

Lightning produced nitrogen oxides in the lower atmosphere – An overview

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Nitrogen oxides are dominant constituents of the atmospheric lower regions namely, troposphere and stratosphere. Lightning phenomenon plays an important role in governing the odd nitrogen oxides in the lower atmosphere and hence in global change process. In this paper, studies made by different workers on the lightning produced nitrogen oxides and other related phenomena have been reviewed. Studies in this direction are very sparse in India. Besides the ground based, balloon-borne and satellite-borne measurements, aircraft measurements in India and elsewhere are essential for providing global information. In order to make reliable measurements the experimental site should be in remote areas free from heavy industrial pollution, so that the contaminations due to direct influence of anthropogenic activities are avoided.

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1 Introduction

Lightning is one of the oldest phenomena¹⁻⁴. It was considered to be a ghostly phenomenon and people were scared of watching the thunderous and impulsive sparking phenomenon. Gradually the interest started growing and the phenomenon was watched from hide out places only because scaring thunderous lightning sound was heard after some time-delay. The importance of lightning grew slowly since it was found to affect the trace gases in both the hemispheres. In 1827, Von Liebig⁵, for the first time, proposed that lightning could be an important source of atmospheric nitrogen fixation and this initial conjecture was later supported by many investigations⁶⁻⁸. Refined features of lightning in the earth's atmosphere were observed and similar phenomena are now known to occur in many planetary atmospheres and also in the atmospheres of some of the planetary moons⁹. Despite these developments, many aspects of this phenomenon and their impact on different parts of earth's atmosphere are not yet fully investigated and understood. The lightning features of cloud-to-ground (CG) and cloud-

to-cloud (CC) have been recorded and the nature of electromagnetic wave generation has been studied. The lightning discharge strokes and their interactions with the upper and the lower atmospheric gases have been studied in some detail. But, the details of cause and effect scenario of the phenomena still remain to be established. The intent of this paper is to project the details of lightning phenomena that have not been adequately explored and studied^{8,10,11}. However, in India the major effort has gone into the recording of lightning waveforms on the ground. Lightning from above the cloud top has not been adequately recorded and studied. In depth study of the effects of lightning associated electric fields and electromagnetic radiations on the gaseous content of lower atmosphere has not been made. Further, airborne measurements of nitrogen oxides in the lower atmosphere, i.e. troposphere and lower stratospheric regions, are lacking in the context of Indian studies. The airborne measuring techniques have been developed by the research groups in USA and initial measurements were made during the last decade, making use of commercial and specially flown probing aircrafts. The

results of various measurements have been published¹⁰⁻¹³. Some of the papers have reported the airborne measurements of lightning and their correlations with *in situ* measurements of tropospheric and stratospheric content of nitrogen oxides. The initial measurements of nitrogen oxides^{12,13} and their correlation with flying aircraft measurements were studied by the National Center for Atmospheric Research (NCAR), Boulder, Colorado, USA and the results were reported by Ridley *et al.*¹⁰ and Zeng *et al.*¹¹. These measurements clearly demonstrated that lightning did influence the chemistry of nitrogen oxides and O₃ density in the troposphere and upper stratosphere^{8,14-18}.

The global variations of lightning occurrence rates are well known, e.g. Central Africa, the low latitude areas of America and the equatorial area of Southeast Asia show a peak occurrence¹⁹ at about 2000 hrs LT. Recording of lightning wave form, their classifications, analysis of various kinds of waveforms and their Fourier components have been studied at Varanasi²⁰⁻²³ (geomagn. long. 154°E, lat. 16°N) since a long time. Apart from waveform classifications and characterization, features of the lightning generated electromagnetic waves were also studied. In association with lightning, whistler wave phenomena were recorded at Srinagar²⁴ and later on were also recorded at Varanasi²⁵. The features of whistlers recorded at low latitudes have been reviewed²⁶. Studies of atmospheric waveforms carried elsewhere and at Varanasi were limited to their classifications only. With the help of balloons and aircraft payload such measurements are recently being made and results are being reported globally²⁷. The emphasis on lower altitude using balloons and aircraft payloads was not available in the initial phase. Now a global programme is launched and it is high time that Indian efforts should be directed to participate in this global programme.

In this paper the role of lightning phenomena in governing the chemical compositions of low latitude lower atmosphere has been highlighted. Various features of lightning generated gaseous density of lower atmosphere as accomplished by some of the research groups are illustrated. An important aspect of this study is to go into the depth of ion-kinetics. In general, the chemical reactions are studied in the laboratories by manifesting the various features of atmospheric regions at lower altitudes. The optical, X-ray and γ -ray radiations are known to change from

one lightning event to another. Therefore, in this paper, the mean values of the parameters have been used and the variations that could be depicted from time to time have been discussed. The outcome of this study is to assert that research groups with previous background should get together to carry out lightning spectral measurements using aeroplane. In addition, the airborne and ground-based measurements of electromagnetic spectrum should also be made. The research effort is novel and its findings may add significant contribution to global efforts that are likely to grow and enrich our understanding of this unique phenomenon.

2 Lightning generated VLF waves

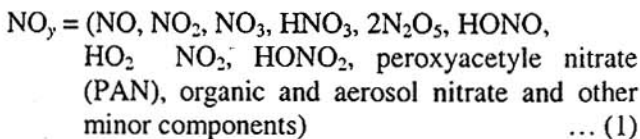
Radio recording observations have revealed close association between whistlers and lightning. Evidences show that many whistlers originate in intense lightning discharges with peak in their energy spectra near 5-8 kHz. Measurements show that as one moves away from the peak there is 6 dB drop in the spectra at 1 kHz and 15 kHz. The non-whistler producing lightning could have spectral peaks either below 1 kHz or above 10 kHz. The total electromagnetic energy in a whistler producing lightning could be approximately 2×10^5 coulombs, about 10 times larger than the average lightning flash. The intensities of electromagnetic waves generated during lightning discharges are known to decrease with frequency f as f^{-2} at lower frequencies and as f^{-1} at higher frequencies. Early observations showed that whistlers were prevalent at higher latitudes and their occurrence rate decreased with decreasing latitudes.

Whistlers and sferics are usually ascribed to return strokes of the lightning discharge, which is considered to be powerful transmitters of ELF and VLF waves. Recording of VLF waves carried out at different locations^{26,28-33} showed that VLF wave activity is confined to the regions near 120° E (South-East Asia), near 30° E (Africa) and 50°-140° W (America), thus establishing a close relation between lightning and whistler activity as far as global coverage is concerned. It is now well known that the stronger lightning signals, apart from generating ELF/VLF waves, also significantly affect the nitrogen oxides in the troposphere and stratosphere. The densities of these oxides do require additional facilities for measurements along with the ground-based detailed spectral measurements of cloud-to-ground lightning discharges.

3 Production of nitrogen oxides and reacting molecules

Study of ion-kinetics in the ionosphere was fully developed since it governed the response of incident electromagnetic waves. The importance of lower atmosphere was not known for a long time since electromagnetic waves were supposed to propagate through this region without any interaction through the process of multiple reflections from ionospheric layers. The frequencies higher than the critical frequency of Es and F2-regions were known to penetrate through and never return. A breakthrough in electromagnetic wave propagation came up and it was known that the very high frequency and microwave signals are affected by the density gradients of atmospheric molecules with excited states. This property of the gas density at lower altitude was used for propagation of microwaves through tropospheric duct. The path of propagation of radio frequency signals to large distances involved large multiple reflections and consequently attenuation of propagating signal became very significant. On the other hand, the microwave signals has tendency of refracting through the tropospheric and stratospheric gas density inhomogeneities. The large distance duct propagation of microwave signals, therefore, needed periodic boosters so as to transmit signals to longer distances.

Nitrogen is a primary constituent of earth's atmosphere with 68% and oxygen distribution of 18%. A large number of constituents form only 3% to 4% of varying population. The nitrogen and oxygen react with each other and form various oxides that are collectively written as



These details were first given by Crosley³⁴. A subclass of NO_y is designated as



The basic reaction producing NO_x along with reaction rates is given in Table 1 (Refs. 34-36).

The above reaction rates³⁴⁻³⁶ are valid for temperature and pressure conditions of 217 K and 0.224 atm. The photolysis rates have been taken for daytime conditions at a height of 11 km. The above reactions are used to estimate the production of NO_x

and NO_y . The emissions of NO_y are mostly in the form of nitric oxide NO. Since photochemical equilibrium between NO and NO_2 is established within a few minutes, it has become customary to use NO_x instead of NO. The stratosphere and troposphere experiment by aircraft measurements (STREAM) at varying altitudes were carried out by Lange *et al.*⁸ and the measured values of NO_y and NO are shown in Fig. 1. From Fig. 1, it is clearly seen that the NO_y is much larger than NO showing that other oxides forming NO_y increase during short duration in the daytime. This is only one-hour data, and therefore, no definite conclusion can be drawn because of short time of flight. Better data over longer period of time can be obtained using balloon data instead of aircraft measurements. Such measurements would be much more revealing and would depict diurnal variations. It is necessary to make detailed measurements under varying cloud cover conditions in presence of active lightning so as to study the effect on the variations of

Table 1 — Basic reaction producing NO_x ($\text{NO} + \text{NO}_2$)

S. No	Reaction	Reaction rate (ppbs) ⁻¹
1	$\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2$	2.39×10^{-5}
2	$\text{NO}_2 + h\nu + \text{O}_2 \rightarrow \text{NO} + \text{O}_3$	0.01 sec^{-1}
3	$\text{HONO} + h\nu \rightarrow \text{OH} + \text{NO}$	0.0033 sec^{-1}
4	$\text{OH} + \text{NO} \rightarrow \text{HONO}$	4.63×10^{-2}
5	$\text{OH} + \text{NO}_2 \rightarrow \text{HNO}_3$	9.18×10^{-2}
6	$\text{HO}_2 + \text{NO} \rightarrow \text{OH} + \text{NO}_2$	8.4×10^{-2}
7	$\text{OH} + \text{CH}_4 + \dots \rightarrow \text{HO}_2 + \text{NO}_2 + \dots$	5.2×10^{-6}

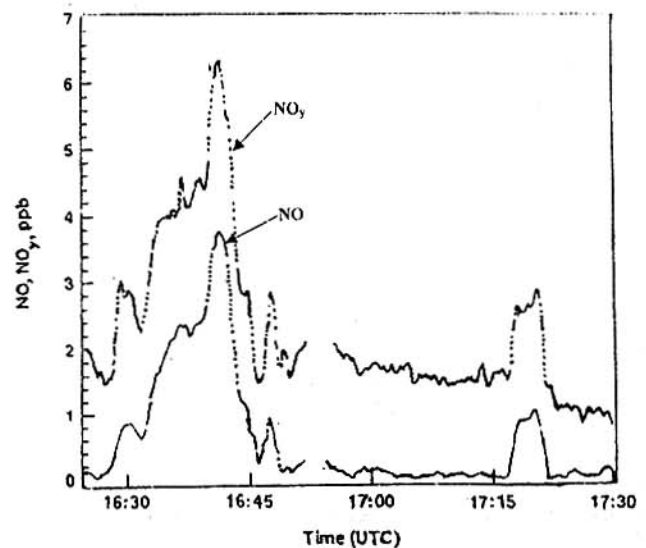


Fig. 1— Time series for the tracers for NO and NO_y particles observed during STREAM flight 7 on 15 July 1998 at zenith angle 25-33°, altitudes 11-13 km in troposphere (Lange *et al.*⁸, 2001).

NO_y and $\text{NO}_x (= \text{NO} + \text{NO}_2)$. Reactive nitrogen oxide is denoted in the tropospheric region by $\text{NO}_x = \text{NO} + \text{NO}_2$, only when both of these oxides are either measured or calculated. It is quite likely that NO_2 measurements are not available due to its comparative intricacies of measurements. In such cases, measured values of NO is used along with calculated values of NO_2 and the sum of these two is denoted by

$$\text{NO}_x = \text{NO} + \text{NO}_2 \text{ (calculated)} \quad \dots(3)$$

Here, also reproduced are the scatter plots of NO_y and NO_z variations with CO concentration variations of 15 s averaged data of all available flights as shown in Fig. 2. Species $\text{NO}_z (= \text{NO}_y - \text{NO}_x)$ is the oxidation products of NO_x . It is found that the NO and NO_2 play a dominant role in the lower atmospheric regions and also at higher altitudes due to incidence of soft solar X-rays as discussed in detail by Barth *et al.*³⁷.

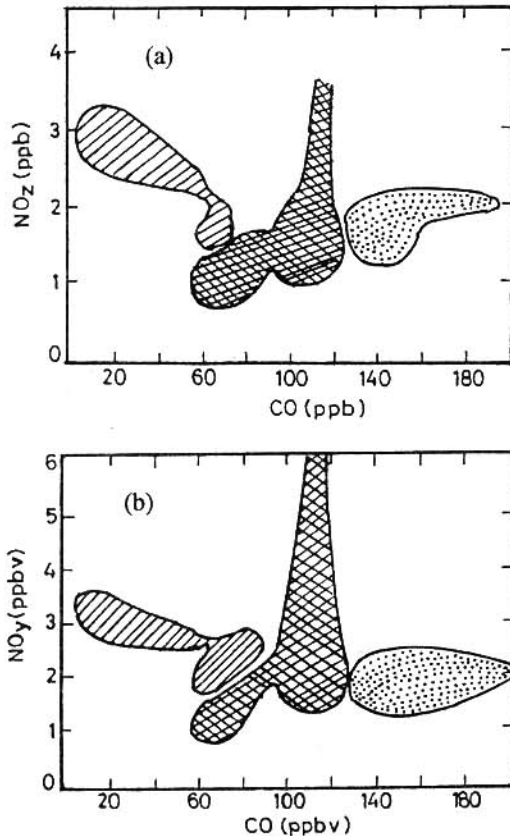


Fig. 2— Scatter plots of 15 s averaged data of all flights [(a) $\text{NO}_z (= \text{NO}_y - \text{NO}_x^*)$ versus CO , and (b) NO_y versus CO . The stratospheric fraction was separated by different contour data points with ozone values > 120 ppb. Stratospherically influenced data represented by slant line hatching, tropospheric data represented by crossed hatching, data with low NO_y and elevated CO are represented by dot hatching (Barth *et al.*³⁷, 1999)].

3.1 Nitric oxide (NO)

The exact measurements of NO mixing ratio as function of altitude are of great diagnostic importance. The NO measurements are carried out using *in situ* and remote sensing techniques. The rapid conversions of $\text{NO} \rightarrow \text{NO}_2$ and $\text{NO}_2 \rightarrow \text{NO}$ near sunset and sunrise complicate the comparison with the model predictions. The NO mixing ratios have been measured using balloon-borne instrumentation and on-board recording. The variation of NO mixing ratio was measured using balloon-borne chemiluminescence technique by Ridley and co-workers³⁸⁻⁴⁰ and is shown in Fig. 3. The measurements carried out for over 2 years were spread between solar zenith angles

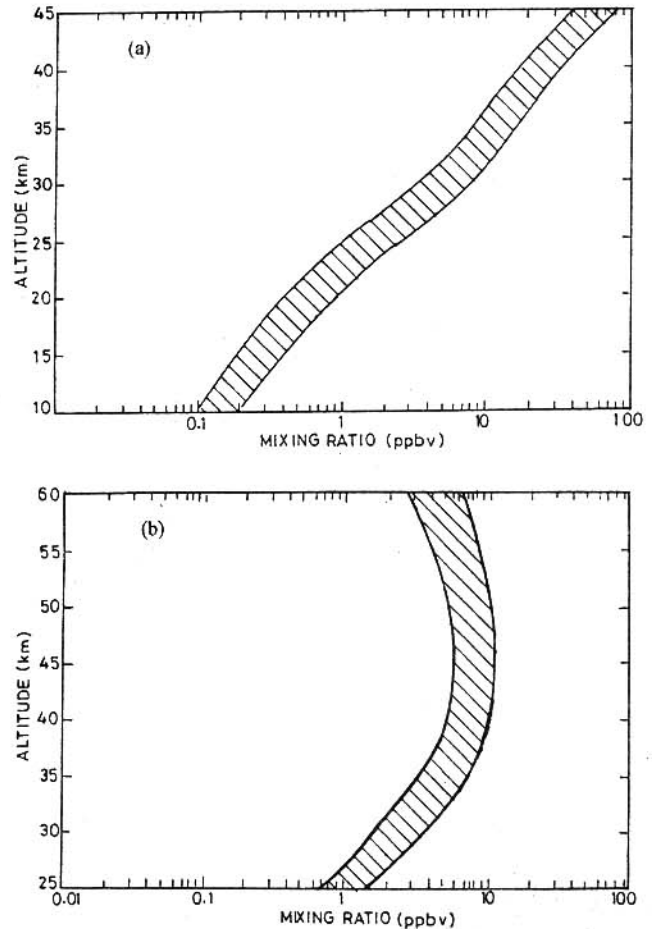


Fig. 3— (a) *In situ* NO mixing ratio measurements of Ridley and coworkers³⁸⁻⁴⁰ [All of the flights were made with instrumentation that incorporated a new inlet and flight calibration procedures. The variation of NO mixing ratio between 32°N and 32°S are bounded by the shaded region between the two lines. The data below 10 km are relatively less in number.] and (b) *In situ* NO mixing ratio rocket-borne measurements by Horvath and Mason⁴¹ [It shows variations above 25 km and extends up to 60 km. All these measurements are at 39°N (Horvath and Mason⁴¹, 1978).]

of approximately 32° S and 32° N with slightly varying temperature variations as shown in Fig. 3(a). The mixing ratio spread is shown to lie between two lines shown in Fig. 3(b). Closer comparisons have confirmed the major NO diurnal variations expected from the stratospheric odd-nitrogen chemistry. Ridley and co-workers³⁸⁻⁴⁰ have carried out extensive measurements in the past. The NO density is found to increase slightly during the daytime. There is not much difference in the results of measurements when one carefully compares the results in the height range that is common to balloon-based and rocket-based measurements as shown in Fig. 3[(a) and (b)]. The NO density is the daytime phenomenon and is controlled by the variations of solar radiated flux. It is known to decrease rapidly as soon as solar radiation becomes weak, as typically shown by the *in situ* measurements by Ridley and Schiff³⁹ and illustrated in Fig. 4. The NO does not show substantial variation in the latitude range varying between 5° and 50° N. The largest difference in this latitude range was found to be about 2.5 that is seen to increase with increasing the northern latitude.

3.2 Nitrogen dioxide (NO₂)

The variations of NO₂ have been measured using ground-based as well as air-borne measurements. The NO₂ is not generally measured in the stratosphere using *in situ* devices. The NO₂ variations are obtained from remote sensing data. These observations particularly are obtained during sunrise or sunset periods when the change of slant column density as a function of the viewing angle is maximum. The sunset mixing ratios of NO₂ are larger than those observed during sunrise.

The NO₂ profile is found to change during the sunrise and sunset periods. The nature of variation of

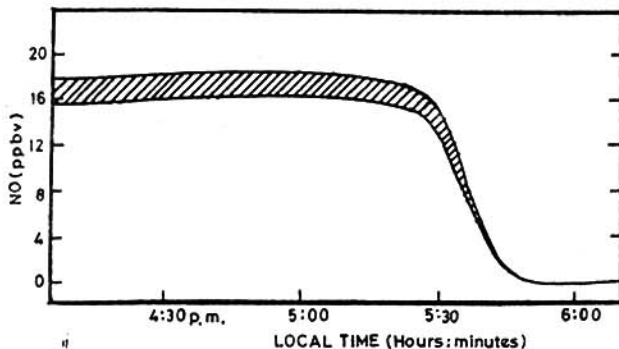


Fig. 4— Sunset *in situ* measurements by Ridley and Schiff³⁹ obtained with a chemiluminescence instrument flown on 08 Nov. 1978 near 32° N, 96° W.

volume mixing ratio is seen to have same trend except some change in the volume mixing ratio as shown and discussed by Evans *et al.*⁴². The NO₂ during winter season, measured by Coffey *et al.*¹⁶, shows significant change in column amount during sunrise and sunset periods as shown in Fig. 5. The NO₂ variations have been illustrated by Murcay and co-workers^{43,44} in a report by Anderson *et al.*⁴⁵. Similar measurements of NO₂ were also reported by them during sunset and sunrise periods. These are shown in Fig. 6[(a) and (b)]. The first measurement [Fig. 6(a)] is at latitude

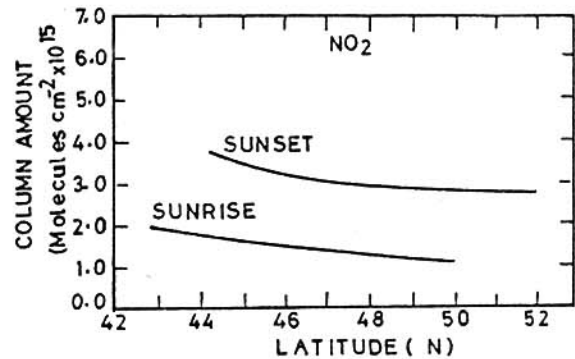


Fig. 5— Sunrise and sunset vertical-column measurements by Mankin and coworkers, who used an infrared absorption apparatus on an aircraft platform (Coffey *et al.*¹⁶, 1981).

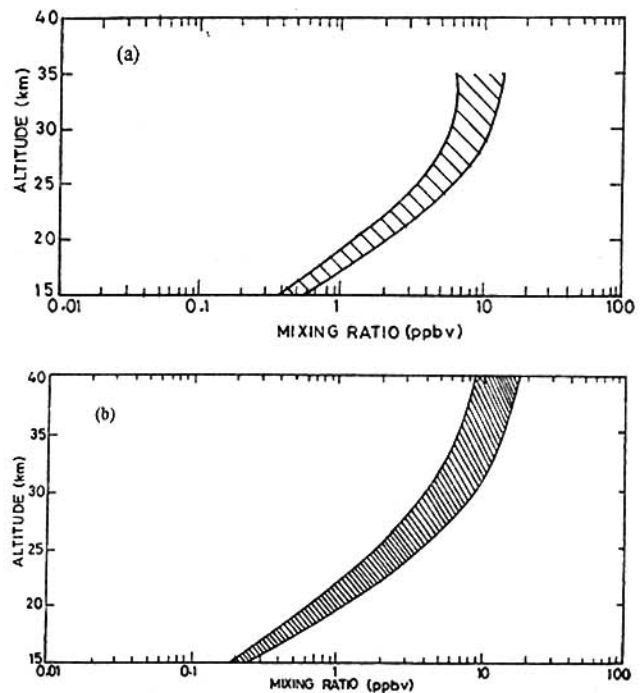


Fig. 6— (a) Remote measurements of the sunset altitude profile of NO₂ made at $51-58^{\circ}$ N latitudes and (b) Remote measurements of the sunset altitude profile of NO₂ made at $32-33^{\circ}$ N latitude (Anderson *et al.*⁴⁵, 1985).

51–58° N, whereas Fig. 6(b) measurements are made at 32–33° N. The two curves show almost similar variations. The solar activity and the cloud cover conditions seem to play important role in the *in situ* measurements at two latitude sites of measurements. The NO₂ does not depict appreciable seasonal variations. The conflict of interpretation of results arises because the presence of cloud cover has not been qualitatively measured. Therefore, a kind of uncertainty remains. It is important that, in future measurements, efforts should be made to resolve this point by considering the nature of cloud cover.

4 Conclusions

From the foregoing discussion, it is quite evident that the ground-based and air-borne measurements of thunderstorm/lightning-generated nitrogen oxides and associated electric field strength would be highly significant, and such information really supplement, to a great extent, to the present understanding of NO_x and ozone chemistry in tropics. Considerable efforts are being made at global levels. It is high time that scientific community and funding agencies should debate and plan out experiment to initiate the study of various features of lower atmosphere at par with ongoing experiments elsewhere. Suitable experimental techniques need to be developed to bridge this gap. It is also essential to develop simultaneous laboratory facilities to perform simulation experiments and to develop realistic models. Moreover, it is of great interest to measure NO density and ionization produced, using multi-experimental techniques and to assess the precision of each of the techniques used. The existing scientific groups in the country need to be augmented with experimental facilities for recording and collecting nitrogen oxides. In addition, future research studies should include aircraft measurements using real-time NO_x analyzers and specially designed probes for recording electric field strength. The feasibility of conducting multi-institutional collaborative field experiments similar to that of SUCCESS (Subsonic Aircraft Contrail and Cloud Effects Special Studies)⁴⁶, SONEX (Subsonic Assessment Ozone and Nitrogen Oxide Experiment)⁴⁷ and STREAM (Stratospheric and Tropospheric Experiments by on-board Aircraft Measurements)⁸ should be ascertained. This kind of measurement missions is a pressing scientific need of the hour and should be acted upon by research supporting agencies and national research and academic institutions in India.

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