

REMOTE DETERMINATION OF THE COMPOSITION, TEMPERATURE, AND VELOCITY OF OUTFLOW OF THE ATMOSPHERIC EMISSION FROM PLANT STACKS USING A RAMAN LIDAR

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Received May 6, 1992*

This paper presents a detailed analysis of the Raman lidar technique for remote sensing of atmospheric emissions from stacks. Analysis made in the paper allows a more correct measurement technique to be proposed. A possibility of measuring simultaneously with the pollutants number densities the emission jet outflow velocity and temperature is shown. This enables one to estimate the gross yields of pollutants at a stack mouth. Thus extended capabilities of the Raman lidar technique are illustrated with experimental results obtained at the West Siberian Steel Production Plant in Novokuznetsk.

INTRODUCTION

As it can be understood from general considerations a control of air quality over industrial areas requires that the parameters of atmospheric emissions from organized emission sources (e.g., plant's stacks) should be known or measured beforehand. It is also clear that some estimates of the amount of pollutants emitted into the atmosphere can be obtained from characteristics of relevant technological processes. At the same time, from the viewpoint of operative air quality control, it is also advisable to have operative and remote means for acquiring data on the composition and intensity of atmospheric emissions from organized emission sources. Among such means the lidars, for example, Raman lidars and differential absorption lidars,^{1,2} are still most advantageous. However, it is worth noting that the lidar techniques developed so far were aimed at measuring only number densities of contaminants. On the other hand it is quite clear that the ecological load on the environment depends not only on the concentration of pollutants but on their gross yields as well.

To estimate the gross yield of the specie from the stack one needs to measure, in addition to its number density, the velocity of the emission outflow from the stack and to know the size of the emission jet.

In this paper we discuss a possibility of extracting information about the emission composition and outflow velocity simultaneously from Raman lidar returns from the emission molecular components. In addition, a possibility of estimating the emission temperature from these same Raman lidar returns revealed in this study is also discussed. We think that thus obtained data on the emission temperature could be useful for remotely assessing the heat pollution of the atmosphere by emissions from plants stacks.

ANALYSIS OF THE RAMAN-LIDAR TECHNIQUE

As early as at the first stage of development of lidar techniques the Raman effect has attracted much attention of many researchers^{3,4,5} by its remarkable property that irradiation of a gas mixture with a monochromatic light results in simultaneously observable spectral responses from the mixture molecular constituents, or in other words in Raman spectrum of the mixture. Moreover, in many

practical situations the so-called Raman lines of different molecular species are spectrally resolved. Just this property of Raman effect enabled different research groups^{6,7,8} to develop remote spectroscopic techniques for detecting gaseous contaminating species in the exhaust products of automobiles and in the atmospheric emissions from plant stacks. A direct proportionality of the Raman line intensity to the number density of the corresponding molecular specie of the mixture makes the interpretation of Raman spectra very simple and in many aspects analogous to laboratory techniques of qualitative and quantitative spectroscopic analysis based on the emission spectra. At the same time a Raman technique can be easily performed remotely that makes it very advantageous compared to standard techniques of a spectroscopic gas analysis. To illustrate this Fig. 1a shows the arrangement of lidar observations of the atmospheric emission from a stack.

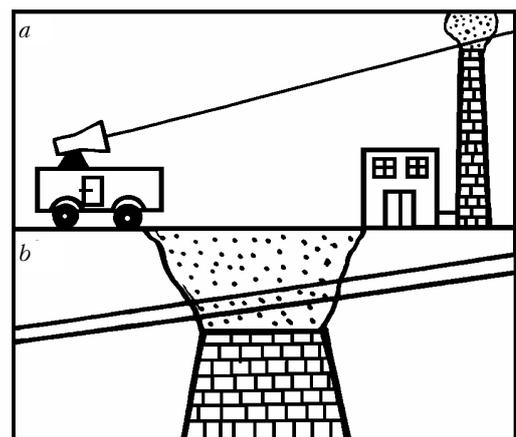


FIG. 1. Optical arrangement of measurements (a) and geometry of a beam interaction with an emission jet of a stack (b).

To begin the analysis of a Raman lidar technique let us first write down the so-called lidar equations² describing the power of Raman backscatter from different molecular

components of the atmosphere collected by the lidar receiving optics as a function of the lidar instrumental parameters and atmospheric parameters, including the profile of a molecular specie along the sounding path, as well

$$P_{N_2}(R) = P_0 \gamma(R) (A/R^2) \sigma_{\pi_{N_2}} n_{N_2}(R) (c\tau/2) T_{\lambda_0}(R) T_{\lambda_{N_2}}(R), \quad (1)$$

$$P_i(R) = P_0 \gamma(R) (A/R^2) \sigma_{\pi_i} n_i(R) (c\tau/2) T_{\lambda_0}(R) T_{\lambda_i}(R), \quad (2)$$

where P_0 is the power of a sounding radiation pulse delivered into the atmosphere from a lidar; $P_{N_2,i}$ is the optical power of a Raman lidar return from molecular nitrogen and i th molecular component of the gas mixture, respectively; R is the range; A is the area of the lidar optical receiver; $\sigma_{\pi_{N_2,i}}$ is the differential cross section of

Raman backscattering by a nitrogen molecule and by a molecule of the i th component of the gas mixture, respectively; $n_{N_2,i}$ is the number density of nitrogen and i th molecular component, respectively; c is the speed of light; τ is the duration of a sounding radiation pulse;

$T_{\lambda_0}(R) = \exp\left(-\int_0^R \alpha_{\lambda_0}(R') dR'\right)$ is the atmospheric transmission at the wavelength λ_0 of sounding radiation;

$T_{\lambda_{N_2},i}(R) = \exp\left(-\int_0^R \alpha_{\lambda_0, \lambda_{N_2}, \lambda_i}(R') dR'\right)$ is the atmospheric transmission at the wavelengths of the Raman lines of nitrogen λ_{N_2} and i th molecular component of the atmosphere λ_i , respectively; $\alpha_{\lambda_0, \lambda_{N_2}, \lambda_i}$ is the atmospheric

extinction coefficient at corresponding to the subscripts wavelengths; and, $\gamma(R)$ is the lidar geometric function describing the overlapping the beam of sounding radiation and viewing cone of the lidar receiver. Taking into account that nitrogen molecules are smoothly and continuously distributed over the atmosphere while the number density of the majority of gaseous pollutants has a pronounced peak above the background value only within a localized region of the atmospheric emission, for instance, from a plant stack one could anticipate, according to Eqs. (1) and (2), the Raman lidar returns from nitrogen and from an i th contaminating gas component of the emission to have the shapes schematically shown in Fig. 2.

As can be seen from Fig. 2 the Raman lidar return from molecular nitrogen is quite a smooth function of range at distances before the emission jet from a stack as well as behind it. A sharp decrease of the signal occurring within the emission jet is explained by a strong extinction of radiation by aerosol emitted into the atmosphere from the stack together with the exhaust gases of a technological process. Of course such a lidar return shape is only observable when no localized turbidity layers are along the sounding path except for the emission jet. The magnitude of such a signal fall off is determined by the values of the extinction coefficients α_{λ_0} and $\alpha_{\lambda_{N_2}}$ of the

emitted aerosol, which, by the way, depend on the aerosol mass density. In contrast to the Raman lidar return from nitrogen the corresponding return signal from a contaminating gaseous specie should have, in accordance with the above said, a pronounced peak above the noise level at the range corresponding to the distance between the lidar and the stack. It should be noted here that in the previous papers devoted to the Raman lidar technique of monitoring air pollutions in

stack plumes, including the above-cited ones, it was assumed as natural that the number density of nitrogen molecules $n_{N_2}(R_{st})$ within the emission jet from a stack is the same as in the atmosphere n_{N_2} outside it, i.e., $n_{N_2}(R_{st}) = n_{N_2}$. The latter value is very often and quite reasonably assumed to be independent of range, of course, for the particular case of sensing emissions from moderately tall stacks. Under this assumption and according to Eqs. (1) and (2) the ratio r_i of the signal $P_i(R_{st})$ to the signal $P_{N_2}(R_{st})$ is

$$r_i = \frac{P_i(R_{st})}{P_{N_2}(R_{st})} = \frac{\sigma_{\pi_i} n_i(R) T_{\lambda_i}(R_{st})}{\sigma_{\pi_{N_2}} n_{N_2}(R_{st}) T_{\lambda_{N_2}}(R_{st})}. \quad (3)$$

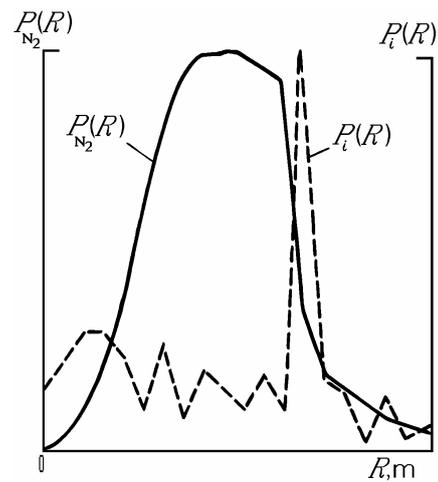


FIG. 2. An expected shape of the Raman lidar returns from molecular nitrogen ($P_{N_2}(R)$) and from a contaminating specie ($P_i(R)$) along a sounding path coming above a stack mouth.

A difference between the values of atmospheric transmission at the wavelengths λ_i and λ_{N_2} can be neglected at least for the case of the radiation extinction due to light scattering by large aerosol particles⁹ as is the case with the stack plumes under consideration. As a result the ratio r_i becomes proportional to the ratio of a sought-for number density of the i th component of the emission to a known value of the molecular nitrogen number density, with the proportionality factor $\sigma_{\pi_i}/\sigma_{\pi_{N_2}}$ being known from the literature for many gases (see, e.g., Ref. 1).

Thus the problem on remote determination of number densities of contaminating gases in the atmospheric emission from a stack is considered to be solved, since the number density of nitrogen can easily be calculated from independently measured atmospheric pressure and temperature using the ideal gas law. Unfortunately, just this point of the previously proposed Raman lidar technique of air pollution control in stack plumes is most doubtful.

In fact, exhaust gases of many industrial technological processes (like, for example, in the complete cycle of steel production) come to stacks being heated to the temperatures attaining 100°C and higher even at a stack mouth. Taking this into account one can readily arrive at a conclusion that the density of a gaseous jet from a stack should be essentially lower than that of ambient air. Thus, assuming that the temperature of an emission jet at a stack mouth is 100°C and the pressure inside the jet is equal to that of

ambient air based on the ideal gas law one easily finds that at temperature of ambient air $T_{\text{air}} = 0^\circ\text{C}$ the ratio of the density of the gas mixture in the jet to the ambient air density is about 0.73.

This is also valid for number density of molecular nitrogen even if pure but merely heated air is emitted from a stack. At a temperature of the emission jet of 150°C what is quite realistic this ratio will falls down to 0.65, other conditions being the same. This example allows one to come to a very important though quite obvious conclusion that in order to make use of a Raman lidar return from nitrogen molecules as an internal standard in a Raman lidar technique of gas analysis of the atmospheric emissions from stacks one should measure the number density of nitrogen in the emission jet itself.

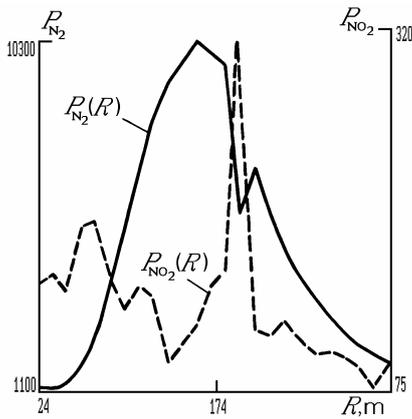


FIG. 3. Experimentally observed Raman lidar returns from nitrogen and NO_2 molecules along the path coming above the mouth of a stack of a coke-ovens battery.

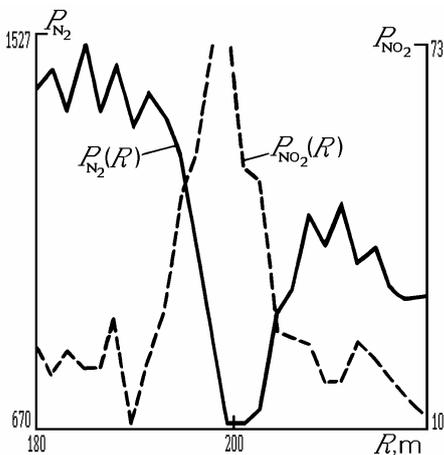


FIG. 4. A portion of signals depicted in Fig. 3 but recorded with a spatial resolution of 1.5 m.

Actually, if the above considerations are valid, the shape of a Raman lidar return from nitrogen measured along an atmospheric path coming just above a stack mouth should have a dip just in the range region within the emission jet from the stack. As a matter of fact, such a shape of Raman lidar returns from molecular nitrogen we have observed in our experiments on monitoring air pollutions emitted into the

atmosphere from a stack of a coke-ovens battery at the West Siberian Steel Production Plant in Novokuznetsk, Russia. A Raman lidar return from nitrogen recorded along a path coming through the emission jet of the stack of the coke-ovens battery is shown in Fig. 3 together with simultaneously recorded Raman lidar return from NO_2 molecules. It can be easily seen from this figure that a dip in the Raman lidar return from nitrogen occurs just at the same range where a peak of the return signal from NO_2 takes place. Return signals shown in Fig. 3 have been recorded with a spatial resolution of 12 m. A portion of these same signals recorded with a spatial resolution of 1.5 m is shown in Fig. 4. The shape and the depth of the dip in Raman signal from nitrogen as well as the drop of the Raman lidar return from atmospheric nitrogen caused by aerosol extinction of radiation are shown in this figure in more detail.

So it is obvious from the above said that Eq. (3) incorrectly describes the ratio of a Raman lidar return from a contaminating specie to a Raman lidar return from molecular nitrogen since the value n_{N_2} entering into its right side is overestimated. In order to correct this situation it is necessary to introduce functional factors into Eqs. (1) and (2) describing distribution of nitrogen and contaminating gases over the diameter of the cross section of the emission jet, i.e., along the sounding beam. Assuming that all the gaseous components and the aerosol of the emission are uniformly mixed over a stack cross section one can come to a conclusion that qualitatively the range behavior of the nitrogen number density and of the number density of the contaminating specie can be described by the functions whose graphs are shown in Fig. 5.

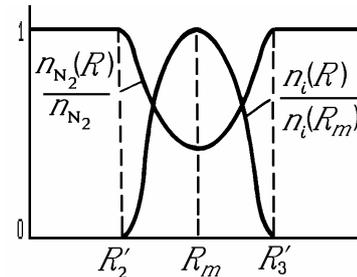


FIG. 5. A view of the nitrogen and a contaminating species distributions over the diameter of an emission jet cross section.

Let us assume for simplicity that the number densities of the contaminating molecular species in the atmosphere outside the emission jet can be neglected. Then the profile of the nitrogen number density along the sounding path can be represented in the general form by

$$n_{\text{N}_2}(R) = n_{\text{N}_2} (1 + B f(R)) \tag{4}$$

and the profile of a contaminating specie number density by

$$n_i(R) = n_{\text{N}_2} G_i f(R) , \tag{5}$$

where n_{N_2} is the number density of nitrogen molecules in the atmosphere outside the emission jet, which is assumed in the below discussion to be constant,

$$B = [(n_{\text{N}_2}(R_m)/n_{\text{N}_2}) - 1] < 0 ; G_i = n_i(R_m)/n_{\text{N}_2}$$

$$f(R) = \begin{cases} 0 & R < R'_2 \cup R \gg R'_3, \\ 1 & R = R_m, \\ f(R) & R \in [R'_2, R'_3]. \end{cases}$$

It should be noted here that usually Raman lidar measurements are performed using a photon counting technique that implies statistical averaging of signals and, as a result, of the sought-for concentrations of molecular species over certain time interval. It is quite natural in this connection to assume that the function $f(R)$ has single maximum at least in the cross section of an emission jet just above a stack mouth, as shown in Fig. 5.

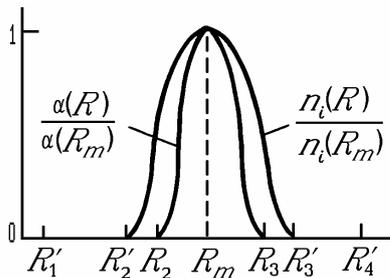


FIG. 6. A comparison of assumed distributions of a gas component and aerosols of the emission over the diameter of the emission cross section.

A bell-shaped curve seems to be most suitable for description of gas component distributions over the cross section of the emission jet because it accounts for natural process of the jet diffusion in the atmosphere just above the stack mouth in the most natural way.

By the way, since the diffusion of gases is obviously a more rapid process than the diffusion of aerosol particles because of their greater mass the aerosol emission jet should be somewhat narrower than the gaseous one in the cross section above the stack mouth, as is schematically shown in Fig. 6. This consideration will be taken into account below in derivation of equations relating the sought-for parameters of the emission jet to the values of the Raman lidar returns from its molecular components. Under these assumptions Eqs. (1) and (2) take the following form

$$P_{N_2}(R) = P_0 \gamma(R) (A/R^2) \sigma_{\pi_{N_2}} n_{N_2} (1 + B f(R)) \times (c\tau/2) T_{\lambda_{N_2}}(R_2) T_{\lambda_0}(R_2) \exp \left(-2 \int_{R_2}^R \alpha(R') dR' \right); \quad (6)$$

$$P_i(R) = P_0 \gamma(R) (A/R^2) \sigma_{\pi_i} n_{N_2} G_i f(R) (c\tau/2) \times T_{\lambda_i}(R_2) T_{\lambda_0}(R_2) \exp \left(-2 \int_{R_2}^R \alpha(R') dR' \right), \quad (7)$$

where $\alpha(R)$ is the extinction coefficient of aerosol in the emission jet, its values at the wavelengths λ_0 , λ_{N_2} , and λ_i being assumed equal what is quite justified for the case of large aerosol particles whose size essentially exceeds the wavelength of sounding radiation. Neglecting a difference

between $T_{\lambda_{N_2}}(R_2)$ and $T_{\lambda_i}(R_2)$ what can be justified for many practical cases because of short (< 1 km) distances from a lidar to a stack for the ratio $F_i(R) = p_i(R)/P_{N_2}(R)$ from Eqs. (6) and (7) one obtains

$$F_i(R) = \sigma_{\pi_i} G_i f(R) / \sigma_{\pi_{N_2}} (1 + B f(R)). \quad (8)$$

From this relation it follows that

$$f(R) = c_i F_i(R) / B(G_i B^{-1} - c_i F_i(R)), \quad (9)$$

where $c_i = \sigma_{\pi_{N_2}} / \sigma_{\pi_i}$. It is quite obvious from relation (9) that the first derivative of $f(R)$ with respect to R equals zero at the same point in which $F'_i = 0$. As a result one obtains for B the relation

$$B = G_i / c_i F_i(R_m) - 1, \quad (10)$$

where R_m is the range at which the functions $f(R)$ and $F(R)$ reach their maxima.

Using combinations of the magnitudes of the Raman lidar returns at ranges R'_2 and R_3 (see the figures)

$$S = R_3^2 P_{N_2}(R_3) / R_2^2 P_{N_2}(R'_2); \quad S_i = R_3^2 P_i(R_3) c_i / R_2^2 P_{N_2}(R'_2),$$

and

$$Q = P_3 R'_3 R'_4 / P_1 R'_1 R'_2 = \exp \left(-2 \int_{R_2}^{R_3} \alpha(R') dR' \right),$$

where

$$P_1 = \int_{R'_1}^{R'_2} P_{N_2}(R) dR; \quad P_3 = \int_{R'_3}^{R'_4} P_{N_2}(R) dR,$$

one obtains that

$$(B/G_i) = (S - Q) / S_i. \quad (11)$$

From this relation, formula (9), and taking into account that $f(R_m) = 1$ we have

$$G_i = (n_i(R_m) / n_{N_2}) = c_i F_i(R_m) S_i / (S_i - c_i F_i(R_m) (S - Q)); \quad (12)$$

$$B + 1 = (n_{N_2}(R_m) / n_{N_2}) = S_i / (S_i - c_i F_i(R_m) (S - Q)). \quad (13)$$

Thus we have derived the equations which relate the sought-for quantities to values that are measurable from Raman lidar returns, and the value c_i which is known for many contaminants from the literature (see, e.g., Ref. 1).

Since the ratio of the molecular nitrogen number density inside the emission jet to its value in the ambient air is also measurable in Raman lidar observations (see Eq. (13)) one can determine the velocity of the emission jet outflow from a stack. Actually, in the case of hot exhaust gases one can assume that the only force making

the gas mixture to ascend along a stack is the Archimedes force. Then one can easily derive the equation for the velocity v of the emission jet outflow from a stack at its mouth

$$v(R_m) = \sqrt{2gH(1 - (n_{N_2}(R_m)/n_{N_2}))}, \quad (14)$$

where g is the acceleration of gravity and H is the height of a stack.

Moreover, using the measured value of the ratio $n_{N_2}(R_m)/n_{N_2}$ and assuming the pressure inside the emission jet above the stack's mouth to be equal to the atmospheric pressure one can readily estimate the temperature of the emission T_e based on the ideal gas law as follows:

$$T_e(R_m) = T_{air} n_{N_2}/n_{N_2}(R_m) \quad (15)$$

provided that temperature of air outside the emission is measured independently.

Now, finishing the discussion of the above approach to the analysis of the Raman lidar technique for sensing the atmospheric emissions from plant stacks we should like to note that one should use the values $n_i(R_m)$ and $v(R_m)$ when evaluating the intensity of an atmospheric emission from a stack because just these axial values (determined by relations (12) and (14)) remain to be the least disturbed compared to their values inside the stack due to diffusion of the emission jet in the atmosphere. The same is valid with estimate of heat emission.

An example of thus estimated values of the emission jet outflow, temperature, aerosol extinction coefficient, and number densities of CO, NO, NO₂, and SO₂ in the emission from a stack of a coke-ovens battery of the above-mentioned steel production plant are presented in Table I.

TABLE I. Measurement data on the concentration of some gases and on the parameters of the emission jet from the coke ovens battery.

A component or a parameter	Concentration, volume, %	Standard error of measurements, %
CO	3	5
NO	3	5
NO ₂	0.19	10
SO ₂	0.55	10
Extinction coefficient, km ⁻¹	20.6	1
Velocity of the jet outflow, m/s	21.2	2.5
Temperature	462 K	±15 K

CONCLUSION

A detailed analysis of the Raman lidar technique of monitoring the atmospheric emissions from plant stacks presented in this paper allowed us not only to correct it but also to reveal, in addition, a possibility of determining from this same set of Raman lidar returns the velocity of the emission jet outflow from a stack. The latter circumstance is quite important for remotely estimating the intensity of atmospheric emissions from stacks. The above approach is applicable to sensing of hot emissions but even so it might be quite practical since hot technological processes are still widely used in industry and their wastes are continuously polluting the atmosphere and the environment as a whole.

An additional information about the emission temperature that can be obtained from Raman lidar returns could, in our opinion, be also useful as an information about state of a technological process and for assessing heat pollution of the atmosphere.

A possibility of simultaneously estimating the aerosol extinction coefficient of the emission along with other its parameters is also worth noting (see expression for the Q value), since it bears information about the aerosol mass concentration that could be extracted using a proper inversion method (see e.g., Ref. 10).

And, finally, we hope that this approach demonstrates good potentialities of the Raman lidar technique to monitor the composition and intensity of the atmospheric emissions from plant stacks.

The authors would like to acknowledge their colleagues D.I. Shefontyuk, A.I. Nadeev, and A.I. Materi from the laboratory of the High-Altitude Lidar Sensing of the Atmosphere at the Institute of Atmospheric Optics, Siberian Branch of the Russian Academy of Sciences for their valuable contributions to creation of the Raman lidar facility which we developed for conducting the experimental study discussed in this paper.

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