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Atmospheric Chemistry and Physics Discussions
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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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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 study explores the effectiveness of simultaneous NOx, VOC and NH3

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emission control on PM2.5 and O3 using CMAQ and a set of polynomial functions. The methods they propose are innovative and computationally efficient, but the presentation of the results and the significance of the findings still need further improvements. Overall, I think there are a number of issues that should be addressed in order to make this paper suitable for publication.

[Response]: We thank the reviewer for recognition of the implications of the results of the analysis presented. We basically followed all the comments and revised manuscript accordingly.

[Comment]: A common problem with statistical polynomial regression is overfitting. The fitting performance will certainly improve with higher orders, but it does not necessarily mean the models represent the true relationships. The authors show a very good fitting performance with inflated R values (0.93 to 1.0, Table 3), but this may actually reflect the models are overfitted. In order for the fitting to be trustworthy, the authors need to prove that the models are not overfitted. You could do so by conducting cross-validation for your model selection by partitioning your data to training and test groups. The test groups should not be used to fit the models, but to evaluate the model performance only.

[Response]: Following the reviewer's suggestion, we conducted the cross-validation for the pf-RSM model. The results are shown in Table R1.

Basically, the statistics of cross-validation are in the same order as shown in out-of-sample validations (OOS100 and OOS15). The performance in pf-RSM gets better along with the increase of sample numbers. Interesting finding is that the pf-RSM with marginal processing exhibits worse performance than that with even sampling method in cross-validation. That is because the samples with marginal processing are located closer to margin areas where is more difficult to predict (Xing et al., 2011). That also implies the samples with marginal processing has better representation of the variability. Nevertheless, the results of validations suggest the pf-RSM with current

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number of samples are not over-fitted, and the training samples selected in fitting the system is recommended to be 40 training samples with marginal processing.

To address the reviewer's concern, we added the discussion in the revised manuscript, as follows:

(Page 6 Line 19) "Method of leave-one-out cross validation (LOOCV) was used to examine whether the statistical polynomial regression is overfitting. The definition of LOOCV is to use a single sample from the original datasets as the validation data, and the remaining sample as the training data to build pf-RSM."

(Page 7 Line 16) Similar results are found in the cross validation (i.e., LOOCV), as the performance in pf-RSM gets better along with the increase of sample numbers. Basically, the statistics of cross-validation are in the same order as shown in out-of-sample validations (OOS100 and OOS15), except for the case of 20 training samples with marginal processing (worse performance due to under-fitting problem). Interesting finding is that the pf-RSM with marginal processing exhibits worse performance than that with even sampling method in cross-validation. That is because the samples with marginal processing are located closer to margin areas where is more difficult to predict (Xing et al., 2011). That also implies the samples with marginal processing has better good representation of the variability. Nevertheless, the results of validations suggest the pf-RSM with current number of samples are not over-fitted, and the training samples selected in fitting the system is recommend to be 40 training samples with marginal processing."

[Comment]: The polynomial functions assume the changes in pollutant concentration only depend on the changes of local emissions, but transport, meteorology, and deposition can also change the concentration. The authors need to provide justification why these factors are not considered.

[Response]: The design of pf-RSM is to investigate the air pollution responses to the emission perturbation which is related to the design of effective control policy. However, as the reviewer mentioned, the processes of transport, meteorology, and deposition can also change the concentration. Thus, we need atmospheric chemical transport model (i.e., CMAQ model in this study) to represent those influences on air pollution. But, contributions from those processes on air pollution are uncontrolled, thus we use fixed meteorological condition to drive all CMAQ simulation runs. To quantify the response of pollution to emission changes, we conducted multiple-CMAQ simulations under different emission scenarios and adopted statistic fitting or regression method to combine those simulations into a statistic model (i.e., RSM) which represents the response of air pollution to emission changes. The contribution of transport, meteorology, and deposition on pollution has already been considered in the CMAQ simulation, though we are not going to build up a response function of concentration to those variables.

To address the reviewer's concern, we clarified this point in the revised manuscript as follows:

(Page 3 Line 13) "We used the same meteorological condition for those multiple scenarios and only the emissions were changed in different scenarios."

[Comment]: The authors did a good job synthesizing their results concisely, but I would recommend the authors provide more insights into the numbers they reported. The interpretation of the results could be improved by: 1) Considering large body literature behind this topic and comparing your results with previous studies, especially those observation-based studies.

[Response]: As the reviewer suggested, we compared the model-based results with observation studies that use indicator to identify the O3 chemistry, as follows:

(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.”

We’ve added the comparison above in the revised manuscript.

Reference

Liu, H., Liu, C., Xie, Z., Li, Y., Huang, X., Wang, S., Xu, J. and Xie, P., 2016. A paradox for air pollution controlling in China revealed by “APEC Blue” and “Parade Blue”. Scientific reports, 6, p.34408.

Jin, X. and Holloway, T., 2015. Spatial and temporal variability of ozone sensitivity over China observed from the Ozone Monitoring Instrument. Journal of Geophysical Research: Atmospheres, 120(14), pp.7229-7246.

Chou, C. C.-K., Tsai, C.-Y., Shiu, C.-J., Liu, S. C. and Zhu, T.: Measurement of NOy during Campaign of Air Quality Research in Beijing 2006 (CAREBeijing-2006): Implications for the ozone production efficiency of NOx, J. Geophys. Res., 114, D00G01, doi:10.1029/2008JD010446, 2009.

[Comment]: 2) Providing more mechanistic reasoning on the results you provided. For example, The authors show the impacts of emission reduction vary in space and time (Section 3.2 and 3.3), but they do not provide any insights for the such variations. There is also no discussion on how the effectiveness of emission control vary with meteorology.

[Response]: As the reviewer suggested, we added following discussion in the revised manuscript:

(Page 9 Line 18) “The day-to-day variability of O3 depends on the budget of O3 source and sink influenced by meteorological variables including actinic flux, temperature, humidity, and precipitation, etc.”

(Page 9 Line 22) “The meteorological condition will also play an important role in the effectiveness of emission controls. Reductions in O3 were noticeable in both control cases, particularly on days when O3 levels were high. However, increases in O3 were observed on July 21-23 (precipitation event occurred across North China Plain), after the controls were applied and when O3 levels were low. This can be explained by the O3 chemistry scheme being in a strong VOC-limited condition on days with low O3 levels, resulting in enhanced O3 from NOx controls (Xing et al., 2011). Thus, the emission controls usually become less effective under unfavorable meteorological condition for O3 production. The pf-RSM also reproduced increases in O3 on those days.”

(Page 11 Line 23) “However, it might need further confirmed by more applications in other regions outside BTH and for a whole year analysis to better represent the seasonality.”

[Comment]: Page 1 Line 10: It looks like you’re talking about the O3 trend since 2010 (after the emission control), but Li et al. analyzed the trend between 2006 and 2011. I suggest cite a more recent paper that reflects the O3 trend since 2010, otherwise it’s misleading.

[Response]: We agree with the reviewer that the original statement is misleading. We clarified it in the revised manuscript as follows:

(Page 2 Line 8) “Since early 2010s (late 2000s in some regions such as Perl River Delta), strict regulations have been implemented on power plants and vehicle emissions, leading to a considerable NO2 reduction witnessed by the declining trend in satellite-retrieved NO2 column densities (i.e., reduced by 32% from 2011 to 2015, Liu et al., 2016).”

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(Page 2 Line 16) “The annual averaged O3 was increased by 0.86 ppb/year from 2006 to 2011 in Guangdong, accompanied by a correspondingly NO2 reduction of 0.61 ppb/year (Li et al., 2014). The recent observation data suggested a continue increasing trend of 8-hour maxima O3 in Zhuhai (from 128 to 142 μg m-3) and Shenzhen (from 122 to 134 μg m-3) in Perl River Delta from 2013 to 2016.”

[Comment]: Page 4 Line 30: Why do you choose 4th degree over 3rd degree? The difference is small, and you didn’t give any statistical justification.

[Response]: We replotted the difference between fitting with 4th degree and 3rd degree, as giving in Figure R1. The statistics show that the fitting with 4th degree has higher R values (R2 in fitting with 4th order is 0.9985 compared to 0.9735 in fitting with 3rd order) and smaller errors (MeanFE in fitting with 4th order is 0.2 compared to 0.6 in fitting with 3rd order).

We clarified this point in the revised manuscript as follows,

(Page 5 Line 2) “Better performance is shown in fitting with 4th order polynomial (R=0.999, MeanFE=0.2) than with 3rd order polynomial (R=0.987, MeanFE =0.6).”

[Comment]: Page 5 Line 30: Why “the training samples need to be as small as possible”? With small number of samples, the coefficients are very likely to be unstable, especially since you’re fitting high-order polynomial functions here.

[Response]: The limitation of RSM model is its heavy computing burden associated with the “samples” development. Each sample represents a CTM simulation under certain emission scenario. One CTM simulation for a typical month simulation period requires 400 CPU-hour, depending on the simulated domain size and selected mechanism. It is true that the for statistic fitting, we want as many as training samples. However, for CTM simulations, the less the better.

One advantage of pf-RSM is that its development requires about only 30% samples of traditional regression-based model, which significantly reduce the computing burden.

We clarified this point in the revised manuscript as follows:

(Page 2 Line 38) “The traditional RSM model is based on regression from thousands of “brute-force” simulations with chemical transport model (CTM) by using a maximum likelihood estimation - experimental best linear unbiased predictors (hereafter referred as “regression-based RSM”). However, such a large amount of CTM simulations (each simulation represents one training sample) required by RSM results in heavy computing burden (usually one CTM scenario for a month simulation needs 400 CPU-hour, depending on the simulated domain size and selected mechanism) which largely limits the application of traditional RSM.”

(Page 6 Line 2) “To minimize the number of CTM simulations (one simulation scenario represents one training sample), the number of training samples needed to be as small as possible, but greater than the number of terms (i.e., unknown coefficients) in the polynomial function.”

(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).”

[Comment]: Page 8 Line 9: It’s not clear to me how you set up the two emission control scenarios. Why do the magnitudes of emission reduction differ among species? How would the agreement between CMAQ and pf-RSM change if the emissions are reduced uniformly?

[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 samples (scenarios) to represent two different control levels, moderate and strict. The different magnitudes of emission reduction among species just present a certain scenario. The validation results might slight change if we change the scenarios (e.g., all pollutants reduced uniformly), 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 5: The word "observe" is misleading. There are no observations in this study.

[Response]: To avoid confusion, we modified the sentence in the revised manuscript as follows:

(Page 10 Line 4) "Larger FR values (slightly lower than 1.0) were shown in the central and southern regions (i.e., Beijing, Tianjin and HebeiS) than in other regions"

[Comment]: Page 9 Line 15: Please be more specific how your study is "consistent with findings of previous studies". It's also worthy mentioning how your study differs from previous studies in terms of methodology, results etc.

[Response]: As the reviewer suggested, we clarified those sentence in the revised manuscript as follows:

(Page 10 Line 8) "In both January and July, most of the urban areas present NH3-rich condition with FR from 0.75-0.95 (Table 4), implying the NH3 is sufficiently abundant to neutralize extra nitric acid produced by an additional 5%-35% (i.e., =1/FR-1) of

NOx emissions. 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."

[Comment]: Page 9 Line 20: I'd suggest the authors compare your model-based findings with observations (e.g. in situ or satellite observations) that use indicator approach to identify the limiting species for the O3 production.

[Response]: As the reviewer suggested, we compared the model-based results with observation studies that use indicator to identify the O3 chemistry, as follows: (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."

We've added the comparison above in the revised manuscript.

Chou, C. C.-K., Tsai, C.-Y., Shiu, C.-J., Liu, S. C. and Zhu, T.: Measurement of NOy during Campaign of Air Quality Research in Beijing 2006 (CAREBeijing-2006): Implications for the ozone production efficiency of NOx, J. Geophys. Res., 114, D00G01, doi:10.1029/2008JD010446, 2009.

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Jin, X. and Holloway, T.: Spatial and temporal variability of ozone sensitivity over China observed from the Ozone Monitoring Instrument. Journal of Geophysical Research: Atmospheres, 120(14), 7229-7246, 2015.

Liu, H., Liu, C., Xie, Z., Li, Y., Huang, X., Wang, S., Xu, J. and Xie, P.: A paradox for air pollution controlling in China revealed by “APEC Blue” and “Parade Blue”. Scientific reports, 6, 34408, 2016.

[Comment]: Page 9 Line 23: The results you show here are just for January and July, but how about other months, especially spring (or fall) when O3 production transitions from VOC-limited (or NOx-limited) to NOx-limited (VOC-limited)? Would you expect the effectiveness of emission control show any seasonality?

[Response]: It is true that the O3 chemistry varies under different meteorological conditions. Even in the same month, the O3 response to precursor reductions varies significantly on either high or low O3 days, as shown in section 3.2. Reductions in O3 were noticeable particularly on days when O3 levels were high. However, increases in O3 were observed on July 21-23, after the controls were applied and when O3 levels were low. This can be explained by the O3 chemistry scheme being in a strong VOC-limited condition on days with low O3 levels, resulting in enhanced O3 from NOx controls (Xing et al., 2011). It is expected that the O3 chemistry will be different in other months such as spring or fall. Further work is necessary to be conducted for a whole cycle year and to get a better representative of O3 seasonality.

Nevertheless, based on the daily analysis of O3 responses to precursor reductions in this study and also in our previous study (Xing et al., 2011), we can see that the effectiveness of emission control varies under different days. Generally, the controls on precursors will be more effective in reducing peak O3 concentrations, and will be less effective for days with low O3 levels which is usually in a strong VOC-limited condition.

We clarified this point in the revised manuscript.

(Page 9 Line 22) "The meteorological condition will also play an important role in the effectiveness of emission controls."

(Page 9 Line 27) "Thus, the emission controls usually become less effective under unfavorable meteorological condition for O3 production."

(Page 11 Line 23) "However, it might need further confirmed by more applications in other regions outside BTH and for a whole year analysis to better represent the seasonality."

[Comment]: Figure 9: How does meteorology affect the day-to-day variability of O3 and the effectiveness of emission controls?

[Response]: The day-to-day variability of O3 depends on the budget of O3 source and sink influenced by meteorological variables including actinic flux, temperature, humidity, and precipitation, etc. For example, there was a precipitation event occurred during July 21-23 in North China Plain, resulting in a lower O3 level across all 5 regions. Besides, the unfavorable meteorological condition for O3 production makes emission controls become less effectiveness. Since NOx become more abundant under unfavorable meteorological condition for photolysis, resulting in a stronger VOC-limited condition (Xing et al., 2011). Thus the emission controls become less effectiveness on low O3 days. We added following discussion in the revised manuscript.

(Page 9 Line 17) "The daily series of the CMAQ-simulated and pf-RSM-predicted 24-hour averaged PM2.5 and 1-hour maxima O3 in baseline and two control scenarios are shown in Figure 9. The day-to-day variability of O3 depends on the budget of O3 source and sink influenced by meteorological variables including actinic flux, temperature, humidity, and precipitation, etc."

(Page 9 Line 22) "The meteorological condition will also play an important role in the effectiveness of emission controls. Reductions in O3 were noticeable in both control cases, particularly on days when O3 levels were high. However, increases in O3 were

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observed on July 21-23 (precipitation event occurred across North China Plain), after the controls were applied and when O3 levels were low. This can be explained by the O3 chemistry scheme being in a strong VOC-limited condition on days with low O3 levels, resulting in enhanced O3 from NOx controls (Xing et al., 2011). Thus, the emission controls usually become less effective under unfavorable meteorological condition for O3 production. The pf-RSM also reproduced increases in O3 on those days."

[Comment]: Table 4: Why are there missing values for HebeiN?

[Response]: Since the PR is larger than 1.2 in HebeiN, the NOx control will always lead to a reduction in O3. Thus it is not necessary to estimate the reduction ratio of VOC to NOx to avoid increasing O3 for HebeiN.

The estimated FR in HebeiN is larger than 1.2, indicating strong NH3 poor condition. The extra benefit from simultaneous reduction of NH3 in HebeiN in July is estimated as 0.074 μg m-3 PM2.5 per 1% reduced NH3.

The values in HebeiN have been added in Table 4 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-AC2-supplement.pdf

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

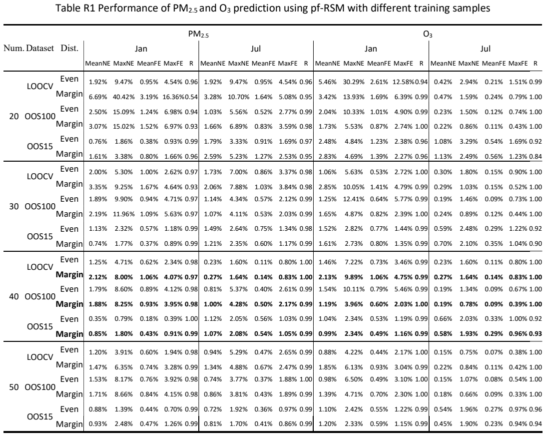

Table R1 Performance of $PM_{2.5}$ and O_3 prediction using pf-RSM with different training samples

Num.	Dataset	Dist.	$PM_{2.5}$										O_3									
			Jan					Jul					Jan					Jul				
			MeanNE	MaxNE	MeanFE	MaxFE	R	MeanNE	MaxNE	MeanFE	MaxFE	R	MeanNE	MaxNE	MeanFE	MaxFE	R	MeanNE	MaxNE	MeanFE	MaxFE	R
20	LOOCV	Even	1.92%	9.47%	0.95%	4.54%	0.96	1.92%	9.47%	0.95%	4.54%	0.96	5.46%	30.29%	2.61%	12.58%	0.94	0.42%	2.94%	0.21%	1.51%	0.99
		Margin	6.69%	40.42%	3.19%	16.36%	0.54	3.28%	10.70%	1.64%	5.08%	0.95	3.42%	13.93%	1.69%	6.39%	0.99	0.47%	1.59%	0.24%	0.79%	1.00
	OOS100	Even	2.50%	15.09%	1.24%	6.98%	0.94	1.03%	5.56%	0.52%	2.77%	0.99	2.04%	10.33%	1.01%	4.90%	0.99	0.23%	1.50%	0.12%	0.74%	1.00
		Margin	3.07%	15.02%	1.52%	6.97%	0.93	1.66%	6.89%	0.83%	3.59%	0.98	1.73%	5.53%	0.87%	2.74%	1.00	0.22%	0.86%	0.11%	0.43%	1.00
	OOS15	Even	0.76%	1.86%	0.38%	0.93%	0.99	1.79%	3.33%	0.91%	1.69%	0.97	2.48%	4.84%	1.23%	2.38%	0.96	1.08%	3.29%	0.54%	1.69%	0.92
		Margin	1.61%	3.38%	0.80%	1.66%	0.96	2.59%	5.23%	1.27%	2.53%	0.95	2.83%	4.69%	1.39%	2.27%	0.96	1.13%	2.49%	0.56%	1.23%	0.84
30	LOOCV	Even	2.00%	5.30%	1.00%	2.62%	0.97	1.73%	7.00%	0.86%	3.37%	0.98	1.06%	5.63%	0.53%	2.72%	1.00	0.30%	1.80%	0.15%	0.90%	1.00
		Margin	3.35%	9.25%	1.67%	4.64%	0.93	2.06%	7.88%	1.03%	3.84%	0.98	2.85%	10.05%	1.41%	4.79%	0.99	0.29%	1.03%	0.15%	0.52%	1.00
	OOS100	Even	1.89%	9.90%	0.94%	4.71%	0.97	1.14%	4.34%	0.57%	2.12%	0.99	1.25%	12.41%	0.64%	5.77%	0.99	0.19%	1.46%	0.09%	0.73%	1.00
		Margin	2.19%	11.96%	1.09%	5.63%	0.97	1.07%	4.11%	0.53%	2.03%	0.99	1.65%	4.87%	0.82%	2.39%	1.00	0.24%	0.89%	0.12%	0.44%	1.00
	OOS15	Even	1.13%	2.32%	0.57%	1.18%	0.99	1.49%	2.64%	0.75%	1.34%	0.98	1.52%	2.82%	0.77%	1.44%	0.99	0.59%	2.48%	0.29%	1.22%	0.92
		Margin	0.74%	1.77%	0.37%	0.89%	0.99	1.21%	2.35%	0.60%	1.17%	0.99	1.61%	2.73%	0.80%	1.35%	0.99	0.70%	2.10%	0.35%	1.04%	0.90
40	LOOCV	Even	1.25%	4.71%	0.62%	2.34%	0.98	0.23%	1.60%	0.11%	0.80%	1.00	1.46%	7.22%	0.73%	3.46%	0.99	0.23%	1.60%	0.11%	0.80%	1.00
		Margin	**2.12%**	**8.00%**	**1.06%**	**4.07%**	**0.97**	**0.27%**	**1.64%**	**0.14%**	**0.83%**	**1.00**	**2.13%**	**9.89%**	**1.06%**	**4.75%**	**0.99**	**0.27%**	**1.64%**	**0.14%**	**0.83%**	**1.00**
	OOS100	Even	1.79%	8.60%	0.89%	4.12%	0.98	0.81%	5.37%	0.40%	2.61%	0.99	1.54%	10.11%	0.79%	5.46%	0.99	0.19%	1.34%	0.09%	0.67%	1.00
		Margin	**1.88%**	**8.25%**	**0.93%**	**3.95%**	**0.98**	**1.00%**	**4.28%**	**0.50%**	**2.17%**	**0.99**	**1.19%**	**3.96%**	**0.60%**	**2.03%**	**1.00**	**0.19%**	**0.78%**	**0.09%**	**0.39%**	**1.00**
	OOS15	Even	0.35%	0.79%	0.18%	0.39%	1.00	1.12%	2.05%	0.56%	1.03%	0.99	1.04%	2.34%	0.53%	1.19%	0.99	0.66%	2.03%	0.33%	1.00%	0.92
		Margin	**0.85%**	**1.80%**	**0.43%**	**0.91%**	**0.99**	**1.07%**	**2.08%**	**0.54%**	**1.05%**	**0.99**	**0.99%**	**2.34%**	**0.49%**	**1.16%**	**0.99**	**0.58%**	**1.93%**	**0.29%**	**0.96%**	**0.93**
50	LOOCV	Even	1.20%	3.91%	0.60%	1.94%	0.98	0.94%	5.29%	0.47%	2.65%	0.99	0.88%	4.22%	0.44%	2.17%	1.00	0.15%	0.75%	0.07%	0.38%	1.00
		Margin	1.47%	6.35%	0.74%	3.28%	0.99	1.34%	4.88%	0.67%	2.47%	0.99	1.85%	6.13%	0.93%	3.04%	0.99	0.22%	0.84%	0.11%	0.42%	1.00
	OOS100	Even	1.53%	8.17%	0.76%	3.92%	0.98	0.74%	3.77%	0.37%	1.88%	1.00	0.98%	6.50%	0.49%	3.10%	1.00	0.15%	1.07%	0.08%	0.54%	1.00
		Margin	1.71%	8.66%	0.84%	4.15%	0.98	0.86%	3.81%	0.43%	1.89%	0.99	1.39%	4.71%	0.70%	2.30%	1.00	0.18%	0.66%	0.09%	0.33%	1.00
	OOS15	Even	0.88%	1.39%	0.44%	0.70%	0.99	0.72%	1.92%	0.36%	0.97%	0.99	1.10%	2.42%	0.55%	1.22%	0.99	0.54%	1.96%	0.27%	0.97%	0.96
		Margin	0.93%	2.48%	0.47%	1.26%	0.99	0.81%	1.70%	0.41%	0.86%	0.99	1.20%	2.33%	0.59%	1.15%	0.99	0.45%	1.90%	0.23%	0.94%	0.94

Fig. 1.

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Discussion paper

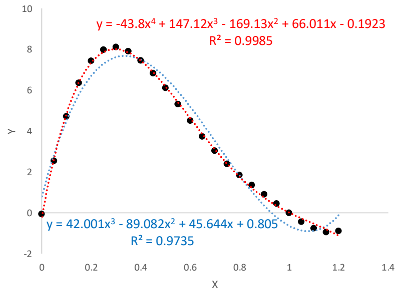

Figure R1 Fitting the PM2.5 responsive function to NOx with a polynomial of a single indeterminate plots with 3rd and 4th order

Fig. 2.