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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Development and application of observable response indicators for design of an effective ozone and fine particle pollution control strategy in China" *by* J. Xing et al.

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[Comment]: The manuscript by Xing et al. on development and application of observable response indicators uses response surface modeling to identify parameters that define key O3 and PM2.5 production regimes, and then correlates these with observable indicators, i.e. ratios of gas and aerosol phase concentrations that are routinely measured. This provides valuable information that could be used to help design effective air quality policy to simultaneously reduce levels of both O3 and PM2.5 which, as the authors point out, has been a challenge in China. The work is thus very relevant

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Discussion paper

and suitable in scope for ACP. The paper is also very clearly written for the most part.

[Response]: We thank the reviewer for recognition of the implications of the results of the analysis presented, and overall positive comments.

[Comment]: My main comments are summarized as follows. The study currently neglect any errors in the polynomial approximations of the full CTM at later stages in the analysis, which I think is an oversight.

[Response]: We agree with the reviewer on the importance of considering the error of polynomial approximations of the CTM. In the development of pf-RSM, we have examined the performance of pf-RSM to ensure its accuracy to meet the criteria of a mean normalized error within 2% and a maximal normalized error within 10%. The large errors are mostly located in the marginal areas where the emissions were reduced to nearly zero and the concentrations will be very small. Thus, the errors in the pf-RSM predictions have limited influence on the shape of nonlinear curve of the response function. To address the reviewer's concern, we have added following description about the error of pf-RSM in the revised manuscript.

(Page 8, Line 164-171) "The pf-RSM performance in predicting PM2.5 and O3 responses has been evaluated in detail using leave-one-out cross validation as well as the out-of-sample validation method, with normalized errors all within 5% for both PM2.5 and O3 across the domain. Relatively large biases occurred for marginal cases, where emissions are controlled by nearly 100% and predicted concentrations are very small. These cases have limited influence on the shape of nonlinear curve of the response function. However, the RSM is developed from a suite of CMAQ simulations, and so uncertainties in the chemical mechanism used in CMAQ might influence the O3 and PM2.5 predictions."

[Comment]: Further, it is not mentioned explicitly that responses being analyzed here are with respect to domain wide emissions perturbations (I suspect, as it isn't explained clearly). This limits the applicability of these responses for evaluation of regional air

quality control strategies, as there would be errors in using these relationships to estimate a response to a regional change in emissions.

[Response]: We agree with the reviewer that the response of PM2.5 and O3 to different regional sources varies significantly. As suggested in our previous study (Xing et al., 2011), the local NOx controls can be either beneficial or unbeneficial in reducing O3, while regional NOx controls usually exhibit benefits in reducing O3. The overall effects are determined by the combination of the selected local/regional control ratios. In this study, the same level of emission perturbations was applied across the country. That is because controls are more likely taken in multiple regions of China rather than only on one single region. In addition, the same level of local and regional reduction is suggested to achieve aggressive air quality goals as demonstrated in our previous study (Xing et al., 2019).

To clarify this point, we have provided additional discussion in the revised manuscript, as follows.

(Page 7, Line 156-161) "Though the responses of O3 and PM2.5 to local or regional emissions vary significantly as suggested in our previous study (Xing et al., 2011), we applied the same change ratio of each pollutant emission to all regions across China. This approach is consistent with the implementation of a multi-regional joint control strategy, which is reasonable for China. The same level of local and regional emission reduction has been recommended to achieve China's aggressive air quality goals (Xing et al., 2019)."

Reference:

Xing, J., Wang, S. X., Jang, C., Zhu, Y., and Hao, J. M.: Nonlinear response of ozone to precursor emission changes in China: a modeling study using response surface methodology, Atmospheric Chemistry and Physics, 11, 5027-5044, 10.5194/acp-11-5027-2011, 2011.

Xing, J., Zhang, F., Zhou, Y., Wang, S., Ding, D., Jang, C., Zhu, Y. and Hao, J.: Least-cost control strategy optimization for air quality attainment of Beijing–Tianjin–Hebei region in China. Journal of environmental management, 245, 95-104, 2019.

[Comment]: Lastly, here are a few definitions / concepts that would be useful for the authors to define upfront (definitions of indicators that general audiences may not be familiar with).

[Response]: As the reviewer suggested, we have defined the indicators including DSN, GR, and AdjGR, and provided references for PR and FR in the revised manuscript.

[Comment]: Overall, the methods and results are interesting and have merit; all of these issues could be addressed with revisions to the text and some additional work on error analysis.

[Response]: We have followed the reviewer's suggestion and made modification correspondingly in the text. Hope the revised manuscript can meet the high standard for ACP journal.

[Comment]: 69: Please define DSN, GR, and AdjGR. Eventually I see later (line 233) that these are defined in the SI, but it would be more useful if they were defined earlier, or at least reference to where their definition can be found provided earlier.

[Response]: As the reviewer suggested, we provide the definition of three indicators at the first time in the text, as follows.

(Page 4 Line 72-75) "Regarding PM2.5 chemistry (more specifically for inorganic PM2.5 sensitivities to NH3 and NOx), indicators such as the degree of sulfate neutralization (DSN), gas ratio (GR), and adjusted gas ratio (AdjGR) have been developed (defined in Text S1) to identify NH3-poor or -rich conditions (Ansari and Pandis, 1998; Takahama et al., 2004; Pinder et al., 2008; Dennis et al., 2008)."

[Comment]: 71: Clarify that by "these" you are referring to indicators for O3. I don't believe this has been done for the SIA indicators such as AdjGR since total nitrate isn't

routinely observable from space.

[Response]: We agree with the reviewer that “these” is referring to O3 indicator. As the reviewer suggested, we have clarified it in the revised manuscript and moved this sentence ahead of the PM chemistry indicator, as follows.

(Page 4 Line 68-72) “The O3 indicators can be derived from surface-monitoring observations (Peng et al., 2006), modeling simulations (Wang et al., 2010), or even satellite retrievals (Jin et al., 2017; Sun et al., 2018), and then examined using three-dimensional chemical transport models (CTMs) (Jiménez et al., 2004; Zhang et al., 2009; Liu et al., 2010; Ye et al., 2016).”

[Comment]: 85: Please define PR and FR.

[Response]: The PR and FR have been defined in our previous study (Xing et al., 2018). As the reviewer suggested, we provide the reference to clarify the definition of PR and FR in the revised manuscript, as follows.

(Page 4 Line 85-87) “Based on the RSM, the chemical response indicators of Peak Ratio (PR) and Flex Ratio (FR) have been designed to identify regimes of O3 and PM2.5 chemistry, respectively (see Xing et al., 2018 for detailed description of PR and FR).”

Reference:

Xing, J., Ding, D., Wang, S., Zhao, B., Jang, C., Wu, W., Zhang, F., Zhu, Y., and Hao, J. Quantification of the enhanced effectiveness of NOx control from simultaneous reductions of VOC and NH3 for reducing air pollution in the Beijing–Tianjin–Hebei region, China, Atmos. Chem. Phys., 18, 7799-7814, https://doi.org/10.5194/acp-18-7799-2018, 2018.

[Comment]: 101: What is meant be severe here? Are the goals to address severe episodes in the winter or address longer-term annual averages? As the chemical mechanisms driving the former are not well know, yet, my guess is the focus of this

article is on the latter, which should be clarified.

[Response]: We agree with the reviewer that our target focuses on reducing long-term annual averages. To clarify this point, we have changed the “severe pollution” to “air pollution” in the revised manuscript, as follows.

(Page 5 Line 101-103) “Notably, accurate quantification of the nonlinear responses of O3 and PM2.5 to their precursor emissions is critical and a prerequisite for effective mitigation of air pollution in China.”

[Comment]: 143 - 146: The cited works here are not published yet, so please provide a brief summary of the performance benchmarks and statistics.

[Response]: We have updated the two cited papers which have been recently published or sent for publication. As the reviewer suggested, we also summarized the performance statistics in the revised manuscript as follows.

(Page 7 Line 148-152) “The normalized mean biases of CMAQ in predicting PM2.5 and O3 are -16.4% and -12.5% compared with monitoring data obtained from the China National Environmental Monitoring Centre. The mean fractional biases for PM2.5 and O3 prediction are -14.2% and -11.1%, respectively (within the benchmark of $\pm$60%). The mean fractional errors for PM2.5 and O3 prediction are 21.6% and 17.0% respectively (within the benchmark of 75%).”

Reference:

Ding, D., Xing, J., Wang, S., Liu, K. and Hao, J.: Estimated Contributions of Emissions Controls, Meteorological Factors, Population Growth, and Changes in Baseline Mortality to Reductions in Ambient PM 2.5 and PM 2.5-Related Mortality in China, 2013–2017. Environmental health perspectives, 127(6), 067009, 2019a.

Ding, D., Xing, J., Wang, S., Chang, X. and Hao, J.: Impacts of emissions and meteorological changes on China’s ozone pollution in the warm seasons of 2013 and 2017, Front. Environ. Sci. Eng. 2019, 13(5): 76, 2019b.

[Comment]: 155 - 263: I have questions about the spatial dimension of the terms in these equations. The manuscripts says that Xi was fit for every grid cell. Does that mean that in each grid cell it was known from the CTM simulations how Conc responded to each of the precursor emission species perturbed specifically in that grid cell? Or is it how Conc response to emissions perturbed uniformly throughout the entire model domain? If the former, that seems like a prohibitively large number of model runs (number of grid cells x 40). In this case then the response is the national average response? If the latter, it seems like the applicability of these equations for policy application is hindered by transport, in that it is now known if the change in concentration is occurring owing to changes in emissions in that location or emissions several hundred km upwind. In essence, a map of the response is not equivalent to a map of where the emissions changes need to be to elicit that response, hence this precludes using this information for region-specific changes to precursor emissions. Unless there are policies that aim to uniformly reduce emissions (from all sectors) the same amount throughout the country, it is hard to envision the direct applicability of these relationships for policy. Thus I'm not sure of the value of the province-specific values like those shown in Fig 11– a PR in a particular province isn't necessarily associated with changes to emissions in that province alone.

[Response]: We thank for the reviewer for raising a critical issue about the spatial match of responding grid cell and controlling grid cell. As the reviewer mentioned, it requires a large number of model runs to identify the controls for each grid cell, which is impossible. Thus, in this study, we applied the same level of emission perturbations to all grid cells across the country. The Xi was still fit for every grid cell, while the control factors represent the emission controls for the whole country, instead of the individual grid cell or region. We agree with the reviewer that the response of PM2.5 and O3 to different regional sources varies significantly. As found in our previous study (Xing et al., 2011), the local NOx controls can have either benefit or dis-benefit in reducing O3, while regional NOx controls usually exhibit benefit in reducing O3. The overall effects are determined by the combination of the selected local/regional control

ratios. However, in China, multi-regional joint controls are more likely conducted rather than only controls on a single region. Besides, our previous study also recommended to apply the same local to regional control level for all regions to achieve air quality attainment with the maximal cost-benefit optimization (Xing et al., 2019). The province-specific values shown in Fig 11 provide an estimate of nonlinear response under the uniform-control case, which suggests the additional action needed for each province to avoid potential risk even after considering the multi-regional controls.

To clarify this point, we have added more discussion in the revised manuscript, as follows.

(Page 7, Line 156-161) "Though the responses of O3 and PM2.5 to local or regional emissions vary significantly as suggested in our previous study (Xing et al., 2011), we applied the same change ratio of each pollutant emission to all regions across China in this study. This approach is consistent with the implementation of a multi-regional joint control strategy, which is reasonable for China. The same level of local and regional emission reduction has been recommended to achieve China's aggressive air quality goals (Xing et al., 2019)."

(Page 22, Line 476-479) "Since the indicators are developed from simulations with spatially uniform emission controls across the country, they are especially useful for providing quick estimates of the potential benefits or risks from uniform controls. These estimates can also provide a basis to design more localized control strategies for particular regions."

Reference:

Xing, J., Wang, S. X., Jang, C., Zhu, Y., and Hao, J. M.: Nonlinear response of ozone to precursor emission changes in China: a modeling study using response surface methodology, Atmospheric Chemistry and Physics, 11, 5027-5044, 10.5194/acp-11-5027-2011, 2011.

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Xing, J., Zhang, F., Zhou, Y., Wang, S., Ding, D., Jang, C., Zhu, Y. and Hao, J.: Least-cost control strategy optimization for air quality attainment of Beijing–Tianjin–Hebei region in China. Journal of environmental management, 245, 95-104, 2019.

[Comment]: Fig 3: Please include units. Also define the domain over which the emissions perturbations are being considered here.

[Response]: As the reviewer suggested, we have included units ("μg m-3" for PM2.5 and "ppb" for O3), and clarified the emissions perturbations in the caption of Figure 3, as follows.

"Isopleth of population-weighted PM2.5 and daytime O3 to precursor emission change in different months. (The x- and y- axes represent precursor emission rates with a baseline of 1, applied to all grid cells in China; background colors represent the population-weighted PM2.5 and daytime O3 concentrations in China, with units of μg m-3 for PM2.5 and ppb for O3)"

[Comment]: 245: I understand why 0 is a lower limit, but why is 2 an upper limit? This seems to cut off a lot of points in April (Fig 5).

[Response]: We agree with the reviewer that more points will be available for regression if we set the upper limit to be larger than 2. In this study, we set the range of emission changes as 0 to 2 to be consistent with our previous studies in which the pf-RSM performance has been well examined. Also, enlarging the upper limit will increase the sampling space, which might also increase the number of cases used to fit the pf-RSM.

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

(Page 8 Line 162-164) "The control matrix is provided in Table S2. The range of emission changes is set as 0 to 2 to be consistent with our previous studies in which the pf-RSM performance has been well examined (Xing et al., 2011; Wang et al., 2011; Xing et al., 2018; Ding et al., 2019b)."

[Comment]: General: If a metric like FR and AdjGR don't agree, the authors are placing the blame entirely on the observable indicator e.g. AdjGR. However, there is some degree of inaccuracy in FR, related to the extent to which the pf-RSM explains the concentration responses. The authors should thus begin the results section with a summary of the accuracy of Eq 1, particularly in terms of discussing the residuals of this functional fit and their magnitudes, showing plots of the change in concentration predicted by FR or PF vs the actual change in concentrations.

Next, the magnitude of these residuals should be taken account when considering figures like 4 and 5. I suspect that the distinction of the 4 quadrants in each panel of Figs 4 and 5 directly along the axis is too strict. Rather, corresponding to the magnitude of the residual error in (1), the comparison for Figs 4 and 5 should be to identify points that lie some distance away from the quadrant boundaries, as points near the boundaries could be impacted by the error FR or PF.

Further, it's not clear in the writeup if the change in concentration in Eq 1 is that from the RSM or the CTM – this should be clarified. If the former, then there's an additional source of error that needs to be stated and accounted for, which is the RSM itself.

Lastly, these sources of error should be kept in mind in the presentation of all of the results comparing observable indicator responses vs RMS responses, e.g., discussion of Figs 7, 9, 10, 12

[Response]: We agree with the reviewer that the disagreement between FR and AdjGR can be influenced from uncertainties in both indicators, and it is importance to consider the error of polynomial approximations of the CTM. In the development of pf-RSM, we have examined the performance of pf-RSM to ensure its accuracy to meet the criteria of a mean normalized error within 2% and a maximal normalized error within 10%, comparing against with CMAQ. As we studied the pattern of the residuals (errors), large errors are mostly located in the marginal areas where the emissions were reduced to nearly zero and the concentration will be very small. Thus, the errors in pf-RSM has

limited impacts on the shape of nonlinear curve of the response function. However, the uncertainties in the chemical mechanism of CMAQ will also contribute to the bias of O3/PM-chemistry determination. As the reviewer suggested, we have clarified the potential errors existed in CMAQ/RSM, and focused on our discussion on the observable indicators, in the revised manuscript, as follows.

(Page 8 Line 164-171) "The pf-RSM performance in predicting PM2.5 and O3 responses has been evaluated in detail using leave-one-out cross validation as well as the out-of-sample validation method, with normalized errors all within 5% for both PM2.5 and O3 across the domain. Relatively large biases occurred for marginal cases, where emissions are controlled by nearly 100% and predicted concentrations are very small. These cases have limited influence on the shape of nonlinear curve of the response function. However, the RSM is developed from a suite of CMAQ simulations, and so uncertainties in the chemical mechanism used in CMAQ might influence the O3 and PM2.5 predictions."

(Page 22 Line 493-497) "We note that the discrepency between the observable indicator and the responsive indicator might also be influcenced by uncertainties in the chemical mechanism of CMAQ as well as prediction errors of the pf-RSM. The new indicators were designed based on the existing chemical mechanism, and the transition values might be refined in the future as our understanding of atmospheric chemical processes improves."

[Comment]: 201: As defined as the ratio of VOCs to NOx, it seems rather circuitous to derive this equation only to show that it reduces to the ratio of the coefficients for the linear VOC and NOx terms (i.e. x5/x6)

[Response]: The ratio of VOCs to NOx (VNr) is designed as the level of simultaneous control of VOCs to prevent an increase in O3 levels from the NOx controls when PR<1 (VOC-limited). Thus we defined the VNr equals the ΔEvoc/ΔEnox which makes first derivative of the ΔConcO3 to ΔEnox equal 0. Considering the NOx controls will be

taken from baseline and ΔEnox is close to 0, we ignore the terms of ΔEnox in the first derivative function above, then the VNr reduces to the ratio of the coefficients for the linear VOC and NOx terms.

We agree with the reviewer that the original description about VNr calculation is a bit ambiguous. We have clarified it in the revised manuscript (see Page 10 Line 219-231).

[Comment]: 228: It is interesting that this change reduces down to just the linear response coefficient of PM2.5 with respect to NH3. This makes me want to see an additional plot in Fig 3 which is NH3 vs NOx.

[Response]: As the reviewer suggested, we have provided the PM2.5 response to NH3 and NOx in the Fig 3

[Comment]: 275: Why is there a seasonal dependence to the performance of HCHO/NO2, particularly with such low performance in April?

[Response]: The seasonality of performance of the observable indicators (such as HCHO/NO2) in predicting O3 chemistry might be associated with the uncertainty of the transition values, as different transition values were reported by different studies or for different location and time (Zhang et al., 2009). In this study, we found that the performance of the HCHO/NO2 can be substantially improved by using the transition value of 0.5 instead of 1, as shown in Table 2. Such result also implies that those indicators by using concentrations of just two species cannot fully consider all factors that determine the O3 chemistry.

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

(Page 14 Line 301-305) “However, the performance of HCHO/NOy and HCHO/NO2 could be greatly improved by using lower transition values, with increased annual success rates as high as 76 %. The change of the transition values implies that such indicators cannot fully consider all factors that determine the O3 chemistry by using

concentrations of just two species."

Reference:

Zhang, Y., Wen, X. Y., Wang, K., Vijayaraghavan, K., and Jacobson, M. Z.: Probing into regional O-3 and particulate matter pollution in the United States: 2. An examination of formation mechanisms through a process analysis technique and sensitivity study, Journal of Geophysical Research-Atmospheres, 114, 31, 10.1029/2009jd011900, 2009.

[Comment]: 384: Could the authors comment on the practicality of this application? I'm having a hard time imagining simultaneous equal %-based reductions to China-wide NH3 and NOx emissions resulting from any real policy, given that these would be coming largely from different sectors, in different locations.

[Response]: We understand the reviewer's concern that the control strategy may not be uniform across the country, since it is impossible to require all regions to follow the same reduction rates, even though it might be cost-efficient for long term air quality attainment (Xing et al., 2019). However, the indicator-based approach, which uses the ambient concentrations of only a few species, can quickly estimate the potential benefit or risk from the uniform controls, which can act as a basis to design more localized control strategies for particular regions. For example, additional simultaneous VOC control with NOx is recommended in regions located at VOC-limited regime.

As the reviewer suggested, we have provided some discussion about the usage of indicator in the revised manuscript, as follows.

(Page 22 Line 476-479) "Since the indicators are developed from simulations with spatially uniform emission controls across the country, they are especially useful for providing quick estimates of the potential benefits or risks from uniform controls. These estimates can also provide a basis to design more localized control strategies for particular regions."

Reference:

Xing, J., Zhang, F., Zhou, Y., Wang, S., Ding, D., Jang, C., Zhu, Y. and Hao, J.: Least-cost control strategy optimization for air quality attainment of Beijing–Tianjin–Hebei region in China. Journal of environmental management, 245, 95-104, 2019.

[Comment]: 420: What are the control pathways considered here? Ah âËŸA ËĞT ok they are mentioned in the figure caption but it would be useful to add to the text.

[Response]: The control pathways considered here are six types of VOC-to-NOx control ratios, including 0, 0.2, 0.4, 0.6, 0.8 and 1.0. As the reviewer suggested, we have added such information in the revised manuscript as follows.

(Page 20 Line 445-447) “To explore the cobenefits of reducing O3 and PM2.5 after simultaneous control of NOx and VOCs, we investigated the effectiveness of six control pathways with various VOC-to-NOx ratios including 0, 0.2, 0.4, 0.6, 0.8 and 1.0 (Fig. 14).”

[Comment]: Fig 14: It’s not clear to me how these results show that simultaneous reductions of O3 and PM2.5 are possible in January – as stated in the text. Rather, it looks like they are not except for all but one scenario (NOx:VOC = 1:1, only at the far end of the pathway). Potentially a very interesting figure here but it needs more explanation.

[Response]: Due to the strong NOx-saturated regime in January, compared to other months, a much larger VOC-to-NOx control ratio and greater NOx emission controls are required to prevent potential disbenefits from NOx controls and to achieve simultaneous reductions of O3 and PM2.5. As demonstrated in Figure 14(a), there is only one pathway that can meet the simultaneous reductions of O3 and PM2.5 (i.e., that with VOC-to-NOx equal to 1 and at the far end of the pathway, with greater emission controls).

As the reviewer suggested, we have provided additional explanation about Figure 14

in the revised manuscript as follows.

(Page 20 Line 447-456) “In general, O3 and PM2.5 concentrations can be reduced in all months through simultaneous control of NOx and VOC emissions, although different VNr and control levels are required in different months. In January (under strongly NOx-saturated conditions), reductions in PM2.5 and O3 require VOC emission controls in addition to NOx controls to prevent potential disbenefits associated with the nonlinear chemistry. The smaller VNr required for PM2.5 ($\sim$0.4) than for O3 ($\sim$1.0) in this case might be associated with the smaller PR for PM2.5 as well as the additional benefit of VOC controls in reducing secondary organic aerosols. Apparently, a larger VNr control ratio and greater emission control is required in January compared with other months. In Fig. 14(a), only one pathway can achieve simultaneous reduction in O3 and PM2.5 concentrations (i.e., the pathway with VNr equal to 1 and at the far end of the pathway, with reduction rates > 80%).”

[Comment]: 37: subscript on NOx.

[Response]: We have corrected it in the revised manuscript.

[Comment]: 51: Seinfeld et al. 2017 not in bibliography. Did the authors mean Seinfeld and Pandis (2012)?

[Response]: We are sorry for the typo. We have updated the reference to “Seinfeld and Pandis, 2012” in the revised manuscript.

[Comment]: 407 - 410: There is perhaps a word missing or something from this sentence, please check.

[Response]: As the reviewer suggested, we have revised this sentence in the revised manuscript as follows.

(Page 20 Line 433-436) “The PR results suggest strong NOx-saturated regimes in northern and eastern China including key regions such as the Sichuan Basin, YRD, and PRD, where simultaneous VOC control with a certain VOC-to-NOx ratio is required

to prevent increases in O3 levels from the NOx controls."

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-183/acp-2019-183-AC2-supplement.pdf

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