Finding a gas plume’s temperature from its infrared spectrum

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Infrared spectrometer measurements can determine the temperature of a stack plume, with the help of fundamental concepts from molecular physics.

Remote sensing of stack plumes can provide vital information about the dispersion of effluents in the atmosphere. We would like our remote measurements to reveal the identities, concentrations, and mass flow rates of various molecular species in the plume. By observing infrared signatures from rotational-vibrational transitions, we can identify CO\textsubscript{2}, H\textsubscript{2}O, and various trace components. Unfortunately, because infrared emission from molecules is a function of both temperature and concentration, it is difficult to get quantitative information from measured intensities of spectral features. A reliable spectroscopic plume ‘thermometer’ would be a first step toward solving this problem.

The realities of remote sensing under field conditions impose practical constraints on our measurements. Commonly used spectroradiometers have insufficient resolution to infer temperature from Doppler broadening of individual spectral lines. Dense plumes are optically thick (that is, their emission is saturated at the blackbody limit) and one can find their temperature from the Planck blackbody equation. However, plumes of practical concern are often optically thin, with insufficient density to absorb much of their own radiation, even at wavelengths near molecular resonances. The blackbody equation does not apply under such circumstances.

We have developed and tested a new approach to the plume-temperature problem, based on the observation that emission from any radiating system shifts toward shorter wavelengths as temperature increases.\textsuperscript{1} In a gas, transitions between well-defined quantum states cause emission, and high-energy transitions radiate at short wavelengths. In cool plumes, most molecular collisions have too little energy to populate these high-energy states, but hot plumes emit much more at short wavelengths. The ratio between the emitted intensity at two wavelengths corresponding to two different transitions can be related to temperature in accordance with basic molecular physics. Taking this ratio removes the dependence on gas concentration, since concentration changes affect both wavelengths equally in optically thin plumes.

The emitted intensity from a molecular transition depends on the population of the upper level, as given by the Boltzmann distribution function. Adapting standard theory,\textsuperscript{2} we derived an equation for temperature $T$ in terms of the measured intensity ratio $R$:

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T = \frac{E_2 - E_1}{k \ln \left( \frac{R}{F(\lambda_1/\lambda_2)} \right)}
$$

where $k$ is Boltzmann’s constant, $\lambda_1$ and $\lambda_2$ are the two wavelengths being measured, and $E_1$ and $E_2$ are the corresponding transition energies. The proportionality factor $F$ appears because of the intrinsic intensities of the two transitions, which depend on quantum-mechanical factors that are difficult to calculate

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from first principles. We found, however, that one can use library gas spectra to calibrate this factor, after making appropriate corrections for the fact that they are absorption, rather than emission, spectra.

We tested the temperature diagnostic in field experiments using two different types of plumes. First, we made small-scale measurements on a sideways-directed plume from an internal combustion engine. Then, in cooperation with the Environmental Protection Agency, we conducted extensive studies using a portable propane-burning plume generator that was set up on a regional airport runway near Waxahachie, TX (see Figure 1). For most of our measurements, the stack was oriented horizontally, allowing us to place the spectrometer close to the plume to minimize absorption of emitted energy by the intervening air. The plumes obtained from these sources spanned the temperature range from 316 to 606K, as measured by hand-held thermocouples and signals from a large three-dimensional thermocouple array.

We tested the spectroscopic temperature diagnostic using emission from two CO$_2$ transitions at wavelengths of 4.2 and 14.5 μm. We chose wavelengths on the wings of the emission bands to avoid absorption of photons by atmospheric CO$_2$ as well as self-absorption by the plume.

The data conformed well to the theory outlined above. The computed spectroscopic ratio behaved as predicted when plotted against thermocouple temperatures. The spectroscopic temperatures were in good agreement with thermocouple values; the RMS error, computed from all data sets, was 22K, on the order of 5% of the absolute temperature values. The greatest source of error was probably the variation in the plume’s location and temperature caused by wind and turbulence within the plume. We found considerable temporal variation in the thermocouple readings: indeed, these measured temperatures often varied by about 20K over the time scales of our measurements. Considering the environmental vagaries inherent in the current experimental procedures, we expect greater accuracy in future more-refined measurements.

Follow-up research will investigate the use of transitions associated with gases other than CO$_2$. Based on preliminary results, methanol, ethanol, and water bands are promising. Ultimately, we hope that the spectroscopic-ratio technique can be used routinely to obtain plume temperatures and absolute concentrations of plume gases.

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**References**