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

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The JMA transport model is used for source/sink inversion atmospheric CO_2 and forward transport modeling. Using the inverse modeling fluxes, Greenhouse Gases Monitoring Information system has been developed to provide simulated CO_2 concentrations in near-real time by the JMA. The forward transport modeling errors due to the used of meteorology and transport model have been estimated using JMA and NIES/FRCGC models. After validating the CCSR/NIES/FRCGC AGCM-based Chemistry-Transport Model (ACTM) for tracer transport at various time and space scales, ACTM is used to simulate CH_4 and N_2O between earth's surface and the mesosphere (~90 km) by implementing simple photo-chemical loss schemes and representing surface fluxes realistically. The model results have been compared with observations in situ near the earth's surface and satellite-based remote sensing measurements in the stratosphere. We also have been developing AGCM-based chemistry and aerosol coupled climate simulations combining a chemistry climate model (CHASER) and an aerosol climate model (SPRINTARS). By introducing nitrate aerosol (NO_3^-) to this model, we find nitrate aerosols play an important role in the observed changes in regional climate like Asian monsoon and imply that the future Asian climate will be affected by nitrate aerosol rather than by sulfate.

Keywords: gases and aerosols, chemistry-transport modeling, source-sink inversion, climate model

1. Inter-comparison of CO_2 forward simulations using two CTMs and flux inversions

This year, we have compared CO_2 transport modeling results using FRCGC/NIES (CTME; Maksyutov et al., 2008) and JMA (CDTM) transport models driven by NCEP and JRA-25 reanalysis meteorology, respectively. The TransCom-3 pre-subtracted CO_2 tracers (fossil-fuel burning, neutral biosphere and air-sea exchange) are simulated for the period of 1988–2001. In case A, we used inter-annually varying (IAV) reanalysis meteorology. In case B, we repeated specific year's meteorology for the entire simulation period (1988–2001). Figure 1 shows the difference between two CO_2 time series in case A for both the CTMs. In most stations, CDTM shows higher seasonal amplitude than CTME. In case B, the standard deviation of CDTM due to inter-annually varying wind shows higher value than that of CTME (Table 1). The standard deviation of the difference between CTME and CDTM is higher than the standard deviation of CDTM (also CTME) due to IAV wind. This means that the between model variability is larger than the inter-annual model variability caused by the meteorology. We should continue model inter-comparison experiment with tracers of better known surface fluxes.

We have developed high resolution inversion system in this project. The system shows better performance (observational data selection rate, etc.) than current inversion system. Also we have gained sufficient knowledge and technique about high resolution inversion system from this intercom-

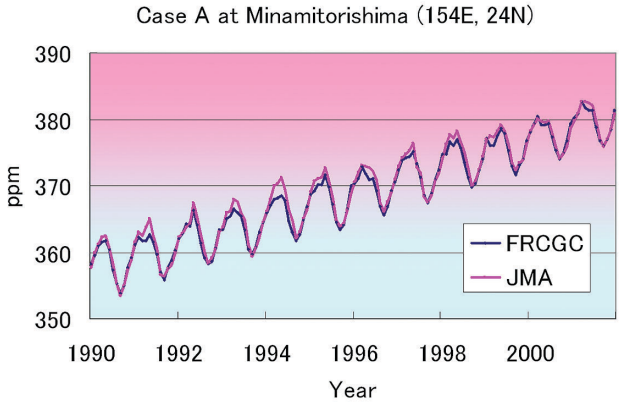

Fig. 1 The difference between two transport model simulation in case A (using IAV meteorology).

Table 1 Standard deviation of "between model" and "between wind" differences (in ppm).

Site Name (Location)	JMA-FRCGC (Case A)	FRCGC (Case A–B)	JMA (Case A–B)
Minamitorishima (154˚E, 24˚N)	0.729	0.439	0.627
South Pole (90˚S)	0.603	0.143	0.394

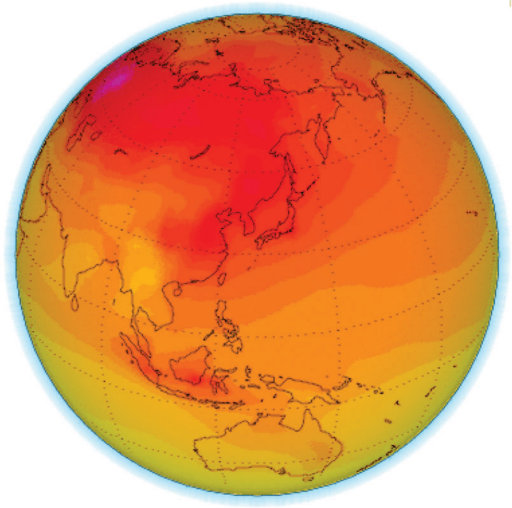

Fig. 2 A sample picture of surface CO_2 concentration is shown from Greenhouse Gas Monitoring Information by JMA.

parison project. JMA has a plan to upgrade Greenhouse Gas Monitoring Information (will be provided from early 2009) using this high resolution inversion system in a few years (Fig. 2).

2. Simulation of nitrous oxide (N_2O) and methane (CH_4) using ACTM

To understand transport timescales of atmospheric constituents within the troposphere, transport times of air parcels from the surface to different regions of the troposphere ("age") are estimated using the newly adopted CCSR/NIES/FRCGC AGCM-based Chemistry-Transport Model (ACTM). The age provides important diagnostics for the tracer transport mechanisms in the troposphere due to advection, cumulus convection and vertical diffusion at daily, weekly and monthly time scales. The model simulations of an "ideal" transport tracer (SF_6) are compared with observations and shown that ACTM is capable of producing near-perfect north-south latitudinal gradients in observed SF_6. Interhemispheric exchange times (τ_{ex}) are calculated from both the observed and simulated SF_6 time series at the 6 observing sites to validate ACTM tracer transport in analysis nudging mode (see Patra et al., 2008 for details).

We used the 67-layers version of ACTM for simulating the gases with stratospheric chemical loss (N_2O, CFC-12, CH_4). The ACTM is nudged with NCEP2 reanalysis for the whole periods of simulations of CH_4 and N_2O (1991–2007). The results were validated by comparing with ground-based observations and satellite data in the stratosphere. In understanding global N_2O cycles, three processes have to be considered, namely, (1) surface fluxes in the troposphere, (2) photochemical loss in the stratosphere and (3) troposphere-stratosphere exchange (STE). Stratospheric N_2O gradients produced by dynamics and photochemical loss control the STE intensity, and thereby the surface concentrations. This is identified as one of the main sources of uncertainty in estimating N_2O surface fluxes by inverse modeling. It should be noted here that most CTMs utilized in N_2O and CH_4 research do not treat the stratospheric chemistry explicitly, and thus are prone to discontinuity at the tropopause region, which can results in unrealistic STE rates. Figure 3 shows latitude-pressure cross-sections of monthly and zonal mean N_2O concentration obtained by model and MLS/Aura satellite observation in January and July of 2005. Generally, the concentration gradients from tropics to higher latitudes and its seasonality are well reproduced by the model. Both in observation and model, regions of enhanced N_2O upwelling from the troposphere into the stratosphere appear at the latitude of 0–30˚ in the summer hemisphere.

Likewise N_2O, the simulation of CH_4 in the lower atmosphere also depends on surface flux distribution, seasonality and STE. However, the largest loss of CH_4 occurs in the troposphere by the reaction with hydroxyl radical (OH). Detailed comparison of CH_4 simulations (using a suitable set of surface fluxes, CHASER OH and ACTM) with ground-based observations have been conducted (Patra et al., 2009). In Fig. 4 we show the comparison of quasi-biennial oscillation (QBO) cycle in stratospheric distribution of atmospheric CH_4. During the easterly QBO phase, CH_4 is efficiently transported upward in the tropical lower stratosphere. On the contrary, meridional CH_4 transport is strongest in the lower stratosphere during westerly QBO phase and relative low CH_4 concentrations can be seen in the middle and upper troposphere. These conditions are well reproduced by the ACTM when nudged with NCEP2 (or ECMWF ERA40) meteorology.

3. Development of chemistry-aerosol coupled climate model

We have been integrating chemistry and aerosol coupled climate simulations combining the chemistry climate model CHASER (Sudo et al., 2002) and aerosol climate model SPRINTARS (Takemura et al., 2003). Both model compo-

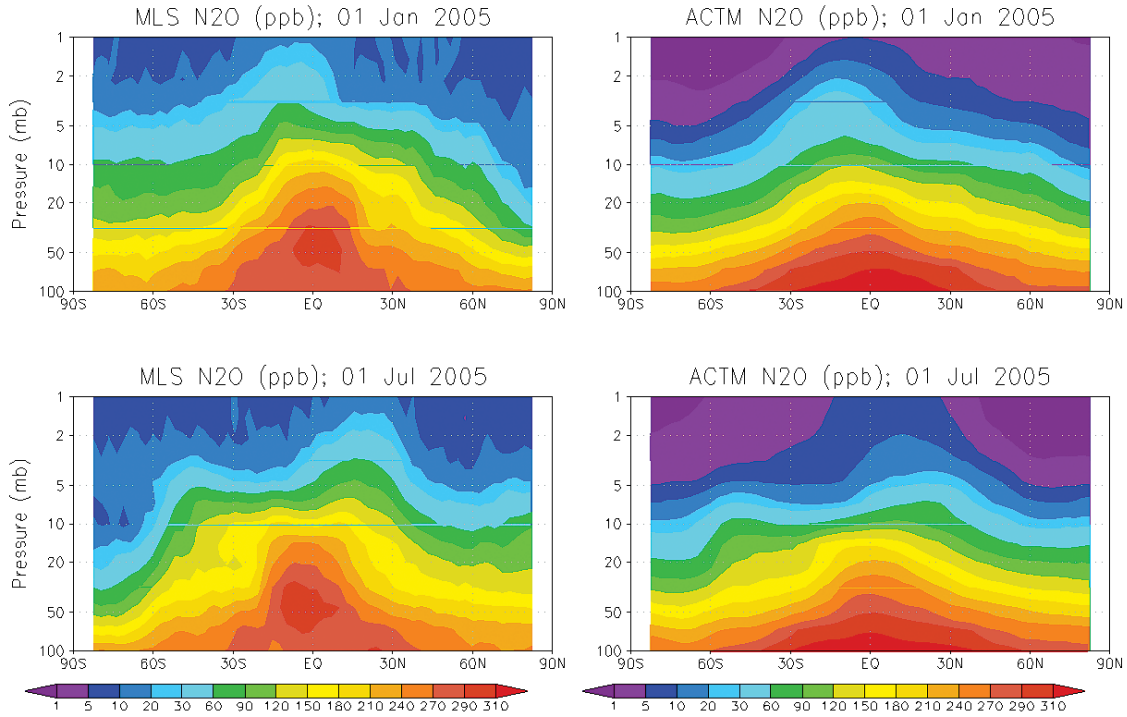

Fig. 3 Comparison of latitude-pressure distributions of N_2O measured by MLS instrument onboard the Aura satellite (source: JPL/NASA, http://mls.jpl.nasa.gov) and ACTM simulation. Model results are sampled on the measurement day and location before averaging.

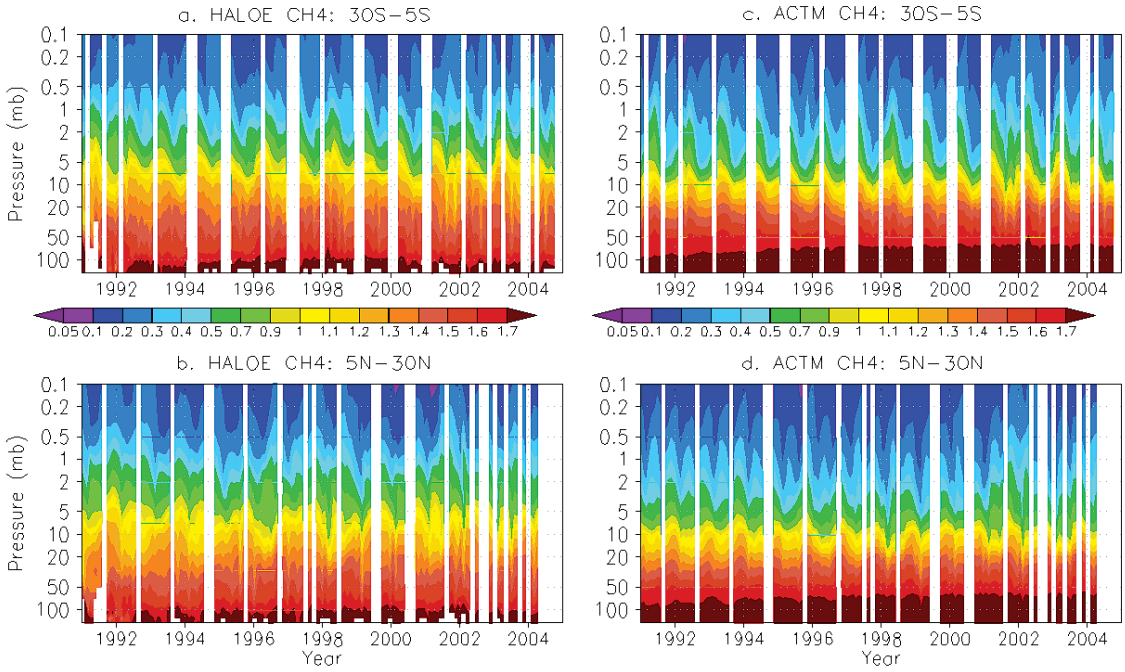

Fig. 4 Comparison of zonal average timeseries of CH_4 as measured by the HALOE/UARS instrument and simulated by ACTM during the period of 1991–2004, averaged over 5–30° latitudes in each hemisphere. Model results are sampled on the measurement day and location before averaging.

nents have been developed in the framework of the CCSR/NIES/FRCGC climate model. In the FY-2008, we newly introduced simulation of nitrate aerosol (NO_3^-) to the global aerosol-climate model using an aerosol thermodynamics module (ISORROPIA). The previous versions of our aerosol model consider only sulfate and organic aerosols as hygroscopic particles that can function as CCN (Cloud Condensation Nuclear). However, nitrate aerosol, produced by oxidation of NO_x, is another important hygroscopic aerosol as well. Our simulation shows that there are anom-

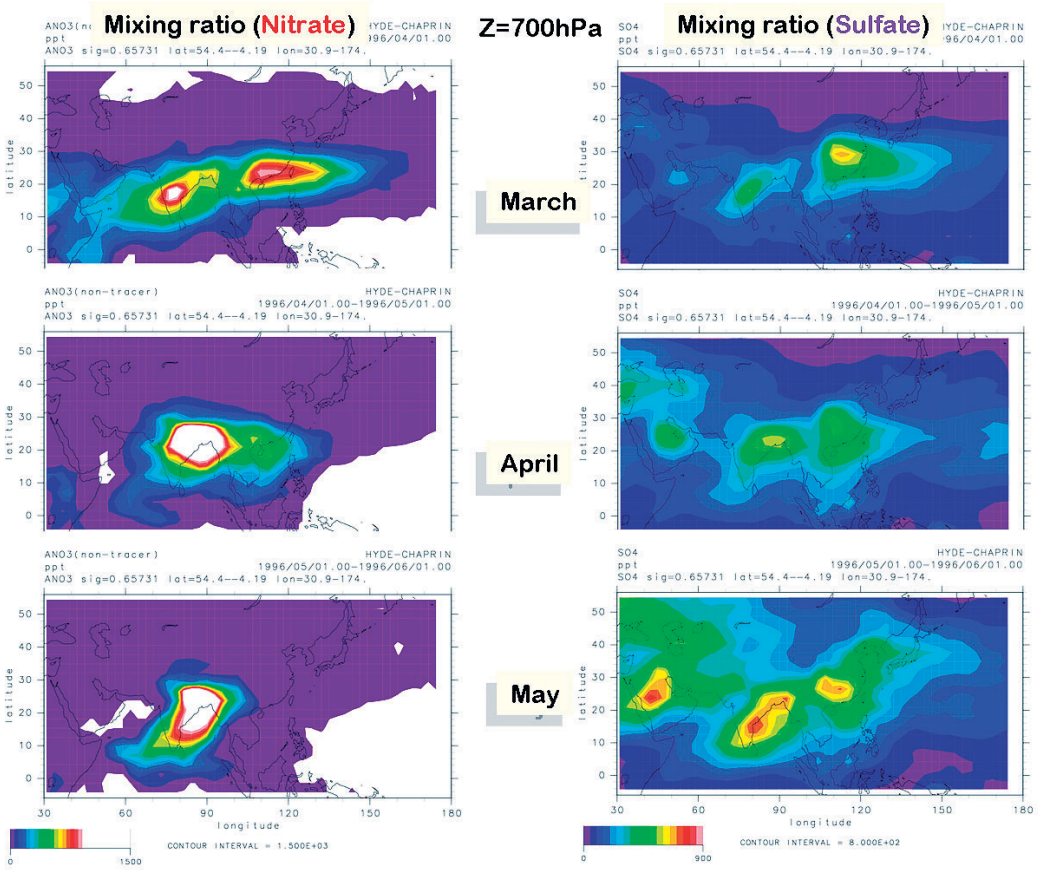

Fig. 5 Calculated nitrate (left) and sulfate (right) mixing ratios at 700 hPa altitude in Asia for March, April, and May.

alously high concentrations of nitrate aerosol in South Asia (particularly around India), coming from abundant ammonium and lower sulfate emission in this region (Fig. 5). In India, free tropospheric mixing ratio and number concentration of nitrate are both larger than those of sulfate in winter to early summer. This result suggests nitrate aerosol may play an important role in the observed changes in regional climate like Asian monsoon. This result further implies that future Asian climate may be affected by nitrate aerosol rather than by sulfate. This is because continued increase in emissions of nitrate precursor gases (NO_X) are expected by most of the emission scenarios, but decreases in sulfate precursors (SO_2) emissions are projected especially in Asia. Development of such a chemistry-aerosol coupled climate model is required for evaluating leverages under different projection scenarios of sulfate and nitrate precursor gases.

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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. 大気中二酸化炭素濃度の前進計算および逆解析の相互比較

2つの大気輸送モデル、FRCGC/NIESのCTMEおよび気象研のJMA CDTMを用いて輸送比較実験を実施した。1988～2001年についてCO_2前進計算を行い、JMA CDTMはFRCGC/NIESよりも大きな季節変動を示す傾向があること、モデル間の輸送の違いは、各モデル内の年ごとに違う気象場に起因する輸送の違いよりも大きいことなどが分かった。同時に本共同実験に基づいて高解像度逆解析システムを開発し、観測データの選別率などで従来のシステムを上回る性能を示した。今回得られた知見は、気象庁が来春に公表開始を予定している温室効果ガス監視情報の高度化に活用する計画である。

2. 大気化学輸送モデルを用いた一酸化二窒素とメタンの計算

大気輸送の時間スケール理解のためにCCSR/NIES/FRCGC 気候モデルをベースとした大気化学輸送モデル(ACTM)を用い、対流圏における空気の年代の計算を行い、移流・対流・拡散からなる大気輸送メカニズムを調べた。さらに六フッ化硫黄(SF_6)を用いて、半球間大気交換時間を計算し観測値と比較した結果、本モデルの輸送は極めて現実に近いことが確認された。このように対流圏－下部成層圏における大気輸送メカニズムを把握した上で、鉛直67層のACTMに単純な化学計算を導入し一酸化二窒素(N_2O)とメタンの計算を行い、地上連続観測や成層圏衛星リモートセンシング観測との比較を行った。N_2Oは熱帯上部対流圏から高高度・極方向への濃度勾配とその季節変化、および夏半球熱帯域の上昇流に伴う濃度変化についてはよく再現されていたが、極渦内の濃度については過大評価気味であった。メタンについては衛星で観測された成層圏内の濃度変動におけるQBOシグナルも本モデルで再現された。

3. 全球エアロゾル・気候モデルの開発

全球エアロゾル・気候モデル SPRINTARSおよび全球化学・気候モデルCHASERを軸とした大気化学・エアロゾル結合気候モデル開発の一環として、本年度は、硝酸塩(NO_3^-)を追加し、硫酸塩計算の改善を行った。エアロゾル熱力学平衡モデル(ISORROPIA)を化学・エアロゾル計算過程に新たに導入し、硫酸塩・アンモニウム塩・硝酸塩の複合系のシミュレーションを可能にし、アンモニア・アンモニウムの硫酸塩(吸湿性)への影響も反映できるようになった。本モデル計算を行った結果、インド・バングラディシュ周辺で硝酸塩エアロゾルが特に高濃度となることが分かった。このような局所的に高濃度の硝酸塩エアロゾルは雲・降水過程にも関与していると考えられ、アジアモンスーンなどにも影響している可能性が示唆される。さらに、硝酸塩の前駆気体である窒素酸化物は、アジア域において今後も増加すると見込まれており、硝酸塩が将来のアジアモンスーンに顕著に影響する可能性があると考えられる。

キーワード：大気中微量成分とエアロゾル, 大気化学輸送モデリング, 逆解析による放出・吸収源推定, 気候モデル