Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-2-AC1, 2018

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Quantification of the enhanced effectiveness of NO_x control from simultaneous reductions of VOC and NH_3 for reducing air pollution in Beijing-Tianjin-Hebei region, China" *by* Jia Xing et al.

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Received and published: 15 April 2018

We thank the reviewer for the detailed and thoughtful review of our manuscript. Incorporation of the reviewer's suggestion has led to a much improved manuscript. Detailed below is our response to the issues raised by the reviewer. We also detail the specific changes incorporated in the revised manuscript in response to the reviewer's comments.

[Comment]: This paper developed a method by fitting multiple CMAQ simulations with

a set of polynomial functions to quantify responses of ambient PM2.5 and O3 concentrations to changes in precursor emissions. The performance of the model looks sound. However, I suggest the authors to include more scientific findings based on the model developed in this study.

[Response]: We thank the reviewer for recognition of the implications of the results of the analysis presented. We followed the reviewer's suggestion and include more discussion on the scientific findings based on the model developed, which is detailed in our response to the reviewer's comment on Section 3.3.

[Comment]: The reason why pollutant responses to emissions can be characterized as a series polynomial functions by the previous developed regression-based RSM has not been clarified.

[Response]: The relationship between pollutant responses to emissions can be quantified by an atmospheric chemical transport model (noted by CTM, e.g., CMAQ) which describes most of the physical and chemical processes in the atmosphere. Studies on multiple CTM simulations under different emission scenarios ("brute force method") can investigate the full range of pollutant responses to emissions. The principle of regression-based RSM model is to build up the full range of pollutant responses to emissions using an advanced statistic method (i.e., response surface method) from a number of CTM simulations. The accuracy of regression-based RSM in representing the nonlinearity in pollutant response to emissions has been examined thoroughly by different methods including cross validation, out-of-sample validation and isopleth validation in previous studies (Xing et al., 2011; Wang et al., 2011; Zhao et al., 2015; Xing et al., 2017; Zhao et al., 2017). Since the relationship between pollutant responses to emissions is followed by the basic chemical functions and physical laws parameterized in CTM (i.e., CMAQ in this study), the function used to represent the relationship in regression-based RSM (implicitly) can be parameterized explicitly in a series of basis functions. This study used a linear combination of polynomial bases (i.e., 1, x, x2, x3...) to characterize the pollutant responses to emissions, and the terms were selected carefully following the procedure described in section 2.2.

To clarify this point, we provided the following discussion in the revised manuscript.

(Page 4 Line 9) "The accuracy of regression-based RSM in representing the nonlinearity in pollutant response to emissions has been examined thoroughly by different methods including cross validation, out-of-sample validation and isopleth validation in previous studies (Xing et al., 2011; Wang et al., 2011; Zhao et al., 2015; Xing et al., 2017; Zhao et al., 2017). The relationship between pollutant responses to emissions followed by the basic chemical functions and physical laws is implicitly represented in the regression-based RSM. In this study, however, we adopted a linear combination of polynomial bases (i.e., 1, x, x2, x3. . .) to parameterize explicitly the pollutant responses to emissions."

[Comment]: The authors use too many self-defined abbreviations in the text, for instance, PR, VNr, FR, which make the paper not very reader-friendly.

[Response]: We defined a few indicators in this study, including (1) peak ratio (denoted as PR) representing VOC-limited or NOx-limited condition, (2) suggested reduction ratio of VOC to NOx (denoted as VNr) to avoid increasing O3 under VOC-limited condition, (3) flex ratio (denoted as FR) representing NH3-poor or NH3-rich condition. Because of the advantage of the pf-RSM method which is able to provide the full range of the pollutant responses to emissions, it not only can qualitatively identify the current status to certain chemical scheme, but also can quantitatively estimate the exact transition point on which the chemical scheme will be transited to the other. That is the reason why we defined and calculated the PR and FR. Actually these two indicators were first developed and already used in our previous publications (Xing et al., 2011; Wang et al., 2011).

However, we agree that too many self-defined abbreviations will reduce the readability of this manuscript. To address this concern, we deleted the abbreviation of VNr and reduced the usage of the PR and FR in the revised manuscript.

[Comment]: Page 8, line 1-5, please explain the reason why the performance of the pf-RSM with less than 40 training samples exhibited a noticeable discrepancy compared with that of the regression-based RSM, but not for those with over 40 training samples..
[Response]: The discrepancy shown in the comparison between pf-RSM with less than 40 training samples with that of regression-based RSM is due to the underfitting issue. It is mainly because the number of training samples is not large enough to capture the nonlinearity in the model system. When the number of training samples increases to over 40, the discrepancy is reduced.

To clarify this point, we added the following discussion in the revised manuscript:

(Page 8 Line 28) "Such discrepancy is caused by the underfitting issue implying the number of training samples is not large enough to capture the nonlinearity in the model system. The issue can be addressed by added more training samples to fit the model. The 40 training samples presented good agreement with the predictions of the regression-based RSM. Improving sampling method is also important for reducing the biases. We can see that additional marginal processing also improved the performance of the pf-RSM."

[Comment]: The uncertainty of the fitting results is missing.

[Response]: The uncertainty of pf-RSM is evaluated by the comparison the pf-RSM prediction against with true CTM simulation (i.e., out-of-sample validation). Five statistical indices representing the performance were calculated including the mean normalized error (MeanNE), maximal normalized error (MaxNE), mean fractional error (MeanFE), maximal fractional error (MaxFE) and correlation coefficient (R). From the comparison with the results of 115 CMAQ simulations, we found that the pf-RSM with 40 training samples can meet the criteria of MeanNE within 2% and MaxNE within 10% (which is comparable to the performance of previous regression-based RSM).

To clarify this point, we added some discussion in the revised manuscript as follows: (Page 8 Line 13) "To meet the criteria of MeanNE within 2% and MaxNE within

10% (i.e., uncertainty of pf-RSM) which is comparable to the performance of previous regression-based RSM, use of 40 training samples with marginal processing (to improve boundary conditions) is recommended."

[Comment]: Section 3.3. This section needs substantial improvement considering this is the only section discussing the application of the method. What are the new findings using the modified model, but not indicated by previous one? What is the advantage of the model compared to the existed ones? Otherwise, this paper seems to be more like a technical document, but lack scientific findings.

[Response]: In this manuscript, we proposed a new method (i.e., pf-RSM) to quantify the pollution response to emissions. Compared to existed methods, the newly developed pf-RSM has two advantages: 1) explicitly represent the response, make it easy to investigate the nonlinearity (e.g., peak value, derivative) of the predicted system, as the indicator (PR, FR) we defined in this study; 2) substantially reduce the computational burden by more than 60%, enable the usage on studies with high spatial and temporal resolution.

In this studies, we adopted the pf-RSM model to investigate the enhanced effectiveness of NOx control from simultaneous reductions of VOC and NH3 for reducing O3 and PM2.5. With the pf-RSM, the enhanced effectiveness was quantified. Strong VOC-limited condition in urban areas in BTH has already been recognized in previous studies, due to the abundance of NOx emissions. However, questions that how many current NOx emissions are overabundant and how many VOC emissions are suggested to simultaneously reduce with NOx were not well addressed. With the newly developed pf-RSM in this study, we can provide a quantitate answer. Our results suggest that the NOx emission reduction rate need be greater than 20%-60% (depends on the location in BTH) to pass the transition from VOC-limited to NOx-limited, and a simultaneous VOC control (the ratio of VOC reduction to NOx reduction is about 0.5-1.2) can avoid increasing O3 during the transition. Similarly, the benefit of NH3 control for PM2.5 reduction is well documented. In this study, we quantified the enhanced benefits in

PM2.5 reductions from simultaneous reduction of NH3 to be 0.04-0.15 μg m-3 PM2.5 per 1% reduction of NH3 along with NOx, with greater benefits in July when the NH3-rich condition is not as strong as in January. Besides, we found the response varies significantly over space and time. All the results are derived from the pf-RSM model. Since the pf-RSM model is more efficient compared to previous method, the potential usage of pf-RSM includes cost-benefit optimization and integrated assessment. We are developing an air pollution control cost-benefit and attainment assessment system (ABaCAS, Xing et al., 2017), and pf-RSM will be one of the core module in the whole system. The comparison between our results and other studies was also added into this section.

Following the reviewer's suggestion, we revised the section 3.3 and emphasize the scientific findings as follows:

(Page 9 Line 34) "The nonlinearity in the pollution response to emissions leads to an either enhanced or reduced effectiveness of emission controls. In previous studies, the concept of NH3-limited/-poor and NOx-/VOC-limited conditions was used widely to demonstrate the influence of NH3 and VOC on effectiveness of NOx controls for reducing PM2.5 and O3, respectively. However, some key questions were not well addressed, such as how much percentage of NOx or NH3 is overabundant and how much percentage of VOC need reduced simultaneously to avoid increased O3. In this study, the newly developed pf-RSM explicitly represents the response and the enhanced effectiveness can be easily quantified. As the indicators defined in Section 2.3 can be used to quantify the nonlinear effectiveness of emission control for reducing PM2.5 and O3."

(Page 10 Line 11) "The result is consistent with our previous study (Wang et al., 2011) which reported that NH3 is sufficiently abundant to neutralize extra nitric acid produced by an additional 25% of NOx emissions in north China Plain based on a traditional regression-based RSM study."

(Page 10 Line 17) "That is consistent with the findings of previous studies (Xing et al., 2011) which used a traditional regression-based RSM and found that the PR changes from 0.8 to 1.2 as the distance from the city center increases."

(Page 10 Line 20) "Our results are consistent with the observational studies that use indicator to identify the O3 chemistry. For example, Liu et al (2016) studied on the ratios of HCHO over NO2 from the satellite retrieves and found that local ozone production in urban Beijing is VOC-limited when there are no substantial changes in NOx emission in 2015. Chou et al. (2009) found that Beijing urban area was "VOC-limited" region based on the observation of NO, NOx and NOy at the Peking University site during August 15 to September 11 in 2006. Jin and Holloway (2015) calculated the ratio of HCHO to NO2 from the OMI instrument aboard the Aura satellite and found the O3 production is more likely to be VOC-limited over urban areas and NOx-limited over rural and remote areas in China from 2005 to 2013."

(Page 11 Line 11) "After the application of a prior knowledge of the pollutant responsiveness to emissions in the RSM system, the cases required for single regional pf-RSM development were substantially decreased to 40 samples, compared with the previous requirement of over 100 samples, imply that the fitting-based RSM (i.e., pf-RSM) is three time faster than previous regression-based RSM (i.e., the number of CTM simulations needed in pf-RSM is 60% less than that required by previous regression-based RSM). The pf-RSM system in this study operates rapidly, and thus can quickly generate responses with high spatial and temporal resolutions, thereby further facilitating cost-benefit optimization and enabling further assessment studies to be conducted (e.g., air pollution control cost-benefit and attainment assessment ABaCAS system described by Xing et al., 2017)."

Reference

Xing, J., Wang, S., Jang, C., Zhu, Y., Zhao, B., Ding, D., Wang, J., Zhao, L., Xie, H., Hao, J.: ABaCAS: an overview of the air pollution control cost-benefit and attainment

assessment system and its application in China. The Magazine for Environmental Managers - Air & Waste Management Association, April, 2017.

[Comment]: The conclusion in the abstract, like "Thus, simultaneously reducing NH3 and VOC emission along with NOx reduction is recommended to assure the control effectiveness of PM2.5 and O3", is too general.

[Response]: To emphasize the scientific findings in this study, we revised this sentence as follows:

(Page 1 Line 34) "Thus, the newly developed pf-RSM model has successfully quantified the enhanced effectiveness of NOx control, and simultaneous reduction of VOC and NH3 with NOx can assure the control effectiveness of PM2.5 and O3."

[Comment]: Page 2, line 5, the grammar of "significantly influences on" is not proper.

[Response]: We fixed the typo, and revised it as "significantly influences" (Page 2 Line 5)

[Comment]: Page 3, line 31- 35, the sentence is too long to read. Please consider rephrasing it.

[Response]: We have reduced the length of the sentence in the revised manuscript, as following

(Page 3 Line 35) "In general, tropospheric O3 and PM2.5 concentrations are contributed by its sources and sinks through a series of atmospheric processes, such as horizontal or vertical advection and diffusion, gas phase chemistry, and deposition. The nonlinear behavior in each of these processes contributes to the nonlinearity in the responses of concentrations to precursor emissions. Similar responsive functions can be expected across regions and time. For example, a universal ozone isopleth diagrams developed using the empirical kinetic modeling approach of the U.S. Environmental Protection Agency (Gipson et al., 1981) represents the general O3 responsiveness to NO, and VOC concentrations."

[Comment]: Page 8, line 9-10. The description about how to design scenarios is missing. For instance, why the moderate control is defined as ENOx, ESO2, ENH3, EVOCs and EPOA = -49%, -45%, -20%, -64%, and -20%? Will the validation results change if you change the definition of scenarios?

[Response]: The scenarios were designed from a 100 Latin Hypercube Sampling method. The two scenarios were selected randomly from the 100 samples, for the purpose of analyzing different location and time. The validation on averages (time and location) are conducted for all 100 samples as we discussed in section 3.1. Here we just pick up two scenarios to represent two different control levels, moderate and strict. The validation results might slight change if we change the scenarios, however, the performance should be similar to the two we presented here.

To clarify this point, we added some discussion in the revised manuscript as follows:

(Page 8 Line 37) "These two scenarios are selected from the OOS100, to represent two kinds of emission levels, moderate and strict respectively, for the purpose of analyzing the pf-RSM performance under different locations and times. Please note that the validation results might slight change if we change the scenarios, however, the performance should be similar to the two we presented here."

[Comment]: Page 9, line 19-20. The sentence is confusing. Please try to rephrase it.?

[Response]: As the reviewer suggested, we revised the sentence as follows:

(Page 10 Line 27) "The PR values calculated in this study also indicate that the control of NOx (with less than 20%-60% reduction, =1-PR) could result in an increase of O3; however, O3 would decrease with substantial control of NOx (with greater than 20%-60% reduction)."

[Comment]: Figure 5. The font size of legend does not fit the graph.

[Response]: Following the reviewer's suggestion, we reduced the font size of legend in the Figure 5 in the revised manuscript.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-2/acp-2018-2-AC1-supplement.pdf

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-2, 2018.