9-23-2003

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Recommended Citation

Herbert J. Mitchell, Gerard P. Jellison, David P. Miller, Carl Salvaggio, Craig J. Miller, "The importance of background in the detection and identification of gas plumes using emissive infrared hyperspectral sensing", *Proc. SPIE 5093, Algorithms and Technologies for Multispectral, Hyperspectral, and Ultraspectral Imagery IX*, (23 September 2003); doi: 10.1117/12.488188; <https://doi.org/10.1117/12.488188>

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The Importance of Background in the Detection and Identification of Gas Plumes Using Emissive Infrared Hyperspectral Sensing

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ABSTRACT

Using a Fourier transform infrared field spectrometer, spectral infrared radiance measurements were made of several generated gas plumes against both a uniform sky and terrestrial background. Background temperature, spectral complexity, and physical homogeneity each influenced the success of emissive infrared spectral sensing technology in detecting and identifying the presence of a gas plume and its component constituents. As expected, high temperature contrast and uniform backgrounds provided the best conditions for detectability and diagnostic identification. This report will summarize some of SITAC's findings concerning plume detectability, including the importance of plume cooling, plumes in emission and absorption, the effects of optical thickness, and the effects of condensing plumes on gas detection.

Keywords: Plumes, IR spectrometry, Effluent Detection

1. INTRODUCTION

SITAC scientists have been developing a modeling tool called the Spectral Architecture Evaluator (SAE)¹, which does an end-to-end estimation of the performance of spectral sensors in various environments against various objects. An important part of this model development effort is its validation against actual data. For data to be used for rigorous model validation, it needs to be well ground truthed. Much of the existing IR field data of plumes lacks a sufficient amount of ground truthing to be useful for validation purposes. To alleviate this shortage, SITAC scientists conducted some small-scale plume experiments using a vehicle exhaust plume that could easily be repeated and could be well ground truthed. Also larger scale experiments were conducted at Waxahachie, TX. In addition to the model validation objective, SITAC sought to use the plume data to develop a model that would adequately predict plume temperatures, species concentrations, and the resultant plume emissions. Such a model could then be used to make future measurement programs more efficient.

2. SIMPLE PLUME GENERATOR EXPERIMENT

SITAC scientists embarked on a plan to produce small-scale plumes at minimal cost and complexity, but with sufficient supporting measurements to ensure their usefulness for model and algorithm validation. SITAC wanted to release some interesting materials into the plumes, but nothing hazardous or otherwise unsafe. For the initial experiments, SITAC chose readily available materials from a local hardware/lumber store. These included the store's brands of propane, methanol and acetone. SITAC needed a heat source to vaporize the methanol and acetone, so the plume emanating from a Ford ExcursionTM was used. The Ford ExcursionTM has a turbocharged 7.3 liter engine whose idle speed is computer controlled at 700 rpm. Measurements showed that at idle it produces about 1.55 moles of 346 degree K exhaust per second, so it has ample flow and heat content to serve as the plume source. The Ford exhaust plume contained no visible particles or other detectable contaminants.

To produce a plume generator, a length of expandable 4-inch diameter aluminum dryer vent hose was connected to the Ford ExcursionTM exhaust pipe. A 3x3cm opening was cut in the hose one meter from exit end for material injection.

Tests showed that this plume generator could fully vaporize modest amounts of methanol and acetone. SITAC measured the temperature and flow rate from the dryer vent pipe and found both quite constant over time and from day to day.

To inject liquid materials into the flow, a common air paint spray gun was used. The contents of the paint container were weighed before and after each timed run with a scale sensitive to 0.1g and this measurement was used to calculate the amount of vapor in the plume during any measurement. As the injected vapor cools the plume due to the heat of vaporization of the acetone or alcohol, SITAC measured the temperature of the plume during runs by using thermocouples at the pipe exit port, near the IR sensor, and at several points further down-plume.

To inject a gas like propane into the plume, an unlit single orifice burner attachment was inserted through the 3x3cm dryer vent opening. As with the other materials, the gas container was weighed before and after each timed run on a scale sensitive to 0.1g. This measurement was used to calculate the amount of propane in the plume.

The injection port was 1 meter from the plume exit point, which allowed time for vaporization of the spray droplets and for mixing of the plume. Various amounts of fluid/gas were injected into the plume. Each injection lasted for 30 seconds. The interferometer was commanded to scan at the start of the timed run. The actual scans occurred over seconds 14 through 30 of the run. The variation in the amount of liquid injected material for the various runs was achieved by turning a knob on the spray gun while holding the hand valve at full open. Changes in the amounts injected for each run were estimated initially by visual examination of the spray, but were quantified after the fact by the weight changes. With a little practice, it became fairly easy to produce plumes with relatively even steps of increasing gas concentration.

For the IR measurements, SITAC used its Designs & Prototype (D&P), Inc. MicroFTIR Model 101 Fourier Transform Spectrometer, running at 4 cm^{-1} spectral resolution over the spectral region from 3 to 18 microns. Measurements were made in accordance with the published SITAC protocols for using this sensor². Before each set of runs where vapors were injected into the plume, the D&P interferometer was calibrated with its attached warm and cold blackbody sources. This calibration was followed by measurement of the background through the Ford ExcursionTM plume with no material injected. Without moving the IR sensor, a few vapor injection runs were then made looking through the plume to the previously measured background. Right after the runs, another measurement was made looking through the plume while nothing was injected. This measurement was followed by a recalibration of the D&P with the blackbody sources. This sequence was repeated so that the measurements spanned several gases at various concentrations. In addition, at least once per day for each background, a measurement was made of the background while the Ford engine was not operating. The instrument was not moved while this sequence of measurements was being made.

The D&P spectrometer was positioned so that it looked either up or down through the plume 10 cm above or below the plume and 10 cm down the plume from the exit plane of the vent pipe. The instrument was never placed directly in the plume and its closeness to the vent exit plane assured that it was always looking through the plume. To further ensure that the D&P was always looking at the plume, the experiment was always run with the D&P positioned downwind of the plume exit port, though the winds on the measurement days were always less than 2.5 m/s.

Figures 1 and 2 show pictures of the plume generator in action. These include the general set-up, the material injection, the weighing, and a picture of the plume when a visible material (talcum powder) was injected.

3. FORD EXCURSIONTM PLUME EXPERIMENTS

3.1 Results with no Injected Material

Figure 3 shows five example spectra of the sky background during one of the run days. Four of these are taken through the Ford ExcursionTM plume and one was taken when the Ford ExcursionTM was not running. It is difficult to discern differences between the five spectra as there are essentially no 7.5 – 14.5 micron emissions from the Ford ExcursionTM plume, save for slight water emissions below 8 microns and some hot carbon dioxide emissions beyond 14 microns. The downward viewed blacktop background also showed no effects of the Ford ExcursionTM plume, other than those

same water and carbon dioxide features. The spectral region between 3 and 7.5 microns was also quite free of emissions except for those from water and carbon dioxide.

3.2 Acetone Plume Upward Viewing Results

Figure 4 shows the data from a typical set of upward viewing runs using acetone. As expected, when the concentration of material in the plume is increased, the radiance from the plume increases.

SITAC used the thermocouple temperature data and the amounts of acetone injected to calculate the expected radiance from these plumes. The calculations were based on the spectral absorbencies of acetone contained in the Hanst spectral library³. For the lower concentrations, excellent agreement was obtained. For the higher concentrations, where the plume was becoming optically thick, the calculated radiances exceeded the measurements. This discrepancy was not caused by transmission effects, which were accounted for in the calculation. Instead, it was determined that the heat of vaporization of the injected acetone was causing the problem by cooling the plume.

The effects of the heat of vaporization on the plume temperature led to increased emphasis on continuous monitoring of plume temperatures for subsequent runs. When the plume was monitored with thermocouples, data clearly showed the cooling effects of vaporization but also showed a lag in this effect due to heat transfer from the walls of the vent pipe. This lag was not predictable and necessitated the continuous temperature monitoring of all subsequent plumes. Using the thermocouple temperature data, much better agreement between the calculations and measurements was obtained. SITAC also made the decision not to attempt measurements using the Ford ExcursionTM for plumes experiments having densities over about 2000 ppmv-m of gases with similar boiling points and heats of vaporization properties as acetone and methanol.

Figure 5 shows an example of a calculation vs. experiment success at a high injection rate when continuous temperature measurements were being made. This plume has reached the point of being optically thick (herein defined as the point where transmission drops to 1/e). A few of the calculations and experimental did not agree, but usually for some explainable reason such as making a run too soon after an aborted very high concentration run which had left some residual liquid in the pipe.

3.3 Methanol Upward Viewing Results

SITAC made several measurements of methanol plumes. Figure 6 shows an assemblage of several of these runs while viewed in the upward direction. With few exceptions, the upward viewing methanol runs produced reasonable results. The plumes would cool with increasing concentration of gas, but they always remained warmer than the sky background.

3.4 Acetone and Methanol Downward Viewing Results

The downward view methanol and acetone measurements allowed both emissive and absorbing plumes to be studied. Runs were done on two days, one where the blacktop background was at a temperature of about 315K, and the other where it was about 290K. On the day where the ground was hotter, the plume vs. background temperature differential was about 25K whereas on the colder day, the differential was about 50K. On the day where the blacktop was hot, at the higher gas concentrations, the plume cooled due to the heat of vaporization of the gas and often would cool to a point where it was below the background temperature. This temperature difference would shift the gas plume from emission to absorption against the background. Figure 8 shows one of these cases. At lower concentrations, the methanol is, as expected, emitting above the background, at the highest concentration, the plume is cooled to the point where it is absorbing the background. Acetone produced similar results.

3.5 Ford ExcursionTM Plume Experiment Summary

Figure 8 plots the measured radiance versus plume concentration for all of the acetone and methanol Ford ExcursionTM plume experiments. On the left of the curves is the background with no injected gas. As gas is injected in increasing amounts, the curves show a definite increase above background. At still higher concentrations, the plume becomes

optically thick and the curves level off. This forms a curve that is mildly “S” shaped. The “S” shape of many of the plots is expected, as the signal from the gas rises above the noise, goes through a nearly linear radiance vs. concentration phase, and then, at higher concentrations flattens out due to increasing optical thickness. When the plume cools as a result of the heat of vaporization of so much injected material, the curves drop to background levels or below, due to absorption. Both curves are at a wavelength where emissions from their respective gas are high. For the methanol curve, the plotted wavelength is 9.48 microns, and it is 8.24 microns for acetone. To show the variations in the measurements for each run, all points are included except for cases where the vaporization was obviously incomplete and liquid poured out of the vent. Calculations of expected emissions/absorption generally agreed with these measurements.

4. WAXAHACHIE, TX EXPERIMENTS

To handle concentrations greater than those that could be injected into the Ford Excursion™ plume, SITAC utilized a portable plume generator constructed and operated by Aero Survey Inc. at a local airport near the town of Waxahachie, TX. This generator is pictured in figure 9, which also shows the general experimental set-up, the D&P Fourier Transform Infrared spectrometer, and the plume made visible during a polyethylene glycol release. On the top of the crane in this figure is a Bomem FTIR spectrometer; the D&P is next to a person standing beneath the Bomem. An array of thermocouples mounted on a frame of PVC pipe can be seen between the generator and the propane supply tank. A weather station is next to the propane tank. The thermocouple array had 25 thermocouples spaced on a 40 cm grid and then the array was duplicated 1 and 3 meters distant. This 2x2x4 meter grid was very useful in obtaining plume temperatures and in aiming the spectrometers and when SITAC acquires the reduced data, it should produce interesting real time thermal “pictures” of the plume.

Several gases were released at Waxahachie, some by themselves and some in combination. These included ethanol, acetone, 1,1,1,2 tetrafluoroethane (F-134A), and polyethylene glycol (to make a visible plume). In addition, ethanol was released in conjunction with water to produce a plume where the water would condense as is typical for moist plumes in cold environments. Pending additional measurements of this plume generator, the species concentration units for all releases at Waxahachie will be given herein as milliliters (ml) of liquid per minute or for gases in units of liters (l) per minute. SITAC has the approximate conversion factors for going to ppmv-m, but they are too uncertain to use without additional measurements.

Ethanol was used for most of the plume experiments, as it is an inexpensive, readily available and safe material. The ethanol was denatured with a small amount of heptane and also had some small amount of allotropic water. Infrared measurements were made of the ethanol plume at four different distances from the stack and at many different ethanol concentrations.

4.1 Waxahachie Ethanol Results

At Waxahachie many plumes were made at various distances from the stack and at many different gas concentrations. To give the reader an idea of the number of measurements, all the upward viewing ethanol runs at 1m from the stack are presented in figure 10. This chart also includes the before and after sky background spectra. The temperature differential between the sky background and the plume at this 1m distance is about 65 degrees K. As the ambient wind varies in direction and speed, variations are seen from run to run even when all else is held constant. For this reason, SITAC has been working to develop a means to obtain plume temperatures from the carbon dioxide emissions from the plume itself⁴.

In addition to measurements at 1m from the stack, measurements were also made viewing at distances of 0.25m, 0.5m, 2m, and 4m. These measurements produced spectra similar to figure 10, when adjusted for ethanol concentration and temperature differential between the background and plume.

Figure 11 shows the approximate variation of the differential between the plume temperature and the sky background for these positions as measured by the thermocouple grid. The plume radiance varied in proportion to these temperatures and the species concentration.

Figure 12 summarizes the results for all the ethanol runs. The radiance plots vs. concentrations weakly show the same “s” curve shape as seen with the vehicle plume. After the curve rises above background, there is a moderately linear increase with concentration, followed by a leveling off as the plume becomes thick. The various inflection points vary, as expected, with the plume to background temperature differential. These curves include all data points and often include cases where a wind gust may have blown the plume out of or off to the side of the field of view of the sensor. For some of the points this has been verified by noting the changes in the radiance of the carbon dioxide emissions at 4.2 microns.

4.2 Other Gases

As previously stated several other gases were released at Waxahachie. Figure 13 shows an interesting result of such a release where two gases were released concurrently in order to test the ability of a processing algorithm to separate the gases and to quantify their relative release rates. Of note in this figure is the washing out of the fine spectral features that should have been seen at 4 cm^{-1} (from library spectral at that resolution). This may be caused by interaction between the gases or between the gases and the water vapor in the plume.

4.3 Steam Plume Measurements

The last series of measurements were of ethanol runs where enough water was added to the plume to make it condensing. These measurements are significant because many real world plumes are condensing. Figure 14 shows three steam ethanol measurements along with four ethanol measurements in non-condensing plumes, at two different plume to background temperature differentials. All these ethanol runs were at a flow rate of 200 ml/min. As expected, the two measurements at ~ 60 and two at ~ 85 degrees K temperature differential (plume to background) produce clearly detectable gas plumes, well above threshold. One of the steam plume measurements at 200 ml/min produces a slight signal above background, the other two were totally in the background. Clearly the water droplets in the condensing plume interfere with the ability of the spectrometer to detect molecular species. The temperature of the steam plume was not measured in these three cases, but the outer surface of the steam plume was at least a few degrees above the sky background, based on the full 3-18 micron radiance curves. The core of the steam plume was probably much hotter, but the liquid water in the condensate masks all but a relatively thin layer of plume.

5. CONCLUSIONS AND LESSONS LEARNED

After each set of plume measurements, SITAC examined the data and generated ideas for some additional measurements. With small-scale experiments such additions were quite easy and inexpensive to implement. After the first set of measurements wherein the plume was significantly cooled by the higher gas concentrations, it was realized that extensive temperature measurements are needed to understand and model the plume data. Plume velocity measurements downwind from the stack were also needed and will soon be implemented. When extensive temperature measurements at a spatial resolution of about 0.4m were made in a plume, it was quickly realized that the thermocouple grid should have been even denser. Response times of the thermocouples need to be carefully measured as plumes can be blown around by the wind in very short time scales. To understand the entrainment phase of a plume in the first one or two stack diameters downwind, higher spatial resolution sensing is needed. The plume cooled by over a hundred degrees in only one stack diameter distance ($\sim 40\text{ cm}$) at Waxahachie. To address this rapid cooling question, measurements will be made with a higher resolution thermocouple grid and, if possible, with MWIR radiometers to document the carbon dioxide temperature profiles in the plume.

These preliminary experiments and analysis yielded some significant new findings, and demonstrated the soundness of SITAC's approach to plume research. The steam plume measurements are quite significant as real world plumes are often condensing and thus may present difficulties for species detection using IR spectral techniques. SITAC has produced some relatively robust data of the IR emissions of gases in plumes and has been able to generally match these measurements to calculations based on library data. SITAC has also developed a means to visually map plumes by injecting an inert material into the plume and then photographing the plume. Preliminary results show the value of simultaneous spectroscopic and thermocouple measurements. Combining photography and thermocouple measurements

provides unprecedented capability to study plume physics in a rigorous manner. SITAC is using this synergistic approach to study plume dynamics, and to compare verified plume behavior with theoretical work dating back to the 50's⁵. Evidence that the plume gas temperatures can be measured directly from the IR is discussed in a companion paper⁴.

Future measurements will be directed at filling in the gaps that have been discovered in the existing data as well as continuing to verify calculations of minimum detectable quantities for gases as a function of the gas/background temperature difference, background complexity, atmospheric path variations, and initial gas temperature. Future measurements will include some means to define plume boundaries as well as developing techniques to map these boundaries remotely.

ACKNOWLEDGEMENTS

The authors would like to thank the Central MASINT Organization, its Technology Coordination Office, SITAC, Los Alamos National Laboratory, and the Environmental Protection Agency (EPA) Region-7 for the sponsoring and support of these collections. These efforts particularly benefited from the efforts and dedication of Mark Thomas of the EPA, participants from AeroSurvey, LANL, Airborne Imaging, McKinzie Environmental, Rochester Institute of Technology, and ABB Bomem. Craig Miller and Joe Leckie from SITAC provided able assistance with the experimental setup and measurements.

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Ford Excursion™ Plume Experiments

General Experiment Set-Up

D&P Spectrometer Down View

30 sec. Of Gas Injection

Before/After Weighing

Figure 1: Photographs of the Ford Excursion™ Plume Experiments.

Ford Excursion™ Plume Experiments

View of Injection into
the Stack
Note: No Visibly Detectable
Aerosols in nor Liquid
Fraction

View of Plume during
Injection of ~2 micron
Particles – Ruler has a
2m scale

Figure 2: Ford Excursion™ plume injection photographs.

Figure 3: Signatures of the background during the Ford Excursion™ plume experiments.

Figure 4: Signature of the sky background, looking through an acetone plume during the Ford Excursion™ plume experiments.

Figure 5: Signatures of measured and calculated acetone.

Figure 6: Signatures of the sky measured through the alcohol plume during the Ford Excursion™ plume experiments.

Figure 7: Signature of the methanol plume against an earth background.

Figure 8: Summary of Ford Excursion™ Plume Experiments.

Waxahachie, TX Plume Experiments

Plume Generator

General Experimental Set-Up

D&P FTIR Spectrometer

Visible Plume Run

Figure 9: Waxahachie Plume Experiment Photographs.

Figure 10: Signature of Ethanol measurements.

Figure 11: Approximate temperatures at Waxahachie.

Figure 12: Ethanol Radiance vs Concentration

13

14

Figure 13: Ethanol and acetone measurements.

Figure 14: Effects of condensing plume.