MONITORING FORMALDEHYDE CONCENTRATION OVER ISLAMABAD USING GROUND BASED AND SATELLITE OBSERVATIONS

Javeria Abbas, Muhammad Fahim Khokhar, and Muhammad Arshad

***Abstract*— Traffic is one of the most important sources of high pollution levels in major cities of Pakistan and therefore the population is prone to high exposure in the ambient environment. Formaldehyde (HCHO) emitted by anthropogenic and biogenic activities is an intermediate product in the oxidation pathways of non-methane hydrocarbons. This study is designed to monitor the HCHO concentrations over Islamabad, Pakistan. Mini MAX-DOAS (Multi Axis – Differential Optical Absorption Spectroscopy) instrument was used to perform ground-based measurements at IESE (Institute of Environmental Sciences and Engineering), NUST (National University of Sciences and Technology) Islamabad (33.6416° N, 72.9835° E) Pakistan. The spectra were acquired for a time period of six months i.e. October 2013-March 2014. Analysis was performed to retrieve HCHO Differential Slant Column Densities (DSCD). Tropospheric vertical column densities (VCD) of HCHO over Islamabad were derived from measured DSCD by using geometric air mass factor approach. Mean Tropospheric HCHO VCDs were found to be 4.32×10^{16} molecules/cm^2. Satellite observations from Ozone Monitoring Instrument (OMI) were used for comparison with the ground-based mini MAX-DOAS observations. Tropospheric HCHO VCDs derived from mini MAX-DOAS measurement presented a fair agreement with the satellite observations over Islamabad, Pakistan. However, satellite observations underestimate the concentration of HCHO by 1 degree. Further, wind data was used to find out the possible pollutants transport corridors over IESE-NUST. Air masses coming from Kashmir highway and Golra Mor were the main source of HCHO pollution over monitoring site.**

***Terms*— Formaldehyde, MAX-DOAS, Troposphere, Urban Air Pollution,**

I. INTRODUCTION

Formaldehyde (HCHO) is considered as one of the most plentiful hydrocarbons in our atmosphere. It plays a very significant part in altering the chemistry of troposphere. It is a very vital indicator of the emissions of non-methane volatile organic compound (NMVOC) including the photochemical activity as well. HCHO is a prime emission product from fossil fuel combustion and biomass burning. The main source in our atmosphere is the photochemical oxidation of both methane and non-methane hydrocarbons. The core removal processes of HCHO during the day are; oxidation by OH radicals (that yields $CO+HO_2+H_2O$) and the most important two photolysis pathways (yielding $CO+H_2$ and $CO+2HO_2$).
Being the main source, methane oxidation is the primary producer of more than half of the worldwide production of HCHO NMVOC oxidation constitutes the remaining production. (Stavrakou *et al.,* 2008). Therefore, the satellite measurements of HCHO can be useful to control and limit NMVOC emissions in current advanced chemical transport models [1-5]. Presently, UV backscatter satellite sensors are used for observing tropospheric formaldehyde [6-13] also from airborne and ground-based in situ instruments [14-15]. Ground-based remote-sensing Fourier transform infrared spectrometers are also used [16-17]. Active and passive DOAS can also be used to measure CH_2O in the UV (see Platt and Stutz, 2008). The novel MAX-DOAS technique has been effectively used for detecting several gases, these include BrO, HCHO, NO_2, SO_2 and CHOCHO [18-24].

Principally, differential absorption structures are used in the MAX-DOAS techniques. These structures of the oxygen collision complex present in the visible wavelength, (O_2-O_2 or O_4) derive information about aerosols. For conducting long-term and automated measurements in a steady method, MAX-DOAS is appropriate. MAX-DOAS measurements can also produce noteworthy information about numerous significant trace gases, such as formaldehyde (CH_2O), nitrogen dioxide (NO_2) and glyoxal [19-21, 25], and therefore have a various possible and important applications for understanding the complex Earth systems.

There is a dire need to explore the ambient air quality in the region especially when bearing in mind that the economic base of Pakistan is being shifted from agriculture to industry This study is considerably important as there is a dire need of air quality baseline data in Pakistan. This will provide a baseline for pollution monitoring in the country. This study mainly focuses on the measurement of formaldehyde (HCHO) column densities over Islamabad. This measurement is done using Max-DOAS. The mentioned instrument was mounted on the roof top of Institute of Environmental Science and Technology, IESE, sector H-12, Islamabad. (latitude =33.6416° N, longitude= 72.9835° E). Satellite data was acquired and the results were then compared with the ground based observations.
This paper is structured as; Section 2 provides the information about the instrument, the recorded data and the satellite that was used for data retrieval and satellite data acquisition. The results are discussed in the Section 3.

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F. Javeria Abbas is with the Institute of environmental sciences and engineering, National University of Sciences and Technology Islamabad, Pakistan (j.abbas280@gmail.com).

S. Muhammad Fahim Khokhar is Assistant professor at , National niversity of Sciences and Technology Islamabad, Pakistan and he is also a Visiting scientist, Satellite Group Max-Planck Institute for Chemistry Mainz, Germany (Tel.: +92-51-90854308, email:fahim.khokhar@iese.nust.edu.pk).

T. Muhammad Arshad is associate professor at Institute of environmental sciences and engineering, National University of Sciences and Technology Islamabad, Pakistan (e-mail:marshad@iese.nust.edu.pk)

II. Data Analysis

A. Study site

The mini max-DOAS instrument was located at the Institute of Environmental Science and Engineering (IESE) at the National University of Science and Technology, (NUST), H-12 Campus, Islamabad, Pakistan. (Latitude: 33.6416° N, longitude: 72.9835° E). The instrument was faced towards the South-East direction. This point towards the industrial Sector of the region, Sector I-10. Sector I-9 and I-10 are the main industrial regions with marble, steel, oil, electrical and textile industries.

Kashmir highway, the main express highway divides Islamabad from West to East. This highway passes through almost all the sectors of the city with major portion of vehicular traffic. This Kashmir highway passes just 500 m away in the North of our monitoring site. It connects to N5 highway in the WEST. The Islamabad Junction Principal (IJP) road, carries both LTV and HTV traffic and passes on south-east of the monitoring site.

B. Instrument

MAX-DOAS instrument is a light weighed and fully automated spectrometer. It is specially intended for the appropriate spectral analysis of scattered sunlight [18]. The dimensions are 13cm × 19cm × 14cm. The entrance optics is controlled by a closed aluminum box. A fiber coupled spectrograph with the controlling electronics resides in the box. A stepper motor controls the elevation of viewing angles. A 40 mm focal quartz lens is present at the entrance optics. For light dispersion, a crossed Czerny-Turner spectrometer (USB2000+, Ocean Optics Inc.) having a spectral range and resolution of 320-465 nm and 0.7 nm respectively is used. The detector used in the instrument is a one-dimensional CCD (Sony ILX511 with 2048 individual pixels). DOASIS software installed on a laptop with Windows XP operating system was used for field measurements.

C. Analysis setting for HCHO

Formaldehyde exhibits absorptions in the UV region ranging from 240 to 360 nm. For the retrieval of HCHO only the bands above 320 nm are usually used due to amplified absorption of ozone below 320 nm. The HCHO differential slant column densities (DSCDs) are retrieved from the measured spectra in UV region by using the DOAS technique. (Platt and Stutz, 2008). The fitting interval of wavelength is generally enhanced to (a) maximizing the sensitivity to HCHO, (b) minimizing the scatter and fitting residuals of the retrieved HCHO slant column densities, and (c) minimize the interference with other present absorbers.

The measured spectra were analyzed by using the DOAS technique [26]. A wavelength analysis window for HCHO retrieval was 323.5–355 nm. For DOAS fit the used trace gas absorption cross sections were; HCHO at 298K [27] NO_2 at 294 K [28], O_3 at 223, 243K [29], O_4 with a Fraunhofer reference spectrum (highly resolved solar spectrum), Ring spectrum (calculated from the Fraunhofer spectrum by using DOASIS) and a fifth order polynomial was used in this spectral fitting process. Fig. 2 displays the trace gas absorption cross sections that are considered vital in the 323.5-355 nm wavelength region. It is noteworthy that all absorption cross sections used were convoluted to the resolution of the instruments by using instrumental slit function. In this analysis the red lines are the measured intensities whereas the black lines are the calculated intensities.

D. Measurements at IESE-NUST

The measurement started from October 2013 to March 2014. These measurements were performed on continuous basis from dawn till dusk. The solar irradiance spectra were recorded at various angles. These were noted at the viewing elevation angles of 1°,2°,4°,5°,6°,10°,15°,30°,45°,90°. The period of a discrete measurement was about one minute (60 sec). The temperature was set at 16°C for all measurements. A root mean square (RMS) of 2.7×10^{-3} was applied to the retrieved differential slant column densities (DSCD) to filter the values qualitatively. Retrieval is strongly sensitive to temperature of the detector (a temperature variation of 0.1^{o} C can cause RMS of 1×10^{-4}, for details see Coburn et al., 2011).

An example of retrieves HCHO DSCD (mol/cm^2) is shown in the Fig. 3. The complete set of elevation angles were taken at a cloud free day of 16 October, 2013. For smaller elevation angles the retrieved DSCDs are higher than the DSCDs measured for the larger elevation angles. This clearly indicates that the HCHO columns were higher close to the ground i.e. the pollution sources and decreases as we move away from the ground. The vertical column densities (VCDs) were derived from the derived DSCD by using geometric air mass factor (AMF) approach [30].

E. Calculation of Tropospheric VCD

Differential air mass factor (DAMF) is used to convert measured DSCDs into VCD_{trop}. This DAMF is defined as the difference of air mass factor (AMF) when viewing angle i.e. $\alpha \neq 90^\circ$ and $\alpha = 90^\circ$. The Air Mass Factor for the off-axis view and zenith can be assessed as 1 and 1/sin α, respectively. Therefore, the tropospheric VCD of a trace gas can be written as:

$$VCD_{geo} = DSCD\,(\alpha)\,/DAMF(\alpha) \qquad \rightarrow \text{eq. 1}$$

$$VCD_{geo} = DSCD\,(\alpha)/(1/\sin\alpha)-1 \qquad \rightarrow \text{eq. 2}$$

F. Satellite Data

OMI onboard NASA’s EOS Aura satellite, is orbiting at 705 km in a Sun-synchronous orbit at an inclination of 98° and 13:38 local equator crossing time [31]. Aura provides 14 to 15 orbits having a period of approximately 99 min with a global coverage on daily basis. The Ozone Monitoring Instrument is a UV/visible nadir solar backscatter CCD spectrometer. It covers a spectral range 270–500 nm with resolution of 0.45 nm that lies between 310 and 365 nm with a swath angle of 114° and covers 2600 km area on Earth. Its 60 pixels have footprints ranging from 13 × 24 km^2 at nadir to 28 × 160 km^2 at swath edges.

A combination of algorithms together with TOMS version 7, DOAS, Hyperspectral BUV Retrieval and forward modeling were used to extract the various OMI data products. The HCHO products distributed under TEMIS project are based on scientific developments being performed at BIRA-IASB Brussels Belgium. In this study, OMI HCHO tropospheric

VCDs level-2 product from TEMIS website (www.temis.nl) were used. Daily data was downloaded and then processed. Gridded data is used to a latitude-longitude grid size of 0.25 by 0.25 degrees. For detailed description of OMI products can be referred [31-32].

III. Results and Discussions

A. Temporal Analysis

Tropospheric HCHO VCDs retrieved from mini Max-DOAS measurements performed from October 2013 till March 2014, are shown in Fig. 4. Time series is presenting daily mean HCHO VCD and black vectors are indicating the mean wind direction for that particular day. Peaks in HCHO VCD indicated very interesting behavior and role of meteorological parameters especially wind direction. These high peaks were mainly induced by favorable wind direction and speed causing the polluted air masses being transported to IESE-NUST site. The peaks in HCHO columns were observed when the wind direction was WNW, NE and WSW. It can be clearly seen that these high peaks are resulted from HCHO pollution being transported from the Kashmir Highway and Golra Moar. Fig. 4 shows the daily averaged temporal variation of HCHO VCDs (molecules/cm^2 on left hand side) and the red lines representing the mixing ratios (ppbv on right hand side) over IESE NUST (October 2013-March 2014) and comparison of HCHO levels with existing WHO standards (WHO-limit-doted red line)

HCHO VCDs were converted into HCHO mixing ratios in the ambient air. It was assumed that the HCHO is located close to the ground surface near to the emission sources. Assuming that HCHO is confined in a box profile between the surface and 500 m. The observed HCHO VCDs were converted into number density by dividing the assumed vertical height:

Firstly the observed HCHO VCD were converted into number density by dividing with assumed vertical height

HCHO Number Density (molec./cm^3) = HCHO VCD (molec./cm^2) / 50000 cm

The calculated HCHO number density can be directly converted into NO_2 mixing ratios according to the following formula:

HCHO mixing ratio (ppbv) = HCHO Number Density (molec./cm^3) / 2.503 10^{10} (molec./cm^3)

The concentration of observed HCHO was higher than the limits prescribed by WHO i.e. around 83 ppbv.

B. Diurnal Profile of Formaldehyde

Fig. 5 shows the average diurnal profile of HCHO over the study site for the time period October 2013-March 2014. The concentration of HCHO is high in the morning as the solar intensity and rate of photolysis is low but the concentration reduces during the noon due to high photolysis rate and it increases again in the evening. The increase in concentration is also attributed to rush hours and traffic during morning and evening and due to the oxidation of NMVOCs.

C. CNG Availability and Formaldehyde

Pakistan is the world’s largest Compressed Natural Gas (CNG) consuming country. More than 3,000 CNG stations are in service in 99 different cities and towns of the country and more than 1000 would be installed in the next two years. A surge in the number of CNG-fuelled vehicles aggravated the winter CNG shortage in Pakistan.

Fig. 6 displays the weekly cycle of formaldehyde concentration over Islamabad for the time period of October 2013-March 2014. CNG is methane stored at high pressure. As mentioned earlier, the key source of Formaldehyde production is the oxidation of Methane and non-methane volatile organic compounds (NMVOC). In this study we tried to create a link between CNG availability and formaldehyde concentration over the study area. CNG was available only during two days in the week i.e. Monday and Thursday. From the bar chart it can be seen that the CNG was available on Monday and hence we get high concentration of HCHO on Tuesday. The concentration of HCHO declined the next day i.e. Wednesday, this can be attributed to the short lifetime of the trace gas. The CNG was again available on Thursday and we can see high concentrations on Friday and the trend continues. There are other sources of HCHO as well, here we just made an effort to develop a link between HCHO and CNG because methane is the main driver of HCHO production.

D. Mini MAX-DOAS Comparison with OMI Observations

Mini MAX-DOAS observations have been compared with satellite observations in various studies conducted worldwide [30].

In the current section, tropospheric HCHO VCD derived from ground-based mini MAX-DOAS observations are compared with OMI satellite observations for the respective days over IESE-NUST site (Fig.7). In this study, OMI HCHO tropospheric VCD level-2 product from TEMIS were used for comparison with the mini MAX-DAOS observation. The ground-based observations averaged only for the time between 12:30 to 13:30 were used in order to correlate to OMI overpass over Pakistan at 13:30 local time. The comparison presented in the fig.7 indicates similar temporal patterns in measured HCHO VCD at IESE-NUST site (with some minor differences). Both observations were selected for cloud free days. The observed difference between the ground-based mini MAX-DOAS values and OMI observations are mainly due to the reason that mini MAX-DOAS observations are point observations while OMI observations are averaged over a ground pixel size of 13 x 24 km^2. Therefore satellite observations are underestimated by one degree of magnitude.

E. Spatial Analysis

The maps show the monthly averaged concentration of formaldehyde for the study period of October 2013-March 2014 (Fig. 8). The concentration over IESE-NUST is represented by the black circle. The maps show the given correlation that exists between the grounds-based and satellite observations.

IV. Conclusion

Mini MAX-DOAS instrument were used to prepare database of HCHO concentrations during the time period October 2013 - March 2014. The concentration of HCHO was higher as compared to the guidelines provided by WHO on various occasion. Wind direction/speed plays an important role in transporting the pollutants away from the emission sources. IESE- NUST is considered as a clear site and high concentrations observed were due to the wind direction that has transported pollutants from the nearby areas towards NUST. High traffic densities on Golra Moar and Kashmir Highway are the main source of air pollution being transported to IESE-NUST site. Similar spatial trends were found when compared with the OMI observations. Differences are mainly due to the reason that MAX-DOAS gives the point observations whereas OMI observations are averaged over larger area.

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Figures

Figure 1: Google map of Islamabad, Rawalpindi and neighboring regions. Red line is indicating IJP road. Yellow lines are indicating the trajectories of Kashmir Highway and grand trunk road. Red balloon is showing the location of IESE-NUST monitoring site shown in inset picture.

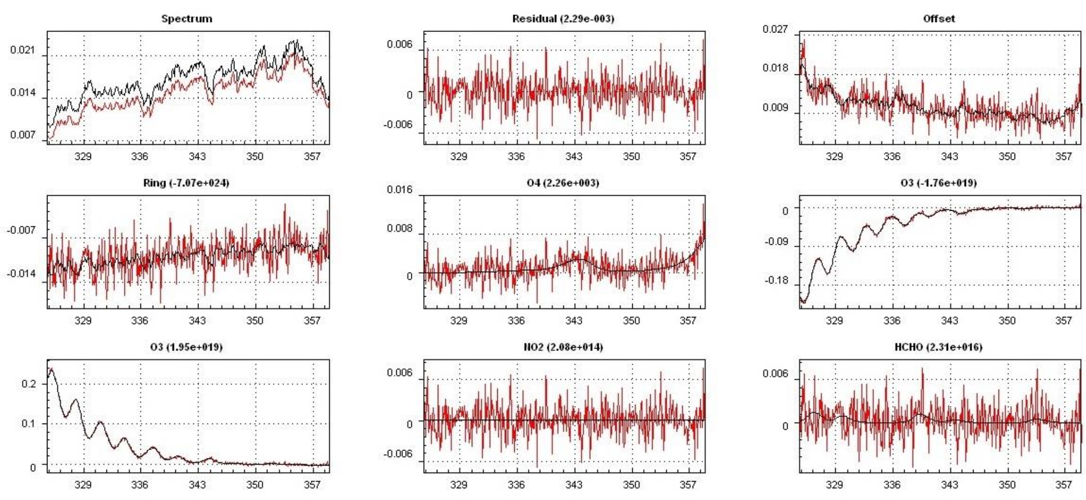

Figure 2: Example of DOAS fit by WINDOAS

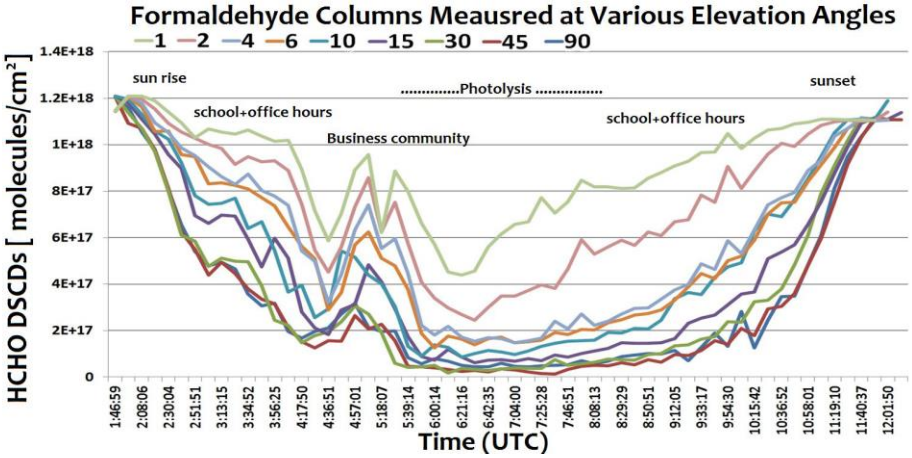

Figure 3: shows the diurnal variation of DSCD (molecule/cm2) retrieved from solar irradiance spectra recorded for 1, 2°, 4°, 6°, 10°, 15°, 30°, 45° and 90° elevation viewing angles during a day of 16 October 2013.

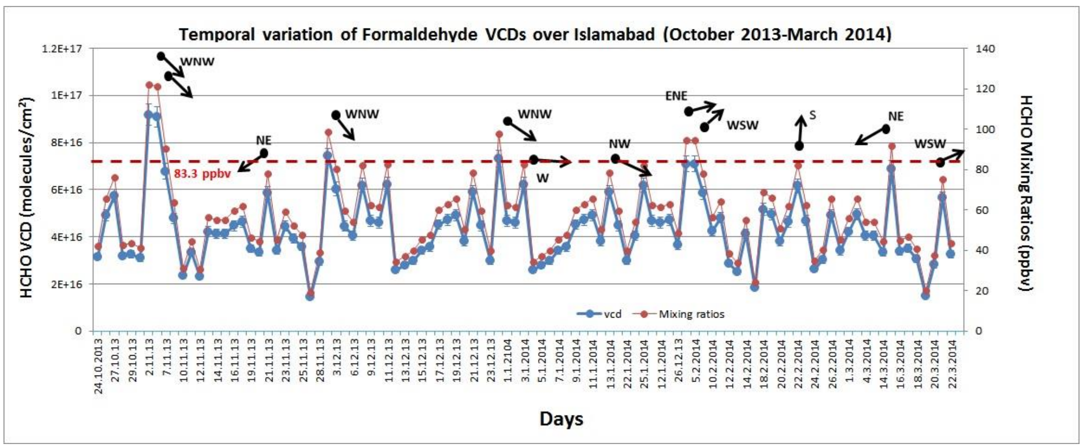

Figure 4. shows the daily averaged temporal variation of HCHO VCDs (molecules/cm2 left hand side) with bars representing the standard deviation and the red lines representing the mixing ratios (ppbv right hand side) over IESE NUST (October 2013-March 2014) and comparison of HCHO levels with existing WHO standards (WHO-limit-doted red line)

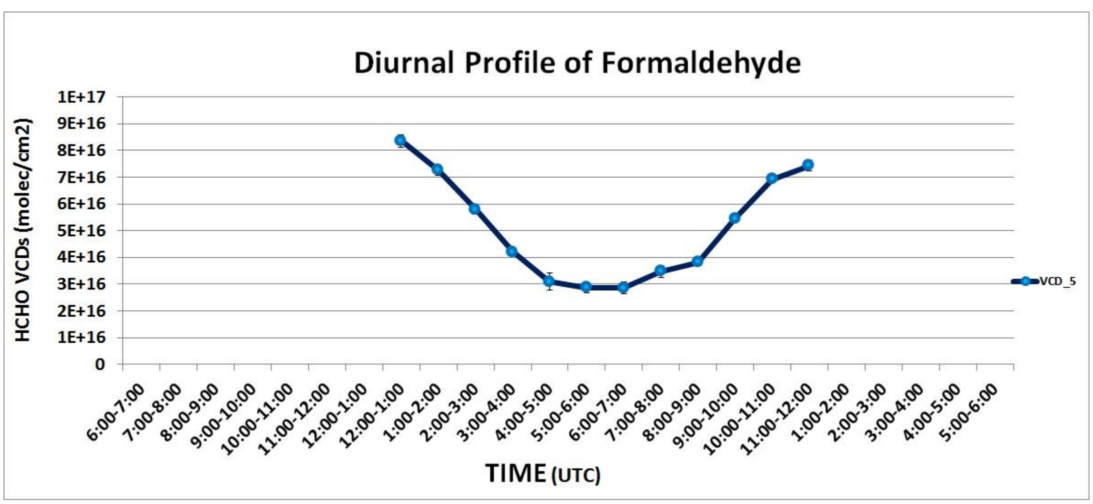

Figure 5. Average diurnal profile of HCHO for the time period October 2013-March 2014.

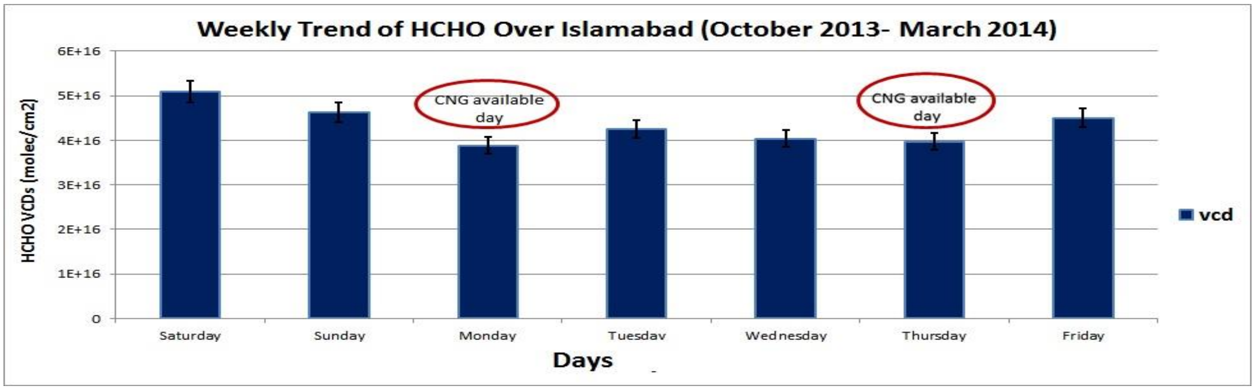

Figure 6. Weekly trend of Formaldehyde column density over Islamabad (October 2013-Marxh 2014). Bars represent the standard deviation.

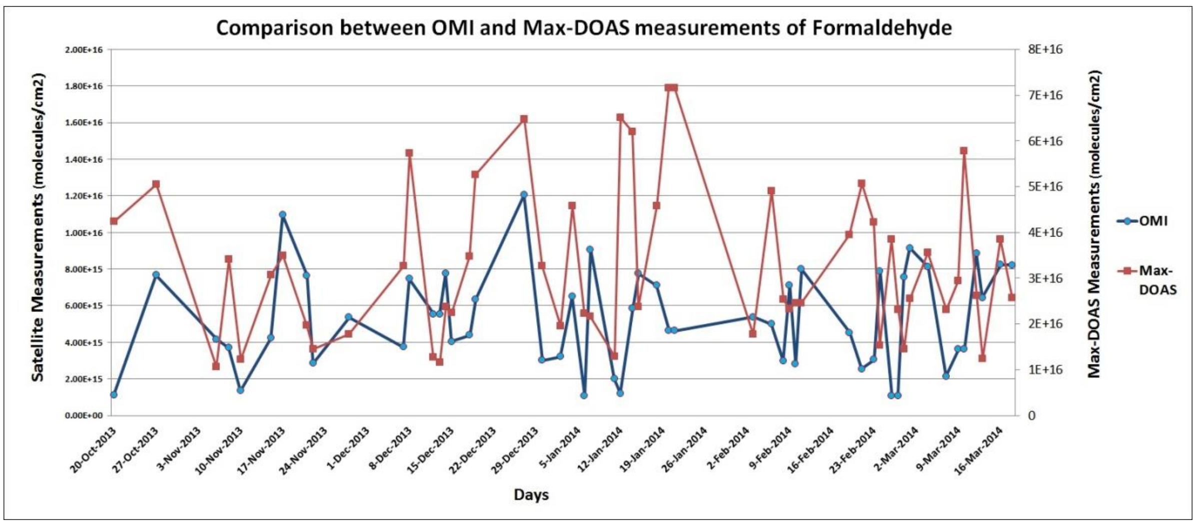

Figure 7. shows comparison of tropospheric HCHO VCD (molecules/cm2) derived from Ground-based mini MAX-DOAS measurements and the OMI satellite observations for respective cloud free days in from October 2013 till March, 2014 over IESE-NUST Islamabad. Mini-MAX-DOAS observations were averaged only for time between 12:30 to 13:30 in order to correlate to OMI overpass over Pakistan at 13:00 local time.

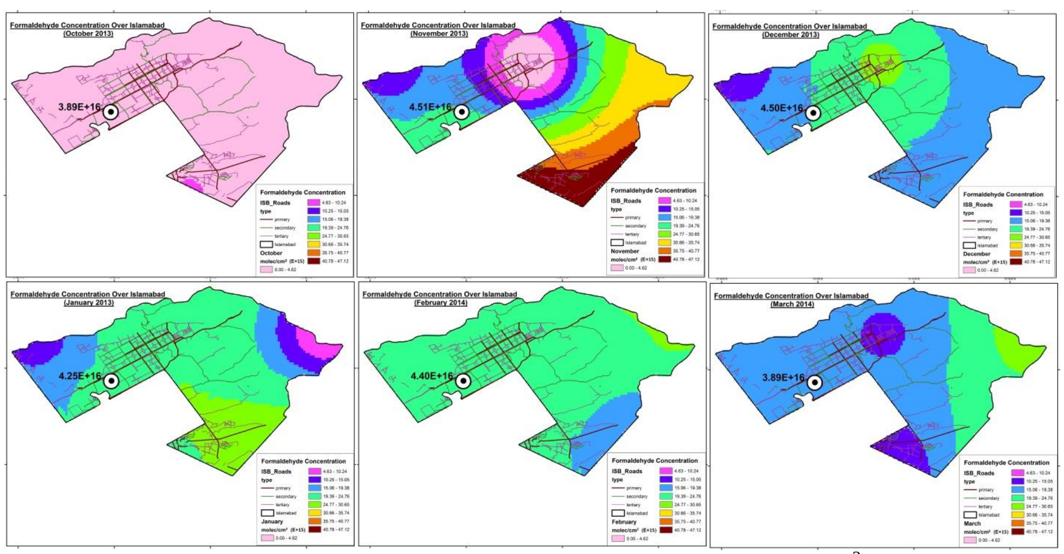

Figure 8. Spatial distribution of monthly mean formaldehyde concentration in molecules/cm^2 over Islamabad during the time period of October 2013-march 2014. The black circle indicates the concentration at IESE, NUST. The monthly maps are prepared by taking the daily concentration of HCHO.