

Remote photometry of the atmosphere using microwave breakdown

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Abstract. A novel method for remote optical diagnostics of the atmosphere at heights 30–60 km is proposed. The method relies on exciting atoms and molecules of minority species by electron impact during and following an ionizing microwave pulse injected from a focused ground-based transmitter. Free electrons produced in the breakdown region are the exciting agents for the atmospheric target molecules. The mixing ratio of the minority species can then be measured by either detecting the direct emission from allowed transitions or by utilizing lidar techniques to measure the excitation level of metastable states. Computer simulations of the intensity of the expected emission, based on kinetic theory of air breakdown, are presented. It is shown that mixing ratios below particle per trillion can be detected using microwave heaters with state of the art effective radiation power and modern detection technology.

1. Introduction

Accurate, simultaneous, and continuous measurements of the atmospheric mixing ratios of active species are critical in determining quantitative links of increased greenhouse gases and climate change. Despite significant effort the database on which predictions and assessments of climate change are based is still insufficient. There are, in general, two types of data acquisition techniques: in situ and remote. The in situ measurements rely on rockets and stratospheric balloons. These are single-shot measurements that cannot be applied to study systematically the temporal dynamics of the concentration of the relevant species. Furthermore, they are hindered by altitude limitations and local contamination problems. The remote techniques rely on ground- or space-based lidars. Despite significant progress, propagation issues, signal to noise ratio limitations, and the high cost and calibration problems of space-based systems limit the extent of passive spectroscopy and lidar-based techniques.

The objective of this paper is to propose a novel concept that allows for continuous monitoring of the strato-

sphere at altitudes of 25–60 km. The concept utilizes ground-based assets, although it can be complemented by space-based platforms and satellites. It is unique in that it can provide simultaneous determination of the relevant atmospheric species over long periods of time. The technique is illustrated in Figure 1. It relies on atmospheric breakdown [Borisov *et al.*, 1986; Tsang *et al.*, 1991] by using a ground-based, microwave source, whose pulse is focused at the desired altitude. The breakdown process creates a weakly ($< 10^{-7}$) ionized plasma and is thus nonperturbing. The relevant mixing ratios of the important species can be measured by passive or active spectroscopy, since the plasma electrons created during breakdown excite a mini-aurora display. The technique is unique since it allows continuous and simultaneous determination of the relevant species. Mixing ratio profiles can be obtained by scanning the breakdown region in altitude. By synchronizing the detection with the ionizing pulse, extraneous noise sources can be eliminated. Observations of the airglow can provide information on wind and turbulence.

The main goal of the paper is to present the concept, indicate the technological requirements, and demonstrate its potential. Detailed engineering designs are beyond the scope of the presented article. The paper is organized in two parts. The first part deals with a brief description of atmospheric breakdown, the resulting atomic and molecular excitation, and the associated emissions. The analysis is performed by using a Fokker-Planck code since the non-Maxwellian features of the electron distribution function are important. The results of this part are a key input in deriving the required characteristics of the microwave

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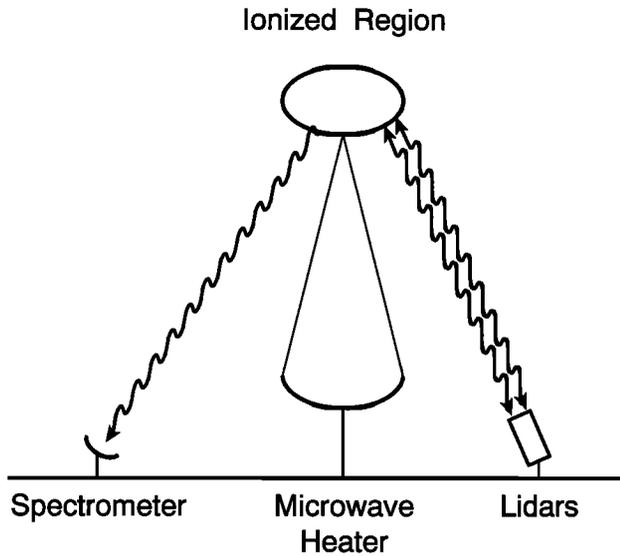


Figure 1. Schematic of the proposed concept.

source. The second part presents a strawman diagnostic facility and discusses the techniques for measuring the relevant mixing ratios. Consistent with the objectives of the paper the analysis is based on two illustrative examples. The paper concludes with a summary and suggestions for future work and implementation.

2. Atmospheric Breakdown and Airglow Production

A microwave pulse focused at an altitude z with power density above the breakdown threshold generates an electron density $n(t)$ which is given by

$$n(t) = n(t=0) \exp[\nu_i t] \quad (1)$$

where ν_i is the ionization rate. The ionization rate ν_i is a function of the microwave frequency ω , the ambient neutral density $N(z)$, and the incident power density P [Short *et al.*, 1990; Tsang *et al.*, 1991]. The ionization process is accompanied by excitation of atoms and molecules by electron impact. The concentration N_{js} of an excited state j of species s is given by

$$\frac{dN_{js}}{dt} = k_{js}n(t)N_s - \frac{N_{js}}{\tau_{js}} + \sum_k \frac{N_{ks \rightarrow js}}{\tau_{ks \rightarrow js}} \quad (2)$$

where k_{js} is the excitation rate of the corresponding atom or molecule whose density is N_s , and τ_{js} is the effective lifetime of the excited electronic state. The last term in the right-hand side of equation (2) represents cascade from higher states. The coefficient of k_{js} is given by

$$k_{js} = 4\pi \int_0^{\infty} v^3 \sigma_{js}(v) f(v, P, \omega) dv \quad (3)$$

where σ_{js} presents the corresponding cross section and $f(v, P, \omega)$ is the stationary electron distribution function as a function of the microwave frequency and incident power density. From the above equations we find that the temporal evolution of N_{js} during the microwave pulse is given by

$$N_{js}(t, z) = \frac{k_{js}N_s(z) n(t=0)}{(1 + \nu_i \tau'_{js})} \tau'_{js} (e^{\nu_i t} - e^{-t/\tau'_{js}}) \quad (4)$$

$$\tau'_{js} = \tau_{js} \left(1 - \sum_k \frac{\tau_{js}}{\tau_{ks \rightarrow js}} \frac{N_{ks \rightarrow js}}{N_{js}} \right)^{-1}$$

For allowed states, with lifetimes much shorter than the microwave pulse length τ , the number of photons n_{js} , emitted per unit volume, per pulse will be given by

$$n_{js}(z) = \frac{1}{\tau_{js}} \int_0^{\tau} N_{js}(t, z) dt = \frac{k_{js}N_s(z)n(t=\tau)}{\nu_i} \frac{\tau'_{js}}{\tau_{js}} \quad (5)$$

with

$$\nu_i = 4\pi N \int_0^{\infty} v^3 \sigma_i(v) f(v, P, \omega) dv \quad (6)$$

where σ_i is the ionization cross section. For the species of interest (Table 1) we considered excitation of high electronic levels, so that allowed cascade transitions from higher states are not available; while the forbidden transitions were selected so that they have long lifetime ($\tau_{ks \rightarrow js} \gg \tau_{js} \simeq \tau'_{js}$). It is convenient to write equation (5) as

$$n_{js}(z) = \lambda_{js} g_s n(\tau) \quad (7)$$

with λ_{js} given by

$$\lambda_{js} = \frac{\int_0^{\infty} v^3 \sigma_{js} f(v) dv}{\int_0^{\infty} v^3 \sigma_i f(v) dv} \quad (8)$$

and the mixing ratio g_s of the species s by

$$g_s \equiv \frac{N_s}{N} \quad (9)$$

$n(\tau)$ is the total electron density created by the pulse. On the basis of equation (7), assuming that over the ionizing volume V the conditions are uniform and neglecting absorption and scattering of visible light photons, the number of emitted photons P_{js} associated with the j state of species s , received by a telescope of area S located at a distance R from the breakdown, will be given by

$$P_{js}(\tau) = \frac{\lambda_{js} g_s (n(\tau) V) S}{4\pi R^2} = (\lambda_{js} g_s) N_e \frac{S}{4\pi R^2} \quad (10)$$

Table. Emissions of Some Minor Atmospheric Species Stimulated by Electron Impact.

Species	Transition Excited by Electron Impact	Excitation Energy, eV	Lifetime, ns	Stimulated	Transition
				Designation	Wavelength, nm λ (Atoms) λ_{∞} (Molecules)
Cl	$3^3P_{3/2} \rightarrow 5^2D_{3/2}$	11.80	22	$5^2D_{3/2} \rightarrow 4^2D_{5/2}$	808.67
H	$1^2S_{1/2} \rightarrow 3^2P_{1/2,3/2}$	12.09	5.3	$3^2P_{1/2,3/2} \rightarrow 2^2S_{1/2}$	656.2
N	$2^4S_{3/2} \rightarrow 4^4F_{3/2-9/2}$	12.98	22	$4^4F_{3/2-9/2} \rightarrow 3^4D_{1/2-7/2}$	1011.4
O	$2^3P_2 \rightarrow 3^3P_{0,1,2}$	10.99	36	$3^3P_{0,1,2} \rightarrow 3^3S_1$	844.6
F	$2^2P_{3/2} \rightarrow 3^2S_{1/2}$	14.68	26	$3^2S_{1/2} \rightarrow 3^2P_{3/2}$	731.1
	$2^2P_{3/2} \rightarrow 3^2D_{3/2,5/2}$	14.59	47	$3^2D_{3/2} \rightarrow 3^2P_{1/2}$	780.02
			128	$3^2D_{5/2} \rightarrow 3^2P_{3/2}$	755.47
ClO	$X^2\Pi_i \rightarrow A^2\Pi_i$	3.99	n.a.	A \rightarrow X	321
		3.92			323
CO	$X^1\Sigma^+ \rightarrow B^1\Sigma^+$ $X^1\Sigma^+ \rightarrow C^1\Sigma^+$	10.78	24	B \rightarrow A	458
		11.40	2	C \rightarrow A	374
NO	$X^2\Pi_r \rightarrow B'^2\Delta_i$ $X^2\Pi_r \rightarrow F^2\Delta$ $X^2\Pi_r \rightarrow C^2\Pi_r$	7.48	75	B' \rightarrow B	700.5
		9.66	90	F \rightarrow C	1,051
		6.46	25	C \rightarrow A	1,243
OH	$X^2\Pi_i \rightarrow A^2\Sigma^+$	4.02	690	A \rightarrow X	310

In equation (10), $N_e = n(\tau)V$ is the total number of electrons created by the microwave pulse. Notice that the factor ($\lambda_{js}g_s$) corresponds to the number of photons per ionization. Equation (10) can be used to determine g_s if the value of λ_{js} as a function of the heater parameters and the ambient neutral density is known.

The physics of ionospheric breakdown and the rate and efficiency with which ionization occurs as a function of the incident power density and frequency has been discussed in two recent papers [Short et al., 1990; Tsang et al., 1991]. We briefly review some results pertinent to our problem and refer the interested reader to the original publications. We start by noting that for high-frequency ionization, in the sense of $\omega \gg \nu_m$, where ν_m is the maximum elastic collision frequency of the electrons with the neutrals given by

$$\nu_m = 1.5 \times 10^{-7} N s^{-1}, \tag{11}$$

the electron distribution $f(v)$ is independent of the ambient neutral density and depends only on the value of

$$\tilde{\epsilon} = \frac{1}{2} m \frac{e^2 E_0^2}{m^2 \omega^2} \tag{12}$$

where E_0 is the amplitude of the electric field and $\tilde{\epsilon}$ is nothing more than the quiver energy of the electrons in the microwave field. It has been shown that $f(v)$ is self-similar

with respect to $\tilde{\epsilon}$ [Gurevich, 1978]. Therefore λ_{js} depends only on $\tilde{\epsilon}$, and σ_{js}/σ_0 .

As an example of the proposed diagnostics, we have used a Fokker-Planck code [Tsang et al., 1991] to determine λ_{js} as a function of $\tilde{\epsilon}$ for three important atmospheric species Cl, H, and CO. The cross sections for the excitation of Cl and H atoms by electron impact were obtained using the generalized Born approximation [Sobelman et al., 1981]. The cross section for the excitation of the CO($B^1\Sigma^+$) electronic state was adopted from Chung and Lin [1974], and the branching ratio for the B \rightarrow A transition was taken as 0.32 [Dotchin et al., 1973]. The results of the calculation of λ_{js} are shown in Figure 2. We have also used the Fokker-Planck code to determine the value of the elastic electron neutral collision frequency $\nu(\tilde{\epsilon})$ as a function of $\tilde{\epsilon}$ for use in later calculations. This is shown in Figure 3.

It is convenient to express the photon flux P_{js} at the detector as a function of the total energy E in the microwave pulse. This can be achieved by noting that for air the energy ϵ_i per ionization pair is a function of $\tilde{\epsilon}$ only. It is given by Figure 4d of Tsang et al. [1991]. In determining the value of ϵ_i in this reference, an initial value of $n(t=0)$ was required which was taken a 1 el/cm³. However, as was noted there, under conditions of above threshold irradiation the effect of $n(t=0)$ on ϵ_i was logarithmically small, and to zero order the value of ϵ_i is independent of $n(t=0)$.

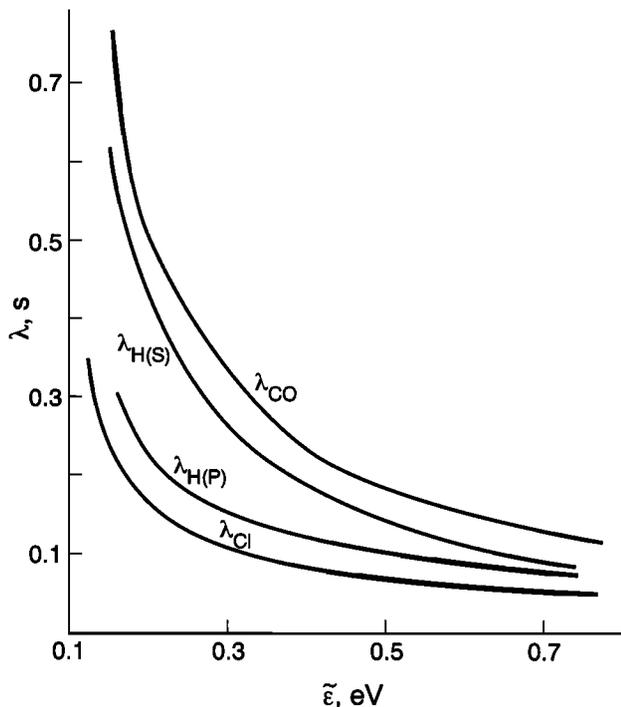


Figure 2. The factor $\lambda_{js} = k_{js}/k_i^{\text{air}}$ for three different species. From top to bottom are the λ factors for the excitation of $\text{CO}(B^1\Sigma^+)$, $\text{H}(2^2S_{1/2})$ (reduced 50 times), $\text{H}(3^2P_{1/2,3/2}^0)$ (reduced 10 times), and $\text{Cl}(5^2D_{3/2})$.

Furthermore, modeling and simple estimates indicate that focused microwaves above the breakdown threshold with the exception of the front of the pulse are totally absorbed [Milikh *et al.*, 1993]. Under these conditions the total number of electrons produced during the breakdown is $N_e = E/\varepsilon_i$. Using practical units, equation (10) for the number of photons per square meter on the ground becomes

$$P_{js} = 5.5 \left(\frac{E}{1 \text{ MJ}} \right) \left(\frac{30 \text{ km}}{R} \right)^2 \left(\frac{S}{\text{m}^2} \right) \left(\frac{g_s}{10^{-12}} \right) \left(\frac{100 \text{ eV}}{\varepsilon_i} \right) \lambda_{js} \quad (13)$$

The energy per photon can be minimized by noting that the function $\lambda_{js}(\tilde{\varepsilon}) \times 100 \text{ eV}/\varepsilon_i(\tilde{\varepsilon})$ reaches its maximum at a quiver energy of $\tilde{\varepsilon} \simeq 0.4 \text{ eV}$. This is seen from Figure 4, which shows the function $\lambda_{js}(\tilde{\varepsilon}) \times 100 \text{ eV}/\varepsilon_i(\tilde{\varepsilon})$ for some species along with the value of $\varepsilon_i(\tilde{\varepsilon})/100 \text{ eV}$ taken from Tsang *et al.*, [1991], as a function of $\tilde{\varepsilon}$. These results are used below as examples of chlorine and hydrogen detection using two different techniques.

3. A Strawman Facility

The potential and the technological requirements of a breakdown facility for atmospheric diagnostics can be best illustrated by referring to a concrete example. The particular example refers to diagnosing an altitude range 30–50 km. In selecting the microwave frequency, we

note that $\omega \gtrsim \nu_m$ can be satisfied with a microwave frequency of 10 GHz. Figure 5 shows the power density as a function of altitude from a microwave facility with power 3 GW focused by a 34-m dish (J.T. Cha and P. Koert, private communication, 1993). In this case the quiver energy reaches 0.3 eV in a focal spot located at 35 km, which is close to the efficiency peak of the discussed diagnostics (see Figure 4). The required pulse length τ was determined from the conditions that τ be comparable to the lifetime τ_{js} of the excited species and exceeds the mean ionization time ($\tau \gg \nu_i^{-1}(\tilde{\varepsilon})$) to result in effective microwave absorption. For the altitude range of interest it corresponds to $\tau \simeq 100 \text{ ns}$, giving 300–400 J/pulse.

Two different techniques for determining the relevant mixing ratios are considered. The first utilizes passive ground-based photometry and applies to species having allowed transitions in the atmospheric window. A list of such species is shown in Table 1, with the main optical parameters taken from Huber and Herzberg [1979] and Radzig and Smirnov [1985]. The second utilizes active lidar spectroscopy and is applicable to species having metastable states excited by electron impact. Relevant molecules are listed in Table 2 with the corresponding optical parameters taken from Radzig and Smirnov [1985].

3.1. Detection of Chlorine Concentration by Passive Photometry

One of the most important active species is Cl. Its mixing ratio is so low that only a few sporadic daytime measurements were successful [Anderson *et al.*, 1977]. Brasseur and Solomon [1984] estimate the mixing ratio as varying between 5×10^{-13} and 3×10^{-11} between 30 and 50 km during day with the concentration reduced by 2 orders of magnitude during night [Turco, 1985]. We discuss below the requirements for detecting Cl at both day and night conditions. We base our detection scheme on measurement of the 808.6-nm emission, which follows the excitation of the electron level $\text{Cl}(5^2D_{3/2})$ by electron impact.

A remote sensor operating under daylight conditions is sub-

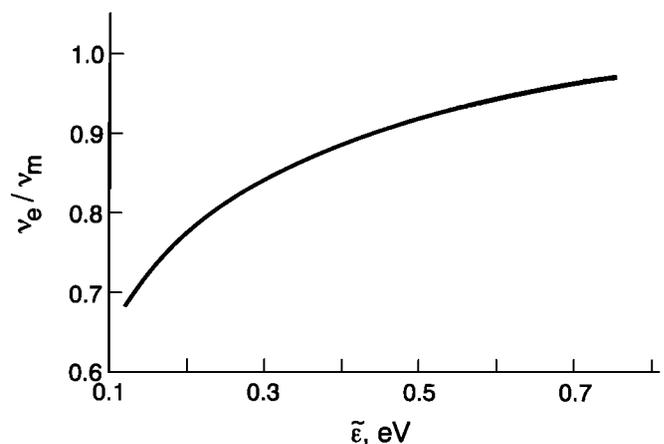


Figure 3. The ratio of ν_e/ν_m as a function of the quiver energy $\tilde{\varepsilon}$.

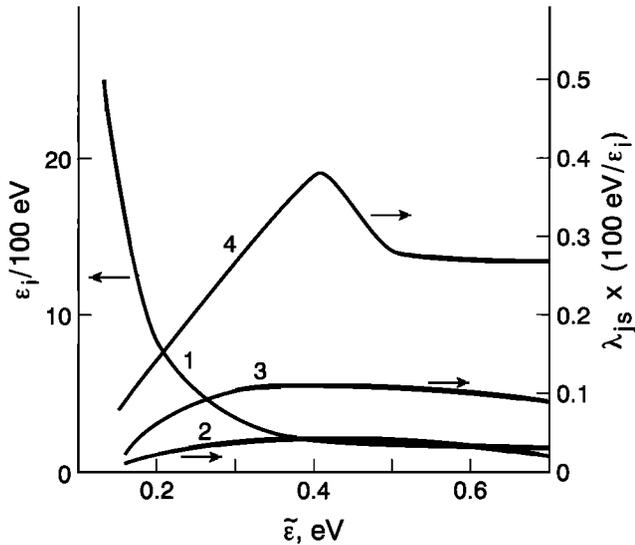


Figure 4. Curve 1 is adopted from Tsang et al. [1991]. It shows the microwave energy required per ionization ϵ_i normalized to 100 eV, as a function of the quiver energy $\tilde{\epsilon}$. Curves 2–4 show the function $\lambda_{js} \times \frac{100 \text{ eV}}{\epsilon_i}$, for the excitation of allowed transition of Cl (curve 2), CO (curve 3), and H (curve 4), reduced by a factor of 2.

jected to background illumination. The number of background photons P_b received per microwave pulse in the effective spectral window is

$$P_b = \frac{R_b S \Delta \lambda_o \tau \Omega}{h \nu_o} \quad (14)$$

where R_b is the sky spectral radiance at $\lambda = \lambda_o$, $h \nu_o$ is the photon energy, and Ω is solid angle defined by the field of view of the receiving system. We assume that the telescope views the region of breakdown caused by microwaves of the intensity within 3 dB of the maximum. From Figure 5 and for $R = 35 \text{ km}$, we find that it corresponds to a value of $\Omega = 1.6 \times 10^{-8} \text{ rad.}$ We also assume that by adjusting the microwave frequency, comparable field can be achieved at other heights so that $\Omega = \text{const.}$

To reduce the background intensity, the measurements could be conducted by looking away from the Sun. In fact, we use the spectral radiance at sea level for a 45° viewing angle under clear sky conditions which is given by $R_b(\lambda = 808.6 \text{ nm}) = 5 \times 10^{-3} \text{ W/m}^2 \text{ 1/nm sr}$ [Karp et al., 1988]. We, further, assume that the bandwidth of the applied optical filter $\Delta \lambda_o$ is of the order of 0.1 nm and the energy of the corresponding Cl photon is $h \nu_o = 1.5 \text{ eV}$. Figure 6 shows the result of calculations using equations (13) and (14). The shaded region in Figure 6 gives the number of Cl photons per microwave pulse measured by a 1 m^2 telescope aperture area as a function of altitude, for a daytime mixing ratio of Cl varying between 5×10^{-13} and 3×10^{-11} at 30–50 km [Brasseur and Solomon, 1984]. The pluses in Figure 6 show the number of background photons intercepting the telescope for a 100-ns pulse. Measurements of low photon input can be performed by using, for example, the automatic guider system (AGS) [Currie, 1977]. This system allows subtraction of the background emission, followed by the amplification of the input signal using photomultipliers. It has been designed as input in a star-tracking telescope. With an input of few photons per

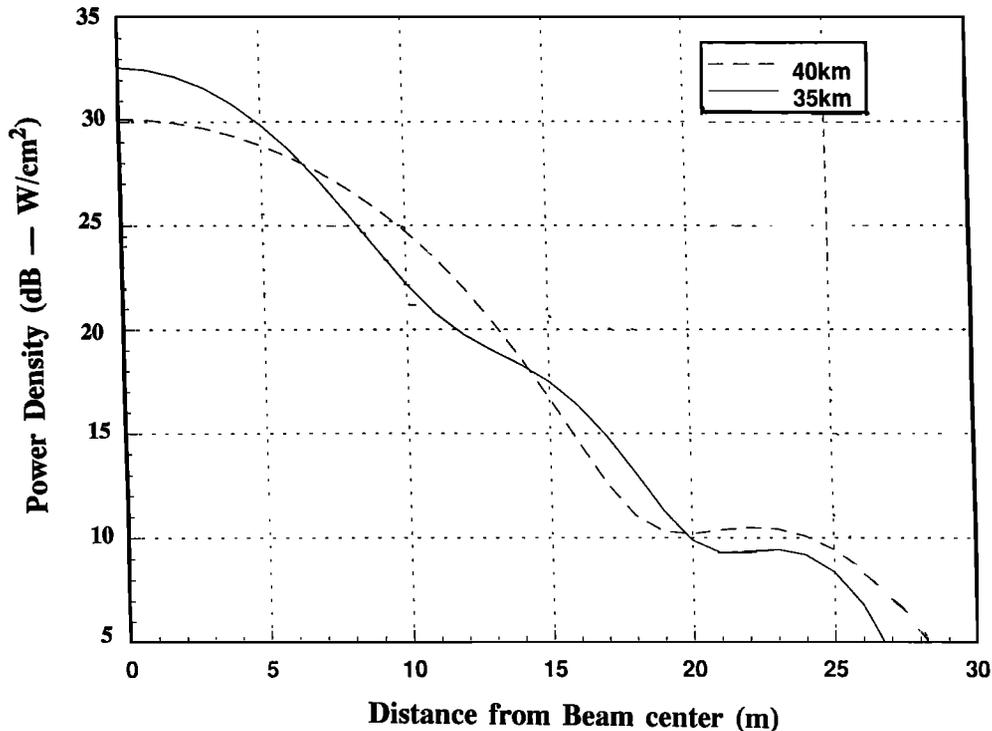


Figure 5. Power density from a microwave facility with power 3 GW focused by a 34-m dish at different heights for a 10 GHz frequency.

Table 2. Optical Parameters of the Target Molecules

Species	Metastable Level			Allowed Transition Matched to the Metastable Level			Wavelength of the Stimulated Transition, nm
	Excitation Energy, eV	Lifetime, ms		Excitation Energy, eV	Lifetime, ns		
CO	$a^3\Pi_r$	6.01	8	$b^3\Sigma^+$	10.39	56	285
NO	$B^2\Pi_r$	5.6	0.0031	$B'^2\Delta_1$	7.43	110	689
OH	$B^2\Sigma^+$	8.48	0.002	$C^2\Sigma^+$	10.94	4	512.2
H	$2s^2S_{1/2}$	10.2	long	$3p^2P^0_{1/2,3/2}$	12.09	5.3	656.2

pulse requires 10^4 consecutive pulses using 1 m² mirror. It means that with a 1-kHz repetition rate the integration time is about 10 s.

Additional difficulties in measuring emission lines can be due to overlapping by strong lines emitted by majority species. For example, the 6-5 transition of the nitrogen first positive band ($\lambda_o=847$ nm) and the Ar I ($\lambda_o=810$ nm) can possibly affect detection of the Cl($\lambda=808.7$ nm) line. Such effects should be studied using breakdown in small laboratory chambers, although, since both lines are due to forbidden transitions, we do not expect serious detection problems.

Since the nighttime concentration of Cl is by 2 orders of magnitude smaller than the day, a correspondingly lower Cl photon flux is expected. A possible nighttime detection scheme utilizes coincidence measurements using two mirrors. Such a scheme operating at a 1-kHz repetition rate could possibly detect mixing ratios below particle per billion (ppb) with an integration time of less than 1 s.

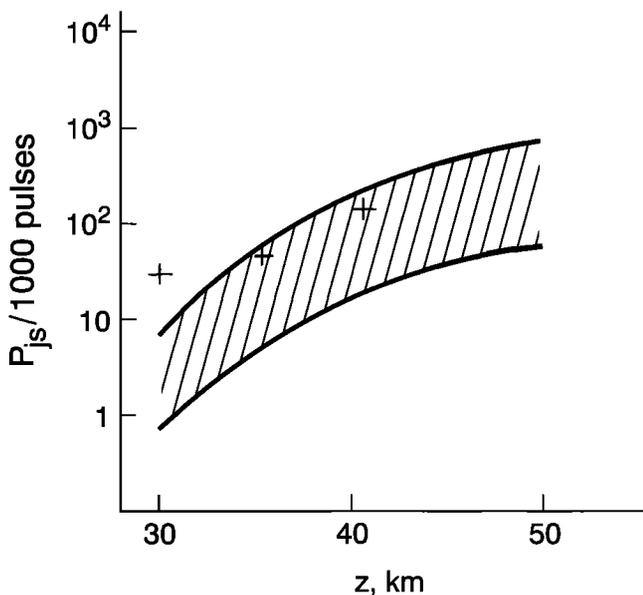


Figure 6. The number of Cl photons of $\lambda=808.6$ nm expected at daytime per microwave pulse per m² of the telescope area. The shaded region is drawn using the expected daytime concentration of Cl. The number of background photons is shown by pulses.

3.2. Detection of Atomic Hydrogen by Active Spectroscopy

Species lacking allowed transitions in the optical range can be measured by using excitation of metastable states by electron impact. The abundance of the metastables is measured by using two laser pulses with slightly different wavelengths and comparing their attenuation, which is called differential absorption lidar (DIAL) [Measures, 1984]. The wavelength of the first laser is chosen to coincide with the strong absorption feature between the metastable and the allowed transition located above the metastable electronic level. The wavelength of the second laser is detuned into the wing of the feature. To reduce the influence of collisional quenching on the concentration of metastables, the laser pulses should be sent at the end of the microwave pulse, and the width of the laser pulses should be short compared to the characteristic time of quenching.

The population of the excited metastable atoms/molecules N_{js}^* can be obtained from equation (4) since the lifetime of such state is long compared to the characteristic ionization time ($\tau_{js}\nu_i \gg 1$). In this case,

$$N_{js}^* = k_{js} N_s n(\tau) / \nu_i \quad (15)$$

Taking into account equations (8), (9), and (15), we can obtain that

$$N_{js}^* = \lambda_{js} g_s n(\tau) \quad (16)$$

The differential absorption of the two-laser return has the form

$$\frac{\Delta E}{E} = \sigma_a^r \lambda_{js} \int n(\tau) dl g_s \quad (17)$$

where σ_a^r is the resonant cross section for the transition of the metastable state to the allowed state, and the integration is along the direction of propagation of the laser signals. The value of the integral $\int n(\tau) dl$ can be evaluated assuming that the depth of the ionized region is limited by the absorption length of the microwave ΔR , i.e., by the distance along which the microwave power is reduced e^2 times

$$\Delta R = 2 \frac{c \omega^2}{\nu_e \omega_e^2} \quad (18)$$

where $\nu_e(\tilde{\epsilon})$ is the frequency of the electron-neutral collisions, and $\omega_e = \sqrt{3 \times 10^9 n(\tau) / \text{cm}^{-3}}$ is the plasma frequency. Using equations (17) and (18), we find

$$\frac{\Delta E}{E} = 2 \sigma_a^r \lambda_{js} g_s \frac{c}{\nu_e} n_c \quad (19)$$

where n_c is the critical plasma density for the frequency ω .

As an application of this technique we discuss the detection of the abundance of atmospheric atomic hydrogen based on the excitation of the metastable level $H(2s^2S_{1/2})$ by electron impact, followed by measurements with lidar wavelength of $\lambda=656.2$ nm. This corresponds to the excitation of the allowed transition $H(2s^2S_{1/2} \rightarrow 3p^2P_{1/2,3/2}^0)$. The cross section of the resonant absorption has the following form [McGee and McIlrath, 1984]:

$$\sigma_r^a(\lambda) = \sqrt{\frac{\ln 2}{\pi}} \frac{\lambda^4}{4\pi c} \frac{1}{\sqrt{\Delta\lambda_D^2 + \Delta\lambda_L^2}} \frac{g'}{g''} \frac{1}{\tau_{js}} \quad (20)$$

where $\Delta\lambda_D$ and $\Delta\lambda_L$ are correspondingly the Doppler linewidth and the laser linewidth; g' and g'' are the degeneracies of the upper and lower electronic states. The radiative lifetime is $\tau_{js} = 5.3$ ns.

The motion of the hydrogen atom at $T=200$ K causes Doppler broadening $\Delta\lambda_D \simeq 6$ pm. For the value of the cross section quoted above and using equations (19) and (20), we find

$$\frac{\Delta E}{E} = 1.2 \times 10^{-8} \left(\frac{f}{10 \text{ GHz}} \right)^2 \left(\frac{g_s(z)}{10^{-11}} \right) \left(\frac{10^{17} \text{ cm}^{-3}}{N(z)} \right) \quad (21)$$

Equation (21) allows us to determine the number of pulses required for detection of the H mixing ratio g_s as a function of the altitude (i.e., value of N) and of detection sensitivity $\Delta E/E$. If, following Measures [1984], we assume that a detection sensitivity $\Delta E/E \gtrsim 10^{-3}$, equation (21) implies that the number of pulses r required by the facility at $f = 10$ GHz will be given by

$$r \approx 10^5 \left(\frac{10^{-11}}{g_s} \right) \left(\frac{N(z)}{10^{17} \text{ cm}^{-3}} \right) \quad (22)$$

For $N(z) \approx 2 \times 10^{16} \text{ cm}^{-3}$ (50 km altitude), mixing ratios of 10^{-11} would require $10^4 - 10^5$ pulses, which at a 1-kHz repetition rate corresponds to integration times of 10–100 s.

4. Conclusions

A novel remote diagnostic technique of the atmosphere was presented. It is based on the atmospheric breakdown by focused microwave pulses. The energetic electron flux generated during breakdown leads to impact excitation of the minority species. Two different methods for monitoring the abundance of the minority species following breakdown were proposed. The first is based on measuring the intensity of optical emissions. The second method relies on use of differential absorption lidar to detect metastable states excited by electron impact. It was shown that a 10-GHz microwave heater with 3 to 4-GW power, a 100-ns pulse length, and 1-kHz repetition rate focused in the atmosphere by a 34-m dish could possibly detect mixing ratios below parts per trillion (ppt) with integration times of 1 s, using optical emission spectroscopy, and of the order of

10 ppt with 10 to 100-s integration time using DIAL. The system discussed here presents far from optimized an example design. In addition to the techniques discussed here, diagnostics using high-altitude platforms and satellites can detect IR, UV, and other emissions. The facility discussed here will be an ideal facility for atmospheric chemistry studies involving heterogeneous chemistry particles in the polar regions.

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