Modifying and Testing

a Length Modulated Radiometer

Used for Measuring

Atmospheric Carbon Monoxide and Methane

by

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ABSTRACT

In order to understand the impact and mitigation of anthropogenic influences on the atmosphere's properties, continual and global observations are an immense asset. The tropospheric concentrations of carbon monoxide (CO) and methane (CH₄) are particularly important for environmental modelling, since they play key roles in atmospheric chemistry. Correlation radiometry is often used for such observations, and a new form, called length modulation radiometry, has been developed at the University of Toronto.

A ground-based length modulated radiometer (LMR) for observing CO and CH₄ was built and tested with the ultimate goal of measuring CO at the 4.7 µm and 2.4 µm bands to a precision of ±10% and CH₄ at the 2.3 µm band to a precision of ±1%. The work presented in this report involved modifying and testing this LMR. Initially, the instrument was tested to ensure that results from previous work were repeatable. Optimal setup parameters to attain the minimum imbalance were the same as those obtained before, and the LMR’s response to various nitrogen and CO pressures matched the theoretical predictions.

Several modifications were made to the LMR to improve its performance. A fibre optic cable to direct the radiation source into the LMR was added, a rotating vane chopper was used, and the optical setup was improved to minimize the imbalance. As well, the digital signal processing software and hardware were upgraded to allow more efficient, and more powerful, data processing and analysis. Following these changes, the optimal parameters for operating the LMR were reestablished. The changes were also implemented to permit the addition of a second detection channel to the LMR. This second channel will allow the reduction of noise in the data stream from solar radiation. The work presented in this report seems to demonstrate that the LMR is likely to be an effective ground-based instrument for measuring CO and CH₄.
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DSP56001  3.2.4 A digital signal processing card made by Motorola.
EAT       2.3.1 Effective Average Transmission.
EDT       2.3.1 Effective Difference Transmission.
EOS       2.4 Earth Observing System.
G         2.3.1 The instrument function of a correlation radiometer, derived from the optical and electrical gains of the radiometer. Unitless.
I         2.2 The intensity or radiance of a radiation beam. Units of energy per area, per time, per frequency, per steradian.
I(\nu)    2.3.1 The intensity of radiation, as a function of wavenumber, incident upon a radiometer’s correlation cell. Units of energy per area, per time, per frequency, per steradian.
J         2.2 The source function in Schwarzschild’s equation. Units of energy per area, per time, per frequency, per steradian.
K         2.3.1 The radiometer’s evacuated balance. Unitless.
LMC       2.3.4 Length Modulated Cell.
LMR       2.3.4 Length Modulated Radiometer.
MOPITT    2.4 Measurements of Pollution in The Troposphere.
PMR       2.3.3 Pressure Modulated Radiometer.
S_{avg}  2.3.1 The integral of the EAT with respect to wavenumber. Units of energy per volume, per time, per frequency, per steradian.
S_{diff} 2.3.1 The integral of the EDT with respect to wavenumber. Units of energy per volume, per time, per frequency, per steradian.
S_{I(ideal)} 4.3 The ideal imbalance, or ratio $S_{\text{diff}}/S_{\text{avg}}$ of a Length Modulated Radiometer. Unitless.
S_{I(measured)} 4.3 The measured imbalance, or ratio $S_{\text{diff}}/S_{\text{avg}}$ of a Length Modulated Radiometer. Unitless.
SCR  2.3.2 Selective Chopper Radiometer.

SPM  4.7 Signal Processing Module.

$X_1$  4.3 A corrective factor for the imbalance, or ratio $S_{\text{eff}}/S_{\text{avg}}$, obtained from measurements with a second detector in a Length Modulated Radiometer. Unitless.

$v$  2.3.1 Wavenumber. Units of inverse length.

$v_1$ and $v_2$  2.3.1 Define the passband of a narrow band-pass filter. Units of inverse length.

$\tau$  2.2 Transmittance of a medium. Unitless.

$\tau_i(v)$  2.3.1 Transmittance as a function of wavenumber of an external narrow band-pass filter. Unitless.

$\tau_{s1}(v)$  2.3.1 Transmittance as a function of wavenumber of the radiometer's lower density state, i.e. with a low amount of gas. Unitless.

$\tau_{s2}(v)$  2.3.1 Transmittance as a function of wavenumber of the radiometer's higher density state, i.e. with a high amount of gas. Unitless.
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1. CARBON MONOXIDE AND METHANE IN THE ATMOSPHERE

1.1 Introduction

Planet Earth comprises four parts: the lithosphere (land), the hydrosphere (water), the atmosphere (air), and the biosphere (life). These four components continually interact, but most of the interaction occurs at the boundaries between them, which is at the Earth’s surface. The biosphere primarily exists in this boundary layer. The biosphere, in fact, has affected the lithosphere and hydrosphere mainly in this boundary region in the form of altering soils’ composition and presence, and influencing surface and ground water flow and contamination.

The atmosphere’s composition, however, has been shaped almost entirely by interaction with the biosphere. During the planet’s youth, protobacteria oxygenated the atmosphere while decaying biological material nitrified it (Keffer, 1996) and since then, plants and animals have played major roles in balancing the oxygen and carbon dioxide contents. The species *homo sapiens* in particular has been the unintentional driving force behind much change since the industrial revolution, and the changes have occurred at a pace far beyond nature’s usual time scales. These atmospheric changes—at both the global and local spatial scales—have had significant, rapid impacts on the lithosphere, hydrosphere, and biosphere. By studying these changes, human beings will learn the problems inherent in such change, and understand how to mitigate them and prevent further detrimental effects.

1.2 The Atmosphere

1.2.1 The Atmosphere’s Structure

The International Union of Geodesy and Geophysics (IUGG) has developed standard nomenclature to distinguish layers in the vertical profile of the Earth’s atmosphere. The layers
are divided according to temperature gradient (Figure #1-1) and are termed, from the ground up, the troposphere in which the temperature decreases from approximately 290 K to 220 K, the stratosphere in which the temperature increases to approximately 260 K, the mesosphere in which the temperature decreases to approximately 190 K, and the thermosphere in which the temperature rises to between 500 K and 2000 K depending on the amount of solar activity (Liou, 1980). The tops of the layers are the local extrema in the temperature gradient and are the tropopause, stratopause, mesopause, and thermopause respectively. Pressure decreases monotonically with altitude as depicted in Figure #1-2.

1.2.2 The Atmosphere’s Composition

The atmosphere is composed of gases with virtually constant concentrations and with varying concentrations, along with aerosols, solid particles, liquid particulates, clouds, and precipitation. On the next page, Table #1-1 from Liou (1992) lists the predominant atmospheric components and their volume percentages. Nitrogen, oxygen, and argon represent 99.966% of the atmosphere’s volume, but it is the trace gases which have the most significant effects on the atmosphere’s chemical and radiative properties. Most of the constituents vary widely with altitude and, as shown in Table #1-1, several have influences only in the troposphere, near the Earth’s surface. Water and ozone also vary immensely with latitude and season. Carbon dioxide exhibits fluctuations correlated with the growing season in each hemisphere (Liou, 1992). Nitrous oxides, carbon dioxide, methane, and chlorofluorocarbons have each shown increasing trends globally over the past few decades (Bunce, 1994; Liou, 1992).
**Figure #1-1:** (adapted from Liou, 1980) Approximate vertical temperature profile of the Earth's atmosphere

**Figure #1-2:** (adapted from Keffer, 1996) Approximate vertical pressure profile of the Earth's atmosphere
Table #1-1: (Liou, 1992)
Predominant atmospheric components and their volume percentages

<table>
<thead>
<tr>
<th>Gases with constant volume percentages</th>
<th>Gases with variable volume percentages</th>
</tr>
</thead>
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<tr>
<td><strong>gas</strong></td>
<td><strong>volume percent</strong></td>
</tr>
<tr>
<td>nitrogen (N₂)</td>
<td>78.084</td>
</tr>
<tr>
<td>oxygen (O₂)</td>
<td>20.948</td>
</tr>
<tr>
<td>argon (Ar)</td>
<td>0.934</td>
</tr>
<tr>
<td>carbon dioxide (CO₂)</td>
<td>0.034</td>
</tr>
<tr>
<td>neon (Ne)</td>
<td>18.18×10⁻⁴</td>
</tr>
<tr>
<td>helium (He)</td>
<td>5.24×10⁻⁴</td>
</tr>
<tr>
<td>krypton (Kr)</td>
<td>1.14×10⁻⁴</td>
</tr>
<tr>
<td>xenon (Xe)</td>
<td>0.089×10⁻⁴</td>
</tr>
<tr>
<td>hydrogen (H₂)</td>
<td>0.5×10⁻⁴</td>
</tr>
<tr>
<td>methane (CH₄)</td>
<td>1.7×10⁻⁴</td>
</tr>
<tr>
<td>nitrous oxide (N₂O)</td>
<td>0.3×10⁻⁴</td>
</tr>
<tr>
<td>carbon monoxide (CO)</td>
<td>0.08×10⁻⁴</td>
</tr>
</tbody>
</table>

* indicates concentrations near the Earth’s surface.

1.2.3 The Importance of the Troposphere: The OH Radical

The troposphere encompasses most of the biosphere along with the boundaries between the atmosphere and both the lithosphere and the hydrosphere. Hence, any substance released into the atmosphere has the first opportunity to react in the troposphere and tropospheric chemistry is subsequently very complex. Chlorofluorocarbons (CFCs) and nitrous oxide (N₂O) are the only anthropogenic substituents in Table #1-1 that have no tropospheric sinks (Bunce, 1994). Other substances become involved in reaction chains which revolve around the hydroxyl (OH) radical.
The OH radical is an extremely powerful oxidizing agent, and so it attacks and destroys most tropospheric constituents in clean and polluted atmospheres. The radical’s involvement in so many tropospheric processes, and the speed of the reactions, means that it has an extremely short lifetime ranging from approximately 1 s in very clean air to approximately 1 ms in very polluted air (O’Brien and Hard, 1993). Therefore, measuring the concentration of OH in the atmosphere is very challenging, but such knowledge is critical to attaining an accurate understanding of tropospheric chemistry.

The current concentration of tropospheric OH, averaged globally, yearly, and diurnally, is measured as approximately 0.03 pptv (Bunce, 1994). The main formation mechanism for OH is depicted in equations [1-1] → [1-4].

1. \[ \text{NO}_2 + h\nu (\lambda < 400 \text{ nm}) \rightarrow \text{NO} + \text{O} \] (\(h\nu\) represents a photon)
2. \[ \text{O} + \text{O}_2 + \text{M} \rightarrow \text{O}_3 + \text{M} \] (M is any molecule)
3. \[ \text{O}_3 + h\nu (\lambda < 320 \text{ nm}) \rightarrow \text{O}_2^* + \text{O}^* \] (* represents an excited state which for O is usually \(^1\text{D}\))
4. \[ \text{O}^* + \text{H}_2\text{O} \rightarrow 2\text{OH} \]

A sudden increase in the OH concentration can also occur at sunrise due to equation [1-5].

5. \[ \text{HNO}_x + h\nu \rightarrow \text{OH} + \text{NO}_{x-1} \] (x=2,3)

Two other pathways occur, as shown in equations [1-6] and [1-7], but they are usually insignificant:

6. \[ \text{O}^* + \text{RH} \rightarrow \text{OH} + \text{R} \] (R is a hydrocarbon chain)
7. \[ \text{H}_2\text{O}_2 \rightarrow 2\text{OH} \]

The destruction of OH occurs either by abstraction of a hydrogen atom from another atom, such as a hydrocarbon chain (equation [1-8]), or by addition to an unsaturated centre, for example the reverse of [1-5] with any other molecule as a catalyst. The acids are then rained
out of the troposphere.

\[ [1-8] \quad \text{OH} + \text{RH} \rightarrow \text{R} + \text{H}_2\text{O} \]

If R in [1-8] is unsaturated, addition can also occur around the double or triple bonds. The main sinks for OH are addition to carbon monoxide and abstraction from methane (Bunce, 1994) and these reactions are part of numerous cycles and chains which create the complexity in tropospheric chemistry, as described in section 1.3.1.

1.3 Atmospheric Carbon Monoxide and Methane

1.3.1 Tropospheric Chemistry of Carbon Monoxide and Methane

The OH radical provides the only known gas-phase tropospheric sink for CO. The reaction seems to follow the two-step process in equation [1-9], which is also the sink for 70% of OH molecules (Bunce, 1994):

\[ [1-9] \quad \text{OH} + \text{CO} \rightarrow \text{HO} + \text{CO}_2 \]

Most of the remaining 30% of OH oxidizes methane in the simplified reaction chain below presented by Bunce (1994) and modified by Evans (1994).

\[ [1-10] \quad \text{OH} + \text{CH}_4 \rightarrow \text{CH}_3 + \text{H}_2\text{O} \]

\[ [1-11] \quad \text{CH}_3 + \text{O}_2 + \text{M} \rightarrow \text{CH}_2\text{OO} + \text{M} \]

\[ [1-12] \quad \begin{align*}
(a) \quad \text{CH}_2\text{O} + \text{NO} & \rightarrow \text{CH}_3\text{ONO}_2 \\
(b) \quad \text{CH}_3\text{O} + \text{NO} & \rightarrow \text{CH}_4 + \text{NO}_2 \\
(c1) \quad \text{CH}_2\text{O} + \text{HO}_2 & \rightarrow \text{CH}_3\text{OOH} + \text{O}_2 \\
(c2) \quad \text{CH}_3\text{OOH} + \text{O}_2 & \rightarrow \text{CH}_4 + \text{OH}
\end{align*} \]

\[ [1-13] \quad \begin{align*}
(a) \quad \text{CH}_3\text{O} + \text{O}_2 & \rightarrow \text{CH}_2\text{O} + \text{HO}_2 \\
(b) \quad \text{CH}_3\text{O} + \text{CH}_4 & \rightarrow \text{CH}_3\text{OH} + \text{CH}_3
\end{align*} \]

\[ [1-14] \quad \begin{align*}
(a) \quad \text{CH}_2\text{O} + \text{OH} & \rightarrow \text{H}_2\text{O} + \text{HCO} \\
(b) \quad \text{CH}_2\text{O} + \text{hv} & \rightarrow \text{H} + \text{HCO}
\end{align*} \]

\[ [1-15] \quad \text{HCO} + \text{O}_2 \rightarrow \text{CO} + \text{HO}_2 \]
This series then finishes with equation [1-9].

\[ \text{[1-9]} \quad \text{OH + CO} \rightarrow \text{H + CO}_2 \]

This series can also serve for other hydrocarbons by replacing CH\textsubscript{4} with RH in equations [1-10], and [1-13b]. The exact sequence of elementary reactions is poorly understood, extremely complicated, and made more difficult by the involvement of most intermediates in other tropospheric chemical cycles, such as the formation of photochemical smog. The reactions can be summarized, though, by stating that hydrocarbons are oxidized by OH to carbon dioxide and water in the series hydrocarbon → alcohol → aldehyde → acid → carbon dioxide/water (Evans, 1994).

When there is a high concentration of NO in the air, and hence photochemical smog is formed, these reactions form four molecules of ozone for each molecule of methane which is oxidized, as shown in equation [1-16] (Bunce, 1994; Evans, 1994):

\[ \text{[1-16]} \quad \text{CH}_4 + 8\text{O}_2 \rightarrow \text{CO}_2 + 2\text{H}_2\text{O} + 4\text{O}_3 \]

Carbon monoxide is an intermediary in the cycles, but as the concentration of NO drops, carbon monoxide becomes a sink for ozone as depicted in reaction [1-17] (Bunce, 1994; Evans, 1994):

\[ \text{[1-17]} \quad \text{CO} + \text{O}_3 \rightarrow \text{CO}_2 + \text{O}_2 \]

Carbon monoxide and methane undoubtedly are key players in tropospheric chemistry.

1.3.2 Tropospheric Carbon Monoxide

Carbon monoxide’s biggest direct effect on health and the environment is that it combines irreversibly with haemoglobin, rendering the blood molecule incapable of carrying oxygen to cells (Bunce, 1994). Carbon monoxide has an affinity for haemoglobin which is 200 times that of oxygen (Keffer, 1996). This effect is very localized when it occurs, and in any case, urban air typically contains CO mixing ratios which are less than one-fifth of the 100 ppmv threshold
health hazard value (Bunce, 1994; Keffer, 1996). Carbon monoxide also contributes to the greenhouse effect even though it is not a greenhouse gas, because its concentration affects the concentration of other tropospheric gases which are greenhouse gases (Houghton, 1990). For example, equation [1-9] illustrates that an increase in CO results in an increase in CO₂, which is a major contributor to the greenhouse effect. This equation is the largest sink for hydroxyl radicals. As well, during periods of high NO, CO joins the reaction cycles which form photochemical smog while during periods of low NO, CO converts to CO₂ as an O₃ sink (equation [1-17]).

The current global CO budget cannot predict future trends of CO in the atmosphere (Khalil, 1995; Drummond, 1992), but recent studies (Khalil and Rasmussen, 1994, 1988; Novelli et al., 1994) indicate that after increasing by approximately 1% per year in the 1980s, global CO is currently decreasing by between 3% and 7% per year. Accepted values for CO are approximately 100 ppbv in the northern hemisphere and approximately 45 ppbv in the southern hemisphere (Khalil and Rasmussen, 1994; Drummond, 1992). The gradient arises because the average atmospheric lifetime of CO is 2 months, but inter-hemispherical mixing requires at least one year (Khalil, 1995). Therefore, the northern hemisphere has either more sources, or fewer sinks, for CO than the southern hemisphere. The former explanation is more probable since more than half of atmospheric sources are anthropogenic (Khalil and Rasmussen, 1994) and the northern hemisphere has a much larger industrialized population than the southern hemisphere. Approximations of the main sinks and sources of tropospheric CO are listed in Table #1-2, which was compiled by Khalil (1995). As Khalil (1995) mentions, there are large uncertainties in the values, hence the totals are not necessarily indicative of the observed trends.
Table #1-2: (Khalil, 1995)
Estimated Sources and Sinks of Tropospheric Carbon Monoxide

<table>
<thead>
<tr>
<th>SOURCES OF CO (10^9 kg/year)</th>
<th>SINKS OF CO (10^9 kg/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Combustion of biomass</td>
<td>Uptake by soils 400</td>
</tr>
<tr>
<td>Combustion of fossil fuels</td>
<td>Migration to the stratosphere 100</td>
</tr>
<tr>
<td>Non-methane hydrocarbon oxidation</td>
<td>210</td>
</tr>
<tr>
<td>Emissions from oceans</td>
<td>150</td>
</tr>
<tr>
<td>Emissions from vegetation</td>
<td>80</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td><strong>TOTAL</strong> 2500</td>
</tr>
</tbody>
</table>

1.3.3 Tropospheric Methane

Methane is an environmental concern because it is a greenhouse gas. Currently, much of the infrared radiation in methane’s main absorption bands of 8.0 μm and 3.2 μm (Goody and Yung, 1989) escapes from the earth because the concentration of methane in the troposphere is fairly low (see below). If the tropospheric CH₄ concentration were to increase significantly, most of the radiation in these bands would then be trapped. In contrast, the high concentration of CO₂ currently in the troposphere ensures that most of the radiation in the CO₂ absorption bands is absorbed. Hence, a large increase in the tropospheric CO₂ concentration will not result in a large increase in absorption of radiation. In fact, a CH₄ molecule currently has 25 times the radiation absorptive effect of a CO₂ molecule (Houghton, 1990) which results in CH₄ contributing 11 times the global warming effect of CO₂ on a mass basis (Bunce, 1994).

Studies (Lowe et al., 1994; Prinn, 1994; Khalil and Rasmussen, 1990 and 1983; Blake and Rowland, 1988) indicate that the atmospheric methane mixing ratio continually increased around the world between 1978 and 1994 but the rate of increase has been declining. Global
atmospheric \( \text{CH}_4 \) was approximately 1.70 ppmv in 1994 (Lowe et al., 1994; Prinn, 1994). \( \text{CH}_4 \) has an atmospheric lifetime of about 10.5 years, but there is a strong inter-hemispheric gradient with the northern hemisphere having an average \( \text{CH}_4 \) mixing ratio in 1990 of close to 1.75 ppmv, 0.20 ppmv more than the southern hemisphere (Prinn, 1994).

Estimates of the main sources and sinks of methane are listed in Table #1-3 (Prinn, 1994) on the next page. Recent experiments (Mango et al., 1994) also show that transition metals in carbonaceous sedimentary rocks can act as catalysts for the reaction between hydrogen and \( n \)-alkenes which forms light hydrocarbons, including methane. As Prinn (1994) states, the values in Table #1-3 are subject to enormous uncertainties—occasionally with a relative error in excess of 100%—and therefore comparing the total values will not necessarily indicate the trend of \( \text{CH}_4 \) in the atmosphere. Some of the \( \text{CH}_4 \) produced by wetlands is oxidized to \( \text{CO}_2 \) and hence does not enter the budget; otherwise \( \text{CH}_4 \) is released by ebullition from the benthal layer as well as through plant stem transport (Hamilton et al., 1995). Although this source is natural, human activities influence the level through their creation and destruction of wetlands as a result of engineering waterways and through the draining of wetlands for agricultural uses.
### Table #1-3: (Prinn, 1994)
Estimated Sources and Sinks of Atmospheric Methane

<table>
<thead>
<tr>
<th>SOURCES OF METHANE (10^9 kg/year)</th>
<th>SINKS OF METHANE (10^9 kg/year)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>NATURAL</strong></td>
<td></td>
</tr>
<tr>
<td>Wetlands</td>
<td>115</td>
</tr>
<tr>
<td>Termites</td>
<td>20</td>
</tr>
<tr>
<td>Ocean</td>
<td>10</td>
</tr>
<tr>
<td>Freshwater</td>
<td>5</td>
</tr>
<tr>
<td>CH4 Hydrate</td>
<td>5</td>
</tr>
<tr>
<td><strong>ANTHROPOGENIC</strong></td>
<td></td>
</tr>
<tr>
<td>Coal mining, Natural Gas, and Petroleum industries</td>
<td>100</td>
</tr>
<tr>
<td>Enteric fermentation</td>
<td>80</td>
</tr>
<tr>
<td>Rice paddies</td>
<td>60</td>
</tr>
<tr>
<td>Biomass burning</td>
<td>40</td>
</tr>
<tr>
<td>Landfills</td>
<td>30</td>
</tr>
<tr>
<td>Animal Wastes</td>
<td>25</td>
</tr>
<tr>
<td>Domestic Sewage Treatment</td>
<td>25</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td>515</td>
</tr>
<tr>
<td><strong>Removal by atmosphere</strong></td>
<td></td>
</tr>
<tr>
<td>(troposphere and stratosphere)</td>
<td>470</td>
</tr>
<tr>
<td><strong>Removal by soils</strong></td>
<td></td>
</tr>
<tr>
<td></td>
<td>32</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td>502</td>
</tr>
</tbody>
</table>
2. DETECTING AND MEASURING ATMOSPHERIC CO AND CH₄

2.1 Introduction

By observing and modelling CO and CH₄ in the atmosphere, their sources and sinks can be clarified and their trends over various spatiotemporal scales can be determined. The most obvious approach is to collect an air sample—with a balloon, aircraft, or rocket—and then to analyze the sample for the desired components. This method, though, is logistically difficult and very expensive. More importantly, it provides data for only a single location at a single time and therefore not only provides a very incomplete data set, but is also subject to unknown vagaries which may make any given sample anomalous.

2.2 Remote Sensing

Remote sensing overcomes many of the problems associated with collecting samples, and satellite instruments provide an unparalleled wealth of continuous, global data for the modelling of environmental systems. Houghton (1990) comments that satellite data "offer the most effective and objective means" for such studies. Remote sensing is essentially observation without physical contact: properties of substances or objects are determined by examining changes in a field which interacts with the substance or object.

Most remote sensing techniques examine modifications to the electromagnetic field by the substance or object being measured. For atmospheric studies, remote sensing measures radiation transfer through the atmosphere. Although the Sun and most artificial radiation sources are not monochromatic, the theory underlying atmospheric remote sensing is developed with equation [2-1], the integral form of Schwarzchild's equation. This equation is valid for only monochromatic radiation but it provides an excellent approximation for remote sensing studies.
\[ I(b) = I(a)\tau(a,b) + \int_{\text{path } a-b} J(x) \frac{d\tau(b,x)}{dx} \, dx \]

a, b are the endpoints of the path through which the beam traverses.
I is the intensity or radiance of the radiation beam.
Units of energy per area, per time, per frequency, per steradian.
J is indicative of any process along the path which adds energy to the beam, and is known as the source function.
Units of energy per area, per time, per frequency, per steradian.
\( \tau \) is the transmittance of the medium along the path. Unitless.

2.3 Correlation Radiometry

2.3.1 Principles of Correlation Radiometry

Correlation radiometry is a remote sensing technique which uses a gas sample as a spectral filter for the incoming radiation. A cell is filled with the atmospheric gas which is being studied, and radiation is passed through this gas cell. The amount of the gas through which the radiation passes is modulated, which in turn modulates the transmission of radiation through the cell at wavelengths corresponding to the gas absorption lines. The radiation is collected by a detector which amplifies and outputs signals that have the same frequency of modulation as the amount of gas through which the radiation passes. This principle is illustrated in Figure #2-1.

A plot of transmission versus wavenumber for a cell containing an arbitrary gas with two absorption lines is displayed in Figure #2-2A. The upper line represents transmission through a low amount of gas while the lower line represents transmission through a high amount of gas. The average transmission over time (Effective Average Transmission, EAT) and the transmission difference between the high and low states (Effective Difference Transmission, EDT) can then be plotted as shown in Figure #2-2B. The EAT is very close to the observed background.
**Figure #2-1:** (Drummond 1992)
A schematic diagram of the principle of correlation radiometry.

![Diagram](image)

**Figure #2-2:** (adapted from Drummond, 1992)
(A) A plot of transmission versus wavenumber for the two gas states.
(B) A plot of the effective average transmission (EAT) and the effective difference transmission (EDT) versus wavenumber, based on the graphs in (A).
radiation, but the EDT is highly spectrally selective and so can be used to produce a large signal in the vicinity of the absorption lines of the gas.

The drawback to this system is the complexity involved in correlating gas concentration with the EDT. Fortunately, there are methods of simplifying the mathematics somewhat. The studies presented here involve a situation using wavelengths (specifically, 2.3 \( \mu \text{m} \) and 2.4 \( \mu \text{m} \) being measured at ground level with the sun as a source) which have both a high starting intensity, \( I(a) \), and a very small source function, \( J(x) \), over the path \( a \rightarrow b \). The transmittance, \( \tau \), is finite over the path \( a \rightarrow b \), hence in equation [2-1]—with \( I \gg J \)—the integral term becomes negligible compared to the first term, \( I \tau \). The EDT and EAT can be approximated by using only the \( I \tau \) term of equation [2-1] and then integrated with respect to wavenumber, yielding the terms \( S_{\text{diff}} \) and \( S_{\text{avg}} \) respectively (equations [2-2] and [2-3]).

\[
[2-2] \quad S_{\text{diff}} = G \int_{v_1}^{v_2} I(v) \tau_f(v) \left[ \tau_{s1}(v) - \tau_{s2}(v) \right] dv
\]

\[
[2-3] \quad S_{\text{avg}} = G \int_{v_1}^{v_2} I(v) \tau_f(v) \left[ \frac{\tau_{s2}(v) + \tau_{s1}(v)}{2} \right] dv
\]

\( G \) is the instrument function derived from the radiometer’s optical and electronic gains. Unitless.

\( I(v) \) is the intensity of the radiation incident upon the correlation cell as a function of \( v \).

Units of energy per area, per time, per frequency, per steradian.

\( S_{\text{avg}} \) is the integral of the EAT with respect to \( v \).

Units of energy per volume, per time, per frequency, per steradian.

\( S_{\text{diff}} \) is the integral of the EDT with respect to \( v \).

Units of energy per volume, per time, per frequency, per steradian.

\( v \) is wavenumber. Units of inverse length.

\( v_1 \) and \( v_2 \) define the passband of a narrow band-pass filter external to the radiometer.

Units of inverse length.

\( \tau_f(v) \) is the transmittance, as a function of \( v \), of an external narrow band-pass filter with a passband extending from \( v_1 \) to \( v_2 \). Unitless.
\( \tau_{sl}(v) \) is the transmittance, as a function of \( v \), of the radiometer's lower density state, i.e. with a low amount of gas. Unitless.

\( \tau_{sh}(v) \) is the transmittance, as a function of \( v \), of the radiometer's higher density state, i.e. with a high amount of gas. Unitless.

The ratio \( S_{diff}/S_{avg} \) is then correlated with the gas concentration between the radiation source and the LMR.

A significant problem with all correlation radiometers is optical imbalance which is any signal produced by the radiometer at the frequency of gas modulation which is not due to the absorption of radiation by the gas in the gas cell. Such signals are often a result of direct and indirect influences from the gas modulation mechanism. Much of the effort described in this research and by Tolton (1990 and 1995) relates to characterizing and reducing optical imbalance in a length modulated radiometer (section 2.3.4). The ideal \( S_{diff}/S_{avg} \)--which is correlated with gas concentration--can actually be calculated from the measured \( S_{diff}/S_{avg} \) if the evacuated imbalance of the radiometer is known (Tolton, 1995) as shown in equation [2-4].

\[
[2-4] \quad \frac{S_{diff\, ideal}}{S_{avg}} = \left( \frac{2 - K}{2} \right) \times \frac{S_{diff\, measured}}{S_{avg}} - K
\]

\( K \) is the radiometer's evacuated imbalance. Unitless.

2.3.2 The Selective Chopper Radiometer

The first form of correlation radiometry was the technique of selective chopping (Smith and Pidgeon, 1964) which implies the name selective chopper radiometer (SCR). The density of gas through which the beam passes is modulated by moving cells containing different gas densities into and out of the optical beam. Either the gas cells are mechanically moved while the beam remains stationary or the beam's path is mechanically switched between two gas cells.

Although SCRs allow a wide range of gas densities to be used, each cell may have different optics resulting in a large optical imbalance. The first SCR instrument was part of the
NIMBUS 4 satellite and it measured temperatures in the stratosphere by chopping the beam between two cells filled with carbon dioxide (Drummond, 1992). Similar data were obtained from the NIMBUS 5 satellite which included an SCR instrument that kept the beam stationary but placed different gas cells in the beam’s path (Drummond, 1992).

2.3.3 The Pressure Modulated Radiometer

Pressure modulated radiometers (PMRs) contain a single gas cell in which the gas pressure is cycled with pistons. There are no moving optics in PMRs, which reduces the imbalance, but the gas emits radiation at the same frequency of the density modulation due to adiabatic heating induced by the pressure changes. The data analysis is further complicated by the pressure cycling because there are an infinite number of pressure states. Finally, the mean pressure in the PMR cannot exceed 10 kPa without problems arising with the pressure modulation mechanism (Tolton, 1995). Consequently, satellite-based PMRs are best suited for remote sensing of the upper atmosphere.

PMR instruments were successfully used for measuring the temperature of the mesosphere and the stratosphere on the NIMBUS 6 and NIMBUS 7 satellites (Taylor, 1983). PMRs on balloons and satellites have been used extensively for composition and temperature measurements of the upper atmosphere, and one instrument was placed aboard the Pioneer Venus orbiter for establishing vertical temperature profiles in the upper atmosphere of Venus (Taylor et al., 1979).

2.3.4 The Length Modulated Radiometer

The latest development in radiometers comes with the length modulated radiometer (LMR) being developed at the University of Toronto (Drummond, 1989). The beam’s path length through the gas is modulated inside the gas cell by rotating an optically inert rotor through the beam. To eliminate large optical differences between the long path and the short path, a
compensator cell and rotor are added into the beam's path, as illustrated in Figure #2-3. The compensator cell is placed after the gas cell, from the perspective of the incoming beam. The compensator rotor is aligned so that when the gas cell rotor intersects the beam (the short gas path), the beam does not travel through the compensator rotor, and vice versa (the long gas path). Ideally, there is no difference inside the length modulated cell (LMC) between the short gas path's optical path and the long gas path's optical path. The compensator cell is either evacuated or filled with a gas which is optically inert at the wavelengths being detected.

LMRs can operate at higher pressure levels than PMRs and so can be used for satellite-based remote sensing of the troposphere. As well, the only moving components of the instrument are the rotors which immensely simplifies the mechanical design and operation of the instrument. Four LMRs are planned for the MOPITT project (section 2.4).

2.4 The MOPITT Project

The MOPITT (Measurements of Pollution in The Troposphere) satellite instrument is scheduled to be launched in 1998 on NASA's Earth Observing System satellite EOS-AM1. EOS-AM1 is one of several satellites in sun-synchronous polar orbit which will form the Earth Observing System, EOS. EOS will contain numerous instruments providing continuous, global, wide-ranging observations of the Earth for at least a decade (ESSC, 1988). Results from MOPITT will assist in the long-term, high-resolution modelling of tropospheric chemistry.

The MOPITT instrument will consist of six correlation radiometers: two pressure modulated radiometers (PMRs) and four length modulated radiometers (LMRs). The objective is to obtain, with a vertical resolution of 3-4 km at altitudes of 0-15 km, tropospheric column measurements of CO to a precision of ±10% and of CH₄ to a precision of ±1% (Drummond,
**Figure #2-3:** (adapted from Drummond, 1989)
A schematic of the length modulated cell (LMC) with two rotors.

The gas path is modulated between A and A-B in the gas cell. The rotors are aligned so that the thickness of rotor through which the radiation passes is always B.
1992). Both the 4.7 µm (2130 cm⁻¹) and 2.4 µm (4170 cm⁻¹) regions will be examined for the CO measurements while the 2.3 µm (4350 cm⁻¹) region will be examined for the CH₄ measurements (Drummond, 1992). The profiles will be used for modelling CO and CH₄ in order to glean a better understanding of these pollutants and of their influence on tropospheric chemistry.
3. THE LENGTH MODULATED RADIOMETER

3.1 Introduction

The Mk III LMC and the LMR optical table setup characterized by Tolton (1995) were used for this research. The Mk III LMC was built by COM DEV for use in a ground-based LMR. The instrument had not been used in approximately one year prior to October 26, 1995, hence a series of tests was run to check for changes in the instrument's behaviour. In order to understand the nature of the investigations discussed in this report, the LMR is briefly described in section 3.2. Further information, including equipment specifications, circuit diagrams, and detailed component characteristics, can be found in Tolton (1995).

3.2 Description of the LMR

The information in this section is a summary of that presented in Tolton (1995).

3.2.1 Overview

This instrument was used mainly for the ground-based remote sensing of atmospheric CO by determining the absorption by atmospheric CO of solar radiation in the 2.4 μm (4170 cm⁻¹) region. The setup of the instrument on the optical table is displayed in Figure #3-1. The table is surrounded by an aluminum box which is insulated with styrofoam, and which is mounted on a cart that also holds most of the vacuum system. Most of the electronics are attached to the optical table, but the main power supply, the computer, and the oscilloscopes sit on an independent cart.

3.2.2 Optical Path

Figure #3-2 is a ray-trace diagram of the instrument. The lenses are 38.1 mm in diameter with focal lengths of 100 mm. The double-pass through the LMC eliminates imbalance due to
Figure #3-1: (Tolton, 1995)
The LMR set up on the optical table.
Figure #3-2: (adapted from Tolton, 1995)
A ray-trace diagram of the length modulated radiometer (LMR).

Figure #3-3: (Tolton, 1995)
The Mk III LMC mounted on an aluminum frame. The gas tube extends out to the left.
non-parallelism between the rotors while the 5° twist reduces imbalance due to single reflections. The filter is a Tor 1/1 narrow band-pass filter, is mounted inside the detector unit, and is cooled by liquid nitrogen to reduce noise. The infrared detector is model SDD-7854-S1 manufactured by Cincinnati Electronics and its 1 mm diameter InSb detector element is cooled by liquid nitrogen.

3.2.3 The Mk III Length Modulated Cell

Figure #3-3 is a photograph of the LMC while a cross-section appears in Figure #3-4. Calcium fluoride (CaF$_2$) was chosen for its optical inertness in the regions of the CO absorption bands (2130 cm$^{-1}$ and 4170 cm$^{-1}$). In these regions, the material has high transmittance—approximately 94% (Musicant, 1985)—low reflectivity, low absorptivity, and a low index of refraction (1.40354 at 2174 cm$^{-1}$ and 1.42168 at 4167 cm$^{-1}$ (Bezuidenhout, 1991)). The three walls (gas cell wall, inner wall, and compensator cell wall) are made of 6061 aluminum and partition the LMC into two cells: the gas cell which is filled with the desired gas or is evacuated, and the compensator cell which in this instrument is not sealed to the atmosphere. There are two optical paths through the LMC, but the upper path in Figure #3-3 is blocked by copper plugs.

The rotors’ rate of rotation is approximately 10 Hz, but can be adjusted with the LMC drive controller (see Figure #3-1). The symmetry of the rotor shape (Figure #3-5) balances the rotor but the four sectors force the beam to switch optical paths (a long-to-short or a short-to-long transition is a switch) at a rate four times the rotors’ rotation rate—approximately 40 Hz. The gas cell and compensator rotors are 90° out of phase to ensure equal optical path lengths through the long and short paths. The gas cell has an optical path length of 10.0 mm and the rotors are 8.0 mm thick, so the optical path through the gas alternates between 10 mm and 2 mm.
Figure #3-4: (Tolton, 1995)
A cross-section of the length modulated cell (LMC).

Correlation Cell
CaF$_2$ Windows
Optical Path
Bearings
Magnetic Drag Coupling
CaF$_2$ Gas Cell Rotor
Optical Path (alternate)
Vacuum Seal
Gas Cell Wall
Inner Wall
Compensator Cell Wall
Compensator Cell
Motor Stator
Motor Rotor
Optical Encoder
CaF$_2$ Compensator Rotor

150.4 mm
54.4 mm
Figure #3-5: (Tolton, 1995)
The shape and dimensions of the gas cell and compensator CaF$_2$ rotors.

Dimensions are in millimeters and are ±0.03 mm.
The rotors are 8.000 mm thick and are ±0.001 mm or better.
Figure #3-3 (but not Figure #3-4) illustrates the flexible tubing that permits evacuation and filling of the gas cell. The tube extends out from the LMC and is attached to a vacuum system. A pressure head is also joined to the vacuum system, as shown in Figure #3-1. The gas temperature in the gas cell is measured with a thermistor attached to the outside of the gas cell wall. The optical coder in Figure #3-4 registers a pulse at every half-turn of the rotors and the pulses are electronically converted to a square wave with a period equivalent to one full rotation.

3.2.4 Electronic Signals

Figure #3-6 displays the tuning fork chopper index, the LMC index, the detector signal, and the accept/reject regions for just more than one full chopper cycle. During transitions between rotor sectors, the beam scatters off the rotor edges reducing the signal, and so these sections are always rejected. The four rotor sectors create four such transitions occurring at a rate four times the rotor rotation speed, or approximately 40 Hz.

Figure #3-7 details the detector signal and chopper index. The sum offset and sum total parameters entered for the FASTC2.C routine (section 3.3.1) are illustrated: the right edge of A represents the offset and the width of B represents the total. The detector's amplifier is inverting, resulting in the negative voltages, and the peaks of the sine wave are flat because the chopper vanes are entirely closed for this period of time (but still moving, so they are overlapping one another).

The data are passed into a Motorola DSP56001 microprocessor digital signal processing card which was programmed in assembler to store the data stream and to calculate average signals during the open and closed states of the chopper. The card is attached to an IBM-compatible computer with an Intel 386 chip. After passing through the DSP56001 card, the data are further processed and graphed in a spreadsheet.
The frequencies are 600 Hz for the tuning fork chopper index bit and approximately 10 Hz for the LMC index which corresponds to the LMC rotation rate. The approximate detector signal is also displayed, but is negative because the detector has an inverting amplifier. This signal from the detector comes from both the 600 Hz tuning fork chopper and the 40 Hz transition between the rotor sectors (there are 4 sectors per LMC rotation). Based on the LMC index, the accept regions and reject regions are shown. The reject regions are user-defined and represent the times at which detector data is rejected due to a transition between rotor states.
Chopper open and closed signals are calculated by summing the detector signals in the extrema of the detector signals. The number of data points summed, and which data points are summed, are defined by the variables A and B which are experimentally determined to minimize noise. Typically, A is 3 data points after a chopper index transition and B is 24 data points long.
3.3 Software

3.3.1 C Programs

Two routines programmed in ANSI Standard C collect data from the LMR: ADC_SUM.C and FASTC2.C.

The ADC_SUM.C routine is run with the chopper off and collects a continuous string of detector signals over one rotation of the LMC. The results are averaged for a user-defined number of rotations, yielding a graph of output signal versus time for one LMC rotation. Optical anomalies on the rotors—such as dust or smudges—can thus be identified by sharp deviations in the signal. The number of rotations used for the tests presented here was ten.

The FASTC2.C routine collects instrument signals with the chopper on for four rotations of the LMC and then removes linear drift in each rotor sector. From these values, the imbalance is calculated in a spreadsheet for a large number (typically 85) of successive measurements. The average and standard deviation of the imbalance measurements are then calculated. This routine can also be run almost continuously (by requesting that several thousand data sets be collected) while optical adjustments are made. In this manner, the effect on imbalance of alterations to the optics can be observed immediately. When running the routine, the user also inputs the parameters for rejection fraction (normally 0.06), fractional shift (normally 0), sum offset (normally 3), and sum total (normally 24).

3.3.2 GENASIS

GENASIS is a series of programs used to calculate the theoretical $S_{diff}/S_{avg}$ ratio. The user defines the spectroscopic instrument, the spectral filters, the atmosphere, the radiation source, the interaction between the atmosphere and radiation, and the spectral range and resolution. Integrations are then completed one spectral line at a time over the defined spectral range at the
defined spectral resolution. The GENASIS results are used to calibrate the LMR each time an atmospheric measurement is taken.

3.4 Re-Investigating the Imbalance of the LMR

The effects of optical setup and software parameters on the LMR’s imbalance were examined, since the LMR had not been functional for approximately one year. The graphs referred to in this section appear at the end of this chapter. Data points on the graphs represent arithmetic mean values and error bars represent one standard deviation above and below the arithmetic mean.

3.4.1 The Effect of Setup and Software Parameters

The LMC was evacuated for these measurements. Aligning the optics and the detector, and optimizing software parameters, resulted in imbalance measurements consistently in the range $1 \times 10^{-4} \rightarrow 3 \times 10^{-4}$ with standard deviations approximately one order of magnitude lower. The optimal setup and software parameters determined in this research compare very well to those attained by Tolton (1995) during his work, and they are summarized in Table #3-1.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Optimal Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>rotation of LMC about its vertical axis</td>
<td>$203^\circ$ ($5^\circ$ from being $\perp$ to the optical axis)</td>
</tr>
<tr>
<td>vertical tilt of the LMC out of the optical plane</td>
<td>none</td>
</tr>
<tr>
<td>sum total for FASTC2.C</td>
<td>24</td>
</tr>
<tr>
<td>sum offset for FASTC2.C</td>
<td>3</td>
</tr>
<tr>
<td>fractional shift for FASTC2.C</td>
<td>0</td>
</tr>
</tbody>
</table>
The effect of rotating the LMC around its vertical axis in order to eliminate imbalance due to single reflections was examined (Graphs #3-1a, #3-1b). With an angle of 208° representing a state with the rotors perpendicular to the optical axis, the optimal angle appeared to be 203° → 205°. The LMC was placed at 203° to retain conditions similar to Tolton’s (1995) work.

The effect of rotating the LMC around the horizontal axis which is perpendicular to the beam was examined to see if the imbalance would be reduced for various detector positions about the detector peak. The results are in Graph #3-2 and indicate that the only effect of tilting the LMC is to change the offset of the imbalance vs detector position curve. The tilting eliminates the effects of single reflection, in a similar manner as rotating the LMC around its vertical axis, but the effect will be the same regardless of the method used; there is no advantage in using both techniques (Tolton, 1996).

The parameters entered in FASTC2.C were briefly examined (Graphs #3-3, #3-4, #3-5). Graph #3-3 shows that with a sum total of 10, the lowest standard deviations are obtained with a sum offset of 10 → 12 implying that the best portion of the wave is bits 15 → 17. In order to retain more data, though, Tolton (1995) recommends using a sum total of 24, which requires a sum offset of 3 → 5 in order to retain the 15 → 17 portion in the centre of the accept region. Graph #3-4 varies offset with a sum total of 24 indicating that the optimum is likely a sum offset of 3. Finally, Graph #3-5 seems to confirm that a fractional shift of 0 yields the lowest standard deviation.

3.4.2 The Effect of Gas and Gas Pressure

The influence of nitrogen pressure on the imbalance was examined by alternating various pressures of nitrogen with a vacuum and running FASTC2.C. Graph #3-6a is reasonably linear,
but masks the fact that the imbalance of the vacuum decreased consistently with time (Graph #3-6b). The difference in imbalance from the vacuum to each pressure, however, was very close to the theoretical values (Graph #3-6c) indicating that the LMR is performing very well.

Carbon monoxide was examined next by cycling through a vacuum, nitrogen at a pressure, and then carbon monoxide at the same pressure. The carbon monoxide results were approximately 2% higher than theoretical predictions by the GENASIS software (Graph #3-7). This discrepancy is most likely a result of neglecting to measure the LMC’s temperature, which then had to be estimated for using GENASIS.
Graph #3-1a: Average Imbalance vs LMC Twist Angle
Graph #3-2: Vertically Tilting the LMC
Average Imbalance vs Detector Position

- Without Tilt
- With Tilt
4. MODIFYING AND TESTING THE LMR

4.1 Using a Fibre Optic Cable

The two halogen lamps and the diffuse reflecting surface have been replaced by a single halogen lamp and a fibre optic cable leading to the LMR as shown in Figure #4-1. The fibre optic cable is a UV-Vis FIBER from Fiberguide Industries, but it also works in the near infrared region of CO and CH₄ absorption. The three irises shown in Figure #4-1 are used both to prevent saturation of the signal processing electronics and to vary the optical beam’s diameter in the LMR. A peak-to-peak voltage of less than 3.5 V was found to avoid saturation problems, and for most of the work presented here, the peak-to-peak voltage was maintained between 2 and 3 V (the detector saturates at 6 V, so there was no fear of saturating the detector at these signal levels). The irises’ diameters were altered in order to determine their effect on the imbalance. All three irises affect the imbalance, but the effects were often very small, and the exact influence of each iris was difficult to determine. Quantitative investigations of the effect of iris size on imbalance were conducted in section 4.7.

The fibre optic cable provides a much smaller beam diameter than the diffuse reflecting surface which could be a problem since Tolton (1995) recommends that a wider beam should be used in order to reduce imbalance from transmission variations over the LMC’s rotor. In actual fact, the small beam diameter did not appear to present much of a problem: the LMC was raised, lowered, or rotated in order to find the most consistent section of the rotor as determined by the ADC_SUM.C results. In order to eschew potential problems with the smaller beam diameter in the future, the optical setup was modified by adding a lens just before the gold roof mirror and realigning the optics to move the beam’s focal point away from the LMC, as detailed in section 4.4 paragraph (e).
Figure #4-1: The LMR with the fibre optic cable (adapted from Tolton, 1996).
4.2 Rotating Vane Chopper

Tolton’s studies (1995) employed a tuning fork chopper in the LMR (see Figure #3-1). The use of this chopper resulted in problems with chopping the beam evenly. In the chopping cycle, the chopper was not closed for very long, meaning that there was not much time to take the measurements and a good signal-to-noise ratio could not be obtained. Additionally, the chopper never opened to a position which permitted the entire beam to pass through. Therefore, the beam intensity reaching the detector was always attenuated by the chopper. This situation is depicted in Figure #3-7. Because the signal continually varies throughout the open state, any time-based jitter in recording the data during the open state will substantially add to the noise level. To summarize, there is a significant deviation from the consistent square wave required for a high signal-to-noise ratio.

To alleviate the problems, the tuning fork chopper was replaced with a Model 300 Super Chopper from Palo Alto research. This chopper is a rotating vane chopper which can be driven by an external frequency. A vane with 14 slots--7 open and 7 closed--and with a frequency range 14 Hz to 746 Hz was run at a chopping rate of 600 Hz (i.e. the rotation rate was 600/7 = 85.7 Hz). The chopper cycle could be phase-locked to the digital signal processing output (see section 4.3) and the vane was shielded with a protective cover. The slots’ outer diameter is 1.52 cm and their inner diameter is 1.50 cm. The chopper slots are much larger than the beam diameter, hence the length of time for switching between open and closed states is less than one tenth the time of the full cycle (Tolton, 1996). The detector signal is therefore much more square than it was with the tuning fork chopper allowing the signal-to-noise ratio to be much higher.

The chopper and its mount form a block which is 21.6 cm perpendicular to the beam horizontally, 8.89 cm parallel to the beam horizontally, and 21.6 cm vertically. This block was
attached to a base on the optical table thereby keeping the chopper steady and raising the blade to a level at which it could chop the beam. The chopper’s drive box was also attached to the optical table. Photographs of the chopper and its drive box are shown in Figure #4-2.

4.3 Reduction of Noise in the Data Stream From Solar Radiation

Fluctuations in the solar radiation at ground level were the largest source of noise in field tests of the ground-based instrument (Tolton, 1996, 1995). The fluctuations were theorized to be caused by rapid changes in the scattering properties of the atmosphere. Much of this noise can be eliminated in theory by adding a second channel to the instrument without a gas cell, as illustrated in Figure #4-3 (with some optical changes; see section 4.4). Detector #2 would measure variations in the incoming radiation. These variations would be factored into the imbalance measured yielding a corrected value for the ideal LMR imbalance from equation [4-1] (Tolton, 1996).

\[
S_{I(ideal)} = \frac{S_{I(measured)} - X_I}{1 - \frac{S_{I(measured)}X_I}{4}}
\]

\[4-1\]

\(S_{I(ideal)}\) is the LMR’s actual imbalance: the ratio \(S_{\text{diff}}/S_{\text{avg}}\) (section 2.3.1). Unitless, \(S_{I(measured)}\) is the LMR’s measured imbalance: the ratio \(S_{\text{diff}}/S_{\text{avg}}\) (section 2.3.1). Unitless. \(X_I\) is the corrective factor obtained from measurements by Detector #2. Unitless.

Note that \(S_{I(ideal)}\) becomes the term "\(S_{\text{diff}}/S_{\text{avg}}\) measured" in equation [2-4]. The main impetus behind the modifications described in further sections of this chapter was to accommodate the second channel. No experiments with the two channels in operation have yet been conducted.
Figure #4-2A:
The rotating vane chopper and its drive box on the optical table.
Figure #4-2B:
The chopper's drive box.

Figure #4-2C:
The rotating vane chopper.
**Figure #4-3:** (modified from Tolton, 1996)
Using a second detector to eliminate noise external to the LMR.
4.4 Physical Layout on the Optical Table

Figure #4-3 and the photographs in Figure #4-4 display the revised layout on the optical table. Figure #4-4A can be contrasted with Figure #3-1 to see the full extent of the alterations. These alterations are:

(a) The rotating vane chopper replaces the tuning fork chopper (section 4.2).

(b) A second channel and detector have been added to reduce noise external to the instrument (section 4.3). A half-mirror splits the signal between the two channels.

(c) The individual power supplies have been replaced by a single power supply box (Figure #4-4B). This box provides numerous power values by choosing two outlets from white (+12 V), black (-12 V), red (+5), blue (-5), and yellow (+1). Green represents ground. The detectors, though, retained their own power supply to minimize the potential for damage due to inaccurate or inconsistent voltage outputs.

(d) The covering box for the optical table had two tiers, so that the section of the box over the detector was higher than the box over the rest of the table. The second detector (section 4.3) was placed underneath the lower section of the covering box, which unfortunately was too low to accommodate the detector. Therefore, a new covering box which is a single height across the entire optical table was necessary. The design and construction of this box is in progress.

(e) An extra lens between the half-gold mirror and gold roof mirror has been added. The new setup eliminates the focal point at the LMC shown in Figure #3-2. Therefore, the beam is more diffuse as it passes through the LMC and any noise due to scratches or dust on the rotors will be smoothed out. Figure #4-5 is a ray-trace diagram of the new optical setup.

(f) The iris preceding the chopper (see Figure #4-1, the chopper is now a rotating vane style) was removed after it was established that it made negligible difference to the signal seen by either detector.

(g) The optical setup was optimized to attain minimum imbalance by repositioning and realigning the anti-reflection coated germanium lenses and both mirrors.

(h) The halogen bulb, first iris, and input to the fibre optical cable were attached to an optical bar which was placed underneath the optical table (Figure #4-4C) thereby separating the optical input to the LMR from the optical table. Therefore, changing the input from the bulb--which will be used to calibrate the instrument--and, for example, a telescope (see section 5.3) will not affect the optical table. The output from the fibre optic cable was aimed through the second iris attached to the optical table as depicted in Figure #4-4D).
Figure #4-4A: Overview of the revised setup on the optical table.

Figure #4-4B: The single power supply box.

This box supplies voltages to all equipment on the optical table except for the detectors. The circuit on top (section 4.7) inverts the LMC pulse so that the SPM can detect it properly.
Figure #4-4C: Input to the fibre optic cable.

Figure #4-4D
Output from the fibre optic cable.

The halogen bulb, first iris, and input to the fibre optic cable attached to an optical bar and placed underneath the optical table.

The output from the fibre optic cable through the second iris and an anti-reflection coated germanium lens on the optical table. The chopper’s drive box is at the top left corner.
Figure #4-5:
A ray-trace diagram of the LMR with the new optical setup.

On the optical table.

From Below the Optical Table
(A single halogen bulb radiation source and an iris precede the fibre optic cable).
4.5 Digital Signal Processing Software

The LMRs use digital, rather than analog, electronics so that: (a) the signal processing can be described mathematically more easily; (b) the circuit behaviour can be replicated between two or more channels built similarly; and (c) the signal detection, processing, and analysis are simplified. The signal processing software was upgraded from the C programs described in section 3.3.1 to LabVIEW® programs written by Dave Trickey and Jim Drummond. The upgrade added accuracy and speed to the data collection by allowing close to 100% of the incoming data to be processed, rather than the ≈10% obtained with the old system (Tolton, 1996).

Viewing the output and state of the system was much more efficient with the new software (see Figure #4-6). For example, the C programs had to be rerun every time a software parameter was changed, as occurred for the studies in section 3.4. With LabVIEW®, these parameters could be easily modified as data was being collected; for example, Figure #4-6B demonstrates changing the chopper parameters (more details are provided in section 4.7 during the discussion of investigating these parameters). As well, data from the C programs had to be imported into and graphed in a spreadsheet. LabVIEW® displays graphs and output values as the program runs (Figure #4-6) allowing the effects of optical or software parameter changes to be determined instantaneously. Although the graphs cannot be saved, the data can be recorded to a file and then imported into a spreadsheet, as with the C programs. To further ease data analysis, Boyd Tolton wrote a C program called CVT.C which calculates the imbalance’s average and standard deviation from a data file recorded in LabVIEW®. The LabVIEW® software was further modified by Dave Trickey to enable simultaneous data processing for two channels, a necessity for reducing noise external to the instrument (section 4.3).
Figure #4-6A:
The LabVIEW® software in operation.

Figure #4-6B:
Changing the chopper parameters with the LabVIEW® software.
4.6 Computer and Electronic Hardware

A signal processing module (SPM) electronic board was built for each LMR channel. The SPMs output data at 375 kHz and combine with the new digital signal processing software (section 4.5) to replace the DSP56001 computer card used by Tolton (1995) for his research (section 3.2.4). The computer with an Intel 386 chip, which was used for the data processing and analysis described in chapter 3, was replaced with a computer that has an Intel Pentium chip running at 100 MHz, so that the computing power required to process incoming data from two channels would be available. The computer and SPMs sit on an independent cart beside the optical table. A schematic of the new electronics setup is shown in Figure #4-7.

One minor modification to the electronic signals was implemented. The LMC index shown in Figure #3-6 is a square wave at the frequency of the LMC rotation rate (=10 Hz), so there are 2 pulses per rotation (one for the "high" signal, one for the "low" signal). In order to adhere to the requirements of the SPM, the LMC had to send out a pulse approximately 800 µs in length, once per rotation. This pulse is actually emitted by the LMC as a "low" anomaly from a state which is usually "high", but the SPM detects a "high" pulse. To solve the problem, a 7414 Hex Schmitt-Trigger was used to invert the signal and this circuit rested atop the power supply as shown in Figure #4-4B.

4.7 Optimizing Setup and Software Parameters

The effects of optical setup and software parameters on the LMR’s imbalance were examined following all the alterations described in this chapter. The LMC was left open to air for these measurements. Aligning the optics and the detector, and optimizing software parameters, resulted in imbalance measurements consistently in the range $1 \times 10^{-4} \rightarrow 2 \times 10^{-4}$ with
Figure #4-7:
Schematic of the electronics setup.
standard deviations approximately the same order of magnitude. The optimal setup and software parameters are summarized in Table #4-1. The graphs referred to in this section appear at the end of this chapter. Data points on the graphs represent arithmetic mean values and error bars represent one standard deviation above and below the arithmetic mean.

Table #4-1:
New Setup and Software Parameters for the LMC

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Optimal Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>diameter of the second iris (the diameter of the first iris was changed to maintain a 2 V peak-to-peak signal and was 6.55 mm wide)</td>
<td>3.55 mm</td>
</tr>
<tr>
<td>rotation of LMC about its vertical axis</td>
<td>212° (4° from being ⊥ to the optical axis)</td>
</tr>
<tr>
<td>vertical position of the LMC</td>
<td>No clear optimum, although a position between 0.00 mm and 4.00 mm from the lowest position seemed preferable.</td>
</tr>
<tr>
<td>horizontal position of the LMC</td>
<td>The position in which the optical axis is approximately centred on the LMC window.</td>
</tr>
<tr>
<td>number of LMC rotations used for averaging data</td>
<td>As many as feasible, given that data processing slows considerably with more rotations, and that the software consistently crashed with 10 rotations or more.</td>
</tr>
<tr>
<td>accept/reject data within a chopper cycle</td>
<td>Reject the first 80, accept 200, reject the next 100, and accept 200.</td>
</tr>
</tbody>
</table>

The effect on imbalance of the diameter of the iris at the fibre optic cable’s output was examined. The diameter of the iris at the fibre optic cable’s input was continually altered to ensure that the detector always reported a 2 V peak-to-peak signal. Graph #4-1 displays the results and shows that the smallest diameter for the iris yields the lowest imbalance. Wider diameters, though, yield a reasonably consistent imbalance which is not much larger.
The effect of rotating the LMC around its vertical axis in order to eliminate imbalance due to single reflections was examined (Graphs #4-2a and #4-2b). With an angle of 208° representing a state with the rotors perpendicular to the optical axis, the optimal angle appeared to be 212° resulting in an imbalance on the order of $10^{-5}$ which was sporadically repeatable. The standard deviation, though, was consistently $3\times10^{-4}$ whenever an average imbalance of $10^{-5}$ was achieved. The LMC was placed at 212° for the rest of the data collection.

The vertical (Graph #4-3) and horizontal (Graph #4-4) positions of the LMC were investigated. The data sets had large scatters, likely because as the LMC moves, the beam intersects different portions of the rotors. Therefore, varying degrees of scratches, dust particles, or other inconsistencies in the beam's path will occur resulting in different imbalances. Vertically, there was no clear optimum, although between 0.00 mm and 4.00 mm up from the lowest position yielded reasonably consistent results. Horizontally, placing the LMC so that the optical axis was as centred as possible on the window seemed to yield the best results.

The parameters entered in LabVIEW® were briefly examined. Graph #4-5 shows that the imbalance's average and standard deviation show a general, though inconsistent, trend to decrease as the number of rotations used for averaging the data increases. Tolton (1996) states that statistical laws dictate that the imbalance decrease is approximately proportional to the square root of the increase in the number of rotations. Seemingly, it is best to average as many rotations as possible. The trade-offs are that (a) the processing time increases as the number of rotations increases, and (b) the LabVIEW® software tends to crash with more than 10 rotations because it cannot handle the volume of incoming data.

The next task was to determine which portions of the signal within the chopper cycle should be retained and used to calculate imbalance. This problem is akin to determining the
variables A and B in Figure #3-7, however, as displayed in Figure #4-6B, the LabVIEW® software requires the user to define the length of both accept and both reject regions (which are delineated by the dotted lines in Figure #3-7; each full chopper cycle (open plus closed) has two accept and two reject regions). The accept regions should be as large as possible to increase the signal-to-noise ratio, but must not encompass any portion of the transition regions between open and closed states or else the noise will be large.

This task was achieved by recording data from an extremely thin accept region. The thin accept region was scanned across a cycle by adjusting the reject regions’ sizes. Therefore, the quality of data at several points throughout the chopper cycle was ascertained. The width of the thin accept region was 10 data points, but because the LabVIEW® software records approximately nine times as much data as the C programs used in section 3.4, the width of accept/reject regions described here cannot be directly compared to the variables A and B from Figure #3-7.

Graph #4-6 displays results for investigating the open portion of the chopper cycle. With the first accept region 10 data points wide and the second accept region 100 data points wide, the imbalance was recorded as a function of the width of the first reject region. As the width of the first reject region was increased, the width of the second reject region was decreased so that the absolute position of the second accept region did not move. From Graph #4-6, setting the first reject region to 80 and the first accept region to 200 appears to provided the largest accept region while avoiding transition sections.

Graph #4-7 displays results for investigating the closed portion of the chopper cycle. The first reject region was set at 80 and the first accept region was set at 200 to use the results from Graph #4-6. With the second accept region set at 10 and the second reject region was increased. The software returned an error—it could not lock onto the data stream—until the second reject
region was 90 data points wide, and so this portion of Graph #4-7 is blank. Recording the region
which provides the most consistent average and standard deviation for the imbalance entails
setting the second reject region to 100 and the second accept region to 300.

4.8 Conclusions

Major changes and additions have been incorporated into the LMR. These changes
simplify some design aspects, improve the optics, streamline the data processing, and permit the
potential for significant reduction of noise. By adjusting setup and software parameters, the
imbalance can consistently be made below $3 \times 10^{-4}$ with a standard deviation close to this
magnitude. Therefore, while there are still many improvements and adjustments to be completed,
preliminary results indicate that the LMC should be able to achieve ground-based measurements
of CO and CH$_4$ within the desired accuracy.
Graph #4-2a: Average Imbalance vs LMC Twist Angle
Graph #4-2b: Imbalance Standard Deviation vs LMC Twist Angle

Imbalance Standard Deviation

LMC Twist Angle
Graph #4-6: Selecting Data Points for the 'Chopper Open' Portion of the Cycle

Average Imbalance

Number of Data Points of the First Reject Region
Graph #4-7: Selecting Data Points for the 'Chopper Closed' Portion of the Cycle
5. FUTURE RESEARCH WITH THE LMR

5.1 Introduction

The LMR will eventually be used as a ground-based instrument for remote sensing of CO and CH\textsubscript{4} in the atmosphere of downtown Toronto. The design was initially developed to provide a model of a single channel of the MOPITT instrument which could be characterized and manipulated in ways that are impossible for the MOPITT instrument. The goals for future research are to continue improving the ground-based instrument and to provide more information for the modelling of the MOPITT instrument. Both instruments will measure the atmospheric CO column to a precision of ±10% and the atmospheric CH\textsubscript{4} column to a precision of ±1% (Drummond, 1992). A major component of the future research with the LMR will therefore be ground-based measurements of atmospheric CO and CH\textsubscript{4}, from the patio of the McLennan Physical Laboratories at the University of Toronto, or any other accessible and useful location.

5.2 Permanent Outdoor Facility

During the summer of 1995, a hut to house the instrument was built on the north roof of the McLennan Physical Laboratories at the University of Toronto. The hut was secured to the roof with chains. The facility was monitored throughout the autumn of 1995 and it became apparent that, likely due to wind, the facility was shifting significantly. As well, some nuts fell off allowing the respective bolts to fall down inside the hut where they are difficult to access. The nuts ended up frozen to ice on the roof. The overall result is that the roof and walls of the hut are not as firmly attached to each other as they were when the facility was first finished. Continual monitoring and maintenance are required.
Due to problems with access to the roof site, the ground-based LMR will have to be dismantled for transporting it up to the outdoor facility. Therefore, the move to this site is virtually permanent and will not occur until the researchers are confident that as much work as possible has been completed in the lab and on the patio of the McLennan Physical Laboratories.

5.3 Using a Telescope to Focus the Sun

An 8" (20.32 cm) Newtonian telescope will be used for actively tracking the sun in outdoor measurements. The telescope has a crystal controlled driver which can be used to ensure that the sun's rays are continually and accurately focussed into the fibre optic cable of the LMR.

5.4 Phase Locking

The chopper, with a frequency of 600 Hz, will be phase-locked to the SPM which operates at a frequency of 375 kHz (section 4.6). The LMC, rotating at ~10 Hz, might eventually be phase-locked to the SPM as well.

5.5 Continuing Investigations into Sources of Imbalance

Attempts are continually underway to reduce the imbalance of the LMR. Therefore, the components in the optical beam (e.g. lenses and mirrors) may be reinvestigated to determine if improvements can be made. As well, McKernan (1996) suggested that if no other sources of imbalance can be discovered, it might be necessary to examine whether or not the gas inside the LMC is well mixed. In the very unlikely event that there are pressure discrepancies within the gas cell itself--perhaps due to compression of the gas by the rapidly rotating rotors--the imbalance may be augmented.
SUMMARY OF RESULTS AND CONCLUSIONS

The length modulated radiometer which uses the Mk III length modulated cell was studied extensively for sources of imbalance in attempts to reduce the imbalance. The optical setup and software used by Tolton (1995) was reexamined in order to determine the optimal parameters for minimizing imbalance. The optimal parameters obtained by Tolton (1995) were confirmed and are summarized in Table #3-1, which is reprinted below.

Table #3-1:
Setup and Software Parameters for the LMC

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Optimal Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>rotation of LMC about its vertical axis</td>
<td>203° (5° from being ⊥ to the optical axis)</td>
</tr>
<tr>
<td>vertical tilt of the LMC out of the optical plane</td>
<td>none</td>
</tr>
<tr>
<td>sum total for FASTC2.C</td>
<td>24</td>
</tr>
<tr>
<td>sum offset for FASTC2.C</td>
<td>3</td>
</tr>
<tr>
<td>fractional shift for FASTC2.C</td>
<td>0</td>
</tr>
</tbody>
</table>

The LMR’s response to various nitrogen and carbon monoxide pressures conformed reasonably well to theoretical predictions.

Several alterations were then done to the LMR. The most significant ones were:

(a) The two halogen bulbs and the diffuse reflecting surface were replaced by a single halogen lamp shining through an iris and then into a fibre optic cable. The output of the fibre optic cable was directed into the LMR.

(b) The tuning fork chopper was replaced by a rotating vane chopper.

(c) Another anti-reflection coated germanium lens was added—primarily to eliminate the beam’s focal point at the LMC—along with another iris, and the optical setup was subsequently reoptimized.
(d) The digital signal processing software was upgraded to permit more efficient data processing and analysis and to allow easier investigations into the effects on imbalance of altering the software parameters.

(e) The digital signal processing computer card (DSP56001) was replaced by a signal processing module which outputs data more rapidly and in a manner compatible with the upgraded software.

(f) The computer with an Intel 386 chip was upgraded to one with an Intel Pentium chip in order to effectively and efficiently handle the data stream.

The LMR with these changes was reexamined in order to determine the optimal parameters for minimizing the imbalance. The optimal parameters are summarized in Table #4-1, which is reprinted below in a slightly modified form.

<table>
<thead>
<tr>
<th>Parameter</th>
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</tr>
</thead>
<tbody>
<tr>
<td>diameter of the iris on the optical table.</td>
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<td>horizontal position of the LMC</td>
<td>The position in which the beam is approximately centred on the LMC window.</td>
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<tr>
<td>accept/reject data within a chopper cycle</td>
<td>Reject the first 80, accept 200, reject the next 100, and accept 200.</td>
</tr>
</tbody>
</table>

Investigations were started into adding a second detection channel to the LMR which would allow the reduction of noise in the data stream from solar radiation. The optical beam is split after passing through the chopper, with one portion directed through the LMC to the first detector and the rest directed towards the second detector. The aforementioned changes (d), (e),
and (f) were enacted based on this plan for the second channel. No results with the second detector had been obtained at the time of writing this report.

This work has yielded results which appear to confirm that the length modulated radiometer used in these studies will become an effective ground-based instrument for measuring atmospheric carbon monoxide and methane. With the success of this LMR, as well as of the MOPITT instruments, one small task in the quest to observe the atmosphere globally and continually for environmental modelling will have been completed.
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