First Simultaneous Observations of Formaldehyde and Glyoxal by MAX-DOAS in the Indo-Gangetic Plain Region

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Abstract

Since January 2017 continuous multi-axis differential optical absorption spectroscopy (MAX-DOAS) observations have been performed for the first time at Pantnagar (29.03°N, 79.47°E), a semi-urban site located in the Indo-Gangetic Plain region in India. Here we report the formaldehyde (HCHO), glyoxal (CHOCHO), and nitrogen dioxide (NO₂) concentrations for the lowest layer (0–1 km) of the retrieved vertical profiles. The ratio of CHOCHO to HCHO concentrations (R_{GF}), an important tracer indicative of changes in volatile organic compound emissions was estimated. During spring and autumn enhanced concentrations of HCHO and CHOCHO were observed under the influence of biomass burning. The mean R_{GF} for the whole observation period (January–November) in Pantnagar was estimated to be 0.029 ± 0.006 . Comparing with similar MAX-DOAS observations in central Thailand and reported literature values, we found that the R_{GF} tends to be $< \sim 0.04$ under the influence of biomass burning and/or anthropogenic emissions.

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1. Introduction

It has been well recognized that volatile organic carbons (VOCs) can change the oxidization capacity of the atmosphere, contribute to the formation of secondary organic aerosols and tropospheric ozone, and lead to the production of cloud condensation nuclei (Houweling et al. 1998; Liakakou et al. 2007; Monks 2005; Poisson et al. 2000; Volkamer et al. 2006). VOCs are emitted from oceans, vegetation, biomass burning, fossil fuel usage, and geochemical processes (Alvarado et al. 2014; Kansal 2009). Two important oxidized products of VOCs are formaldehyde (HCHO) and glyoxal (CHOCHO), which are thought to be useful tracers of VOC fluxes to the atmosphere (e.g., Hoque et al. 2018).

HCHO is the most abundant and simplest aldehyde in the atmosphere (Kansal 2009). It is predominantly a photochemical degradation product of methane and non-methane hydrocarbons (NMHCs) emitted from natural and anthropogenic sources (Fu et al. 2007; Jenkin et al. 1997). Primary sources of HCHO include direct emissions from biomass burning, industrial processes, fossil fuel combustion (Andreae and Merlet 2001; Lee et al. 1997; Holzinger et al. 1999; Yokelson et al. 1999; Geiger et al. 2002), and vegetation (Seco et al. 2007). The sinks of HCHO include photolysis, oxidization by hydroxyl (OH) radicals, and wet deposition (Arlander et al. 1995).

CHOCHO is the smallest and one of the most prevalent dicarbonyl compounds in the atmosphere. The main source of CHOCHO is the oxidation of biogenic VOCs such as isoprene and terpenes followed by anthropogenic VOCs such as acetylene, benzene, toluene, and xylene (Fu et al. 2008; Myriokefalitakis et al. 2008). CHOCHO can also be formed during the use of biomass and biofuels (Fu et al. 2008). The sinks of CHOCHO are similar

to those for HCHO, i.e., photolysis reactions (Fu et al. 2008), reaction with OH radicals, and dry and wet depositions (Myriokefalitakis et al. 2008). In addition, reversible and irreversible uptakes of CHOCHO by aqueous aerosols were reported (Kroll et al. 2005; Galloway et al. 2009; Ervens and Volkamer et al. 2010).

Due to the similar sinks and sources of HCHO and CHOCHO, the CHOCHO to HCHO concentration ratio (R_{GF}) is considered to be a possible tracer of changes in VOC emission sources (e.g., Hoque et al. 2018). But discrepancies remain in studies reporting the response of R_{GF} to different VOC emission sources. MacDonald et al. (2012) conducted long-path differential optical absorption spectroscopy (DOAS) measurements of HCHO and CHOCHO in a tropical forest, and reported an $R_{GF} > 0.2$. Using multi-axis DOAS (MAX-DOAS) measurements in Mainz, Ortega et al. (2015) observed R_{GF} values lower than 0.04 at a site dominated by anthropogenic emissions. Using satellite observations, Vrekoussis et al. (2010) found that the R_{GF} ranges between 0.04 and 0.06 in regions with strong biogenic emissions, whereas it can be below 0.03 in regions influenced by anthropogenic emissions. In contrast, DiGangi et al. (2012) conducted in-situ measurements of HCHO and CHOCHO during two observation campaigns in Pinus Ponderosa forests and found higher R_{GF} from anthropogenic reactive VOC mixtures and biomass burning compared with biogenic reactive VOC mixtures. Kaiser et al. (2015) observed higher R_{GF} over areas with strong monoterpene emissions and attributed lower R_{GF} to strong isoprene emissions during an aircraft campaign over the southeastern U.S. Thus, the R_{GF} response to different types of VOC emissions sources is yet under discussion. Here, we report almost one-year (January 2017 to November 2017) MAX-DOAS observations of HCHO and CHOCHO at Pantnagar, India (29.03°N, 79.47°E, 237 m.a.s.l). Besides, estimating the R_{GF} , we compare the R_{GF} response with the result from similar MAX-DOAS observations at a rural site in central Thailand (Hoque et al. 2018).

2. Observation site and methodology

The Indo-Gangetic Plain (IGP) is one of the most populated regions in the world. Pantnagar is a semi-urban site situated in the IGP region in India. It is located ~ 225 km northeast of the capital city New Delhi (Fig. 1). The Himalayan mountains lie in the north and east direction of the site, while low altitude plains are situated in the south and west. The major roadway connecting Pantnagar to nearby cities and a small local airport (~ 3 km) are situated close to the site. Few small-scale industries are located in Rudrapur (~ 12 km southwest of Pantnagar) and Haldwani (~ 25 km northeast of Pantnagar). The climate in Pantnagar is divided into four seasons: (1) winter (December to February), (2) spring (March to May), (3) summer monsoon (June to August), and (4) autumn (September to November).

A MAX-DOAS instrument similar to the one used by Hoque et al. (2018) for observations at Phimai, Thailand was installed in a building at Biotech Bhavan, Haldi in Pantnagar and its continuous observation started on 17 January 2017. The MAX-DOAS instrument measures ultraviolet-visible (UV-VIS) spectra of scattered sunlight at different elevation angles (ELs). The detailed description of the instrument is provided elsewhere (Irie et al. 2011, 2015; Hoque et al. 2018). The EL sequence of 3°, 4°, 5°, 6°, 8°, and 70° was used, and the same sequence of ELs was repeated

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Fig. 1. Location of Pantnagar which is situated ~225 km northeast of New Delhi. The map has been produced using the Google Earth application.

every 15 min. To minimize the possible systematic error in oxygen collision complex (O_4), MAX-DOAS off-axis EL measurements were limited to below 10° , while the measurement sensitivity in the lowest layer was kept maximum (Irie et al. 2015). The spectra between 310 and 515 nm were recorded. The wavelength calibration was performed using a high-resolution solar spectrum from Kurucz et al. (1984). The spectral resolution (full width at half maximum) was about 0.4 nm at 357 and 476 nm. Measured spectra were analyzed using the Japanese vertical profile retrieval algorithm version 2 (JM2) (Irie et al. 2011, 2015). The retrieval procedures and error estimates are described in detail by Irie et al. (2008a, 2008b, 2011, 2015). A short description is provided here.

First, the measured spectra were analyzed utilizing the DOAS technique (Platt 1994) to retrieve the differential slant column density (Δ SCD) of trace gases. Δ SCD is defined as the difference between the spectral measurement at an EL and that at the reference EL at 70° . HCHO and CHOCHO Δ SCD values were retrieved from fitting windows of 336–359 and 436–457 nm, respectively. The closest fitting windows containing significant O_4 absorptions around 360 and 477 nm were utilized to retrieve the O_4 Δ SCD values (Irie et al. 2009, 2011). The absorption cross-section data used and the absorbers fitted in the HCHO and CHOCHO fitting windows are identical to those used by Hoque et al. (2018).

Following the DOAS analysis, the retrieved O_4 Δ SCD values were converted to aerosol optical depth (AOD) and vertical profiles of aerosol extinction coefficient, using the aerosol retrieval algorithm based on optimal estimation (Irie et al. 2008a; Rodgers 2000). The radiative transfer model JACOSPAR (Irie et al. 2015) was used to create a lookup table (LUT) for the box air mass factor (A_{box}) vertical profile, which is defined as the ratio of the slant to vertical columns for a given layer. JACOSPAR is based on the Monte Carlo Atmospheric Radiative Transfer Simulator (MCARaTS) (Iwabuchi 2006). The results of MCARaTS have been validated against other radiative transfer models (Wagner et al. 2007). Utilizing the A_{box} LUT, the optimal aerosol loading and the A_{box} profiles which corresponds to the O_4 Δ SCD values at all ELs, were determined. Thus, this is different from simple geometric calculations. Following the similar procedure as the aerosol retrieval, HCHO and CHOCHO Δ SCD values were converted to their vertical column densities (VCDs) and vertical profiles of HCHO and CHOCHO, respectively. The A_{box} profiles in the LUT were calculated for 357 and 476 nm, which were slightly different from the trace gas wavelengths. To address such wavelength differences, the AOD at the desired trace gas wavelength was estimated from the retrieved AOD at the closer aerosol wavelength, assuming an Ångström exponent of unity. Then, the recalculated AOD was used to select the corresponding A_{box} profiles from the

Table 1. Estimated errors (%) for the retrieved CHOCHO and HCHO VMRs for the 0–1-km layer.

Component	Total error	Random error	Systematic error	Error due to bias in EL
CHOCHO	25	14	20	5
HCHO	37	20	31	5

LUT.

The estimated total error consisted of random and systematic components. The random error was estimated from the residuals that arose from the fittings of the HCHO (or CHOCHO) Δ SCD. The systematic error was estimated assuming changes in the retrieved AOD (corresponding A_{box} values), with values as high as 50 and 30%, respectively (Irie et al. 2011). The estimated errors, including the uncertainty in the ELs, are summarized in Table 1. To minimize the impact of clouds, retrievals for retrieved AOD greater than 3, corresponding to the highest value in the LUT, were excluded. These are likely due mostly to optically thick clouds. Further data screening was performed based on residuals of the O_4 Δ SCD ($< 10\%$), CHOCHO Δ SCD ($< 50\%$), HCHO Δ SCD ($< 50\%$), and the degree of freedom of the signal greater than 1.02.

3. Results and discussion

To characterize HCHO and CHOCHO data (at 0600–1800 local time (LT)) for the 0–1 km layer retrieved by MAX-DOAS at Pantnagar for 2017, their seasonal variations are shown in Fig. 2. Significant enhancements of both HCHO (~6 ppbv) and CHOCHO (~0.2 ppbv) concentrations were observed during spring and autumn. While biomass burning activities vary with geographical locations within India, they are usually predominant in spring (Venkataraman et al. 2006). Significant biomass burning activities during spring and autumn in Pantnagar and its nearby areas have been also reported (Kumar et al. 2011; Bhardwaj et al. 2016). To confirm them, the daily and monthly mean fire radiative power (FRP) data within 100 km of Pantnagar were analyzed (Fig. 2c). FRP is a measure of outgoing radiant heat from fires (Justice et al. 2002). The data were obtained from the MODIS Active Fire Detections data website (<https://firms.modaps.eosdis.nasa.gov>). Most of the fires were observed during spring and autumn, in correspondence to enhancements in HCHO and CHOCHO. In Fig. 2d, MAX-DOAS retrieved HCHO data are also shown together

Fig. 2. Time series of monthly means of (a) HCHO and (b) CHOCHO VMRs for the 0–1 km layer retrieved from the MAX-DOAS observations in Pantnagar in 2017. The error bars represent the 1σ standard deviation of the data used to calculate the monthly mean. HCHO and CHOCHO observations from 0600–1800 LT were used to calculate the monthly mean. (c) Daily and monthly mean fire radiative power (FRP) averaged over a distance of 100 km centred at Pantnagar. The black line and red dots indicate monthly and daily mean FRP values, respectively. (d) Comparison between HCHO time series obtained from our MAX-DOAS measurements and GOME-2 observations. GOME-2 observations were averaged over a distance of 50 km centred at Pantnagar. Data with cloud fraction (CF) < 0.2 were used. The monthly means were calculated using days in common between GOME-2 and morning MAX-DOAS (0900–1100 LT) data. The error bars represents the 1σ of the data used to calculate monthly means.

Fig. 3. (a) Time series of monthly mean NO₂ VMRs for the 0–1 km layer and the estimated monthly mean R_{GF} values in Pantnagar. Observations from 0600–1800 LT were used to calculate the monthly means. (b) Diurnal variation of R_{GF} in Pantnagar during different seasons. (c) Time series of monthly mean NO₂ VMRs for the 0–1 km layer and the estimated monthly mean R_{GF} values for the dry (January–April) and wet season (June–September) in Phimai in 2016. Observations from 0700–1700 LT were used to calculate the monthly mean. (d) Diurnal variation of R_{GF} in Phimai during the dry season (January–April) in 2015 and 2016. The error bars in (a), (b), (c), and (d) show the 2σ standard error.

with those from the GOME-2 satellite observational product, published by the German Aerospace Centre (Hassinen et al. 2016). The overpass time of GOME-2 at the equator is 0930 LT. The days in common between GOME-2 and morning MAX-DOAS (0900–1100 LT) data were used to calculate the monthly averages. GOME-2 data were averaged within 50 km. Good positive correlation ($R = 0.71$) between the observations was found. The enhancements of HCHO during the spring and autumn were also seen in the satellite observations. These correspondences support the potential link between biomass burning activities in IGP and the observed enhancement of both HCHO and CHOCHO.

The mean R_{GF} for the whole observation period was estimated to be 0.029 ± 0.006 . The daily mean CHOCHO and HCHO VMRs

for the 0–1-km layer were used for the estimation. The uncertainty was estimated following the procedure of Hoque et al. (2018). The estimated R_{GF} for the Pantnagar site was in the range of values reported in literatures for different conditions of VOC emission sources (Munger et al. 1995; Grosjean et al. 1996; Possanzini et al. 2007; Choi et al. 2010; Irie et al. 2011; DiGangi et al. 2012; Kaiser et al. 2015). For detailed understanding of the change in R_{GF} at Pantnagar, R_{GF} values for different seasons were estimated (Fig. 3a). R_{GF} values at Pantnagar were found to have varied between ~ 0.020 and ~ 0.039 . Utilizing satellite measurements, Vrekoussis et al. (2010) reported high R_{GF} values (0.04–0.06) where dominant VOC sources were biogenic, whereas R_{GF} values (< 0.03) were lower for regions with enhanced NO₂ level. Moreover, Kaiser

et al. (2015) also observed a high R_{GF} (0.04–0.07) under strong biogenic VOC emissions. The Pantnagar site can be influenced by biomass burning during the spring and autumn. The seasonal variation in NO_2 , which was retrieved simultaneously with HCHO and CHOCHO using JM2 utilizing the fitting window of 460–490 nm, showed enhanced NO_2 concentrations during spring and autumn (Fig. 3a). Biomass burning can contribute to the high NO_2 observed over the IGP region during spring and autumn (Ghude et al. 2013). Fu et al. (2007) found significant contribution of biomass burning to the observed HCHO variability over India during spring. However, based on our dataset, it is hard to assess the relative contribution of anthropogenic and pyrogenic sources in Pantnagar. Thus, the lower R_{GF} during spring and autumn could be potentially due to the combined influence of anthropogenic emissions and biomass burning. Although IGP is known as a high vegetation area (Mahajan et al. 2015), lower correlation among temperature, HCHO, and CHOCHO were observed in Pantnagar (Fig. S1). Seasonal variation in normalized difference vegetation index (NDVI) and enhanced vegetation index (EVI) were also poorly correlated with HCHO and CHOCHO (Fig. S2). Over strong vegetation areas, Vrekoussis et al. (2010) found positive correlations among EVI, HCHO, and CHOCHO. With lower correlations among temperature, HCHO, CHOCHO, NDVI, and EVI, biogenic emission sources can potentially be less significant in Pantnagar. Thus, the overall R_{GF} (< 0.04) observed in Pantnagar is likely mainly due to strong influence from biomass burning and anthropogenic emissions.

Figure 3b shows the diurnal variation in R_{GF} at Pantnagar. No significant differences in the diurnal R_{GF} values were observed in different seasons and most of the time the values were less than ~ 0.04 . Figure 3d shows the diurnal variation in R_{GF} estimated (0–1 km layer) from MAX-DOAS observations of HCHO and CHOCHO at Phimai (15.18°N, 102.56°E), a rural site in Thailand (Hoque et al. 2018). Pronounced biomass burning occurred during the dry season (January–April) in Phimai (Sugimoto et al. 2015; Hoque et al. 2018). The R_{GF} values in Phimai during dry seasons were mostly lower than 0.03. The estimated mean R_{GF} values during the dry seasons in Phimai and during all seasons in Pantnagar were similar (Table 2). Comparing the R_{GF} values at both sites, it is observed that, under the influence of biomass burning and anthropogenic emissions R_{GF} values tend to be $< \sim 0.04$. DiGangi et al. (2012) reported R_{GF} value ~ 0.03 during a biomass burning event. The average R_{GF} reported by Zarzana et al. (2017) from agricultural biomass burning was ~ 0.038 , although the observed R_{GF} had a wide range from 0.008 to 0.11. The mean R_{GF} values < 0.04 for region associated with anthropogenic emissions

Table 2. Estimated R_{GF} during seasons influenced by biomass burning in Phimai and Pantnagar. The errors represent the 2σ standard error.

Season and site	R_{GF}
Winter (JF), Pantnagar	0.029 ± 0.003
Spring 2017 (MAM), Pantnagar	0.030 ± 0.002
Summer Monsoon (JJA), Pantnagar	0.030 ± 0.004
Autumn 2017 (SON), Pantnagar	0.026 ± 0.002
Dry Season 2015 (JFMA), Phimai	0.030 ± 0.001
Dry Season 2016 (JFMA), Phimai	0.028 ± 0.002

were also observed by Ortega et al. (2015) and Miller et al. (2014). Thus, under the influence of biomass burning and anthropogenic emissions, the mean R_{GF} value tend to be less than ~ 0.04 . However, R_{GF} under the influence of biomass burning can vary with the type of biomass fuel (Akagi et al. 2013; Stokwell et al. 2014). At both sites, crop waste was the main biomass fuel. Different crop residue types can also yield different R_{GF} values (Kyle Zarzana, personal communication). If biomass burning was the dominant VOC source during spring and autumn in Pantnagar, then $R_{GF} < 0.04$ could potentially be representative of crop residues as biomass fuel. A detail classification of the biomass fuels, burning conditions, and VOC sources in Pantnagar is required to draw more definitive conclusion.

The correlation between NO_2 and R_{GF} for Pantnagar was not as strong that for Phimai (Fig. 3a). Figure 4 shows the correlation of HCHO and CHOCHO concentrations against NO_2 for both sites. For Phimai, a rural site, the only significant NO_2 source was most likely biomass burning, whereas Pantnagar can be influenced by local, regional, vehicular emissions, and biomass burning. This might lead to different correlations observed among the trace gases at both sites. Modeling studies by Li et al. (2014) suggested higher sensitivity of R_{GF} to lower NO_2 concentration. Kaiser et al. (2015) found no significant impact of NO_x on R_{GF} over urban areas. The overall NO_x emissions in IGP are higher even within India (Ghude et al. 2008; Ojha et al. 2012), whereas the Phimai site is characterized by low NO_2 (< 1 ppbv) environment. Thus, the higher NO_2 concentrations in Pantnagar could be a probable reason of the R_{GF} response becoming less sensitive to the NO_2 . To understand the relative contribution of different VOC sources to the R_{GF} value in Pantnagar, detailed characterization of relative VOC sources is required, which will be addressed in our future study.

Fig. 4. Correlation among NO_2 , HCHO, and CHOCHO at Pantnagar ((a) and (b)) and Phimai ((c) and (d)). The daily mean values are used for the correlation plots. Data for the whole observation period at both sites are included in the plots.

4. Conclusions

We reported simultaneous MAX-DOAS observations of HCHO, CHOCHO, and NO₂ conducted for the first time at a semi-urban site situated in the IGP region in India. The highest concentration of HCHO (~6 ppbv) and CHOCHO (~0.2 ppbv) were observed during spring and autumn. The monthly mean NO₂ was less than 1.5 ppbv. Such enhancements were potentially due mainly to biomass burning activities. The estimated R_{GF} for the Pantnagar site was 0.029 ± 0.006 . Estimated R_{GF} values for different seasons in Pantnagar were < 0.04 . The comparison with similar MAX-DOAS observations at a rural site in central Thailand and few reported literatures showed that under the influence of biomass burning and anthropogenic emission the R_{GF} tends to be $< \sim 0.04$. The low correlation of NDVI and EVI with HCHO and CHOCHO was also suggestive of less significance of the biogenic VOC emission sources compared to biomass burning and anthropogenic emission sources.

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