

Response to Co-Editor's comments

Dear Editor, we would like to thank you once again for your comments and suggestions which are useful for maintaining the quality of the manuscript and improve its write up. The authors' response and changes in the manuscript are presented in "blue" color.

Comments to the Author:

Additional technical corrections are needed prior to publication and are listed below by line number:

Thank you very much for pointing out these technical corrections which are important to improve the quality of the manuscript.

Lines 306, 311, and 313: Clarify what is meant by "changes" and "variabilities." As stated these are too vague and it is unclear what specific measures are being discussed. Perhaps "relative standard deviation" is the correct term?

We would like to apologize for creating confusion on changes and variabilities. We also agree with your suggestion using "relative standard deviation" instead of changes or variabilities. We have changed it throughout the manuscript.

Line 756: The directions "East-South/North East" is confusing. Please break this up into parts to omit the slash.

45: This comparison suggested that

213: iron, fabrics, etc.

215: 4 km to the west of Bode.

220: located approximately 4.5

234: that results in high sensitivity. (Omit precision here, as this is not affected by the path length.)

236: interest to reduce drift.

238: precisions for CO₂...

244: data every 5 seconds and... corrected data for CO₂

247: field measurements.

254: simultaneous measurements...

258: The Horiba...
265: A statistically...
271: deemed unnecessary.
272: Besides being highly...
292: from entering into...
293: an AWS....
299: 3.5 discuss the interrelation...
306: mixing ratio were higher...
327: spell out "ca."
344: Valley, the measurement...
348: (small-medium) and biomass...
356: The small differences...
357: the smaller number...
373: than in the pre-monsoon....
383: all seasons...
387: CH4 emissions...
390: brick kilns
396: suppresses emissions.
398: and, where relevant, reduction...
400: during high intensity
409: related to little or no rainfall
412: relatively low during...
426: increased by...
430, 432: East-Northeast (E-NE)
447: local sources...
462: emissions form the main...
464: higher values of all...
487: farm fires...
492: were coincident
498: diurnal patterns
506: decayed

509: may explain...

533: verb is needed to complete the clause following “and”

546: from the plains to the mountains

548: Although the CO...

565: Omit “Measurement in”

583: change in the mixing later

596: vegetation fires

771: seasons and wind directions suggested

Table 6 caption: and CO₂ and their lower and upper bounds (LB and UB).

Table 5 caption: change “gram of gas” to “mass of gas”

Table 5 caption: “, except for the transport sector”

Table 1: use capitalization consistently in the headings.

Figure 9 caption: in all seasons except...

Thank you for providing detail technical corrections. We have incorporated all the technical corrections mentioned above in the revised manuscript. Furthermore, we have checked the ACP’s manuscript preparation guidelines and style once more carefully, and consistently followed the style, for example units and time format, throughout the manuscript (for example, 10 % NOT 10%, 09:00 instead of 9 am).

1 **Seasonal and diurnal variations of methane and carbon dioxide in the Kathmandu Valley**
2 **in the foothills of the central Himalaya**

3 Khadak Singh Mahata^{1,2}, Arnico Kumar Panday^{3,4}, Maheswar Rupakheti^{1,5*}, Ashish Singh¹,
4 Manish Naja⁶, Mark G. Lawrence^{1,2}

5 [1] Institute for Advanced Sustainability Studies (IASS), Potsdam, Germany

6 [2] University of Potsdam, Potsdam, Germany

7 [3] International Centre for Integrated Mountain Development (ICIMOD), Lalitpur, Nepal

8 [4] University of Virginia, Virginia, USA

9 [5] Himalayan Sustainability Institute (HIMSI), Kathmandu, Nepal

10 [6] Aryabhata Research Institute of Observational Sciences (ARIES), Nainital, India

11

12 *Correspondence to: M. Rupakheti (maheswar.rupakheti@iass-potsdam.de)

13

14 **Abstract**

15 The SusKat-ABC (Sustainable Atmosphere for the Kathmandu Valley- Atmospheric Brown
16 Clouds) international air pollution measurement campaign was carried out during December
17 2012-June 2013 in the Kathmandu Valley and surrounding regions in Nepal. The Kathmandu
18 Valley is a bowl-shaped basin with a severe air pollution problem. This paper reports
19 measurements of two major greenhouse gases (GHGs), methane (CH₄) and carbon dioxide
20 (CO₂), along with the pollutant CO, that began during the campaign and were extended for a year
21 at the SusKat-ABC supersite in Bode, a semi-urban location in the Kathmandu Valley.
22 Simultaneous measurements were also made during 2015 in Bode and a nearby rural site
23 (Chanban), ~25 km (aerial distance) to the southwest of Bode, on the other side of a tall ridge.
24 The ambient mixing ratios of methane (CH₄), carbon dioxide (CO₂), water vapor, and carbon
25 monoxide (CO) were measured with a cavity ring down spectrometer (Picarro G2401, USA),
26 along with meteorological parameters for a year (March 2013 - March 2014). These

27 measurements are the first of their kind in the central Himalayan foothills. At Bode, the annual
28 average mixing ratios of CO₂ and CH₄ were 419.3 (± 6.0) ppm and 2.192 (± 0.066) ppm,
29 respectively. These values are higher than the levels observed at background sites such as Mauna
30 Loa, USA (CO₂: 396.8 \pm 2.0 ppm, CH₄: 1.831 \pm 0.110 ppm) and Waliguan, China (CO₂: 397.7 \pm
31 3.6 ppm, CH₄: 1.879 \pm 0.009 ppm) during the same period, and at other urban and semi-urban
32 sites in the region such as Ahmedabad and Shadnagar (India). They varied slightly across the
33 seasons at Bode, with seasonal average CH₄ mixing ratios being 2.157 (± 0.230) ppm in the pre-
34 monsoon season, 2.199 (± 0.241) ppm in the monsoon, 2.210 (± 0.200) ppm in the post-monsoon,
35 and 2.214 (± 0.209) ppm in the winter season. The average CO₂ mixing ratios were 426.2
36 (± 25.5) ppm in pre-monsoon, 413.5 (± 24.2) ppm in monsoon, 417.3 (± 23.1) ppm in post-
37 monsoon, and 421.9 (± 20.3) ppm in winter season. The maximum seasonal mean mixing ratio of
38 CH₄ in winter was only 0.057 ppm or 2.6 % higher than the seasonal minimum during the pre-
39 monsoon period, while CO₂ was 12.8 ppm or 3.1 % higher during the pre-monsoon period
40 (seasonal maximum) than during the monsoon (seasonal minimum). On the other hand, the CO
41 mixing ratio at Bode was 191 % higher during the winter than during the monsoon season. The
42 enhancement in CO₂ mixing ratios during the pre-monsoon season is associated with additional
43 CO₂ emissions from forest fire and agro-residue burning in northern South Asia in addition to
44 local emissions in the Kathmandu Valley. Published CO/CO₂ ratios of different emission sources
45 in Nepal and India were compared with the observed CO/CO₂ ratios in this study. This
46 comparison [suggested](#) that the major sources in the Kathmandu Valley were residential cooking
47 and vehicle exhaust in all seasons except winter. In winter, the brick kiln emissions were a major
48 source. Simultaneous measurement in Bode and Chanban (15 July - 3 Oct 2015) revealed that
49 the mixing ratio of CO₂, CH₄ and CO mixing ratios were 3.8 %, 12 %, and 64 % higher in Bode
50 than Chanban. Kathmandu Valley, thus, has significant emissions from local sources, which can
51 also be attributed to its bowl shaped geography that is conducive to pollution build-up. At Bode,
52 all three gas species (CO₂, CH₄ and CO) showed strong diurnal patterns in their mixing ratios
53 with a pronounced morning peak (ca. 08:00), a dip in the afternoon, and again gradual increase
54 through the night until the next morning, whereas CH₄ and CO at Chanban did not show any
55 noticeable diurnal variations.

56 These measurements provide the first insights into diurnal and seasonal variation of key
57 greenhouse gases and air pollutants and their local and regional sources, which are important
58 information for the atmospheric research in the region.

59 **1 Introduction**

60 The average atmospheric mixing ratios of two major greenhouse gases (GHGs), CO₂ and CH₄,
61 have increased by about 40 % (from 278 to 390.5 ppm) and about 150 % (from 722 to 1803
62 ppb) respectively since pre-industrial times (~1750 AD). This is mostly attributed to
63 anthropogenic emissions (IPCC, 2013). The current global annual rate of increase of the
64 atmospheric CO₂ mixing ratio is 1-3 ppm, with average annual mixing ratios now exceeding a
65 value of 400 ppm at the background reference location in Mauna Loa (WMO, 2016). Between
66 1750 and 2011, 240(±10) Pg C of anthropogenic CO₂ was accumulated in the atmosphere of
67 which two thirds were contributed by fossil fuel combustion and cement production, with the
68 remaining coming from deforestation and land use/land cover changes (IPCC, 2013). CH₄ is the
69 second largest gaseous contributor to anthropogenic radiative forcing after CO₂ (Forster et al.,
70 2007). The major anthropogenic sources of atmospheric CH₄ are rice paddies, ruminants and
71 fossil fuel use, contributing approximately 60 % to the global CH₄ budget (Chen and Prinn, 2006;
72 Schneising et al., 2009). The remaining fraction is contributed by biogenic sources such as
73 wetlands and fermentation of organic matter by microbes in anaerobic conditions (Conrad,
74 1996).

75 Increasing atmospheric mixing ratios of CO₂ and CH₄ and other GHGs and short-lived climate-
76 forcing pollutants (SLCPs) such as black carbon (BC) and tropospheric ozone (O₃) have caused
77 the global mean surface temperature to increase by 0.85 °C from 1880 to 2012. The surface
78 temperature is expected to increase further by up to 2 degrees at the end of the 21st century in
79 most representative concentration pathways (RCPs) emission scenarios (IPCC, 2013). The
80 increase in surface temperature is linked to melting of glaciers and ice sheets, sea level rise,
81 extreme weather events, loss of biodiversity, reduced crop productivity, and economic losses
82 (Fowler and Hennessy, 1995; Guoxin and Shibasaki, 2003).

83 Seventy percent of global anthropogenic CO₂ is emitted in urban areas (Fragkias et al., 2013).
84 Developing countries may have lower per capita GHG emissions than developed countries, but
85 the large cities in developing countries, with their high population and industrial densities, are
86 major consumers of fossil fuels and thus, emitters of GHGs. South Asia, a highly populated
87 region with rapid growth in urbanization, motorization, and industrialization in recent decades,
88 has an ever increasing fossil fuel demand and its combustion emitted 444 Tg C yr⁻¹ in 2000
89 (Patra, et al., 2013), or about 5 % of the global total CO₂ emissions. Furthermore, a major
90 segment of the population in South Asia has an agrarian economy and uses biofuel for cooking
91 activities, and agro-residue burning is also common practice in the region, which are important
92 major sources of air pollutants and greenhouse gases in the region (CBS, 2011; Pandey et al.,
93 2014; Sinha et al., 2014).

94 The emission and uptake of CO₂ and CH₄ follow a distinct cycle in South Asia. By using inverse
95 modeling, Patra et al. (2011) found a net CO₂ uptake (0.37 ± 0.20 Pg C yr⁻¹) during 2008 in
96 South Asia and the uptake (sink) is highest during July-September. The remaining months acts as
97 a weak gross sink but a moderate gross source for CO₂ in the region. The observed variation is
98 linked with the growing seasons. Agriculture is a major contributor of methane emission. For
99 instance, in India it contributes to 75 % of CH₄ emissions (MoEF, 2007). Ambient CH₄
100 concentrations are highest during June to September (peaking in September) in South Asia which
101 are also the growing months for rice paddies (Goroshi et al., 2011). The minimum column
102 averaged CH₄ mixing ratios are in February-March (Prasad et al., 2014).

103 Climate change has impacted South Asia in several ways, as evident in temperature increase,
104 change in precipitation patterns, higher incidence of extreme weather events (floods, droughts,
105 heat waves, cold waves), melting of snowfields and glaciers in the mountain regions, and
106 impacts on ecosystems and livelihoods (ICIMOD, 2009; MoE, 2011). Countries such as Nepal
107 are vulnerable to impacts of climate change due to inadequate preparedness for adaptation to
108 impacts of climate change (MoE, 2011). Decarbonization of its economy can be an important
109 policy measure in mitigating climate change. Kathmandu Valley is one of the largest
110 metropolitan cities in the foothills of the Hindu Kush-Himalaya which has significant reliance on
111 fossil fuels and biofuels. In 2005, fossil fuel burning accounted for 53 % of total energy

112 consumption in the Kathmandu Valley, while biomass and hydroelectricity were 38 % and 9 %,
113 respectively (Shrestha and Rajbhandari, 2010). Fossil fuel consumed in the Kathmandu Valley
114 accounts for 32 % of the country's fossil fuel imports, and the major fossil fuel consumers are
115 residential (53.17 %), transport (20.80 %), industrial (16.84 %), and commercial (9.11 %)
116 sectors. Combustion of these fuels in traditional technologies such as Fixed Chimney Bulls
117 Trench Kiln (FCBTK) and low efficiency engines (vehicles, captive power generator sets etc.)
118 emit significant amounts of greenhouse gases and air pollutants. This has contributed to elevated
119 ambient concentrations of particulate matter (PM), including black carbon and organic carbon,
120 and several gaseous species such as ozone, polycyclic aromatic hydrocarbons (PAHs),
121 acetonitrile, benzene and isocyanic acid (Pudasainee et al., 2006; Aryal et al., 2009; Panday and
122 Prinn, 2009; Sharma et al., 2012; World Bank, 2014; Chen et al., 2015; Putero et al., 2015;
123 Sarkar et al., 2016). The ambient levels often exceed national air quality guidelines (Pudasainee
124 et al., 2006; Aryal et al., 2009; Putero et al., 2015) and are comparable or higher than ambient
125 levels observed in other major cities in South Asia.

126 Past studies in the Kathmandu Valley have focused mainly on a few aerosols species (BC, PM)
127 and short-lived gaseous pollutants such as ozone and carbon monoxide (Pudasainee et al., 2006;
128 Aryal et al., 2009; Panday and Prinn, 2009; Sharma et al., 2012, Putero et al., 2015). To the best
129 of authors' knowledge, no direct measurements of CO₂ and CH₄ are available for the Kathmandu
130 Valley. Recently, emission estimates of CO₂ and CH₄ were derived for the Kathmandu Valley
131 using the International Vehicle Emission (IVE) model (Shrestha et al., 2013). The study
132 estimated 1554 Gg of annual emission of CO₂ from a fleet of vehicles (that consisted of public
133 buses, 3-wheelers, taxis and motor cycles; private cars, trucks and non-road vehicles were not
134 included in the study) for the year 2010. In addition, the study also estimated 1.261 Gg of CH₄
135 emitted from 3 wheelers (10.6 %), taxis (17.7 %) and motorcycles (71 %) for 2010.

136 This study presents the first 12 months of measurements of two key GHGs, CH₄ and CO₂ along
137 with other trace gases and meteorological parameters in Bode, a semi-urban site in the eastern
138 part of the Kathmandu Valley. The year-long measurement in Bode is a part of the SusKat-ABC
139 (Sustainable Atmosphere for the Kathmandu Valley – Atmospheric Brown Clouds) international
140 air pollution measurement campaign conducted in and around the Kathmandu Valley from

141 December 2012 to June 2013. Details of the SusKat-ABC campaign are described in Rupakheti
142 et al. (2017, manuscript in preparation). The present study provides a detailed account of
143 seasonal and diurnal behaviors of CO₂ and CH₄ and their possible sources. To examine the rural-
144 urban differences and estimate the urban enhancement, these gaseous species were also
145 simultaneously measured for about three months (July - October) in 2015 at Chanban, a rural site
146 about 25 km (aerial distance) outside and southwest of Kathmandu Valley. The seasonality of the
147 trace gases and influence of potential sources in various (wind) directions are further explored by
148 via ratio analysis. This measurement provides unique data from highly polluted but relatively
149 poorly studied region (central Himalayan foothills in South Asia) which could be useful for
150 validation of emissions estimates, model outputs and satellite observations. The study, which
151 provides new insights on potential sources, can also be a good basis for designing mitigation
152 measures for reducing emissions of air pollutants and controlling greenhouse gases in the
153 Kathmandu Valley and the region.

154 **2 Experiment and Methodology**

155 **2.1 Kathmandu Valley**

156 The Kathmandu Valley consists of three administrative districts: Kathmandu, Lalitpur, and
157 Bhaktapur, situated between 27.625° N, 27.75° N and 85.25° E, 85.375° E. It is a nearly circular
158 bowl-shaped valley with a valley floor area of approximately 340 km² located at an altitude of
159 1300 meter above sea level (masl). The surrounding mountains are close to 2000-2800 m in
160 height above sea level with five mountain passes located at about 200-600 m above the valley
161 floor and an outlet for the Bagmati River southwest of the Kathmandu Valley. Lack of
162 decentralization in in Nepal has resulted in the concentration of economic activities, health and
163 education facilities, the service sector, as well as most of the central governmental offices in the
164 Kathmandu Valley. Consequently, it is one of the fastest growing metropolitan areas in South
165 Asia with a current population of about 2.5 million, and the population growth rate of 4 % per
166 year (World Bank, 2013) Likewise, approximately 50 % of the total vehicle fleet (2.33 million)
167 of the country is in Kathmandu Valley (DoTM, 2015). The consumption of fossil fuels such as
168 liquefied petroleum gas (LPG), kerosene for cooking and heating dominates the residential
169 consumption, while the rest use biofuel (fuel wood, agro-residue, animal dung) for cooking and

170 heating in the Kathmandu Valley. The commercial sector is also growing in the valley, and the
171 latest data indicate the presence of 633 industries of various sizes. These are mainly associated
172 with dyeing, brick kilns, and manufacturing industries. Fossil fuels such as coal and biofuels are
173 the major fuels used in brick kilns. Brick kilns are reported as one of the major contributors of air
174 pollution in the Kathmandu Valley (Chen et al., 2015; Kim et al., 2015; Sarkar et al., 2016).
175 There are about 115 brick industries in the valley (personal communication with Mahendra
176 Chitrakar, President of the Federation of Nepalese Brick Industries). Acute power shortage in the
177 Valley is common all around the year, especially in the dry season (winter/pre-monsoon) when
178 the power cuts can last up to 12 hours a day (NEA, 2014). Energy demand during the power cut
179 period is met with the use of small (67 % of 776 generators surveyed for the World Bank study
180 was with capacity less than 50 kVA) but numerous captive power generators (diesel/petrol),
181 which further contribute to valley's poor air quality. According to the World Bank's estimate,
182 over 250,000 such generator sets are used in the Kathmandu Valley alone, producing nearly 200
183 MW of captive power, and providing about 28 % of the total electricity consumption of the
184 valley (World Bank, 2014). Apart from these sources, trash burning, which is a common practice
185 (more prevalent in winter) throughout the valley, is one of the major sources of air pollutants and
186 GHGs.

187 Climatologically, Kathmandu Valley has a sub-tropical climate with annual mean temperature of
188 18°C, and annual average rainfall of 1400 mm, of which 90 % occurs in monsoon season (June-
189 September). The rest of the year is dry with some sporadic rain events. The wind circulation at
190 large scale in the region is governed by the Asian monsoon circulation and hence the seasons are
191 also classified based on such large scale circulations and precipitation: Pre-Monsoon (March-
192 May), Monsoon (June-September), Post-Monsoon (October-November) and Winter (December-
193 February). Sharma et al. (2012) used the same classification of seasons while explaining the
194 seasonal variation of BC concentrations observed in the Kathmandu Valley. Locally in the
195 valley, the mountain-valley wind circulations play an important role in influencing air quality.
196 The wind speed at the valley floor is calm ($\leq 1 \text{ m s}^{-1}$) in the morning and night, while a westerly
197 wind develops after 11:00 in the morning till dusk, and switches to a mild easterly at night
198 (Panday and Prinn, 2009; Regmi et al., 2003). This is highly conducive to building up of air
199 pollution in the valley, which gets worse during the dry season.

200 **2.2 Study sites**

201 Two sites, a semi-urban site within the Kathmandu Valley and a rural site outside the Kathmandu
202 Valley, were selected for this study. The details of the measurements carried out in these sites is
203 described Table 1 and in section 2.2.1 and 2.2.2.

204 **2.2.1 Bode (SusKat-ABC supersite)**

205 The SusKat-ABC supersite was set up at Bode, a semi-urban location (Figure 1) of the
206 Madhyapur Thimi municipality in the Bhaktapur district in the eastern side of the Kathmandu
207 Valley. The site is located at 27.68° N latitude, 85.38° E longitude, and 1344 masl. The local
208 area around the site has a number of scattered houses and agricultural fields. The agriculture
209 fields are used for growing rice paddies in the monsoon season. It also receives outflow of
210 polluted air from three major cities in the valley: Kathmandu Metropolitan City and Lalitpur
211 Sub-metropolitan City, both mainly during daytime, and Bhaktapur Sub-metropolitan City
212 mainly during nighttime. Among other local sources around the site, about 10 brick kilns are
213 located in the east and southeast direction, approximately within 1-4 km from the site which are
214 operational only during dry season (January to April). There are close to 20 small and medium
215 industries (pharmaceuticals, plastics, electronics, tin, wood, aluminum, iron, fabrics etc.)
216 scattered in the same direction. The Tribhuvan International Airport (TIA) is located
217 approximately 4 km to the west of Bode.

218 **2.2.2 Chanban**

219 Chanban is a rural/background site in Makwanpur district outside of the Kathmandu Valley
220 (Figure 1). This site is located ~25 km aerial distance due southwest from Bode. The site is
221 located on a small ridge (27.65° N, 85.14° E, 1896 masl) between two villages - Chitlang and
222 Bajrabarahi - within the forested watershed area of Kulekhani Reservoir, which is located
223 [approximately](#) 4.5 km southwest of the site. The instruments were set up on the roof of 1-storey
224 building in an open space inside the Nepali Army barrack. There was a kitchen of the army
225 barrack at about 100 m to the southeast of the measurement site. The kitchen uses LPG,
226 electricity, kerosene, and firewood for cooking activities.

227 **2.3 Instrumentation**

228 The measurements were carried out in two phases in 2013-2014 and 2015. In phase one, a cavity
229 ring down spectrometer (Picarro G2401, USA) was deployed in Bode to measure ambient CO₂,
230 CH₄, CO, and water vapor mixing ratios. Twelve months (6 March 2013 - 5 March 2014) of
231 continuous measurements were made in Bode. The operational details of the instruments
232 deployed in Bode are also provided in Table 1. In phase two, simultaneous measurements were
233 made in Bode and Chanban for a little less than 3 months (15 July to 03 October 2015).

234 The Picarro G2401 analyzer quantifies spectral features of gas phase molecules by using a novel
235 wavelength-scanned cavity ring down spectroscopic technique (CRDS). The instrument has a 30
236 km path length in a compact cavity that results in high sensitivity. Because of the high precision
237 wavelength monitor, it uses absolute spectral position and maintains accurate peak
238 quantification. Further, it only monitors the special features of interest to reduce drift. The
239 instrument also has water correction to report dry gas fraction. The reported measurement
240 precisions for CO₂, CH₄, CO and water vapor in dry gas is < 150 ppb, < 30 ppb, < 1 ppb and <
241 200 ppm for 5 seconds with 1 standard deviation (Picarro, 2015).

242 In Bode, the Picarro analyzer was placed on the 4th floor of a 5-storey building with an inlet at
243 0.5 m above the roof of the building with a 360 degree view (total inlet height: 20 m above
244 ground). The sample air was filtered at the inlet to keep dust and insects out and was drawn into
245 the instrument through a 9 m Teflon tube (1/4 inch ID). The Picarro analyzer was set to record
246 data every 5 second and recorded both directly sampled data and water corrected data for CO₂
247 and CH₄. In this paper, only water-corrected or dry mixing ratios of CH₄ and CO₂ were used to
248 calculate the hourly averages for diurnal and seasonal analysis.

249 The instruments were factory calibrated before commencing the field measurements. Picarro
250 G2401 model is designed for remote application and long term deployment with minimal drift
251 and less requirement for intensive calibration (Crosson, 2008) and thus was chosen for the
252 current study in places like Kathmandu where there is no or limited availability of high quality
253 reference gases. Regular calibration of Picarro G2401 in field during 2013-2014 deployment was
254 not conducted due to challenges associated with the quality of the reference gas, especially for
255 CO and CH₄. One time calibration was performed for CO₂ (at 395 and 895 ppm) in July 2015
256 before commencing the simultaneous measurements in Bode and Chanban in 2015. The

257 difference between CO₂ mixing ratio reported by the analyzer and the reference mixing ratio was
258 within 5 %. CO observations from Picarro G2401 were compared with observations from
259 another CO analyzer (Horiba, model AP370) that was also operated in Bode for 3 months
260 (March - May 2013). The Horiba CO monitor was a new unit, which was factory calibrated
261 before its first deployment in Bode. Nevertheless, this instrument was inter-compared with
262 another CO analyzer (same model) from the same manufacturer prior to the campaign and its
263 correlation coefficient was 0.9 [slope of data from the new unit (y-axis) vs the old unit (x-axis) =
264 1.09]. Primary gas cylinders from Linde UK (1150 ppb) and secondary gases from Ultra-Pure
265 Gases and Chemotron Science Laboratories (1790 ppb) were used for the calibration of CO
266 instrument. Further details on CO measurements and calibration of Horiba AP370 can be found
267 in Sarangi et al. (2014; 2016). A statistically significant correlation ($r = 0.99$, slope = 0.96) was
268 found between Picarro and Horiba hourly average CO mixing ratio data (Supplementary
269 Information Figure S1). Furthermore, the monthly mean difference between these two
270 instruments (Horiba AP370 minus Picarro G2401) was calculated to be 0.02 ppm (3 %), 0.04
271 ppm (5 %) and 0.02 ppm (4 %) in March, April and May, respectively. For the comparison
272 period of 3 months, the mean difference was 0.02 ppm (4 %). Overall differences were small to
273 negligible during the comparison period and thus, adjustment in the data was deemed
274 unnecessary.

275 Besides being highly selective to individual species, Picarro G2401 has a water correction
276 function and thus accounts for the any likely drift in CO, CO₂ and CH₄ mixing ratios with the
277 fluctuating water vapor concentration (Chen et al., 2013; Crosson, 2008). Crosson (2008) also
278 estimated a peak to peak drift of 0.25 ppm. Further, Crosson (2008) observed a 1.2 ppb/day drift
279 in CO₂ after 170 days from the initial calibration. For a duration of one year the drift will be less
280 than 1 ppm, which is less than 1 % of the observed mixing ratio in (hourly ranges: 376-537 ppm)
281 Bode even if the drift was in same magnitude as in case of Crosson (2008). Crosson (2008)
282 reported 0.8 ppb peak to peak drift in CH₄ measurements for 18 days after the initial calibration.

283 There were other instruments concurrently operated in Bode; a ceilometer for measuring mixing
284 layer height (Vaisala Ceilometer CL31, Finland), and an Automatic Weather Station (AWS)
285 (Campbell Scientific, USA). The ceilometer was installed on the rooftop (20 m above ground) of

286 the building (Mues et al., 2017). For measuring the meteorological parameters, a Campbell
287 Scientific AWS (USA) was set up on the roof of the building with sensors mounted at 2.9 m
288 above the surface of the roof (22.9 m from the ground). The Campbell Scientific AWS measured
289 wind speed and direction, temperature, relative humidity and solar radiation every minute.
290 Temperature and rainfall data were taken from an AWS operated by the Department of
291 Hydrology and Meteorology (DHM), Nepal at the Tribhuvan International Airport (TIA, see
292 Figure 1), ~4 km due west of Bode site.

293 At Chanban, the inlet for Picarro gas analyzer was kept on the rooftop ~3 m above the ground
294 and the sample air was drawn through a 3 m long Teflon tube (1/4 inches ID). The sample was
295 filtered at the inlet with a filter (5-6 μm pore size) to prevent aerosol particles from entering into
296 the analyzer. An AWS (Davis Vantage Pro2, USA) was also set up in an open area, about 17 m
297 away from the building and with the sensors mounted at 2 m above ground.

298 **3. Results and discussion**

299 The results and discussions are organized as follow: Sub-section 3.1 describes a year round
300 variation in CH_4 , CO_2 , CO and water vapor at Bode; sub-sections 3.2, 3.3 present the analysis of
301 the observed monthly and seasonal variations and diurnal variation. Sub-sections 3.4 and 3.5
302 discuss the interrelation of CO_2 , CH_4 and CO and potential emission sources in the valley and
303 sub-section 3.6 compares and contrasts CH_4 , CO_2 , CO at Bode and Chanban.

304 **3.1 Time series of CH_4 , CO_2 , CO and water vapor mixing ratios**

305 Figure 2 shows the time series of hourly mixing ratios of CH_4 , CO_2 , CO, and water vapor at
306 Bode. Meteorological data from Bode and the Tribhuvan International Airport are also shown in
307 Figure 2. Data gaps in Figure 2a and 2b were due to maintenance of the measurement station. In
308 general, the fluctuations in the mixing ratio for CO were higher (in terms of % change) than in
309 CH_4 and CO_2 during the sampling period. CO mixing ratios decreased and water vapor mixing
310 ratios increased significantly during the rainy season (June-September). For the entire sampling
311 period, the annual average (\pm one standard deviation) of CH_4 , CO_2 , CO, and water vapor mixing
312 ratios were 2.192 (± 0.066) ppm, 419.3 (± 6.0) ppm, 0.50 (± 0.23) ppm, and 1.73 (± 0.66) %,
313 respectively. The relative standard deviation (RSD) for the annual average of CH_4 , CO_2 and CO

314 were thus 3 %, 1.4 % and 46 %, respectively. The **RSD** at Mauna Loa were CH₄: 6 % and CO₂:
315 0.5 % and at Waliguan were CH₄: 0.48 %, CO₂: 0.9 %. The high variability in the annual mean,
316 notably for CO in Bode could be indicative of the seasonality of emission sources and
317 meteorology. The annual CH₄ and CO₂ mixing ratios were compared to the historical
318 background site (Mauna Loa Observatory, Hawaii, USA) and the background site (Waliguan,
319 China) in Asia, which will provide insight on spatial differences. The selection of neighboring
320 urban and semi-urban sites, where many emission sources are typical for the region, for
321 comparison provides information on relative differences (higher/lower), which will help in
322 investigating possible local emission sources in the valley. As expected, annual mean of CH₄ and
323 CO₂ mixing ratios in the Kathmandu Valley were higher than the levels observed at background
324 sites in the region and elsewhere (Table 4). We performed a significance test at 95 % confidence
325 level (t-test) of the annual mean values between the sites to evaluate whether the observed
326 difference is statistically significant ($p < 0.05$), which was confirmed for the annual mean CH₄
327 and CO₂ between Bode and Mauna Loa, and between Bode and Waliguan. CH₄ was nearly 20 %
328 higher at Bode than at Mauna Loa observatory (1.831 ± 0.110 ppm) (Dlugokencky et al., 2017)
329 and **calculated (ca.)** 17 % higher than at Mt. Waliguan (1.879 ± 0.009 ppm) for the same
330 observation period (Dlugokencky et al., 2016). The slightly higher CH₄ mixing ratios between at
331 Bode and Waliguan than at Mauna Loa Observatory could be due to rice farming as a key source
332 of CH₄ in this part of Asia. Thus, it could be associated with such agricultural activities in this
333 region. Similarly, the annual average CH₄ at Bode during 2013-14 was found comparable to an
334 urban site in Ahmedabad (1.880 ± 0.4 ppm, i.e., **RSD**: 21.3 %) in India for 2002 (Sahu and Lal,
335 2006) and 14 % higher than in Shadnagar (1.92 ± 0.07 ppm, i.e., **RSD**: 3.6 %), a semi-urban site
336 in Telangana state (~70 km north from Hyderabad city) during 2014 (Sreenivas et al., 2016).
337 Likewise, the difference between annual mean mixing ratios at Bode (419.3 ± 6.0 ppm, 1.4%
338 **RSD**) vs. Mauna Loa (396.8 ± 2.0 ppm, 0.5% **RSD**) and Bode vs. Waliguan (397.7 ± 3.6 ppm,
339 0.9% variability) (Dlugokencky et al., 2016a) was statistically significant ($p < 0.05$).

340

341 The high CH₄ and CO₂ mixing ratios at Bode in comparison to Ahmedabad and Shadnagar could
342 be due to more than 115 coal-biomass fired brick kiln, some of them are located near the site
343 (less than 4 km) and confinement of pollutants within the Valley due to bowl shaped topography

344 of the Kathmandu Valley. Although Ahmedabad is a big city with high population larger than
345 Kathmandu Valley, the measurement site is far from the nearby heavy polluting industries and
346 situated in plains, where ventilation of pollutants would be more efficient as opposed to the
347 Kathmandu Valley. The major polluting sources were industries, residential cooking and
348 transport sector in Ahmedabad (Chandra et al., 2016). Shadnagar is a small town with a
349 population of 0.16 million and major sources were industries (small-medium) and biomass
350 burning in residential cooking (Sreenivas et al., 2016).

351 The monthly average of CO₂ mixing ratios in 2015 in Chanban (Aug: 403.4, Sep: 399.1 ppm)
352 were slightly higher than the background sites at Mauna Loa Observatory (Aug: 398.89 ppm,
353 Sep: 397.63 ppm) (NOAA, 2015) and Mt. Waliguan (Aug: 394.55 ppm, Sep: 397.68 ppm)
354 (Dlugokencky et al., 2016a). For these two months in 2015, CH₄ mixing ratios were also higher
355 in Bode (Aug: 2.281 ppm, Sep: 2.371 ppm) and Chanban (Aug: 2.050 ppm, Sep: 2.102 ppm)
356 compared to Mauna Loa Observatory (Aug: 1.831 ppm, Sep: 1.846 ppm) (Dlugokencky et al.,
357 2017)) and Mt. Waliguan (Aug: 1.915 ppm, 1.911 ppm) (Dlugokencky et al., 2016). The small
358 differences in CO₂ between Chanban and background sites mentioned above indicate the smaller
359 number of and/or less intense CO₂ sources at Chanban during these months because of the lack
360 of burning activities due to rainfall in the region. The garbage and agro-residue burning activities
361 were also absent or reduced around Bode during the monsoon period. However, high CH₄ values
362 in August and September in Bode, Chanban and Mt. Waliguan in comparison to Mauna Loa
363 Observatory may indicate the influence of CH₄ emission from paddy fields in the Asian region.

364

365 **3.2 Monthly and Seasonal variations**

366 Figure 3 shows the monthly box plot of hourly CH₄, CO₂, CO and water vapor observed for a
367 year in Bode. Monthly and seasonal averages of CH₄ and CO₂ mixing ratios at Bode are
368 summarized in Table 2 and 3. CH₄ were lowest during May-July (ranges from 2.093-2.129 ppm)
369 period and highest during August-September (2.274-2.301 ppm), followed by winter. In addition
370 to the influence of active local sources, the shallow boundary layer in winter was linked to
371 elevated concentrations (Panday and Prinn, 2009; Putero et al., 2015, Mues et al., 2017). The low
372 CH₄ values from May to July may be associated with the absence of brick kiln and frequent

373 rainfall in these months. Brick kiln were operational during January to April. Rainfall also leads
374 to suppression of open burning activities in the valley (see Figure 2b). The CH₄ was slightly
375 higher (statistically significant, $p < 0.05$) in monsoon season (July–September) than in the pre-
376 monsoon season (unlike CO₂ which was higher in pre-monsoon), and could be associated with
377 the addition of CH₄ flux from the water-logged rice paddies (Goroshi et al., 2011). There was a
378 visible drop in CH₄ from September to October but remained consistently over 2.183 ppm from
379 October to April with little variation between these months. Rice-growing activities are minimal
380 or none in October and beyond, and thus may be related to the observed dip in CH₄ mixing ratio.

381 Comparison of seasonal average CH₄ mixing ratios at Bode and Shadnagar (a semi-urban site in
382 India) indicated that CH₄ mixing ratios at Bode were higher in all seasons than at Shadnagar:
383 pre-monsoon (1.89 ± 0.05 ppm), monsoon (1.85 ± 0.03 ppm), post-monsoon (2.02 ± 0.01 ppm),
384 and winter (1.93 ± 0.05 ppm) (Sreenivas et al., 2016). The possible reason for lower CH₄ at
385 Shadnagar in all seasons could be associated with geographical location and difference in local
386 emission sources. The highest CH₄ mixing ratio in Shadnagar was reported in post-monsoon
387 which was associated with harvesting in the Kharif season (July – October), while the minimum
388 was in monsoon. Shadnagar is a relatively small city (population: ~0.16 million) compared to
389 Kathmandu Valley and the major local sources which may have influence on CH₄ emissions
390 include bio-fuel, agro-residue burning and residential cooking.

391 The seasonal variation in CO₂ could be due to (i) the seasonality of major emission sources such
392 as brick kilns (ii) seasonal growth of vegetation (CO₂ sink) (Patra et al., 2011) and (iii)
393 atmospheric transport associated with regional synoptic atmospheric circulation (monsoon
394 circulation and westerly disturbance in spring season) which could transport regional emission
395 sources from vegetation fire and agriculture residue burning (Putero et al., 2015), and a local
396 mountain-valley circulation effect (Kitada and Regmi, 2003; Panday et al., 2009). The
397 concentrations of most pollutants in the region are lower during the monsoon period (Sharma et
398 al., 2012, Marinoni, 2013; Putero et al., 2015) because frequent and heavy rainfall suppresses
399 emissions sources. We saw a drop in the CO₂ mixing ratio during the rainfall period due to
400 changes in various processes such as enhanced vertical mixing, uptake of CO₂ by vegetation and
401 soils, and, where relevant, reduction in combustion sources. CO₂ can also dissolve into rainfall,

402 forming carbonic acid, which may lead to a small decrease in the CO₂ mixing ratio as has been
403 observed during **high** intensity rainfall (Chaudhari et al., 2007; Mahesh et al., 2014). Monsoon is
404 also the growing season with higher CO₂ assimilation by plants than other seasons (Sreenivas et
405 al., 2016). In contrast, winter, pre-monsoon and post-monsoon season experiences an increase in
406 emission activities in the Kathmandu Valley (Putero et al., 2015).

407
408 The CO₂ mixing ratios were in the range of 376 - 537 ppm for the entire observation period.
409 Differences with CH₄ were observed in September and October where CO₂ was increasing
410 (mean/median) in contrast to CH₄ which showed the opposite trend. The observed increase in
411 CO₂ after October may be related to **little** or no rainfall, which results in the absence of rain-
412 washout and/or no suppression of active emission sources such as open burning activities.
413 However, the reduction in CH₄ after October could be due to reduced CH₄ emissions from paddy
414 fields, which were high in August-September. CO₂ remains relatively low during July-August,
415 but it is over 420 ppm from January to May. Seasonal variation of CO₂ in Bode was similar in
416 seasonal variation but the values are higher than the values observed in Shadnagar, India
417 (Sreenivas et al., 2016).

418 The variations in CO were more distinct than CH₄ and CO₂ during the observation period
419 (Figure 3). The highest CO values were observed from January-April (0.71-0.91 ppm). The
420 seasonal mean of CO mixing ratios at Bode were: pre-monsoon (0.60 ± 0.36 ppm), monsoon
421 (0.26 ± 0.09 ppm), post-monsoon (0.40 ± 0.15 ppm), and winter (0.76 ± 0.43 ppm). The
422 maximum CO was observed in winter, unlike CO₂ which was maximum in pre-monsoon. The
423 high CO in winter was due to the presence of strong local pollution sources (Putero et al., 2015)
424 and shallow mixing layer heights. The addition of regional forest-fire and agro-residue burning
425 augmented CO₂ mixing ratios in pre-monsoon. The water vapor mixing ratio showed a seasonal
426 pattern opposite of CO, with a maximum in monsoon (2.53 %) and minimum in winter (0.95 %),
427 and intermediate values of 1.56 % in pre-monsoon and 1.55 % in post-monsoon season.

428 There were days in August-September when the CH₄ increased by more than 3 ppm (Figure 2).
429 Enhancement in CO₂ was also observed during the same time period. In the absence of tracer
430 model simulations, the directionality of the advected air masses is unclear. Figure 4 shows that

431 during these two months, CO₂ mixing ratios were particularly high (> 450 CO₂ and > 2.5 ppm
432 CH₄) with the air masses coming from the East-Northeast (E-NE). CO during the same period
433 was not enhanced and didn't show any particular directionality compared to CH₄ and CO₂
434 (Figure 4c). Areas E-NE to Bode are predominantly irrigated (rice paddies) during August-
435 September, and sources such as brick kilns were not operational during this time period. Goroshi
436 et al. (2011) reported that June to September is a growing season for rice paddies in South Asia
437 with high CH₄ emissions during these months and observed a peak in September in the
438 atmospheric CH₄ column over India. Model analysis also points to high methane emissions in
439 September which coincides with the growing period of rice paddies (Goroshi et al., 2011, Prasad
440 et al., 2014). The CH₄ mixing ratios at Bode in January (2.233 ± 0.219 ppm) and July ($2.129 \pm$
441 0.168 ppm) were slightly higher than the observation in Darjeeling (January: 1.929 ± 0.056
442 ppm; July: 1.924 ± 0.065 ppm), a hill station of eastern Himalaya (Ganesan et al., 2013). The
443 higher CH₄ values in January and July at Bode compared to Darjeeling could be because of the
444 influence of local sources, in addition to the shallow boundary layer in Kathmandu Valley. Trash
445 burning and brick kilns are two major sources from December until April in the Kathmandu
446 Valley while emission from paddy fields occurs during July-September in the Kathmandu
447 Valley. In contrast, the measurement site in Darjeeling was located at higher altitude (2194 masl)
448 and was less influenced by the local emission. The measurement in Darjeeling reflected a
449 regional contribution. There are limited local sources in Darjeeling such as wood biomass
450 burning, natural gas related emission and vehicular emission (Ganesan et al., 2013).

451 The period between January and April had generally higher or the highest values of CO₂, CH₄
452 and CO at Bode. The measurement site was impacted mainly by local Westerly-Southwesterly
453 winds (W-SW) and East-Southeast (E-SE). The W-SW typically has a wind speed in the range
454 $\sim 1 - 6$ m s⁻¹ and was active during late morning to afternoon period ($\sim 11:00$ to $17:00$ NST,
455 supplementary information Figure S2 and S3). Major cities in the valley such as Kathmandu
456 Metropolitan City and Lalitpur Sub-metropolitan City are W-SW of Bode (Figure 1c). Wind
457 from E-SE were generally calm (≤ 1 m s⁻¹) and observed only during night and early morning
458 hours ($21:00$ to $8:00$ NST). The mixing ratio of all three species in air mass from the E-SE was
459 significantly higher than in the air mass from W-SW (Figure 4). There are 10 biomass co-fired
460 brick kilns and Bhaktapur Industrial Estate located within 1-4 km E-SE from Bode (Sarkar et al.,

461 2016). The brick kilns were only operational during January-April. Moreover, there were over
462 100 brick kilns operational in the Kathmandu Valley (Putero et al., 2015) which use low-grade
463 lignite coal imported from India and biomass fuel to fire bricks in inefficient kilns (Brun, 2013).

464 Fresh emissions from the main city center were transported to Bode during daytime by W-SW
465 winds which mainly include vehicular emission. Compared to monsoon months (June-August),
466 air mass from W-SW had higher values of all three species (Figure 4) during winter and pre-
467 monsoon months. This may imply that in addition to vehicular emission, there are other potential
468 sources which were exclusively active during these dry months. Municipal trash burning is also
469 common in the Kathmandu Valley, with a reported higher frequency from December to February
470 (Putero et al., 2015). The frequency in the use of captive power generator sets are highest during
471 the same period, which is another potential source contributing to air coming from W-SW
472 direction (World Bank, 2014; Putero et al., 2015).

473 Regional transport of pollutants into the Kathmandu Valley was reported by Putero et al. (2015).
474 To relate the influence of synoptic circulation with the observed variability in BC and O₃ in the
475 Kathmandu Valley, 5-day back trajectories (of air masses arriving in the Kathmandu Valley)
476 were computed by Putero et al., (2015) using the HYSPLIT model. These individual trajectories
477 which were initialized at 600 hPa, for the study period of one year and were clustered into nine
478 clusters. Of the identified clusters, the most frequently observed clusters during the study period
479 were the Regional and Westerly cluster or circulation (22 % and 21 %). The trajectories in the
480 regional cluster originate within 10° x 10° around the Kathmandu Valley, whereas the majority of
481 trajectories in this westerly cluster originated broadly around 20-40° N, ~60° E. Putero et al
482 (2015) found that the regional and westerly synoptic circulation were favorable for high values
483 of BC and O₃ in the Kathmandu Valley. Other sources of CO₂ and CH₄ could be due to
484 vegetation fires which were also reported in the region surrounding the Kathmandu Valley
485 during the pre-monsoon months (Putero et al., 2015). Similarly, high pollution events, peaking in
486 the pre-monsoon, were observed at Nepal Climate Observatory-Pyramid (NCO-P) near Mt.
487 Everest, which have been associated with vegetation fires in the Himalayan foothills and
488 northern IGP region (Putero et al., 2014). MODIS derived forest counts (Figure 5), which also
489 indicated high frequency of forest fire and farm fires from February to April and also during

490 post-monsoon season. It is interesting that the monthly mean CO₂ mixing ratio was maximum in
491 April (430 ± 27 ppm) which could be linked to the fire events. It is likely that the westerly winds
492 ($> 2.5 - 4.5 \text{ m s}^{-1}$) during the daytime (supplementary information Figure S2, S3) bring additional
493 CO₂ from vegetation fires and agro-residue burning in southern plains of Nepal including the
494 IGP region (Figure 5). Low values of CO₂ and CH₄ during June-July (Figure 3) were coincident
495 with the rainy season, and sources such as brick kiln emission, trash burning, captive power
496 generators, and regional agriculture residue burning and forest fires are weak or absent during
497 these months.

498 3.3 Diurnal Variation

499 Figure 6 shows the average seasonal diurnal patterns of CH₄, CO₂, CO, and water vapor mixing
500 ratios observed at Bode for four seasons. All the three gas species had a distinct diurnal patterns
501 in all seasons, characterized by maximum values in the morning hours (peaked around 7:00-
502 9:00), afternoon minima around 15:00-16:00, and a gradual increase through the evening until
503 next morning. There was no clear evening peak in CH₄ and CO₂ mixing ratios whereas CO
504 shows an evening peak around 20:00. The gradual increase of CO₂ and CH₄ in the evening in
505 contrast to the increase until evening peak traffic hours and later decay of CO may be indicative
506 of a few factors. As pointed out earlier, after the peak traffic hours, there are no particularly
507 strong sources of CO, especially in the monsoon and post-monsoon season. It is also likely that
508 some of the CO is decayed due to nighttime katabatic winds which replace polluted air masses
509 with cold and fresh air from the nearby mountain (Panday and Prinn, 2009). As for the CO₂, the
510 biosphere respiration at night in the absence of photosynthesis can add additional CO₂ to the
511 atmosphere which especially in the very shallow nocturnal boundary layer may explain part of
512 the increase of the CO₂ mixing ratio. The well-defined morning and evening peaks observed in
513 CO mixing ratios are associated with the peaks in traffic and residential activities. The CH₄ and
514 CO₂ showed pronounced peaks in the morning hours (07:00 - 09:00) in all seasons with almost
515 the same level of seasonal average mixing ratios. CO had a prominent morning peak in winter
516 and pre-monsoon season, but the peak was significantly lower in monsoon and post-monsoon.
517 The CO ($\sim 1 - 1.4$ ppm) around 08:00 - 09:00 in winter and pre-monsoon were nearly 3-4 times
518 higher than in monsoon and post-monsoon season. It appears that CH₄ and CO₂ mixing ratios

519 were continuously building up at night until the following morning peak in all seasons. The
520 similar seasonal variations in CH₄ and CO₂ across all seasons could be due to their long-lived
521 nature, as compared to CO, whose diurnal variations are strongly controlled by the evolution of
522 the boundary layer. Kumar et al. (2015) also reported morning and evening peaks and an
523 afternoon low in CO₂ mixing ratios in industrial, commercial, and residential sites in Chennai in
524 India. The authors also found high early morning CO₂ mixing ratios at all sites and attributed it
525 to the temperature inversion and stable atmospheric condition.

526
527 The daytime low CH₄ and CO₂ mixing ratios were due to (i) elevated mixing layer height in the
528 afternoon (Figure 7), (ii) development of upslope wind circulation in the valley, and (iii)
529 development of westerly and southwesterly winds which blows through the valley during the
530 daytime from around 11:00 to 17:00 (supplementary information Figure S2), all of which aid in
531 dilution and ventilation of the pollutants out of the valley (Regmi et al., 2003; Kitada and Regmi,
532 2003; Panday and Prinn, 2009). In addition, the daytime CO₂ minimum in the summer monsoon
533 is also associated with high photosynthetic activities in the valley as well as in the broader
534 surrounding region. In the nighttime and early morning, the mixing layer height was low (only
535 around 200-300 m in all seasons) and remains stable for almost 17 hours a day. In the daytime it
536 grows up to 800-1200 m for a short time (ca. from 11:00 to 18:00) (Mues et al., 2017). Therefore
537 the emissions from various activities in the evening after 18:00 (cooking and heating, vehicles,
538 trash burning, and bricks factories in the night and morning) were trapped within the collapsing
539 and shallow boundary layer, and hence mixing ratios were high during evening, night and
540 morning hours. Furthermore, plant and soil respiration also increases CO₂ mixing ratio during
541 the night (Chandra et al., 2016). However, Ganesan et al. (2013) found a distinct diurnal cycle of
542 CH₄ mixing ratios with twin peaks in the morning (7:00 - 9:00), and afternoon (15:00 - 17:00)
543 and a nighttime low in winter but no significant diurnal cycle in the summer of 2012 in
544 Darjeeling, a hill station (2194 masl) in the eastern Himalaya. The authors described that the
545 morning peaks could be due to the radiative heating of the ground in the morning, which breaks
546 the inversion layer formed during night, and as a result, pollutants are ventilated from the
547 foothills up to the site. The late afternoon peaks match wind direction and wind speed (upslope
548 winds) that could bring pollution from the plains to the mountains.

549 The diurnal variation of CO is also presented along with CO₂ and CH₄ in Figure 6c. CO is an
550 indicator of primary air pollution. Although the CO mixing ratio showed distinct diurnal pattern,
551 it was different from the diurnal patterns of CO₂ and CH₄. CO diurnal variation showed distinct
552 morning and evening peaks, afternoon minima, and a nighttime accumulation or decay.
553 Nighttime accumulation in CO was observed only in winter and pre-monsoon and decay or
554 decrease in monsoon season and post-monsoon season (Figure 7). The lifetime of CO (weeks to
555 months) is very long compared to the ventilation timescales for the valley, so the different
556 diurnal cycles would be due to differences in nighttime emissions. While the biosphere respire
557 at night which may cause a notable increase in CO₂ in the shallow boundary layer, most CO
558 sources (transport sector, residential cooking) except brick kilns remain shut down or less active
559 at night. This also explains why nighttime values of CO drop less in the winter and pre-monsoon
560 than in other seasons. Furthermore, the prominent morning peaks of CO in pre-monsoon and
561 winter compared to other seasons results from nighttime accumulation, additional fresh
562 emissions in the morning and recirculation of the pollutants due to downslope katabatic winds
563 (Pandey and Prinn, 2009; Panday et al., 2009). Pandey and Prinn (2009) observed nighttime
564 accumulation and gradual decay during the winter (January 2005). The measurement site in
565 Pandey and Prinn (2009) was near the urban core of the Kathmandu Valley and had significant
566 influence from the vehicular sources all over the season including the winter season. Bode lies in
567 close proximity to the brick kilns which operate 24 hours during the winter and pre-monsoon
568 period. Calm southeasterly winds are observed during the nighttime and early morning (ca. 22:00
569 – 8:00) in pre-monsoon and winter, which transport emissions from brick kiln to the site (Sarkar
570 et al., 2016). Thus the gradual decay in CO was not observed in Bode.

571 The timing of the CO morning peak observed in this study matches with observations by Panday
572 et al. (2009). They also found CO morning peak at 8:00 in October 2004 and at 9:00 in January
573 2005. The difference could be linked to the boundary layer stability. As the sun rises later in
574 winter, the boundary layer stays stable for a longer time in winter keeping mixing ratios higher in
575 morning hours than in other seasons with an earlier sunrise.

576 The morning peaks of CO₂ and CH₄ mixing ratios occurred around 6:00 -7:00 local time in the
577 pre-monsoon, monsoon, and post monsoon season, whereas in winter their peaks are delayed by

578 1-2 hours in the morning; CH₄ at 8:00 and CO₂ at 9:00. The CO showed that its morning peak
579 was delayed compared to CO₂ and CH₄ morning peaks by 1-2 hour in pre-monsoon, monsoon
580 and post-monsoon (at 8:00) and in winter (at 9:00). The occurrence of morning peaks in CO₂ and
581 CH₄ 1-2 hours earlier than CO is interesting. This could be due to the long lifetimes and
582 relatively smaller local sources of CH₄ and CO₂, as CO is mainly influenced by emissions from
583 vehicles during rush hour, as well as from biomass and trash burning in the morning hours. Also,
584 CO increases irrespective of change in the mixing layer (collapsing or/rising, Figure 7) but CO₂
585 and CH₄ start decreasing only after the mixing layer height starts to rise. Recently, Chandra et al.
586 (2016) also reported that the CO₂ morning peak occurred earlier than CO in observations in
587 Ahmedabad City India. This was attributed to CO₂ uptake by photosynthetic activities after
588 sunrise but CO kept increasing due to emissions from the rush hour activities.

589 The highest daytime minimum of CO₂ was observed in the pre-monsoon followed by winter
590 (Figure 6b). The higher daytime minimum of CO₂ mixing ratios in the pre-monsoon season than
591 in other seasons, especially winter, is interesting. The local emission sources are similar in pre-
592 monsoon and winter and the boundary layer is higher (in the afternoon) during the pre-monsoon
593 (~1200 m) than in winter (~900 m) (Mues et al., 2017). Also, the biospheric activity in the
594 region is reported to be higher in the pre-monsoon (due to high temperature and solar radiation)
595 than winter (Rodda et al., 2016). Among various possible causes, transport of CO₂ rich air from
596 outside the Kathmandu Valley has been hypothesized as a main contributing factor, due to
597 regional vegetation fires combined with westerly mesoscale to synoptic transport Putero et al.
598 (2015). In monsoon and post-monsoon seasons, the minimum CO₂ mixing ratios in the afternoon
599 drops down to 390 ppm, this was close to the values observed at the regional background sites
600 Mauna Loa and Waliguan.

601

602 **3.4 Seasonal interrelation of CO₂, CH₄ and CO**

603 The Pearson's correlation coefficient (r) between CO₂ and CO was strong in winter (0.87),
604 followed by monsoon (0.64), pre-monsoon (0.52) and post-monsoon (0.32). The higher
605 coefficient in winter indicates that common or similar sources for CO₂ and CO and moderate
606 values in pre-monsoon and monsoon indicates the likelihood of different sources. To avoid the

607 influence of strong diurnal variations observed in the valley, daily averages, instead of hourly,
608 were used to calculate the correlation coefficients. The correlation coefficients between daily
609 CH₄ and CO₂ for four seasons are as follows: winter (0.80), post-monsoon (0.74), pre-monsoon
610 (0.70) and monsoon (0.22). A semi-urban measurement study in India also found a strong
611 positive correlation between CO₂ and CH₄ in the pre-monsoon (0.80), monsoon (0.61), post-
612 monsoon (0.72) and winter (0.8) (Sreenivas et al., 2016). It should be noted here that Sreenivas
613 et al., (2006) used hourly average CO₂ and CH₄ mixing ratios. The weak monsoon correlation at
614 Bode, which is in contrast to Sreenivas et al. (2016), may point to the influence of dominant CH₄
615 emission from paddy field during the monsoon season (Goroshi et al., 2011). Daily CH₄ and CO
616 was also weakly correlated in monsoon (0.34) and post-monsoon (0.45). Similar to CH₄ and
617 CO₂, the correlation between CH₄ and CO were moderate to strong in pre-monsoon (0.76) and
618 winter (0.75).

619 Overall, the positive and high correlations between CH₄ and CO mixing ratios and between CH₄
620 and CO₂ mixing ratios in the pre-monsoon and winter indicate common sources, most likely
621 combustion related sources such as vehicular emission, brick kilns, agriculture fire etc., or the
622 same source regions (i.e. their transport due to regional atmospheric transport mechanisms).
623 Weak correlation, between CH₄-CO₂ and between CH₄-CO, during monsoon season indicates
624 sources other than combustion-related may be active, such as agriculture as a key CH₄ source
625 (Goroshi et al., 2013)

626 **3.5 CO and CO₂ ratio: Potential emission sources**

627 The ratio of the ambient mixing ratios of CO and CO₂ was used as an indicator to help
628 discriminate emission sources in the Kathmandu Valley. The ratio was calculated from the
629 excess (dCO and dCO₂) relative to the background values of ambient CO and CO₂ mixing ratios.
630 The excess value was estimated by subtracting the base value which was calculated as the fifth
631 percentile of the hourly data for a day (Chandra et al., 2016).

632 Average emission ratios from the literature are shown in Table 5, and average ratios of
633 dCO/dCO₂ are shown in Table 6, disaggregated into morning hours, evening hours, and seasonal
634 values. It must be stated that due to the large variance in the calculated ratio from this study
635 (Table 6) as well as the likely variation in the estimated ratio presented in Table 5, the

636 interpretation and conclusion about sources should be cautiously drawn and will be indicative.
637 Higher ratios were found in pre-monsoon (12.4) and winter (15.1) season compared to post-
638 monsoon (8.3) and monsoon (7.5). These seasonal differences in the dCO/dCO_2 ratio are
639 depicted in Figure 8, which shows a clear relationship with the wind direction and associated
640 emissions, with the highest values especially for stronger westerly winds. Compared to the other
641 three seasons, the ratio in winter was also relatively high for air masses from the east, likely due
642 to emissions from brick kilns combined with accumulation during more stagnant meteorological
643 conditions (supplementary information Figure S2, S3). In other seasons, emission emanating
644 from the north and east of Bode were characterized by a dCO/dCO_2 ratio below 15. Air masses
645 from the west and south generally have a ratio from 20 to 50 in all but post-monsoon season,
646 where the ratio sometimes exceeds 50. A ratio of 50 or over is normally due to very inefficient
647 combustion sources (Westerdahl et al., 2009; Stockwell et al., 2016), such as agro-residue
648 burning, which is common during the post-monsoon season in the Kathmandu Valley.

649 For interpretability of emission ratio with sources, the ratio was classified into three categories:
650 (i) 0 – 15, (ii) 15 – 45, and (iii) greater than 45. This classification was based on the observed
651 distribution of emission ratio during the study period (Figure 8) and a compilation of observed
652 emission ratios typical for different sources from Nepal and India (see Table 5). An emission
653 ratio below 15 is likely to indicate residential cooking and diesel vehicles, and captive power
654 generation with diesel-powered generator sets (Smith et al., 2000; ARAI, 2008; World Bank,
655 2014). The emission from brick kilns (FCBTK and Clamp kilns, both common in the Kathmandu
656 Valley), and inefficient, older (built before 2000) gasoline cars fall in between 15 - 45 (ARAI,
657 2008; Weyant et al., 2014; Stockwell et al., 2016). Four-stroke motorbikes and biomass burning
658 activities (mixed garbage, crop-residue and biomass) are one of the least efficient combustion
659 sources, with emission ratios higher than 45 (ARAI, 2008; Westerdahl et al., 2009; Stockwell et
660 al., 2016).

661 Although ratio of dCO/dCO_2 is a weak indicator of sources and the mean ratio has large variance
662 (See Table 6), the conclusions drawn, from using Figure 8 and the above mentioned
663 classification, are not conclusive. The estimated dCO/dCO_2 ratio tentatively indicates that the
664 local plume impacting the measurement site (Bode) from the north and east could be residential

665 and/or diesel combustion. The estimated dCO/dCO_2 ratio of the local plume from the south and
666 west generally falls in the 15-45 range which could indicate emissions from brick kilns and
667 inefficient gasoline vehicles. Very high ratios were also estimated from the south west during the
668 post-monsoon season. Among other possible sources, this may indicate agro-residue open
669 burning.

670 The emission inventory for CO identifies (aggregate for a year) residential, and gasoline related
671 emission from transport sector (Sadavarte et al., 2017, in preparation). The inventory is not yet
672 temporally resolved, so no conclusion can be drawn about the sources with respect to different
673 seasons. From the 1 km x 1 km emission inventory of the Kathmandu Valley for 2011, the
674 estimated sectoral source apportionment of CO is residential (37 %), transport sector (40 %) and
675 industrial (20 %). The largest fraction from the residential sector is cooking (24 %) whereas the
676 majority of transport sector related CO in the Kathmandu Valley is from gasoline vehicles.

677 The dCO/dCO_2 ratio also changes markedly between the morning peak hours (07:00-09:00,
678 except in winter season when the peak occurs during 08:00-09:00) and evening peak hours
679 (19:00-21:00 pm) (Table 6). Morning and evening values were lowest (2.2, 8.0) during the
680 monsoon and highest (11.2, 21.6) in the winter season, which points to the different emission
681 characteristics in these two seasons. This feature is similar to Ahmedabad, India, another urban
682 site in south Asia, where the morning/evening values were lowest (0.9/19.5) in monsoon and
683 highest in winter (14.3/47.2) (Chandra et al., 2016). In the morning period, the ratio generally
684 falls within a narrower range, from less than 1 to about 25, which indicates a few dominant
685 sources, such as cooking, diesel vehicles, and diesel gen-sets (see Figure 9). In the evening
686 period, the range of the ratio is much wider, from less than 1 to more than 100, especially in
687 winter. This is partly due to the shallower boundary layer in winter, giving local CO emissions a
688 chance to build up more rapidly compared to the longer-lived and well-mixed CO_2 , and also
689 indicating the prevalence of additional sources such as brick kilns and agro-residue burning.

690 **3.6 Comparison of CH_4 and CO_2 at semi-urban site (Bode) and rural site (Chanban)**

691 Figure 10 shows time series of hourly average mixing ratios of CH_4 , CO_2 , CO and water vapor
692 observed simultaneously at Bode and Chanban for the period of 15th July to 3rd October 2015.

693 The hourly meteorological parameters observed at Chanban are shown in supplementary Figure
694 S4. The hourly temperature ranges from 14 to 28.5° C during the observation period. The site
695 experienced calm winds during the night and moderate southeasterly winds with hourly
696 maximum speed of up to 7.5 m s⁻¹ during the observation period. The CH₄ mixing ratios at
697 Chanban varied from 1.880 ppm to 2.384 ppm, and generally increased from the last week of
698 July until early September, peaking around 11th September and then falling off towards the end
699 of the month. CO followed a generally similar pattern, with daily average values ranging from
700 0.10 ppm to 0.28 ppm. The hourly CO₂ mixing ratios ranged from 375 to 453 ppm, with day to
701 day variations, but there were no clear pattern as observed in trend like CH₄ and CO mixing
702 ratios.

703 The CH₄, CO₂, and CO mixing ratios were higher in Bode than in Chanban (Figure 10, Table 4),
704 with Chanban approximately representing the baseline of the lower envelope of the Bode levels.
705 The mean CO₂, CH₄ and CO mixing ratios over the entire sampling period of nearly three
706 months at Bode are 3.8 %, 12.1 %, and 64 % higher, respectively, than at Chanban. The
707 difference in the CO₂ mixing ratio could be due to the large uptake of CO₂ in the forested area at
708 Chanban and surrounding regions compared to Bode, where the local anthropogenic emissions
709 rate is higher and less vegetation for photosynthesis. The coincidence between the base values of
710 CO and CH₄ mixing ratios at Bode and the levels observed at Chanban implies that Chanban CO
711 and CH₄ mixing ratios are indicative of the regional background levels. A similar increase in CO
712 and CH₄ mixing ratios at Chanban from July to September was also observed at Bode, which
713 may imply that the regional/background levels in the broader Himalayan foothill region also
714 influences the baseline of the daily variability of the pollutants in the Kathmandu Valley,
715 consistent with Panday and Prinn (2009).

716 Figure 11 shows the comparison of average diurnal cycles of CO₂, CH₄, CO and water vapor
717 mixing ratios observed at Bode and Chanban. The diurnal pattern of CO₂ mixing ratios at both
718 sites is similar, but more pronounced at Bode, with a morning peak around 06:00-07:00, a
719 daytime minimum, and a gradual increase in the evening until the next morning peak. A
720 prominent morning peak at Bode during the monsoon season indicates the influence of local
721 emission sources. The daytime CO₂ mixing ratios are also higher at Bode than at Chanban

722 because of local emissions less uptake of CO₂ for photosynthesis in the valley in comparison to
723 the forested area around Chanban. Like the diurnal pattern of CO₂ depends on the evolution of
724 the mixing layer at Bode, as discussed earlier, it is expected that the mixing layer evolution
725 similarly influences the diurnal CO₂ mixing ratios at Chanban. CO, on the other hand, shows
726 very different diurnal patterns at Bode and Chanban. Sharp morning and evening peaks of CO
727 are seen at Bode, indicating the strong local polluting sources, especially cooking and traffic in
728 the morning and evening peak hours. Chanban, in contrast, only has a subtle morning peak and
729 no evening peak. After the morning peak, CO sharply decreases at Bode but not at Chanban. The
730 growth of the boundary layer after sunrise and entrainment of air from the free troposphere, with
731 lower CO mixing ratios, causes CO to decrease sharply during the day at Bode. At Chanban, on
732 the other hand, since the mixing ratios are already more representative of the local and regional
733 background levels which will also be prevalent in the lower free troposphere, CO does not
734 decrease notably during the daytime growth of the boundary layer as observed at Bode.

735 Similarly, while there is very little diurnal variation in the CH₄ mixing ratios at Chanban, there is
736 a strong diurnal cycle of CH₄ at Bode, similar to CO₂ there. At Chanban, the CH₄ mixing ratio
737 only shows a weak minimum at around 11:00, a slow increase during the day until a its peak
738 around 22:00, followed by a slow decrease during the night and a more rapid decrease through
739 the morning. The cause of this diurnal pattern at Chanban is presently unclear, but the levels
740 could be representative of the regional background throughout the day and show only limited
741 influences of local emissions.

742

743 4. **Conclusions**

744 A cavity ring down spectrometer (Picarro G2401, USA) was used to measure ambient CO₂, CH₄,
745 CO, and water vapor mixing ratios at a semi-urban site (Bode) in the Kathmandu Valley for a
746 year. This was the first 12-months of continuous measurements of these four species in the
747 Kathmandu Valley in the foothills of the central Himalaya. Simultaneous measurement was
748 carried out at a rural site (Chanban) for approximately 3 months to evaluate urban-rural
749 differences.

750 The measurement also provided an opportunity to establish diurnal and seasonal variation of
751 these species in one of the biggest metropolitan cities in the foothills of Himalayas. Annual
752 average of the mixing ratio of CH₄ and CO₂ in Bode revealed that they were higher than the
753 mixing ratios at the background sites such as the Mauna Loa, USA and Mt. Waliguan, China, as
754 well as higher than urban/semi-urban sites in nearby regions such as Ahmedabad and Shadnagar
755 in India. These comparisons highlight potential sources of CH₄ and CO₂ in the Kathmandu
756 Valley, such as brick kilns in the valley.

757 Polluted air masses were transported to the site mainly by two major local wind circulation
758 patterns, East-South and North-East and West-Southwest throughout the observation period.
759 Strong seasonality was observed with CO compared to CO₂ and CH₄. Winter and pre-monsoon
760 high CO are linked to emission sources active in these seasons only and are from east-southeast
761 and west-southwest. Emission from the east-southeast are most likely related to brick kilns
762 (winter and pre-monsoon), which are in close proximity to Bode. Major city-centers are located
763 in the west-southwest of Bode (vehicular emission) which impact the site all-round the year,
764 although higher during winter season. Winter high was also observed with CO₂ and CH₄, which
765 are mostly local influence of brick kilns, trash burning and emission from city-center. Nighttime
766 and early morning accumulation of pollutants in winter due to a shallow stable mixing height (ca.
767 200 m) also contribute to elevated levels than other seasons. Diurnal variation across all seasons
768 indicates the influence of rush-hour emissions related to vehicles and residential emissions. The
769 evolution of the mixing layer height (200 -1200 m) was a major factor which controls the
770 morning-evening peak, afternoon low and night-early morning accumulation or decay. Thus the
771 geographical setting of the Kathmandu Valley and its associated meteorology play a key role in
772 the dispersion and ventilation of pollutants in the Kathmandu Valley. The ratio of dCO/dCO₂
773 across different seasons and wind directions suggested that emissions from inefficient gasoline
774 vehicles, brick kilns, residential cooking and diesel combustion are likely to impact Bode.

775 The differences in mean values for urban-rural measurements at Bode and Chanban is highest for
776 CO (64 %) compared to CO₂ (3.8 %) and CH₄ (12 %). Low values of CH₄ and CO₂ mixing ratios
777 at the Chanban site could represent a regional background mixing ratios.

778 This study has provided valuable information on key greenhouse gases and air pollutants in the
779 Kathmandu Valley and the surrounding regions. These observations can be useful as ground-
780 truthing for evaluation of satellite measurements, as well as climate and regional air quality
781 models. The overall analysis presented in the paper will contribute along with other recent
782 measurement and analysis to providing a sound scientific basis for reducing emissions of
783 greenhouse gases and air pollutants in the Kathmandu Valley.

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Table 1. Instruments and sampling at Bode (semi-urban site) and Chanban (rural site)

Site	Instrument	Species	Sampling interval	Measurement period	Inlet/sensor height above ground (m)
Bode	i. Cavity ring down spectrometer (Picarro G2401, USA)	CO ₂ , CH ₄ , CO, water vapor	5 sec	06 Mar 2013 - 05 Mar 2014 14 Jul 2015 - 07 Aug 2015	20
	ii. CO monitor (Horriba AP370, USA)	CO	5 min	06 Mar 2013 – 07 June 2013	20
	iii. Ceilometer (Vaisala CL31, Finland)		15-52 min	06 Mar 2013 – 05 Mar 2014	15
	iv. AWS (Campbell Scientific, USA)		1 min		23
	a. CS215	RH, T		06 Mar 2013 – 24 Apr 2013	
	b. CS300 Pyranometer	SR		06 Mar 2013 - 05 Mar 2014 14 Jul 2015 - 07 Aug 2015	
	c. RM Young 05103-5	WD, WS		06 Mar 2013 - 05 Mar 2014 14 July 2015 - 07 Aug 2015	
	v. Airport AWS (Envirodata, Australia)				
	a. TA10	T		18 Jun 2013 – 13 Jan 2013	
	b. RG series	RF		06 Mar 2013 – 15 Dec 2013	
Chanban	i. Cavity ring down spectrometer (Picarro G2401, USA)	CO ₂ , CH ₄ , CO, water vapor	5 sec	15 July 2015 - 03 Oct 2015	3
	ii AWS (Davis Vantage Pro2, USA)	RH, T, SR, WD, WS, RF, P	10 min	14 July 2015 - 07 Aug 2015	2

AWS: Automatic weather station, RH: ambient relative humidity, T: ambient temperature, SR: global solar radiation, WS: wind speed, WD: wind direction, RF: rainfall, P: ambient pressure

Table 2. Summary of monthly average CH₄ and CO₂ mixing ratios observed at Bode, a semi-urban site in the Kathmandu Valley during March 2013 to Feb 2014 [mean, standard deviation (SD), median, minimum (Min.), maximum (Max.) and number of data points of hourly average values]

Month	CH ₄ (ppm)					CO ₂ (ppm)					Data points
	Mean	SD	Median	Min.	Max.	Mean	SD	Median	Min.	Max.	
Mar	2.207	0.245	2.152	1.851	3.094	426.6	26.4	418.3	378.8	510.8	596
Apr	2.183	0.252	2.094	1.848	3.121	430.3	27.4	421.0	397.0	536.9	713
May	2.093	0.174	2.040	1.863	2.788	421.7	22.1	413.4	395.9	511.2	725
Jun	2.061	0.142	2.017	1.869	2.675	417.9	21.3	410.4	390.5	495.7	711
Jul	2.129	0.168	2.074	1.893	2.770	410.3	18.2	406.3	381.0	471.0	500
Aug	2.274	0.260	2.181	1.953	3.219	409.9	22.8	405.3	376.1	493.1	737
Sep	2.301	0.261	2.242	1.941	3.331	414.9	30.2	404.0	375.9	506.2	710
Oct	2.210	0.195	2.156	1.927	2.762	417.0	25.1	411.8	381.9	486.7	743
Nov	2.207	0.203	2.178	1.879	2.705	417.2	20.7	415.7	385.7	478.9	717
Dec	2.206	0.184	2.193	1.891	2.788	417.7	17.3	418.0	386.7	467.6	744
Jan	2.233	0.219	2.198	1.889	2.744	424.8	20.9	422.3	392.7	494.5	696
Feb	2.199	0.223	2.152	1.877	2.895	423.2	22.0	417.9	392.2	484.6	658
Annual	2.1923	0.066	2.140	1.848	3.331	419.3	6.0	413.7	375.9	536.9	

Table 3. Summary of CH₄ and CO₂ mixing ratios at Bode across four seasons during March 2013 to Feb 2014 [seasonal mean, one standard deviation (SD), median, minimum (Min.) and maximum (Max.)]

Season	CH ₄ (ppm)					CO ₂ (ppm)				
	Mean	SD	Median	Min.	Max.	Mean	SD	Median	Min.	Max.
Pre-Monsoon	2.157	0.230	2.082	1.848	3.121	426.2	25.5	417.0	378.8	536.9
Monsoon	2.199	0.241	2.126	1.869	3.331	413.5	24.2	407.1	375.9	506.2
Post-Monsoon	2.210	0.200	2.167	1.879	2.762	417.3	23.1	414.1	381.9	486.7
Winter	2.214	0.209	2.177	1.877	2.895	421.9	20.3	419.3	386.7	494.5

Table 4. Comparison of monthly average CH₄ and CO₂ mixing ratios at a semi-urban and a rural site in Nepal (this study) with other urban and background sites in the region and elsewhere.

Site Setting	Bode, Nepal (Urban)				Chanban, Nepal (Rural)		Mauna Loa, USA (Background) ^c		Waliguan, China (Background) ^d	
	CO ₂	CH ₄	*CO ₂	*CH ₄	*CO ₂	*CH ₄	CO ₂	CH ₄	CO ₂	CH ₄
Unit	Ppm	ppm	Ppm	Ppm	Ppm	Ppm	Ppm	ppm	Ppm	ppm
Mar 2013	426.6	2.207					397.3	1.840	399.5	1.868
Apr	430.3	2.183					398.4	1.837	402.8	1.874
May	421.7	2.093					399.8	1.834	402.5	1.878
Jun	417.9	2.061					398.6	1.818	397.4	1.887
Jul	410.3	2.129					397.2	1.808	393.3	1.888
Aug	409.9	2.274	411.3	2.281	403.4	2.050	395.2	1.819	392.0	1.893
Sep	414.9	2.301	419.9	2.371	399.1	2.102	393.5	1.836	393.1	1.894
Oct	417.0	2.210					393.7	1.836	395.6	1.876
Nov	417.2	2.207					395.1	1.835	397.1	1.875
Dec	417.7	2.206					396.8	1.845	398.6	1.880
Jan 2014	424.8	2.234					397.8	1.842	398.8	1.865
Feb	423.2	2.199					397.9	1.834	401.1	1.878
<i>Annual</i>										
Bode	419.3	2.192								
Mauna Loa							396.8	1.832		
Waliguan									397.7	1.880
Shadnagar (2014) ^a	394.0									
Ahemadabad (2013-2015) ^b	413.0	1.920								

*The monthly values for CO₂ and CH₄ in 2015, ^aSreenivas et al., 2016, ^bChandra et al., 2016, ^cDlugokencky et al., 2017; NOAA, 2015, ^dDlugokencky et al., 2016; Dlugokencky et al., 2016a.

Table 5. Emission ratio of CO/CO₂ (ppb ppm⁻¹) derived from emission factors (mass of gas emitted from per kilogram of fuel burned, “except for the transport sector” which is derived from gram of gases emitted per kilometer distance travelled)

Sectors	Details	CO/CO ₂	Reference
1. Residential/Commercial			
i. LPG		4.8	Smith et al. (2000)
ii. Kerosene		13.4	Smith et al. (2000)
iii. Biomass		52.9 - 98.5	*
iv. Diesel power generators	< 15 year old	5.8	The World Bank (2014)
	> 15 year old	4.5	
2. Transport			
a. Diesel			
i. HCV diesel bus	> 6000cc, 1996-2000	4.9	**
	post 2000 and 2005	5.4	
ii. HCV diesel truck	> 6000cc, post 2000	7.9	
b. Petrol			
i. 4 stroke motorcycle	< 100 cc, 1996-2000	68	
	100-200 cc, Post 2000	59.6	
ii. Passenger cars	< 1000 cc, 1996-2000	42.4	
iii. Passenger cars	< 1000 cc, Post 2000	10.3	
3. Brick industries			
i. BTK fixed kiln		17.2	Weyant et al. (2014)
ii. Clamp brick kiln		33.7	Stockwell et al. (2016)
iii. Zigzag brick kiln		3.9	Stockwell et al. (2016)
4. Open burning			
i. Mixed garbage		46.9	Stockwell et al. (2016)
ii. Crop-residue		51.6	Stockwell et al. (2016)

* Westerdahl et al. (2009)

** http://www.cpcb.nic.in/Emission_Factors_Vehicles.pdf

Table 6. Average (SD) of the ratio of dCO to dCO₂, their Geometric mean (GeoSD) over a period of 3 hours during (a) morning peak (b) evening peak and (c) seasonal (all hours) of the ambient mixing ratios of CO and CO₂ and their lower and upper bound (LB and UB).

Period	Season	Mean (SD)	Median	N	Geomean (GeoSD)	LB	UB
a. Morning hours (7:00-9:00)	Pre-monsoon	7.6 (3.1)	7.8	249	11.3 (1.5)	5.2	24.8
	Monsoon	2.2 (1.6)	1.9	324	9.9 (1.9)	2.7	36.3
	Post-monsoon	3.1 (1.4)	2.8	183	11.1 (1.5)	4.7	26.3
	Winter*	11.2 (4.4)	11	255	11.4 (1.5)	5.3	24.2
b. Evening hours (19:00-21:00)	Pre-monsoon	15.1 (9.0)	12.7	248	10.5 (1.7)	3.5	31.6
	Monsoon	8.0 (5.2)	6.3	323	10.2 (1.8)	3.1	33.5
	Post-monsoon	11.5 (5.6)	10.6	182	11.0 (1.6)	4.4	27.6
	Winter	21.6 (14.1)	18.2	254	10.2 (1.8)	3.1	33.6
c. Seasonal (all hours)	Pre-monsoon	12.2 (13.3)	8.8	1740	8.2 (2.4)	1.4	48.4
	Monsoon	7.5 (13.5)	2.9	2176	5.9 (3.3)	0.5	65.6
	Post-monsoon	8.3 (12.4)	4.4	1289	6.8 (3.0)	0.8	59.2
	Winter	15.1 (13.3)	12.5	1932	9.2 (2.1)	2.0	41.7

*The morning peak was one hour delayed in winter, thus the 8:00-10:00 period data was used in the analysis.

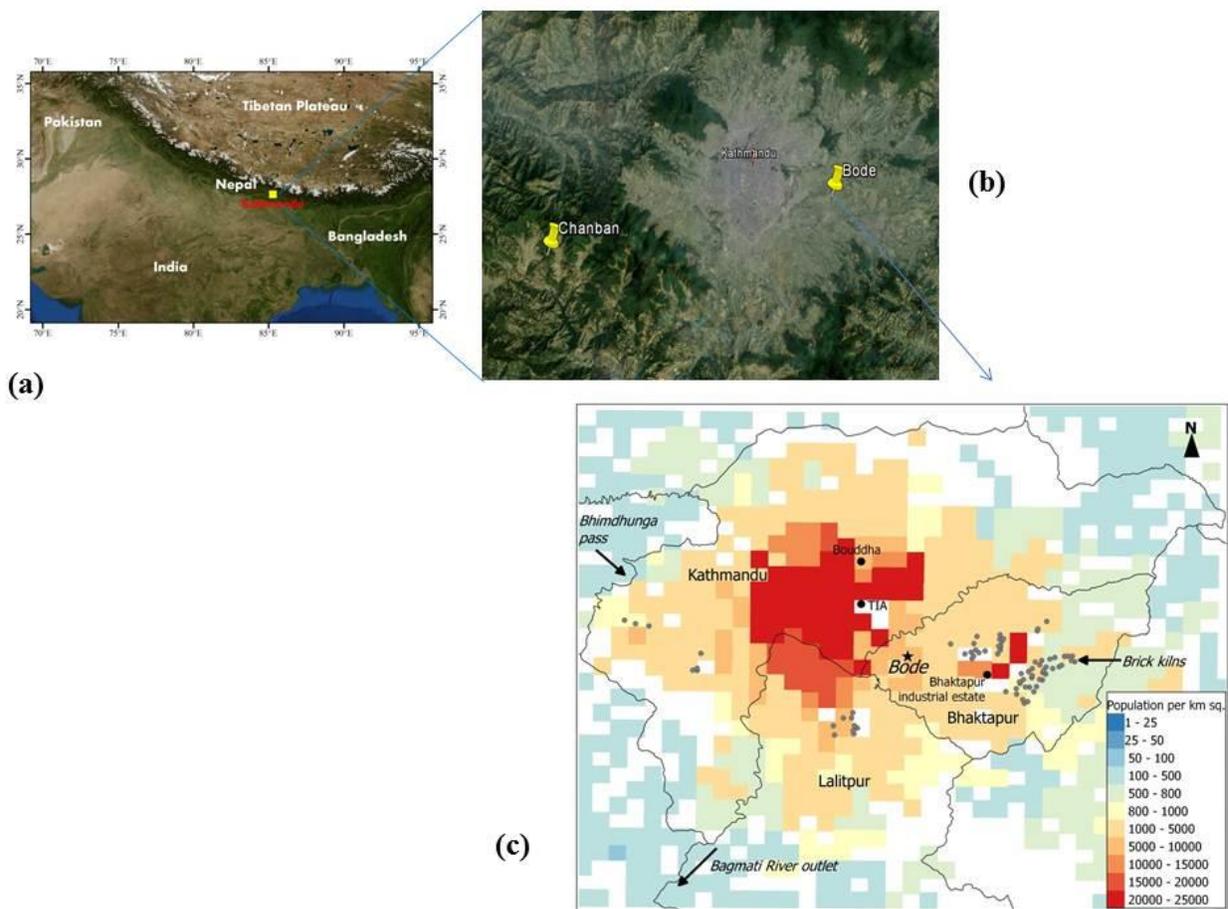


Figure 1. Location of measurement sites: (a) Kathmandu Valley (b) semi-urban measurement site at Bode in Kathmandu Valley, and a rural measurement site at Chanban in Makawanpur district Nepal, (c) general setting of Bode site. Colored grid and TIA represent population density and the Tribhuvan International Airport, respectively.

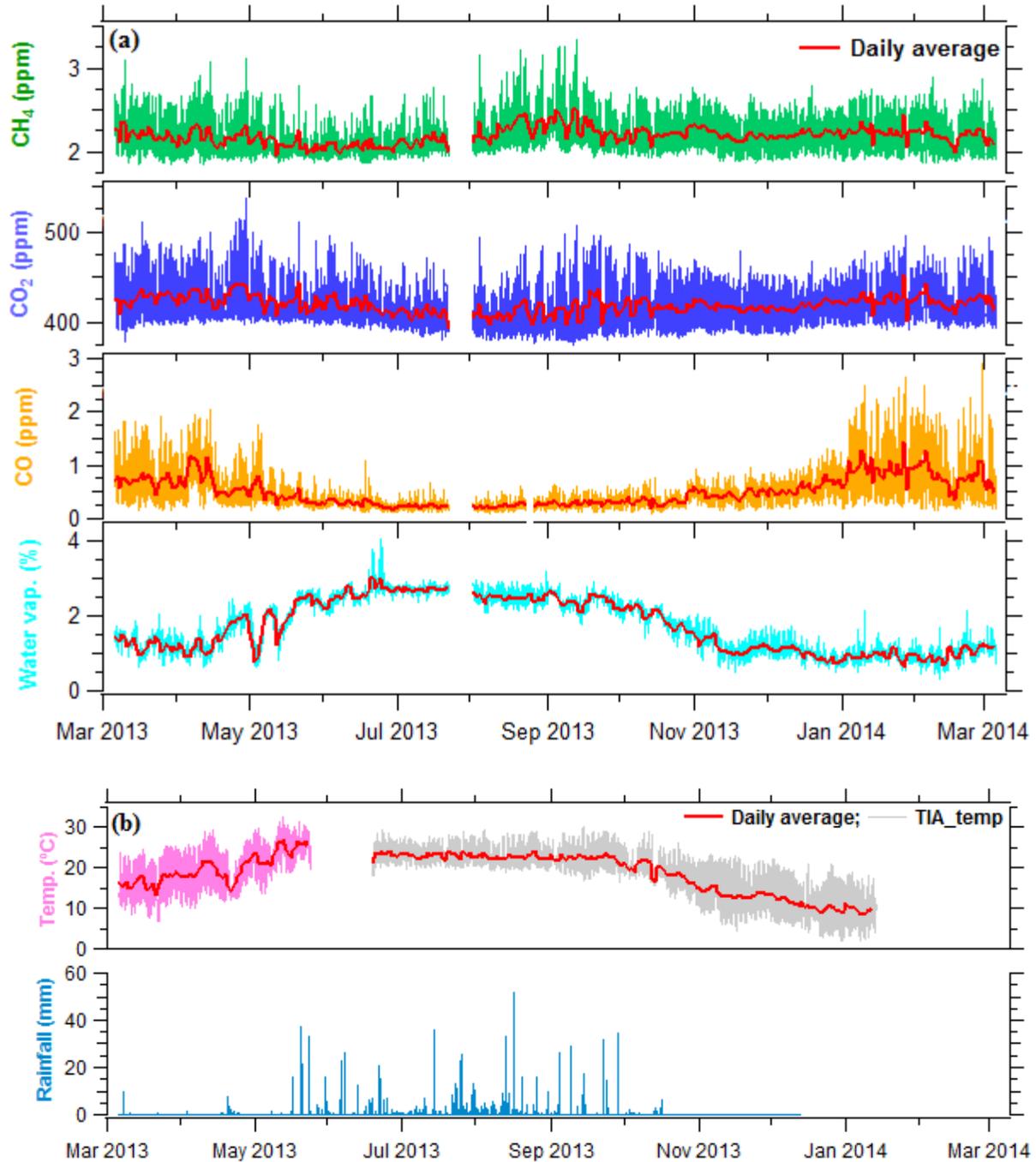


Figure 2. Time series of hourly average (a) mixing ratios of CH₄, CO₂, CO, and water vapor measured with a cavity ring down spectrometer (Picarro G2401) at Bode, and (b) temperature and rainfall monitored at the Tribhuvan International Airport (TIA), ~4 km to the west of Bode site in the Kathmandu Valley, Nepal. Temperature shown in pink color is observed at Bode site.

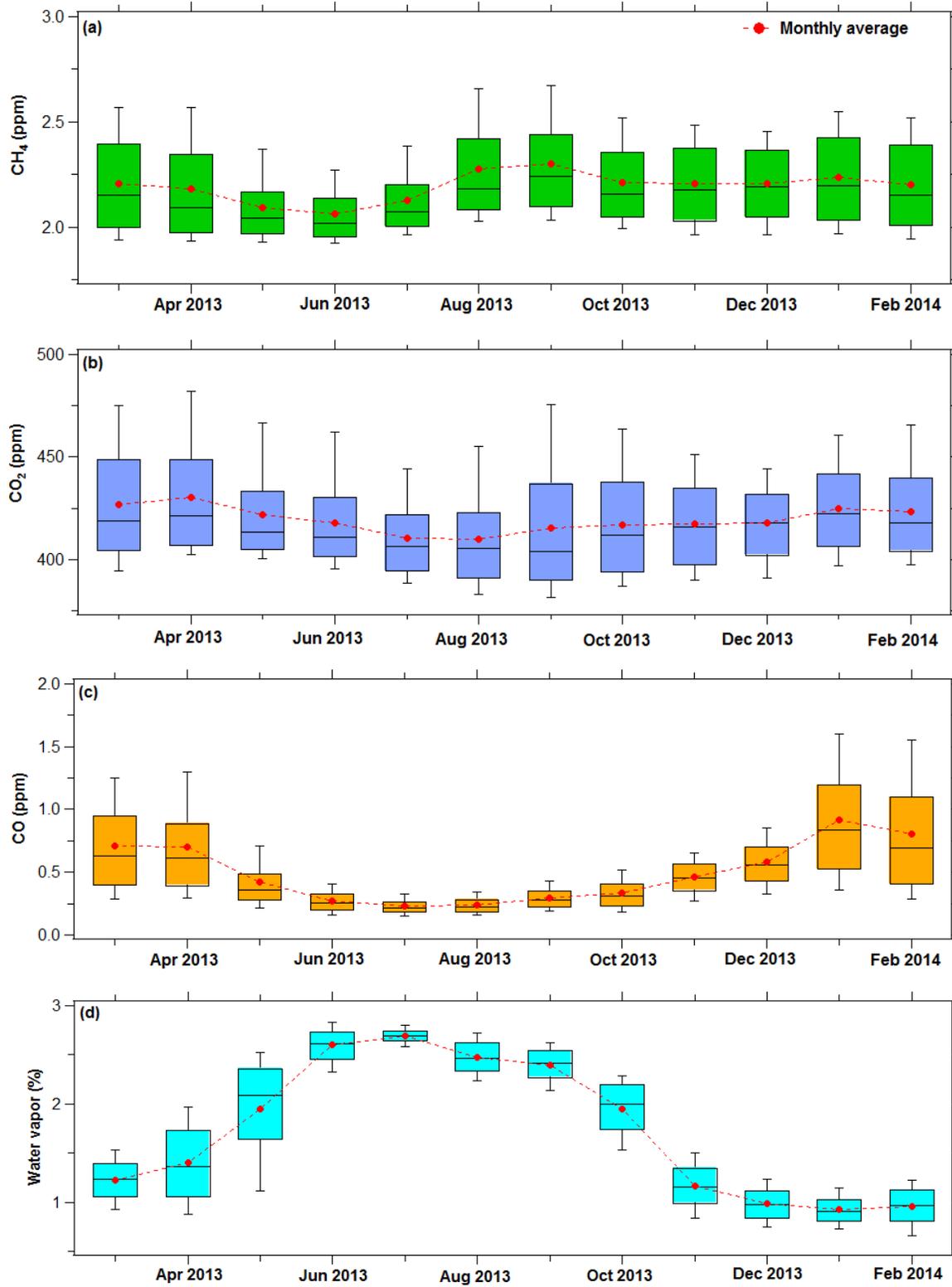
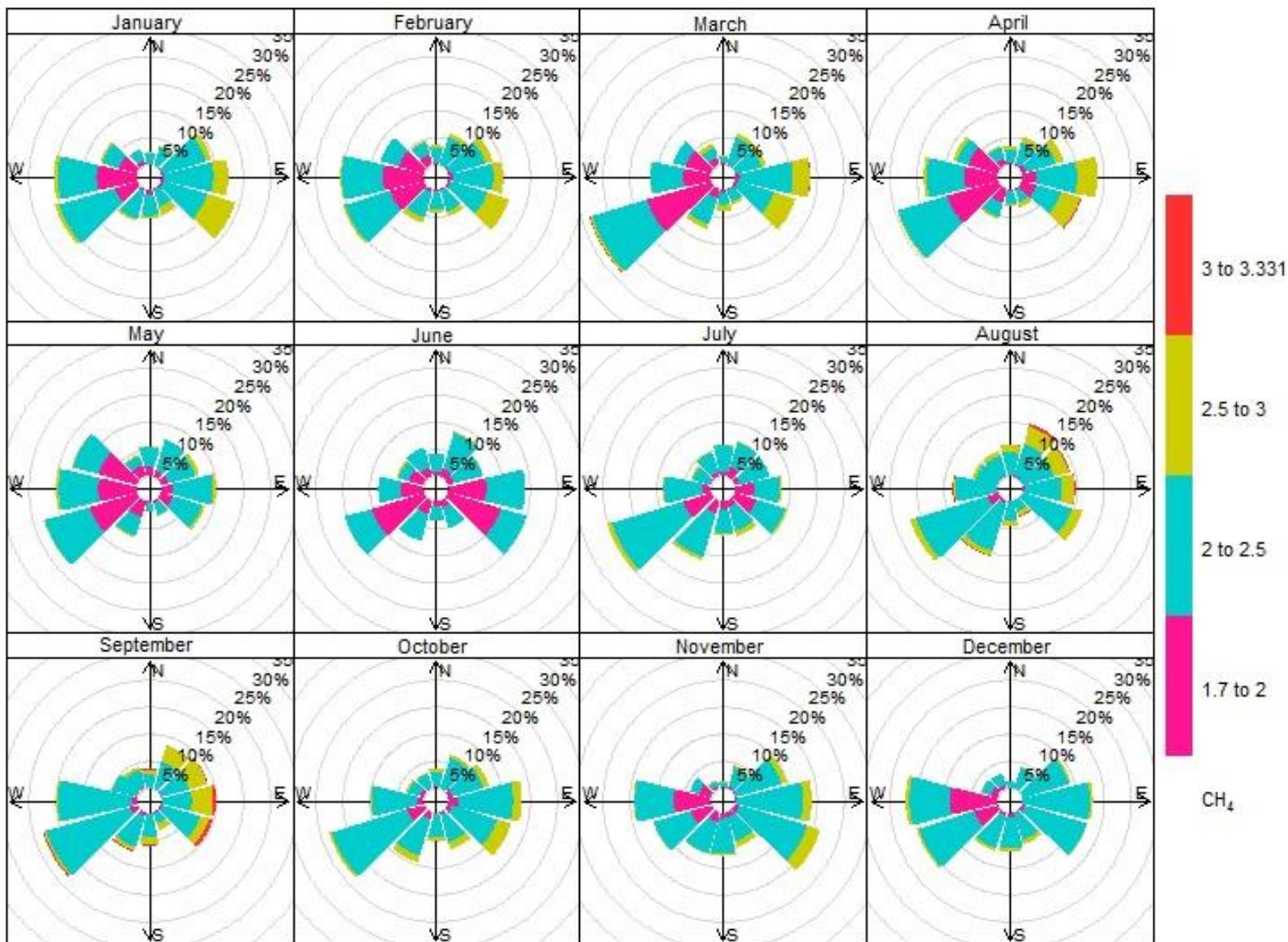
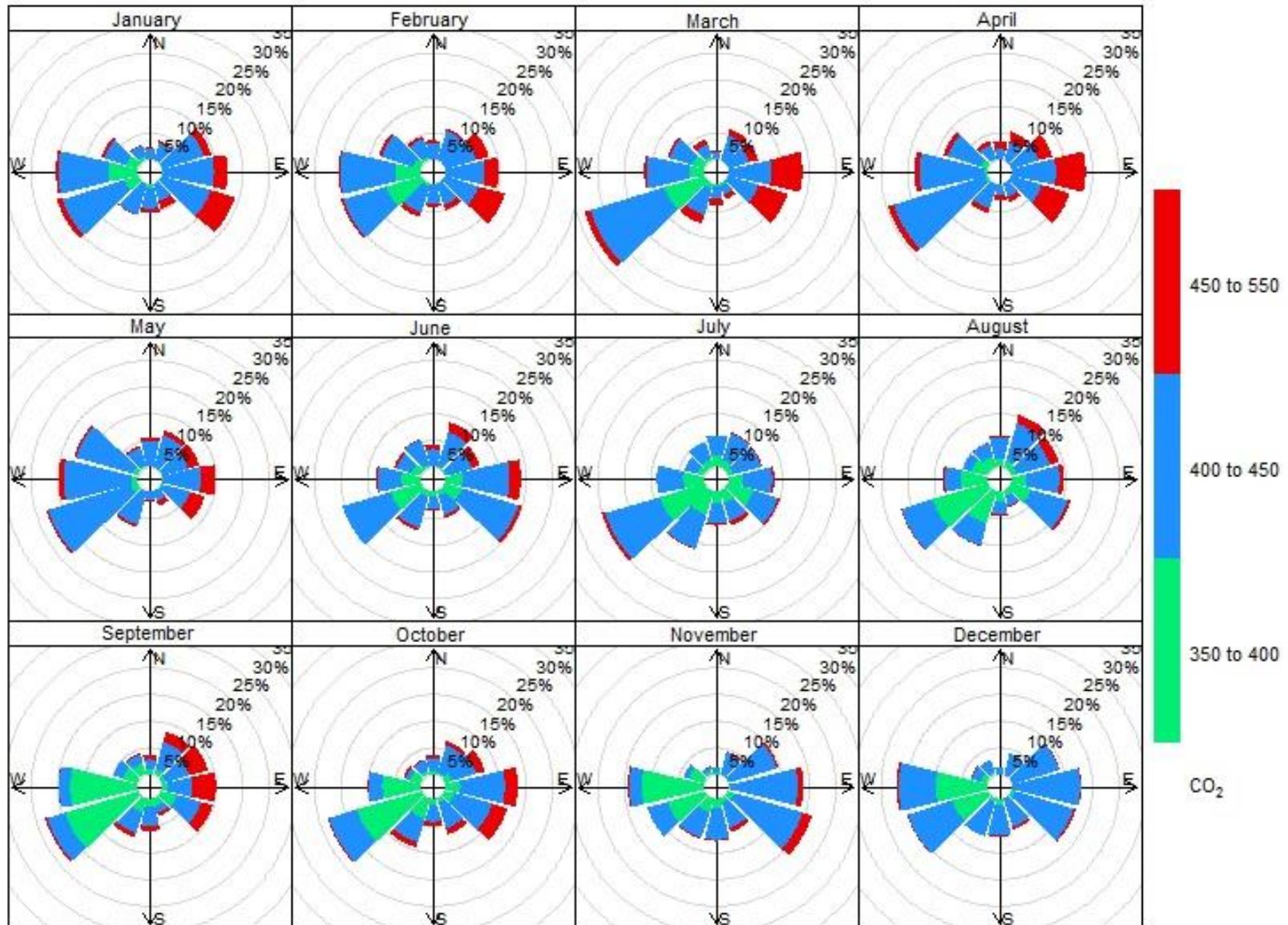


Figure 3. Monthly variations of the mixing ratios of hourly (a) CH₄, (b) CO₂, (c) CO, and (d) water vapor observed at a semi-urban site (Bode) in the Kathmandu Valley over a period of a year. The lower end and upper end of the whisker represents 10th and 90th percentile, respectively; the lower end and upper end of each box represents 25th and 75th percentile, respectively, and black horizontal line in the middle of each box is the median for each month while red dot represents mean for each month.

(a)



(b)



(c)

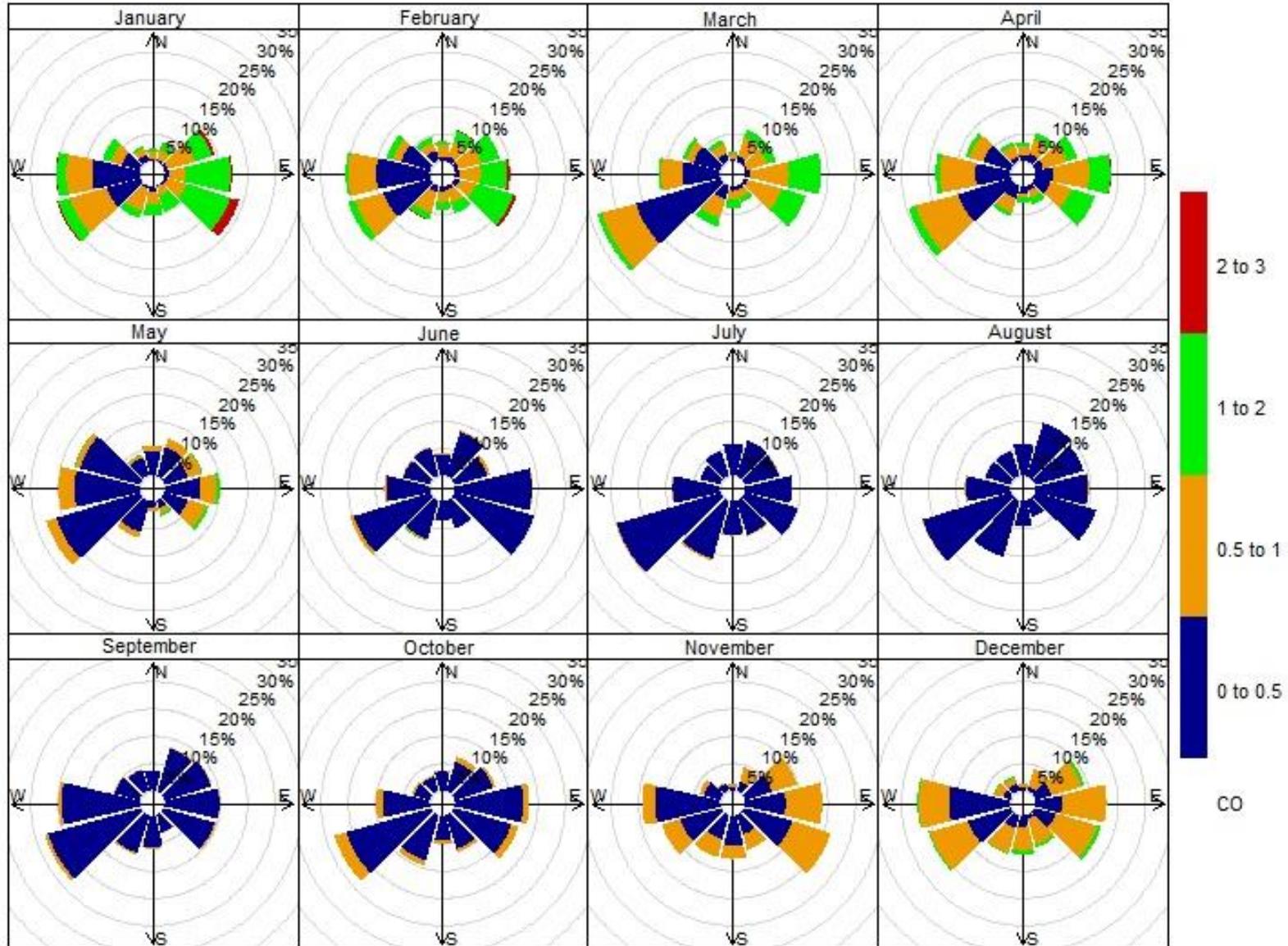


Figure 4. Relation between mixing ratios and wind direction observed at Bode in the Kathmandu Valley (a) CH₄, (b) CO₂ and (c) CO from March 2013 to February 2014. The figure shows variations of CH₄, CO₂ and CO mixing ratios based on frequency counts of wind direction (in %) as represented by circle. The color represents the different mixing ratios of the gaseous species. The units of CH₄, CO₂ and CO are in ppm.

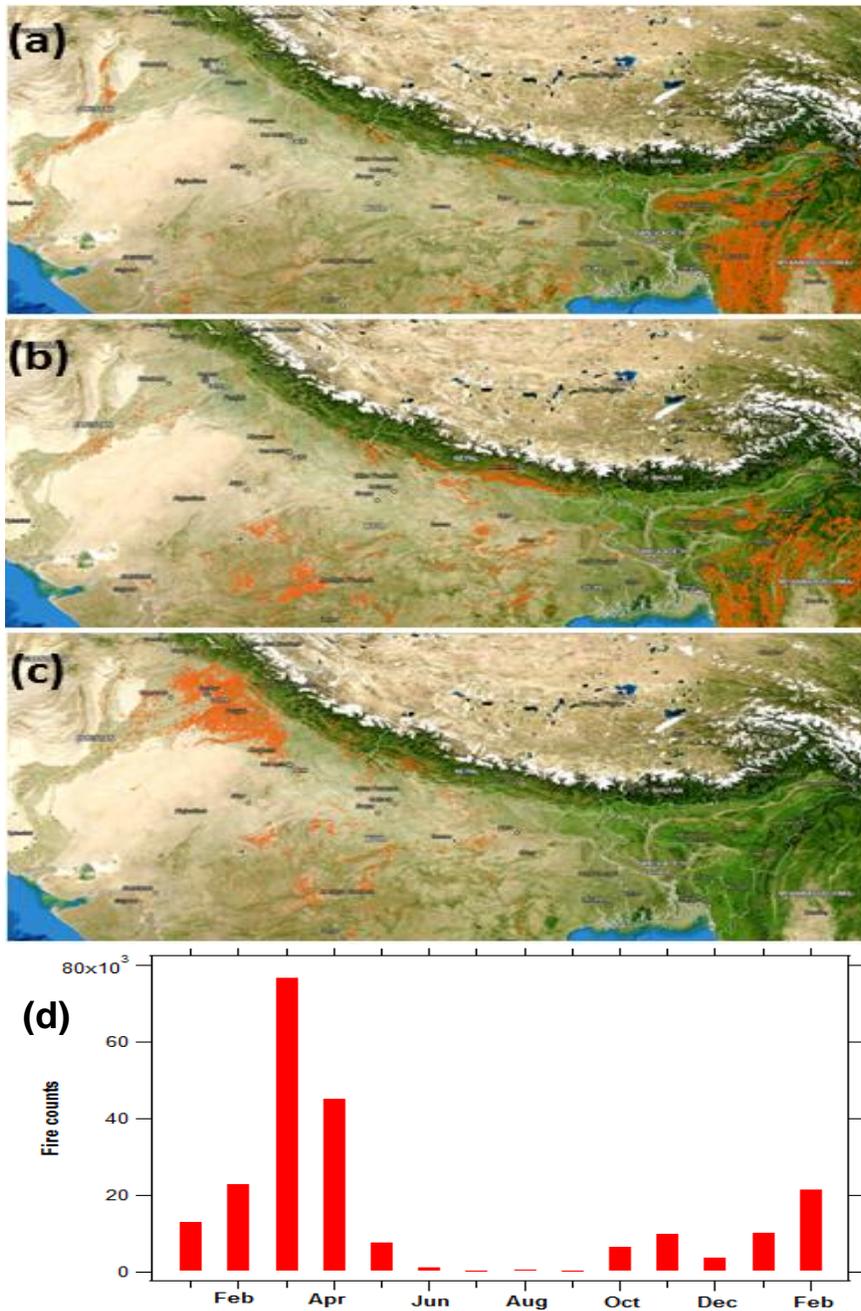


Figure 5. Satellite detected fire counts in (a) Mar, (b) Apr, (c) May 2013 in the broader region surrounding Nepal and (d) total number of fire counts detected by MODIS instrument onboard the Aqua satellite during Jan 2013-Feb 2014. Source: <https://firms.modaps.eosdis.nasa.gov/firemap/>

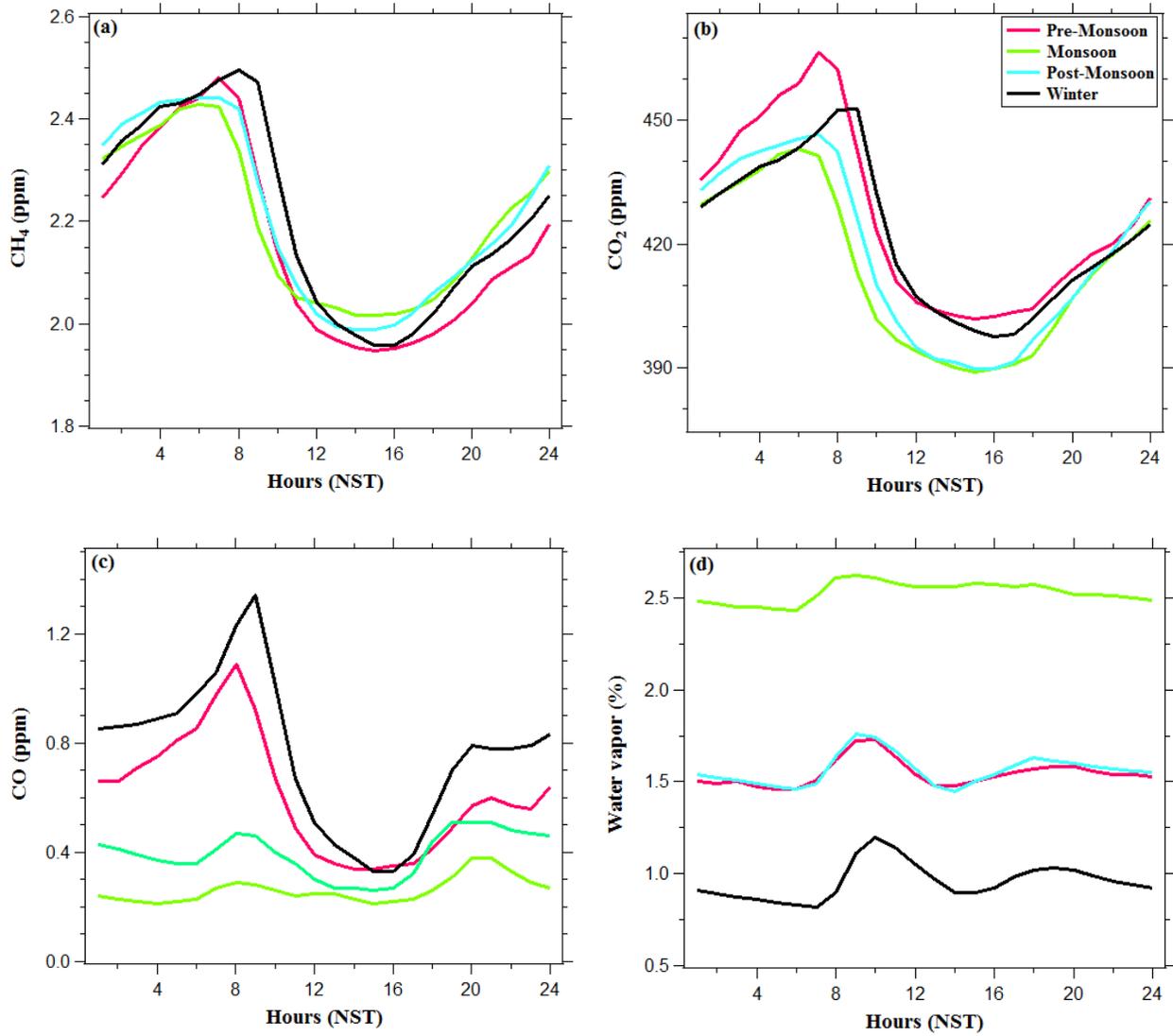


Figure 6. Diurnal variations of hourly mixing ratios in different seasons (a) CH₄, (b) CO₂, (c) CO, and (d) water vapor observed at Bode (semi-urban site) in the Kathmandu Valley during March 2013-February 2014. Seasons are defined as Pre-monsoon: Mar-May, Monsoon: Jun-Sep, Post-monsoon: Oct-Nov, Winter: Dec-Feb. The x axis is in Nepal Standard Time (NST).

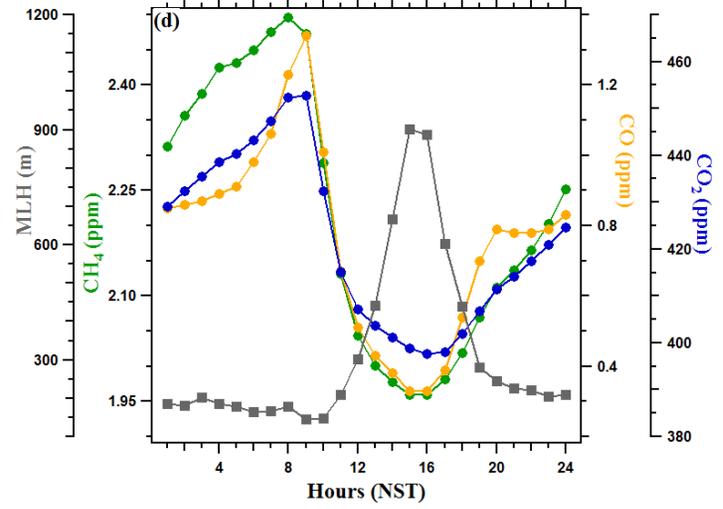
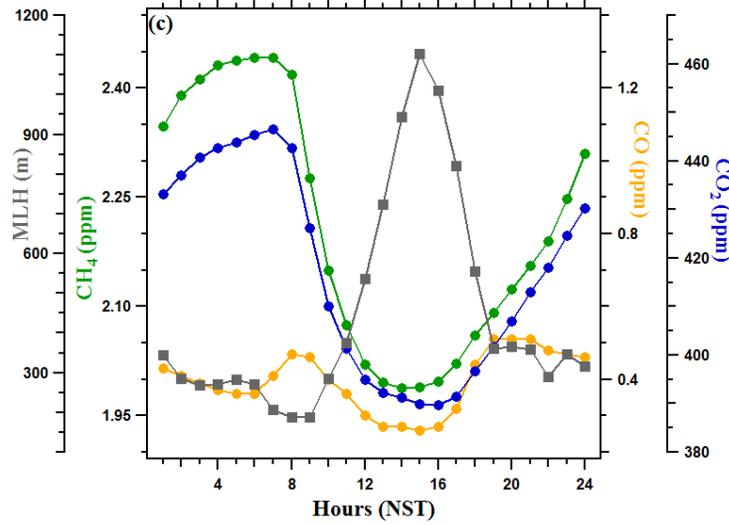
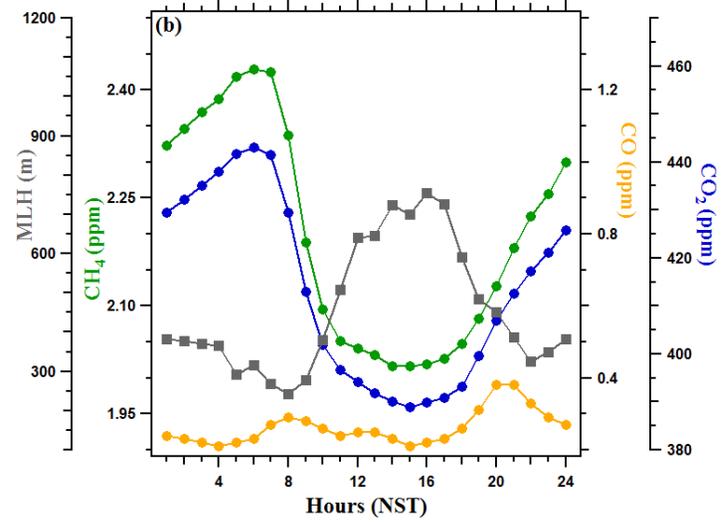
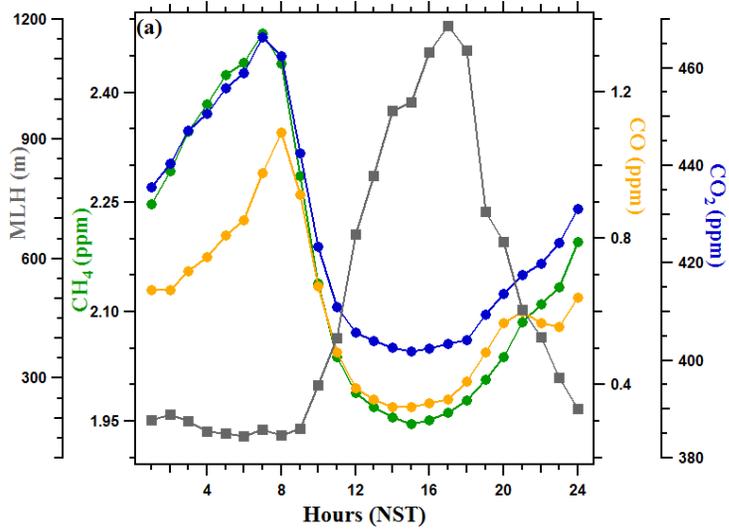


Figure 7. Diurnal variations of hourly mixing ratios of CH₄, CO₂, CO, and mixing layer height (MLH) at Bode (a semi-urban site in the Kathmandu Valley) in different seasons (a) pre-monsoon (Mar-May), (b) monsoon (Jun-Sep), (c) post-monsoon (Oct-Nov) and (d) winter (Dec-Feb) during March 2013- Feb 2014.

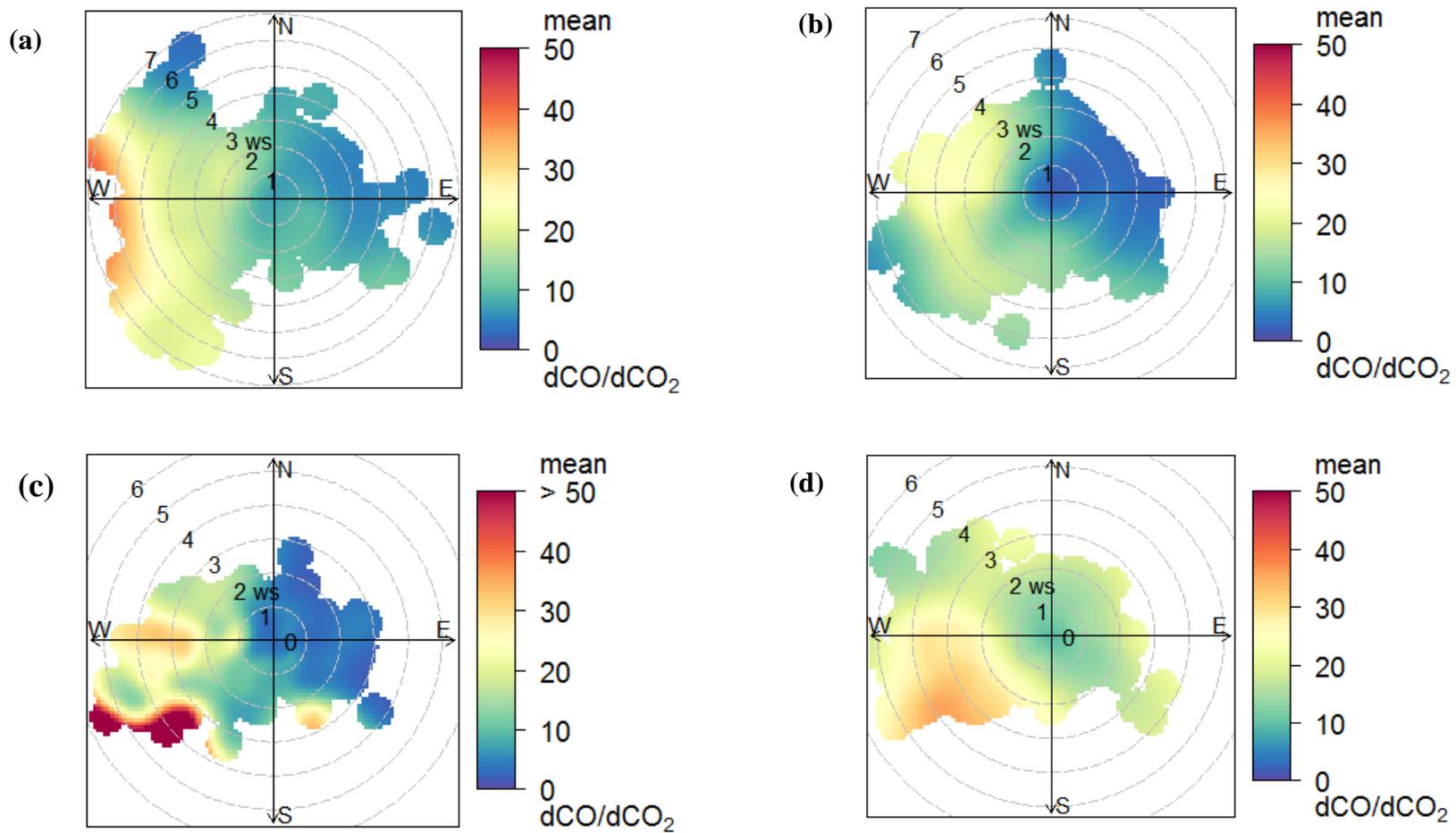


Figure 8. Seasonal polar plot of hourly dCO/dCO_2 ratio based upon wind direction and wind speed: (a) pre-monsoon, (b) monsoon, (c) post-monsoon and (d) winter seasons.

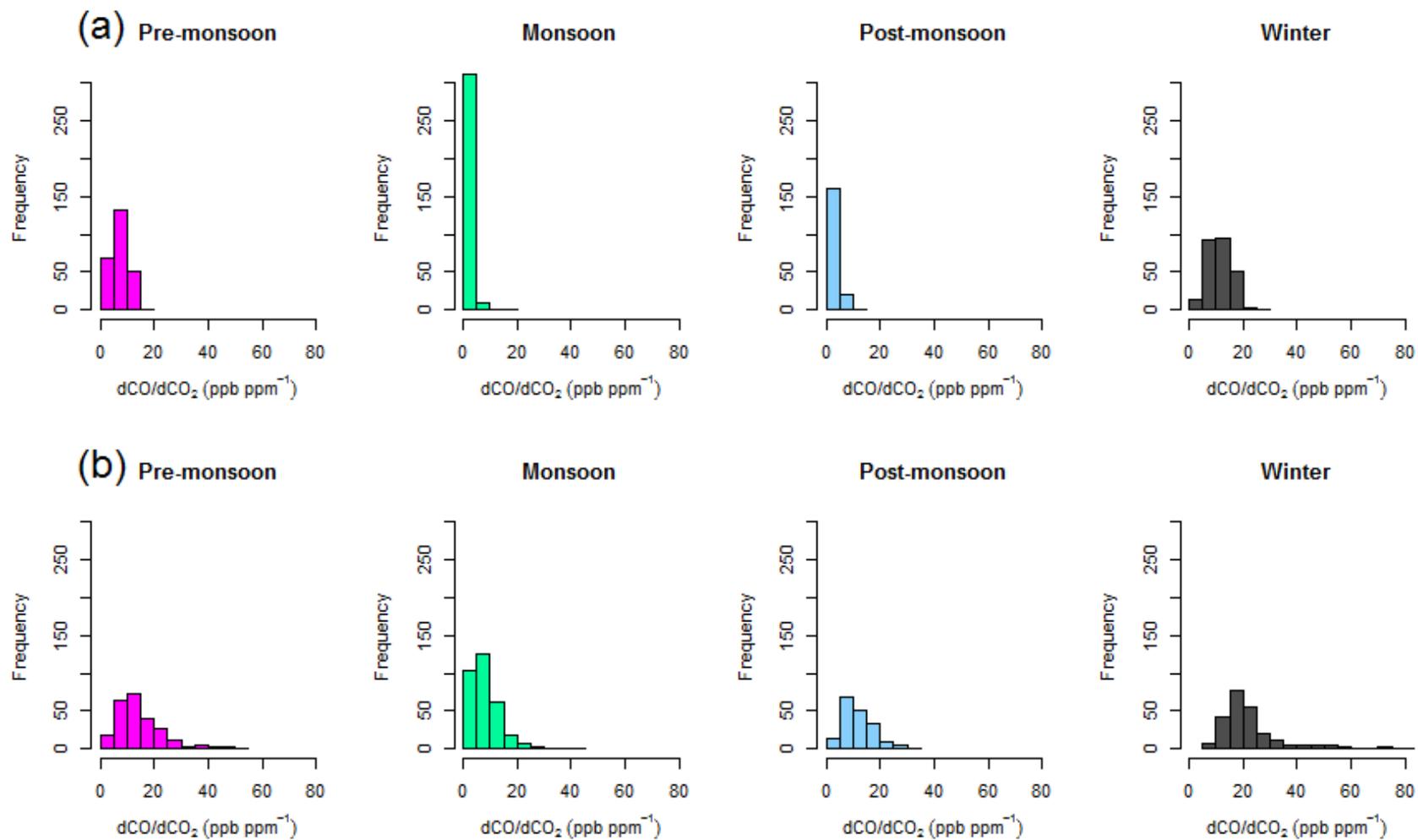


Figure 9. Seasonal frequency distribution of hourly dCO/dCO₂ ratio (a) morning hours (7:00-9:00) in all seasons except winter (8:00-10:00), (b) evening hours (19:00-21:00)

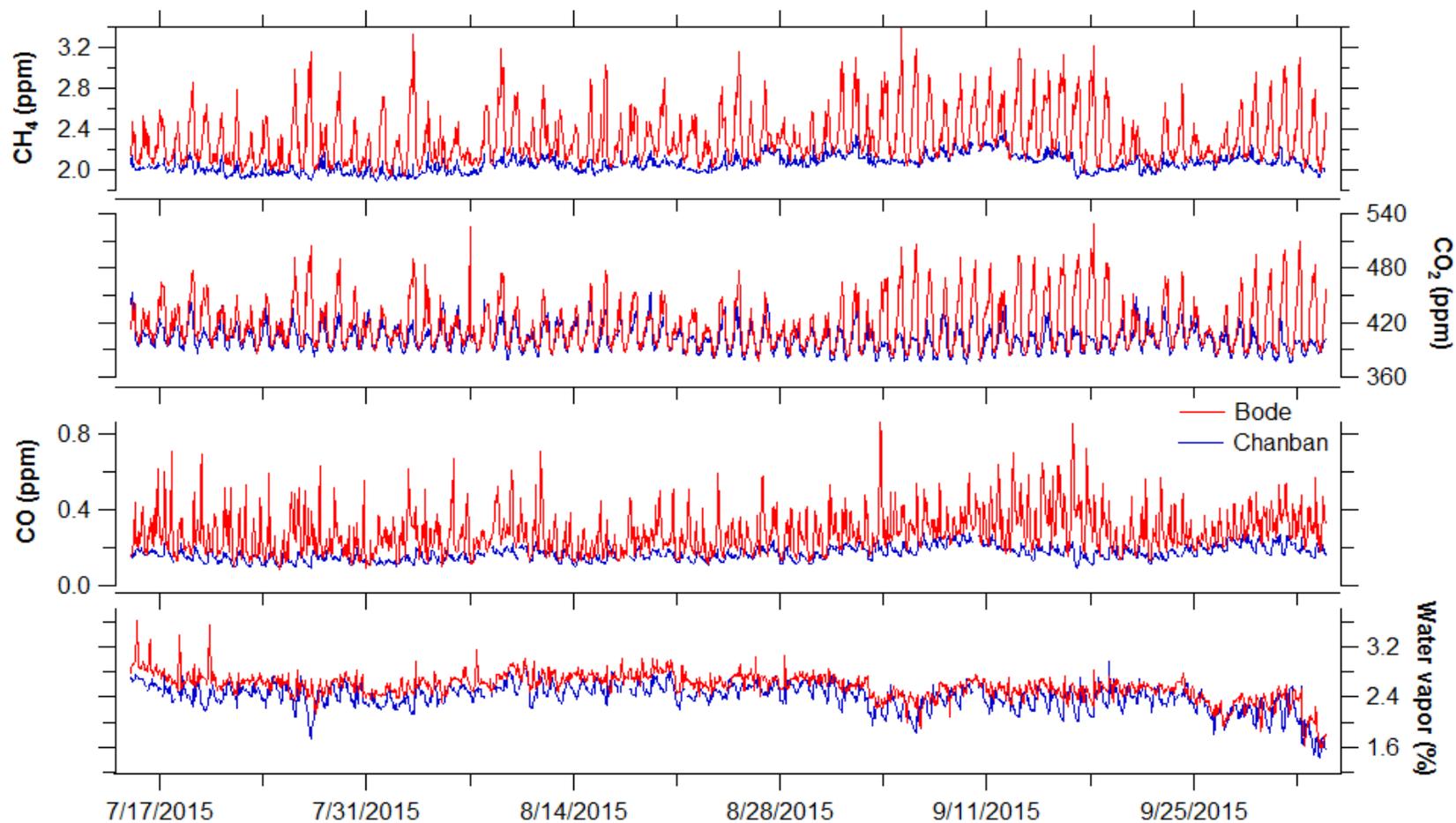


Figure 10. Comparison of hourly average mixing ratios of CH₄, CO₂, CO, and water vapor observed at Bode (a semi-urban site) in the Kathmandu Valley and at Chanban (a rural/background site) in Makawanpur district, ~ 20 km from Kathmandu, on other side of a tall ridge.

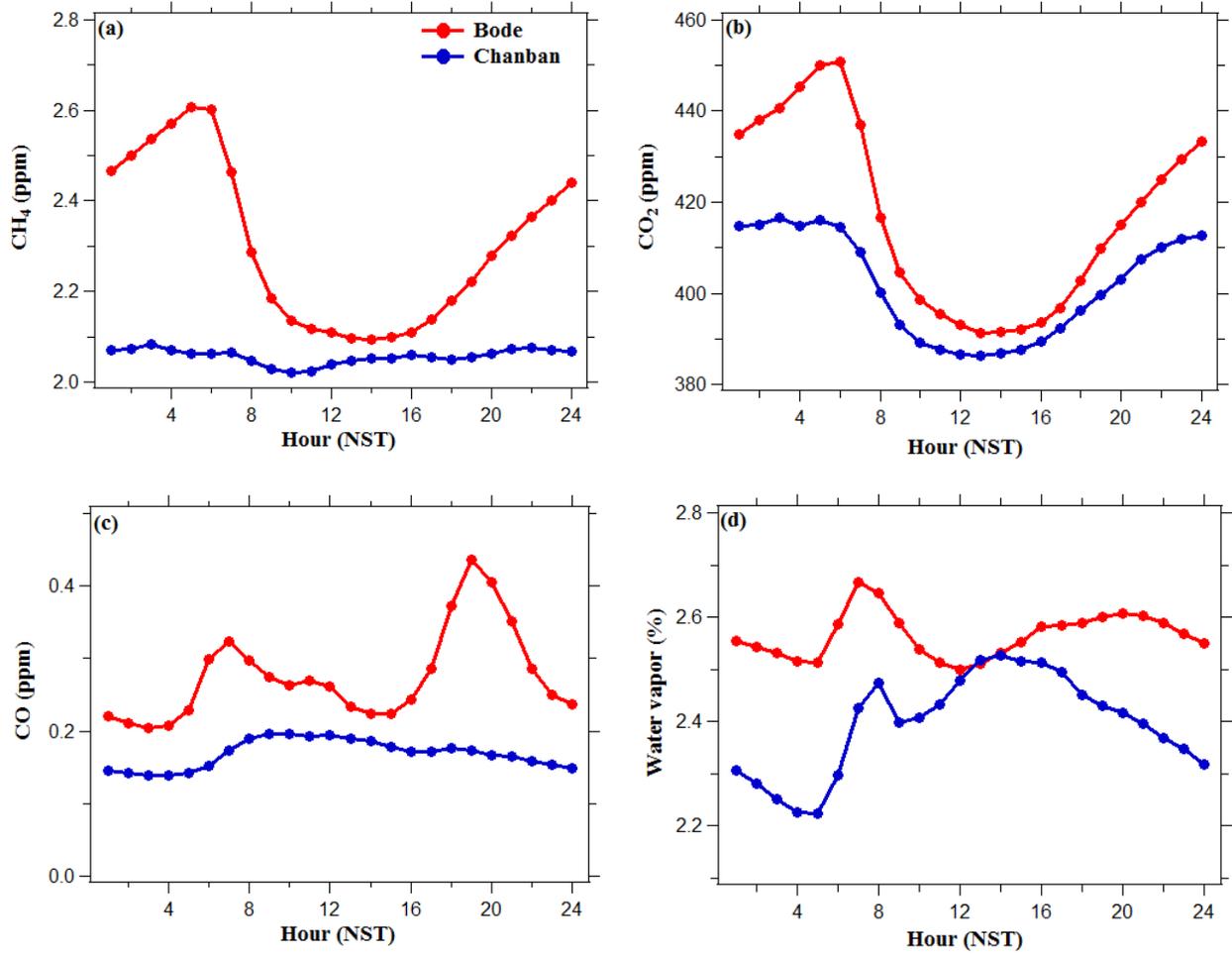


Figure 11. Diurnal variations of hourly average mixing ratios of (a) CH₄, (b) CO₂, (c) CO and (d) water vapor observed at Bode in the Kathmandu Valley and at Chanban in Makawanpur district during 15 July- 03 October 2015.