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# Environmental Geochemical Cycle Modelling Research

## Project Representative

Takashi Sekiya Earth Surface System Research Center, Research Institute for Global Change, Japan Agency for Marine-Earth Science and Technology

## Authors

Jagat S. H. Bisht <sup>\*1</sup>, Prabir K. Patra <sup>\*1,2</sup>, Masayuki Takigawa <sup>\*3</sup>, Yugo Kanaya <sup>\*1</sup>, Masahiro Yamaguchi <sup>\*4,1</sup>

<sup>\*1</sup>Earth Surface System Research Center, Research Institute for Global Change, Japan Agency for Marine-Earth Science and Technology, <sup>\*2</sup>Research Institute for Humanity and Nature, Kyoto, <sup>\*3</sup>Institute of Arctic Climate and Environment Research, <sup>\*4</sup>Earth System Division, National Institute for Environmental Studies Tsukuba

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## 1. Introduction

CO<sub>2</sub> is a well-mixed and long-lived greenhouse gas (GHG) in the atmosphere which has both anthropogenic and natural sources. CO<sub>2</sub> concentration is increasing steadily in the atmosphere because emissions by anthropogenic activity [1]. The concentration of CO<sub>2</sub> in the Earth's atmosphere has been steadily rising due to various human activities, primarily the burning of fossil fuels (such as coal, oil, and natural gas) for energy, deforestation, and industrial processes.

To estimate gridded CO<sub>2</sub> emissions from various sources, such as industrial, residential, commercial, and transportation processes, anthropogenic CO<sub>2</sub> emission inventories have been developed and are regularly updated and improved for better accuracy [2]. Several studies have demonstrated that the current concentration of CO<sub>2</sub> in the atmosphere is largely due to human activities, particularly the burning of fossil fuels [1]. It has been reported that more than 60% of global fossil-fuel CO<sub>2</sub> emissions are produced in cities [3], making them important targets for mitigation efforts.

In addition to anthropogenic CO<sub>2</sub> emissions, atmosphere-biosphere carbon exchange significantly affects the atmospheric CO<sub>2</sub> concentration and is equally important to understand the atmospheric carbon cycle. At sub-daily scale, atmosphere-ecosystem CO<sub>2</sub> exchange is mainly determined by physiological and phenological responses driven by meteorological conditions such as solar radiation, temperature, and humidity. A regional model fully coupled with meteorology that can run in a kilometer scale or below could be used here to address the mesoscale transport of carbon dioxide (CO<sub>2</sub>) and its flux exchange between the biosphere and the atmosphere [4]. Ahmadov et al. [5] coupled Vegetation Photosynthesis and Respiration Model (VPRM) [6] module with the WRF model, and conducted CO<sub>2</sub> modeling over Europe. This framework has also been utilized in other studies [7, 8], which have demonstrated the effectiveness of the atmosphere-biosphere coupled model in capturing mesoscale CO<sub>2</sub> transport at regional and local scales with significant improvements. VPRM CO<sub>2</sub>

fluxes are required to be fine-tuned using observed vegetation fluxes for the land use types in the region [9].

This study is performed to evaluate the performance of WRF-GHG over Japan, specifically the Kanto region, centered around Tokyo. The Kanto region is the largest economic area in Japan, that includes Tokyo, which boasts one of the largest economies of any city in the world. The Kanto region alone accounts for about 40% of Japan's GDP (gross domestic product), where the chemical, steel, machine industries, thermal power plants, automobile traffic, residential facilities account for a high proportion of CO<sub>2</sub> emissions. Several sites over Kanto region have continuous CO<sub>2</sub> surface data and aircraft observations that can be used to better evaluate the model performance.

## 2. Materials and Methods

We use WRF with coupled chemistry (WRF-Chem version 4.2.1) model, which uses the GHG module to simulate the transport of CO<sub>2</sub>, methane (CH<sub>4</sub>), and carbon monoxide (CO) (hereafter referred as WRF-GHG). The module includes VPRM to simulate the CO<sub>2</sub> biogenic emissions. The WRF-GHG set-up details are given in Jagat et al. [10]. We used the similar setup here.

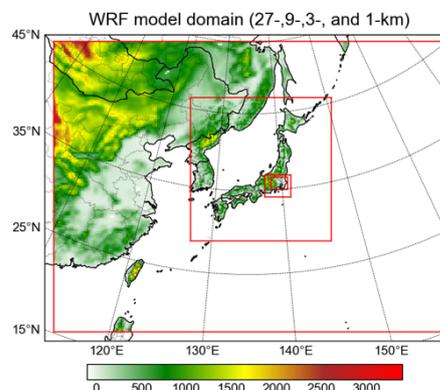


Figure 1. The diagram illustrates the domain configurations for the model simulations (four domains: 27, 9, 3, and 1 km) and displays the terrain height for each domain.

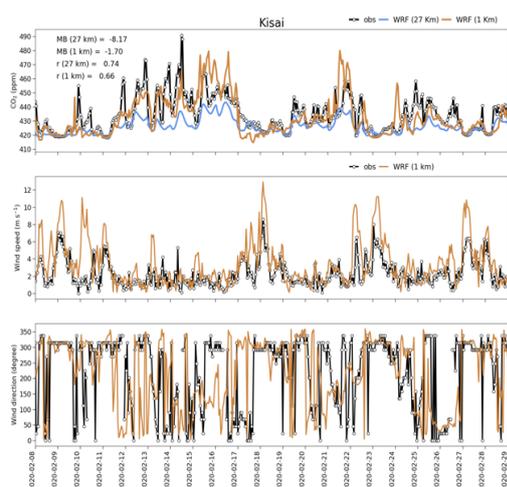


Figure 2. CO<sub>2</sub> concentrations and winds at Kisai observation site during February 2018. The observations (black) shown along with model simulation with EAGrid for domain 01 (27 km) and domain 04 (1 km). Statistics of model observation comparison is given in the top-panel.

### 3. CO<sub>2</sub> concentration observations

Atmospheric CO<sub>2</sub> hourly concentration in-situ data is analyzed at Kisai (36.10°N, 139.57°E, altitude; 34 m). The in-situ CO<sub>2</sub> concentration data recorded with VIA-510R (HORIBA Ltd.) with measurement uncertainty of ~0.3 ppm at Kisai observations sites is obtained from the World Data Centre for Greenhouse Gases (WDCGG) operated by the Japan Meteorological Agency (JMA). We also use CONTRAIL Continuous CO<sub>2</sub> Measuring Equipment (CME) CO<sub>2</sub> concentration data aboard Japan Airlines’ commercial airliner flights [11] for flights arriving and departing from Haneda (HND) airport in Japan ([10]; Figure 1).

### 4. Results and Discussion

We compared the WRF-GHG simulation for two spatial resolutions; coarser resolution (27 km) and finer resolution (1 km). In the case of coarser resolution, the model simulations are performed for the outermost domain independently (Fig. 1; 27 km) during February 2018 without taking other domains into account. We may notice instances (for e.g., 02-03 February 2018) where both 27 km and 1 km resolution model simulations significantly underestimated the CO<sub>2</sub> concentration. We have also shown the wind plots in Figure 2 for Kisai which display wind direction mismatches between the model simulations and observations during 02-03 February 2018. The observed wind direction is from the North-West, while the simulated wind direction is from North-East. We have noticed a hotspot over the North-West direction in the emission map (Figure not shown). Similarly, a wind direction mismatch could be noticed during 16-17 January 2018 between the model simulations and observations. In the wind plots (Fig. 2), we may notice dominant model winds are from South-West (greater Tokyo region), whereas the dominant observed winds are from North-East

direction in 1 km simulations. Additionally, we noticed that, when the prolonged synoptic condition associated with high CO<sub>2</sub> concentrations (Fig.2; 07-10 February 2018; wind speed < 4 m s<sup>-1</sup>), the model simulations with 1 km resolution better capture the observed variability compared to 27 km model simulations.

We have also demonstrated the model-observation comparison for CO<sub>2</sub> concentration and winds during February 2020 (Fig. 3). Here we also noticed that, the prolonged synoptic condition (10-16 February 2020; wind speed < 4 m s<sup>-1</sup>) associated with high CO<sub>2</sub> concentrations leads to better CO<sub>2</sub> simulations with a 1 km model horizontal resolution compared to 27 km. We concluded that, the high-resolution simulation amplifies the model bias which is introducing more transport error, and reduces the model-observation correlation. Additionally, the high-resolution modeling found to better reproduce the observed variability for prolonged synoptic conditions that involves stagnant wind (wind speed < 4 m s<sup>-1</sup>).

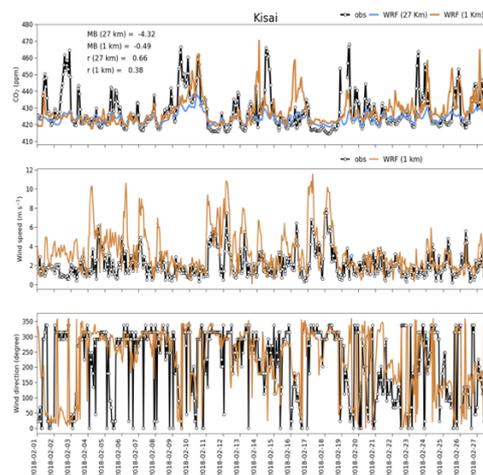


Figure 3. Same as Figure 2 but for the year 2020.

We compared the WRF-GHG simulations with CONTRAIL aircraft observations for two spatial resolutions; coarser resolution (27 km) and finer resolution (1 km) (Fig. 4a). It may be noted that coarser resolution simulations largely underestimated the observed CO<sub>2</sub> concentration up to an altitude range of approximately 2400 m (Fig. 9a). Above that, the 1 km and 27 km model simulations are similar. The under-estimation of CO<sub>2</sub> concentration in coarser resolution WRF-GHG simulations could be attributed to the under-representation of fine scale vertical transport processes [12] such as: vertical diffusion and convection. On the other hand, 1 km simulations reasonably reproduced the observed variability in the vertical distribution of CO<sub>2</sub> concentration.

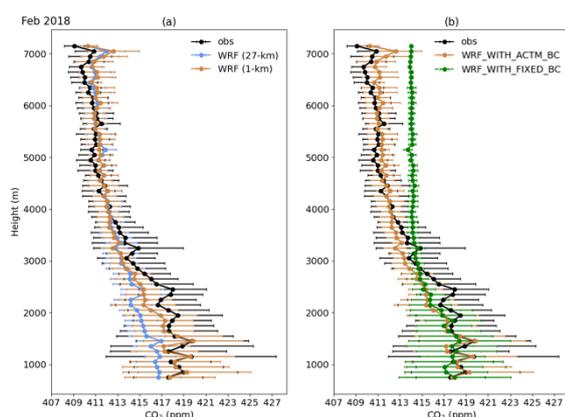


Figure 4. Comparison of CO<sub>2</sub> vertical distribution between CONTRAIL and WRF-GHG simulations during February 2018 for: (a) finer (1 km) and coarser (27 km) model domains, (b) fixed (a constant value) initial and lateral boundary conditions and with MIROC4-ACTM initial and lateral boundary conditions to WRF-GHG. The error bar represents the standard deviation.

Our study also included a sensitivity analysis of boundary conditions, where we conducted CO<sub>2</sub> concentration simulations using fixed boundaries instead of MIROC4-ACTM (Fig. 4b). The analysis showed that, beyond an altitude of 3200 m, a systematic bias of approximately 4 ppm exists in the CO<sub>2</sub> profile when fixed (a constant value) boundary conditions are applied, as compared to the results obtained when using boundary conditions from MIROC4-ACTM. Furthermore, when using fixed lateral boundary conditions, plume-like signatures as observed in the CO<sub>2</sub> profile around 7000 m (Fig. 4b) are not reproduced. We conclude that the selection of a model field with a wider domain (MIROC4-ACTM for this study) for lateral boundary conditions to WRF-GHG is critically important.

## 5. Summary

The model sensitivity to horizontal resolution was examined. We noticed that, model simulations are sensitive to model resolution at surface observation site. High-resolution modeling reproduced the observed variability during prolonged synoptic conditions (wind speed < 4 m s<sup>-1</sup>) associated with high CO<sub>2</sub> concentration.

The advantage of high-resolution modeling was evident in aircraft profile comparisons, indicating a clear superiority over coarser resolution model simulations. The vertical profile comparison shows that above certain altitude from surface (700m in our case) the vertical transport processes (such as convection and vertical diffusion) are more dominant which are better represented in high resolution modeling.

Another significant finding is the model's sensitivity to lateral boundary conditions. By comparing with aircraft observations, we can pinpoint the altitude from where boundary condition

effects become paramount. The study reveals a persistence systematic bias (~ 4 ppm) beyond an altitude of 3200 m (February 2018) when fixed boundary conditions are applied. Our lateral boundary sensitivity results suggest that CO<sub>2</sub> simulations beyond 3200 m are influenced by long-range transport from Eurasia.

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