

The authors are grateful to the reviewer's valuable comments that improved the manuscript.

### **Anonymous Referee #1**

#### General Comments

This is a good paper that uses the WRF-CHEM model to analyse observations (predominantly airborne measurements) from the ICARTT field campaign, to investigate the transport and evolution of pollution from the New York area out over the Atlantic. A key feature is the transport of the pollution plumes above the stable marine boundary layer, decoupling the pollutants from the surface. WRF-CHEM is run with different chemical initial and boundary conditions (IC/BCs), showing that anthropogenic and biogenic emissions from N. America are important on the background levels of pollutants observed and that biomass burning plumes also impacted the area during the campaign. The paper helps to further understand the ICARTT observational data and also illustrates the importance of IC/BC in regional model studies. This paper is well written. On the whole the description of the work performed and the discussion is detailed and thorough. The paper should be published in ACP, subject to a few minor points below.

#### Specific Comments

A major aspect of the paper is the impact of the IC/BCs, specifically dynamic IC/BCs. However, what is not very clear to me is the way these are treated in the control run. There are a few places in the text (e.g. page 14040, lines 22-25; page 14055, lines 18) which seem to imply that the control run had no chemical IC/BCs, where as in fact prescribed static chemical IC/BCs were employed. i.e. the text states that chemical IC/BCs are used in the sensitivity runs so thus implies that they were not in the control run.

In order to clarify chemical IC/BCs in the control simulation, the title of Sect. 3.1 'WRF-Chem model and experimental setup' was replaced with 'WRF-Chem model and experimental setup for control simulation'. In addition, further explanations on the chemical IC/BCs were added in the section, including some reference papers. Below is the revised paragraph describing the chemical IC/BCs in the control simulation.

Initial mixing ratios of chemical species are assigned following McKeen et al. (2002) and Liu et al. (1996) which consist of spatio-temporally invariant vertical profiles of gaseous and aerosol components representing Northern Hemispheric, mid-latitude, clean environmental conditions. O<sub>3</sub> mixing ratios are set to 30 ppbv from the surface to about 500 m, increasing gradually to 60 ppbv at 12 km, then increasing exponentially to about 170 ppbv at 15 km which remains constant to the model top (~20 km). CO conditions are set to 80 ppbv homogeneously up to 14 km, then decreasing exponentially to 70 ppbv at the model top. Based on the prescribed time-invariant ('static') vertical profiles, the model lateral boundary mixing ratios of chemical species are determined by constant inflow and zero-gradient outflow conditions (Skamarock et al., 2008).

The only description of the BCs that I can find for the control run is on page 14040, lines 15-16, which states “Model lateral boundary conditions applied for chemical species are constant inflow and zero gradient outflow conditions in the coarse domain”. It is not clear to me what this means. A more detailed description is required.

Further descriptions on the lateral boundary conditions were added in Sect. 3.1 (See the answer above). Skamarock et al. (2008) can be referred for numerical techniques regarding lateral boundary conditions. The reference was added in the revised manuscript for clarity.

Although (page 14040, line 22-) describes the experimental set up of the chemical IC/BCs for the sensitivity runs, it never defines this as being “dynamic” or exactly what this term means in this context. The text needs to be much clearer and consistent in the defining the type of IC/BCs used.

In order to differentiate from the control simulation using static (‘time-invariant’) chemical IC/BCs, the words “dynamic (‘time-variant’)” were used for sensitivity simulations in the title of Sect. 3.2 and in context, which might make the manuscript clearer for the use of chemical IC/BCs.

The other implication of these bits of text is that using IC/BCs (or more specifically dynamic IC/BCs) increases the simulated ozone. It does in the simulations considered, but surely this depends on the IC/BCs that might otherwise be used in the control run. Be careful not to imply more than is justified.

Authors agree with the reviewer. After reviewing the manuscript, one sentence in Sect. 5 (shown below) was revised for clarity as follows:

‘All sensitivity simulations with chemical IC/BCs increased background O<sub>3</sub> levels.’ (previous)

‘All sensitivity simulations with dynamic chemical IC/BCs increased background O<sub>3</sub> levels compared to the control simulation using prescribed static IC/BCs.’ (revised)

A brief description of how the WRF-CHEM data for the flight tracks was extracted from the model output would be useful.

The aircraft measurements with high spatial and temporal resolutions were projected on the model grid, and then median values of the observations at each model grid point were selected and compared to the simulated values. This sentence was added in Sect. 2 in the revised manuscript.

#### Technical Corrections

Page 14044 , line 8, it would be useful to state in brackets after the MBE values quoted which day they each relate to.

Specific date was added in each MBE value in the revised manuscript.

Page 14044 , line 14, “at” the two urban stations

It was corrected in the revised manuscript.

Page 14055, line 9-16, discusses Figure 14. Although comparing panels a and b of the figure demonstrates that the model captures the general chemical aging behaviour observed, I would be interested to see correlation plots between the model and observed data of say  $\text{NO}_x/\text{NO}_y$  to assess this more quantitatively.

Figure A1 shows comparison of the observed and simulated  $\text{NO}_x/\text{NO}_y$  ratios for two days. The model well reproduces the chemical aging behavior, showing a high correlation coefficient ( $R=0.94$ ). This is identical as explained in Fig. 14. This explanation was added in the paragraph in Sect. 4.3.

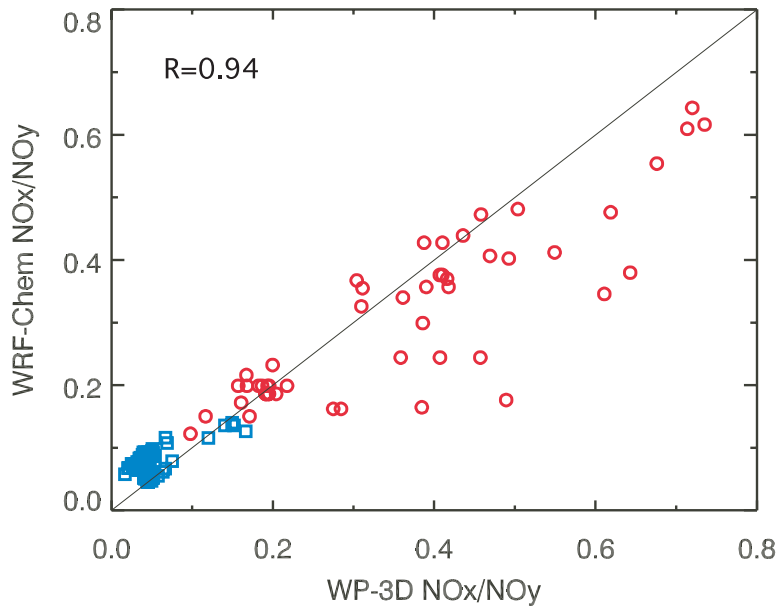


Fig A1. Comparison of the observed and simulated  $\text{NO}_x/\text{NO}_y$  for 20 July (red circle) and 21 July (blue square). The correlation coefficient between the modeled and observed  $\text{NO}_x/\text{NO}_y$  is given.

The authors are grateful to the reviewer's valuable comments that improved the manuscript.

## Anonymous Referee #2

The authors presented a nice analysis to show the evolution of urban plumes from NYC using airborne measurements and model results. They did solid work to demonstrate the importance of the right IC/BCs in regional air quality simulation, which is one major aspect of the paper. Information on model simulations needs to be provided more clearly to avoid confusion.

Specifically:

1. Which simulations were used for Section 4.2? This sort of information should be provided explicitly up front.

The analyses on simulated meteorology and chemical plumes, shown in Sect 4.1-4.3, are based on the nested domain (9 km resolution) of the control simulation. This sentence was included in Sect. 3.1 for clarity.

2. How were the background levels of O<sub>3</sub> and CO defined? Page 16, line 4: the simulated CO mixing ratios averaged along the flight track were lower than the observed by ~40 ppbv, and the authors stated that it indicated a lower "background level" in the simulated mixing ratio. It could be many things that contributed to this model underprediction, among which lower emissions of CO could be one. Why is it necessarily due to the underpredicted background level only? And what is this "background" level? A clear definition is needed.

In this study, the terminology 'background levels' was used as baseline mixing ratios that are discriminated from those of the strong urban plumes we are focusing. Even though its definition is rather qualitative than quantitative, we think that it can be clearly understood within context as is the case in many research articles.

In terms of the sentence in page 16 line 4, it was rewritten in the revised manuscript without the use of 'background level' for clarity as follows.

'The simulated CO mixing ratios along the flight track are generally lower than the observations by about 40 ppbv on average (Fig. 4b). The under-predictions of simulated CO may be caused by several factors such as uncertainties in both local and remote emissions, errors in their transport to the area of interest, and other physical and chemical processes.'

Thanks for the comment.

3. Page 16, lines 6-9: again, which simulations were being analyzed here? After one finishes reading the whole paper, it became pretty obvious that the controlled run was used in this section. If information like this was clearly given up front, it spares readers from guessing.

That point was explicitly described in Sect. 3.1 as stated in the answer of comment 1 above.