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First observations of light non-methane hydrocarbons (C_2–C_5) over a high altitude site in the central Himalayas

Tapaswini Sarangi [a,c], Manish Naja [a,*], S. Lal [b], S. Venkataramani [b], Piyush Bhardwaj [a], N. Ojha [a], R. Kumar [a,d], H.C. Chandola [c]

[a] Aryabhatta Research Institute of Observational Sciences, Nainital, India
[b] Physical Research Laboratory, Ahmedabad, India
[c] Department of Physics, D. S. B. Campus, Kumaun University, Nainital, India
[d] Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, USA

HIGHLIGHTS

- First observations of light NMHCs over the central Himalayas.
- Winter high and summer low in light NMHCs with somewhat different composition.
- NMHCs levels and seasonal cycle controlled mainly by OH oxidation at Nainital.
- Higher levels of some NMHCs over this region when compared with other global sites.

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ABSTRACT

This study presents observations of methane (CH_4) and light non-methane hydrocarbons (NMHCs) for the first time from a high altitude site Nainital (29.4°N, 79.5°E, 1958 m amsl) in the central Himalayas. The whole air samples collected with a frequency of 3 samples per week during April 2009–December 2011 are analyzed using a Gas Chromatograph equipped with Flame Ionization Detector (GC-FID). Additionally, samples were collected from two semi-urban sites (Haldwani and Pantnagar) in the adjoining Indo Gangetic plain region. CH_4 and NMHCs show a distinct seasonal cycle over this region with more frequent observations of higher levels during winter (DJF) and late autumn (SON) and lower levels during the summer–monsoon (JJA). Different NMHCs exhibit better correlations during autumn/winter as compared to the summer–monsoon season. The annual mean mixing ratios of methane, ethane, ethene, propane, propene, i-butane, n-butane, acetylene, and i-pentane at Nainital are measured to be 1.9 ± 0.1 ppmv, 1.8 ± 1.0, 0.7 ± 0.9, 0.6 ± 0.8, 0.6 ± 0.7, 0.6 ± 0.7, 0.5 ± 0.6, 1.0 ± 0.8, and 0.5 ± 0.6, respectively (all in ppbv). The seasonal cycle of CH_4 at Nainital is found to be similar to other global high altitude sites (Jungfraujoch and Mauna Loa) but somewhat different than a high altitude site Mt. Abu in India. NMHCs, other than ethane and propane, are found to be higher over this central Himalayan region than other sites. Additionally, composition of NMHCs is shown to be different over the study region when compared with other sites in the IGP region. A correlation study between ln((n-butane)/(ethane)) and ln((i-butane)/(ethane)) showed that oxidation by the OH radical is the main removal mechanism of these species over the central Himalaya and dilution maintains the ratios of these species. The lowest slope of propane and acetylene with CO during summer and spring are indicating absence of fresh air mass over this region. This study fills a major gap in observational data for light NMHCs in the Himalayas and has implications for better understanding of tropospheric chemistry over this region.

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1. Introduction

Methane (CH_4) and Non-methane hydrocarbons (NMHCs) are very important atmospheric trace gases in the context of air quality, atmospheric chemistry and global climate. CH_4 is the second

* Corresponding author. ARIES, Manora Peak, Nainital, Uttarakhand, 263129, India.
E-mail address: manish@aries.res.in (M. Naja).

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most important greenhouse gas after CO_2 (Forster et al., 2007) and along with NMHCs produces ozone (e.g. Crutzen, 1995; Kleinman et al., 2005), which is an air pollutant and a greenhouse gas. The photolysis of ozone produces hydroxyl radical (OH), known as the detergent of the atmosphere. The potential of NMHCs to produce ozone is much higher (10–13) compared to carbon monoxide (CO) and CH_4 (Chameides et al., 1988), however, ozone production depends non-linearly on NMHCs (Brasseur et al., 1999). The oxidation of hydrocarbons also contributes to the global atmospheric CO burden (Granier et al., 2000) and formation of secondary organic aerosols (SOA) (Kanakidou et al., 2005; Kroll and Seinfeld, 2008). The wide range of atmospheric lifetimes (few hours to several tens of days) possessed by NMHCs make them useful tracers of atmospheric transport and indicators of photochemical aging (e.g. Parrish et al., 1992; McKeen and Liu, 1993; Jobson et al., 1998; Kleinman et al., 2003; Arnold et al., 2007).

The hydrocarbons are emitted from both natural and anthropogenic sources. The anthropogenic sources of CH_4 include vehicular exhaust, leaks of commercial natural gas, emissions from industries and rice paddies, while the natural sources include wetlands, oceans, forest vegetation and fires, termites (Singh and Zimmerman, 1992; EPA, 2010). The major sink of CH_4 is the reaction with OH with relatively small contributions from destruction in the stratosphere and soil (Born et al., 1990). Generally, methane emissions from paddy account for about 6% of the total emissions. However, emissions estimates from paddies have large range and vary between 39 and 112 Tg CH_4 yr^{-1} (Denman et al., 2007). In warm weather conditions, and water-logged soil, rice paddies acting as wetlands emit significant amount of methane. The methane emissions in India have been reported to have grown by 9% during 1985–2008 (Garg et al., 2011) and its concentration by 30 ppbv yr^{-1} (Lal et al., 2004).

The major sources of NMHCs include natural gas, petrochemical industries, automobile exhaust, gasoline, industrial processes, burning of agricultural waste and other plant materials, and brush fires. The measurements of NMHCs have been mostly conducted in North America, Europe and East Asia, (e.g. Goldstein et al., 1995; Jobson et al., 1994; Swanson et al., 2003; Yates et al., 2010; Zhang et al., 2014), while these are severely limited in South Asia, where biofuel and biomass burning play a disproportionately large role in the emissions of trace gases including NMHCs (Lawrence and Lelieveld, 2010). The measurements of NMHCs have been reported only from very few sites in South Asia and these include Ahmedabad and Mt Abu in western India (Sahu and Lal, 2005, 2006), Mohali (Sinha et al., 2014), Bombay (Rao et al., 1997), and Kathmandu and Nagarkot (Sharma et al., 2000a). In addition, NMHCs measurements have been reported from the Bay of Bengal (Mallik et al., 2013) and 1–2 months campaign mode observations in the Indo-Genetic Plain (IGP) region (Lal et al., 2012). These measurements have provided important information about the absolute levels and variability of NHMCs over this region. However, the measurements of NMHCs with a complete seasonal cycle are nearly non-existent in northern part of India, where different satellite retrievals and model results show elevated pollution levels (e.g. Fishman et al., 2003; Jethva et al., 2005; Kumar et al., 2013).

In light of the above conditions, measurements of CH_4 and NMHCs were initiated for the first time at a high altitude site Nainital located in the central Himalayas. The observed variations of trace gases at Nainital, due to its unique location and minimal local anthropogenic emissions, are suggested to be a manifestation of the activities in the surrounding regions and can provide invaluable information about the inflow of pollutants into the Himalayas. In addition, we also conducted NMHCs measurements at two nearby low altitude semi-urban sites (20–40 km from Nainital) Haldwani and Pantnagar located in the IGP region. This manuscript is organized as follows. We begin with a brief description of the observation site and an overview of data analysis and methodology. The seasonal variations of CH_4 and NMHCs at Nainital, Pantnagar and Haldwani are discussed next and compared with selected available observations around the world. We then use inter-species correlations to examine dominant sources of NMHCs. Results from this study are summarized at the end.

2. Experimental details

2.1. Observations sites and general meteorology

The whole air samples were collected at a remote mountain top called Manora Peak (29.37°N, 79.45°E, 1958 m amsl) at Nainital, located in the central Himalayas (Fig. 1). A detailed description of this site can be seen in Kumar et al. (2010), Sarangi et al. (2014), and Naja et al. (2014), where it is shown that this site offers relatively cleaner atmosphere during most of the time and observations at this site can be considered as representative of a larger region of the northern part of South Asia. A few air samples were collected from two nearby semi-urban sites called Haldwani (29.2°N, 79.5°E, 450 m amsl) and Pantnagar (29.0°N, 79.5°E, 231 m amsl) (30–50 km from Nainital). Haldwani and Pantnagar are located due South of Nainital and have small scale industries. The nearest megacity Delhi is about 225 km due West of these sites.

The general meteorology prevailing at Nainital has been discussed in detail in previous studies (Kumar et al., 2010; Sarangi et al., 2014; Naja et al., 2014) and here we provide only a brief overview. Solar radiation shows a systematic increase from winter to spring and it becomes most intense in April. It shows lowest values during summer–monsoon period (July–August). Average temperature shows more or less similar variations as in the solar radiation; however minimum values of temperature are observed in winter season. Highest values of relative humidity and simultaneous lowest values of solar radiation during summer–monsoon mark the arrival of monsoon at Nainital and are associated with substantial amount of rainfall. The wind patterns are westerly/northwesterly in winter and gradually shifts to southwesterly during June–July, while the air masses circulate over the Northern Indian region in May and September.

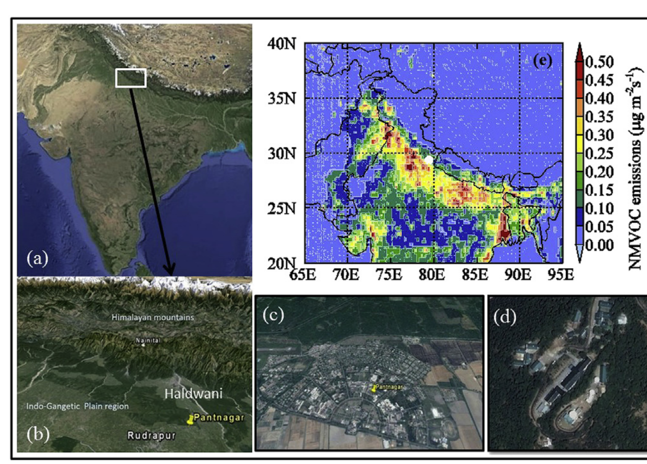

Fig. 1. Images (google) showing part of (a) India and study regions with location of three sites (b) Nainital, Haldwani and Pantnagar (c) Pantnagar University campus and (d) ARIES and (e) map showing the spatial distribution of total non-methane volatile organic compounds (NMVOC) anthropogenic emissions obtained from EDGAR.

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