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Trace gases behaviour in sensitive areas of the northwestern Himalaya–A case study of Kullu-Manali tourist complex, India

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Surface concentration of the three important trace gases, ozone (O_3), nitrogen dioxide (NO_2) and sulphur dioxide (SO_2) were measured at three different tourist locations, namely Kullu, Manali and Kothi in the northwestern Himalayan region, which are located at 1220 m, 2050 m and 2530 m above the mean sea level mainly to asses the anthropogenic impact. The surface O_3 was monitored for four years during the period 1998 - 2002 and 2004 at the time of peak tourist season (May-June), and SO_2 and NO_2 were measured during the entire period in 2003. The peak O_3 concentrations reached close to 50 ppb level, while the annual mean concentrations of O_3, SO_2 and NO_2 remained within the United States Environmental Protection Agency's (USEPA's) National Ambient Air Quality Standards (NAAQS). The peak hourly average values of O_3 was 44 ppb at Manali and 32 ppb at Kothi during evening (1700 hrs IST), while that at Mohal (near Kullu) was 32 ppb in the afternoon (1500 hrs IST) period. The seasonally average value of maximum concentration of NO_2 was 3.8±0.6 μg m^{-3} at Kothi and 7.6±1.0 μg m^{-3} at Mohal in autumn (October-November), while that of SO_2 was 21.4±1.8 μg m^{-3} at Kothi and 18.8±1.3 μg m^{-3} at Mohal during the monsoon (July-September) and summer (April-June) periods, respectively. Vehicular emissions and biomass burning for heating and cooking during the winter period (especially when power failure is common) as well as during forest fires could be the major contributors for increased emissions of these trace gases. However, the influence of long-range transport may also be important.

Keywords: Surface ozone, Nitrogen dioxide, Sulphur dioxide, Kullu-Manali, Northwestern Himalaya

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1 Introduction

Recent studies have indicated that the concentrations of ozone in the troposphere is increasing by about 1-2% per year[1-2] and may triple within the next 30-40 years, especially, in industrialized locations[3]. When the level of O_3 in the atmosphere is more than 70 parts per billion (ppb) for a period of 20-90 min, coughing, choking and severe fatigue in human health are noticed[4]. Ozone is a phytotoxic air pollutant that can lead to serious injury to plant tissues and reductions in their growth and productivity[5]. It has been shown that exposure to ozone concentrations of more than 50 ppb is sufficient to cause damage to certain species of vegetation[6]. Ozone exposure to plants during the day brings about also bio-chemical changes in them resulting in decreased photosynthesis[7]. Ozone and other photochemical oxidants are formed in air by the photolysis of nitrogen oxides and reactive organic compounds[8-10] and is a source of the hydroxyl radical (OH) which reacts rapidly with most of the air pollutants and trace species found in the atmosphere.

It has been argued that ozone transportation from the free troposphere exerts a strong influence on concentrations especially at elevated sites[11]. Such atmospheric changes can produce transient peak ozone concentrations around 100 ppb at sea level and concentrations more than 250 ppb in mountain region[12]. An increase in tropospheric O_3 through this process has also been observed globally over the past century[13]. The increase in the concentration of surface O_3 at high altitude sites can thus mainly be due to influence of photochemistry from anthropogenic pollutants, and transportation of pollutants from nearby sources[14].

The nitrogen dioxide (NO_2) is relatively an inert gas and moderately toxic. The major health impacts of NO_2 are increased incidence of lower respiratory tract infection in children and decreased airway responsiveness in asthmatics[15]. This irritates the alveoli of the lungs[5]. Children, the elderly, asthmatics and individuals with chronic obstructive pulmonary disease are more responsive to NO_2 than others in the community[16]. Sulphur dioxide (SO_2) is often a local pollutant, but can also be subject to long-range transport process when oxidized in sulphate form. The SO_2 tends to irritate the mucous membrane of the respiratory tract and expedites the development of chronic respiratory disease, particularly, bronchitis and pulmonary emphysema[5].

According to the current estimates, the emission strength of NO_x and SO_2 in Asia are in the same range as in North America and Europe[17]. Asia is characterized by its rapid growth of emissions in recent decades due to increase in population as well as increase in per capita emission rates. Whereas the emission rates of NO_x and SO_2 in North America and Europe have levelled off, at least in the last decade, judging from fuel consumption and the effort of emission controls[18]. Preliminary estimates of SO_2 emissions in India are 0.48 tera grams (Tg) yr^{-1} during 1997 (Ref. 19). Himachal State, where the present study region locates, contributes to 1.1% (0.005 Tg yr^{-1}) of the total SO_2 emissions in the country of which more than 80% is through burning of fossil fuels[20].

The important world mountains, particularly, the Indian Himalaya have so far been lacking such studies like surface ozone, nitrogen oxides and sulphur dioxides. Thus, the present attempt could be helpful much more in unfolding its current status which would work as the background values for further research, particularly, when the air quality might have been comparable to other deteriorated hill towns. It is important to measure pollution in an area where anthropogenic pressure exerted by tourists is high and tourism potential to harness in other new areas is abundant. The objectives of the present study are (i) to understand diurnal variation of the surface ozone during peak season of tourists, and (ii) to determine the seasonal variations in the concentration of other trace gases such as NO_2 and SO_2.

2 Experimental sites

Kullu and Manali tourist spots in the Kullu valley comprise the headwater Beas basin of the hill State of Himachal Pradesh in the north-western Himalaya (Fig. 1). The Kullu valley which is 80 km long[21] with a minimum width of 2 km begins from Larji (957 m in the lower Beas basin) and stretches up to Rohtang crest (4038 m in the upper Beas basin).

Kullu town is located at 31°38' N and 77°60' E with a geographical area of 7 km^2 inhabited by about 18,000 permanent residents[22]. Kullu and Manali are the major tourist spots in this valley. Mohal is 5 km south to Kullu town and is primarily famous for agro-farming (orchards) and characterized with semi-urban settlement (by converting arable land into house constructions for dwelling). Manali tourist resort, located at 32°24'30" N and 77°10'6" E having geographical area of about 3.5 sq. km, is inhabited by about 6,000 permanent residents[22] and has a temperate climate. Approximately, 11 lakh visitors visit Manali and about 2 lakh (22.53%) Kullu annually[23]. The surrounding land use at Manali is characteristic for a large scale hotel constructions, business establishments, apple orchard and sparse coniferous tree species. Kothi, which is 12 km away from Manali, is the last habitable settlement in the upper Kullu valley.

3 Measurements (materials and methods)

The UV Photometric O_3 Analyser (Thermo Environmental Instruments, Inc., USA) was used to monitor surface ozone. Ozone concentration was determined by measuring the attenuation of light at 254 wavelength with an accuracy of 1 ppb. The O_3 concentration was measured during the summer months of May and June for 5-7 days in a month at each of these locations.

The concentration of NO_2 and SO_2 were measured with the help of gaseous impingers attached with High Volume Sampler (APM 430) every alternate days at Mohal and Kothi in different months during the year 2003. From this, 180 samples were available for SO_2 and NO_2 at Mohal (14-16 samples per month) and 153 samples for Kothi (8-16 samples per month). Standard colorimetric methods were used[24-26] for estimating the concentrations of NO_2 (Ref. 25) and SO_2 (Ref. 26).

4 Results and discussion

4.1 Surface ozone (O_3)

The diurnal mean value of surface ozone at Mohal is 20.2 ppb, while that at Manali and Kothi are 28.9 ppb and 27.8 ppb, respectively. The United States Environmental Protection Agency's (USEPA's) National Ambient Air Quality Standards (NAAQS) has fixed a permissible limit for surface ozone

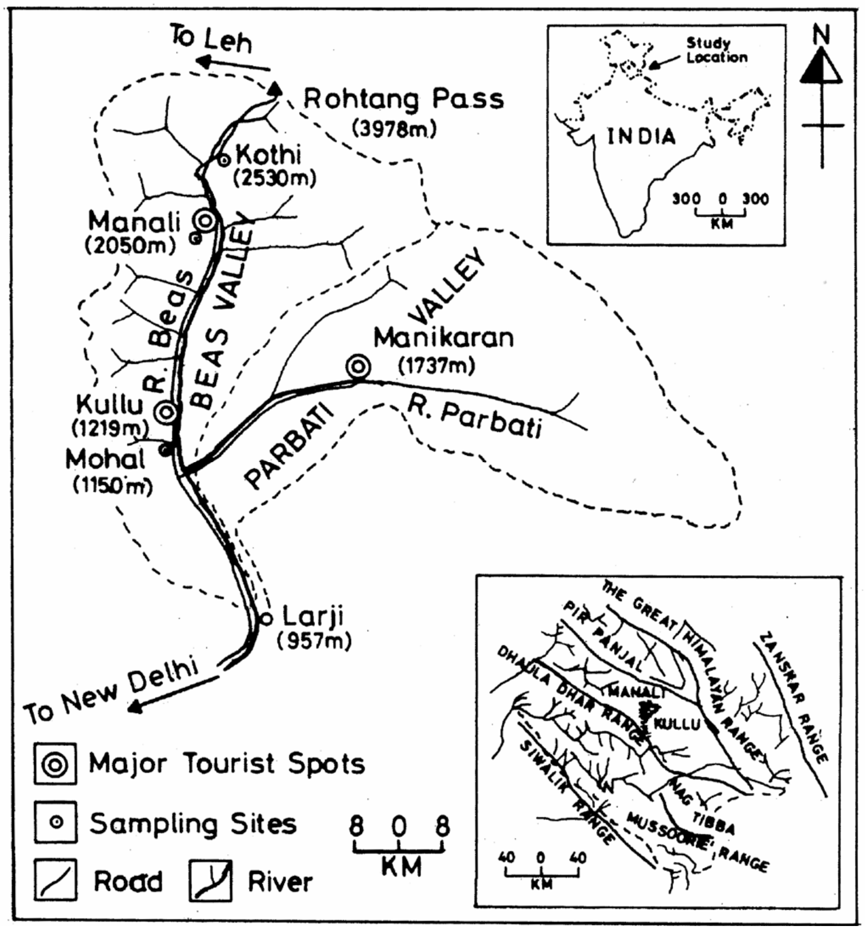

Fig. 1—Geographical location of sampling sites, Kullu-Manali Tourist Complex and their adjoining areas

concentrations with 80 ppb for 8 hours average and 120 ppb for 1 hour's exposure[27] and the above measured mean values are within the allowable range. The mean diurnal variations of O_3 at these three locations are presented in Figs 2-4. At Mohal (lowest altitude), the O_3 concentration varies from 9.4 ppb at 0600 hrs IST to 32 ppb at 1500 hrs IST with an amplitude of 22.6 ppb.

These values are lower than the USEPA standard (80 ppb for 8 hours' average, and 120 ppb for 1 hour)[27], and primary air quality standard from WHO[28] (56.5 ppb). At Manali, the mean O_3 concentration (Fig.3) varies from 15.5 ppb at 0400 hrs IST to 44 ppb at 1700 hrs IST with a mean amplitude of 28.5 ppb (marginally more than that at Mohal). The surface O_3 concentration, in general, is large at Manali. At Kothi, the O_3 concentration (Fig. 4) varies from 24.1 ppb at 0400 hrs IST to 32.1 ppb at 1700 hrs IST with diurnal amplitude of 8 ppb. The diurnal amplitude is lowest at Kothi, a station located at the highest altitude (Fig. 4).

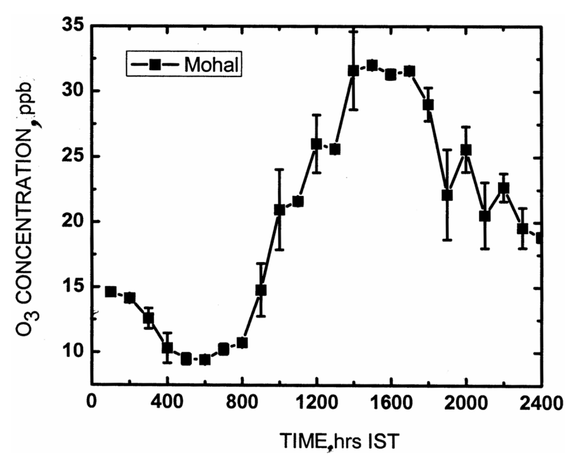

Fig. 2—Surface ozone (O_3) concentrations at Mohal (1150 m asl) in the northwestern Himalaya

A large number of plying vehicles passing through Mohal (2,900 day^{-1} from 6 a.m. to 6 p.m.) and Kothi (1,300 day^{-1} from 6 a.m. to 6 p.m.) during peak tourist season (June 2005-2006) and biomass burning due to

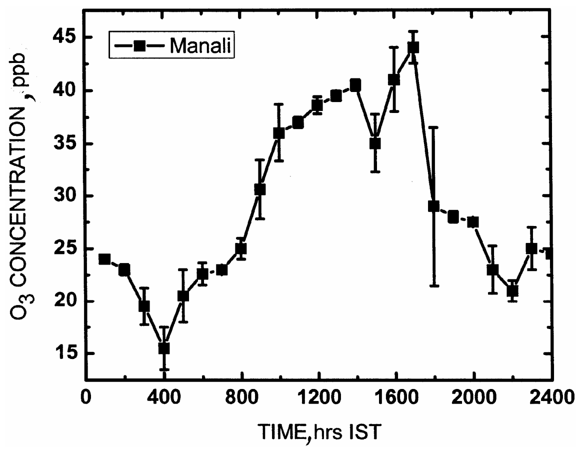

Fig. 3—Surface ozone (O_3) concentrations at Manali (2050 m asl) in the northwestern Himalaya

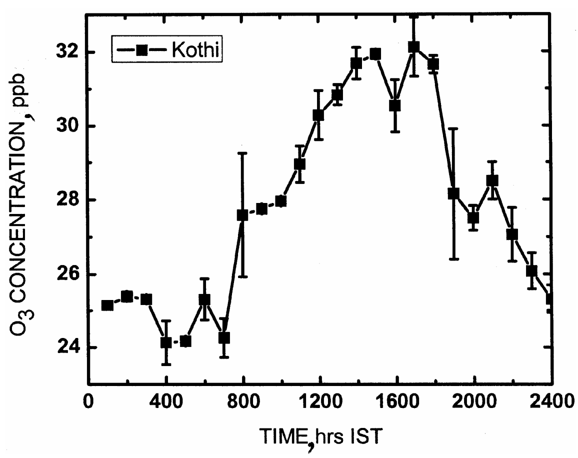

Fig. 4—Surface ozone (O_3) concentrations at Kothi (2530 m asl) in the northwestern Himalaya

forest fires, cooking and heating during power failures in the hill spots and their surroundings (remote rural villages) are the important favouring factors of NO_2 formation. The NO_2 works as an important precursor in photochemistry of surface ozone in presence of sunlight. According to our recent study, the contribution of diesel operated vehicles at Mohal ranged from 37% (February-March 2006) to 52% (September 2005); and at Kothi from 48% (February 2005) to 74% (October 2005) of the total plying vehicles a day during 2005-2006. Biomass burning included accidental forest fires as well as man made ones, fuel burning for agriculture[29], cooking and heating purposes.

The surface ozone concentrations at Manali, the mid-altitudinal site compared to two others, showed a wide range of variations of 28.5 ppb from minimum to maximum within 24 h. Broadly, the O_3 concentration is low between 0100 hrs IST and 0800 hrs IST and high between 1000 hrs IST and 1900 hrs IST. The low concentration is observed when photochemical process is less active [in absence of sunlight and low abundance of NO_2 and other volatile organic compounds (VOC)]. At the same time, most of the NO_x emissions from vehicles occur in the form of NO, which actually consume ozone by the reaction $NO+O_3 > NO_2+O_2$. As a consequence, ozone levels are often lower than normal, close to such sources. It is only after several hours that the photochemical processes involving NO_x and hydrocarbons start to produce substantially elevated ozone levels. But the highest diurnal O_3 concentration remains between 1000 hrs IST and 1700 hrs IST when conditions remain much more conducive to photochemistry process due to increase in NO_2 concentrations from automobile exhausts, abundant sunlight and low humidity. Thus, anthropogenic source of tropospheric ozone is chiefly due to the photolysis of NO_2 where ambient ozone can largely be increased due to increasing emissions of its precursors, NO_x, VOC and carbon monoxides (CO). A similar study conducted at Gibbs (2006 m) in Mitchell State Park of North Carolina in the Southern Appalachians also showed that high-ozone episodes occurred during the passage of synoptic high pressure, which results due to low wind speed (< 5 m s^{-1}), warm temperature (daily maximum of >25°C) with low relative humidity (< 60%) which are conducive for the photochemical process[6].

4.2 Nitrogen dioxide (NO_2)

The natural sources of NO_2 are bacterial action in soil, volcanic action and lightning, while the anthropogenic sources include combustion of oil, coal, gas in both automobiles and industry and forest fires[30]. The mean NO_2 levels at the low altitude (Mohal) site is relatively large compared to the other site (Kothi) (Table 1). However, these values are within national (15 μg m^{-3} annual average for sensitive areas)[31] and USEPA's NAAQS (100 μg m^{-3} or 53 ppb annual arithmetic mean) standards[27].

On an average, at Mohal, NO_2 concentration ranged from 2.7±0.9 to 7.6±1.0 μg m^{-3} in winter and autumn seasons, respectively (Fig. 5). Similarly, maximum and minimum values of NO_2 concentration at Kothi ranges between 1.0±0.1 μg m^{-3} (monsoon) and 3.8±0.6 μg m^{-3} (autumn). The major sources contributing to NO_2 values, under the present study region, are mainly automobile exhausts, biomass burning and forest fires.

4.3 Sulphur dioxide (SO_2)

At both these locations (Mohal and Kothi) highest concentration of SO_2 are observed in June and July, respectively. On seasonally averaging, the SO_2 concentration at Mohal varies from 10.1±1.0 μg m^{-3} in autumn to 18.8±1.3 μg m^{-3} in summer, while at Kothi these values were, respectively, 7.3±0.2 μg m^{-3} in autumn and 21.4±1.8 μg m^{-3} in monsoon (Fig. 6 and Table 1). Though, the values of SO_2 concentration crossed the Indian National Ambient Air Quality Standards (15 μg m^{-3} annual average) set for sensitive areas[30] during the monsoon period, they were within the USEPA's NAAQS standards (56.6 μg m^{-3} or 30 ppb annual arithmetic mean)[27].

The main source of SO_2 in this region could be combustion of fuels. Its contribution in the atmosphere depends much on sulphur content of the fuel used for heating and power generation. Besides, the open burning of refuse, municipal incinerators and prevailing hot water sulphur springs at Vashist (Manali), Ramshila (Kullu) and Manikaran in the Kullu valley also contribute some amount of sulphur dioxide to the atmosphere. Incomplete combustion of fossil fuels, combustion of oil, coal, gas in both automobiles and industry and volcanic gases contribute to SO_2 concentrations in ambient air [30].

Table 1—Seasonal variation of trace gases (μg m^{-3}) monitored in 2003 in the hill spots of the Kullu valley

Seasons	Mohal (Kullu)		Kothi (Manali)	
	SO_2	NO_2	SO_2	NO_2
Summer (April-June)	18.8±1.3	3.8±0.6	12.1±5.6	2.2±0.3
Monsoon (July–September)	13.8±2.6	3.1±1.3	21.4±1.8	1.0±0.1
Autumn (October–November)	10.1±1.0	7.6±1.0	7.3±0.2	3.8±0.6
Winter (January-March and December)	13.7±4.3	2.7±0.9	12.4±2.5	1.5±0.6

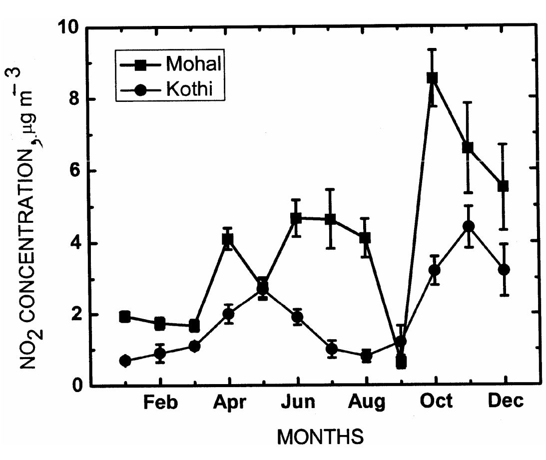

Fig. 5—Nitrogen dioxide (NO_2) concentrations (in 2003) at Mohal (1150 m asl) and Kothi (2530 m asl) in the northwestern Himalaya

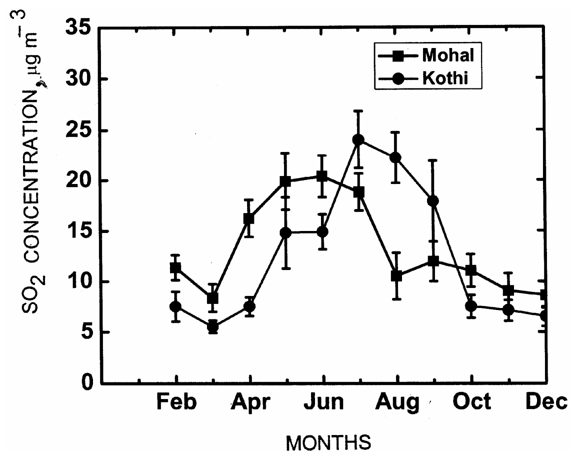

Fig. 6—Sulphur dioxide (SO_2) concentrations (in 2003) at Mohal (1150 m asl) and Kothi (2530 m asl) in the northwestern Himalaya

5 Conclusions

Following are the main conclusions emerging from this study:

(i) The concentrations of O_3, NO_2 and SO_2 are within permissible limit set by USEPA's standards though the concentration of SO_2 in summer and monsoon period crossed marginally the standard set by Indian NAAQS.

(ii) On an average, surface ozone concentrations at Mohal, Manali and Kothi were 20.2 ppb, 28.9 ppb, 27.8 ppb, respectively. The mean diurnal variation is small at high altitude station Kothi compared to the other two locations. The photochemical process in presence of NO_2 is primarily responsible for the observed increase in O_3 during the day. However, long range transport may also be important.

(iii) The seasonal variation of NO_2 is almost similar at Mohal and Kothi during September-March period but differs later. The abnormal increase in NO_2 during October-December at Mohal could be attributed to the increased anthropogenic activity during Dussehra season. While NO_2 concentration at Kothi shows a minimum in August and it peaks at Mohal in July.

(iv) The seasonal variation of SO_2 at Mohal and Kothi are similar in nature though shifted in periods. The concentration of SO_2 at Mohal is ~1.8 times larger in summer and at Kothi 2.9 times larger in monsoon as compared to the respective values during autumn season when its concentration reaches a minimum at these sites. Increased tourism activities could particularly be responsible for this feature.

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