

Simultaneous *in situ* measurement of CO, H₂O, and gas temperatures in a full-sized coal-fired power plant by near-infrared diode lasers

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We present what is to our knowledge the first near-infrared diode-laser-based absorption spectrometer that is suitable for simultaneous *in situ* measurement of carbon monoxide, water vapor, and temperature in the combustion chamber (20-m diameter, 13-m path length) of a 600-MW lignite-fired power plant. A fiber-coupled distributed-feedback diode-laser module at 1.56 μm served for CO detection, and a Fabry-Perot diode laser at 813 nm was used to determine H₂O concentrations and temperature from multiline water spectra. Despite severe light losses (transmission, $<10^{-3}$) and strong background radiation we achieved a resolution of 1.9×10^{-4} (1σ) fractional absorption, equivalent to 200 parts in 10^6 by volume of CO (at 1450 K, 10^5 Pa) with 30-s averaging time. © 2003 Optical Society of America

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1. Introduction

The phasing out of nuclear energy in many industrialized nations in combination with large stocks of coal in the ground has generated strong interest in coal combustion as a source of electricity, so coal will remain a primary energy source of high significance for some decades to come. However, the increasing demand to reduce CO₂ emissions according to the Kyoto Protocol and to minimize the overall cost of combustion calls for an increase in combustion efficiency and service lifetime of power plants as well as for simultaneous minimization of pollutant emission. CO is a key molecule that directly affects these concerns.

The presence of CO indicates incomplete combustion and chemically reducing combustion conditions. Incomplete combustion leads to enhanced pollutant formation. Reducing combustion conditions causes two additional important problems: First, the oxidic protective layer of the metallic combustor walls are attacked, causing increased corrosion of the combustor walls and of the heat exchangers. Second, the

probability of slagging is increased because of the disadvantageous viscosity properties of the slag under reducing conditions.^{1,2}

In power plants and in other industrial settings, CO is measured chiefly with extractive sensors based on nondispersive infrared (NDIR) photometers.³ These devices always require extraction of a gas sample, which is typically done locally through a single access point to the flue gas duct. The gas sample is filtered to remove particles and cooled to well below room temperature to remove water, prevent condensation, and protect the sensor from hot gas. The sample is then transported to the measurement cell via a sampling tube that is sometimes as much as 100 m long. This sampling process creates numerous difficulties that affect the usefulness of the species signal for process control. These difficulties can range from slow temporal response to problems in obtaining representative concentrations caused by effects such as adsorption, condensation, chemical reaction, and fractionation during the sampling process. Furthermore, the NDIR sensors suffer, especially in the presence of complicated gas mixtures, from a lack of specificity that is caused, for instance, by the low spectral resolution of the optical filters used in some of these instruments. Extractive sensors in general and NDIR sensors in particular also generate a rather higher cost of ownership because they require routine calibration with reference gases.

Laser-based optical species sensors, especially if they are able to avoid gas sampling and can measure

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in situ within the combustion chamber, offer many advantages such as high speed and corrosion resistance. A promising technique for measuring CO is tunable-diode-laser absorption spectroscopy, which is based on diode lasers in the near-infrared (NIR) and the mid-infrared spectral regions. Because of their line-of-sight character, absorption-based methods provide more-representative results than do point sampling techniques, a factor that is important for the surveillance of large measurement volumes.

The high resolution and high sensitivity that are possible with tunable-diode lasers (TDLs) is widely documented in the literature, and first experiments that used the advantageous spectroscopic properties of TDLs date back as far as the 1970s, when CO detection was first achieved for environmental applications by use of cryogenically cooled mid-infrared TDLs.⁴ This technique was then quickly extrapolated to solution of combustion problems.^{5,6} Today mid-infrared TDLs are still of great importance for basic⁷ and combustion⁸ research. However, with the availability of mass-produced, room-temperature (RT) operated TDLs, for telecommunication purposes the research began to concentrate on the NIR spectral range, taking advantage of the high spectral quality, simple operation, and low cost associated with NIR TDLs. But NIR TDL applications suffer to a certain extent from the low absorption coefficients that are provided by most overtone and combination bands. This is especially true for the second overtone of CO near 1.5 μm .^{9,10} But, despite recent progress in the development of RT InSb TDLs for the first vibrational overtone band at 2.3 μm ,^{11–13} and even of near-RT quantum cascade lasers that are suitable for the fundamental vibrational transition at 4.6 μm ,^{14,15} NIR TDLs still seem to be a good compromise for applications that offer long absorption paths and require only moderate sensitivity. This is particularly true as long as InSb lasers are not commercially available and the quantum cascade lasers suffer from unpredictable long-term reliability, lack of RT detectors, and the need for special pulsing electronics.

Many studies of the detection of CO in a well-controlled laboratory environment have been published: They were based either on extractive gas sampling in RT absorption cells^{8,11,14–20} or on operation in small (a few centimeters in diameter) and highly stable flame environments^{5–7,12,13,21–23} such as McKenna, Hencken, or Wolfhard-Parker burners.

But, even though such studies have often aimed at industrial combustion applications, only a few *in situ* CO measurements made under industrial conditions, in which combustion chambers had diameters of as much as 20 m, have been published. For instance, Allen *et al.*²⁴ reported on a NIR-TDL-based fiber-coupled *in situ* spectrometer used for rapid CO detection. The measurement path (30-cm path length) was located outside the furnace in the flue gas duct of a high-temperature 750-kW pilot furnace. Despite the short optical absorption path, which was located outside the combustion zone, Allen *et al.* reported a

much higher noise in their signal compared with laboratory flame conditions, as a result of particulates in the flue gas and fluctuations in radiative background emissions. However, no amounts of absolute particulate loading or transmission of the measurement path were specified. In another study Muta *et al.*²⁵ presented measurements of CO in gas cells (sensitivity, 5 ppm \times m/Hz), where ppm is parts in 10^6 , as well as in a waste incinerator (2.2-m diameter). From that paper it can be estimated that they achieved a CO detectivity of 500 ppm at 1000 °C. However, they worked at a rather low average particulate load, well below 200 mg/(STP m³).

There is strong interest in *in situ* CO measurement for coal combustion processes. But typical combustion plants have diameters of 10–20 m. In addition, because these plants are fired by coal dust, absorption measurement faces many problems because of the very high dust load (well above a few grams per cubic meter) and the high luminosity of the process. We have found in the literature no reports of successful *in situ* CO measurements under such demanding conditions.

Over the past few years it was demonstrated during fire-suppression tests,²⁶ in waste incinerators,²⁷ in large gas-fired power plants,^{28,29} and even in high-pressure combustion processes³⁰ that diode lasers not only are suitable for highly sensitive extractive gas analysis³¹ under well-controlled conditions but, in combination with suitable engineering and data evaluation NIR TDLs, are fascinating and powerful tools well suited for *in situ* gas analysis in harsh industrial environments. Additionally, the range of transmission conditions usable with TDL spectrometers was recently improved, and what is to our knowledge the first *in situ* species measurement in a mid-sized coal-fired power plant was made.³²

Here we report on a new NIR TDL spectrometer that facilitates simultaneous *in situ* measurement of two species concentrations and temperatures under highly adverse conditions, namely, at the end of the combustion chamber (20-m diameter) of a full-scale (600-MW) power plant fired with pulverized coal. We demonstrate that it is possible to determine small molecular absorptions, of the order of a few percent, in the presence of a quickly fluctuating combustion process that absorbs or scatters $\sim 99.9\%$ of the emitted laser light and produces more background light on the detector than is received from the laser itself. The spectrometer and the spectroscopy behind are explained, and simultaneous measurement of CO and water will be demonstrated.

2. Theory

We use direct absorption spectroscopy for diode-laser-based *in situ* gas analysis in complicated environments. That means that, unlike with other techniques such as wavelength-modulation spectroscopy,^{18–21,25,33} frequency-modulation spectroscopy,^{8,23,34} and balanced detection,^{17,31} the decoupled detector signal, including all offsets and disturbances, is digitized and evaluated. No further

modulation of the laser wavelength is used besides the scanning modulation needed to recover the complete absorption line shape.

The major advantage in using direct absorption spectroscopy is that it is possible to perform a calibration-free measurement of absolute gas concentrations. The trade-off is lower sensitivity than for double modulation techniques (wavelength-modulation or frequency-modulation spectroscopy). The basic setup is simple: The beam of a diode laser is directed through the measurement volume onto a photodetector. The laser wavelength is scanned over the absorption line, and the area of the absorption line is extracted to yield the number density of absorbers. However, great care has to be taken to correct the *in situ* signals for various strong disturbances found under combustion conditions: Most important are strong fluctuations of the overall transmission $Tr(t)$, where t is time, of the measurement path caused by broadband absorption, scattering by particles, or beam steering owing to refractive-index fluctuations. Furthermore, there is intense thermal background radiation $E(t)$ from burning coal particles that increases the detector signals. These effects change rapidly in time and have to be included in an extended version of Beer's law:

$$I(\lambda) = I_0(\lambda)\exp[-S(T)g(\lambda - \lambda_0)NL]Tr(t) + E(t).$$

Detected intensity $I(\lambda)$ is given by initial intensity $I_0(\lambda)$ times an exponential decay that is due to molecular absorption and the time-dependent broadband transmission $Tr(t)$. $E(t)$ describes the thermal background radiation. The molecular absorption signal itself is described by temperature-dependent line strength $S(T)$, number density N of molecular absorbers, absorption path length L , and a normalized (area = 1) function $g(\lambda - \lambda_0)$, which describes only the shape of the absorption line, which is centered at wavelength λ_0 .

We can effectively separate the transmission and emission effects from the molecular absorption and correct them by taking advantage of two important features of diode lasers, their fast wavelength tunability (by modulating the laser current) and their simultaneous current-induced output power modulation. By tuning the laser much faster than the fluctuations in transmission and emission we can assume that these disturbances will be constant during the wavelength scan and correct them by subtracting the offsets and dividing the offset-corrected scan through the baseline function. Phase-locked averaging of the wavelength scans, equivalent to using a comb filter at the modulation frequency and its overtones, is used for further noise reduction.

3. Line Selection

The CO absorption band overlaps nicely the commercially available relatively inexpensive fiber-coupled telecommunication diode lasers that are used for wavelength-division multiplexing. These lasers offer high spectral quality, good availability, and ad-

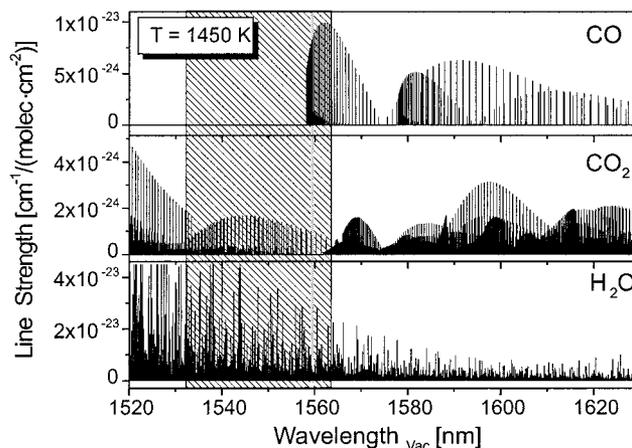


Fig. 1. Line-strength spectra of CO, CO₂, and H₂O near 1560 nm taken from the Hitran'96/Hitemp database. The hatched regions indicate the telecommunication C band; the vertical line at 1560 nm marks the region of CO line selected for the present study.

vantageous (butterflytype) diode-laser packages with integrated Peltier coolers, optical isolators, and high optical output power. Thus we decided to choose the 1.56- μm band for the detection of CO. Nearly 2-orders-of-magnitude stronger CO absorption and less water interference can be found in the region of the first overtone at 2.3 μm . However, these research-grade Fabry-Perot-type laser diodes are not commercially available. In addition, they show badly reproducible spectroscopic properties (mode hops), so these lasers are not well suited for industrial measurements.

Figure 1 shows the NIR line-strength spectrum of CO and the major combustion products CO₂ and H₂O in the spectral region of the second vibrational overtone of CO near 1.6 μm from the Hitran'96/Hitemp data base.³⁵⁻³⁸ We modeled a small section of spectrum in Fig. 1 with good candidates for *in situ* mea-

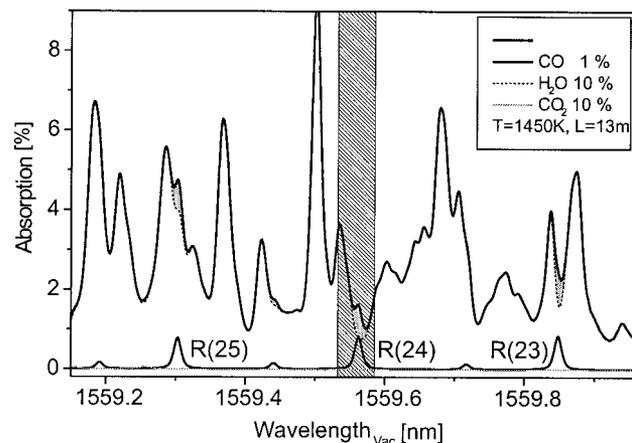


Fig. 2. Absorption spectrum for 0.5% CO, 10% H₂O, and 10% CO₂ at $T = 1450\text{ K}$, $p = 1\text{ atm}$, and $L = 13\text{ m}$ calculated with Hitran'96/Hitemp data and Voigt line shapes. The R(24) transition seems to be best suited for *in situ* CO detection at 1450 K. The hatched area indicates the wavelength interval accessible by current tuning of the DFB diode laser used in this study.

measurements, using Voigt line shapes. Figure 2 shows the calculated absorption spectrum for power-plant-specific combustion conditions ($T = 1450$ K, 0.5% CO, 10% H₂O, 10% CO₂, $p = 10^5$ Pa, $L = 13$ m). The R(24) line was selected for our measurements because it is best isolated from neighboring high-temperature H₂O lines.

Because of the lack of suitable temperature measurement in some power plants, we investigated the possibility of spectroscopic laser-based temperature determination by two-line thermometry.^{28,32} Neighboring CO absorption lines could not be used for that purpose because of their similar temperature behavior and their large spectral separation. Water absorption in the immediate vicinity of the selected CO line is also unsuitable for temperature determination. Therefore a H₂O line triplet in the 813-nm region was selected that had been used for temperature determination in previous studies.^{28,29,32} These lines are well suited for two-line thermometry because of their differing temperature behavior in the expected region. In addition, the 813-nm laser has a relatively high output power of 40 mW, which is quite helpful in these strongly scattering combustion environments.

4. Laser Characterization

We had to investigate several electronic and optical properties to select suitable lasers for *in situ* measurements. A pigtailed multiquantum-well distributed-feedback (DFB) laser-diode module with an optical isolator (Mitsubishi) and a peak optical output power of 15 mW was selected for the CO absorption measurement. The integrated thermoelectrical cooler guaranteed excellent temperature stabilization even under power-plant conditions. For the water-vapor measurement near 813 nm we used a single-mode Fabry-Perot laser (Sharp) with 40-mW maximum power.

We could tune the diode-laser wavelength slowly and coarsely by changing the diode laser's temperature or quickly and finely by adjusting the operating current. The static and dynamic tuning properties of both lasers were characterized. A wavemeter (Burgleigh WA 1000) was used for the static temperature- and current-tuning coefficients depicted in Fig. 3. Whereas DFB-type lasers show a nice continuous tuning behavior, the tuning of a Fabry-Perot laser is interrupted by mode hops. This effect hinders access to the selected absorption lines and requires selection of laser diodes.³⁰

For the 1560-nm DFB laser we determined a static temperature-tuning coefficient of 0.40 cm⁻¹/K and a static current-tuning rate of 0.0242 cm⁻¹/mA. The temperature-tuning coefficient of the 813-nm Fabry-Perot laser (Sharp) was 0.995 cm⁻¹/K within a mode. Its static current-tuning coefficient was 0.112 cm⁻¹/mA. The mode hops of this laser were stable and reproducible. The wavemeter used was based on a Michelson interferometer. Adding some self-made timing electronics and a digital oscilloscope to calculate the fast Fourier transform of the interference

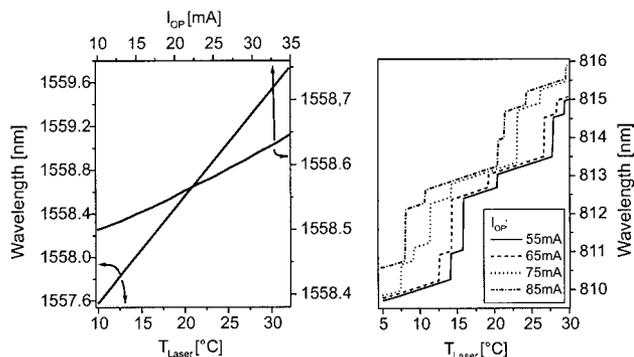


Fig. 3. Left, static temperature- and current-induced wavelength tuning of the 1560-nm DFB laser (Mitsubishi; 0.40 cm⁻¹/K, 0.0242 cm⁻¹/mA). Right, static temperature-induced wavelength tuning of the 813-nm Fabry-Perot laser (Sharp). Sections with continuous tuning are interrupted by mode hops. The temperature-tuning rate within a mode was 0.995 cm⁻¹/K. The static current-tuning rate within a mode was 0.112 cm⁻¹/mA.

fringe pattern allowed the wavemeter to be used also to record the emission spectra of the laser diodes. The side-mode suppression of the selected lasers was well above 25 dB for the Fabry-Perot laser and better than 30 dB for the DFB laser.

With increasing modulation frequency, the current-induced wavelength tuning at a fixed current modulation amplitude decreased. Furthermore, the tuning became increasingly nonlinear owing to the finite thermal relaxation of the heat deposited in the semiconductor crystal. This dynamic tuning rate must be known precisely for calibration-free evaluation of the absorption signals. Therefore we used an air-spaced Fabry-Perot etalon ($L = 10$ cm) to determine the dynamic tuning rate by counting the interference fringes, using Labview software. Figure 4

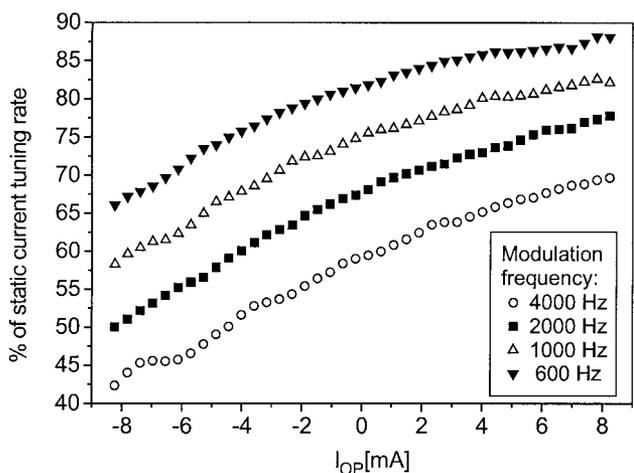


Fig. 4. Dynamic tuning behavior of the 1560-nm DFB diode laser under triangular modulation for several modulation frequencies (only the up scan is shown) at constant modulation amplitude. The tuning rate is normalized with respect to the static current-tuning coefficient (0.0242 cm⁻¹/mA). Nonlinear tuning behavior over the wavelength scan is evident, as well is a narrowing of the wavelength span with increasing modulation frequency.

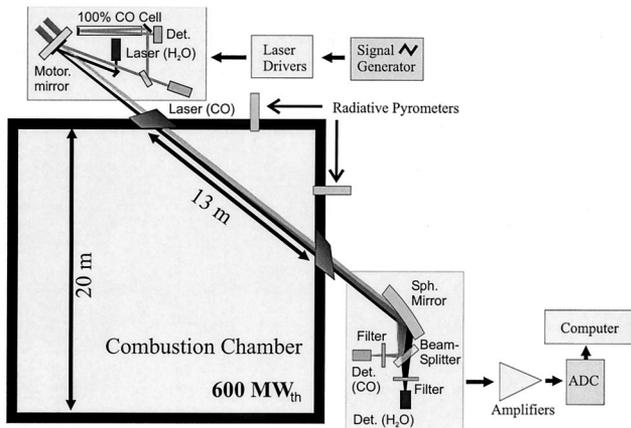


Fig. 5. Schematics of the experimental setup during *in situ* CO/H₂O measurements at a coal-fired power plant. Det.'s, detectors; Sph., spherical.

shows the dynamic tuning properties of the DFB laser for various modulation frequencies at a fixed current modulation amplitude. At modulation frequencies of as high as 5 kHz the dynamic tuning rate is reduced by as much as 50% of the static tuning coefficient. The tuning curves determined in this study can be used further to linearize the wavelength axis of the recorded absorption signals to improve the data evaluation.

5. *In Situ* Measurement

The *in situ* measurements of CO and water vapor were performed in an industrial full-scale lignite-fired power plant with a thermal power of 600 MW. The combustion chamber was ~100 m high and had a square cross section of 20 m by 20 m. The plane of measurement was localized 65 m above the floor of the combustion chamber, immediately below the first heat exchangers. In the first campaign we measured diagonally across one of the corners of the furnace, which generated an absorption path length of 13 m. Recently we also obtained an absorption path through the center of the combustion chamber ($L = 20$ m). The results presented here were all measured with the 13 m-path. Figure 5 shows schematics of the spectrometer setup.

All spectrometer components were mounted directly onto the furnace walls by use of water-cooled flanges with an inner diameter of 100 mm. The use of water-cooled flanges was unavoidable because the RT at the location of the electronics was 320–330 K and even higher at the site of the optics components near the combustor walls. Highly stable current sources were used for the Peltier elements controlling the laser temperature and to provide a low-noise laser current. The two lasers were synchronously current modulated with triangular 5-kHz signals from two signal generators (SRS DS345). The laser beams were collimated and superimposed, forming a composite dual-wavelength beam. Then the beam was directed through the combustion chamber via a mirror. Brewster windows (~6-cm effective diame-

ter) permitted optical access to the combustion chamber. A small fraction (4% per surface) of the 1560-nm beam was split off the main beam with an uncoated window and was double passed through a reference cell (30 cm long) filled with pure CO to permit reliable identification of the *in situ* CO line. The high initial-state energies of the water lines did not permit use of a near-RT reference cell with water.

The transmitted laser light was collected with a 3" (7.62-cm) spherical mirror (25-cm focal length) and separated into its individual wavelengths by a dichroic beam splitter. Optical narrow-band interference filters (10-nm FWHM) were used to reduce the strong thermal background radiation and to prevent cross talk between the two wavelength channels.

Visual alignment of the *in situ* laser beam over a distance of 13 m was nearly impossible because of the high luminosity of the combustion and the low transmission. Time-consuming manual alignment was not practical because of the high ambient temperatures of as much as 60 °C near the furnace walls. Therefore we developed an automatic alignment tool based on a motor-driven ϑ - ϕ mirror mount and a computerized line-by-line search algorithm. By isolating the modulated laser radiation from the background by phase-sensitive detection we achieved excellent immunity to noise and obtained perfect alignment within a few minutes, even under difficult conditions. For other applications under unstable geometric conditions, a permanent alignment control loop has already been achieved by use of a position-sensitive detector.²⁸

Thermoelectrically cooled InGaAs detectors (Hamamatsu, 3-mm diameter) were employed to convert the 1560-nm signals. Uncooled silicon photodiodes (5-mm diameter) detected the 813-nm laser light. The electrical signals were amplified (Femto DLPCA-S amplifier) and digitized with a sampling rate of 5 Msamples/s by a 12 bit analog-to-digital converter (ADC) board (National Instruments, 6110E) for further computer-based data evaluation. Two hundred successive scans were co-added within a 3-s interval and stored to disk. As the desired response time was only 30 s, we averaged ten 3-s scans before correcting for transmission–emission and evaluating the line shape. The total number of single scans used per concentration value was 2000, yielding a duty cycle of only 1/150. Employing digital signal processors with high data throughput should drastically improve this value in the near future. The laser intensity modulation and the wings of the neighboring absorption lines not included in the main absorption line fits were modeled by a third-order polynomial. Linear multiline Voigt fits were used to derive the absorption line areas. We converted the line areas into concentrations by applying an extended Beer's law and the ideal gas law and taking into account the pyrometric temperatures provided by the combustion control system.

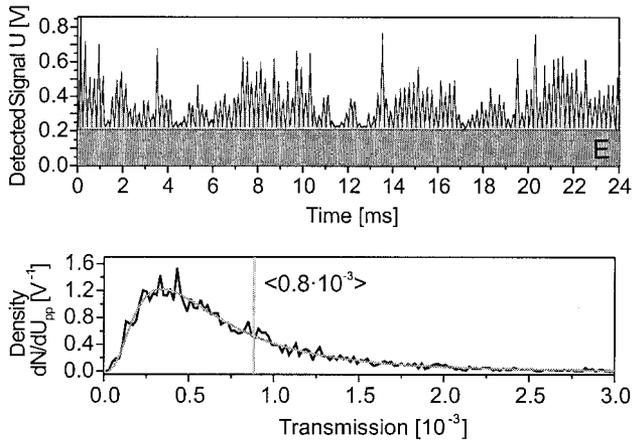


Fig. 6. Top, dc-coupled *in situ* detector signal over a high number of wavelength scans (each sharp spike corresponds to an up and a down scan). The amplitude modulation of the signal is modified by strong transmission losses along the absorption path. In addition, the detector signal is increased by thermal background emission (*E*). Bottom, transmission levels found over a longer time period, indicating an average *in situ* transmission of the measurement path (vertical line) of 8×10^{-4} .

6. Results

The total measurement time was 5 days. The power plant was permanently running under full load conditions. However, the combustion air supply was modified intentionally to generate variations in the *in situ* CO concentrations. All results shown were achieved during the same 8-h time slot.

A typical short segment of raw data on a millisecond time scale is depicted in Fig. 6. The triangular amplitude modulation of the 1560-nm laser generated by the 5-kHz current modulation is clearly recognizable. Strong variations in the amplitude modulation result from fast changes of the transmission level. The entire signal rides on a relatively slowly varying offset (*E*), which is generated by thermal background radiation. We have shown the statistics of the transmission variations in Fig. 6 by plotting the frequency dN of occurrence of a certain transmission interval of width dU versus the transmission. The mean transmission of the *in situ* path was less than 8×10^{-4} O.D.; the most probable transmission was even less, of the order of 4×10^{-4} O.D.

Long-term variations in transmission and background emission ratio are shown in Fig. 7. The lighter curves are 30-s averages, and the darker curves are adjacent averages over time intervals of 30 min. The background emission itself (not shown) is fairly stable and fluctuates by only 10%. The ratio of thermal emission to transmitted laser power is also plotted in Fig. 7 and indicates that the thermal background emission that passes through the narrow-band filters is as much as 10 times higher than the transmitted laser light. However, efficient techniques to correct for fluctuations in transmission and background emission have been developed as mentioned above, so the absorption signals could be corrected precisely for these effects.

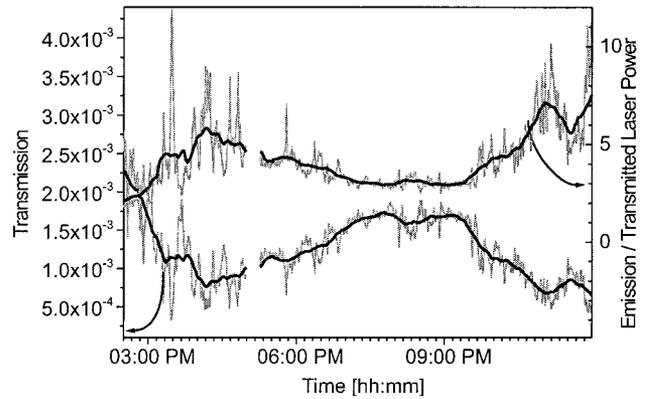


Fig. 7. Time series of the transmission [$Tr(t)$] through the combustion chamber (left axis) and ratio of the detected thermal background emission behind the filters ($1.1 \times 10^{-5} \text{ W} \pm 10\%$) to the transmitted laser power (right axis). The interruption at 5:00 PM is due to window cleaning and realignment. (Lighter trace, 30-s average; darker trace, 30-min moving average.) Here and below, [hh:mm] indicates hours and minutes.

Typical *in situ* absorption profiles of CO and water vapor acquired under combustion conditions are depicted in Fig. 8. Both scans have been corrected for background emission and broadband transmission. The absorption of the *R*(24) CO line (left) with a peak absorption of 1.15% is equivalent to a CO concentration of 1.2% by volume. The absolute volume fraction was derived from the area of the Voigt line shape fitted to the measured line profile by use of Beer's law and the ideal gas law. Further parameters needed for the conversion are the dynamic current tuning rate [described above ($0.012 \text{ cm}^{-1}/\text{mA}$)], the line strength [$S(T)$] of the transition, which was calculated from Hitran'96 data, and finally the pyrometric gas temperature. For the CO line there is a small water line lying underneath, so the resulting CO con-

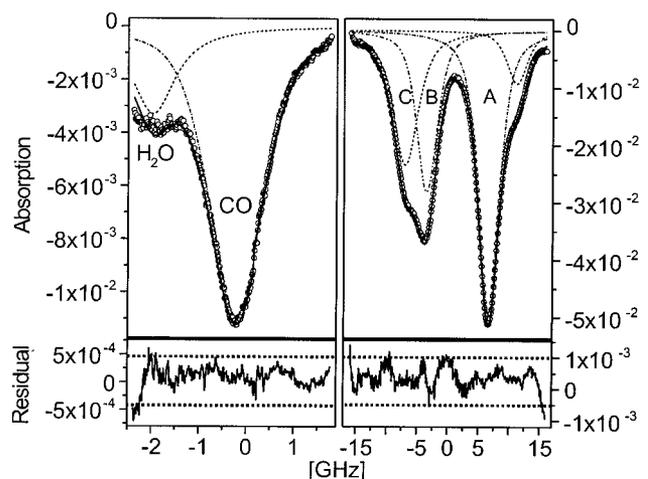


Fig. 8. Typical *in situ* line shapes (30-s average) of the CO *R*(24) line at 1559.5 nm and H_2O at 813 nm measured at a coal-fired power plant. The scans are transmission and emission corrected and normalized to the initial laser intensity. Multiline Voigt fits were used for concentration determination. A detectivity of 10^{-3} to 10^{-4} O.D. can be derived from the residual.

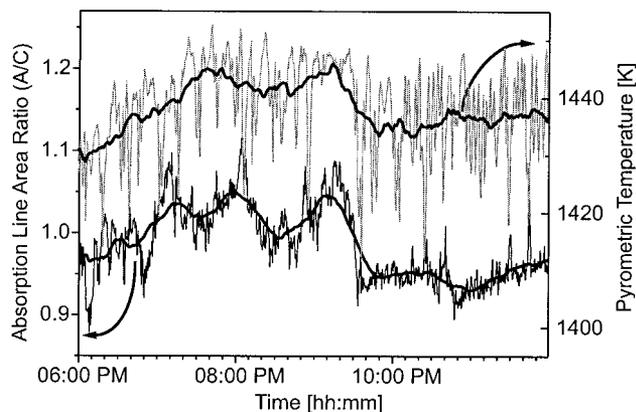


Fig. 9. Time series of the line area ratio of H₂O lines A and C in Fig. 8 compared with temperature data from the radiative pyrometer taken looking into the combustion chamber.

centration has to be reduced by 70 ppm by volume for each percent by volume of H₂O detected in the flue gas. For the conditions shown this contribution is not significant. However, the correction could be performed with simultaneous water measurement when the CO concentration is low. We evaluated the residual between the Voigt fit and the acquired scan to estimate the detectivity of our spectrometer. For CO the standard deviation (1σ) is 1.9×10^{-4} , which converts to a detection limit of 200 ppm by volume CO at a 30-s averaging time (=2000 scans averaged).

The right-hand plot in Fig. 8 shows the water-vapor spectrum recorded at the same time. We used line A (13 0 13 ← 14 0 14, 211 ← 000) to determine the concentration because a single-line fit is more reliable than a dual-line fit. Because of the temperature dependence of this line, a correction for ambient water vapor outside the combustor was not necessary. The H₂O concentration derived in the presented spectrum was 14.5% by volume. A minimum detectable absorption of 3.3×10^{-4} (1σ) could be determined, which corresponds at that temperature to a resolution of 0.1% by volume H₂O.

As a temperature indicator we used the line area ratio of the H₂O lines A (13 0 13 ← 14 0 14, 211 ← 000), B (625 ← 634, 112 ← 000), and C (735 ← 634, 211 ← 000) in Fig. 8. The results of the temperature measurements are shown in Fig. 9. Whereas the area ratio $A/(B + C)$ was used in previous studies,^{28,29,32} we found a better correlation with the radiative pyrometer data by using the ratio A/C . The reason for this is still under investigation. In addition, it has been reported²⁸ that Hitemp and Hitran data are erroneous for lines B and C, so we performed calibration measurements to determine the line ratios at various temperatures. These calibrations have been made only up to 1270 K. Therefore we are conservative and will generate relative temperatures from our measurements only, until we manage to extend the calibration measurements to 1500 K.

For this reason we used the pyrometric temperature data of the power plant for evaluation of gas

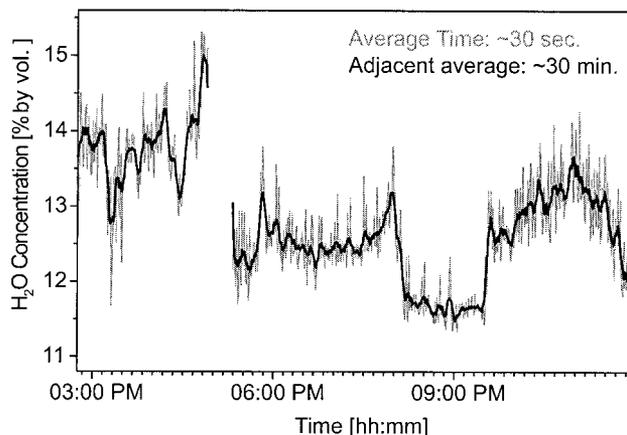


Fig. 10. Time history of the *in situ* water-vapor concentration (30-s average and 30-min moving average). The interruption at 5:00 PM is due to window cleaning and realignment.

concentrations of water and CO. The temporal evolution of the water-vapor concentrations derived from line A is shown in Fig. 10 for 30-s and 30-min averages. One important property of the water signal is that it can be used to correct for water interferences in the CO signal, which will be particularly important at lower CO concentrations. However, it is again necessary to extend the experimental data on the line strength to higher temperatures to verify the Hitemp data.

Finally, Fig. 11 demonstrates the results (for both time resolutions) of the *in situ* measurement of CO in the lignite-fired power plant. We generated several CO bursts near 3:30 and 5:00 PM by manipulating the secondary airflow into the combustion chamber. Figure 11 shows the *in situ* data generated when diode-laser absorption spectroscopy was used in comparison with a conventional extractive CO measurement. The probing location of the NDIR device was in the flue gas duct ~100 m behind the combustor. At that sampling point we found, as expected, much lower CO concentrations than in the combustion

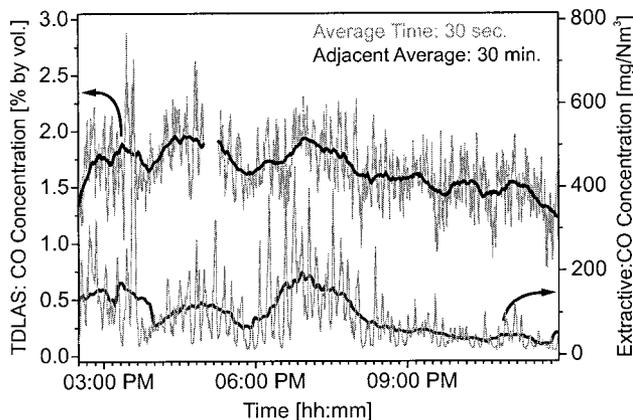


Fig. 11. Comparison of the laser-based *in situ* CO concentration measurement (left axis) with extractive CO detection by an NDIR sensor (right axis). The NDIR sampling point was located 100 m downstream from the combustor.

chamber. Nevertheless, we found a close correlation (better than 96% over this 8-h period) between the two CO measurements, which we determined by a cross correlation of the CO signals. The cross correlation also yielded a time lag between the two signals, which is consistent with the flue gas velocity and the distance between the measurement points. Both facts support the validity of our measurement, despite the large concentration difference.

7. Conclusion

We have demonstrated what is to our knowledge the first NIR-diode-laser-based dual-species *in situ* spectrometer for simultaneous measurement of CO and H₂O concentrations and gas temperature during industrial full-scale coal-fired combustion. The spectrometer was successfully tested over two 60-h periods in a 600-MW full-scale lignite-fired power plant (absorption paths, 13 and 20 m). A fractional absorption resolution of better than 10⁻³ with a time resolution of 30 s could be achieved, despite severe disturbances (transmission 10⁻³ to 10⁻⁴, strong background radiation even behind a narrow-band interference filter) and high temperatures within the *in situ* measurement path. This is equivalent to a resolution (1σ) in the CO (H₂O) concentration of 200 ppm by volume (0.1% by volume). A new automatic alignment setup based on a motorized mirror mount was demonstrated that was able to align lasers and detectors within a few minutes even at near-zero visibility along the beam path.

The spectrometer is easily extendable to other species such as O₂, NH₃, HCL, and HF by a simple change of laser diodes, beam splitters, and optical filters. Therefore it has the capability to become a universal tool for industrial gas analysis in advanced combustion control applications.

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 38. Further information on HITRAN and HITEMP available from www.hitran.com.