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Diurnal Variation of Ambient NH₃ in Relation with Agricultural Activities and Meteorological Factors at a Rural Site in North India

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Abstract

Ammonia is a chemically active gas which accelerates particulate matter formation by combining with nitrate (NO₃⁻) and sulphate (SO₄²⁻) in acid cloud droplets, thereby reducing air quality. Since pre-industrial times, NH_a emissions have more than doubled globally, owing to increase in agricultural activities and fertilizer usage. In this study, ambient NH₃ monitoring was done during selected periods on event basis in summer season (kharif crop) at a rural site of Jhajjar district of Haryana. Collected gaseous NH₂ samples in absorbing solution (1.4ml H₂SO₄ in 1 litre water) at a flow rate of 1 LPM were prepared with the indo phenol-blue method and analyzed using spectrophotometer at 630nm. Here, we present the day-night variation in ambient NH₃ concentrations emitted from various agricultural activities such as synthetic fertilizers, animal manure, biological N-fixation, the crop residue in the field after harvest, biomass burning, etcin relation with meteorological parameters. Its emission was recorded as 1 to 45; 63 to 190; 98 to 187 and 56 to 249 µg m⁻³ during sowing, fertilizer addition, grain filling and biomass burning respectively. Concentration of NH, during the sowing period i.e. 1 to 45 µg m⁻³ can be considered as baseline values. The Concentration of ambient NH_a reached its maxima at night and minima duringmidday. NH₃ concentration was observed to be high during night time which might be due to reduced dispersion as the atmospheric conditions are stable at night. Concentration of NH₃ is majorly influenced by wind speed and wind direction & its dependency on these meteorological parameters suggested a local source influence indicating that the nearby agricultural



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fields might be the major NH_3 contributors at the observational site. This study suggests that the knowledge of NH_3 levels measured at various stages can help in implementing N efficient management system and emissions can be reduced by minimizing the Nitrogen (N) input during different stages. These measurements are also helpful in making fertilizer policy, and guidelines for farmers.

Introduction

The biogeochemical cycling of nitrogen is of vital importance due to the role of nitrogen in both aquatic and terrestrial ecosystems. However, global nitrogen cycle is being disturbed by the unchecked increase in food and energy production resulting in the accumulation of Reactive Nitrogen (Nr) in various environmental matrices. Generally, Nr refers to any formof nitrogen compound that is biologically active, photo chemically reactive and radioactively active in the biosphere and atmosphere of theearth.1-2 Nr includes inorganic reduced form of N (NH_3 , NH_4^+), inorganic oxidized forms (NO_x, NO₃⁻, N₂O, HONO) & organic nitrogen compounds.³ However, these Nr species are extremely important in atmospheric chemistry. Hence forth, the monitoring of Agricultural nitrogen air emissions and understanding their behaviour in atmosphere is crucial due to impending environmental and human health concern of the latter. One of the major sources of the incessant increase in the nitrogen air emissions is the agriculture activities followed by livestock, human waste, transport sector. Ammonia is one of the most important reactive Nitrogen (Nr) compounds emitted from agricultural sources. Other minor anthropogenic sources of atmospheric NH₃ emissions include animal manure, slash burning and industries⁴ along with the natural sources such as forest fires, soils and oceans.5-6 In India, the Indo-Gangetic plain (IGP) has been reported as hotspot of both NH₃ and NH₄⁺ emissions due to intense agricultural activities in the region.7 The major contributor for NH₃ emissions in agricultural systems includes inorganic fertilizers, livestock manure, increased biological N-fixation, biomass burning and residue added to the field after crop harvesting. For instance, as reported by Misselbrook et al.,8 agricultural sector alone is responsible for about 90% of the total atmospheric NH₃ emissions. According to Vitousek et al.,9 human interventions have approximately doubled the fluxes of Nr species viz NH₃, NOx,NH₄⁺, NO₃⁻, and N₂O.¹⁰ Human activities contribute approximately 170 Tg year-1 of Nr into agroecosystems, where inorganic fertilizer additions account for about 80 Tg Nr annually.¹¹ For Indian Scenario, Aneja et al.,12 reported the highest contribution of fertilizer application and livestock towards NH₃ as 2696.6 Gg and 1704.8 Gg respectively. Among various inorganic fertilizers, urea is the most widely used N fertilizer in south Asia.13 Furthermore, urea is the highest contributor (~ 94%) of NH₃ because of its large scale use and nitrogen content is maximum among all other fertilizers being used in India.14 Urea alone contributes 2481.5 Gg NH₃ whereas di-ammonium phosphate (DAP), NPK, ammonium phosphate contributes 124.8 Gg, 89.7 Gg and 0.6 Gg NH₃ respectively, annually. Also, the rate of increase of urea in agricultural fields is estimated to be 92% per year, thus adding to the Nr budget.13 45-55% of these Nr inputs are recovered by crop biomass whereas remaining is lost from agricultural systems via. leaching, erosion (32-45 Tg N year-1) and denitrification(26-60 Tg N year-1).10-11 Hence directly or indirectly influences the Nitrogen cycle.

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A remarkable portion of N-fertilizer or excessreactive soil N (Nr) is lost through volatilization in the form of ammonia(NH₃) and surface run off which significantly contributes to the Nitrogen loss. Bouwman et al.,15 reported that 10-30% of the applied N fertilizer is lost through volatilization process. Once NH, is applied to the soil by fertilizer addition, NH,* gets retained on the exchange sites. NH₄⁺ is either nitrified to nitrate (NO3-), or decomposed to NH3, depending on soil and environmental conditions. High temperature inhibits the nitrification process as reported by Grunditz and Dalhammar,¹⁶ and hence, ammonia volatilization increases with increase in temperature. In agroecosystems, NH, emissions can be reduced by minimizing the N input, increasing the efficiency of N fertilizers.17

Some studies have been carried out by research groups for NH₂ inventories in Asian region¹⁸ which

mainly relied on emission factors. However, no country-specific emission factor for NH_3 is available, there is high uncertainty in the emission estimates of NH_3 in India.¹⁹ As reported by Parashar *et al*²⁰ NH_3 emissions from fertilizers and livestock are estimated to be 1175 Gg and 1433 Gg, respectively. According to the estimates given by Yang *et al*,²¹ the annual concentration of gaseous ammonia measured in an agricultural field varied from 1.3 to 17.2 µg m⁻³ and observed to be higher in summer than in winter season.

Fate of atmospheric ammonia (NH₂) is highly variable. NH, is the most abundant alkaline constituent in the atmosphere²² and a precursor molecule for secondary aerosol formation causing severe air pollution in East and South Asia by contributing ambient levels of fine particulate matter (PM_{2,5}).²³ Transformation of atmospheric NH₂ into NH₄⁺ via chemical processes of precursor gases (NH₃, SO₂, NOx) occurs either by condensation or by direct nucleation. The major inorganic compounds formed through the gas-to-particle formation process are ammonium bisulfate (NH, HSO,), ammonium sulfate (NH₄)₂SO₄, ammonium nitrate (NH₄NO₂) and ammonium chloride (NH₄Cl).Due to the substantial increment in atmospheric NH₃ emissions and its transformation, more ammonium (NH_{4}^{+}) is returned to terrestrial and aquatic ecosystems via dry and wet deposition mechanisms. In temperate regions, precipitation is the main scavenging mechanism whereas in dry regions (with no or little rainfall) dry deposition is the dominating mechanism for the removal of atmospheric pollutants. Since, in India~ 90 % of precipitation occurs during monsoon period (June to September), dry deposition is the dominating mechanism for the scavenging of atmospheric pollutants throughout the year.24 Moreover, the fate of Nr species is determined by atmospheric acidity, particulate loading, land use dynamics, and photochemistry of the atmosphere.²⁵ The deposition of NH₂ and NH₄⁺ in various compartments across the globe causes a cascade of environmental problems such as eutrophication of terrestrial and aquatic ecosystem, biodiversity loss, forest damage, water and soil acidification.²⁶⁻²⁷ Hence its role in atmospheric phenomenon such as neutralisation of cloud water, precipitation and aerosol formation typically in the fine particle size range is pivotal.28

In addition, the formation of ammonium particulates increases the residence time of NH, in the atmosphere thereby influencing the geographic distribution of acidic species. Also, deposition of these fine particles deep into the lungs causes morbidity and mortality in humans, alterations in visibility and climate.29 Henceforth, the monitoring of atmospheric NH, in agricultural fields is one of the decisive parameters that needs to be considered. For this the study was conducted at a rural site in Jhajjar district of Haryana with the following objectives- i). to quantify the NH₂ concentration and its variation during different growing stages of crops sown in our study area and ii). to study the effect of meteorological parameters in order to identify the possible transformations of NH₂. The NH₂ emission rate from various agricultural activities and its downwind concentration mainly depends on meteorological conditions. There exists a positive relationship between incident solar radiation/ air temperature and NH₃ emission from surfaceapplied fertilizers.³⁰⁻³² The Solar radiation increases The NH₃ emission by increasing the atmospheric turbulence and hence, NH₃ evaporates from the surface. Apart from that evaporation of water due to increasing air temperature increases the total N concentration at the surface. Consequently, the NH, at the surface is transported upwards by atmospheric turbulence and sideways, by advection. Hence, NH₃ emission levels are related to wind speed, relative humidity and temperature.32 Higher humidity and lower temperature favours the formation of NH,⁺ aerosols in presence of NO₂ and SO₂.

Methodology

Site Description

Astudy was conducted at a rural site in Chhuchhakwas village of Jhajjar district in Haryana (Fig. 1). It lies between 28°22' - 28°49' North latitudes, and 76°18' -76°59' East longitudes. The village is located at a 13 kms from Jhajjar district which lies in the south-east part of the Haryana state covering total geographical area of 1834 sq.km. The climate characteristics of this area are hot summer, cold winter and moderate rainfall of about 444 mm. This area comprises approximately 87.03% land under agriculture and 6.77% build up area, indicating very less development in terms of urbanization. The district covers an forest area of 41 km², net sown

area of 1670 km^2 and cultivable area of 1760 km^2 (Agriculture department Jhajjar). The sampling site

represents a typical rural atmosphere surrounded by agricultural fields in all directions.



Fig. 1: Map of the sampling site

 Table 1: Monthly variation of temperature, wind speed and relative humidity during the sampling period

Month	Temperature (°C)	Wind speed (mph)	Relative humidity (%)
July, 2017	37.40	9	39.46
August, 2017	35.83	7	52.13
September, 201	7 35.28	4	38.06
October, 2017	34.86	3	32.40

Meteorological Data

The Temperature, relative humidity and wind data of the study region for every month (July to October) were downloaded from the world weather online.com. Table 1 shows the mean values of all above mentioned meteorological parameters during the sampling period.

Experimental Setup

Collection of gaseous NH_3 samples was performed on 6 hr basis during both daytime and night time (8am-2pm and 10pm-4am) from July to October 2017 using a sampling assembly (Fig. 2) consisting of a low volume pump operating at a flow rate of 1 LPM. Ammonia gas was absorbed in absorbing solution (20 ml of 25 mM H_2SO_4) in a standard impinger for 6 hours.³³ The aerosol samples were collected on the PTFE filters (diameter 47 mm and pore size 0.2µm) placed upstream to the impingers. Filter pack was connected to the impinger through silicon tubing. Collected gaseous NH₃ samples in absorbing solution were transferred into centrifuge tubes, preserved in refrigerator and analyzed by catalyzed Indophenol-blue method. It includes photometric determination of NH₃ which is based on reaction with phenol and hypochlorite producing Indophenol, intensely blue in alkaline medium which is further analysed using spectrophotometer at 630nm. The collection efficiency of impinger technique for NH_3 capture was estimated by using two impingers in series (Fig. 2), using the formula:

Collection efficiency =
$$\eta_2 * 100/(\eta_1 + \eta_2)$$
 ...(1)

where η_1 and η_2 are the values of optical density in impingers 1 and 2, respectively (Fig. 2)



Fig. 2: Flow diagram of sampling assembly

Hence, the collection efficiency of NH_3 using this method was estimated to be 67%. Furthermore, certainty and reliability in data collection was maintained by rejection of daily samples in case of any disruption or reduction of post-sampling flow rate by more than 10% of the pre-sampling flow rate.

Agriculture and Cropping Pattern

The farming practices in Jhajjar district are dominated by agriculture activities and animal husbandry. The main crops sown in this region are Kharif cropsand Rabi crops. Kharif crops also known as monsoon or autumn crops, are cultivated at the onset of the monsoon season around June and harvested by September or Octobereg. Oryza Sativa (Paddy), pearl millet (bajra), gossypium (cotton), Cyamopsistetragonoloba (guar) etc. and Rabi crops are agricultural crops that are sown in winter in month of November and harvested in Marcheg. Triticum aestivum (Wheat), Brassica juncea (mustard), Hordeum Vulgare(barley), etc.

Results and Discussion

Concentration of NH₃ Over the Study Period

Table 2 gives the concentration of NH₃ over various periods (growing, post-fertilizer, grain filling, post-

harvest) for the kharif crop (mainly pearl millet in our study area). Growing period in the month of July shows the least NH₃ concentration with an average value 29.68 µg m⁻³ as compared to all other time phases. These values can be assumed as background values. Nitrogen fertiliser can either be incorporated into the soil (basal dressing) before growing the crop or applied to the soil surface (top dressing). In basal dressing, nitrogen fertiliser is placed below the soil layer which limits the rates of nitrification, denitrification, volatilization and therefore, enhance efficiency of N fertilizer use.34 As Diammonium phosphate (DAP) fertilizer is mainly used in basal dressing (30 kg/hectare) prior to growing, the levels were very less as compared to the levels after top dressing (commonly urea) fertilizer application. The levels increased markedly after urea was applied on the soil surface on 1st August with an application rate~100kg/hectare. Among various fertilizers being used in India, the NH₃ concentrations peaked in conjunction with the application of urea. Contribution from application is highest not only because of its excessive usage in Indian agriculture but also due to the large emission factor for NH₃

emissions.³⁵ The average were measured as 96 μ g m⁻³ and 149 μ g m⁻³ during day and night time respectively. The levels of NH₃ are very high during grain filling and post-harvest period. As plant uptake of soil-N decreases towards maturity,

 NH_3 volatilization from soil increases. This may be responsible for higher NH_3 emission towards maturity.For further analysis during these months, variation of NH_3 was related with wind directions as described below (Fig. 7).

Date	Sampling periods	Day time NH ₃ conc.(µgm⁻³)	Night time NH ₃ conc.(µgm ⁻³)	
09 July 2017		1	-	
10 July 2017		34	-	
11 July 2017	Sowing period	41	-	
12 July 2017		45	-	
13 July 2017		25	-	
02 Aug 2017		115	163	
03 Aug 2017	Post-fertilizer	122	88	
04 Aug 2017	addition	84	190	
05 Aug 2017		63	154	
12 Sept 2017		141	170	
13 Sept 2017	Grain filling	187	139	
14 Sept 2017		125	144	
15 Sept 2017		98	121	
08 Oct 2017		56	176	
09 Oct 2017	Post-harvest	104	234	
10 Oct 2017		117	249	
11 Oct 2017		151	184	

Table 2: Day-Night concentration of NH₃ over various time periods of the kharif crop (pearl millet in our study area)

Diurnal Variation of NH₃ Over the Study Period Fig.3 shows the diurnal variability in NH₃ concentration over the study period. NH₃ concentration is higher during night time as compared to the day time. Similar variation in NH₃ emissions were also reported by other researchers.³⁶ During day time NH₃ concentration varied from 1 to 187µgm⁻³ and during night time it lies in the range 88 to 249 μ gm⁻³. The mean concentration of NH₃ for the day and night time was found to be 89 and 168µgm-3 respectively. Fig.4 shows the variation of average day and night time concentration of NH₃ over the sampling months. Higher NH₃ concentrations observed during night time might be due to stable atmospheric conditions (less turbulence) which resulted in reduced dispersion of gaseous NH₃ in the atmosphere because of the accumulation and inefficient vertical mixing within a relative shallow boundary. This pattern is in consistent with the studies by other researchers.³⁷⁻³⁸

Meteorological Data

The Mean temperature during the sampling periods over the month of July, August, September, October observed to be 37.4°C, 35.83°C, 35.28°C and 34.86°C respectively whereas the average relative humidity levels were found to be 39.46%, 52.125%, 38.06% and 32.40% respectively. The prevailing wind speed and directions were mostly in the range 2–7 mph with distinct patterns in W, E, NW and ESE directions.



Day & Night variation

Fig. 3: Diurnal variation of NH₃ concentration over the study period.



Fig. 4: Average day- night time variation of NH₃ over the sampling months.

Variation of NH_3 with temperature and relative humidity is as represented in Fig. 5. There was no significant correlation observed between NH_3 concentration with temperature and relative humidity in our catchment (Fig. 6). The Levels of $\rm NH_3$ are negatively correlated with temperature and relative humidity during day time. In contrast, $\rm NH_3$ concentration are positively correlated with temperature & negatively with relative humidity during night time. The relation can be explained

on the fact that high temperature and low relative humidity conditions favours the formation of NH_3 from NH_4^+ . The reason being, in summer, particulate

ammonium nitrate is volatile, and hence, $\rm NH_4NO_3$ will be in gaseous phase. 39



Fig. 5: Variation of NH₃(µg m⁻³) with RH (%) and temperature (°C) during day time and night time



Fig. 6: Correlation of NH₃ (µg m⁻³) with RH (%) and temperature (°C) during day time and night time

Possible Sources of NH,

Since, NH_3 is either subjected to dry deposition or readily converted to NH_4^+ . Therefore, high levels

of gaseous NH₃ are expected close to the surface and at less distance from the emission sources.⁴⁰⁻⁴¹ Hence, there might be some local emission sources near the observation site. The wind rose plots suggested higher concentration from the west direction of the sampling site in the month of July and August (Fig.7). Application of large amount of inorganic nitrogen fertilizers in the nearby agricultural fields might be contributing to ambient NH₃ concentration in these sampling months. Several researchers from various parts of the world also reported similar observations.⁴²⁻⁴³ During the grain filling period, in the month of September, the level of

ammonia was considerably high, and the prevailing wind direction was mainly northwest (Fig.7c). As the village is in the northwest direction of the sampling site, livestock population, solid waste generation might be contributing towards higher NH_3 levels. In addition, wind was very diverse majorly from E and ESE direction during post harvest period in October. In addition to biomass burning, there might be various sources like animal waste, solid waste, etc. from nearby areas.



Fig. 7: Wind rose plot a) July 2017 b) August 2017 c) September 2017 d) October 2017















Fig. 8: Day & night wind rose plots (e, f) 8 October 2017 (g,h) 9 October 2017 (i, j)10 October 2017 (k, l) 11 October 2017, respectively

It is observed that the NH₃ concentration during day & night time on 8 October, 2017 was 56 μ g m⁻³ & 176 μ g m⁻³ (Table 2). Wind rose diagrams were plotted to understand the significant gap between the diurnal values on this particular day (Fig.8). It can be inferred that there were only minor changes in wind directions

during day and night time. Hence, the high NH_3 levels starting from night time (8 October, 2017) can be attributed to the onset of biomass burning during post harvest period. However, to rule out any other possibility further investigation is required.

Site	Background	Period	NH ₃ (μg m ⁻³)	Reference
North Carolina (USA)	Agricultural	July-September 2004	11.1	Wilson and Serre ⁴⁴
Dongbeiwang (China)	Suburban- agricultural		9.5	Shen <i>et al</i> .45
Beijing (China)	Urban	August 2007	24.3	Lanniello <i>et al</i> .46
Dhangadi (Nepal)	Regional	September 1999 -June 2001	14.6	Carmichael <i>et al</i> .47
Agra (India)	Urban	July-September November-February (1997-1998)	10.8	Parmar <i>et al</i> .48
Okhla, New Delhi (India)	Urban industrial	October 2012 -September 2013	40.7	Singh and Kulshrestha ¹⁴
Mai, U.P (India)	Rural-agricultural	October 2012 -September 2013	50.5	Singh and Kulshrestha ¹⁴
Chhuchhakwas, Haryana (India)	Rural-agricultural	July 2017 Aug 2017 Sept 2017 Oct 2017	29.7 122.9 141.2 159.5	Present study

Fable 3: Comparison of ambient NH	çoncentrations at various	characteristic sites worldwide
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Comparison of Concentrations of Gaseous NH₃ With other Studies

Generally, the level of atmospheric NH_3 is higher in tropical regions as compared to temperate regions. This is probably due to high temperature in tropical regions resulting in higher evaporation rates of gaseous NH_3 from agricultural soils, animal waste and other sources. In addition, abundance of alkaline dust in tropics favours the existence of alkaline NH_3 in its gaseous form. In contrast, dominance of H_2SO_4 in acidified atmosphere in most of the temperate areas results in the formation of NH_4^+ and SO_4^{-2} aerosols.³⁸ Table 3 gives a comparison of NH_3 concentrations at various sites worldwide.

Conclusion

The average NH₃ level in the growing period, when DAP was incorporated into the soil was very less i.e. 29.68 μ gm⁻³. This is because addition into the soil increases the efficiency of fertilizer use. The concentration reached to considerably high levels, 96.53 μ gm⁻³ and 149.40 μ gm⁻³ during day and night time respectively, after addition of N-fertilizer (urea) on the soil surface. The wind direction indicated nearby agricultural fields to be the main source of these high levels. Average NH₃ concentration during day and night time were observed to be 138.41 μ g m⁻³ and 144.01 μ g m⁻³ respectively prior to crop harvesting. Such high

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levels might be due to the contribution from livestock, sewage, etc. as the winds were predominantly from North-west (village) direction during this sampling period. Ambient NH_3 concentration reached its maxima at night and minima during midday.

The study suggested that to reduce anthropogenic forcing of N cycle globally, there is need to address the inefficiency of N fertilization. In addition, there is a need to examine the formation of ammonium aerosols in the air around the agricultural areas in a comprehensive manner to address the health and climate issues related to ammonium aerosols.

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Conflict of Interest

The authors do not have any conflict of interest.

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