Validation of Global Climatologies of Trace Gases Using NASA Global Tropospheric Experiment (GTE) Data

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Methane is an important trace gas because it is a greenhouse gas that affects the oxidative capacity of the atmosphere. It is produced from biological and anthropogenic sources, and is increasing globally at a rate of approximately 0.6% per year [Climate Change 1992, IPCC]. By using National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostics Laboratory (NOAA/CMDL) ground station data, a global climatology of methane values was produced. Unfortunately, because the NOAA/CMDL ground stations are so sparse, the global climatology is low resolution. In order to compensate for this low resolution data, it was compared to in-situ flight data obtained from the NASA Global Tropospheric Experiment (GTE). The smoothed ground station data correlated well with the flight data. Thus, for the first time it is shown that the smoothing process used to make global contours of methane using the ground stations is a plausible way to approximate global atmospheric concentrations of the gas. These verified climatologies can be used for testing large-scale models of chemical production, destruction, and transport. This project develops the groundwork for further research in building global climatologies from sparse ground station data and studying the transport and distribution of trace gases.
INTRODUCTION

The concentration of methane (CH₄) in the atmosphere, an important greenhouse gas, is currently increasing at the rate of approximately 0.6% per year [Climate Change 1992, IPCC]. Table 1 shows the global sources of methane. Between 356 and 875 teragrams (Tg, 10¹² grams) of methane are produced globally each year. Of this amount, as shown by the table, between 240 and 575 Tg are produced from anthropogenic sources. This amount constitutes 2/3 of all methane emissions globally. Because the concentration of methane is steadily increasing, and anthropogenic sources are such a large fraction of total production, it is important to understand man-kind's impact on the atmosphere. In order to aid in this understanding, it is shown that it is possible to determine global methane concentrations using sparse ground station data.

The basis of the project lies in methane measurements obtained from two sources. The first data source was the NASA Global Tropospheric Chemistry Program (GTCP), which was developed in recognition of the central role of tropospheric chemistry in global change. Its goal is to promote an understanding of the troposphere and to assess the susceptibility of the global atmosphere to chemical change. One component of the GTCP is the Global Tropospheric Experiment (GTE). Focusing on the study of the global troposphere, GTE consists of a series of airborne field experiments designed to (1) evaluate the capability of instrument techniques to measure concentrations of key chemical species in the atmosphere, and (2) systematically address tropospheric chemistry issues relevant to global change. The GTE project has encompassed four major airborne experiments: (1) the Chemical Instrumentation Test and Evaluation (CITE) experiments were conducted to evaluate the measurement techniques of some atmospheric gases; (2) the Atmospheric Boundary Layer Experiment (ABLE) explored the atmospheric boundary layer above major ecosystems that are known to influence global chemistry; (3) the Pacific Exploratory Mission-West (PEM-West) examined the impact of natural and anthropogenic emissions over the Pacific Ocean; and (4) the Transport and Chemistry near the Equator in the Atlantic (TRACE-A) investigated the distribution of trace gases over the tropical south Atlantic.

The second data source was the National Oceanic and Atmospheric Administration/Climate Monitoring and Diagnostics Laboratory (NOAA/CMDL). In April of 1983, NOAA/CMDL expanded its air flask sample analysis to include the collection and study of methane in the atmosphere. Globally, there are 27 permanent ground sites dedicated to this study. The Global Cooperative Air Sampling Network, which includes 44 total sites, collects samples approximately once per week (see Figure 1). All of these samples were analyzed for methane and referenced to a standard scale at the NOAA/CMDL laboratory in Boulder, Colorado. This data is now available via electronic archive.

The LARSS project involved data reduction and analysis of data from NOAA/CMDL and NASA/GTE. From a public database, NASA/GTE and NOAA/CMDL data were obtained. Using information about the ground stations and the measurements taken there, global contour plots of methane levels were constructed. The findings show that this low-resolution data can be smoothed and produce an accurate representation of global methane levels. The basis of the project lies in the verification of this contouring method. In order to verify the routine, NASA/GTE in-situ flight data was used, and its findings were compared to those generated by the contour plots. It is shown that good climatologies can be made using NOAA/CMDL ground station data, and that the high-accuracy and resolution in-situ aircraft data from the NASA/GTE missions can be used to validate the smoothing and contouring routine.
DESCRIPTION OF DATA

The average values for monthly methane levels were obtained from the NOAA/CMDL data archive for the years 1991-1993. A 72 by 36 array was used to represent a 5° by 5° global grid. Because the data was limited, many missing points needed to be replaced. To calculate these missing points, the program Spyglass Transform was used. This routine replaced the missing methane values with values interpolated from those that did exist. The weighted fill method was used to determine these missing data points. It replaced the missing data with a weighted average of the real data within a certain range (8). A spherical distribution model was used, in which the weight dropped off as the reciprocal of the distance squared (1/d^2) from the missing data value. The weighted fill was run twice more on the new arrays created, both with a range of 4. The defined data points were preserved so they would not be lost during this process. The final array created a global climatological map of methane with no missing data points. Some sample plots are show as Figures 2-14, in which the low values of methane are indicated by blues, intermediate values are green to orange, and high values are red.

The data files for the various GTE missions were obtained by electronic means from the Distributed Active Archive Center (DAAC) at the NASA Langley Research Center. For each flight, methane levels were measured and recorded every five seconds. This high resolution data were then time correlated with flight navigational data, and the CH\textsubscript{4} measurements were separated into 5° by 5° blocks and the number of CH\textsubscript{4} samples taken in each block, their average, and their standard deviation was then calculated.

The key step in this process was to verify the validity of the smoothed contour plots. To do this, a correlation between the NOAA/CMDL ground station data and the in-situ GTE flight data needed to be found. Since there was no set program to make these comparisons, necessary IDL routines were developed on a DECstation 5000/200. The code produces graphs of each global area studied, comparing the flight data to the ground station measurements. This routine helped in making analyses for the objective.

DISCUSSION

Some measurements taken during some of the flights exhibited unexpected variations. During Flight 4 of the PEM-West A mission, which went from the Ames Research Center in California to Anchorage, Alaska, the methane measurements showed a sharp decrease at approximately 2100 GMT (see Figure 15). But at this same time the plane increased its altitude to over 35,000 ft., and a decrease was observed in some other trace gas species being measured. A significant increase in ozone concentration also occurred at this same time. This simultaneous increase in ozone, and a decrease in other trace gases indicates that the flight entered the tropopause and possibly the stratosphere. In this example, further evidence was provided by a temperature profile taken as the plane descended from an altitude of over 35,000 ft. to approximately 1500 ft. This profile showed a decrease in temperature with decreasing altitude from 35,000 ft to about 30,000 ft., which is characteristic of the stratosphere. Since data taken while the flight was not in the troposphere would not be representative of tropospheric methane levels, this data was excluded from the analysis and comparison with the globally contoured methane levels.
RESULTS

In general, the contours produced using the NOAA/CMDL ground station data compared favorably with the flight data from GTE. Figures 16-18 show some of these comparisons. The contour lines usually showed a 10-30 ppb higher methane concentration than the levels determined from the flight data. The contoured methane levels show the same trends seen in the flight measurements. In Figure 18, from PEM-West A Flight 4, it can be seen that methane levels increased by approximately 72 ppb during the flight and while the contour data show a 47 ppb change.

Figure 19 shows a plot of latitude versus the difference between the predicted and measured methane levels. It can be seen that the difference between the predicted contour and the measured flight data varies with latitude. At about 10°-15° N the average difference is approximately zero, while above and below this latitude the predicted levels are higher than the measured concentrations. In PEM-West A Flight 4, for example, the difference between the two methane values started out at approximately 35 ppb at the beginning of the flight and decreased to approximately 10 ppb at the end of the flight. A compilation of all the flight measurements taken showed that the average difference between measured and predicted values at the 40°-50° N latitude (the flight's starting point) was 23-33 ppb and that the average difference between the values at the 55°-65° N was 8-20 ppb.

One point to consider is the global variation of methane versus errors in calculation. On average, based on the actual values determined, there was a bias of 10%-20%. When looking a global variations of methane (see Figures 2-13), it is noted that there is a consistent concentration of 1650-1800 ppb. Therefore globally, there is an error of magnitude of approximately 20%. The average difference between the flight data and ground station data then becomes significant when considering global variations. For example, from season to season and from pole to pole, one can expect to see a maximum variation of approximately 200 ppb. Thus, if an error of over 20 ppb were found, one can deem the calculation inaccurate. If a constant bias of approximately 10-20 ppb were removed from all of the comparisons, this method of producing global climatologies and their verification using in-situ flight data becomes more accurate.

CONCLUSIONS

Several issues remain to be examined from the project. It is noted in Figure 4 that the difference between the contours and the measured flight data varied with latitude. The graph shows a "double-hump" distribution of values, with a minimum value at 10°-20° N. The cause of this "double-hump" can be explained by examining both Figures 1 and 19. At 10°-20° N, there are virtually no ground stations. Therefore, one can predict that measurements in that area obtained from the smoothing process will be somewhat inaccurate. In order to compensate for this, it would be beneficial to place further ground stations in Africa, South America, and northern and central Australia. This would dramatically improve the resolution quality of contouring using ground station data.

The contouring routine treated the globe as a cylinder, and not a flat plane. As shown below, this was necessary to do this in order to connect data points that are at opposite ends of a map that are actually beside each other.
However, with limited software, it was difficult to treat the globe as a sphere. Because of this, the predicted values in the high latitudes may be suspect. Therefore, it may be beneficial to construct a spherical contouring routine to examine what changes became evident using this new method. It must also be mentioned that only the northern and central pacific regions, the United States, and the equatorial Atlantic regions during the months of September and October were examined. Greater validity could be lent to the contouring method if other areas and times of the year were be examined.

The capacity to validate global climatologies is significant for chain studies. Since methane has a long lifetime in the atmosphere, and is therefore well-mixed, it is not practical to use global climatologies to determine the sources of the gas. Instead, these climatologies can be used to examine seasonal trends of methane, as seen in Figures 2-14. The work can be further expanded to compare these seasonal cycles to estimated sources from biomass burning. The methods used for this project can also be used to contour other trace gases, such as carbon dioxide (CO$_2$) and nitrous oxide (N$_2$O). In conclusion, the project shows that using aircraft data, especially if globally distributed, is a good method to validate global climatologies produced from sparse ground station data. It lays the groundwork for further research in building global climatologies from sparse ground station data and studying the transport and distribution of trace gases.
REFERENCES

### Estimated Sources and Sinks of Methane (Tg methane per year)

<table>
<thead>
<tr>
<th>Sources</th>
<th>Amount (Tg methane per year)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Natural</strong></td>
<td></td>
</tr>
<tr>
<td>Wetlands</td>
<td>115 (100-200)</td>
</tr>
<tr>
<td>Termites †</td>
<td>20 (10-50)</td>
</tr>
<tr>
<td>Ocean</td>
<td>10 (5-20)</td>
</tr>
<tr>
<td>Freshwater</td>
<td>5 (1-25)</td>
</tr>
<tr>
<td>CH₄ Hydrate</td>
<td>5 (0-5)</td>
</tr>
<tr>
<td>Coal Mining, Natural Gas &amp; Pet. Industry †</td>
<td>100 (70-120)</td>
</tr>
<tr>
<td>Rice Paddies †</td>
<td>60 (20-150)</td>
</tr>
<tr>
<td>Enteric Fermentation</td>
<td>80 (65-100)</td>
</tr>
<tr>
<td>Animal Wastes †</td>
<td>25 (20-30)</td>
</tr>
<tr>
<td>Domestic Sewage</td>
<td>25 (?</td>
</tr>
<tr>
<td>Treatment †</td>
<td>30 (20-70)</td>
</tr>
<tr>
<td>Landfills †</td>
<td>40 (20-80)</td>
</tr>
</tbody>
</table>

| Anthropogenic            |                              |
| Atmospheric (tropospheric + stratospheric) removal † | 470 (420-520) |
| Removal by soils         | 30 (15-45)                   |
| Atmospheric Increase     | 32 (23-37)                   |

† indicates revised estimates since IPCC 1990

Ground Stations Available for Methane
1991-1993

Figure 1
Figure 2
Figure 3
Figure 14
TRACE A Flight 19

Figure 15

Predicted Methane Levels from Ground Station Data

Measured Methane Values from Flight Data
PEM-West A Flight 5

Figure 16
Predicted Methane Levels from Ground Station Data

Measured Methane Values from Flight Data

Figure 17
Difference Between the Predicted and In-Situ Methane Values (ppb)

Figure 18