B－5．2 Modeling of Material Transport in the Troposphere

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Abstract

A global chemical transport model is used to evaluate the emission and extinction of green house gases in the atmosphere. Tropospheric OH amount is revised using annual trend of methyl chloroform. Emission estimate for Methane is evaluated using latitude gradient of surface concentrations. Seasonal variations of CO2 exchange of biosphere is examined using forward calculations and inversion methods.

Tropospheric OH estimated by Spivakofsky is to be reduced 36 % in NIRE-CTM. The source estimate of Methane by Taylor has larger emission rate in the Southern Hemisphere. CO2 exchange of biosphere by Fung starts uptake in growing season later than the correct time.

Key Words Chemical transport model, OH radical, methane, carbon dioxide

1. Introduction

Chemical composition of atmosphere is changing with solar-terrestrial variations and human activities. Concentrations of greenhouse gases remote from the source region are important for understanding their radiative effects on the climate as well as the equilibrium state of chemical compositions, both at the present day and in the future. Correct representations of emission, transport, and extinction are crucial to obtain realistic distributions of those gases.

National Institute for Resources and Environment developed a chemical transport model, NIRE-CTM-93. Prominent feature of this model is the use of analyzed meteorological data. Using analyzed data, the model can simulate time variations of compositions which can be comparable with observations. The model is tested by long-lived species with constant emission scenarios, such as CFC-11. It is not examined with a compositions with chemical reactions in the atmosphere, nor emission with seasonal variations so far.

2. Research Objectives

The objective of this study is to evaluate the preset day knowledge for emission and chemical reactions using NIRE-CTM-93. The model has 2.5 horizontal resolution and 15 vertical layers with 6 hours time intervals. The wind data to drive this model are the analysis of European Center for Medium Range Weather Forecasts. Transport method is semi-Lagrangian with linear interpolation.

OH radical is the most active agent to oxidize methyl chloroform, methane, carbon monoxide etc. Direct measurement of the amount of OH radical has developed only recently and is not available so far. Alternatively, the amount in the equilibrium state has been estimated using a chemical transport model with a reactions. The first objectives is to quantify the current estimate of OH radical amount using NIRE-CTM-93. In general, because chemical reaction rate is sensitive to the ambient temperature, equilibrium state is the functions of atmospheric temperature. Equilibrium OH radical amount in the NIRE-CTM-93 may be different from that in GISS (Goddard Institute for Space Studies) model in which Spoivakofsky estimated the OH radical amount.

Methane in the atmosphere is emitted from wetland, landfills, and termite, etc. The emission estimates with the seasonal variations are provided from Taylor at Australian National University. The source contains anthropogenic emissions, from fossil fuel combustion landfill, rice paddies and natural emissions form termites and wetland. Each component of the emissions has large uncertain and refinement of the emission estimate using NIRE-CTM-93 is an interesting excirsize. This is the second objectives. The third objectives is to evaluate the emission and absorption of carbon dioxide. Carbon dioxide is emitted from fossil fuel combustion and has been increasing since industrial revolution. Each year, about 7 giga tons in terms of carbon are emitted from fossil fuel combustion and the desctraction of tropical rain forest. Only 3 giga tons of emitted CO2 is remained in the atmosphere. The fate of remaining CO2 is not well understood. Concentrations in the atmosphere exhibit a significant seasonal variations probably due to the seasonal variations of emission and absorption of terrestrial biosphere. An estimate of exchange between atmosphere and biosphere is made by I.Fung at Virginia University, Canada. An estimate of emission from fossil fuel combustion is provided from Marland at Ork Ridge National Laboratory, U.S.A.. Revision of those estimates and making estimate of emission and absorption from ocean are another interesting excircize.

3. Research Method

Three compositions are transported in the model; Methyl Chloroform (CH3CCl3), Methane (CH4), and Carbon Dioxide (CO2). One component, OH radical, is given in

monthly mean zonal mean form. CTM is used for forward calculations and an inversion technique is applied to CO2 sources.

3-1. Methyl Chloroform (1994)

Source distribution of Methyl Chloroform is provided from Hartley at Georgia Institute of Technology. Annual emission is estimated by Alternative Fluorocarbon Environmental Acceptability Study (AFEARS). Zonal mean, monthly mean OH radical amount is provided from Spivakiofsky. Reaction rate between Methyl Chroloform and OH radical is obtained from a table compiled by Jet Propulsion Laboratory.

Initial condition of integration is uniform concentrations of 110 pptv in the Northern Hemisphere and 70pptv in the Southern Hemisphere. Concentrations are integrated using respective emission of eleven years from 1978 to 1989. Annual trends and seasonal variations are compared with the observations by Atmospheric Lifetime Experiment (ALE).

3-2. Methane and carbon dioxide (1995)

Adjusted OH radical amount is used to make methane concentrations simulations using an emission estimated by Taylor et al.(1991).Initial condition is uniform concentrations of 1600 ppbv. Annual emission is constant value of 524Tg. Annual trends and seasonal variations are compared with the observations by Climate Monitoring and Diagnostics Laboratory (CMDL) of National Oceanic and Atmospheric Agency (NOAA).

Constant emission from fossil fuel combustion and seasonal cycles of emissions and absorption of CO2 from land ecosystems are given to NIRE-CTM-93. Initial condition is uniform concentrations of 350 ppmv. Annual trends and seasonal variations are compared with the observations by CMDL/NOAA and Atmospheric Environmental Service(AES) , Canada.

3-3. Inversion of CO2 (1996)

Inversion calculation of sources and sinks of CO2 consists of forward calculations of each source components and inversion calculations to make a fit to observed concentrations. There are four major components in the source; constant emissions of fossil fuel comsbustions, oxidation of CO in the atmosphere, absorption and emission s of land ecosystem, and those of ocean.

Seasonal variations of 19 ecosystems are produced by applying vegetation type map o f Mathews (1x1) and total absorption and emission estimate of Fung et al.(5x4, 1987). Emission and uptake are integrated separately. Integration for uptake are treated as

fertilization of that ecosystem. Ocean is divided into 12 parts, and each components are integrated separately. Produced concentrations signals are decomposed into five parts; latitude gradients, annual oscillations (sin and cosine), and semi-annual oscillations (sin and cosine). Measurement function is produced with the weighted sum of the difference between observed components and simulated components. Best combinations of sources are produced to make the model predictors minimum in least square sense.

4. Results

4-1. OH radical amount

In the first experiment, the increase rate of total methyl chloroform in the model atmosphere is significantly smaller than the observations. The proper amount of OH radicals to explain the growth rate is estimated and 64% of the original estimate. The modified OH radical amount is used hereafter.

Concentrations of Methyl chloroform are compared with five ALE/GAGE sites in monthly mean base. Minimum concentrations in the CTM at Ireland agree well with observations if OH radical amount is reduced by 36 %.

Maximum concentrations in the CTM at Oregon agree well with observations. At other three sites, Barbados, Samoa, Tasmania, concentrations agree well in the earlier half periods. Seasonal variations at Tasmania are reproduced in the CTM.

Methane concentrations are calculated for seven years from 1983 to 1989 and concentrations at five GMCC sites are compared with the observations. Annual growth rate is consistent with the observations if 524Tg/yr source is given to the CTM for seven years. Concentrations show significant seasonal variations. At Alert CTM shows maximum in summer and observed maximum is in winter. At other four sites, seasonal variations in CTM are consistent with the observations. Concentrations in the Southern Hemisphere is larger than the observations, suggesting the rate of emission from the Southern Hemisphere is larger than the correct distributions.

Carbon dioxide concentrations are calculated using a source from fossil fuel combustion and seasonal exchange of CO2 between atmosphere and land ecosystem. Concentrations at four CMDL sites are compared with daily observations. Seasonal variations are consistent with observations if seasonal exchange of land ecosystem is given to the CTM. At Samoa, seasonal variations of synoptic time scale fluctuations are reproduced in the both experiment, fossil fuel and vegetation, suggesting the seasonal fluctuations are caused by the fluctuations of Inter Tropical Convergence Zone (ITCZ). In the Northern Hemisphere, spring time maxima in CTM is twice as

large as the observed one. The timing of draw down of CO2 in the growing season start earlier than the observations.

Source component are divided into 19 land ecosystems, 9 oceanic regions, fossil fuel combustion, and oxidization of carbon monoxide. Each source is given to CTM and CTM produced signals at CMDL sites. They are used to reconstruct total source component to fit the observations. This process is called inversions.

The process includes to constrain the emission estimate with previous knowledge with its allowance. If the allowance is given loosely, the estimate showed a turn over of the seasonal variations.

5. Discussion

NIRE-CTM-93 showed a reasonable concentrations for three greenhouse gases, Methyl Chroloform, Methane, Carbon Dioxide with the estimated of their emission distributions and extinction procedures, while there remained some problems. Tropospheric OH in Spivakofsky's value is too high compared to the present estimates. Methane concentrations indicating the emission from the Southern hemisphere in Taylor's estimate may be too large. Biospheric uptake may be too sharp in Fung's estimates.

Reference

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