

Review of "Evaluation of Anthropogenic Emissions and Ozone Pollution in the North China Plain: Insights from the Air Chemistry Research in Asia (ARIAs) Campaign" by Hao He et al.

MS Number: acp-2019-248

Summary:

This paper discusses aircraft and surface observations of important air pollutant species over China. Such measurements are critically important for understanding the poor air quality in China, which adversely affects the health of 100's of millions of people. However, this paper does not adequately describe the measurements, and the results are poorly discussed, in some cases reaching contradictory conclusions. In at least one instance, the results are compromised by a fundamental math error. The results of the photochemical grid modeling appear to be extremely poor. I recommend that the paper be rejected, and the authors encouraged to resubmit the paper, if the issues detailed below can be adequately addressed.

Major issues and comments:

- 1) Much more detailed information regarding the aircraft measurements must be given, perhaps in the Supplementary Material. The single paragraph in Section 2.1 is inadequate. Questions that immediately occurred to me include: Were the instruments zeroed and calibrated in flight? How can ozone be measured at 1 Hz frequency when the specifications of the ozone instrument state that the response time is 20 seconds with a 10 second lag time? How can formaldehyde be measured at 1 Hz frequency when the specifications of that instrument state that the time resolution is 90 sec with a delay time ~300 sec. How were the lag times of the various instruments synchronized with the GPS system? What was the aircraft air speed (i.e., 1 Hz measurement frequency corresponds to what spatial resolution)? What was the rate of ascent and descent (this is important given the evident time resolution of some of the instruments)? Given the disparate time resolution of the instruments, how were comparable spatial average concentrations calculated? What are the accuracies and precisions of the 1 Hz measurements? (Please provide a table listing these instruments parameters and explanations or references of how these parameters were determined.) Can any evidence be provided to demonstrate these tabulated accuracy and precision parameters are realistic (e.g., the results of in-flight instrument comparisons with another aircraft)? A full description of these and all such instrument issues must be provided for the interested reader.
- 2) Lines 264-276: This paragraph suggests that underestimation of ozone precursors in CMAQ could lead to the poor model performance. Poor simulation of the boundary layer depth also could lead to the poor model performance; a balanced discussion of both of these issues should be given.
- 3) Figure S2d seems to show a comparison of measured versus modeled NO and NO_y, yet the measurement of these two species is not described in Section 2.1. What is going on here? Similarly Figure 4c gives NO measurements. If NO_y was measured, why is its model-measurement comparison not given in Figure 4? Clearly the description of the aircraft measurements must be improved as noted in Comment 1 above.
- 4) Figure 4 compares 10-minute averages of measured concentrations with model results. A discussion of this averaging must be given, as the aircraft covers a significant distance (~30 km?) and can cover a significant altitude range in 10 minutes. Is this really a reasonable comparison?

- 5) Lines 264-276: This paragraph is confused, contradictory and inaccurate. Figure 4 shows that NO is neither under- nor over-estimated by CMAQ. Figure S2 indicates that CMAQ under-estimates (not over-estimates) NOy. This description requires rethinking and rewriting.
- 6) It is not possible for me to assess the validity of the analysis in Section 3.2 because 1) only a very brief discussion of the approach and results are given, 2) the reference for the method (Halliday et al., 2018) is “to be submitted” and is thus not available, and 3) no illustrations of the EF calculations are shown in the paper or in the Supplementary Material. The description of this analysis must be improved.
- 7) Lines 307-328: In these paragraphs the authors apparently make an elementary math error. They appear to be comparing the average of many, independently determined emission ratios (EFs) based on measurements with the ratio of the total EDGAR inventory emissions. Such a comparison is not valid because in general the arithmetic average of a distribution of ratios is not equal to the ratio of the means of the numerators and denominators of the ratios. The underlying numbers must be corrected and the discussion modified accordingly.
- 8) Section 3.3 compares satellite measurements with CMAQ model results for NO₂, CH₂O and CO. The discussion proceeds with no considerations of systematic uncertainties in either the satellite measurements or the model results. The authors note that over the aircraft campaign area CMAQ predicts 81% of NO₂ satellite column measurement and has good agreement with CH₂O (<20% underestimation). To me this seems to be excellent agreement, and that it is not legitimate to interpret <20% differences as indications of emission uncertainties. This section is not acceptable without robust quantitative discussion of the systematic uncertainties in both the satellite measurements and the model results.
- 9) Sections 3.1 and 3.3 seem to reach inconsistent conclusions. Section 3.1 suggests that CMAQ generally underestimates observed concentrations of major air pollutants, often by large factors (Line 273: factors of 2 to 4 for all air pollutants; Line 287: a factor of 5 for VOCs). Yet as noted in the previous comment, Section 3.3 finds agreement within 20%. Section 3.4 goes back to the idea that the CMAQ run substantially underestimates the concentrations of ozone and its major precursors in the NCP. Such inconsistencies must be fully and quantitatively addressed before emissions within the CMAQ modeling can be objectively adjusted.
- 10) Section 3.4 is not satisfactory. Figures 10 and S3 present time series of observations and model results, but the agreement is quite poor regardless of the model run. These comparisons should be based on an objective measure of overall model performance so that the reader can appreciate how well or poorly each of the model simulations actually reproduced the observations.
- 11) Figure 11 compares observations and model result for a selected flight segment. The agreement appears to be extremely poor. Again, these comparisons should be based on an objective measure of overall model performance (for all flights) so that the reader can appreciate how well or poorly the model simulation actually reproduced the observations.
- 12) Much of the Conclusions and Discussion section is speculative and/or not quantitatively supported by the results discussed previously.

Minor issues:

- 1) Line 122: The country of the Environmental Protection Agency (EPA) should be indicated.

China (where the research is located) has an EPA, but I assume that this sentence refers to the U.S. EPA.

- 2) Figure 1 should be improved. It is not possible for a reader unfamiliar with Chinese geography to easily understand the region of China actually covered by the flight tracks.
- 3) Lines 245-248: The statement on these lines is not accurate. The description is not of the generally observed concentrations, but rather reflects the maximum concentrations observed.
- 4) The Sections are not properly numbered; two are labeled Section 3.3.