Hybrid Monte Carlo-Gear's Solver to Retrieve the Vertical Profiles of Minor Atmospheric Constituents for Cloud Microphysical Modelling

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Valiamala P.O., Thiruvananthapuram – 695 547, Kerala, India

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Kavita Patnaik

(SC19D019)

Under the joint supervision of

Dr. Amit P. Kesarkar

Scientist/ Engineer- SF

NARL, Gadanki

Dr. Chandrasekar A.

Outstanding Professor

IIST, Thiruvananthapuram

Research work carried out at

National Atmospheric Research Laboratory

Department of Space, Government of India, Gadanki, India- 517 112

Abstract

Determining the number concentration of minor constituents in the atmosphere is very important as it controls the chemistry processes of the entire troposphere. These constituents may act as cloud condensation nuclei (CCN) and ice nuclei (IN), impacting heterogeneous nucleation inside the cloud. However, the estimations of the number concentration of CCN/IN in cloud microphysical parameters are associated with uncertainties. In the present work, a hybrid Monte Carlo Gear solver has been developed to retrieve profiles of CH_4, N_2O, and SO_2. The idealized experiments have been carried out using this solver to retrieve the vertical profiles of these constituents over four megacities, viz., Delhi, Mumbai, Chennai, and Kolkata. Community Long-term Infrared Microwave Coupled Atmospheric Product System (CLIMCAPS) dataset around 0800 UTC (2000UTC) has been used for initializing the number concentration of CH_4, N_2O, and SO_2 for daytime (nighttime). The daytime (nighttime) retrieved profiles have been validated using 2000 UTC (next day 0800 UTC) CLIMCAPS products. ERA5 temperature dataset has been used to estimate the kinematic rate of reactions with 1000 perturbations determined, using Maximum Likelihood Estimation (MLE). The retrieved profiles and CLIMCAPS products are in very good agreement, as evidenced by the percentage difference between them, within the range of 1.3×10^{-5} – 60.8 %, and the coefficient of determination mainly within the range between 81-97 %. However, during the passage of tropical cyclone and western disturbance, its value became as low as 27% and 65% over Chennai and Kolkata, respectively. The development of synoptic scale systems, such as western disturbances, tropical cyclone Nivar, and easterly waves, led to disturbed weather over these megacities. The retrieved profiles during disturbed weather conditions have large deviations in vertical profiles of N_2O. However, the profiles of CH_4 and SO_2 showed lower deviation. It is inferred that incorporating the above methodology in the dynamical model will help simulate the realistic vertical profiles of the minor constituents in the atmosphere.

The accurate forecast of the diurnal cycle of the number concentration of trace gases is vital due to their influence on precipitation processes, where they control the number concentration of cloud condensation nuclei (CCN). A 1-D hybrid Monte Carlo-Gear solver was developed to retrieve vertical profiles of the number concentration of CCNs,

for microphysics modeling has been tested for representation of the diurnal cycle in this study. The retrieved profiles of CH_4 and SO_2 have been tested with the Copernicus Atmosphere Monitoring Service (CAMS) model at 3-hour time intervals over four megacities for rainy and non-rainy days. The retrieved profiles have shown diurnal variation up to 18 UTC at all pressure levels with either lead or lag time, similar to the CAMS model. After 18 UTC, there is observed a rapid increase in the number concentrations.

During non-rainy days, the 1-D model slightly overestimated (underestimated) the maximum (minimum) number concentrations of CH_4 over Delhi, whereas concentrations are overestimated over Kolkata, Chennai, and Mumbai. Forecasted CH_4 has a good (weak) correlation over Chennai (Mumbai), respectively. The 1-D model overestimated (overestimated) the maximum (minimum) number concentrations of SO_2 over Delhi. However, the maximum (minimum) concentrations are underestimated (overestimated) in Kolkata, Chennai, and Mumbai. The number concentrations of SO_2 have shown a good correlation for all megacities except Delhi. CH_4 number concentration is overestimated during rainy days. Delhi and Kolkata show a good correlation of CH_4 during rainy days. SO_2 during rainy days is underestimated, except over Chennai, and both models show a good correlation, except over Mumbai. Overall, it can be stated that the 1-D hybrid solver successfully simulates the monthly mean diurnal variation of vertical profiles of CH_4 and SO_2.

A hybrid Monte-Carlo Gear solver, developed earlier, has been improved to retrieve the vertical profiles of CH_4 and N_2O during disturbed weather situations, such as western disturbances, tropical cyclones, and heavy rainfall events, over the four megacities. Due to rapid changes in the temperature, during the passage of these systems over megacities, the percentage differences of CH_4 and N_2O number concentrations were large as compared to the Community Long-term Infrared Microwave Coupled Atmospheric Product System (CLIMCAPS). Hence, the hybrid solver has been modified by improving the maximum likelihood estimates of vertical temperature profiles. The number concentrations of CH_4 and N_2O during these weather events since 2012 have been obtained from the CLIMCAPS dataset for bias correction. It was found that the modified methodology has improved the retrieval of CH_4 and N_2O vertical profiles by reducing the error percentages during daytime and nighttime over these megacities. The percentage

error in the estimated number concentrations of CH_4 and N_2O is decreased significantly during (i) the passage of the western disturbance and rainy days of August 2020 over Delhi, (ii) the rainy days of June 2020 over Kolkata, (iii) the influence of supercyclonic storm Nivar (24 and 25 Nov 2020) over Chennai and (iv) rainy days of July 2020 over Mumbai. Implementing the above solver in the global model may lead to more accurate retrievals of the vertical profiles of the number concentrations.