

Reviewer 3:

This paper used an Extended Response Surface Modeling (ERSM) technique to assess the source contributions of various chemical precursors, emission sectors, source regions, and their combinations to the PM_{2.5} concentrations over the BTH area. It extended the previous conventional RSM model and pursued more than 1000 simulation scenarios. It is informative and valuable to the air pollution controls over the heavily polluted BTH area. I would suggest this paper to be published after minor revision.

Response: We appreciate the reviewer's valuable comments which help us improve the quality of the manuscript. We have carefully revised the manuscript according to the reviewers' comments. Point-to-point responses are given below. The original comments are in black, while our responses are in blue.

(1) In the abstract, page 2, line 6, "primary inorganic PM_{2.5} is the single pollutant which makes the largest contribution (24-36%) to PM_{2.5} concentrations." What is the exact mean of the word "single"?

Response: We thank the reviewer for this valuable comment. In the context of this sentence, a "single" pollutant means one of the six pollutants (or pollutant groups) considered in the RSM/ERSM prediction systems, i.e., NO_x, SO₂, NH₃, NMVOC+IVOC, POA, and primary inorganic PM_{2.5}. Primary inorganic PM_{2.5} is defined as the chemical components of primary PM_{2.5} other than POA. To avoid confusion, we have revised the preceding sentence as follows in the revised manuscript (Page 2, Line 5-7).

Among all air pollutants, primary inorganic PM_{2.5} makes the largest contribution (24-36%) to PM_{2.5} concentrations.

(2) In the Table S4, "Statistical results for the comparison of monthly PM_{2.5} concentrations", the variable calculated in the statistics is hourly PM_{2.5} concentrations, right?

Response: The original data used in the statistical analysis are daily PM_{2.5} concentrations. We have clarified this in the footnote of the revised table.

(3) In Table S4 and S5, please add the number of data pairs, especially in S5.

Response: We have added the number of data pairs used in statistics in the revised Table S4 and S5.

1 (4) I would suggest the authors add a discussion on the limitations or uncertainties of this
2 study at the end of the conclusion section.

3 Response: Following the reviewer's suggestion, we have added a paragraph about the
4 limitations and uncertainties of the present study at the end of the manuscript (Page 21, Line
5 13-27). The added paragraph is shown as follows.

6 The present study has a few limitations. First, the establishment of ERSM requires several
7 hundred or over 1000 emission scenarios, although the scenario number needed for a specific
8 number of control variables has already been dramatically reduced as compared to the
9 conventional RSM technique. Studies are needed to further reduce the scenario number but
10 retain the accuracy of the ERSM technique. Second, the current ERSM technique has not
11 considered the impact of meteorological variations on ambient concentrations. Third,
12 although the responses of PM_{2.5} concentrations to precursor emissions predicted by ERSM
13 have been demonstrated to agree well with chemical transport model simulations, evaluating
14 the predicted responses against the actual situation in the real atmosphere still represents a
15 major challenge, because it is extremely difficult to artificially perturb emissions in the
16 atmosphere. Last but not the least, the NMVOC and IVOC emissions have been lumped
17 together in this study to reduce the number of control variables. Considering their differences
18 in sources and SOA formation potentials, a detailed quantification of the individual
19 contributions of NMVOC and IVOC emissions from various sources to PM_{2.5} concentrations
20 is required in the future to better inform NMVOC/IVOC control policies.