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THREE FIELD TESTS OF A GAS FILTER CORRELATION RADIOMETER

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By

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Joseph C. Casas
and
Estelle P. Condon

Interim Report

Prepared for the
National Aeronautics and Space Administration
Langley Research Center
Hampton, Virginia 23665

Under
Research Grant NSG 1127
Henry G. Reichle, Jr., Technical Monitor
Atmospheric and Environmental Sciences Division

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Norfolk, Virginia 23508

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ABSTRACT

The results of three test flights to remotely measure non-urban carbon monoxide (CO) concentrations by gas filter correlation radiometry are presented and discussed. The flights took place in May 1974 over the western edge of the Gulf Stream, and in June 1975 over the Albemarle Sound, North Carolina. The inferred CO concentrations obtained through use of the Gas Filter Correlation Radiometer (GFCR) agreed with independent measurements obtained by gas chromatograph air sample bottle analysis to within 20 percent. The equipment flown on board the aircraft, the flight test procedure, the gas chromatograph direct air sampling procedure, and the GFCR data analysis procedure are discussed.
ACKNOWLEDGMENTS

The work reported in this paper represents the combined efforts of personnel from the National Aeronautics and Space Administration/Langley Research Center (NASA/LaRC), Northrop Services, Inc., and Old Dominion University. The Old Dominion University personnel participated in the field tests and were primarily responsible for research performed in the areas of reduction of data obtained by the Gas Filter Correlation Radiometer (GFCR), and the analysis of air sample bottles through gas chromatography. The work described in the report was performed under NASA research grants NSG 1127, NSG 1394, and NSG 1395.

NASA/LaRC personnel on board the C-54 aircraft included Mr. W.D. Hesketh, Mr. J.A. Holland, Mr. S.M. Beck, and Mr. J.C. Nelms. Their responsibilities included the operation of the GFCR, the taking of air grab samples, the operation of the additional experiment supporting equipment, and the planning of the flight pattern. On board the R.V. Annandale, the NASA/LaRC personnel included Dr. H.G. Reichle, Jr., and Dr. H.D. Orr, III, who participated in the taking of surface measurements and in the test planning. The Northrop personnel included Mr. R.E. Hughes, Ms. A.L. Walker, and Mr. V.A. Letourneau, who were responsible for the taking of CO concentration measurements using a gas chromatograph. On board the surface vessel for the Albemarle Sound flights was Dr. H.G. Reichle, Jr., who participated in the test planning.
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THREE FIELD TESTS OF A GAS FILTER CORRELATION RADIOMETER

By
Shirley A. Campbell¹, Joseph C. Casas¹, and Estelle P. Condon¹

SUMMARY

The results of three flight tests of a Gas Filter Correlation Radiometer (GFCR) are presented and discussed. This instrument is used in the remote detection of atmospheric trace gases by nondispersive infrared technology. For these tests, it was used for the measurement of carbon monoxide (CO) concentrations in June 1975 over the Albemarle Sound, North Carolina, and in May 1974 over the western edge of the Gulf Stream.

The major purposes of these flights were to verify all phases of operation of the GFCR, including the optical alignment, to develop a data reduction procedure, and to test the experiment verification procedure which utilizes gas chromatography in the analysis of air sample bottles. One minor purpose was to demonstrate that the GFCR technique is feasible over surfaces of uniform radiance, e.g., water surfaces.

The results of the air sample bottle analysis are shown in comparison to CO concentrations inferred through analysis of the GFCR data. The results of the GFCR agreed with the air sample bottle data to within 20 percent. The equipment flown on board the aircraft and the data-taking procedure are discussed. The data reduction procedures are described, and the final results and conclusions are presented.

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SYMBOLS

E  monochromatic upwelling radiance, watts cm$^{-2}$sr$^{-1}$
f  Chapman function, dimensionless
h  sensor altitude index, dimensionless
h' uppermost layer altitude index, dimensionless
H_s wavenumber dependent sun irradiance at the top of the atmosphere, watts cm$^{-1}$sr$^{-2}$cm$^{-1}$
N^O Planck blackbody radiation, watts cm$^{-2}$sr$^{-1}$
T  layer temperature, Kelvin
T_s surface temperature, Kelvin
z  altitude index, dimensionless
ε  wavenumber dependent surface emittance, dimensionless
τ  gaseous transmittance at a particular altitude, dimensionless
θ  sun zenith angle, degrees
ω  wavenumber (inverse wavelength), cm$^{-1}$

INTRODUCTION

The purpose of this paper is to present the results of an application of gas filter correlation radiometry to the remote measurement of nonurban carbon monoxide (CO) concentrations. After feasibility studies (refs. 1,2,3) had proven favorable to the concept of remote measurement of tropospheric gaseous pollutant concentrations from aircraft and Earth-orbiting platforms, the National Aeronautics and Space Administration (NASA) at Langley Research Center undertook the development of the gas filter correlation principle. The major objective of the Measurement of Air Pollution from Satellites (MAPS) program has been the development of the Gas Filter Correlation Radiometer (GFCR) for the quantitative remote measurement of CO in the atmosphere. Several
instrument techniques and methods of applying the GFCR principle have been under development since 1972 (refs. 1,2,3).

The GFCR principle is based upon the nondispersive infrared gas analyzers that have been used for several years, and upon the Selective Chopper Radiometer (SCR) flown on board Nimbus IV (ref. 4). The operational principles of several versions of the GFCR have been thoroughly explained by Reichle and Hesketh (refs. 5 and 6).

In order to relate the output signals of the GFCR to the gas burden present in the atmosphere, theoretical models must be applied, because it is not currently possible to simulate accurately in the laboratory the infrared activity (emission and absorption) for long atmospheric paths. The primary tool in this area of research has been the basic atmospheric radiative transfer calculations determined by band or line-by-line mathematical absorption models. Many computer algorithms utilizing either the approximate solutions of band models or the more rigorous line-by-line calculations have been developed for the purpose of calculating the upwelling infrared radiance from planetary surfaces. Band model algorithms calculate the average spectral attenuation of the pollutant band strength by assuming either systematic or random spectral line strengths and line spacing. Line-by-line calculations consider individual spectral line parameters which have been observed experimentally or have been theoretically fitted to spectral constants. While the time required for band model calculations is generally less than for the line-by-line model calculations, the increased accuracy of the spectral line-by-line calculations enhances the quality of the data analysis for high spectral resolution instrumentation such as the GFCR.

Since the GFCR has both day and night measurement capabilities in the 4.6 micrometer spectral region, the one-dimensional equation of radiative transfer for a nadir viewing sensor can be expressed as:
\[ E(\omega) = \varepsilon(\omega) N^0(\omega, T_s) \tau(\omega, h) \]
\[ + \int_0^h N^0(\omega, T(z)) \frac{d\tau(\omega, z)}{dz} \, dz \]
\[ + \frac{1}{\pi} [1 - \varepsilon(\omega)] \cos \theta H_s(\omega) \]
\[ \cdot \left[ \tau(\omega, h) \right] \left[ \tau(\omega, h') \right] f(\theta) \]

where \( \varepsilon(\omega) \) is the wavenumber dependent surface emittance, \( N^0(\omega, T_s) \) is the Planck blackbody function which is dependent on wavenumber and surface temperature, \( T_s \), or radiating gas temperature at a particular altitude, \( T(z) \). The monochromatic transmittance of the atmosphere between the emitting surface \( z \) and the altitude of the sensor, \( h \), is represented by \( \tau(\omega, h) \), and the monochromatic vertical transmission of the entire modeled atmosphere is represented by \( \tau(\omega, h') \). The solar zenith angle is \( \theta \) and the wavenumber dependent sun irradiance at the top of the atmosphere is \( H_s \). The Chapman function (ref. 1), \( f(\theta) \), is equal to \( \sec \theta \) for \( 0^\circ < \theta < 60^\circ \), and is equal to the Chapman polynomial for \( \theta > 60^\circ \). The first, second, and third terms, respectively, represent the thermal radiation attenuated by the atmosphere reaching the sensor, the thermal radiation emitted or absorbed by the infrared active gases in the atmosphere, and the solar radiation attenuated by the entire atmosphere and then reflected by the Earth's surface and transmitted to the sensor.

The line-by-line model used for the purpose of analysis of data for the GFCR is contained in the Simulated Monochromatic Atmospheric Radiative Transfer (SMART) program (ref. 7). The SMART program performs the task of calculating atmospheric transmittance and upwelling radiance as a function of wavenumber, altitude, and variation in atmospheric/Earth surface parameters. These calculations are then used to describe the predicted GFCR
output response curves as a function of the changing atmospheric CO gas burden between the nadir viewing instrument and the surface of the Earth.

Personnel from NASA/LaRC and Old Dominion University have been involved in the analysis of GFCR data obtained during test flights conducted in 1974 and 1975. Two of the major purposes of these test flights were to examine several problem areas associated with the GFCR instrument design, and to test and verify the data analysis procedure for the GFCR technique. The instrument operational problem areas have already been discussed by Reichle and Hesketh (refs. 5 and 6). The raw data obtained during these flights served as a first exercise for the new software developed. The purpose of the software was to reduce these data to atmospheric CO burden concentrations as a function of aircraft altitude and of variable surface/atmospheric radiating characteristics, i.e., surface temperature, vertical temperature, and water vapor mixing ratio profiles, etc. The third major purpose of the flights served to verify the GFCR inferred CO concentration levels by comparing them with measurements made via a direct air sample bottle method which incorporates gas chromatography. A brief discussion of the data reduction procedure and the gas chromatograph direct sampling procedure are presented in subsequent sections.

FLIGHT TEST PROCEDURES

For each of the 1974 and 1975 flight tests, a NASA C-54 aircraft was equipped with the GFCR instrument, analog recording system, an aircraft flight hygrometer, an in-flight outside air temperature resistance thermometer, and a Precision Radiometric Thermometer (PRT-5). This thermometer operates in the 11.0 micrometer spectral window region and measures radiometric surface temperature.

The first flight test took place on May 4, 1974, over the Gulf Stream approximately 75 km southeast of Cape Lookout, North
Carolina, as shown in figure 1. This test utilized the Wallops stationed C-54 aircraft and the R. V. Annandale, a 28.8 meter research vessel under charter to Wallops Flight Center. This vessel stationed on the western edge of the Gulf Stream served as a surface reference point for aircraft navigation and as a meteorological information platform for National Oceanic and Atmospheric Administration (NOAA) radiosonde launches and surface condition observations. Also, during this flight test, continuous CO surface concentration measurements were made on board using a gas chromatograph.

During the period of this flight (from approximately 1100 hours to 1345 hours), the prevailing surface winds were generally from the northeast. The mode of operation for the aircraft began with the aircraft approaching the ship from the south and upwind from the test site area. Approximately one kilometer from the ship, each data run began and continued for three to four minutes of flight time. After each data run, the aircraft descended to the next test altitude and repeated the same flight direction and ground track. This procedure was performed for altitudes between 5335 m and 150 m. Unfortunately, due to an equipment malfunction, all data taken above the 2850 m altitude were lost.

The second and third flights on June 9 and 10, 1975, consisted of a daylight flight (from approximately 1254 hours to 1407 hours) and a nighttime flight (from approximately 2120 hours to 2325 hours) over the Albemarle Sound 11 km southeast of Edenton, North Carolina. A 5.8 m Old Dominion University research vessel was stationed halfway between the northern and southern Sound shoreline and approximately midway between the railroad and state Route 32 bridge crossings, as shown in figure 2. Unlike the R.V. Annandale, this vessel did not have the capability of radiosonde launching or direct CO gas chromatograph analysis. The vessel did serve as a reference point for the aircraft and as a platform for obtaining meteorological surface observations. In addition, it was
Figure 1. Test site for Gulf Stream flight on May 4, 1974.
Figure 2. Test site for Albemarle Sound flights on June 9 and 10, 1975.
equipped with a direct air sampling mast for obtaining grab samples to be used in a CO gas chromatograph analysis. The aircraft was also equipped with similar bottles used to obtain grab samples at each data run altitude.

The surface winds were generally out of the east for both the day and night flights. The aircraft data runs consisted of west to east tracks between the railroad and State Route 32 bridges, maintaining a flight path just south of the boat location, as shown in figure 2. The altitudes of flight were from 3150 m to 450 m during the daylight hours and from 2550 m to 450 m for the nighttime flight. Extensive cloud coverage was encountered during the night flight at altitudes above 1350 m; therefore, some of the data above this altitude were lost due to cloud interference.

DATA REDUCTION PROCEDURE

The solution of the radiative transfer equation by the SMART program requires a detailed knowledge of the meteorological conditions, i.e., atmospheric temperature and water vapor vertical profiles and surface temperature, corresponding to the time and geographical location of the GFCR measurements. As previously stated, the aircraft was equipped with outside air and dewpoint temperature sensors. The data obtained from these sensors were used in corroboration with information from NOAA radiosondes launched from the surface vessel directly before and after the test flight. The atmospheric temperature and water vapor volume mixing ratio data are shown in figures 3, 4, and 5.

The surface temperature was inferred using radiometric instrumentation (PRT-5) flown on board the aircraft. Thermistor measurements of the water's bulk temperature taken from a vessel located below the aircraft during each flight were 2° C lower than the inferred radiometric temperature. This difference is thought to be due to the fact that the PRT-5
Figure 3. Temperature and water vapor mixing ratio profiles for Albemarle Sound flight on June 10, 1975.
Figure 4. Temperature and water vapor mixing ratio profiles for Albemarle Sound night flight on June 9, 1975.
Figure 5. Temperature and water vapor mixing ratio profiles for Gulf Stream flight on May 4, 1974.
measures actual infrared active skin temperature, which is often less than 1 mm thick (refs. 8 and 9), while thermistor measurements indicate the bulk temperature of the water below the infrared radiating sea/air interface. After having obtained the atmospheric profile and surface temperature information, a line-by-line program (ref. 7) was run simulating the GFCR's predicated response to ten different concentrations of CO as a function of flight altitude. An example of the GFCR response curve for an altitude of 3200 meters over the Albemarle Sound for June 10, 1975, is shown in figure 6. The program's output includes instrument balance and calibration radiance information in watts cm\(^{-2}\) sr\(^{-1}\). (Detailed explanations of the instrument procedures are described in refs. 1 and 10.) These radiance values were then equivalenced to the corresponding GFCR output signals in volts, thus establishing the linear relationship between the predicted response and the actual instrument output. The GFCR's data in volts were then converted to watts cm\(^{-2}\) sr\(^{-1}\) by linearly interpolating on the calibration curves. A cubic spline interpolation algorithm using the atmospheric response curves previously generated by the radiative transfer program was then employed to obtain concentrations of CO in parts per million (ppm) from the GFCR's output in watts cm\(^{-2}\) sr\(^{-1}\).

Uniformity of the vertical distribution of the pollutant gas below the aircraft is determined by the vertical atmospheric thermal stability. For example, the Albemarle Sound data were reduced assuming a single uniformly mixed layer of CO. The final results are presented in figures 7 and 8 and Appendixes A and B. For the Gulf Stream flight, the meteorological conditions dictated two uniformly mixed layers of CO. The layer below 1800 m was uniformly mixed at .215 ppm (as indicated by the GFCR measurements), and the layer above 1800 m was assumed uniformly mixed, but at a different concentration of CO. The concentration of the upper layer was then inferred. The final results are presented in figure 9.
Figure 6. GFCR response curve for 3200 m over the Albemarle Sound on June 10, 1975.
Figure 7. Average CO concentrations inferred by GFCR and direct air sample bottle CO concentrations obtained by a gas chromatograph as a function of altitude for the Albemarle Sound day flight on June 10, 1975.
Figure 8. Average CO concentrations inferred by GFCR and direct air sample bottle CO concentrations obtained by a gas chromatograph as a function of altitude for the Albemarle Sound night flight on June 9, 1975.
Figure 9. Average CO concentrations inferred by GFCR as a function of altitude for the Gulf Stream flight on May 4, 1974. The one-layer model assumed uniform mixing of CO below the aircraft. The two-layer model assumed uniform mixing at .215 ppm below 1800 m, and the concentrations above 1800 m were inferred. The surface concentration was measured with a gas chromatograph.
GAS CHROMATOGRAPH DIRECT SAMPLING PROCEDURE

In order to verify the inferred CO concentrations obtained by the GFCR, a direct method for measuring CO incorporating gas chromatography in the analysis of air sample bottles was utilized for the Albemarle Sound flights. The bottles, which are commercially available, are manufactured of 304 stainless steel and are 300 ml by volume. Prior to use, each bottle was subjected to a cleaning and heat treatment process which consisted first of a degreasing for 10-12 hours with trichloroethylene. They were then heated to 1010° C in a vacuum furnace at 10^-4 torr. The temperature was held constant for five minutes and then the bottles were allowed to cool slowly to room temperature while still under vacuum. This procedure served to remove any oil film or oil residue that might contaminate the air samples.

For the actual test flight, a sampling station was located in a small boat on the Albemarle Sound. Here surface air samples were collected in the stainless steel bottles while the aircraft made overpasses in a spiral descent mode. Air sample bottles were also taken on board the aircraft at selected altitudes where measurements were made with the GFCR. During the test, each bottle was pressurized with ambient air three times to 2.41 x 10^5 N/m². The pressure was released twice, and the third sample was kept for analysis. All bottles were returned to the laboratory for analysis.

The apparatus and analysis procedure described below were modeled after that used by John W. Swinnerton, et al., of the Naval Research Laboratory (refs. 11, 12, 13, 14). A gas chromatograph equipped with dual flame ionization detectors was used for the analysis of the air samples. A cold trapping system using dry ice-acetone served to preconcentrate the gas of interest, CO, from a 100 ml sample loop. The sample gas was transferred from the sample loop onto the cold trap. The
cold trap was heated to 100° C to expel the CO into the chromatograph where it was separated in time from the residual air peak. The data were recorded on a strip chart with the CO peak separated in time from the residual air. The amount of the pollutant gas present in the sample is directly proportional to the area under the peak. The area is approximated by multiplying the height of the peak by the width at half height. Gas standards of known concentration were analyzed and the areas under their peaks were computed. A calibration curve was plotted of area vs. concentration of CO, and the unknown concentrations of the samples were then inferred from the plot. The accuracy of this procedure has been shown to be within 15 percent. The Albemarle Sound data are shown in figures 7 and 8.

RESULTS

The results of the Gulf Stream flight are shown in figure 9. Unfortunately, there were no direct air samples taken at the flight altitudes and the inferred CO concentrations could not be checked. There was, however, a surface concentration measurement by a gas chromatograph, and good agreement was found between this measurement and the GFCR result (see figure 9).

As previously explained in the Data Reduction Procedure Section, a two layer atmospheric model was shown to better simulate actual meteorological conditions (fig. 3). Here, the lower layer, below 1800 m, was uniformly mixed at .215 ppm (as indicated by the GFCR measurement), and the upper layer, above 1800 m, was inferred to be approximately .11 ppm. This result is within 10 percent of the average value for a clean atmosphere for this latitude (ref. 15).

Figure 7 and Appendix A show the results of the Albemarle Sound day flight. The gas chromatograph direct air sampling procedure had been developed and was operative. Grab samples were collected at each flight test altitude in stainless steel
bottles for later analysis with a gas chromatograph. The surface CO concentration was obtained by averaging five bottle samples taken on board the boat during the duration of the test. The accuracy of the direct sampling procedure has been shown to be within 15 percent. A comparison can be made between the CO bottle sample measurements and the GFCR column density measurements after determining the GFCR's "weighting function," i.e., a mathematical description derived from the GFCR's signal designating the location in the atmosphere below the aircraft from which the instrument receives most of its information. For these flights, the "weighting function" determined that the major portion of the instrument's signal was obtained from the atmosphere directly below the aircraft resulting in the agreement of the GFCR data with the bottle sample data to within 20 percent. Modifying the characteristics of the GFCR weighting function by changing instrument parameters is under further study.

The results of the Albemarle Sound night flight are shown in figure 8 and Appendix B. Since the instrument operates in the thermal infrared (4.6 μm), day and night operations are possible. Again, as in the day flight, good agreement is obtained between the GFCR results and the air sample bottle analysis done with the gas chromatograph. During the first 80 seconds of the flight at the 1372 m altitude, it was observed that clouds were contaminating the instrument's field of view. Thus, this data reduced under assumption of no cloud cover, results in an underestimation of CO concentration. For flight altitudes below 1372 m, the CO optical mass between the aircraft and the Earth's surface decreases; therefore, the GFCR's signal to noise ratio (SNR) decreases resulting in an increase in the standard deviation from the mean inferred CO concentration. This is evident in the plots of Appendixes A and B.

CONCLUSIONS

The results of three test flights to remotely measure nonurban carbon monoxide concentrations using a Gas Filter Correlation Radiometer have been presented. The flights were
made on May 4, 1974, over the western edge of the Gulf Stream, and on June 9 and 10, 1975, over the Albemarle Sound, North Carolina. The three major purposes of these flights were to verify all phases of operation of the GFCR including the optical alignment, to develop a data reduction procedure, and to test the experiment verification procedure which utilizes gas chromatography in the analysis of air sample bottles. The results of these flights showed that the GFCR's optical and electronic components functioned properly, the gas chromatograph direct sampling procedure operated as designed, and a reliable and efficient data reduction procedure was developed. One minor purpose of these flights was to demonstrate that the GFCR technique is feasible over surfaces of uniform radiance, e.g., water. This was verified by the agreement between the results of the GFCR and the air samples for the Albemarle Sound flights to within 20 percent. Test flights were also made over land surfaces in the Albemarle Sound area, but the data was not reducible due to instrument problems.

The gas filter correlation technique has been shown to be capable of measuring carbon monoxide over water. Since the time that these data were reduced, an improved version of a GFCR has been received at Langley (ref. 6). This instrument will allow CO measurements to be extended to areas over land.
APPENDIX A

Results of the test flight over the Albemarle Sound on June 10, 1975 (daytime). Included are point plots of all data taken over water for each altitude flown showing concentration of CO versus time.
Figure A1. Concentration of CO versus time for an altitude of 3200 m.
Figure A2. Concentration of CO versus time for an altitude of 2591 m.
Figure A3. Concentration of CO versus time for an altitude of 1981 m.
Figure A4. Concentration of CO versus time for an altitude of 1372 m.
Figure A5. Concentration of CO versus time for an altitude of 762 m.
Figure A6. Concentration of CO versus time for an altitude of 457 m.
APPENDIX B

Results of the test flight over the Albemarle Sound on June 9, 1975 (nighttime). Included are point plots of all data taken over water for each altitude flown showing concentration of CO versus time.
Figure B1. Concentration of CO versus time for an altitude of 2591 m.
Figure B2. Concentration of CO versus time for an altitude of 1981 m.
Figure B3. Concentration of CO versus time for an altitude of 1372 m.
Figure B4. Concentration of CO versus time for an altitude of 762 m.
Figure B5. Concentration of CO versus time for an altitude of 457 m.
REFERENCES


