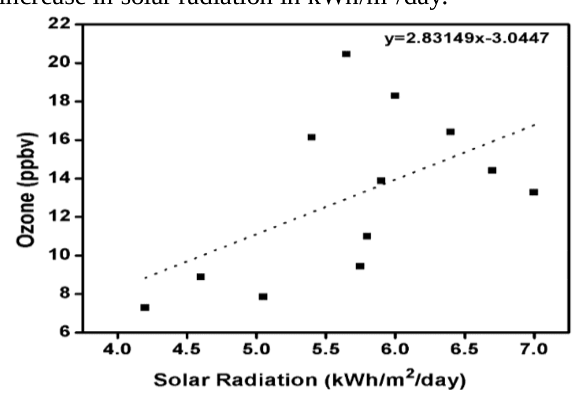


Figure 3: Regression analysis between monthly mean O_3 and monthly mean solar radiation at KUC during the period of study

Depending on the location, solar radiation can have a pronounced effect on the type and rate of chemical reactions in the atmosphere. The intensity of sunlight has an important influence on the rate of the chemical reactions that produce the smog. In the troposphere, O_3 formation is a photochemical reaction which is initiated by an attack of OH radicals on volatile organic compounds. Figure 4 represents the photochemical ozone production. These reactions are strongly influenced by solar radiations. Higher surface ozone concentrations can be related to the high intensity of solar radiation and high temperature levels which promote the photochemical generation of O_3. Similar reactions are also followed by methane (CH_4) and non-methane hydrocarbons (NMHCs) and lead to ozone formation.

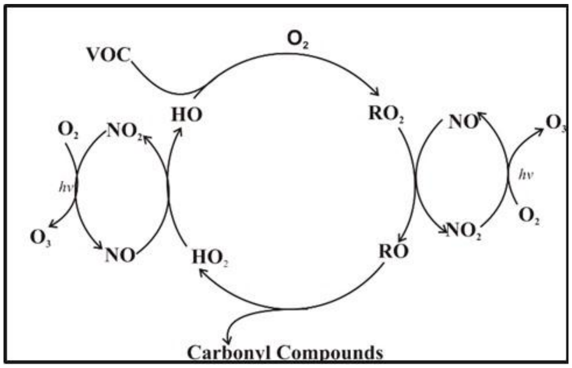


Figure 4: Schematic representation of photochemical ozone production

Temperature

Figure 5 represents the scattered variation of different monthly mean ozone values against corresponding monthly mean temperature during the period of study at KUC. Ozone exhibits a positive correlation with temperature. The positive correlation between ozone and temperature is due to the fact that the radiation controls the temperature and hence the photolysis efficiency will be higher. It clearly depicts that concentrations of ozone increased with increase in temperature, which also supports the slow increasing trend of both the ozone concentration and temperature over Kannur for the said period. The correlation between ozone and temperature shown in the figure which yields a linear relationship with a correlation coefficient 0.27. The rate of increase of O3 was 0.6090 ppbv per unit increase in temperature in °C.

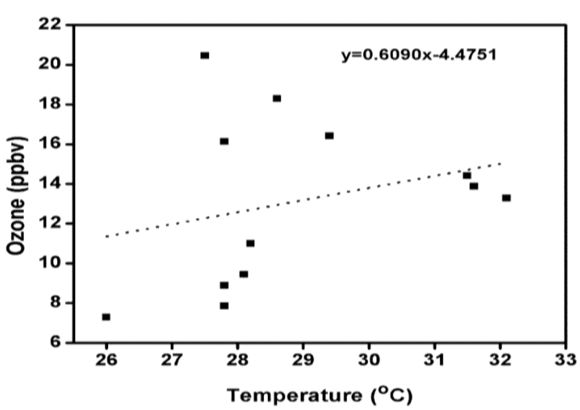


Figure 5: Regression analysis between monthly mean O_3 and monthly mean temperature at KUC during the period of study

The temperature and sunlight (solar radiation) have critical role in the chemical reactions that occur in the atmosphere to form photochemical smog from other pollutants. Ozone formation reaction is endothermic (ΔH_f = 143 kJ/mol O3). Higher temperatures will favour higher equilibrium concentration of O_3. If temperature decreases the rate of formation of O_3 will also decrease. The higher temperature enhances the production of tropospheric ozone, which in turn again increases global warming as a greenhouse molecule. The formation of ozone is also favoured the presence of oxides of nitrogen, through the following reactions R1 to R4. However, ozone levels do not always increase temperature, such as when the ratio of VOCs to NOx is low [12]. As higher temperatures will change some of the reactions, not all of the complex chemical reactions involved in ozone production in the troposphere such as those involving methane. Because of the short- lived nature of these chemical constituents, there is uncertainty to make predictions.

Relative Humidity

Ozone exhibits a good positive correlation with temperature and solar radiation and negatively correlated with relative humidity. Figure 6 depicts the scattered variation of different monthly mean ozone values against corresponding monthly mean relative humidity during the period of study at KUC. It clearly shows that concentration of ozone decreased with increase in relative humidity. The correlation between ozone and relative humidity shown in the figure which yields a linear relationship with a correlation coefficient -0.85 which is significant. The rate of decrease of O_3 was 0.339 ppbv per unit increase in relative humidity in %.

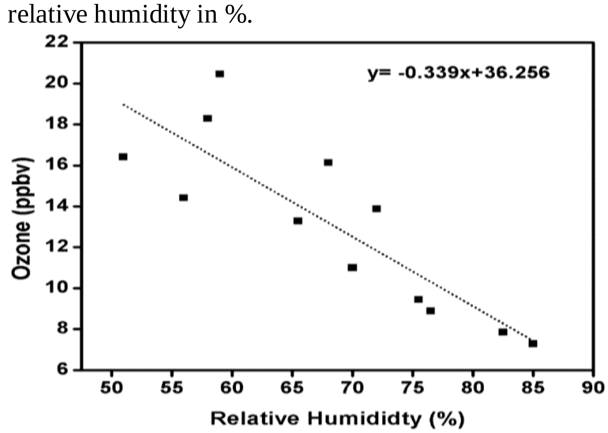


Figure 6: Regression analysis between monthly mean O_3 and monthly mean relative humidity at KUC during the period of study

The solar radiation controls the temperature, which raises the photolysis efficiency resulting in a positive correlation between ozone and temperature. Relative humidity plays an important role in affecting the series of chemical reactions which result in ozone formation. Photolysis of ozone followed by the reaction of O (^{1}D) with water vapour produces OH radical (reactions (R5) and (R6)) that is conducive to subsequent photochemical reactions resulting in further ozone production through oxidation of CO and CH_4.

$CO + OH\cdot \rightarrow CO_2 + H\cdot$
$H\cdot + O_2 + M \rightarrow HO_2\cdot + M$
$HO_2\cdot + NO \rightarrow NO_2 + OH\cdot$
$NO_2 + h\gamma\ (\lambda < 420\ nm) \rightarrow NO + O\ (^3P)$
$O\ (^3P) + O_2 + M \rightarrow O_3 + M$
Net reaction: $CO + 2O_2 \rightarrow CO_2 + O_3$

$CH_4 + OH\cdot \rightarrow CH_3\cdot + H_2O$
$CH_3\cdot + O_2 + M \rightarrow CH_3O_2\cdot + M$
$CH_3O_2\cdot + NO \rightarrow CH_3O\cdot + NO_2$
$CH_3O\cdot + O_2 \rightarrow CH_2O + HO_2\cdot$
$HO_2\cdot + NO \rightarrow NO_2 + OH\cdot$
$NO_2 + h\gamma\ (\lambda < 420\ nm) \rightarrow NO + O\ (^3P)$ (2×)
$O\ (^3P) + O_2 + M \rightarrow O_3 + M$ (2×)
Net reaction: $CH_4 + 4O_2 \rightarrow CH_2O + H_2O + 2O_3$

In these reactions, carbon dioxide and hydrocarbons provide the peroxy radicals through their oxidation and are consumed in the process, while NOx is conserved and thus acts as catalyst in ozone formation. Instead of methane (CH_4), other organic compounds (NMHC, VOC) can also be participating in this reaction chain, where carbonyl species or a ketone formed next to the ozone. But the negative correlation between ozone and relative humidity at the present measurement site suggests that the chemical loss of ozone, following the reactions given below, is the dominant process during high humidity[23].

$O_3 + h\gamma\ (\lambda < 320\ nm) \rightarrow O\ (^1D) + O_2$
$O\ (^1D) + H_2O \rightarrow 2OH\cdot$
$O_3 + OH\cdot \rightarrow HO_2\cdot + 2O_2$
$O_3 + HO_2\cdot \rightarrow OH\cdot + 2O_2$

Moreover, higher humidity levels are associated with greater cloud abundance, atmospheric instability and low incoming solar radiation, slowing down photochemical processes. Also the surface ozone is depleted through deposition of its molecules of water droplets, which are higher during high humidity levels, hence the O_3 concentration has a strong dependence on humidity in this location.

Seasonal correlation of O_3 with solar radiation, temperature and relative humidity

In the troposphere, ozone act as a secondary pollutant, which depends on regional topography and climate. The Indian Meteorological Department (IMD) designated India into 4 seasons: winter, summer, monsoon and post-monsoon. Winter occurred from December to February, summer lasting from March to May, and May is the hottest month of the year. Monsoon or rainy season enduring from June to September, and contains lots of humidity. Post-monsoon season lasts from October to November, followed by winter. The seasons are the main parameters which affect the concentration of ozone naturally. Other than this, meteorological parameters like solar radiation, temperature and relative humidity are also responsible for ozone variation throughout the season. Seasonally, ozone exhibits a good positive correlation with solar radiation and temperature, except in winter season and negatively correlated with relative humidity. The correlation coefficient between seasonal O_3 concentration with solar radiation, temperature and relative humidity were 0.75, 0.37 and -0.99 respectively, at KUC during the period of observation.

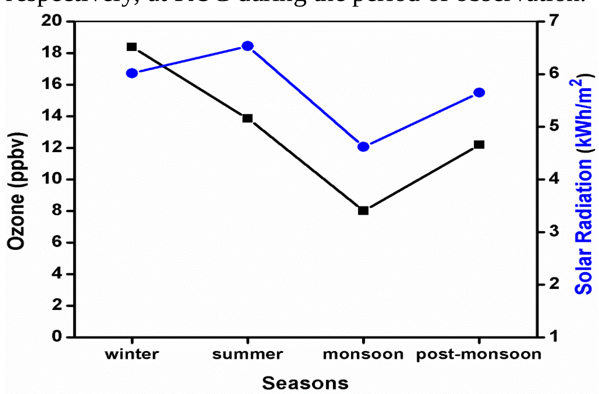


Figure 7: Seasonal correlation of O_3 with solar radiation at KUC during the period of study

It is observed in figure 7, 8 and 9 that higher levels of surface ozone are observed in winter and summer seasons as compared to monsoon and post monsoon season. The highest levels of ozone at summer season are attributed to maximum photochemical reactivity taking place due to maximum solar radiation. On the other hand, very high levels of ozone are observed in winter season, although photochemical activity is considerably reduced in summer season. The highest O_3 observed in winter is attributed to a lower mixing height resulting in the trapping of pollutants near the earth's surface due to temperature inversion[24].

In winter season, the ozone concentration is higher due to the elevated amount of precursor gases and the maximum sunshine hours of more than 9-10 hours/day due to the clear sky. Another possible reason for this could be the long range transport of ozone and its precursors, in addition to its photochemical formation. It is obvious that the air mass movements during the winter and post monsoon months were from east of this site and confined from the west during the summer and monsoon seasons. It is further observed that air masses appear to originate from the eastern part of Kannur from where transports of pollutants from nearby industries occur. There is also a possibility of enhanced transport of ozone from the stratosphere during the winter season [25-28].

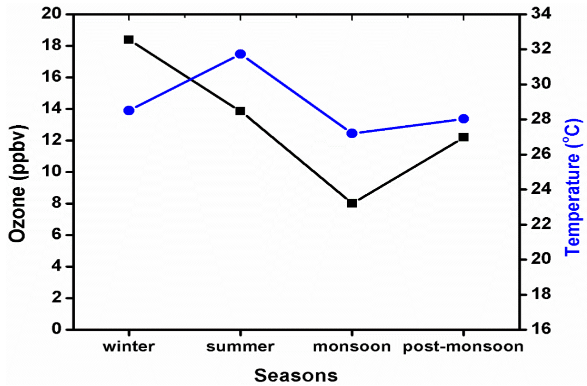


Figure 8: Seasonal correlation of O3 with temperature at KUC during the period of study

The highest levels of ozone at summer season are attributed to maximum photochemical reactivity taking place due to maximum solar radiation. During summer and monsoon seasons, the movement of air masses originated over the Arabian Sea and marine air mass was relatively clean compared to the continental air. Owing to the oceanic influence, the air masses enriched with hydroxyl radicals (OH) may trigger the removal of ozone to a larger extent. This may be one of the reasons, for the reduction of ozone observed in summer season at this location in spite of high solar radiation. The convective motion and increased cloud cover over this location during summer induced a relatively small mixing ratio observed in summer than in winter.

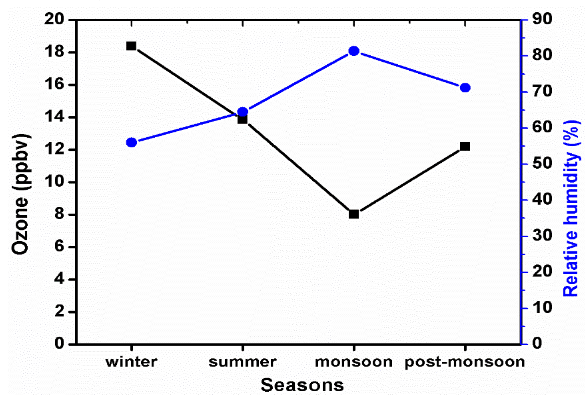
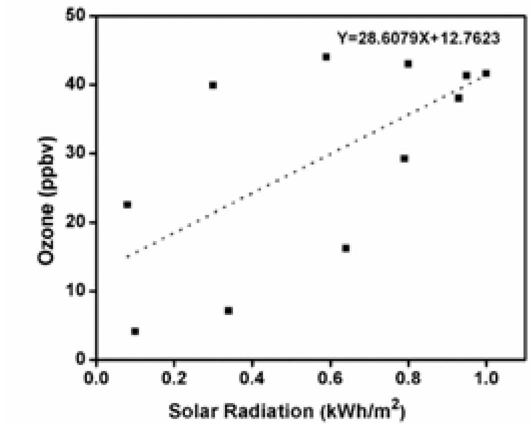


Figure 9: Seasonal correlation of O_3 with relative humidity at KUC during the period of study

A relatively low amount of surface ozone was observed during monsoon seasons compared to summer and winter. During the monsoon seasons, there is low solar radiation due to the cloud cover, low temperature and high relative humidity, which reduce the ozone concentration at this season. The maximum humidity was measured during monsoon and its minimum was observed in the winter months.

Diurnal correlation of O_3 with solar radiation, temperature and relative humidity - their comparison at rural and urban areas of Kannur

The above seasonal study of Kannur shows higher ozone concentration in winter. For this comparative study, the variations of meteorological factors and surface ozone observed during winter months in 2009 and 2010 have been used. This study carried out at two locations of different characterization: at Kannur University Campus (KUC), a rural location and at Kannur Town (KT), an urban location.



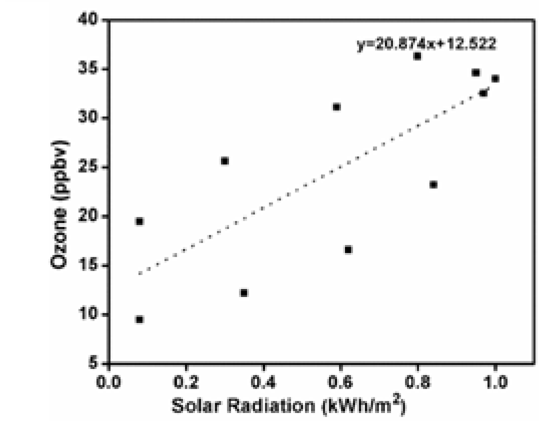


Figure 10: Regression analysis between O_3 and solar radiation at KUC and KT during the period of study

A linear correlation between O_3 and meteorological parameters like solar radiation, temperature and relative humidity during the period of observation has been made at KUC and KT, and the correlation graphs are shown in the following figures 10, 11 and 12. Diurnally, surface ozone exhibits a good positive correlation with solar radiation and temperature and negatively correlated with relative humidity. At KUC and KT, the correlation between O_3 and solar radiation, temperature and relative humidity were 0.65 and 0.76, 0.6862 and 0.6871, and -0.5138 and -0.3085 respectively. It is realized that solar radiation at the rural area (KUC) is less than the urban area (KT), the difference may be due to cloud cover. Temperature in the rural area (KUC) is slightly less than the urban area (KT), the difference may be due to the slight variation in solar radiation and also due to less amount of air pollution. But relative humidity in the rural area (KUC) is greater than the urban area (KT). Higher humidity levels are associated with atmospheric instability and large cloud cover, the photochemical process is slow and the surface ozone is depleted by deposition on water droplets in this location, hence the ozone concentration has a strong dependence on humidity. The correlation between O_3 and humidity is more noticed at KUC since it is a rural location with less amount of pollution.

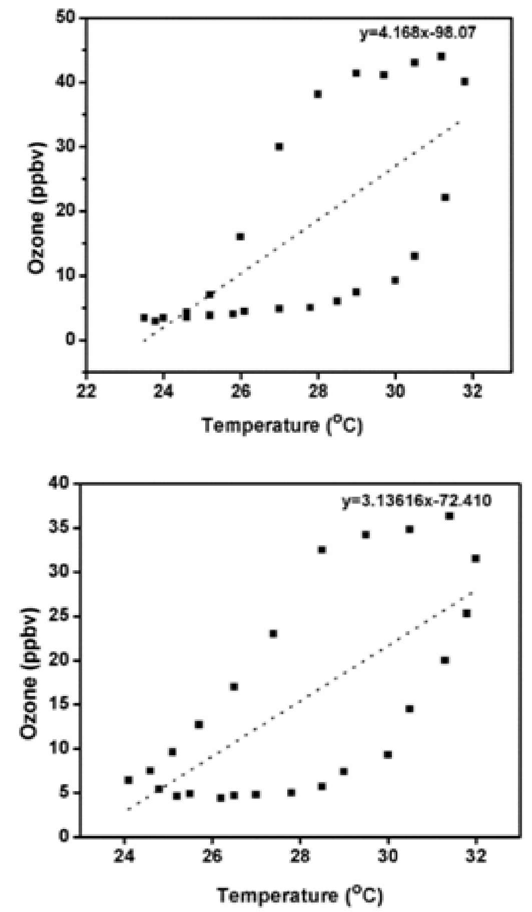
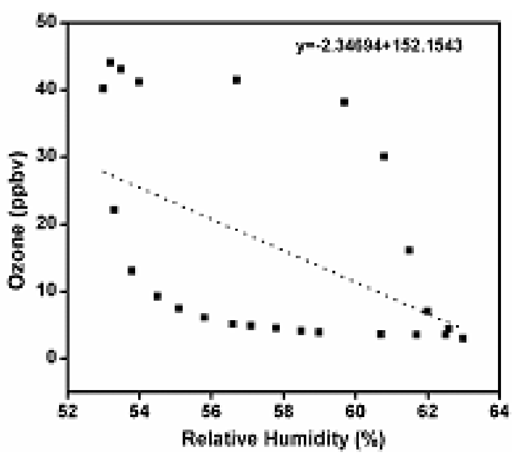


Figure 11: Regression analysis between O_3 and temperature at KUC and KT during the period of study



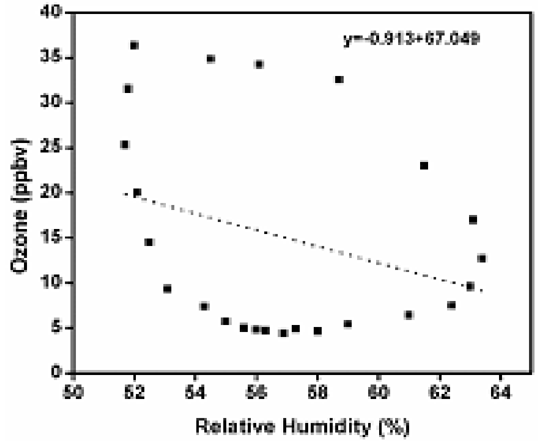


Figure 12: Regression analysis between O_3 and relative humidity at KUC and KT during the period of study

The observations revealed that surface ozone abundance is higher at rural area (KUC) than that at urban area (KT): Two mechanisms have been proposed to account for the higher surface ozone concentration at rural area[29,30]. One is the direct transport of ozone from urban areas and the other is the transport of its precursors NOx and NMHCs, followed by in situ photochemical ozone production. The enhanced O_3 concentration observed at KUC, during the winter season was mainly due to the easterly airflow that favours advection of precursors from inland locations that induces active photochemistry, in addition to in situ photochemical production of O_3.

Conclusion

Energy is a pre-requisite for chemical reactions. Energy can appear in many forms, such as: light, heat, kinetic, etc. We examined the effect of heat (temperature), light (solar radiation), presence of reactant such as water vapour (humidity) for the production and destruction of ozone, which is a tracer in the atmosphere in air quality studies, since it is produced from air pollutant gases. Tropospheric ozone is a major concern of research because it is an important constituent of photochemical smog, which is harmful for plants and animals Ozone is a secondary pollutant that is not emitted directly by sources, but is formed in the atmosphere through reactions among the primary pollutants like VOCs and NOx in the presence of solar radiation. Ozone helps to produce hydroxyl (OH) radicals, which destroy pollutants through oxidation. It is the hydroxyl radical, which initiates the majority of organic oxidations in the troposphere, and, thus is a controlling element in environmental chemistry. Ozone exhibits a good positive correlation with temperature and solar radiation and negatively correlated with relative humidity. From this study, it is revealed that there is a significant influence of humidity on the production and removal of ozone in

Kannur. By this study, it is evident that O_3 exhibits a distinct seasonal variation over this site, which may not only be controlled by solar radiation, but also by atmospheric dynamics. The results of this study are preliminary and need confirmation with more observations to explore the O_3 chemistry. Studying the interaction between ozone and climate change and predicting the consequences of change, requires enormous computing power, reliable observations and robust diagnostic abilities. The efficiency of future research depends on an integrated strategy, with more interactions between scientists, observations and mathematical models.

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