

# Atmospheric Composition Change and its Climate Effect Studied by Chemical Transport Models

Project Representative

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We have used the CCSR/NIES/FRCGC atmospheric general circulation model (AGCM) based transport model for simulations of greenhouse gases (GHGs), e.g., CO<sub>2</sub>, N<sub>2</sub>O, SF<sub>6</sub>, and a transport diagnostic tracer <sup>222</sup>Radon (half-life 3.8 days). These simulations are now being analyzed comparing with observations and preparing strategies for future inverse modeling of GHG fluxes. The JMA CDTM is used for tracer transport in inversion intercomparison.

**Keywords:** carbon dioxide, nitrous oxide, chemical transport model, planetary boundary layer, interannual variations

## 1. Diagnostics of model transport across the planetary boundary layer

Fig. 1 shows the daily average model simulations of CO<sub>2</sub>, Radon, and SF<sub>6</sub> at different time averaging of hourly data (lower three rows). Due to short atmospheric lifetime Radon is used for diagnosing model transport across the planetary boundary layer (PBL), and variations between summer/day (higher mixing height) and winter/night (stronger inversion layer). When a temperature inversion occurs near the ground, Radon builds up within the PBL because its emission is prescribed to be uniform from this land region. This information is useful for selecting the most appropriate model level when the modeled CO<sub>2</sub> are to be used along with observation for estimating surface sources/sinks of CO<sub>2</sub> by inverse modeling at lower bias. On the other hand since SF<sub>6</sub> has very heterogeneous emission distribution, following the power grid locations which are closely located to the fossil fuel consumption, and no known loss in the troposphere, its vertical profiles indicate seasonal and synoptic changes in transport patterns.

The comparison of model simulations and observations of CO<sub>2</sub> reveals several important features: The daily averages using all data (left column; top two rows) indicate seasonal variations in CO<sub>2</sub> arising from summer-time biospheric carbon uptake by the terrestrial biosphere exceeding anthropogenic emission (low values in Jun-Sep; blue shade), and winter-time respiratory release from the terrestrial biosphere and anthropogenic emissions (high values in red shading). A similar comparison using only the afternoon (13–16 LT) val-

ues depicts a vertically well mixed condition extending beyond the tower height. Thus a comparison between model and data is easier done by selecting data from any tower sampling height and lower model levels. This is important as long as the coarse vertical (and horizontal) resolution transport models are in use. For the nighttime (03–06 LT) model-data comparison it appears that modeled vertical gradient is stronger than that observed.

## 2. Seasonal variation of atmospheric N<sub>2</sub>O

Atmospheric nitrous oxide (N<sub>2</sub>O) concentration was simulated using a chemistry-coupled AGCM nudged with reanalyzed meteorological fields for the period 1983–2002. Attempts have been made to determine most realistic emission fields for reproducing observed N<sub>2</sub>O latitudinal distribution and seasonal cycle. We found that implementation of stratospheric chemistry is crucial to simulate seasonal cycle and interannual variations (IAVs) in N<sub>2</sub>O. Fig. 2 shows comparisons of simulated and observed IAVs at 9 surface sites widely located around the globe. It is clearly seen that simulations without N<sub>2</sub>O photochemistry (broken lines) produce weaker or no IAVs and those with chemistry (solid lines) are able to generate similar IAV amplitudes as well as phase. This is due to the fact that N<sub>2</sub>O gradients across the tropopause and stratosphere-troposphere exchange anomalies linked with the climate variations, such as ENSO in troposphere and QBO in stratosphere, are required for realistic simulation of IAVs.

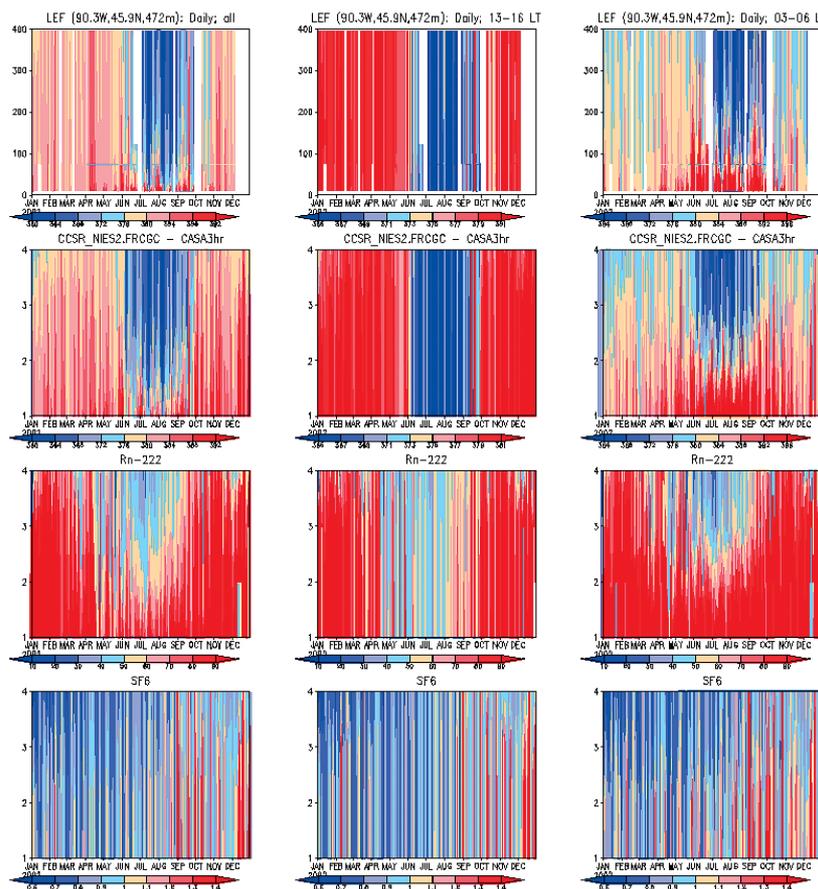


Fig. 1 Vertical profiles of  $\text{CO}_2$  from tall tower at Park Falls, Wisconsin (LEF; top row), model simulations (2<sup>nd</sup> row from top), and modeled profiles of  $^{222}\text{Rn}$  (3<sup>rd</sup> row from top) and  $\text{SF}_6$  (bottom row) are shown. Left, center, and right columns are due to different averaging schemes, all 24 hr data in a day, afternoon values only (13–16 LT), nighttime values only (03–06 LT), respectively. The y-axis for observed profiles is in meter, while the model profiles are shown as model level (surface to  $\sigma$ -layer 5; approximately 0–500 m). The AGCM, nudged with NCEP/NCAR reanalysis temperature and winds, is run at T106 horizontal resolution and 32 vertical layers.

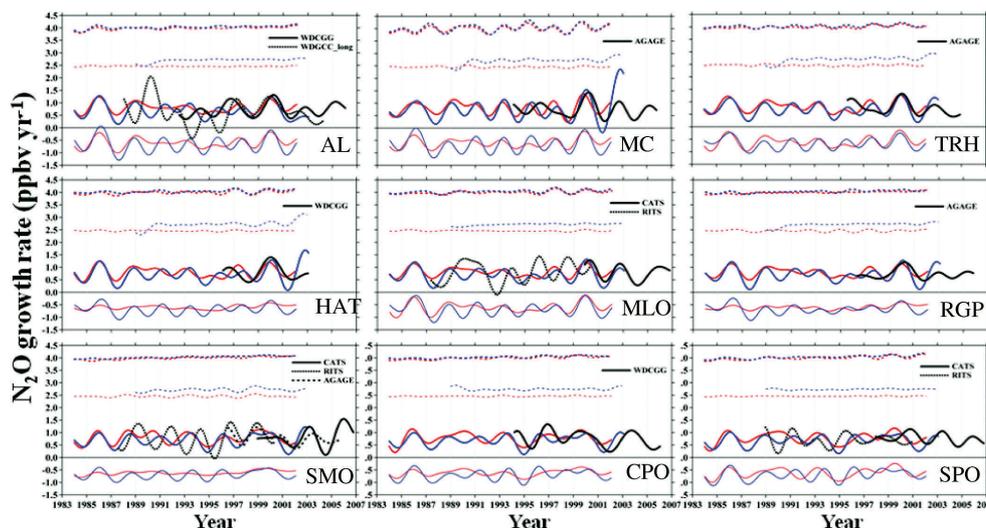


Fig. 2 Simulation of  $\text{N}_2\text{O}$  growth rates using AGCM at T42 horizontal resolution and 67 vertical layers are compared with observations at 6 northern hemisphere (top two rows) and 3 southern hemisphere (bottom row) stations. Several types of observations are shown when available for greater confidence on observations (see legends). The cases of model simulations are due to different implementation of  $\text{N}_2\text{O}$  chemistry in the stratosphere (broken lines are for no chemistry cases with different emission scenarios, solid lines are for model runs with different photochemistry schemes). Blue and red lines are for AGCM transport nudged with NCEP2 and ECMWF reanalyzed meteorology, respectively.

### 3. Analysis of CO<sub>2</sub> flux variability by FRCGC and JMA transport models

This year, we adopted the 64-region inversion method [1, 2] and simulate tracer transport with low resolution (2.5° in horizontal) CDTM with JMA reanalysis (JRA) meteorological fields. This number of region is one of the highest resolutions in current carbon-cycle research. We have estimated CO<sub>2</sub> monthly mean fluxes from 1990 to 2000 using time-dependent inversion [3] with two transport models (FRCGC and JMA). The 82 observational sites (Fig. 3) are selected from WMO/WDCGG on the condition that the data selection rate by the inversion is larger than 60%.

Fig. 4 shows our analysis results. We find that there is little difference in estimated CO<sub>2</sub> flux variability in global scale between FRCGC and JMA transport model. In regional scale, the estimated CO<sub>2</sub> fluxes show the similar phase and amplitude of CO<sub>2</sub> flux variability but there are some growing differences in northern and tropical land region between two models.

The less constrained land areas (South–West Temperate

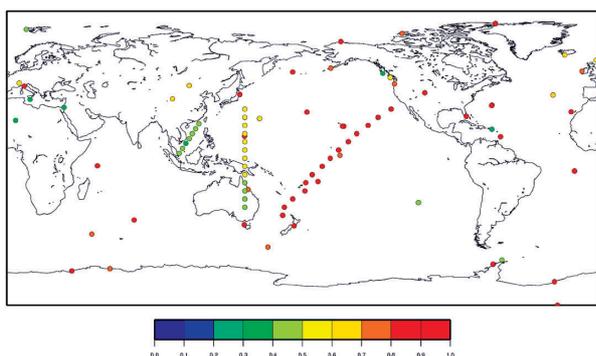


Fig. 3 The observational sites used in the inversion intercomparison (source: <http://gaw.kishou.go.jp/wdcgg.html>). The color shows the data availability rate in the analysis period (1990–2000). The colour bar is shown at an interval 0.1 and full range of 0–1.

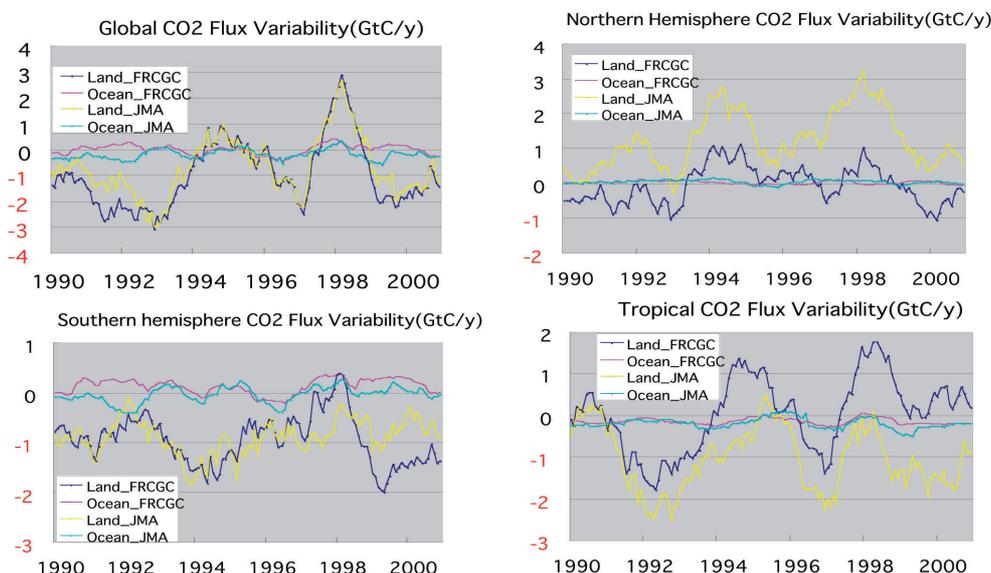


Fig. 4 CO<sub>2</sub> flux estimation by time dependent inversion with FRCGC and JMA transport model.

Asia, South–West Australia and North–West Boreal Asia) tend to show larger difference in estimated fluxes. These growing differences seem to come from the differences in meteorological data and tracer transport schemes. We have a plan to use finer horizontal resolution (1 × 1°) transport model in the intercomparison. In addition, as both models participate in another tracer transport model intercomparison project (TransCom), we could compare our models in several aspects in future.

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### References

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# 全球・地域スケール化学輸送モデルによる大気組成変動とその気候影響の研究

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## 1. 境界層を横切るモデル輸送の診断

境界層 (PBL) を横切るモデル輸送の診断を、CO<sub>2</sub>, ラドン, SF<sub>6</sub> について行った。地上付近で温度逆転の起こりやすい冬季の夜間には、大気中寿命の短いラドンはPBL中に滞留するので、ラドンをを用いた診断の情報は、逆モデルによってCO<sub>2</sub>の地表におけるソース・シンクを議論するのに適したモデルを選択する上で非常に有用であることが分かった。他方、SF<sub>6</sub>はその排出が空間的に非常に不均一であり、且つ対流圏に消失過程が全くないので、CO<sub>2</sub>などの領域スケールでの季節変化パターンへの輸送診断に有効であることが分かった。

## 2. 大気中N<sub>2</sub>Oの季節変化

大気中N<sub>2</sub>O濃度のシミュレーションを1983–2002年について行った。その結果、N<sub>2</sub>O濃度の緯度分布、及び季節変化の再現には、成層圏化学が決定的に重要な要因となっていることが分かった。N<sub>2</sub>Oの成層圏光化学を入れない場合には、N<sub>2</sub>Oの年々変動はほとんど生じず、実測を全く再現できないが、成層圏化学を入れることによって年々変動の幅と位相を良く再現できる。これは圏界面を横切る成層圏-対流圏交換のアノーマリが、対流圏のENSO、成層圏のQBOなどの気候変化とリンクして、地表付近のN<sub>2</sub>O濃度の年々変動を引き起こすためであることが分かった。

## 3. FRCGC及びJMA輸送モデルによるCO<sub>2</sub>フラックス変動の解析

64地域分割逆モデルとJMA再解析気象場を用いて2.5°分解能での輸送のシミュレーションを行った。二つの輸送モデル (FRCGC 及びJMA) による時間依存逆モデル計算により1990–2000年のCO<sub>2</sub>月平均フラックスを推定した。一般的にはこれらのモデル間には、CO<sub>2</sub>フラックス変化の振幅、位相に大きな差は見られなかったが、北半球高緯度域及び熱帯の陸域においては、これら二つのモデル間に差が見られた。

キーワード: 二酸化炭素, 亜酸化窒素, 化学輸送モデル, 境界層, 年々変動