Methane: Interhemispheric concentration gradient and atmospheric residence time

(Interhemispheric concentration gradient and atmospheric transport/Amazon basin)

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ABSTRACT The ground level concentrations of methane in the atmosphere have been measured to be in the range from 1.45 to 1.62 parts per million by volume (ppmv) of dry air in remote locations between 62°N and 54°S latitudes during the time period from November 1977 to July 1979. The average (±rms) concentration for the northern hemisphere was 1.57 ± 0.02 ppmv in January 1978 and 1.39 ± 0.02 in July 1979. The average concentration in the southern hemisphere was lower—1.47 ± 0.02 in January 1978 and 1.51 ± 0.01 in July 1979. The ratio of concentrations between the two hemispheres was 1.09 ± 0.016 in January 1978 and 1.65 ± 0.13 in July 1979, for an average of 1.06 ± 0.01. The higher concentrations in the northern hemisphere require either that the sources of methane lie preferentially in the northern hemisphere or that the removal processes operate more rapidly in the northern hemisphere or both. The primary removal process for CH4 is reaction with tropospheric OH radicals and its estimated atmospheric lifetime is 10.5 ± 1.8 yr. The observed interhemispheric gradient is consistent with this lifetime and preferential release of methane in the northern hemisphere. Measurements taken in the Amazon basin region indicate the presence of a substantial source of methane in that area.

The major hydrocarbon present in the natural atmosphere of the earth is methane, which is found in a relative concentration of ~1.6 parts per million by volume (ppmv) throughout the troposphere (1-3). Most hydrogen and most carbon in the atmosphere are found in oxidized forms, and CH4 is subject to relatively rapid oxidation to other chemical forms. The major sources for the continuous replenishment of atmospheric CH4 certainly involve gaseous evolution from biological material under anaerobic conditions—e.g., rice paddies, swamps, enteric fermentation, etc.—but are not known in any precise quantitative detail. The overall biological origin of most of the atmospheric CH4 is inferred from the near-equivalence between the 14C/12C ratio in it to that in living material that draws both 13CO2 and 14CO2 from the atmosphere (1-4). Any carbonaceous material isolated for >20,000 yr from exchange with the cosmic ray-produced atmospheric 14C reservoir will have a negligible 14C content. Hence, the contribution to atmospheric CH4 from the flaring of natural gas, or by volcanoes, earthquakes, etc. (5), cannot be major because these sources should be essentially devoid of 14C. Ehhalt (3) has placed an upper limit of 10% on the nonbiogenic sources of CH4.

Methane has been known as a component of the atmosphere since 1945 (6), and several groups have reported (Table 1) higher concentrations in the northern than in the southern hemispheres (7-15, 4). A consensus developed in the late 1970s that a concentration gradient does indeed exist between the two hemispheres. Ehhalt (10) has further demonstrated that the presence of N/S hemispheric gradients in methane concentrations provides some boundary conditions on the atmospheric residence time of CH4 and upon the N/S distribution of its sources and sinks.

During 1977-1979 we carried out three series of measurements of CH4 concentrations in tropospheric samples collected in remote locations at the surface between 62°N and 54°S latitudes, and we have consistently observed a N/S gradient with ~5-7% higher concentrations in the north than in the south. The removal of CH4 from the atmosphere is usually assumed to occur almost entirely by reaction 1 with OH radicals,

$$\text{OH} + \text{CH}_4 \rightarrow \text{H}_2\text{O} + \text{CH}_3$$

with most of this attack occurring in the lowest 6 km of the atmosphere. Quantitative estimates of the overall rate of this reaction are limited by the current imprecision in the knowledge of worldwide concentrations of OH in both time and space. In the absence of better information concerning sources and sinks, the atmospheric residence time of CH4 has not yet been well established and further data can be very useful. Previous estimates of the atmospheric residence time for CH4 have ranged from 1.5 (16) or 2 (17) yr to much longer times of 21.5 (12) or 29 (18) yr. Ehhalt (10) has recently estimated a most probable value of about 5 yr with a range from 2.5 to 11 yr.

The variations in CH4 concentrations in our samples correlate well with the variations in CH3CCl3 concentrations measured in the same canisters, for which a considerably larger N/S gradient was observed (19). Because the major removal process for CH3CCl3 also involves reaction with OH radicals in the troposphere, as in Eq. 2,

$$\text{OH} + \text{CH}_3\text{CCl}_3 \rightarrow \text{H}_2\text{O} + \text{C}_2\text{H}_2\text{Cl}_3$$

measurement and evaluation of CH4 and CH3CCl3 concentrations from the same air parcels can provide still further information about the sources for CH4 and the geographical location of the sinks for both compounds. The CH3CCl3 is released to the atmosphere anthropogenically, with about 95% going first into the northern hemisphere, most of it in the temperate zone. The annual release rates for CH3CCl3 have risen sharply during the past decade.

The observation of a N/S hemispheric gradient in the concentration of CH4 requires that atmospheric transport from the Southern to the Northern Hemisphere be occurring on a time scale only slightly longer than the residence time. Thus, the transport process must be a slow one in comparison to the lifetime of CH4 in the atmosphere.

Abbreviations: ppmv, concentration in parts per 10^6 by volume; ITCZ, Intertropical Convergence Zone, meteorologically separating northern and southern hemispheres; N/S, north/south; NBS, National Bureau of Standards.

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centration of a trace gas requires either (i) the source of the gas to be predominantly in one hemisphere or (ii) the removal rate to be more rapid in one hemisphere than the other. The former is certainly the case for many anthropogenic gases that are released largely in the northern hemisphere, including CO₂ from fossil fuel combustion (20), ⁸⁵Kr from nuclear fission (21), and CCl₃F, CCl₂F₂, and other industrially significant chlorine-containing molecules (22). There have also been suggestions that the average tropospheric OH concentration might be higher in the southern hemisphere than in the northern (23-27), such that removal of CH₄ and CH₃CCl₃ by reaction with OH would therefore occur more rapidly in the south than in the north. Because most of the known biological sources of CH₄ are essentially land-based, the primary release of atmospheric CH₄ into the northern hemisphere has been strongly indicated (10). Although biological in character and of long-standing historically, the release of CH₄ from cultivated rice fields and from much of the enteric fermentation can also be assigned as essentially of anthropogenic origin. Both the world land area devoted to the cultivation of rice and the number of domesticated large mammals have increased during the past two decades (28, 29).

Experimental Procedures

We have collected ground-level air samples from rural locations (identified by nearby communities in Tables 2 and 3) between 62°N and 54°S latitudes, usually within a period of several weeks for N/S comparisons. The samples were collected in 2-liter stainless steel canisters each equipped with a single Nupro stainless steel valve. The canisters were evacuated in the laboratory, transported to and opened in the remote environments, and then returned to the laboratory for analysis by gas chromatography. Aliquots were first measured for CH₄ and CCl₃F concentrations by electron-capture gas chromatography. Further aliquots were then used for CH₄ analysis with flame ionization as the detection technique, and finally, still others were used for CCl₂F₂ measurements by electron capture detection.

Two different procedures were used in the analysis of the November 1977 to February 1978 samples. In the first procedure, aliquots were taken directly from the stainless steel sample canisters at room temperature and were analyzed by using helium as the carrier gas in the flame-ionization gas chromatography. The CH₄ concentrations so measured were then corrected to ppmv in dry air from the absolute humidities measured at the time of collection. These absolute humidities varied from 3 torr (1 torr = 1.333 × 10⁵ pascal) (Idaho) to 21 torr (St. Croix), a variation of almost 3% in the contribution of H₂O to the total measured pressure. These data were reported by Mayer et al. The second procedure, used for all of the data reported here, involves freezing out less volatile components (including H₂O) at -20°C, with N₂ as the carrier gas for a more stable baseline and greater sensitivity in the flame-ionization measurements. The CH₄ concentrations listed in the tables are the average of alternate data taken usually from five aliquots of the sample and five aliquots from a secondary standard.

The CH₄ concentrations for these samples are summarized in Tables 2 and 3, expressed as ppmv of dry air, and are grouped generally by the dates of collection. The precision of the measurements of the CH₄ content of each individual sample canister is estimated to be ±0.01 ppmv. All of the sample measurements in each of the groups of Table 2 and those of Table 3 were made in the laboratory during time periods of about 1 month to permit more accurate between-sample comparisons, and are estimated to have a precision within each group of ±0.01 relative to one another. Repetitive measurements of the same set of samples 1-3 months later showed no changes in either the absolute or relative values beyond normal statistical variability, indicating that the air samples preserve the integrity of the CH₄ concentrations over long periods of storage. The Ivanpah sam-

<table>
<thead>
<tr>
<th>Time period</th>
<th>Ref.</th>
<th>N/S ratio</th>
<th>Concentration</th>
<th>Latitude</th>
<th>Concentration</th>
<th>Latitude</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>CH₄ concentration in atm cm⁻¹</td>
<td></td>
<td>CH₄ concentration in ppmv</td>
<td></td>
</tr>
<tr>
<td>Mar.-Apr. 1975</td>
<td>7</td>
<td>1.2</td>
<td>1.46-1.58</td>
<td>13°N-57°N</td>
<td>1.22-1.40</td>
<td>23°S-67°S</td>
</tr>
<tr>
<td>Nov.-Dec. 1972</td>
<td>8, 9</td>
<td>1.06</td>
<td>1.44 ± 0.04</td>
<td>7°N-33°N</td>
<td>1.36 ± 0.04</td>
<td>5°N-75°S</td>
</tr>
<tr>
<td>Nov.-Dec. 1972</td>
<td>10</td>
<td>1.40</td>
<td>± 0.02</td>
<td>0°-30°N</td>
<td>1.30 ± 0.01</td>
<td>5°N-75°S</td>
</tr>
<tr>
<td>Nov.-Dec. 1972</td>
<td>11*</td>
<td>1.08</td>
<td>1.68</td>
<td></td>
<td>1.56</td>
<td></td>
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<tr>
<td>Sep.-Dec. 1977</td>
<td>12, 13</td>
<td>1.03</td>
<td>1.43 ± 0.07</td>
<td>0°-65°N</td>
<td>1.39 ± 0.05</td>
<td>0°-90°S</td>
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<tr>
<td>Jan. 1978</td>
<td>†</td>
<td>1.07</td>
<td>1.58</td>
<td>10°N-56°N</td>
<td>1.47</td>
<td>20°S-54°S</td>
</tr>
<tr>
<td>Apr.-May 1978</td>
<td>14</td>
<td>1.06</td>
<td>1.72</td>
<td>N of 20°N</td>
<td>1.62</td>
<td>S of ITCCZ</td>
</tr>
<tr>
<td>May 1979</td>
<td>15</td>
<td>1.06</td>
<td>1.58</td>
<td>45°N</td>
<td>1.51</td>
<td>43°S</td>
</tr>
<tr>
<td>Jul. 1978</td>
<td>This work</td>
<td>1.068 ± 0.016</td>
<td>1.565 ± 0.017</td>
<td>10°N-56°N</td>
<td>1.466 ± 0.015</td>
<td>20°S-54°S</td>
</tr>
<tr>
<td>July 1979</td>
<td>This work</td>
<td>1.055 ± 0.013</td>
<td>1.593 ± 0.020</td>
<td>10°N-62°N</td>
<td>1.509 ± 0.005</td>
<td>8°S-54°S</td>
</tr>
</tbody>
</table>

N/S, north/south; ITCCZ, Intertropical Convergence Zone.
* Data of ref. 10 corrected upward by 1.2 from calibration change.
† Footnote 4.
The period freeze-out method, both the cause correlation induced most the CH$_4$ carbons intermediate hemispheric" the 2, opportunity procedure utilizes 0° operated over... (unpublished that from... 1.491 ± 0.009 in August–September 1978. We omit the Amazon basin measurements in Table 3 and calculate a southern hemispheric concentration of 1.509 ± 0.005 for July 1979 over the latitude range from 8°S to 54°S. The difference in CH$_4$ concentrations between January 1978 and July 1979 appears to be significant (30). Our northern hemispheric average for January 1978 is 1.565 ± 0.017 and for July 1979 is 1.593 ± 0.020. The corresponding ratios of N/S concentrations are 1.068 ± 0.016 in January 1978 and 1.055 ± 0.013 in July 1979 for an average value of 1.06 ± 0.01. Our average value for the interhemispheric gradient is in good agreement with other recent measurements insofar as the ratio of concentrations is concerned.

### ABSOLUTE CONCENTRATIONS

Our absolute concentrations were determined by direct comparison with secondary standards prepared from a primary standard furnished by Matheson and stated to have an absolute concentration of 48.9 ppbv with an absolute accuracy of ± 1%, as determined by comparison with a National Bureau of Standards (NBS) standard. Cross-comparison showed the absolute concentration in one such secondary standard as 1.499 ppbv on the basis of vacuum-line dilution from the Matheson standard and 1.499 on the basis of direct comparison with a 0.97 ppbv NBS standard. The close agreement suggests that laboratory preparation of secondary standards can be done with a precision equal to or better than the accuracy of the absolute concentration measurements. The response of the flame ionization detector is linear with concentration over the range between two separate NBS standards at 0.97 and 4 ppbv within the accuracy of the measurements (±0.5%) and should certainly be linear over the observed 1.45- to 1.62-ppmv range of CH$_4$ concentrations found in these samples of background tropospheric air.

Our data in Tables 2 and 3 are given in ppmv of dry air, whereas some of the other measurements of Table 1 appear to be expressed in terms of air with natural humidity, judging from the experimental descriptions. Because some of these measurements are made at surface level in the tropics and others are from aircraft flying at altitudes with ambient temperatures below 0°C, the natural humidity undoubtedly varies widely. In our own data, the 21-torr water vapor concentration in the St. Croix sample corresponds to a dry air concentration of CH$_4$.

### Table 3. Ground-level tropospheric concentrations during 1979

<table>
<thead>
<tr>
<th>Latitude</th>
<th>Sampling location</th>
<th>Date (ppmv)</th>
</tr>
</thead>
<tbody>
<tr>
<td>61.8°N</td>
<td>Chickaloon, AK</td>
<td>6/18/79</td>
</tr>
<tr>
<td>61.7°N</td>
<td>Willow, AK</td>
<td>6/18/79</td>
</tr>
<tr>
<td>55.5°N</td>
<td>Ketchikan, AK</td>
<td>6/19/79</td>
</tr>
<tr>
<td>55.5°N</td>
<td>Ketchikan, AK</td>
<td>6/19/79</td>
</tr>
<tr>
<td>47.2°N</td>
<td>Copalis Beach, WA</td>
<td>6/20/79</td>
</tr>
<tr>
<td>44.2°N</td>
<td>Roosevelt Beach, OR</td>
<td>6/20/79</td>
</tr>
<tr>
<td>40.4°N</td>
<td>Cape Mendocino, CA</td>
<td>6/21/79</td>
</tr>
<tr>
<td>25.0°N</td>
<td>Islamorada, FL</td>
<td>7/14/79</td>
</tr>
<tr>
<td>22.8°N</td>
<td>Long Island, Bahamas</td>
<td>7/16/79</td>
</tr>
<tr>
<td>20.7°S</td>
<td>Lusubaboe, HI</td>
<td>4/7/79</td>
</tr>
<tr>
<td>17.8°N</td>
<td>St. Croix, VI</td>
<td>7/17/79</td>
</tr>
<tr>
<td>13.3°N</td>
<td>North Point, Barbados</td>
<td>7/18/79</td>
</tr>
<tr>
<td>10.5°N</td>
<td>Biche, Trinidad</td>
<td>7/21/79</td>
</tr>
<tr>
<td>10.3°N</td>
<td>Mayaro, Trinidad</td>
<td>7/21/79</td>
</tr>
<tr>
<td>5.4°N</td>
<td>Paukaka, Surinam</td>
<td>7/20/79</td>
</tr>
<tr>
<td>1.2°S</td>
<td>Belen, Brazil</td>
<td>6/9/79</td>
</tr>
<tr>
<td>3.1°S</td>
<td>Manaus, Brazil</td>
<td>6/10/79</td>
</tr>
<tr>
<td>3.1°S</td>
<td>Manaus, Brazil</td>
<td>6/10/79</td>
</tr>
<tr>
<td>3.6°S</td>
<td>Fortaleza, Brazil</td>
<td>6/8/79</td>
</tr>
<tr>
<td>8.1°S</td>
<td>Recife, Brazil</td>
<td>7/7/79</td>
</tr>
<tr>
<td>13.0°S</td>
<td>Salvador, Brazil</td>
<td>6/7/79</td>
</tr>
<tr>
<td>19.2°S</td>
<td>Vitoria, Brazil</td>
<td>6/5/79</td>
</tr>
<tr>
<td>23.5°S</td>
<td>Antofagasta, Chile</td>
<td>6/2/79</td>
</tr>
<tr>
<td>41.4°S</td>
<td>Puerto Montt, Chile</td>
<td>5/29/79</td>
</tr>
<tr>
<td>53.1°S</td>
<td>Punta Arenas, Chile</td>
<td>5/31/79</td>
</tr>
</tbody>
</table>

These five samples to the presence of a substantial Amazon basin source of CH$_4$ from the anaerobic decay of biological material. Approximate estimates from these very limited data suggest that the Amazon basin could be the source for as much as 10% of the atmospheric methane.

![Fig. 1. Correlation of measured concentrations of CH$_4$ and CH$_3$CCl$_3$ in tropospheric samples in mid-1979. ppbv, concentration in parts per 10$^{12}$ by volume.](image-url)
demonstrates that validity provides from 1.51 to 1.56 of 6.9 constants for gases with decrease the temperature.

Although the concordance has been removed by the discovery of an error of 1.2 in a calibration standard used in the National Center for Atmospheric Research measurements before 1974 (11). When this correction factor is included, one set of these 1972 measurements then lies above our present values and one lies below it. We are unable to explain the discrepancy among these 1972 values and therefore unable to draw any conclusions about possible longer term trends in CH4 concentrations except those occurring internally within our own data set. It seems probable from the spectroscopic records back to 1948 (6) that no large variation (±20%) has occurred over the past 30 yr.

**ATMOSPHERIC LIFETIME OF CH4**

If reactions 1 and 2 are the predominant mechanisms for atmospheric removal of CH4 and CH3CCL3, respectively, then the removal rates are proportional to the reaction rate constants, k1 and k2, at each location in the atmosphere, and the relatively uncertain OH concentrations in the atmosphere can be essentially canceled out in a relative calculation. The best current rate constant equations for these reactions are (31) k1 = 2.4 × 10^{-12} exp(-1710/T) and k2 = 5.4 × 10^{-12} exp(-1820/T) cm^3 mol^{-1} s^{-1}. Although the available temperature range in the troposphere covers about 90°C, ≈80% of the removal of these molecules occurs below an altitude of 6 km, at temperatures above 275 K, with the median altitude for such reaction at about 2 km (32). At the higher altitudes the lower temperatures reduce the rate constants for reactions 1 and 2 so that they are 1/8th as fast at 217 K as at 298 K. The lower absolute concentrations of well-mixed gases (factor of 4 between altitudes of 0 and 12 km) further decrease the actual reaction rates until the removal rate at 12 km for a given OH concentration is about 1/30th of that for the same concentration at the surface. The activation energies for reactions 1 and 2 are very similar, the ratio k2/k1 varies only from 1.51 to 1.56 between 275–300 K, and the value at 250 K, 1.52, is an appropriate average value for the whole tropospheric reaction competition. By using the atmospheric lifetime of 6.9 ± 1.2 yr calculated for CH3CCL3 from its measured atmospheric loading in 1978 (19), the estimated lifetime for CH4 is 1.52 times longer, or 10.5 ± 1.8 yr. The CH3CCL3 lifetime estimate is based upon removal by all processes—not just reaction with tropospheric OH—but is certainly dominated by the latter.

The existence of a N/S gradient in concentrations of CH4 also provides some limitations on its atmospheric lifetime, through examination of the location and magnitude of its sources and sinks (10). Considerable insight into these relationships can be gained through a two-box N/S model of the atmosphere, as shown by Ehnhalt (10). The observation of essentially only two atmospheric concentrations (one north of 10°N and one south of 0°) for CCL3F and CCL2F2 in July 1979 (unpublished data) demonstrates that the two-box model approach has substantial validity in the real atmosphere for discussions of concentration gradients for trace gases.

Ehnhalt has developed a formula relating in steady state the average atmospheric lifetime, τa, through Eq. 3,

\[ \tau_a = \tau_m \frac{(\alpha - R)}{(\alpha + 1)(R - 1)} \]  

[3]

to the average interhemispheric mixing time, τm, the concentrations in the north and south (Mn, Ms), and the ratio, α, of the source strengths in the northern and southern hemispheres (10). He then adopted τm as 1 yr and calculated τa from his 1972 data (Table 1) in which Mn = 1.40 ppmv and Ms = 1.30 ppmv for a difference of 0.10 ppmv. An equivalent formula to Eq. 3 is given in Eq. 4, in which the interhemispheric concentration ratio,

\[ R = \frac{M_n}{M_s} \]  

[4]

has been used instead of the individual concentrations. Because most biogenic CH4 is land-based, production must be predominantly in the northern hemisphere. Ehnhalt postulated that CH4 production might be approximately proportional to the land area in each hemisphere (α = 2.06) or more likely to that below 60° in each hemisphere (α = 2.4), and he calculated the most probable lifetime for CH4 to be 5 yr. Values of 2.5 and 11 yr were calculated for interhemispheric gradients of 0.15 and 0.05 ppmv, respectively, on the old calibration scale. The change in the National Center for Atmospheric Research absolute calibration (11) changes the absolute values of Mn and Ms, but not those of R or τa.

The formulation of Eqs. 3 and 4 implicitly adopts the additional assumption that the rate of removal of CH4 is the same in both the northern and southern hemispheres—i.e., that there is no hemispheric asymmetry in the average OH radical concentrations. Several atmospheric model calculations have been performed which lead to questioning of the hemispheric equivalence in OH concentrations (23-27, 32), with some hypothesizing as symmetries as large as a factor of 3. The observed CH3CCL3 gradient in concentrations has led to suggestions that the OH concentrations are twice as large in the southern as in the northern hemisphere (26). Other analyses place the ratio between 1.0 and 1.3 (27).

We have expanded the Ehnhalt two-box model to include the additional possibility of hemispheric asymmetry in the removal of CH4 and CH4CCL3 by reaction with OH, and we have defined the interhemispheric mixing rate as k = (τm) and the two removal rates as Λn and Λs, with the ratio Λ = Λn/Λs. With different removal rates in the two hemispheres the average lifetime, τa, is no longer rigorously defined because the lifetime is now dependent upon the hemisphere in which the trace gas is released. However, with lifetimes of 5 yr or longer and interhemispheric mixing times of about 1 yr, the average atmospheric lifetimes are at most a few percent different for the same molecule introduced into the northern versus the southern hemispheres. We therefore define an average loss rate in steady state, Λa, by Eq. 5,

\[ \Lambda_a = \frac{(\Lambda_n R + \Lambda_s)}{R + 1} \]  

[5]

and an average lifetime, τa = 1/Λa, and assume its approximate applicability to any source distribution of gases introduced into the model. The differential equations for variation in the concentrations (Mn, Ms) are then given by Eqs. 6 and 7,

\[ \frac{d(M_n)}{dt} = P_n - k(M_n - M_s) = \Lambda_a(M_n) \]  

[6]

and

\[ \frac{d(M_s)}{dt} = P_s + k(M_n - M_s) = \Lambda_a(M_s) \]  

[7]

in which Pn and Ps are the source functions into the northern and southern hemispheres, respectively, and α = Pn/Ps. At steady state, both Eqs. 6 and 7 are equal to zero and R is given by Eq. 8.
\[ R = \alpha \left( \lambda_k + k \right) + k \]
\[ \left( \lambda_k + k \right) + k \]

Equation [8] represents the relationship between the rate of change of concentration and the parameters involved in the atmospheric budget of methane. The term \( \alpha \) represents the emission rate, \( \lambda_k \) is the decay rate, and \( k \) is the rate of change due to other processes. This equation is used to model the distribution of methane in the atmosphere, taking into account both anthropogenic and natural sources.

The source strengths for \( \text{CH}_4 \) are certainly now of prime importance in establishing more quantitatively the details of the atmospheric methane cycle.

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