TECHNICAL REPORT NO. LWL-CR-05C70A

ACQUISITION OF SMALL NO_2 SOURCES WITH A UV CORRELATION SPECTROMETER

Final Report

By

Hugh T. Reilly
U.S. Army Land Warfare Laboratory
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Gilbert Newcomb
Environmental Measurements Inc.
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ABSTRACT

The feasibility of remotely sensing NO_2 emissions from a single source was explored using a UV Correlation Spectrometer manufactured by Barringer Research Ltd., Toronto, Canada. Detections were made of a 2.5 ton truck exhaust at 200 meters range and a 30 KW diesel generator at 1050 meters range to demonstrate feasibility of detecting small sources of NO_2.

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Introduction

The capability of a UV correlation spectrometer to detect SO_2 and NO_2 emissions in the atmosphere from small sources was explored under contract with Barringer Research Ltd, Rexdale, Ontario, Canada. The effort was reported in Technical Report NO. LWL-CR-05C70 "Spectrometers Sensor Investigation", dated April 1971 (AD 884736). Since that time, improvements in the spectrometer have resulted in an approximately five fold increase in sensitivity to NO_2 in particular.

This report covers efforts to determine the effect of the improved sensitivity on the capability to detect NO_2 emissions from the exhausts of internal combustion engines. The field measurements were by Environmental Measurements Inc. of San Francisco, California under Contract DAAD05-73-M-9332 with a Barringer Research Ltd, COSPEC IV instrument at Aberdeen Proving Ground, MD in tests conducted and supported by the U.S. Army Land Warfare Laboratory.

Test Program

Three targets were available for investigation. These were:

1 - Truck 2½ ton 6x6 M35A2
1 - Helicopter UH-1
1 - 30 KW portable motor/generator diesel powered with a 20 KW resistive load

The NO_2 gases produced by the target exhausts were sought in this investigation. NO_2 is not a direct exhaust product in significant quantities i.e. (greater than 10 ppm). The NO_x products, primarily NO, must oxidize to NO_2 by action of oxygen present in the air to establish concentration levels sufficient for reliable detection. To realize such detection levels from a single vehicle source requires the engines to be operating under load and a low wind velocity. This would permit a maximum amount of NO_x production and a minimum of plume dilution during the process of oxidation from NO_x to NO_2.

The COSPEC IV instrument used during this study period was considerably improved by comparison to the COSPEC III used in the 1970 study. The significant area of improvement was in the optical section of the system where greater light throughput due to improved optical coatings resulted in a lower noise level and therefore a higher short term sensitivity.

Field Test Results

The first meaningful results were obtained when the 30 KW generator was loaded with a 20 KW load. The loading was thermally regulated by a sensor within the load unit. When the load temperature reached the critical point, the load would automatically disconnect from the generator. On the afternoon of June 15 the air temperature was in the mid to high 80's with very slight wind velocity. This condition caused frequent cycling of the load

resulting in a modulation of the generator fuel consumption rate and resulting NO_2 emission levels. By maintaining radio contact with an observer at the generator site who called out each loading cycle and by observing the COSPEC output variations, one could correlate the load cycling to detected NO_2 level variations. By this method the following approximate levels were detected at the ranges indicated. Best results were obtained in all cases by observing at a point approximately two meters directly down the line of exhaust emission flow.

Table I

30 KW generator with 20 KW loading

Range (meters)	Approximate Signal Level (ppm-m)
225	7.5
510	3.0
1050	1.8

Oil was injected into the exhaust of the generator to produce a visible plume. By this means it was determined that the plume depth at the point of observation was approximately 1 to 1.5 meters.

In all of the field testing the direct viewing cassegrain telescope was used. A single entrance slit was also used. This combination provided the minimum field of view of 10 milliradians by 3 milliradians with adequate power input to maintain a noise level of approximately 1 ppm-m with an integration time of 4 seconds. When attempting to detect a gaseous trace element the target would ideally fill the full field of view. This means that at a range of 1000 meters the minimum target would have to cover an area approximately 10 meters by three meters and be properly sighted to cover the complete field of view. Targets which occupy less than the full field of view are linearly attenuated by the ratio of available to occupied field of view. This fact is quite apparent in that an emission source consisting of one exhaust port from one vehicle was very quickly attentuated in linear fashion to the limit of detectability in a relatively short range. The loaded generator concentration pathlength product that was detected decreased with increasing range in approximately linear fashion.

Table 2

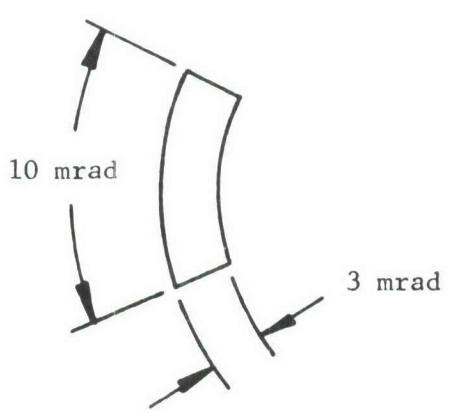


Table 2 (Continued)

Range (meters)	Signal Level (measured)	% Field Obscured	Signal at Full View
225	7.5	30%	25
510	3.0	20%	15
1050	1.8	10%	18

Many attempts were made to detect the 2½ ton truck at ranges from 200 to 500 meters. The following methods were applied as aides in detectability.

The target was positioned with the engine off; then vertical scans were made at equal increments of azimuth over the region of interest which spanned a horizontal sector of 45° and a vertical sector of approximately ± 10° about the target. Likewise horizontal scans were made over the 45° arc covering the 20° vertical range of interest. This exercise defined the background character very nicely.

The vehicle engine was started and allowed to operate at idle speed. All scans, both vertical and horizontal, were repeated. The results obtained were compared against the "engine off" scans made earlier. The logic of this method was to define any existing NO_2 by making it visible as an anomaly which would be apparent as a change in scan character. When an anomaly could not be detected the engine speed was increased to a high speed no load condition. Azimuth scans were repeated and compared against the "engine off" condition. No NO_2 was detected.

The scans which were the result of identifying the background characteristic produced variations in COSPEC output which were equivalent to 20 ppm-m peak to peak for a typical scan. This magnitude of variation made it very difficult to observe the very low levels which were available from the truck. The reason for this large variation in COSPEC output is due in large part to the fact that the viewing line was at a shallow vertical angle to the horizontal, an angle of 1 to 2 degrees. With this view a large color variation gradient was present which introduced non-linear spectral disturbances causing the 20 ppm-m variation in output.

This condition would be greatly minimized if the viewing angle to the plane of interest was near 90° or if the background contained less gradient. The ideal viewing angle would occur if one were viewing from a helicopter or other platform which allowed a direct view of the ground or if the background was the sky, obtained by sighting on a target up hill of the sensor.

Summary and Conclusions

It is concluded from the results of this field study that the feasibility of detection by remote sensing has again been clearly established. The parameters which define the limitation for distance ranging were experienced and recorded.

Having detected the feeble NO_2 emissions from a single source at a range of 1050 meters has established the instrument as an extremely sensitive tool. The primary considerations which define the ranging limit are:

1. The ratio of the anticipated source concentration and line of sight depth; i.e., source concentration path length product (CL), to the detection system noise level (N) over the time interval of interest, CL/N.

2. The ratio of target size (A) to field of view (FOV) which varies linearly as a function of range, (A/FOV) f(R).

By defining a minimum S/N ratio, one can compute the ranging limit by solving the following equation for range;

$$S/N = CL/N\ (A/FOV)\ f(R)$$

Where the ratio of A/FOV is inversely linear with range and FOV = viewing angle height x viewing angle width (in radians) x range.

It remains now to develop application techniques and to choose gaseous targets most ideally suited for this detection method and to then optimize a COSPEC system for this purpose.

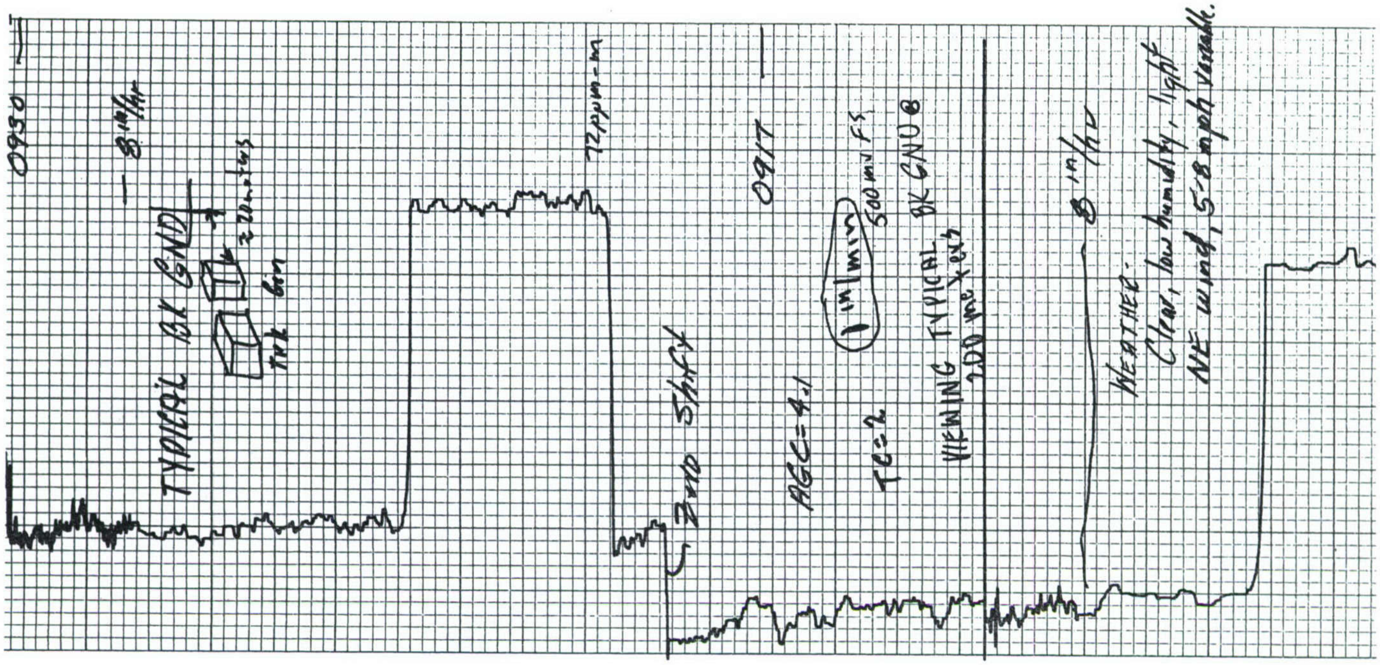
The data obtained by the reliable sighting of the load modulated generator can be extrapolated for any desired target considering the target concentration path length product and distance from the sensor. By this means the sensitivity of detection of the target can be estimated.

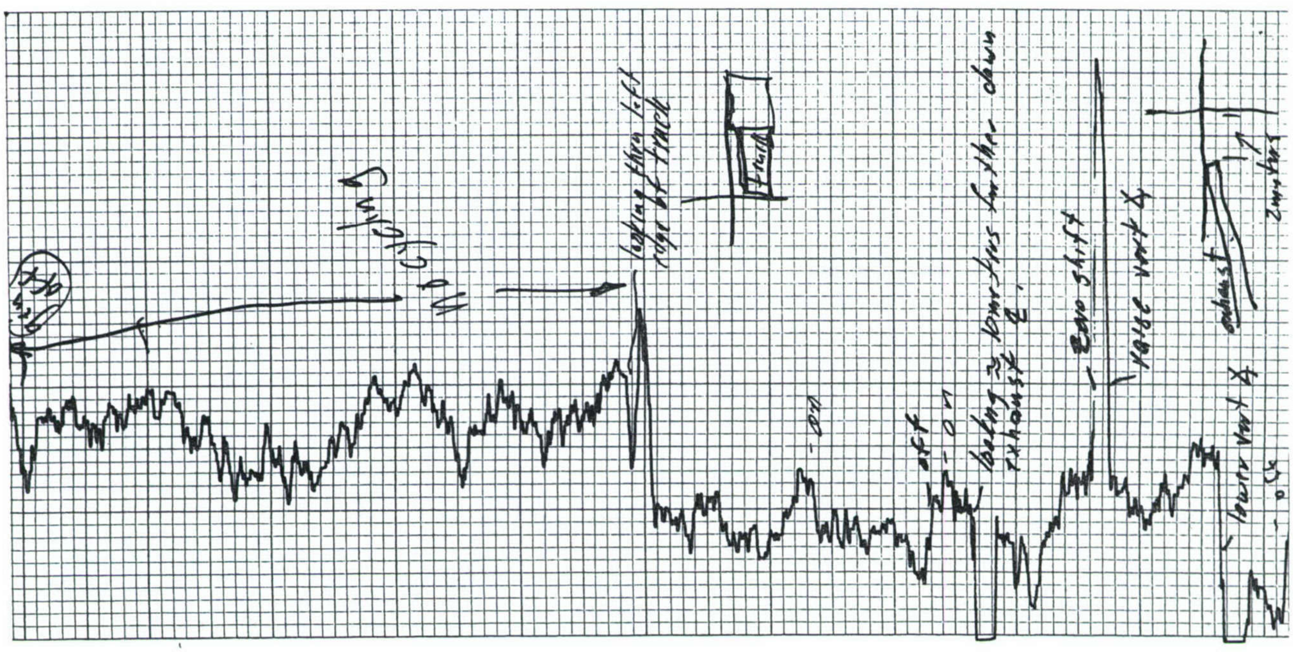
APPENDIX A

FIELD RECORDS OF SIGHTINGS

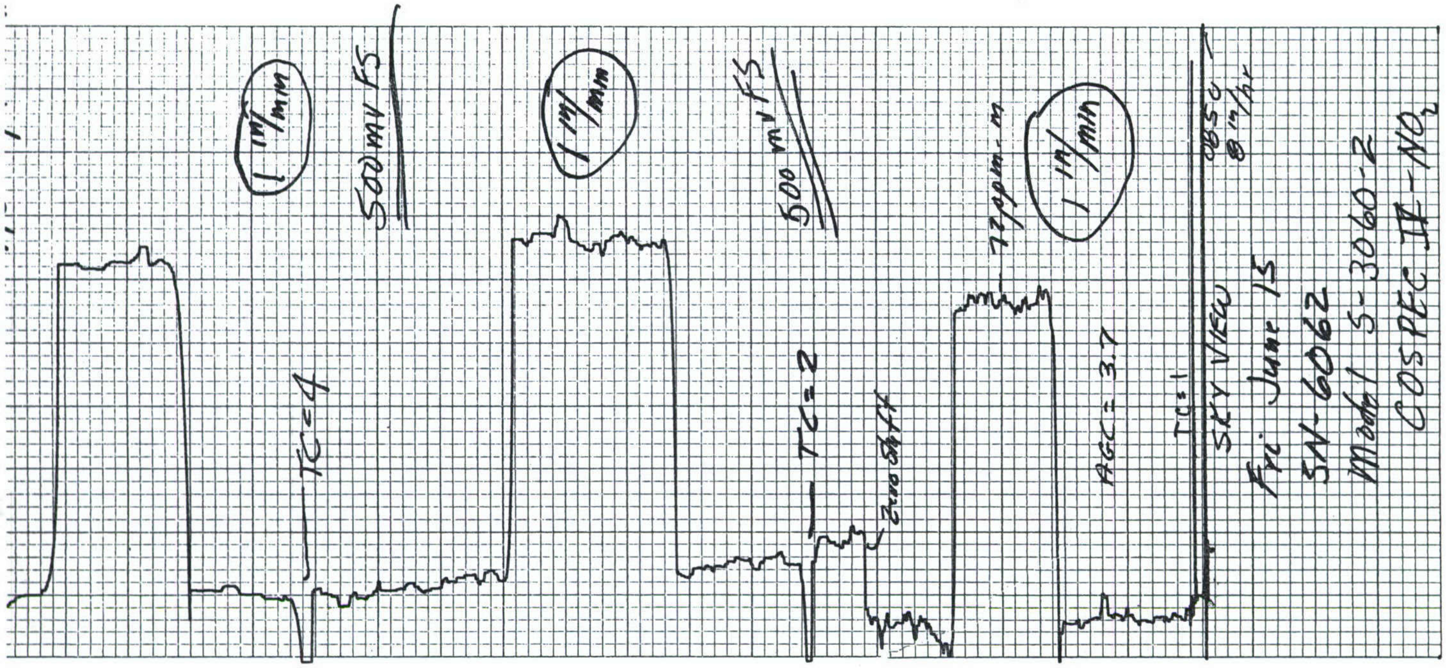
The following pages are arranged consecutively as increasing time during the field day of June 15. These records contain the sightings of the loaded generator at ranges of 225, 510 and 1050 meters as well as the direct view of the exhaust emissions during one stop, start, stop cycle of the generator when the wind direction was directly from generator to COSPEC IV.

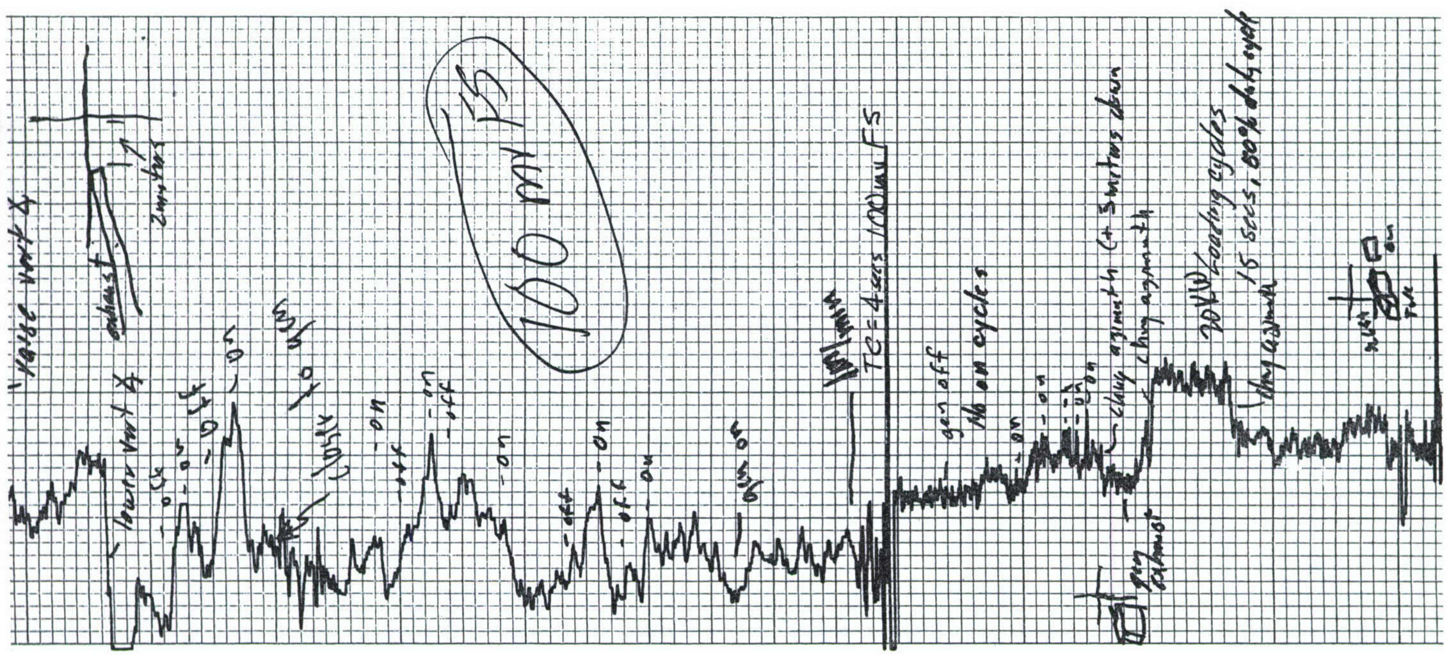
Sensor	COSPEC IV, SN 6062
Range	225, 510, 1050 meters
Winds	Light from approximately 270°
Sky	Clear
Target	30 KW diesel powered generator with 20 KW thermally disconnected loading.

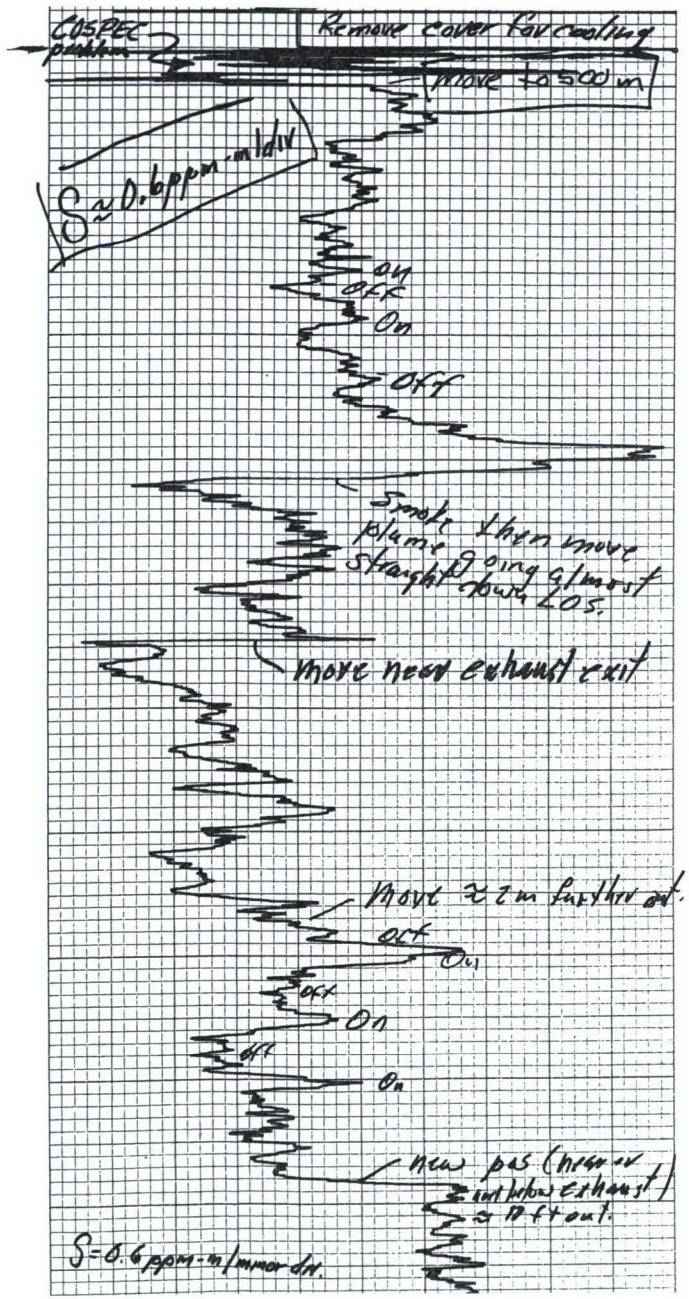

0930
TYPICAL BK GND
72 ppm-m
0917
Zero Shift
AGC = 4:1
TC = 2
1 in/min
500 mV F.S.
VIEWING TYPICAL BK GND
WEATHER:
Clear, low humidity, light
NE wind, 5-8 mph variable.

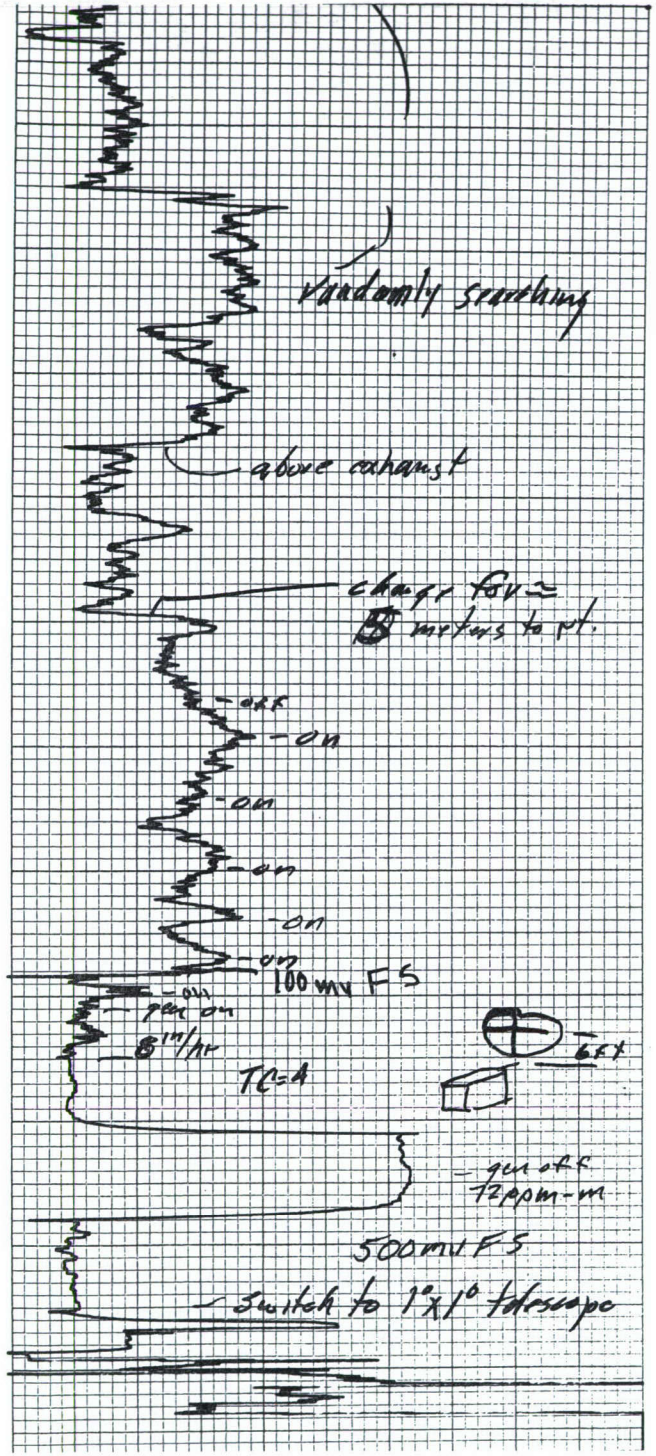

Zero shift
raise vert ∡
lower vert ∡

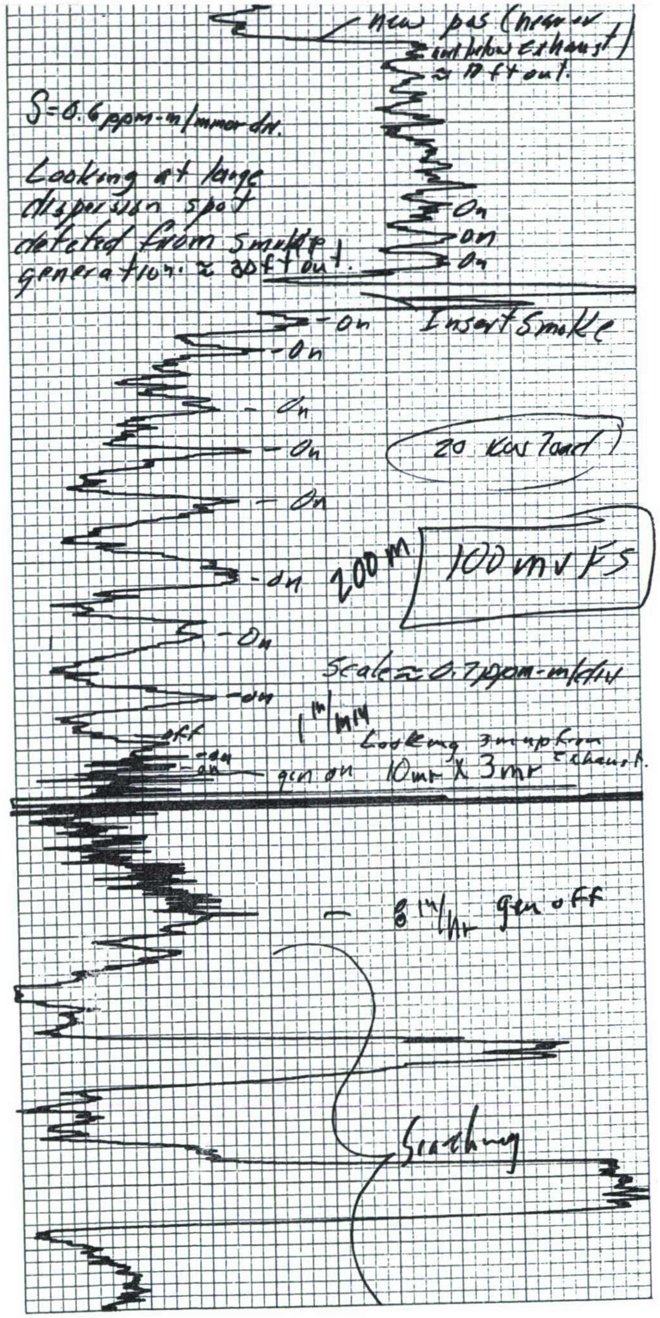
2

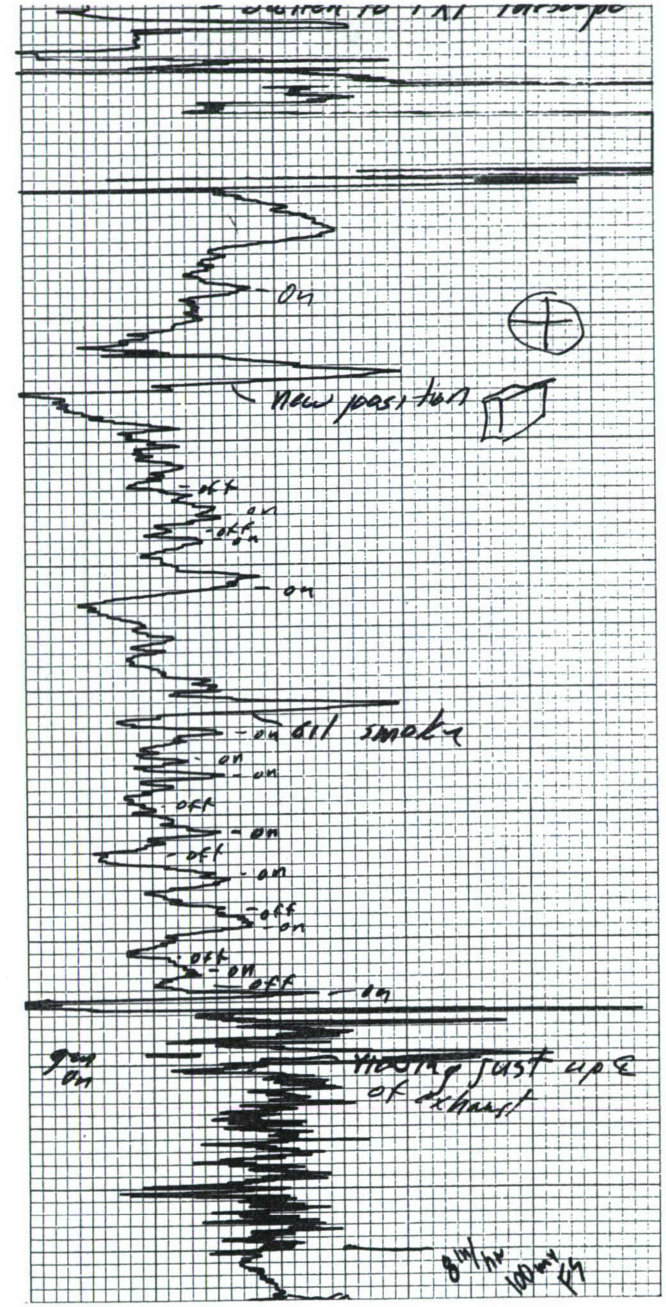

1 in/mm
500 mv FS
TC=4
1 in/mm
500 mv FS
TC=2
Zero Shift
72 ppm-m
1 in/min
AGC = 3.7
TC=1
SKY VIEW
OBSC
8 in/hr
Fri June 15
SN-6062
Model 5-3060-2
COSPEC II-NO2

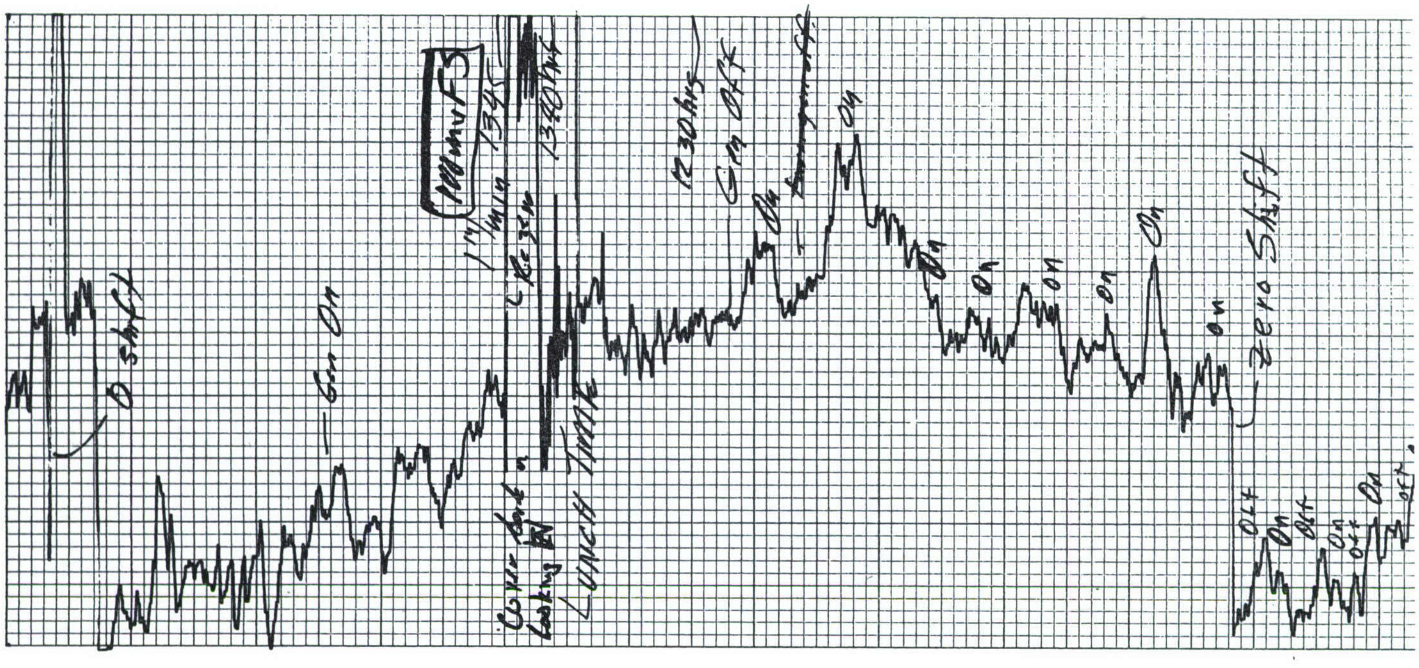

raise vert ∡
lower vert ∡
exhaust
2 meters
- off
- on
- off
- on
closer to gen
- on
- off
- on
- off
- on
- off
- on
- off
- on
gen on
1 in/min
TC = 4 secs 100 mv FS
100 mv FS
gen off
No on cycles
- on
- on
chng azimuth (+ 5 meters down
chng azimuth
gen exhaust
20 KW loading cycles
15 secs, 80% duty cycle
chng azimuth


Cospec position
Remove cover for cooling
move to 500 m
S ≈ 0.6 ppm-m/div
on
off
On
off
Smoke then move plume straight going almost down LOS.
more near exhaust exit
Move ≈ 2 cm further out.
off
On
off
On
off
new pos (near or just below exhaust) ≈ 10 ft out.
S=0.6 ppm-m/minor div.

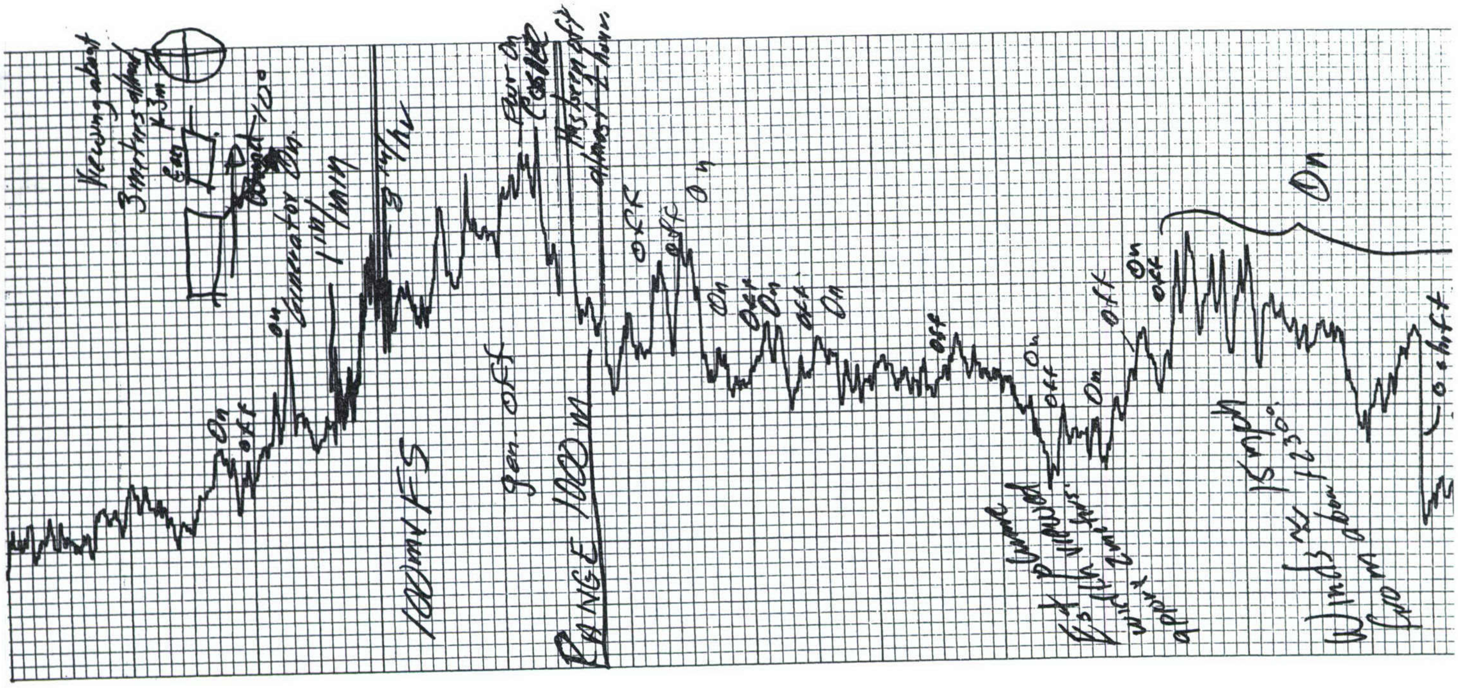

randomly searching
above exhaust
change for ≈ 5 meters to pt.
off
on
on
on
on
on
100 mv FS
gen on
8 in/hr
TC=4
ext
gen off
12 ppm-m
500 mv FS
Switch to 1°x1° telescope


new pos (nearer outlet below exhaust) ≈ 10 ft out.
S=0.6 ppm-m/mm or div.
Looking at large dispersion spot detected from smoke generation ≈ 20 ft out.
On
on
On
Insert Smoke
On
On
On
On
On
on
200m
100 mV FS
On
Scale ≈ 0.7 ppm-m/div
on
1 in/min
Looking 3 m up from exhaust.
off
on
on
gen on 10 mr X 3 mr
8 in/hr gen off
Scatching

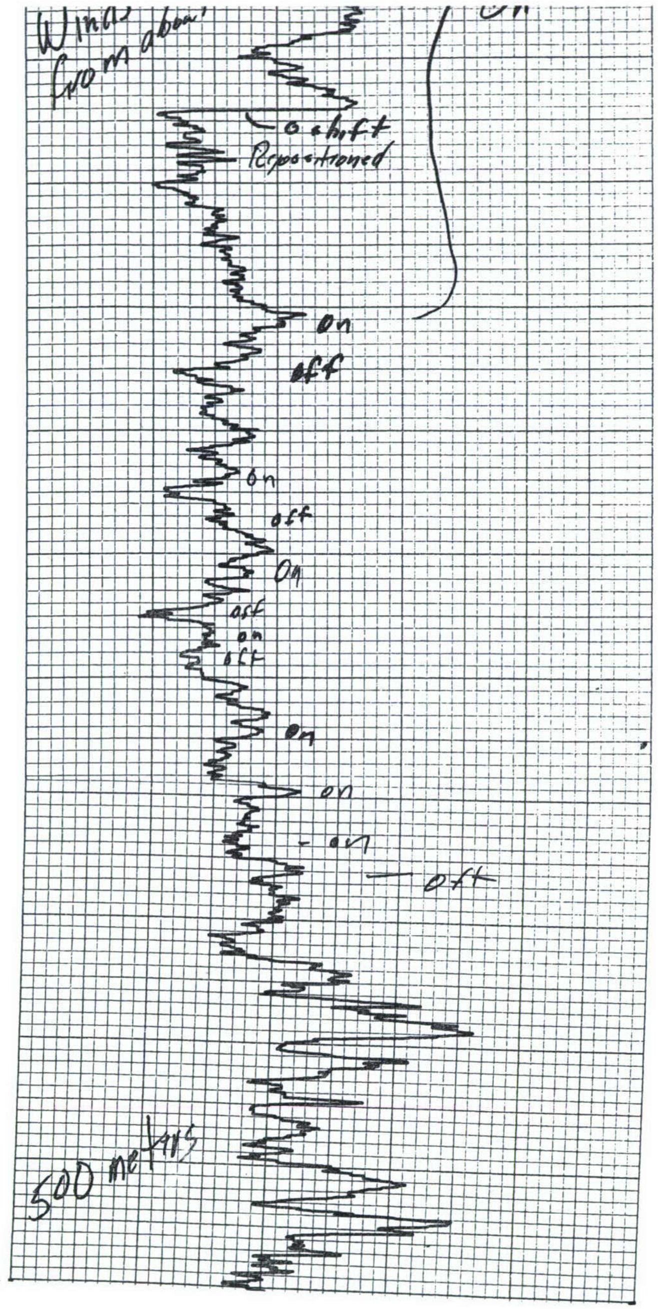

On
new position
off
on
off
on
on
on oil smoke
on
on
off
on
off
on
off
on
off
on
off
on
gen on
just up of exhaust
8 in/hr
100 mV FS

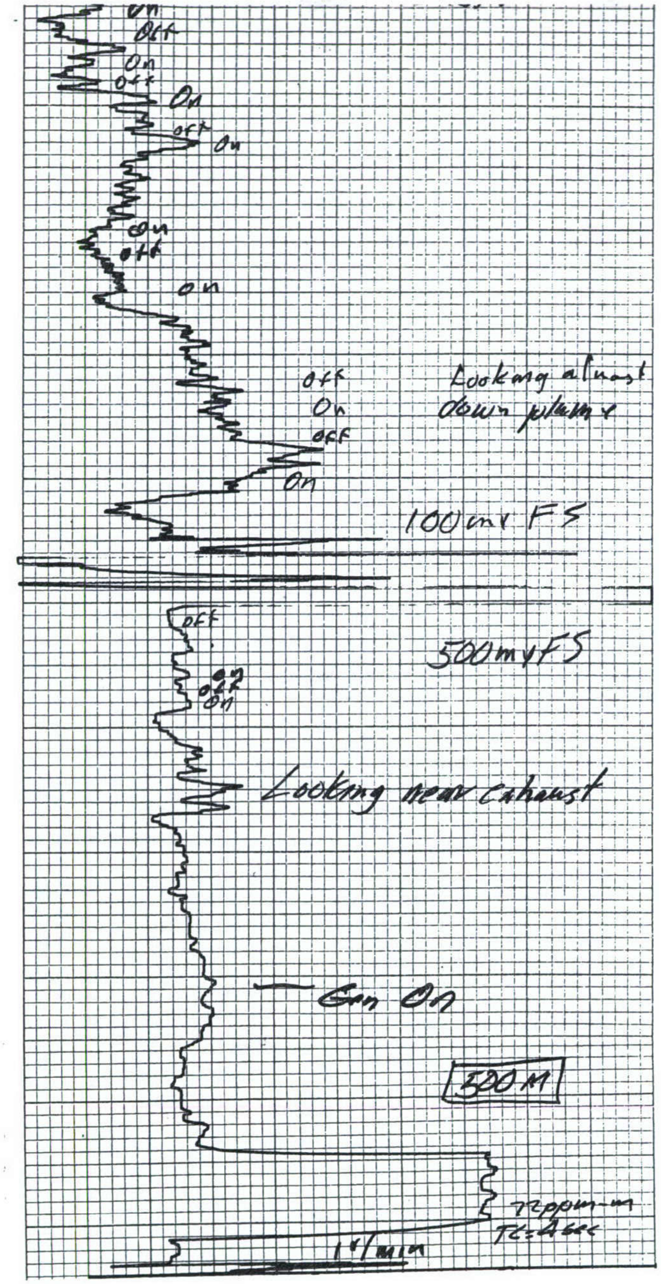


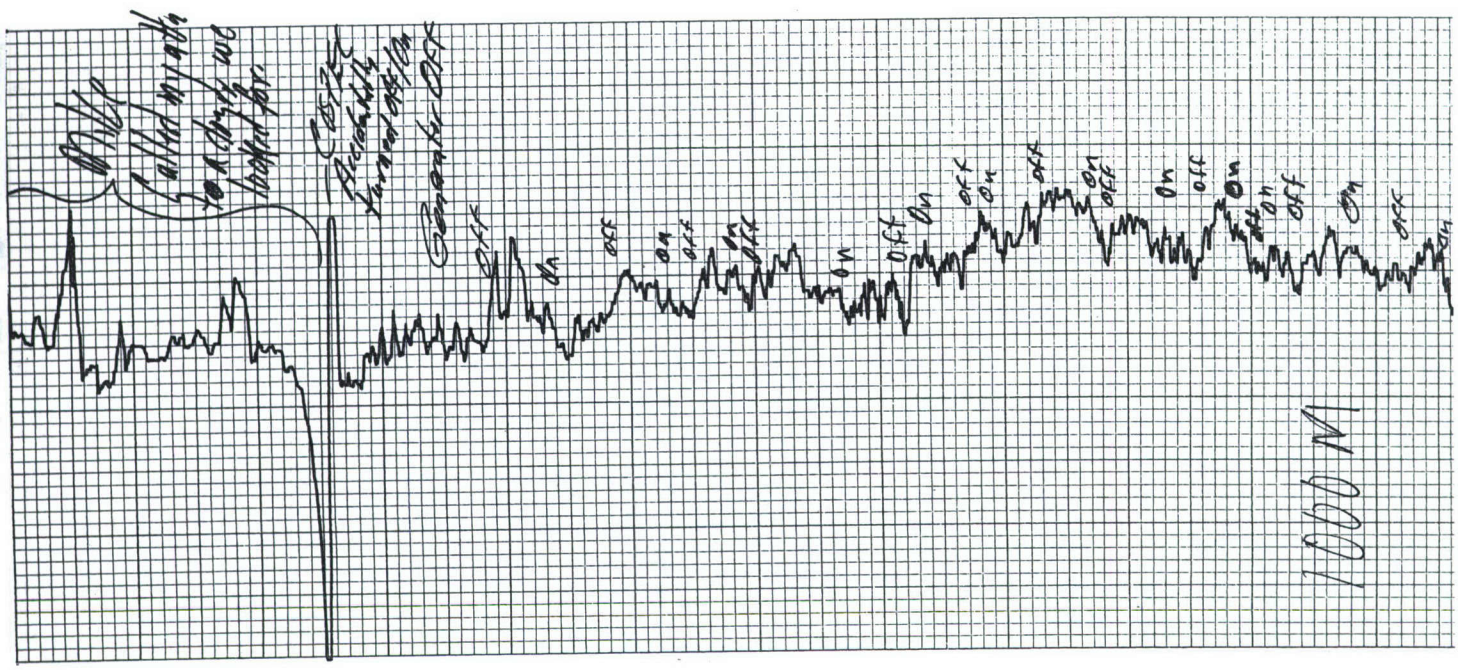
0 shift
Gen On
LUNCH TIME
Gen Off
Zero Shift

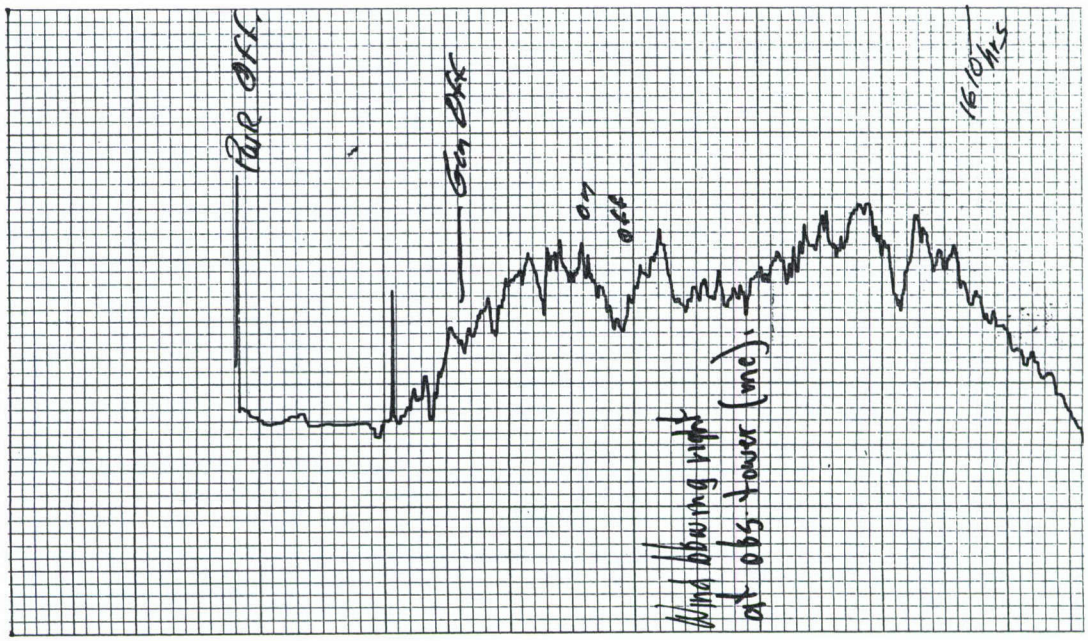


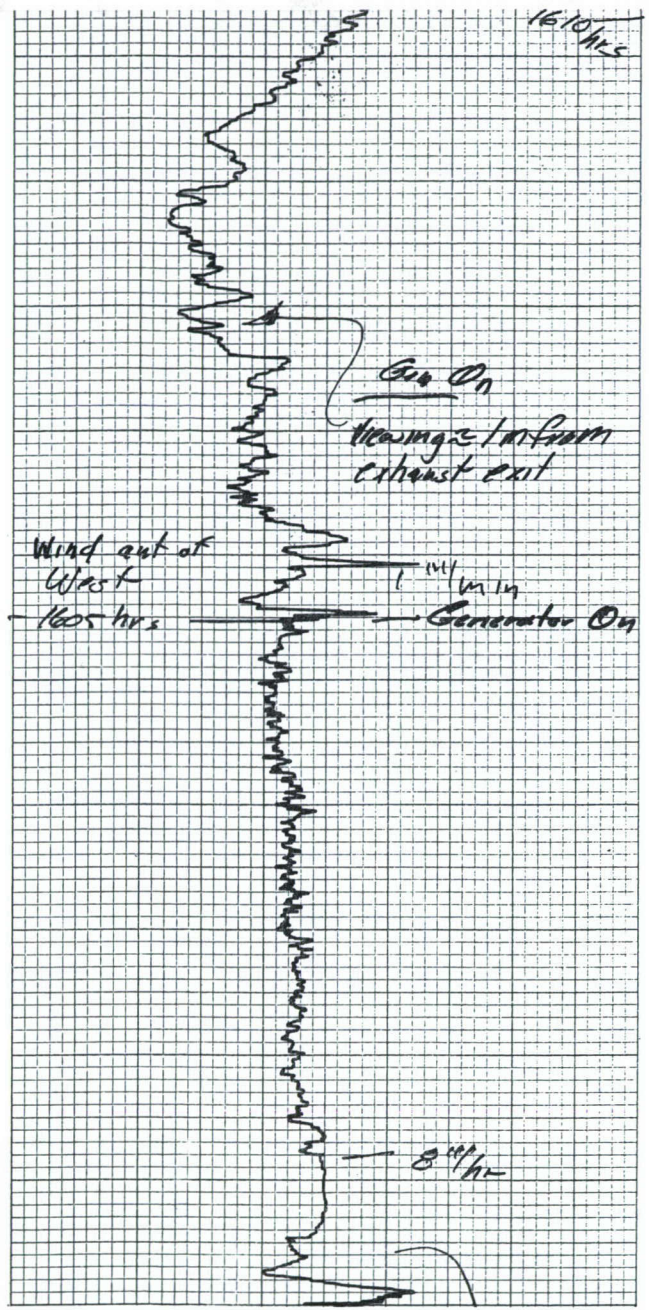
100mi FS
Gen Off
RANGE 1000m
0 shift

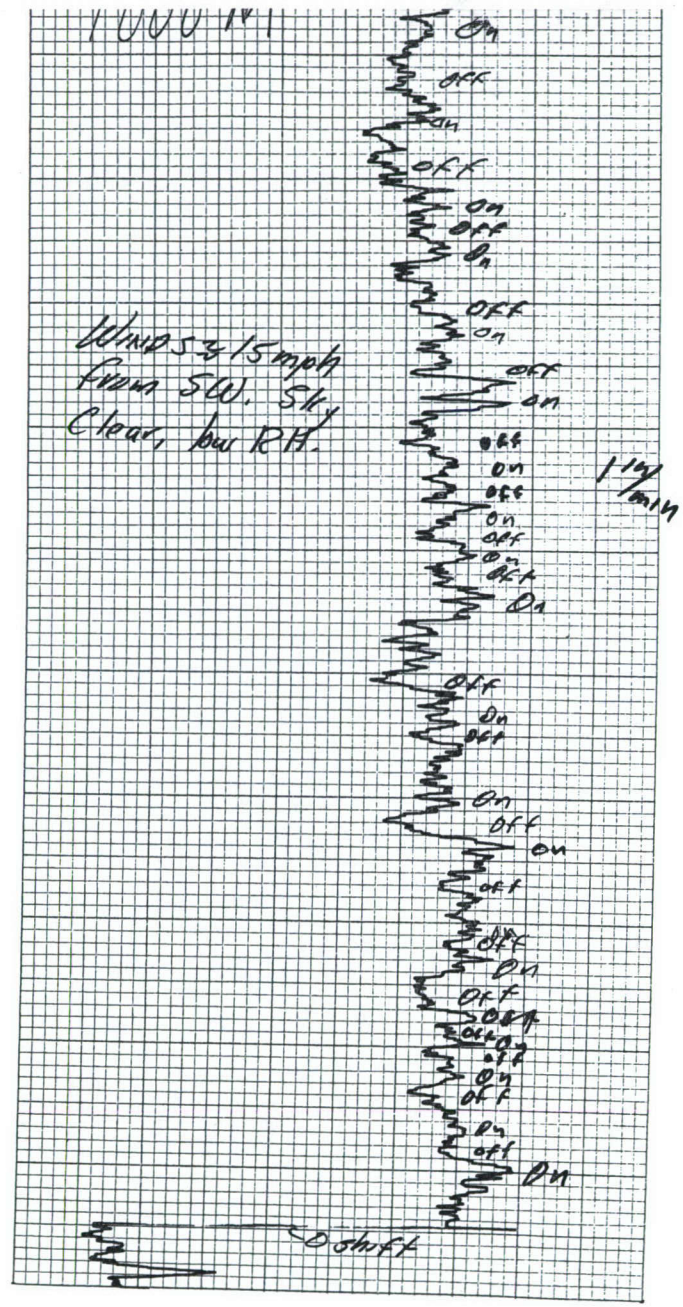

Winds from about
0 shift
Repositioned
On
off
On
off
On
off
on
off
On
on
- on
— off
500 meters


On
Off
On
Off
On
off
On
On
off
On
Off
On
Off
On
Looking almost down plume
100mv FS
off
500mvFS
Looking near exhaust
— Gen On
300 M
1"/min


1000 M


Gun off
16/10/45


1610 hrs
Gen On
Measuring Inst from exhaust exit
Wind out of West 1605 hrs
1 mi/min
Generator On
8 mi/hr


1000 hrs
Wind 5-7 15 mph from SW. Sky Clear, low RH.
On
Off
0 shift
1 mi/min

Appendix B

COSPEC IV

General Description:

The Barringer Correlation Spectrometer type COSPEC IV Model No. PJ308-1-0 is a portable remote sensing instrument intended for the quantitative measurement of sulfur dioxide or nitrogen dioxide (SO_2 or NO_2) in an optical path between a suitable source of ultraviolet (UV) radiant energy. The sensor is designed for maximum versatility in the remote measurement of gas clouds in the atmosphere and is suitable for a fixed ground station, ground mobile, or airborne installation. Model PJ308-1-0 differs from Model PJ308-0-0 in that it is designed for measurement of NO_2 as an alternative to SO_2. The user may convert from one gas to another by means of interchangeable components. Model PJ308-0-0 is suitable for SO_2 only.

The instrument consists of two telescopes to collect light from a distant source, selectable multiple-slit entrance masks, a grating spectrometer for dispersion of the incoming light, a disc-shaped multiple-slit exit mask and an electronics system. The disc correlator functions as a high contrast reference spectrum for matching against the incoming absorption spectra and is comprised of arrays of circular slits photo-etched in aluminum on quartz. The slit arrays are designed to correlate sequentially in positive and negative sense with absorption bands of the target gas by rotation of the disc in the exit plane. The light modulations so produced are detected by a photomultiplier tube and the resulting signals processed in the electronics to produce a voltage output which is proportional to the optical depth or burden (ppm-meters) of the gas cloud under observation. The system automatically compensates for changes in average light intensity.

COSPEC IV is designed to utilize the ultraviolet portion of natural diffused sunlight where well defined banded structure of SO_2 absorption occurs namely the 3,000 A to 3,160A region. For NO_2, operation is in the blue-visible region between 4250A and 4500A.

Applications:

COSPEC IV may be used for monitoring the vertical burden of SO_2 or NO_2 in the atmosphere. When located at a fixed monitoring station and the line of sight directed at the zenith sky, the output represents variations with time of the total amount of gas in the atmosphere overhead. When mounted in a moving vehicle the output is a measure of the spatial distribution of SO_2 across the traverse line. This enables the total production of SO_2 within bounded areas, eg.g industrial complexes, towns, cities, etc., to be determined. When the dimensions of the gas cloud are known the average concentration can be determined by simply dividing the sensor reading in ppm-meters by the optical pathlength through the cloud. For stack effluent measurements the stack diameter is known and remote measurements of the plume at the stack exit can be readily reduced to gas concentrations in the stack gas.

Mounting the remote sensor in a helicopter provides greater mobility and versatility of remote measurements. High stacks may be monitored with greater ease and accuracy than from ground positions and plume dispersion studies may be conducted in three dimensions.

Installed in a fixed wing aircraft the sensor can be used to provide near-synoptic contours of pollutant distribution and the mass transport of pollutants within air sheds and across regional boundaries.

Specifications:

Model:	COSPEC IV Model No. PJ308-0-0	COSPEC IV Model No. PJ308-1-0
Target Gas:	Sulfur Dioxide (SO_2)	SO_2 and NO_2
Light Source:	Diffused solar illumination	Same
Optics:	Two cassegrain telescopes on front turret selectable 1° x 1° or 3mR x 10mR. A right angle mirror attachement is provided for vertical look operation. A sighting telescope is provided for alignment on the target plume or light source. Same both models.	

	Model PJ308-0-0	Model PJ308-1-0
Light Source:	Diffused solar illumination	Same
Optics:	Two cassegrain telescopes on front turret selectable 1° x 1° or 3mR x 10mR. A right angle mirror attachment is provided for vertical look operation. A sighting telescope is provided for alignment on the target plume or light source. Same both models.	
Dyamic Range:	Low concentration 1 - 1000 ppm-m SO_2	Low concentration only 1 - 1000 ppm-m NO_2
	High concentration* 100 - 10,000 ppm-m SO_2	1 - 1000 ppm-m SO_2

* High concentration correlator for SO_2 available as an option

Sensitivity:	Low concentration: 3 ppm-m for 1:1 S/N ratio, 8 sec. integration time and for cloudless sky with solar altitude greater than 10 degrees. High concentrations: 100 ppm-m with 8 sec. integration time and same sky conditions as for low concentration.	Same
Integration Time: (Response Time)	1 to 32 seconds adjustable in 5 steps	Same
Outputs:	SO_2/NO_2 signal (0 - 10 VDC dynamic range at 10K source impedance for each channel compatible with 0-1 ma. strip chart recorder SO_2/NO_2 AGC volt. (((	
	Same both models	
Indicators:	One panel meter for SO_2/NO_2 burden One panel meter for AGC voltage	
Calibration:	Basic instrument alignment including optics and electronic adjustments are performed at BRL. Scale Factor - two fused-silica cells containing carefully calibrated amounts of target gas are incorporated in the instrument for sensor sensitivity calibration (volts per ppm-meter). Wavelength - a lockable micrometer screw is provided for precise adjustment of the grating.	
Reference Lamp:	A Quartz-iodine reference lamp is provided for routine checks on scale factor, wavelength and instrument zero calibration.	
Zero Adjustments:	Two separate signal zeroing controls are located on the front panel, one main OFFSET control for routine positioning of the recorder pen and a second screw drive-adjusted control for establishing sky and artificial source reference levels.	

Controls:

Power ON/OFF

Zero offset adjust

Entrance mask selector

Mask position control

Reference cell selector

Meter Scale selector

Time constant selector

Reference lamp ON/OFF

Test point selector

115V 60 Hz/12VDC/24VDC

Power selector

Reference zero adjust

Grating position control

Mechanical: Size - 29" x 12" x 17" (35" x 12" x 17" including reference lamp attachment)

Weight - 38 lbs. including isolators and mounting plate.

Mounting - mounting plate provided with standard tripod mounting holes

Environmental: Vibration isolation is provided using Barry type T 44 per MIL-S 901C (general purpose) or Barry type H44-AA per MIL-STD-810 (helicopter) to be specified by customer.

Temperature - 0 to 120°F.

Power Requirements: 20 watts nominal for normal monitoring, or 75 watts with reference lamps on at 115 volts 60 Hz or 12VDC. 20 watts only at 24VDC. (Reference lamp not operational on 24VDC.)

Signal Processing:

The optical system provides a converging cone of light at the spectrometer exit aperture and the dispersed spectrum is focused in the plane of the correlator disc. If the incoming light has passed through a gas cloud containing the molecular species of interest, the resulting absorption bands appearing in the dispersed spectrum will correlate in a positive and negative sense with the correlation masks, i.e., slit patterns engraved in the aluminum-on-quartz correlator disc, and the resulting modulated light is detected by the XP1118 PMT. The PMT signals are applied directly to a current-to-voltage amplifier consisting of a dual FET. The resulting voltage wave is a box car type waveform produced by the four sets of slits which make up the four masks on the disc. The energy received through each mask is designated P1, P2, P3 and P4. This modulated dc waveform is fed directly to a high voltage inverter where a synchronous gate pulse is used to accept only the P1 portion of the video signal. The amplitude of P1 is then compared with a predetermined dc reference level. The voltage difference that may exist is then integrated and applied as control of a dc to dc converter. The converter will deliver a maximum of 1500 volts negative to the PMT cathode and dynode chain in the absence of radiation at the PMT photocathode. Under normal operating conditions there will be sufficient radiant energy and hence sufficient video signal to exceed the pre-set dc threshold. The integrated driving signal then forces the high voltage output lower thereby reducing the gain of the PMT and lowering the value of P1 until it equals the pre-set reference value. P1 then is held to a constant value by this automatic gain control operation and since it is sampled once per cycle of the 4 mask correlation process, the same PMT gain applied to all four portions of the signal (P1, P2, P3 and P4) in a given cycle. This type of automatic gain control is then termed the "primary AGC".

The box car signal is also applied to FET gates where P1 and P2 are individually gated and separated from P3 and P4. The amplitude of P3 and P4 is then automatically adjusted by a photon-coupled variable attenuator followed by amplification. This operation forms a secondary automatic gain control system (AGC #2). P3 is synchronously gated then integrated and applied as negative feedback to the photon-coupled variable alternator in such a manner as to maintain the amplitude of 3 essentially constant over a wide dynamic range.

P1, P2, P3 and P4 are then fed to a product detector. Synchronous gating pulses from the correlation disc are applied to the product detector as well. P3 and P4 are inverted before summing with P1 and P2. The final function of the product detector is integration of the resultant video signal. A final amplifier provides the major portion of signal amplification apart from the photomultiplier tube. If P1' P4' is now considered to represent high absorption regions by the target molecule and P2' P3' low absorption regions then the sensor will demonstrate high sensitivity to the presence of a gas cloud. Also if we now consider the case of zero gas and that a monotonic and linear change in spectral energy occurs across the spectral bandwidth, i.e., that a linear spectral gradient is present in the light source, and that P1 through P4 represent regions of progressively

longer wavelength then a moments reflection will show that whereas four such regions can be made to produce additive gas signals they can also be used to produce subtraction gradient signals and hence the sensor will have maximum sensitivity to the target molecule and minimum sensitivity to changes in spectral character of the light source.

Note that the operation of the product detector on the video signals P1, P2, P3 and P4 is such as to take the difference of the differences, i.e., the amplitude difference of P1 and P2 is subtracted from the amplitude difference of P3 and P4.

In simplified terms the overall signal processing can be considered to result in a sensor response R_S described by:

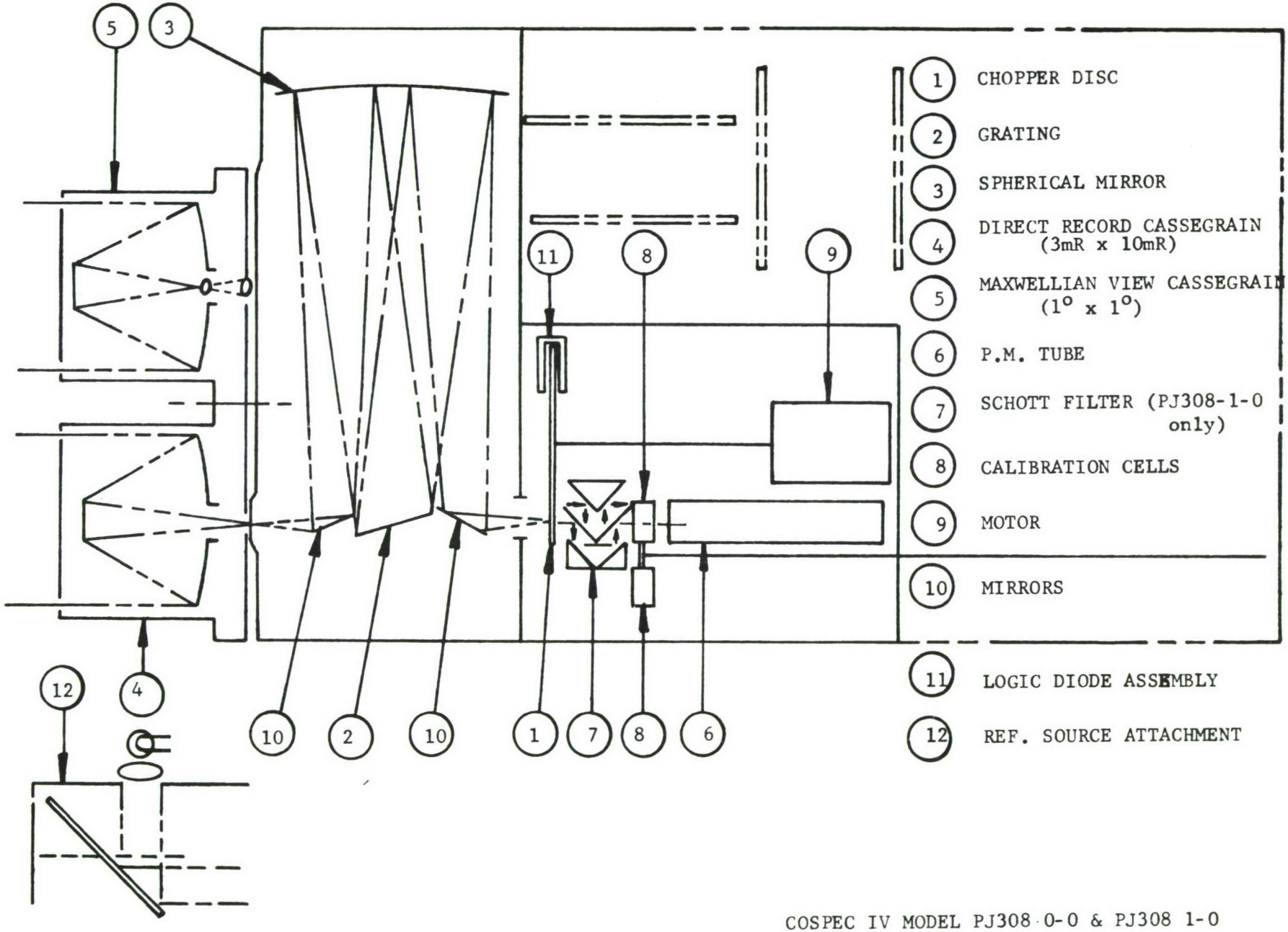
$$R_S = \frac{P1' - P2'}{P1'} - \frac{P3' - P4'}{P3'}$$

where primed terms are radiant power levels at the PMT photo cathode OR

$$R_S = \frac{P4'}{P3'} - \frac{P2'}{P1'}$$

In model PJ308-0-0 a miniature toggle switch S1 is provided on PCB4 to permit X10 increase of signal amplification.

For zeroing purposes an adjustable dc offset voltage is injected into the signal channel through dual FET Q4 and panel-mounted potentiometers R11 and R16.



COSPEC IV MODEL PJ308-0-0 & PJ308 1-0

Oxides of Nitrogen from Internal Combustion Engines:

The internal combustion engine combustion process forms nitrogen oxides (NO_x) which are emitted primarily as an exhaust emission. Other emission ports such as crankcase leakage, etc., only constitute approximately 1% of the total emissions. The two most abundant compounds making up the total NO_x group are nitric oxide (NO) and nitrogen dioxide (NO_2).

NO is formed by the fixation of oxygen and nitrogen at high temperatures according to the reaction $N_2 + O_2 \text{ —— } 2\ NO$. The burning of petroleum fuels in internal combustion engines produces an emission which is primarily NO. NO, which is colorless, reacts at ordinary temperatures with oxygen in the air to form the brownish dioxide NO_2 according to the reaction $NO + \frac{1}{2}\ O_2 \text{ — } NO$.

The rate of formation of NO_2 from NO and oxygen in air increases as the square of the NO concentration. The reaction proceeds very rapidly at high NO concentrations and very slowly at low NO concentrations. At a concentration of 1000 ppm NO, about 5 minutes are necessary to convert one half of the NO to NO_2. At a concentration of 10 ppm, 1000 hours are required for one half conversion. In the presence of hydrocarbons and when irradiated by sunlight, the conversion of NO to NO_2 is much faster.

In the internal combustion engine such factors as air/fuel ratio, compression ratio, ignition timing and manifold pressure all affect peak combustion temperatures. In general for an engine in good adjustment these factors contribute to maximize NO_x emissions during periods of heavy engine loading and during periods of acceleration.

The following is based in part on data contained in the November 1970 report to the Aberdeen Proving Grounds submitted by Barringer Research. The data used here relates to fuel consumption rates and subsequent potential NO_2 emissions.

Considering the 2½ ton personnel carrier, the MB5A2, the following rationale is used to determine the potential NO_2 available for detection under the testing conditions used. For the MB5A2;

Fuel consumption, full load = 3.1 gms/sec
which produces approximately 50 mgms/sec NO_x
which potentially will produce approximately 3000 ppm NO_2.

Fuel consumption at 2200 rpm (no load) = 0.31 gms/sec
which produces approximately 3 mgms/sec NO_x
which potentially will produce approximately 200 ppm NO_2.

Considering these approximations in light of required oxidation times, it would appear that at full loading detectable NO_2 levels would be immediately detectable by COSPEC IV where the threshold of detectability is on the order of 1 ppm-m. In a time span of a few minutes large concentrations of NO_2 should be detectable not considering atmosphere conditions which would drastically dilute the NO. For the no load condition where the potential NO_2 is of the order of 200 ppm, it would appear that the required

oxidation time would be at least one hour for half conversion and on the order of two to five minutes to reach the threshold of COSPEC IV detectability. These times illustrate the extreme difficulty in detecting NO_2 from the unloaded engine especially when natural atmospheric dilution is considered.

The rationale can be carried further to explain the detectability of the diesel generator when loaded to 2/3 full load. Under these loading conditions, combustion temperatures increase producing higher NO_2 levels which provide detectable NO_2 as a direct exhaust emission.

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The feasibility of remotely sensing NO_2 emissions from a single source was explored using a UV Correlation Spectrometer manufactured by Barringer Research Ltd., Toronto, Canada. Detections were made of a 2.5 ton truck exhaust at 200 meters range and a 30 KW diesel generator at 1050 meters range to demonstrate feasibility of detecting small sources of NO_2.

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