Analysis of Laser Measurement of High-Altitude Aircraft Emissions

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Accurate prediction of the environmental impact of stratospheric aircraft operation requires highly sensitive and specific measurement of effluents which have long residence times and possible catalytic interactions with the ambient atmosphere. Calculations have been performed to analyze three narrow bandwidth tunable laser measurement techniques for high altitudes where molecular absorption lines are narrow. Two ended absorption measurements using state-of-the-art tunable infrared diode lasers can provide sensitivities of $\simeq 0.02$–0.2 ppm over 10 m for O_3, CO, NO, H_2O, and SO_2 with minimal interference effects. Heterodyne radiometry of radiation from a hot near wake is capable of measuring initial wake concentrations, while heterodyne radiometry of absorption of solar radiation is sensitive to < 0.01 ppm of NO and CO in the far wake. These heterodyne measurements, with tunable diode laser local oscillators, will require development of milliwatt per mode diode lasers. Remote (1 km) resonance fluorescence measurements using dye lasers are capable of measuring to 0.15 ppm NO_2. Sensitivities can be improved to close to ambient levels during wake penetration.

Introduction

THE future of high-altitude aircraft fleet operations over the United States depends strongly on our ability to understand and predict the environmental impact of such operations. In 1971 Johnston[1] identified the catalytic reduction of ozone by oxides of nitrogen as a potential result of SST operations in the stratosphere. Such a reduction in ozone concentration could result in an increase in ultraviolet radiation at the Earth. A recent National Academy of Sciences report[2] indicates that the biological impact of an increase in ultraviolet radiation could be considerable.

Recent modeling using coupled wake diffusion and chemistry has indicated that the effect of oxides of nitrogen in reducing ambient ozone may be lessened by a lack of mixing due to slow diffusion of the wake effluents.[3] For the fraction of the effluent's residence time in the stratosphere that this model is correct, the NO in the inner portion of the wake is ineffective in removing ozone from the surrounding atmosphere. Reduced ozone within the wake may be an important and sensitive indication of the NO wake concentration, since this model predicts that as long as the wake diffusion is not complete the interior of the wake will be essentially free of ozone. Due to the catalytic nature of the interaction of effluents with the atmosphere combined with long stratospheric residence times, a program to validate this and other stratospheric models must be capable of both measuring the concentration of aircraft effluents in the near wake and monitoring the diffusion and chemistry of these effluents until their concentrations are reduced to ambient levels.

Wake concentrations at the exhaust are expected to be 90,000 ppm H_2O vapor, 500 to 5000 ppm CO, 50 to 200 ppm NO, and 1 to 10 ppm SO_2. Ambient levels at 20 km are of the order of 3 ppm H_2O vapor, 0.01 to 0.20 ppm CO, 0.0001 to 0.003 ppm NO, 0.0001 to 0.001 ppm SO_2, and $\simeq 2$ ppm O_3.

Several authors have discussed the use of the high spectral resolution offered by narrow bandwidth laser systems for sensitive pollutant detection.[4–9] At high altitudes molecular absorption line widths are reduced, and the use of lasers tunable to the center of absorption lines allows highly sensitive and specific detection of effluents and their effect on quantities such as the stratospheric ozone concentration. This paper presents the results of a study of techniques applicable to flight measurements of high-altitude effluents. The results of this study have indicated that three techniques have potential use in flight measurement programs in the near future. These are double-ended absorption measurements for local detection of wake constituents, heterodyne radiometry for remote detection using radiation from the near hot wake or absorption of solar radiation by the far wake, and laser excited resonance fluorescence for single-ended measurement of NO_2 in the wake.

Infrared Absorption Calculations

For an analysis of the two infrared measurement techniques, double-ended absorption and heterodyne radiometry, a detailed knowledge of the high spectral resolution atmospheric absorption and transmittance is required. For the discussions and results presented in this paper, the absorption coefficients and transmittances have been obtained using a standard infinite spectral resolution line-by-line absorption calculation technique. Effects of both pressure and Doppler broadening have been included through use of Lorentz and Voight profile[10] absorption line shapes, with the Voight profile taken when Doppler broadening effects are important. Published absorption line data for H_2O vapor,[11] CO,[12,13] NO,[14,15] and O_3[16] have been used along with the best available estimates of SO_2 line parameters. The effects of pressure and temperature variations on the absorption line intensities and half-widths have been accounted for by the relations[11]

$$S = S_o\left(\frac{T_o}{T}\right)^m \exp\left[-\frac{hcE''}{k}\left(\frac{1}{T}-\frac{1}{T_o}\right)\right] \tag{1}$$

and

$$\alpha = \alpha_o(p/p_o)(T_o/T)^n \tag{2}$$

where S is the line intensity at temperature T, E'' is the energy of the lower state of the transition, h is the Planck constant, c is the speed of light, k is the Boltzmann constant, α is the line half-width at half maximum for pressure p and temperature

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T, and the subscript o refers to a reference condition. The quantity m in Eq. (1) is equal to 1.0 for diatomic or linear polyatomic molecules and 1.5 for nonlinear polyatomic molecules. For n in Eq. (2) a value of 0.5, corresponding to an assumption of temperature-independent collision diameters, has been used for all cases, except for H_2O vapor where a value of 0.62 (Ref. 11) has been used. A 1966 U.S. 45°N July Standard Atmosphere has been assumed to provide the altitude variation of temperature, pressure, and H_2O vapor and O_3 concentrations.

In calculating the absorption coefficient for particular lines of H_2O vapor, SO_2 and O_3, which have many closely packed absorption lines, absorption due to the wings of overlapping lines has been included. In addition, the effects of interference from overlapping lines of gases other than the gas being monitored have been investigated. In the cases presented here, the interference effects have been found to be negligible.

Some of the analytical absorption coefficients used in this paper were verified by experimental results from high resolution diode laser studies under way at NASA Langley Research Center[5] and other labs.[9] A complete lab measurement of line shape vs temperature and pressure will be performed for all lines to be used in flight experiments.

Double-Ended Absorption Measurements

Laser absorption measurements are performed by propagating the laser through the pollutant either directly to a detector or to one or more reflectors and then to a detector. The fraction of the signal absorbed is a function of the path length through the pollutant, the density of the pollutant, and the absorption coefficient at the laser frequency. Figure 1 indicates the calculated variation of absorption coefficient with wave number as a function of altitude for a strong water vapor absorption line. The reduction of line width with altitude indicates that the chance of finding a coincidence between a fixed frequency laser line and an absorption line of a species of interest is much lower at high altitude than at ground level. However, tunable diode lasers are available which can be composition tuned to the frequency region of an effluent line and then current tuned through the line center. These lasers presently are available with powers of >0.1 mw per mode and are being developed into the mw per mode range.

The ratio of the signal received to laser power, s/P of an absorption measurement is the transmittance τ

$$\tau = s/P = \eta G\, e^{-\rho k_\nu l}\, e^{-jl} \tag{3}$$

where η is the system efficiency, G represents the geometric losses, l is the length through the pollutant, k_ν is the pollutant absorption coefficient, j is the attenuation coefficient due to scattering and absorption by interfering gases, and ρ is the pollutant density. For small changes in laser frequency, all factors expect k_ν can be considered constant. The ratio of the transmittance of two frequencies on and off line center is

$$\frac{\tau_{\text{on}}}{\tau_{\text{off}}} = \frac{s_{\text{on}}}{s_{\text{off}}} = \exp-(k_{\nu,\text{on}} - k_{\nu,\text{off}})\rho l \tag{4}$$

The minimum detectable density is then

$$\rho_{\text{MIN}} = -\frac{(\ln s_{\text{on}}/s_{\text{off}})_{\text{MIN}}}{(k_{\nu,\text{on}} - k_{\nu,\text{off}})l} \simeq \frac{(\Delta s/s)_{\text{MIN}}}{(k_{\nu,\text{on}} - k_{\nu,\text{off}})l} \tag{5}$$

where $(\Delta s/s)_{\text{MIN}}$ is the minimum fractional signal change measurable by the system. $(\Delta s/s)_{\text{MIN}}$ can be limited by laser instability, detector noise, atmospheric effects and vibration of optics. Laboratory experiments have demonstrated that diode laser instability is not a severe limitation. A $(\Delta s/s)_{\text{MIN}}$ of one in 10^5 was achieved for a 6 μw diode laser and a 6×10^{-11} w absorption in a C_2H_4 cell. In this case the limitation was probably due to detector noise which was of the order of the absorbed power.[9] Liquid nitrogen cooled detectors are commercially available which are capable of measuring a signal change of one part in 10^6 with a signal power of 5 μw.[7] With heterodyne detection the signal power can be reduced to $\simeq 10^{-10}$ w. For the purpose of calculations in this paper, the minimum signal change has been set at one in 10^3 in order to allow for noise due to vibration and atmospheric turbulence. This limitation is arbitrary, and current lab experiments and breadboard flight system development underway at our lab for evaluation of noise sources may allow more optimistic estimates. Path length has been set at 20 m for an aircraft penetrating the wake carrying a laser and detector in the cabin and a reflector on an external surface at 10 m distance. Since sensitivity increases with path length through the wake, a two-aircraft system scanning the wake would allow an increase in sensitivity. The two-aircraft system, however, would not be capable of making localized measurements and would add considerable operational difficulty. A multiple path reflection system on one aircraft could also be used to increase the effective path length, however, multiple reflections may also increase noise due to vibration and turbulence. The frequency separation between the on and off absorption line center measurements has been chosen as 10^9 Hz, or 0.03 cm^{-1}. This separation can be accomplished applying a few milliamps a.c. current to a diode laser operating at ~ 1 amp d.c. The detection system will measure either the signal change or its derivative as the laser frequency is varied.

Figure 2 is a block diagram of the breadboard flight system currently being developed. A reference gas cell and wavelength stabilizer control the diode d.c. current in order to lock ν_{on} to the center of the diode scan. A programable power supply controls the diode a.c. current level to optimize $\nu_{\text{on}} - \nu_{\text{off}}$ and sets the a.c. frequency to a quiet portion of the noise spectrum. The signal difference at $\nu_{\text{on}} - \nu_{\text{off}}$ is sensed at a synchronous

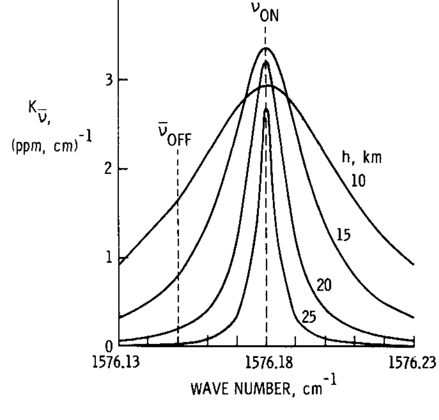

Fig. 1 Altitude variation of absorption coefficient vs wave number for a strong water vapor line.

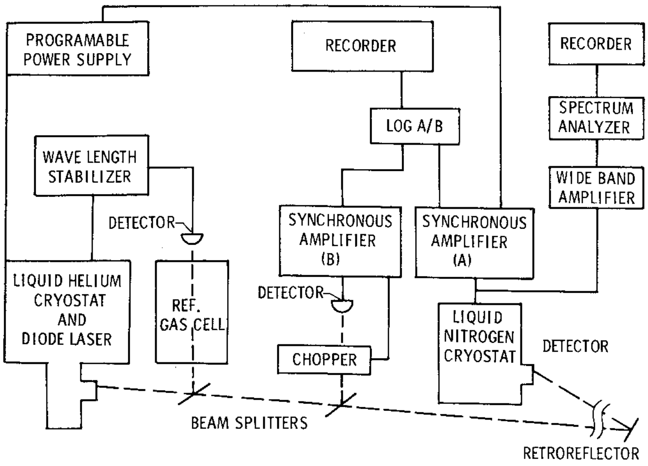

Fig. 2 Double-ended diode laser absorption system breadboard development.

Table 1 Double-ended absorption wave numbers and calculated 20 km absorption coefficients

	ν_{on} cm^{-1}	ν_{off} cm^{-1}	$k_{\nu,\text{on}}$ cm^2/g	$k_{\nu,\text{off}}$ cm^2/g
CO	2172.762	2172.732	6.43×10^5	0.14×10^5
H_2O	1576.180	1576.150	5.41×10^5	0.33×10^5
NO	1900.136	1900.166	1.29×10^5	0.03×10^5
SO_2	1373.019	1373.049	4.13×10^4	0.55×10^4
O_3	1050.872	1050.842	3.55×10^4	0.36×10^4

analyzer and normalized to any laser power difference between the two frequencies. A wide-band detector and spectrum analyzer measure noise at the detector.

Calculations have been made for detection of NO, H_2O, CO, O_3, and SO_2 at 10, 15, 20, and 25 km altitude. Table 1 lists the wave numbers used for detection of each molecule and the absorption coefficients at 20 km. Figures 3 and 4 present minimum detectable concentrations (X_{MIN}) and mass densities (ρ_{MIN}) for each molecule as a function of altitude. The indicated sensitivities are sufficient to measure significant changes in the ambient level of H_2O vapor and O_3 in the wake. CO sensitivity is of the order of the expected ambient levels, but NO and SO_2 sensitivities are not sufficient to measure to the ppb ambient levels. However, the NO sensitivity is sufficient for measurement of expected concentrations for wake dilutions up to the order of 5000 to 1 by the ambient atmosphere. Beyond this point the NO wake distribution can be implied by the concentrations of CO and H_2O or by the reduction of O_3. Low SO_2 sensitivity combined with the expected low wake concentrations will likely limit SO_2 measurements to the near wake region.

The double-ended absorption technique is capable of real-time measurement of wake constituents. The system thus has an additional capability of providing an indication of wake penetration. This can be used both to aid in keeping the chase aircraft in the wake and to selectively control intakes of atmospheric samples to instruments within the aircraft.

Heterodyne Detection

Heterodyne detectors have been used in the infrared for laser communications receivers[17] and for solar measurements.[18] They have also been considered for use in pollution measurements.[5,6,9] The heterodyne radiometer operates by mixing a laser local oscillator with a radiation signal at a high-frequency detector. The detector responds to the square of the electric field

$$R \propto [E_{LO} \cos(2\pi\nu_{LO} t) + E_S \cos(2\pi\nu_S t)]^2 \tag{6}$$

R is the detector response, E_{LO} and E_S are the field strengths of the local oscillator and source, ν_{LO} and ν_S are their frequencies, and t is time. The cross term in this equation results in the detector output containing signal spectral information transformed from the infrared to the radio frequency range at a difference frequency $\nu_D = |\nu_S - \nu_{LO}|$.

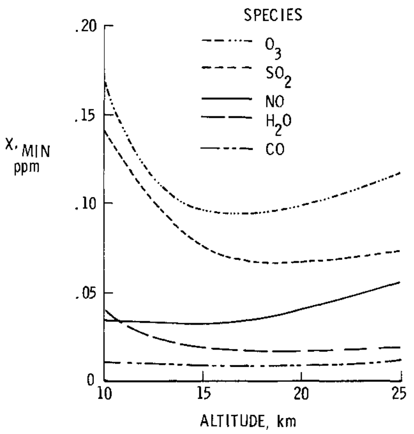

Fig. 3 Diode laser absorption, minimum detectable concentration vs altitude ($l = 20$ m, $\Delta s/s = 0.001$).

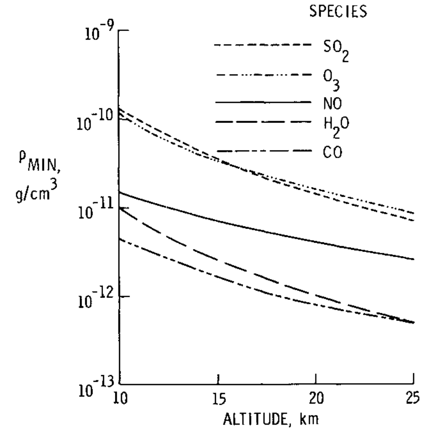

Fig. 4 Diode laser absorption, minimum detectable density vs altitude ($l = 20$ m, $\Delta s/s = 0.001$).

The output of the detector at a particular difference frequency will be proportional to the energy in the signal at frequency ν_S multiplied by the energy of the local oscillator at ν_{LO}. The detector, however, cannot distinguish the sign of the difference; thus, for a difference frequency ν_D and an analyzer integration bandwidth β, the energy measured by the system will be from source frequencies in the ranges

$$\left(\nu_{LO} - \nu_D - \frac{\beta}{2}\right) < \nu_S < \left(\nu_{LO} - \nu_D + \frac{\beta}{2}\right)$$

$$\left(\nu_{LO} + \nu_D - \frac{\beta}{2}\right) < \nu_S < \left(\nu_{LO} + \nu_D + \frac{\beta}{2}\right)$$

Using a tunable laser as a local oscillator ν_D can be set slightly greater than $\beta/2$, and the local oscillator can be set at line center. If a fixed frequency laser is used as a local oscillator, ν_D must be set at the difference between the laser frequency and the pollutant frequency, and energy radiated or absorbed at the mirror image of the difference frequency may interfere with the measurement. The maximum value of ν_D is limited to $\simeq 10^9$ Hz by detector frequency response.

Heterodyne detection requires phase coherence in the local oscillator and the source signals. For a thermal source, the area that appears coherent to the detector is

$$A_{\text{coh}} = r^2\lambda^2/A_{\text{det}} \tag{7}$$

where r is the distance and λ the wavelength, and A_{coh} and A_{det} are the coherence and detector areas. The coherent power received from a source that radiates a power P_{out} per steradian per unit area is P_{rec}

$$P_{\text{rec}} = P_{\text{out}} A_{\text{coh}}(A_{\text{det}}/r^2) = P_{\text{out}}\lambda^2 \tag{8}$$

The coherent power received is independent of distance and detector area so long as the coherent field of view of the detector is smaller than the source. The signal-to-noise ratio of a heterodyne radiometer with a detector efficiency η, an integration time τ, a noise power P_{noise} and a thermal induced current i_{thermal} is[18]

$$SNR = \frac{\eta P_{\text{rec}}(\beta\tau)^{1/2}}{2h\nu\beta\left(1 + \dfrac{P_{\text{noise}}}{P_{LO}} + \dfrac{i^2_{\text{thermal}}}{i^2_{LO}}\right)} \tag{9}$$

With sufficient local oscillator power the noise power and thermal currents become negligible compared to the local oscillator terms, and the detector operates at a quantum limited signal-to-noise ratio. The local oscillator power required to overcome the noise is of the order of 1 to 10 mw. Diode laser development is currently moving rapidly toward higher powers, and diode laser local oscillators with these powers should be available within 1 to 2 yr. Approximate coincidences ($\Delta\nu < 10^9$ Hz) between fixed

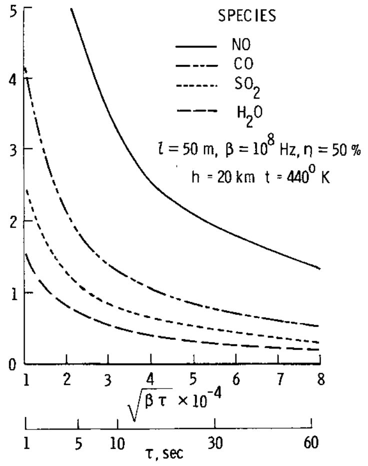

Fig. 5 Heterodyne radiometer, minimum detectable concentration vs $(\beta\tau)^{1/2}$, 20 km 440°K wake ($l = 50$ m, $\beta = 10^8$ Hz, $\eta = 50\%$).

frequency lasers and absorption lines may allow earlier demonstration of heterodyne detection for some gases.[6]

For a thermal source the heterodyne signal-to-noise ratio for wake measurements is[9]

$$SNR = \eta \frac{(1 - e^{-k_\nu \rho l})}{2} \left[\frac{1}{e^{h\nu/kT_w} - 1} - \frac{\varepsilon_B}{e^{h\nu/kT_B} - 1} \right] (\beta\tau)^{1/2} \quad (10)$$

where η is the detector efficiency, T_w is the wake temperature, ε_B is the background emissivity, T_B is the background temperature, ν is the measured frequency, and k_ν is the absorption coefficient at ν. The term containing T_w is due to radiation from the wake, while the term containing T_B is due to absorption by the wake of background radiation. It is assumed in this equation that the local oscillator laser power is sufficient to overcome the system noise and that there is no emission or absorption outside the wake by the effluent being measured.

Since the output of the heterodyne radiometer is proportional to the product of the local oscillator and signal powers, instability of the local oscillator can adversely affect the signal-to-noise ratio. However, this effect can be minimized by normalizing the radiometer output to the local oscillator power. The effect should then be no worse than for diode laser absorption measurements where signal changes of one part in 10^5 have been measured.[9]

Two cases are considered: the heterodyne radiometer measuring the emission from a hot near wake and the radiometer measuring the absorption of solar thermal radiation by a far wake cooled to ambient temperatures. For the hot wake a temperature of 440°K was chosen. The actual temperature of the wake will strongly affect the sensitivity, and line shape measurements may be required to determine the temperature at the point being measured. A wake diameter of 50 m was assumed for these calculations. Absorption coefficients were calculated at 440°K for a 20-km altitude. The bandwidth used for the calculations was 10^8 Hz, or 3.3×10^{-3} cm^{-1}, and the absorption coefficient was integrated over this bandwidth. Integration times of from 1 to 60 sec were used; calculations were performed for minimum detectable concentrations using a signal-to-noise ratio of 1 and a 50% efficient detector. The assumption that there is no emission or absorption by the ambient effluents outside the wake may not be valid. Although both the temperature and concentration in the wake are higher than the ambient levels, the detector will receive energy from ambient molecules along its entire line of sight through the atmosphere. A scanning optical system to measure the signal change as the optical path is scanned through the wake may be needed to remove atmospheric effects from the data. Figures 5 and 6 present results for NO, CO, H_2O vapor, and SO_2. The sensitivities calculated are sufficient for measurements of initial effluent concentrations in the near wake. An attractive mode of operation for near wake measurements would be to mount a rearward viewing radiometer on the aircraft producing the wake.

Table 2 Predicted minimum detectable concentrations and densities for solar absorption 100 m wake

	1 sec		10 sec	
	X_{MIN} ppm	ρ_{MIN} g/cm^3	X_{MIN} ppm	ρ_{MIN} g/cm^3
CO	0.006	4.8×10^{-13}	0.002[a]	1.6×10^{-13}[a]
H_2O	2.3	1.3×10^{-10}	2.1[a]	1.2×10^{-10}[a]
NO	0.008[a]	8.0×10^{-13}[a]		
SO_2	0.013[a]	2.8×10^{-12}[a]		
O_3	2.7[a]	4.3×10^{-10}[a]		

[a] Indicates $\rho kl = 0.001$.

The 5000°K sun offers an excellent source of absorption measurements. Solar absorption has the disadvantage of measuring the total burden of a species between the radiometer and the sun. The optical path must again be scanned in and out of the wake to distinguish between the atmospheric burden and the wake contribution. In the case of reduced ozone in the wake a decrease in absorption through the wake is expected. For the strong lines of ozone and water vapor there is a marked atmospheric attenuation, and it is therefore necessary to choose weaker lines for the solar absorption measurement. This, of course, lowers the attenuation through the wake. Calculations have been performed using weak lines of H_2O vapor and O_3 at 1833.28 cm^{-1} and 1002.011 cm^{-1}, respectively, and the previously selected strong lines for NO, CO, and SO_2. For H_2O vapor and O_3, the atmospheric weak line transmission above 20 km was calculated and substituted for ε_B in Eq. (10). The transmission was arbitrarily set at 0.1 for CO and at 0.5 for NO and SO_2. The bandwidth was again set at 10^8 Hz. Table 2 presents the minimum detectable concentrations and mass densities for a signal-to-noise ratio of 1 and a 50% efficient detector for 1- and 10-sec integration times. The a-noted values have been limited arbitrarily to a minimum one part in 10^3 signal change through a 100-m wake to account for atmospheric variations and turbulence. H_2O vapor and O_3 sensitivities for this technique are limited by the weak lines used but are of the order of the ambient concentrations. CO can be measured to ambient levels and NO to $\simeq 4$ times ambient levels. SO_2 sensitivity is limited by absorption line strength but may be sufficient in the near wake or if anomalously high concentrations of SO_2 are present in the far wake.

Resonance Fluorescence

NO_2 displays fluorescence when exposed to visible radiation. Thus, measurement of this important molecule in catalytic ozone reduction is possible using a remote lidar technique.[19]

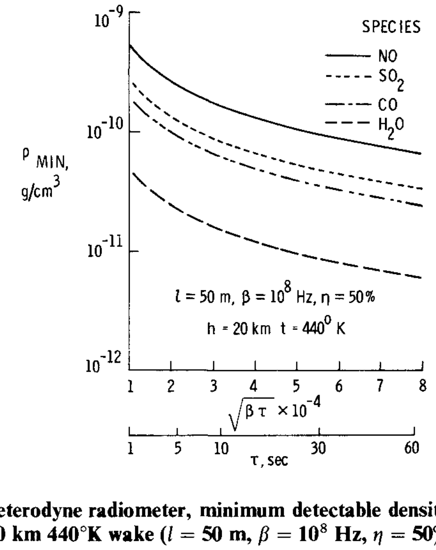

Fig. 6 Heterodyne radiometer, minimum detectable density vs $(\beta\tau)^{1/2}$ 20 km 440°K wake ($l = 50$ m, $\beta = 10^8$ Hz, $\eta = 50\%$).

Since NO_2 is dissociated by sunlight, measurements of wake NO_2 must be performed at night. Night-time operation, however, does have the advantage of avoiding background signal due to scattered sunlight. The resonance fluorescence will be performed using a dye laser and telescope receiver on an aircraft near the wake. The signal received is

$$s = \underset{1}{\frac{E}{h\nu} t_1 \rho k_\nu l} \quad \underbrace{q(\Delta\nu)\frac{1}{1+\tau_f/\tau_q}}_{2} \quad \underbrace{\frac{t_2 A\eta}{4\pi r^2}}_{3} \tag{11}$$

$E/h\nu$ is the number of photons in the laser pulse, t_1 and t_2 are the atmospheric transmissions at the laser and fluorescence frequencies, ρ is the NO_2 density, k_ν is the absorption coefficient, l is the resolution length, $q(\Delta\nu)$ is the partition function, τ_q and τ_f are the quenching and fluorescence lifetimes, A is the telescope area, η is the efficiency, and r is the distance.

Term one is the number of photons absorbed per resolution length; term two is the fraction of this number which results in fluorescent photons in the frequency range received; and term three is the fraction of these fluorescent photons that reach the telescope and are measured. The factor $1/[1+(\tau_f/\tau_q)]$ is the loss due to atmospheric quenching. The absorption coefficient k_ν is not a strong function of pressure or temperature in the visible portion of the spectrum. For NO_2 the quenching lifetime at ground level is 1.85×10^{-10} sec[19] and increases with the inverse of the pressure. Thus the minimum detectable density decreases with the pressure as altitude increases; the sensitivity per ppm remains approximately constant with increasing altitude. The measured value of fluorescence lifetime for NO_2 is 75 μsec and is much longer than calculated values. Recent experiments indicate that 0.5 μsec may be achieved with excitation using narrow line width ($\simeq 0.01$ Å) lasers tuned to particular absorption features.[20] Currently, special dye lasers and measurement equipment are being developed at our laboratory to test this concept.

Calculations were performed for one pulse of a 1-joule dye laser operating at 4550 Å using a 1-km distance, a 100-m resolution depth, a $q(\Delta\nu)$ of 0.1, a 25-cm-diam telescope, and an efficiency of 10%. The absorption coefficient is 1.12×10^{-5} cm^{-1} ppm^{-1} (5450 cm^2/gm).[19] Minimum detectable concentrations are based on a received signal which generates four photoelectrons per resolution depth. The minimum detectable concentration is 0.15 ppm (1.8×10^{-11} g/cm^3 at 20 km). This value is not sufficient for far-wake measurements. An increase in sensitivity to the order of the near ambient levels in the far wake can be achieved by reducing the range of the measurement from 1 km to 100 m, thus making the measurement during wake penetration. Remote fluorescence measurement of the far wake will require a large increase in sensitivity. This can be provided if "fast" fluorescence, which is under laboratory investigation, can be achieved, or by the development of high energy, high pulse repetition rate flight-qualified lasers.

Conclusions

The accurate prediction of the environmental impact of high-altitude aircraft effluents requires high sensitivity wake constituent measurements. Three laser techniques have been identified with potential for performing these measurements. Double-ended laser absorption using a single aircraft penetrating the wake is capable of measuring H_2O vapor, CO, and O_3 to ambient levels and of measuring NO far into the wake. This technique can be accomplished using state-of-the art technology and early flight tests are possible.

Heterodyne radiometry allows measurement without penetrating the wake. Sensitivities using hot near wake radiation are sufficient to measure initial wake constituents. Heterodyne measurement of the absorption of solar radiation by a 100-m wake has reduced sensitivity compared to 20 m path length double-ended absorption for constituents such as O_3 and H_2O vapor with large atmospheric burdens outside of the wake but has an increased sensitivity for the minor constituents CO, NO, and SO_2. Heterodyne radiometry may be initially tested for some gases using fixed frequency lasers where approximate coincidences with absorption lines exist. The further development of diode lasers to the milliwatt per mode range will allow the selection of lines with the highest sensitivity and minimum atmospheric interference.

Resonant fluorescence currently is capable of measuring NO_2 at 1-km range in the near wake and during penetration of the far wake. Remote far wake measurements will require lab demonstration of "fast" fluorescence, or the development of high energy, high pulse rate flight-qualified lasers.

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