Response to the referee comments

Referees' comments:

Referee #1 (ACPD-15-C11505-2016)

The manuscript of Tang et al. elucidates potentials and limits of the Ensemble Kalman filter (EnKF) for chemical data assimilation (DA) and cross-correction of reactive gases and emissions (O_3 and NOx) in the framework of air-quality forecasts. The first part of the paper provides an extended validation of the previous study of Tang et al. (2011) with a focus on NO_2 forecasts. The observed degradation of NO_2 forecasts at some locations motivates the authors to examine the behavior of EnKF in a simplified model setting. DA experiments in such a controlled environment permit to identify the likely cause of the degradation, i.e. strong non-linearities between the controlled NOx emissions and the observed/assimilated O_3 concentration.

First, I appreciated the fact that the authors further validated their previous study and published these new results, even if this partially question the method that was employed in Tang et al. (2011). The EnKF is a powerful and flexible DA algorithm but requires particular care when applied to correct unobserved variables or parameters in complex models. Studies that use EnKF to cross-correct unobserved variables or model parameters should more often try to provide in-depth validation of assimilation results, as the authors did here.

Second, I liked the methodology that was used by the authors, i.e. reproduce the observed behavior within a simplified model. This allowed a reasonable scientific explanation for the NO_2 degradation and, more in general, permitted to highlight the effect of strong non-linearities in chemical DA. As the authors also stated, this topic is often not well discussed in the chemistry DA literature and deserves further research. It would have been nicer if the authors could propose an algorithm to automatically detect strong non-linear regimes and at least avoid the analysis degradation within the EnKF. This limits a bit the impact of the study for the air-quality DA community.

The manuscript is concise and well structured, although multiple sentences should be rewritten in a better English. Hence, I recommend publication in ACP as a companion paper of Tang et al. (2011), after the following comments are considered.

Response: Great thanks to the reviewer for the valuable comments. Accordingly, the manuscript has been revised with improvement of the language. A point-by-point response to the reviewer's comments is given as follows.

Specific comments:

1) Page 35694, line 27: 'the fast variability of the relationship between ozone concentrations and NOx emissions' is not very clear. The O_3-NOx emissions 'relationship' is a result of complex chemical reactions involving other species, radiation, temperature etc. Therefore, the 'relationship' is by *definition not unique and saying that it varies 'fast' has not a precise scientific meaning. I suggest the authors to either remove this sentence or rephrase to make it scientifically sound.*

Response: We agree. We have revised this sentence in the revised manuscript (P.1, line 25-28). *"The mixed effects observed in the cross-variable DA, i.e., positive DA impacts on NO_2 forecast over some urban sites, negative DA impacts over the other urban sites and weak DA impacts over suburban sites, highlighted the limitations of the EnKF under strong nonlinear relationships between chemical variables."*

2) Page 35695, line 14-15: '... the divergence of the influences of the initial condition optimization ...' is not clear. Do the authors mean that the initial condition has a weak influence on chemical forecasts? Please rephrase. It is also worth reminding that chemical species have a large range of life-times and can depend on different processes (emissions, photolysis etc.). This implies that this statement is not very informative without saying to which species and which forecast's duration we refer to.

Response: We agree. This sentence has been corrected in the revised manuscript (P.2, line 9-11). "*One of the major challenges in CDA is that the impact of the initial conditions on the forecast of air pollutants such as ozone decreases with simulation time (Gaubert et al., 2014; Jimenez et al., 2006).*"

3) Page 35695, line 24: I could not find demonstrations of improvements of ozone forecasts in Hanea et al (2004). Please remove the reference if not pertinent to the text.

Response: Thanks for this comment. We have removed this reference in the revised manuscript as suggested (P.2, line 17-18). "… *their applications have provided significant improvement of ozone forecasts (e.g., Tang et al., 2011).*"

4) Page 35698, line 8-9: 'fully supports nonlinear evolution of a model...' might lead to a wrong interpretation since the EnKF is based on Gaussian hypothesis and, as the authors show, it fails when non-linearities become too prominent. I guess the authors mean that EnKF can be implemented quite easily because the full non-linear model is employed during the ensemble forecast step. Please rephrase.

Response: Thanks for this comment and suggestion. We have rewritten this sentence in the revised manuscript as suggested (P.4, line 16-18). *"EnKF can directly calculate the background error covariance from the ensemble forecasts of the highly nonlinear model, which is very suitable for data assimilation in complex high-dimensional models (Carmichael et al., 2008)."*

5) Page 35698, line 16: see comment 3 for Hanea et al. 2004, Lin et al. 2008 is missing in the list of references and van Loon et al. 2000 does not demonstrate improved forecast skills for ozone (this concerns also page 35709, line 1). I suggest the authors to provide a more complete list of references that demonstrate the successful improvement of reactive gases forecasts through DA. Otherwise the authors should acknowledge that more research is needed in this regard.

Response: We have provided two new references to support the statement for improving forecasts through DA in the revised manuscript (P.4, line 22-24). The reference for Lin et al. (2008) is also added to the list of references (P.16, line 32-33). ***"Further applications of the EnKF in improving dust and ozone forecast skills through emission optimization have been reported (e.g., Constantinescu et al., 2007; Eben et al., 2005; Lin et al., 2008; Tang et al., 2011)."***

6) Page 35699, line 9-10: are the samples extracted from a normal distribution? Can the authors also precise the criteria that have been used to choose an ensemble of 50 members. How were the assimilation performances evaluated?

Response: Thanks for these comments. The samples are extracted from a normal distribution using the method proposed by Evensen (1994). The ensemble size is chosen after several sensitivity tests for the O_3 data assimilation (DA). Figure 1 displays the root mean square errors (RMSEs) of analyzed O_3 concentrations in the O_3 DA experiments with the EnKF under different ensemble members. The model domains and observation network is the same as in this study. As can be seen, the RMSEs in the tests with the ensemble size less than 30 are significantly higher than those in the other tests, which may be related to the spurious correlation induced by the small ensemble size. The RMSEs decreased with the increase of the ensemble size. However, due to the linear increase of the computational cost with the ensemble member, we took 50 members as a relatively good balance between computational efficiency and assimilation performance of the O_3 analysis. Furthermore, previous studies (e.g., Carmichael et al., 2008; Constantinescu et al., 2007) applying EnKF in chemical transport model took this ensemble size for ozone data assimilation. Due to space limit in the Journal, the sensitivity result presented in Fig.1 is not showed in the revised manuscript. However, we have clarified these issues in the revised manuscript (P.5, line 8-14).

"The random samples were extracted from a normal distribution using the method proposed by Evensen (1994). N is the ensemble size. The ensemble size (set as 50) was chosen based on several sensitivity experiments of ozone data assimilation. The experiments were performed with the same model domains and observation network as those employed in this study. The results suggest that an ensemble of 50 members keeps good balance between computational efficiency and assimilation performance of ozone analysis."

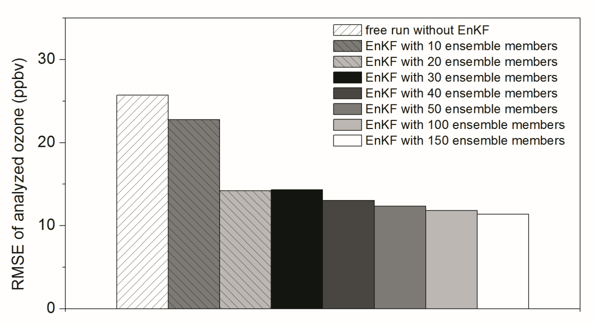

Figure 1. Root mean square errors (RMSEs) of the analyzed ozone concentrations over Beijing and its surrounding areas in the ozone data assimilation experiments that are conducted with ensemble Kalman filter (EnKF) for different ensemble members.

7) Page 35701, Sec. Data assimilation algorithm: Are the authors using some inflation and/or localization technique for the EnKF? If yes please describe it briefly in the text.

Response: Thanks. We have added some sentences to clarify this issue (P.5, line 15-17; P.7, line 15-18). ***"In order to avoid filter divergence, the NO_2 photolysis rate and vertical diffusion coefficient were perturbed by Gaussian distributed random noise, and the NOx emissions (to be updated by the EnKF) were perturbed by a time-correlated Gaussian distributed random noise." "To reduce the spurious impact caused by the finite ensemble size, localization was performed for analysis and only observations within a localization scale were used to update the NOx emissions at a model grid. The localization scale was set as 45km following the configuration of Tang et al. (2011)."***

8) Page 35702, Sec. Surface observation network: the authors should report some information about the measurement method and instrumental uncertainties of the employed in-situ NO_2 measurements. The issue of representativity of NO_2 measurements for the model grid should also be briefly discussed. Compared to O_3, NO_2 measurements in urban environment can be largely affected by local pollution and be not representative of a 10km model pixel. For example, are some of the used NO_2 sites exposed to heavy road traffic?

Response: Thanks! We have added some sentences in the revised manuscript with regard to this issue (P.7, line 28-29; P.8, line 1-6). ***"The measurements of NO_2 and O_3 were observed by online instruments (Model 42C& 42I NO-NO_2-NOx Analyzer and Model 49C&49I O_3 Analyzer from Thermo Scientific). The O_3 observations were assimilated hourly into the model to adjust NOx emissions. The direct comparison between the simulated and observed NO_2 data often suffered from the representativeness errors of the NO_2 measurements. In this study, the stations close to the main roads with heavy traffic were not included in order to reduce the influence of the representativeness errors of the NO_2 measurements. Nevertheless, under certain resolutions (9km for example), the representativeness errors still persisted in NO_2 measurements over urban areas."***

9) Page 35703, lines 10-13: It is not very clear to me why small emissions of NOx cannot undergo 'significant' changes with DA. If the variance of the ensemble is set as a percentage of the NOx emissions themselves, the DA correction is expected to be also proportional to the emissions and, therefore, locally significant. This should be the case unless the O_3 is not sensitive to NOx in low NOx regimes. Can the authors provide more insights on this? Looking at the corresponding O_3 ensemble spread and EnKF correction at suburban sites could also help.

Response: Thanks for raising this issue. According to your comment, Fig. 2 shows the hourly NO_2 concentrations from the observation, the simulation without DA and the simulation with

DA at the suburban site (Yongledian as an example). Figure 3 displays the ensemble spread of the hourly NO_2 forecasts at YLD in the data assimilation experiment using the EnKF. As can be seen in Fig. 2, the simulation without DA significantly underestimated the NO_2 concentrations at YLD, which is probably caused by the very low emission rates of NOx in the model. Under this situation, the perturbations on the NOx emissions still resulted in a relative small ensemble spread (shown in Fig. 3) in the DA using the EnKF, and the ensemble spread is significantly smaller than the errors in the real case. This would lead to weak corrections to the NOx emission over the suburban areas. On the other hand, the DA brought out significant errors of the NO_2 forecast at YLD during some period (especially on August 10 and 16), which may be induced by some wrong adjustments of the NOx emission over urban areas. Therefore, the minor changes of the RMSEs after DA are mainly caused by the above two reasons. We have clarified this issue in the revised manuscript (P.9, line 14-22). ***"At the suburban sites, the DA showed minor influence on NO_2 forecasts and had no statistically significant impacts on the RMSEs over 5 of the 6 suburban sites. Such minor DA impacts over the suburban sites could be explained firstly, by the fact that emission rates of NOx in the model were very low over suburban regions and the simulation without DA significantly underestimated the NO_2 concentrations. Even with the perturbations on the NOx emission, the ensemble spread was significantly weaker than the errors in the real case, and thereby reduced the DA impacts of the EnKF. On the other hand, in regards to the influences of the air pollutants transport from urban regions, observed negative DA impacts over some urban areas may have induced significant errors into the NO_2 forecasts."***

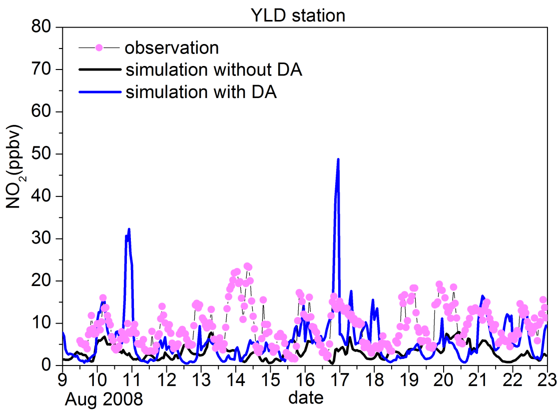

Figure 2. Time series of the hourly NO_2 concentrations obtained from the observation (magenta dots), the simulation without data assimilation (DA) (black line) and the simulation with DA (blue line) at the suburban site of Yongledian (YLD).

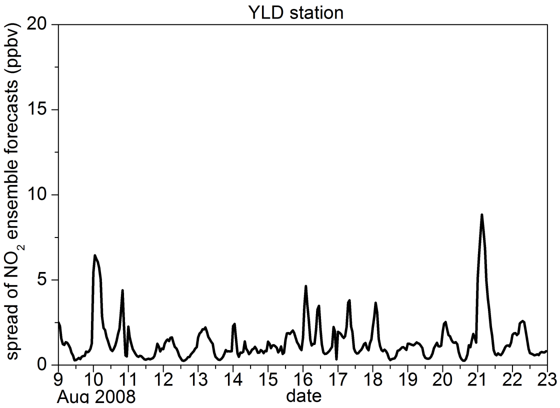

Figure 3. Ensemble spread of the hourly NO_2 ensemble forecasts at the suburban station of Yongledian (YLD) in the data assimilation with the EnKF.

10) Page 35704, lines 23-24: larger errors of modeled NO_2 in ppb units can also just be related to larger values of NO_2 concentration, which normally occurs in early morning and late evening, when NO_2 photo dissociation is not active and the boundary layer is shallow. Is the percentage error showing the same behavior?

Response: Thanks for your comments. According to your comment, we provide Fig. 4 showing daily variation of the root mean square errors (RMSEs) and the relative errors of the NO_2 forecast in the free model run over the urban stations (BY, CP, IAP, TJ and YF) with negative DA impacts. The relative errors present a similar daily variation as the RMSEs. The relative errors of the NO_2 forecasts in night and morning are also much higher than those during the daytime.

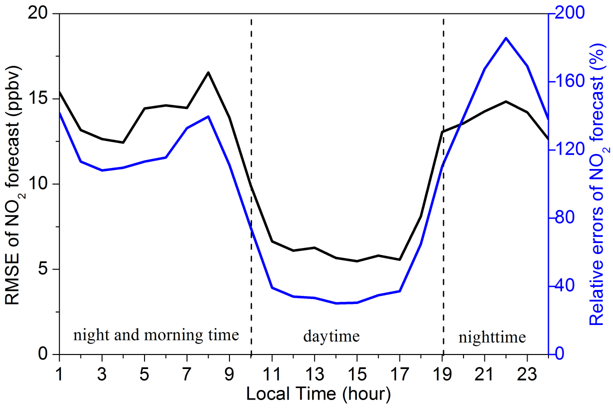

Figure 4. Daily variation of the NO_2 forecast errors in the free run of model at the urban stations (BY, CP, IAP, TJ and YF) with negative DA impacts. The black line represents the root mean square errors (RMSEs) and the blue line is the relative errors (percentage error).

11) Page 35708, lines 6-7: ' ... except for dealing with the non-linear relationship ...'. this part of the sentence is not clear, please clarify what you mean by 'except' and rephrase in case

Response: Thanks. We have rewritten this part in the revised manuscript (P.13, line 4-7). *"Note that above IDA experiments do not consider the complex model errors (e.g., errors in boundary layer or transport modeling). In the real case, model errors exist, and the DA scheme needs to properly quantify model uncertainties and deal with the nonlinearity between assimilated observations and adjusted variables simultaneously. Model errors may affect the results of the real DA."*

12) Page 35708, line 23: 'rapid variations' see comment n. 1

Response: We have revised this sentence in the manuscript (P.14, line 16-17). "*This suggests the variability of nonlinearity of the chemical system leads to different DA impacts during different periods of the day.*"

13) Page 35709, lines 17-20: The largest non-linearities arise from the chemical mechanism. Please explain why changing the model resolution would affect the non-linear behavior of the system and therefore the results of DA.

Response: Thanks for raising this issue. Thunis et al. (2015) reported some (minor) impacts of the spatial model resolution on the non-linearity behavior of the regional air quality modeling. However, the affect is still not very clear, and we have removed this part in the revised manuscript.

14) Page 35709, lines 19-20: *'Except for inversely estimating emissions ...* ' I cannot understand the exception. Doesn't this study show that the estimation of NOx emissions assimilating O_3 observation deals with chemical non-linearities? Please clarify this sentence.

Response: We have removed this sentence in the revised manuscript.

Technical corrections:

Please consider proof-reading the manuscript by an English native speaker. I provide here some suggestions for some sentences that should be ameliorated.

Response: Thanks for your suggestion. We have asked an English native speaker to improve the language of this manuscript. Please see the revised manuscript.

1) *Page 35694, lines 2-3 '... that has been validated as an efficient approach for improving ozone forecast' -> ' that has been used in the companion study to improve ozone forecasts over Bejing and surrounding areas'*

Response: We have revised this in the revised manuscript as suggested (P.1, line 11-12). *"… that has been used in the companion study to improve ozone forecasts over Beijing and surrounding areas."*

2) *page 35694, line 16: remove 'as a further investigation'*

Response: We have removed this in the revised manuscript as suggested.

3) *page 35695, line 7: '... that closely integrates ... is recognized ... ' > ' ... integrates ... and is recognized ...'*

Response: We have revised this as suggested (P.2, line 3-4). *"Chemical data assimilation (CDA) integrates models and observations to better represent the chemical state of the atmosphere and is recognized as a technique …"*

4) *page 35700, lines 8-9: remove 'provide various ... initial estimations) and '*

Response: We have removed this in the revised manuscript as suggested.

5) page 35704, line 8: ' *varies from the day to the night and the morning*' > ' is different between day-time, night-time and morning hours'

Response: We have revised this as suggested (P.10, line 9-10). *"… was different between daytime, nighttime and morning hours."*

6) *page 35706, lines 11-13: '... are combined by EnKF to produce linear correlations between them during the calculation of ...' does not sound very well in English, please rephrase*

Response: We have revised this sentence in the revised manuscript (P.12, line 6-7). "*At the analysis step, the ensemble samples of O_3 concentrations and NOx emissions were integrated into the EnKF to calculate the background error covariance in Eq. (5).*"

7) page 35706, lines 24-28: same as above

Response: We have revised this sentence in the revised manuscript (P.12, line 16-20). ***"From the results in Fig. 4(a-c), the most plausible cause of the negative DA impact on NOx emission estimation is the linearizing analysis of the EnKF in dealing with the cross-variable (O_3 to NOx emission) DA problem of a highly nonlinearly chemical system. With large bias in the a priori estimation of NOx emissions, the cross-variable assimilation may induce enhancement of the bias in NOx emissions."***

References

Carmichael, G., Chai, T., Sandu, A., Constantinescu, E., Daescu, D.: Predicting air quality: Improvements through advanced methods to integrate models and measurements, J. Comput. Phys. 227, 3540–3571, 2008.

Constantinescu, E.M., Sandu, A., Chai, T.F. and Carmichael, G.R.: Ensemble-based chemical data assimilation. II: Covariance localization, Q. J. R. Meteorol. Soc. 133: 1245–1256, 2007.

Evensen, G.: Sequential data assimilation with a nonlinear quasi-geostrophic model using Monte-Carlo methods to forecast error statistics, J. Geophys. Res. 99, 10143-10162, 1994.

Lin, C., J. Zhu, and Z. Wang: Model bias correction for dust storm forecast using ensemble Kalman filter, J. Geophys. Res., 113, D14306, doi:10.1029/2007JD009498, 2008.

Thunis, P., Clappier, A., Pisoni, E., Degraeuwe, B.: Quantification of non-linearities as a function of time averaging in regional air quality modeling applications, Atmos. Environ., 103, 263-275, 2015.

Referee #3 (ACPD-15-C10942-2015)

The manuscript investigates the results of across variable NOx emissions adjustment in an EnKF surface ozone data assimilation on NO_2 forecasts in Beijing and surrounding areas during the 2008 Summer Olympics. The main finding is that the assimilation of ozone data improved the NO_2 estimates during night and early morning but led to a significant deterioration during daytime over some urban sites, compared to surface measurements. The authors provide a possible explanation of this mixed effect by running and analyzing an idealized data assimilation experiment in which a similar effect is a result of a strong nonlinearity in the daytime NOx-O_3 chemistry combined with the presence of bias in the assumed model emissions.

The following is my take on the potential importance of this study. The theory of data assimilation makes a number of assumptions regarding linearity (although not necessarily in the case of EnKF and probability distributions but these are not always satisfied in reality. The question is how far can we push the limits? For example, typically we assume that observations and backgrounds are unbiased while they really are and assimilation still works. In this case it is important to know how much bias is too much or to what extent the assumptions can be violated without the results breaking down. As I understand it, the present study attempts to answer this question for a particular (and very important case of air quality estimation. I really like the idealized data assimilation experiment: I think this part of the analysis is quite convincing (if lacking some minor details, although it is less clear how it relates to the real data assimilation experiment (see my general comments 2 and 3. I also like the overall logic of the presentation. However, I do have a number of critical comments and suggestions, some more serious than others. I recommend the manuscript for publication after these are addressed.

Response: We very much appreciate the reviewer's valuable comments. The reviewer's comments play a very important role in improving the manuscript. We have revised the manuscript accordingly. A point-by-point response to the review's comments is as follows.

General comments

1. *The manuscript fits the criteria for a technical note. I'm not sure if it really qualifies as a research article. I would suggest publishing it as a technical note.*

Response: Thanks for this comment. This manuscript highlights a potential scientific issue in linkage with emission bias, data assimilation and air quality forecast. This systematically calls for a scientific debate on bias reduction in data assimilation process and further improvement of existing method. The manuscript therefore aims at contributing to the scientific progress and publishing in the form of a research article. Nevertheless, we also do not mind publishing it as a technical note suggested by the reviewer.

2. *The study decisively attributes the mixed effects of ozone data assimilation on forecast NO_2 to nonlinearities in the model based solely on an idealized experiment done with a very different and much simplified model. I think all we can say is that the idealized experiment offers a possible*

explanation. Given the simplified nature of the experiment there may be other factors that influence the results of the real data assimilation run, for example transport, which is not included in the idealized case.

Response: Thanks for raising this issue. Model errors from other processes (e.g., transport) are a key issue for the DA experiment and may affect the results of the real data assimilation. Following your comments, we have conducted additional idealized experiments to investigate the influences from the errors of other processes. Because it is quite difficult to simulate the transport process in the box model, we investigated the influences from the errors of the NO_2 photolysis rates that were found to be the top five uncertainty sources of ozone modeling over Beijing and surrounding areas during the Beijing Olympic Games (Tang et al., 2010).

In order to investigate the DA performance of adjusting NOx emissions under the presence of biases on other factors, we assumed that the NO_2 photolysis rate was overestimated by 20% in the idealized box modeling. Firstly, we were blind to the bias of the simulated NO_2 photolysis rate, so that no perturbation was operated on it in the DA experiment. The NOx emission was adjusted in the same way as the above-idealized experiments. Fig. 5a displays the results of the DA experiment under the error scenario of 30% overestimation in the a priori NOx emission. The DA corrected the NOx emission, but led to an underestimation of the emission. This over-correction of NOx emission by the DA could be associated with the bias in simulated NO_2 photolysis rate. Therefore, in the second experiment (Fig. 5b), we considered the uncertainty of the simulated NO_2 photolysis rate and perturbed the NO_2 photolysis rate in the DA. The error scenario was the same as in the first experiment. Under that condition, the DA performed better than that of the first experiment, without over-correction of NOx emission. The results of above experiments suggest that considering the model errors is crucial for the assimilation performance; otherwise the DA leads to over-correction to the state variable. In order to deal with this issue, simulated NO_2 photolysis rates and vertical diffusion coefficients (considered as the key uncertainty sources of the O_3 modeling) were perturbed to account their uncertainties into the real DA experiment. The third DA experiment is quite similar to the second one, but we increased the bias of the a priori NOx emission to 100% overestimation. The results are shown in Fig. 5c. Under large bias in the a priori NOx emission, the DA deteriorated NOx emission estimation. In short, in sight of considering the influence of the model errors, the limitations of the DA method in dealing with the large bias of a highly nonlinear system are still persistent. We have incorporated the above results into the revised manuscript to investigate this issue (P.13, line 4-30).

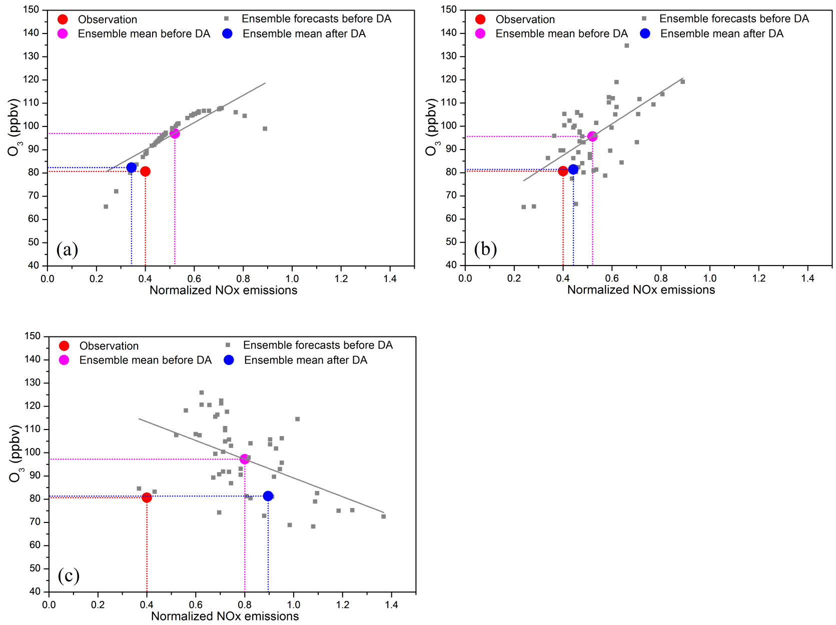

Figure 5 (a-c) O_3 concentrations (ppbv) and NOx emissions (no unit, normalized by the true NOx emission) before and after data assimilation (DA) and their ensemble samples before DA at 12:00 LT on August 12, 2008 in the three ideal DA experiments. The NO_2 photolysis rate is assumed to being overestimated by 20%. (a) The prior NOx emission is overestimated by 30% and adjusted by the DA. The uncertainty of the NO_2 photolysis rate is missed (without perturbations on it) in the DA. (b) The same as the DA in (a), but the uncertainty of the NO_2 photolysis rate is taken into account through perturbing it. (c) The same as the DA in (b), but the bias in the prior NOx emission is increased to 100%. The magenta dot represents the ensemble mean of the O_3 concentrations and NOx emissions before DA, and the gray squares denote the ensemble forecasts of O_3 concentrations corresponding to the perturbations of the NOx emissions. The gray line represents a linear relationship calculated from the ensemble samples of O_3 concentrations and NOx emissions. The red dot represents the true state of NOx emission and the observed O_3 concentration. The ensemble mean of the O_3 concentration and NOx emission after DA are denoted by the blue dot.

3. *I don't understand why all three idealized simulations are run with error scenarios in which the NOx emissions are underestimated compared to the truth. Is it expected to be the case for the real data assimilation experiment? Since the latter uses INTEX-B 2006 emissions I would rather expect them to be higher relative to the period of assimilation as, presumably, the air was less polluted during the Olympics than it was in 2006 (e.g. Wang etal. 2009, there maybe more suitable references. Possibly, I've misunderstood something.*

Response: Thanks for raising this issue. In the real case for the free run of the model, the NO_2 concentrations were overestimated at most of the urban stations but were underestimated at some of the urban stations. In the previous manuscript, we mainly considered the error scenarios for the underestimations of the NOx emissions in the three idealized simulations. Following your comment, in order to consider error scenarios with overestimations of NOx emission, four idealized DA experiments in which NOx emission was assumed to being overestimated by 10%, 30%, 50% and 100% respectively were performed. The results were shown in Fig. 6(a-d). In the first three experiments with 10%, 30% and 50% overestimations of the a priori NOx emission, the DA worked well and significantly reduced the biases of the emission. In the fourth experiment with the largest bias in the a priori emission estimation, the DA enhanced the bias of the emission estimation in daytime. These mixed DA effects under different biases of the a priori emission estimation are similar to those observed in previous idealized experiments conducted with underestimate scenarios. Both underestimate and overestimate scenarios confirm the mixed effects of the DA. The results of the new experiments have been added into the revised manuscript (P.12, line 25-30; P.13, line 1-3).

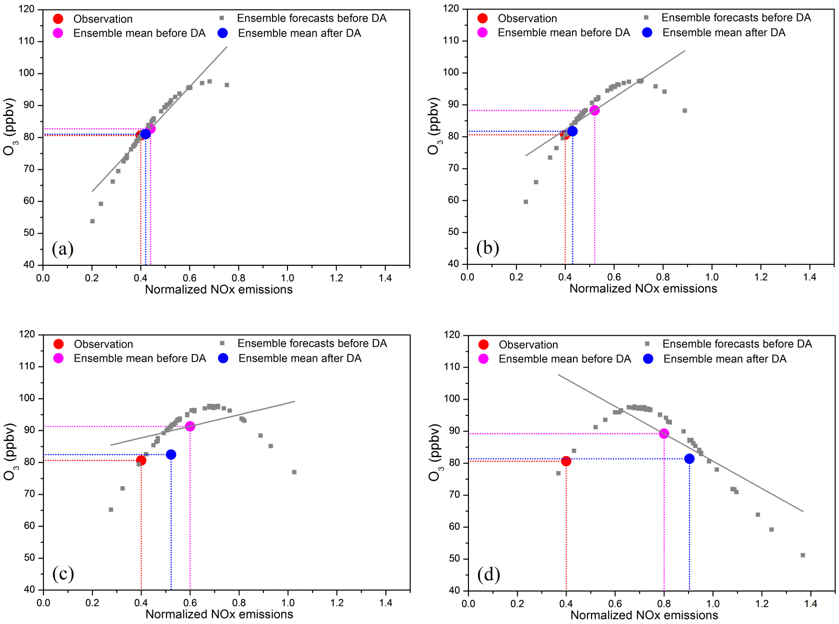

Figure 6 (a-d) O_3 concentrations (ppbv) and NOx emissions (no unit, normalized by the true NOx emission) before and after data assimilation (DA) and their ensemble samples before DA at 12:00 LT on August 12, 2008 in the four idealized DA experiments. (a) DA experiment with 10% overestimation in the a priori NOx emission; (b) DA experiment with 30% overestimation in the a priori NOx emission; (c) DA experiment with 50% overestimation in the a priori NOx emission; (d) DA experiment with 100% overestimation in the a priori NOx emission estimation. The magenta dot, the gray squares, the gray line, the red dot and the blue dot represent the

same as in Fig. 1.

4. *The authors focus on nonlinearity as the sole cause of the mixed results but the idealized experiment simply that it is the presence of a bias in the NOx emissions which leads to problems in a strongly nonlinear model. So it seems that the main culprit here is there action of the nonlinear system to the bias, not the nonlinearity by itself. Isn't EnKF supposed to work well with highly nonlinear systems? This point is important for conclusions and recommendations stemming from the study: in the real world cases, where nonlinearity may be hard to avoid, bias correction is essential.*

Response: Thanks for this important comment. We agree with you. Your suggestions are very good for summarizing the main results of this study. Therefore, we have revised the abstract, the conclusions and the other related contents in the revised manuscript.

Revisions in the abstract (P.1, line 25-30; P.2, line 1) ***"The mixed effects observed in the cross-variable DA … highlighted the limitations of the EnKF under strong nonlinear relationships between chemical variables. Under strong nonlinearity between daytime ozone concentrations and NOx emissions uncertainties (with large biases in the a priori emission), the EnKF may come up with inefficient or wrong adjustment to NOx emissions. The present findings reveal that bias correction is essential for the application of the EnKF in dealing with the DA inconsistency over strong nonlinear system."***

Revisions in the conclusions (P.14, line 29; P.15, line 1-9) ***"Through idealized DA experiments, the mixed effects were found to be strongly associated with the difficulty in dealing with highly nonlinear DA problem especially under large model biases. The results highlighted critical limitation of the EnKF for the chemical DA despite its strong performance for improving ozone forecasts (e.g., Tang et al., 2011). The results suggest that bias correction is crucial for the application of the EnKF in highly nonlinear chemical DA problem. Alternatively, avoiding the cross-variable DA between two strong-nonlinearly related variables such as NOx emissions and O_3 is also a possible way to overcome this issue. For example, assimilating NO_2 observations directly to optimize NOx emissions might produce better result than assimilating O_3 observations to improve the NO_2 forecasts and NOx emission estimations."***

5. *The use of English could use some polishing but I'm not going to focus on this aspect.*

Response：Thanks. We have asked an English native speaker to polish the language of this manuscript. Please see the revised manuscript.

Specific comments & technical corrections

P35696 L11 'indicates gaps'-indicate that gaps

Response: We have revised it to "*reveal some gaps*" in the revised manuscript (P.3, line 1-2).

P35696 L13 'calls'-call

Response: We have revised this as suggested in the revised manuscript (P.3, line 2).

P35698 L8. 'The simplicity in...' I'm not sure what this sentence means

Response: We have revised this sentence in the revised manuscript (P.4, line 16-18). *"EnKF can directly calculate the background error covariance from the ensemble forecasts of the highly nonlinear model, which is very suitable for data assimilation in complex high-dimensional models (Carmichael et al., 2008)."*

P35698 L10. 'Its implementation is very simple...' This sentence needs to be edited for grammar

Response: We have revised this sentence in the revised manuscript (P.4, line 18-20). *"Its implementation is very simple and does not require an adjoint model which is a very cumbersome task for complex high-dimensional model."*

P35699 L21. 60 sounds like a lot! I would like to see a more quantitative justification for that number. Also, 'the changes of emissions mover Beijing (...) during the (...) Olympic Games ' are likely to be systematic, i.e. the assumed INTEX-B estimates are probably biased (high) compared to the situation in 2008.

Response: Thanks for this comment. We have added new reference information to justify the estimation of the NOx emission uncertainties in the revised manuscript (P.5, line 18-26). *"Estimating the uncertainty of the NOx emissions used for the modeling during the Beijing Olympic Games was a hard task. The INTEX-B Asia inventory (Zhang et al., 2009) was estimated to contain 31% uncertainty in NOx emission estimation. But the base year of this inventory is 2006. Another key factor affecting the emission uncertainty is the temporary air pollution control measures during the Beijing Olympic Games. The control measures were estimated to reduce the NOx emissions by 36% to 47% (Wang et al., 2009; 2010). This would induce large biases into the emission inventory and lead to significant increase of the uncertainties of the emission inventory. Therefore, we estimated the uncertainty of the NOx emissions to be 60 % of the first guess emission rates, about twice the uncertainty in the INTEX-B Asia inventory."*

P35700 top of the page. Do the perturbations have zero mean?

Response: We have clarified this in the revised manuscript (P.6, line 4-6). *"Based on the method suggested by Evensen (1994), the perturbations of the variables in three dimensions were implemented through adding a pseudo smooth random field. The random samples were Gaussian distributed with zero mean."*

P35701 Eq(7). Shouldn't U be U', consistent with the notation used in Eqs. (4) and(5) ?

Response: We have revised this in the revised manuscript (P.7, line 6).

“ $U^a(i) = U'(i) + K(y'(i) - HU'(i)), i = 1, 2, \ldots, N$ （7）**”**

P35701 L20. I assume the ensemble mean ($U^a(i)$ averaged over i=1,..., N) is then used as the output analysis state for comparisons (e.g. the blue dots in Figures 4 and 5). Can you clarify this?

Response: Thanks. We have clarified this in the revised manuscript (P.7, line 13-15). “*The ensemble mean of $U^a(i)$ was taken as the best estimation after assimilating observations and was used as the output analysis state for comparisons (e.g. the blue dots in Figures 4 and 5).*”

P35702 L7. So surface ozone observations are assimilated every hour, correct?

Response：

Thanks. We have clarified this in the revised manuscript (P.8, line 1-2). *“The O_3 observations were assimilated hourly into the model to adjust NOx emissions.”*

P35703 L5. Here, 'forecast' is the mean of the ensemble of forecasts, correct?

Response: Yes. We have clarified this in the revised manuscript (P.8, line 28-29). *“Figure 2 compares the root mean square errors (RMSEs) of the 1 h ensemble mean forecast of NO_2 at the 17 stations in the RDA experiment with the RMSEs in the NonDA experiment.”*

*P35703 L5. How many observation forecast differences went into each RMSE? I'm getting ~14 *24 = 336 observations per location. Please provide these numbers here and in the caption of Figure 2. Would the result be different if, say, only the second week of assimilation was used in the RMSE computations, allowing assimilation to spin up? Are the reported differences between the RMSEs at different stations statistically significant?*

Response: Thanks for this comment. The observations used for the RMSE's calculation were a little different at different stations, because some observations were removed due to the quality control process for the data. We have listed the number of the observations used for each station in the revised manuscript (P.9, line 1-3; P.20, line 7-9). *“The RMSE of each site was calculated based on the hourly differences between NO_2 observation and the ensemble mean forecast of NO_2 from 00:00 LT 9 August to 00:00 LT 23 August in 2008. The number of valid observations used for each station is listed in Figure 2.” “The number of the valid observations used for the calculation is 336 at QHD, SJZ, TS, IAP, LF, YF and XH, and the numbers are 292, 226, 326, 317, 326, 320, 333, 321, 311, 323 at BD, PEK, BY, CZ, CP, TJ, XL, YJ, YLD and YuF respectively.”*

In order to investigate the sensitivity of the DA impacts to the period of the calculation, we did similar comparisons as in Figure 2 of the previous manuscript but focused on the first week and the second week independently. Figure 7a displays the result for the first week and Fig. 7b shows the results of the second week. Although the values of the RMSEs at the stations during

the first week were different from those during the second week, the impacts of the DA were similar during the two periods. The DA increased the RMSEs of the NO_2 forecast over the stations of TJ, BY, IAP, YF and CP, while it reduced the RMSEs over the stations of TS, PEK, SJZ, QHD and CZ. This result is also very similar to that shown in Figure 2 of the previous manuscript. Therefore, the figures for the two periods were skipped in the revised manuscript and a sentence was added into the revised manuscript to clarify this issue (P.9, line 27-30). ***"Further investigations were conducted on the variation of such mixed effects of the data assimilation on NO_2 forecasts over both first week (from 00:00 LT 9 August to 00:00 LT 16 August in 2008) and second week (from 00:00 LT 16 August to 00:00 LT 23 August in 2008). As a result, the DA mixed effects were relatively stable during the Beijing Olympic Games."***

We have checked the significance of the differences between the RMSEs at different stations and incorporated the information into the revised manuscript (P.9, line 3-7). ***"The differences of the RMSEs before and after DA were statistically significant over 11 stations (TJ, BY, YF, IAP, CP, XH, CZ, PEK, QHD, SJZ and TS) at the 95% level of the t-test, while there were no statistically significant differences of the RMSEs before and after DA over 6 stations (XL, YuF, YJ, YLD, LF and BD)."***

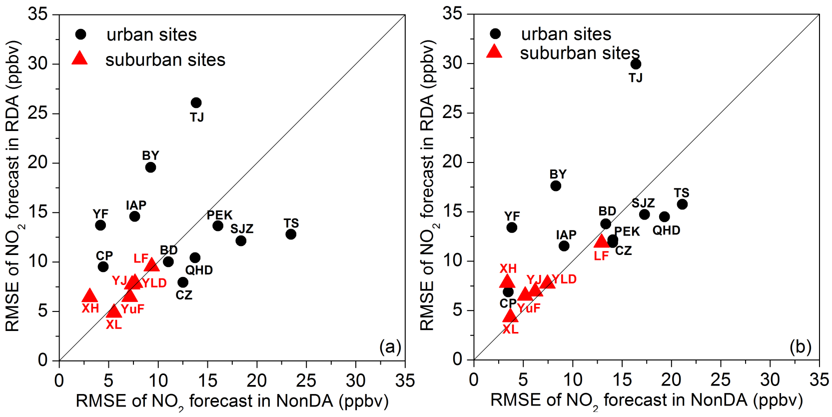

Figure 7 (a-b). Comparison of the root mean square errors (RMSEs) (ppbv) of 1 h NO2 forecasts at the 17 stations of Beijing and its surrounding areas in the real data assimilation (RDA) experiments and those in the reference (NonDA) experiment with a free run of the model (a) during the period of 00:00 LT 9 August to 00:00 LT 23 August in 2008 and (b) during the period of 00:00 LT 9 August to 00:00 LT 23 August in 2008. The comparisons at urban sites are denoted by the dots and those over suburban stations are represented by the triangles. The abbreviations of the station names are displayed close to the marks.

P35703.Was the RMSE dominated by a bias or random error? If it's a bias then is it low or high?

Response: Thanks for this comment. We have clarified this in the revised manuscript (P.9, line 7-11). ***"The RMSEs of the NO_2 forecasts in the free run of the model were dominated by the biases which accounted for 55~90% of the RMSEs (Bias/RMSE). Biases noticed in simulations performed***

over urban sites are relatively larger than those over the suburban ones. The free model run overestimated NO_2 concentrations at most of the urban stations, while underestimated it at most of the suburban ones."

P35705 L2. I wouldn't call it 'in-depth analysis'. The expression suggests analyzing every detail of the problem. What is really done here is one possible explanation of the results using a much idealized experiment.

Response: We have revised this in the revised manuscript (P.10, line 28-29; P.11, line 1-2). ***"An ideal experiment with a known true state provided a simple way to investigate the potential consequences of some key inspected factors in a highly complex system. In order to investigate the possible cause of observed mixed effects in RDA experiment, this study employed a simplified box model including the main chemical processes of NAQPMS (Xiang et al., 2010)."***

P35705. Do I understand correctly that the IDA experiment is just a single analysis step with a single ozone observation? Was the box model forecast run for 1hour or longer? Please, clarify.

Response: We have clarified this in the revised manuscript (P.11, line 13-17). ***"Ensemble runs of the box model were initialized by the ensemble forecasts of the chemical species of NAQPMS at 19:00 LT on 11 August 2008; NOx emissions were perturbed to provide ensemble samples of emissions during the following ensemble runs of the model. At 12:00 LT on 12 August 2008, the artificial O_3 observation was assimilated into the box model to adjust the NOx emissions."***

Figure 4. Is the magenta dot the result of averaging the grey dots? Is 'before DA' the same as 'forecast'?

Response: We have clarified this in the revised manuscript (P.22, line 6-9). ***"The grey squares denote the ensemble forecast O_3 concentrations corresponding to the perturbations of the NOx emissions (ensemble forecasts before DA), and the magenta dot represents the result of the ensemble mean of the grey squares (ensemble mean before DA)."***

P35709 L7. '...due to the needs of linearization at the analysis step, the assimilation should avoid the linearization...'. If DA requires linearization how can it avoid it? I think what the authors mean is that one should avoid problems in which very strong nonlinearities exist (as explained a few lines below. But then how does it jibe with the usual wisdom that the EnKF methodology works well for nonlinear problems? This sentence should be rephrased or dropped.

Response: We have revised this sentence in the revised manuscript (P.15, line 5-9). ***"Alternatively, avoiding the cross-variable DA between two strong-nonlinearly related variables such as NOx emissions and O_3 is also a possible way to overcome this issue. For example, assimilating NO_2 observations directly to optimize NOx emissions might produce better result than assimilating O_3 observations to improve the NO_2 forecasts and NOx emission estimations."***

Conclusions. Based on this analysis is it seems that the problem is the presence of a large bias in a highly nonlinear system.

Response: Thanks. We agree with you. Please see our response to the general comment 4.

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Limitations of ozone data assimilation with adjustment of NOx emissions: mixed effects on NO_2 forecast over Beijing and surrounding areas

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Abstract

This study investigates a cross-variable ozone data assimilation (DA) method based on an ensemble Kalman filter (EnKF) that has been used in the companion study to improve ozone forecasts over Beijing and surrounding areas. The main purpose is to delve into the impacts of the cross-variable adjustment of nitrogen oxides (NOx) emissions on the nitrogen dioxide (NO_2) forecasts over this region during the 2008 Beijing Olympic Games. A mixed effect on the NO_2 forecasts was observed through application of the cross-variable assimilation approach in the real-data assimilation (RDA) experiments. The method improved the NO_2 forecasts over almost half of the urban sites with reductions of the root mean square errors (RMSEs) by 15%~36% in contrast to big increases of the RMSEs over other urban stations by 56%~239%. Over the urban stations with negative DA impacts, improvement of the NO_2 forecasts (with 7% reduction of the RMSEs) was noticed in night and morning versus significant deterioration in daytime (with 190% increase of the RMSEs), suggesting that the negative DA impacts mainly occurred during daytime. Ideal data assimilation (IDA) experiments with a box model and the same cross-variable assimilation method confirmed the mixed effects found in the RDA experiments. In the same tendency, NOx emission estimation was improved in night and morning even under large biases in the prior emission, while deteriorated in daytime (except for the case of minor errors in the prior emission). The mixed effects observed in the cross-variable DA, i.e., positive DA impacts on NO_2 forecast over some urban sites, negative DA impacts over the other urban sites and weak DA impacts over suburban sites, highlighted the limitations of the EnKF under strong nonlinear relationships between chemical variables. Under strong nonlinearity between daytime ozone concentrations and NOx emissions uncertainties (with large biases in the a prior emission), the EnKF may come up with inefficient or wrong adjustment to NOx emissions. The present findings reveal that bias correction is essential for the application of the

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EnKF in dealing with the DA inconsistency over strong nonlinear system.

1. Introduction

Chemical data assimilation (CDA) integrates models and observations to better represent the chemical state of the atmosphere and is recognized as a technique for improving the simulations and forecasts of air pollutants such as ozone and aerosols (Carmichael et al., 2008; Sandu et al., 2011; Zhang et al., 2012). The role of CDA in optimizing initial and boundary conditions has been explored in several applications to improve forecasts of ozone and aerosol (Gaubert et al., 2014; Pagowski et al., 2014). Nevertheless, significant challenges persist in CDA.

One of the major challenges in CDA is that the impact of the initial conditions on the forecast of air pollutants such as ozone decreases with simulation time (Gaubert et al., 2014; Jimenez et al., 2006). To overcome such obstacle, emissions with large uncertainties and strong impacts on air quality modeling, identified as the crucial sources of uncertainties and considered to be the key control variables (Beekmann and Derognat, 2003; Hanna et al., 2001), have been integrated into the CDA. The importance of emissions as control variables in the CDA has also been documented recently (Carmichael et al., 2008; Koohkan et al., 2013; Zhang et al., 2012). Accordingly, advanced CDA techniques that enable inverse or cross-variable adjustments of emissions have been established and their applications have provided significant improvement of ozone forecasts (e.g., Tang et al., 2011).

However, the performances of such advanced CDA on the forecasts of other pollutants related to ozone are rarely reported and have not aroused enough attention. In this field, few studies stand out (Elbern et al., 2007; van Loon et al., 2000). Elbern et al., (2007) carried out two sets of data assimilation experiments with a four dimensional variational inversion method: (1) assimilation of ozone (O_3) and nitrogen dioxide (NO_2) observations simultaneously, and (2) assimilation of only O_3 observations. Both experiments resulted in reductions of nitrogen oxides (NOx) emissions after data assimilation in most cases even if the model underestimated the NOx concentrations before data assimilation. Similar results were reported by van Loon et al. (2000) through the assimilation of O_3 observations and adjustments of sulfur oxides (SOx) emissions using an ensemble Kalman filter. The method enhanced the emission rates of SOx when significant over-prediction of SO_2 concentrations subsisted. Such inconsistencies, i.e., the emissions enhanced under the overestimation of

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concentrations or the emissions reduced under the underestimation of concentrations, reveal some gaps between ozone forecast improvement and precursor emission optimization and call for a comprehensive evaluation of the cross-variable chemical data assimilation techniques.

Tang et al. (2011) employed a high horizontal resolution (9km) model to perform the assimilation of O_3 observations with the ensemble Kalman filter and the adjustment of NOx emissions for O_3 forecast improvement over Beijing and its surrounding areas. However, the impact of ozone assimilation on the precursor (NO_2 & volatile organic compounds) uncertainty was not elucidated. This paper (as an extension of Tang et al (2011)), based on the assimilation experiments performed by Tang et al., (2011), attempts to analyze in detail the impacts of the cross-variable ozone data assimilation on NO_2 forecasts over Beijing and surrounding areas during the 2008 Beijing Olympic Games. Both real O_3 data assimilation (with a 3-dimensional chemical transport model) and ideal O_3 data assimilation experiments (with a box model) are performed to investigate the state of NO_2 and NOx emissions during assimilation processes in order to provide further insights into the scientific potential of the assimilation method.

Section 2 describes the chemical transport model employed, the data assimilation algorithm and the surface observation network. Results from the real data assimilation experiments and the ideal data assimilation experiments are presented in Sect. 3. Section 4 presents conclusions and discussion.

2. Methodology

(1) Chemical transport model

The chemical transport model used for O_3 simulations was the Nested Air Quality Prediction Modeling System (NAQPMS) (Wang et al., 2001). Several applications of NAQPMS have been reported for simulating the chemical processes and transports of ozone, modeling the processes of aerosol and acid rain, and providing operational air quality forecasts in megacities such as Beijing and Shanghai (Wang et al., 2006). It contains modules for modeling the processes of emissions, advection, diffusion, dry and wet deposition, gaseous phase, aqueous phase, heterogeneous and aerosol chemical reactions. The gas-chemistry processes were simulated by the Carbon-Bond Mechanism Z (CBM-Z) which includes 133 reactions for 53 species (Zaveri and Peter, 1999). The dry deposition modeling followed the scheme of Wesely (1999). The vertical eddy diffusivity was parameterized based on a scheme by Byun and Dennis (1995). The O_3 simulations were configured

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with three nested domains and the horizontal resolutions were 81km, 27km and 9km respectively. The first domain covered East Asia with a 81km resolution and the second domain contained North China with a 27km resolution. The third domain displayed in Fig. 1 covered Beijing and its surrounding areas with 9km resolution. Vertically, the model was set as twenty terrain-following layers, nine of which were within the lowest 2 km of the atmosphere and the height of the first layer near the surface was 50 m. The Fifth-Generation National Center for Atmospheric Research (NCAR)/Penn State Mesoscale Model (MM5; Grell et al., 1994) was employed to provide the hourly meteorological inputs for NAQPMS. The regional emission data of the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) Asia inventory for 2006 with 0.5° × 0.5° resolution (Zhang et al., 2009) and the local high-resolution emission inventory were combined to provide the emission data for NAQPMS (Tang et al., 2011).

(2) Data assimilation algorithm

The assimilation algorithm employed was the ensemble Kalman filter (EnKF) proposed by Evensen (1994). The main feature of this method consists of a series of ensemble samples generally produced via ensemble forecasts to calculate the background error covariance of state variables. It serves as an approximate version of the Kalman filter (Kalman, 1960). EnKF can directly calculate the background error covariance from the ensemble forecasts of the highly nonlinear model, which is very suitable for data assimilation in complex high-dimensional models (Carmichael et al., 2008). Its implementation is very simple and does not require an adjoint model which is a very cumbersome task for complex high-dimensional model. It can be used for combined state and parameter estimation (Evensen, 2009). In the field of air pollution, the EnKF has been shown to be an efficient method in optimizing concentrations. Further applications of the EnKF in improving dust and ozone forecast skills through emission optimization have been reported (e.g., Constantinescu et al., 2007; Eben et al., 2005; Lin et al., 2008; Tang et al., 2011).

In the present study, the EnKF was employed to assimilate ozone observations for the corrections of NOx emissions. The main purpose is to elucidate the performances of that method during the cross-variable assimilation of O_3 observations. The sequential algorithm proposed by Houtekamer and Mitchell (2001), as a variant of EnKF, was adopted for its efficiency in computation. The first step of the implementation was to perturb ozone concentrations, NOx emissions and other key uncertainty sources of ozone modeling, i.e., photolysis rates and vertical diffusion coefficients, as

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described by the following equations:

$$\boldsymbol{x}'(\boldsymbol{i}) = \boldsymbol{x}^{\boldsymbol{b}} + \boldsymbol{\zeta}(\boldsymbol{i}),\ i = 1, 2, \ldots, N \quad (1)$$

$$\boldsymbol{e}'(\boldsymbol{i}) = \boldsymbol{e}^{\boldsymbol{b}} + \boldsymbol{\varepsilon}(\boldsymbol{i}),\ i = 1, 2, \ldots, N \quad (2)$$

$$\boldsymbol{q}'(\boldsymbol{i}) = \boldsymbol{q}^{\boldsymbol{b}} + \boldsymbol{\phi}(\boldsymbol{i}),\ i = 1, 2, \ldots, N \quad (3)$$

where $\boldsymbol{x}$, $\boldsymbol{e}$, and $\boldsymbol{q}$ are ozone concentrations, emissions, and other parameters (NO_2 photolysis rates and vertical diffusion coefficients) respectively, and the superscript b represents their background values in the model. The superscript $'$ represents the ensemble samples of these variables after perturbing the background values by random samples of $\boldsymbol{\zeta}$, $\boldsymbol{\varepsilon}$, and $\boldsymbol{\varphi}$. The random samples were extracted from a normal distribution using the method proposed by Evensen (1994). N is the ensemble size. The ensemble size (set as 50) was chosen based on several sensitivity experiments of ozone data assimilation. The experiments were performed with the same model domains and observation network as those employed in this study. The results suggest that an ensemble of 50 members keeps good balance between computational efficiency and assimilation performance of ozone analysis.

In order to avoid filter divergence, the NO_2 photolysis rate and vertical diffusion coefficient were perturbed by Gaussian distributed random noise, and the NOx emissions (to be updated by the EnKF) were perturbed by a time-correlated Gaussian distributed random noise. Estimating the uncertainty of the NOx emissions used for the modeling during the Beijing Olympic Games was a hard task. The INTEX-B Asia inventory (Zhang et al., 2009) was estimated to contain 31% uncertainty in NOx emission estimation. But the base year of this inventory is 2006. Another key factor affecting the emission uncertainty is the temporary air pollution control measures during the Beijing Olympic Games. The control measures were estimated to reduce the NOx emissions by 36% to 47% (Wang et al., 2009; 2010). This would induce large biases into the emission inventory and lead to significant increase of the uncertainties of the emission inventory. Therefore, we estimated the uncertainty of the NOx emissions to be 60 % of the first guess emission rates, about twice the uncertainty in the INTEX-B Asia inventory. The uncertainties of vertical diffusion coefficients in ozone modeling have been estimated by Beekmann and Derognat (2003), Hanna et al. (1998) and Moore et al. (2001), ranging from 25% to 50%. We estimated the uncertainty of vertical diffusion coefficients to be 35% of the first guess values which are close to the average estimation of

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the above three estimations. Also with reference to the studies of Hanna et al. (1998) and Moore et al. (2001), the uncertainty of the modeled photolysis rates was estimated to be 30%. The uncertainty of the modeled O_3 concentrations at the initial time was estimated to be 50% after comparing the modeled O_3 concentrations with observations. Based on the method suggested by Evensen (1994), the perturbations of the variables in three dimensions were implemented through adding a pseudo smooth random field. The random samples were Gaussian distributed with zero mean. The horizontal and vertical scales of initial error correlations could be effectively controlled using this method. The scales were set as 54 km in the horizontal and 3 model grids in the vertical (approximately 200 m) as in Tang et al. (2011).

Ensemble samples of the emissions, the vertical diffusion coefficients, the photolysis rates and the O_3 concentrations were used to derive ensemble forecasts of ozone. In order to achieve cross-variable adjustment for NOx emissions, an extended state variable was defined as:

$$\boldsymbol{U}'(i) = \begin{vmatrix} \boldsymbol{x}'(\boldsymbol{i}) \\ \boldsymbol{e}'(\boldsymbol{i}) \end{vmatrix}, i = 1, 2, \ldots, N \qquad (4)$$

where $\boldsymbol{x}'(\boldsymbol{i})$ and $\boldsymbol{e}'(\boldsymbol{i})$ represent the ozone concentrations and the emissions after perturbations as in Eq. (1). Through the ensemble forecast $\boldsymbol{x}'(\boldsymbol{i})$ is strongly dependent on $\boldsymbol{e}'(\boldsymbol{i})$, which makes it convenient for estimating the correlation between $\boldsymbol{x}$ and $\boldsymbol{e}$ and for cross-variable adjustment of NOx emissions. The background error covariance of the extended variable could be directly calculated from the ensemble forecast results during the simulation period:

$$\boldsymbol{P} = \frac{1}{N-1}\sum_{i=1}^{N}(\boldsymbol{U}'(i) - \overline{\boldsymbol{U}'})\,(\boldsymbol{U}'(i) - \overline{\boldsymbol{U}'})^{T} \qquad (5)$$

where $\overline{\boldsymbol{U}'}$ is the mean of the ensemble samples of the extended state variable and N is the ensemble size.

This algorithm treats the observations as random variables and perturbs them to prevent filter divergence of the EnKF (Houtekamer and Mitchell, 1998). When ozone observations are available, they were perturbed according to the observation errors (Gaussian with mean zero and covariance $\boldsymbol{R}$, including both measurement errors and representativeness errors):

$$\boldsymbol{y}'(\boldsymbol{i}) = \boldsymbol{y} + \boldsymbol{\Upsilon}(\mathbf{i}), \mathrm{i} = 1, 2, \ldots, \mathrm{N} \qquad (6)$$

$$\boldsymbol{\Upsilon} \in N(0, \boldsymbol{R}).$$

As suggested by von Loon et al. (2000), the observation errors were assumed to be within 10% of the

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original observation value and uncorrelated in time and space. It is worth noting that some other variants of the EnKF (e.g., the ensemble square root filter (EnSRF) proposed by Whitaker and Hamill, 2002) do not need the perturbations on observations but can also provide accurate analyses.

Then the ensemble samples of the extended variables from the ensemble forecasts could be updated through assimilating the ozone observations:

$$\boldsymbol{U^a(i) = U'(i) + \mathrm{K}(y'(i) - HU'(i)), i = 1, 2, \dots, N} \quad (7)$$

$$\boldsymbol{K = PH^T(HPH^T + R)^{-1}} \quad (8)$$

where $\boldsymbol{H}$ represents a linear operator mapping the extended state variable from model space to observational space, and $\boldsymbol{K}$ is the Kalman weight calculated based on the background error covariance and the observation error covariance. $\boldsymbol{U^a(i)}$ is the updated ensemble sample of the extended state variable and was used for the sequential ozone forecast. The updating of the ensemble ensembles of the extended variables was conducted one time every 1 hour (1h), and the updated NOx emissions were then used for the NO_2 forecast of the next hour. The ensemble mean of $U^a(i)$ was taken as the best estimation after assimilating observations and was used as the output analysis state for comparisons (e.g. the blue dots in Figures 4 and 5). To reduce the spurious impact caused by the finite ensemble size, localization was performed for analysis and only observations within a localization scale were used to update the NOx emissions at a model grid. The localization scale was set as 45km following the configuration of Tang et al. (2011).

(3) Surface observation network

We employed a regional surface air quality network over Beijing and its surrounding areas during the 2008 Beijing Olympic Games including 17 stations established by the Beijing Environment Monitoring Center and Chinese Academy of Science (Xin et al., 2010). Figure 1 displays the distributions of these stations and the non-industrial NOx emission rates of the observation regions in the third model domain. As can be seen, 11 urban stations (CP, PEK, BY, IAP, YF, BD, CZ, QHD, SJZ, TS, TJ) are located in the urban areas with high non-industrial NOx emission rates, and the other 6 (LF, XH, XL, YJ, YuF, YLD) are in the suburban areas with relatively low non-industrial NOx emission rates. The network provides observations of O_3 and NO_2 at the same temporal resolution as the model (i.e., 1h). The measurements of NO_2 and O_3 were observed by online instruments (Model 42C& 42I NO-NO_2-NOx Analyzer and Model 49C&49I O_3 Analyzer

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from Thermo Scientific). The O_3 observations were assimilated hourly into the model to adjust NOx emissions. The direct comparison between the simulated and observed NO_2 data often suffered from the representativeness errors of the NO_2 measurements. In this study, the stations close to the main roads with heavy traffic were not included in order to reduce the influence of the representativeness errors of the NO_2 measurements. Nevertheless, under certain resolutions (9km for example), the representativeness errors still persisted in NO_2 measurements over urban areas. In order to independently validate the assimilation results, three of the observation stations were withdrawn from the assimilation and were used for the validation. NO_2 observations not used in the assimilation were also used to assess the impacts of the cross-variable assimilation on the NO_2 forecasts.

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3. Results

3.1 Real data assimilation experiment

The real data assimilation (RDA) experiment assimilated the surface ozone observations over Beijing and surrounding areas to adjust the NOx emissions over these areas in the NAQPMS. The experiment was based on the study of Tang et al. (2011) in which the assimilation of real O_3 observations with the EnKF was performed to correct NOx emissions. The experiment focused on a two-week period from 00:00 LT 9 August to 00:00 LT 23 August in 2008. The initial conditions of the simulation were from a two-week spin-up model run. The initial conditions of ozone, NOx emissions and vertical diffusion parameters were perturbed at 19:00 LT on 8 August 2008 according to the equations (1), (2) and (3) and were used to derive ensemble runs of NAQPMS. After 5h free ensemble runs, the observed ozone data started at 00:00 LT on 9 August to be assimilated hourly into the third model domain (displayed in Fig. 1) of NAQPMS to adjust the NOx emissions. Adjusted factors of the NOx emissions were then used for the NO_2 forecast of the next hour. Both daytime and nighttime observations were assimilated. By considering possible large errors in the modeling of vertical profiles of air pollutants, we only adjust the variables in the first three vertical layers near the surface, which could reduce the influence of the modeling errors of vertical mixing on data assimilation. A free run of NAQPMS without data assimilation (NonDA) was also performed as a reference run to validate the assimilation results of the RDA experiment.

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Figure 2 compares the root mean square errors (RMSEs) of the 1 h ensemble mean forecast of NO_2 at the 17 stations in the RDA experiment with the RMSEs in the NonDA experiment. The

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RMSE of each site was calculated based on the hourly differences between NO_2 observation and the ensemble mean forecast of NO_2 from 00:00 LT 9 August to 00:00 LT 23 August in 2008. The number of valid observations used for each station is listed in Figure 2. The differences of the RMSEs before and after DA were statistically significant over 11 stations (TJ, BY, YF, IAP, CP, XH, CZ, PEK, QHD, SJZ and TS) at the 95% level of the t-test, while there were no statistically significant differences of the RMSEs before and after DA over 6 stations (XL, YuF, YJ, YLD, LF and BD). The RMSEs of the NO_2 forecasts in the free run of the model were dominated by the biases which accounted for 55~90% of the RMSEs (Bias/RMSE). Biases noticed in simulations performed over urban sites are relatively larger than those over the suburban ones. The free model run overestimated NO_2 concentrations at most of the urban stations, while underestimated it at most of the suburban ones. The DA impacts on the NO_2 forecast varied substantially from the suburban to the urban stations. At urban station such as BD, PEK, CZ, QHD, SJZ, and TS, the RMSEs were reduced by 15%~36% after DA, resulting in improvement of NO_2 forecasts in contrast to large increases, ranging from 56~239% of the RMSEs at CP, BY, IAP, YF and TJ. At the suburban sites, the DA showed minor influence on NO_2 forecasts and had no statistically significant impacts on the RMSEs over 5 of the 6 suburban sites. Such minor DA impacts over the suburban sites could be explained firstly, by the fact that emission rates of NOx in the model were very low over suburban regions and the simulation without DA significantly underestimated the NO_2 concentrations. Even with the perturbations on the NOx emission, the ensemble spread was significantly weaker than the errors in the real case, and thereby reduced the DA impacts of the EnKF. On the other hand, in regards to the influences of the air pollutants transport from urban regions, observed negative DA impacts over some urban areas may have induced significant errors into the NO_2 forecasts. The above results suggest the adjustment of the NOx emission by the ozone data assimilation has a mixed effect on the NO_2 forecast (i.e., weak DA impacts over suburban sites, positive DA impacts over some urban sites and negative DA impacts over others). Nevertheless, the assimilation produced significant improvement of ozone forecasts over all these sites, as reported by Tang et al. (2011).

Further investigations were conducted on the variation of such mixed effects of the data assimilation on NO_2 forecasts over both first week (from 00:00 LT 9 August to 00:00 LT 16 August in 2008) and second week (from 00:00 LT 16 August to 00:00 LT 23 August in 2008). As a result, the DA mixed effects were relatively stable during the Beijing Olympic Games. Figures 3 (a-c)

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display daily variation of the 1h NO_2 forecast RMSEs in RDA experiment and NonDA experiment over the urban stations with positive DA impacts (CZ, PEK, QHD, SJZ, and TS), those with negative DA impacts (BY, CP, IAP, TJ and YF) and the suburban stations (LF, XH, YLD, YJ and YuF with weak DA impacts). At the suburban stations, the cross-variable DA also showed very weak impacts on the NO_2 forecast in both the daytime and nighttime. At the urban stations with positive DA impacts, the cross-variable assimilation presented consistent positive DA impacts in daytime, nighttime and morning, with a 23% reduction of RMSEs during daytime and a 21% reduction in night and morning.

At the urban sites with negative DA impacts, the performance of the DA was different between daytime, nighttime and morning hours. Adjusting NOx emissions improves the forecasts of NO_2 concentrations during most of the night and the morning time by reducing 7% of the RMSEs in contrast to the deterioration of the forecast in the daytime with 190% increase of the RMSEs. This finding suggests that the impacts of the cross-variable assimilation on the NO_2 forecast during daytime are opposite to those in night and morning at these urban sites. In clear, negative DA impacts mainly occur in the daytime. As described by Tang et al. (2010b), daytime ozone is strongly nonlinearly related to high NOx emissions over urban areas (in particular over central Beijing), whereas nighttime ozone is mainly controlled by the titration reaction of O_3-NO with weak nonlinearity. Due to the obvious discrepancy between daytime ozone and nighttime ozone chemistry, further experiments were carried out in following section to elucidate the impact of the chemistry on the cross-variable assimilation.

Another phenomenon observed in Figs. 3(a-b) is that the errors in NO_2 forecasts with the free model run in night and morning were much higher than those in daytime. This might due to the large uncertainties in modeling of nighttime boundary layer over urban regions (Kleczek et al., 2014). Although the modeling of vertical diffusion was taken as a key uncertainty source in our data assimilation, its uncertainty was not constrained by the data assimilation. Therefore, high errors still subsisted in the nighttime NO_2 forecasts after data assimilation, as shown in Figs. 3(a-b).

3.2 Ideal data assimilation experiment

An ideal experiment with a known true state provided a simple way to investigate the potential consequences of some key inspected factors in a highly complex system. In order to investigate the

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possible cause of observed mixed effects in RDA experiment, this study employed a simplified box model including the main chemical processes of NAQPMS (Xiang et al., 2010). Within conducted ideal data assimilation (IDA) experiments, the true state of ozone concentrations and NOx emissions were assumed to be known. The main purpose is to closely monitor the impacts of ozone chemistry on the cross-variable assimilation method experimented in the RDA. However, this investigation did not take into account complex transport processes and the removal processes were simulated by multiplying the concentrations by removal coefficients. The experiments with the box model were conducted on the IAP station where negative impact on NO_2 forecasts is observed in the RDA experiment. Emission rates and meteorological parameters are from the inputs used by NAQPMS.

Firstly, the IDA experiments focused on the negative DA impacts on the daytime NO_2 forecasts. The a priori emission rates from NAQPMS and their corresponding O_3 concentrations modeled with the box model were assumed to be the true state and were used for validation of the optimized emissions from DA. Ensemble runs of the box model were initialized by the ensemble forecasts of the chemical species of NAQPMS at 19:00 LT on 11 August 2008; NOx emissions were perturbed to provide ensemble samples of emissions during the following ensemble runs of the model. At 12:00 LT on 12 August 2008, the artificial O_3 observation was assimilated into the box model to adjust the NOx emissions. Artificial O_3 observations were generated through adding slight random errors to the true state of O_3 concentrations. To be consistent with the RDA experiment, the random errors for perturbing observations were also assumed to be within 10% of the true value. Three error scenarios for NOx emissions (10%, 30% and 50% underestimations) were assumed and separately applied to simulations of the box model. In order to avoid dealing with complex model errors, the errors in NOx emissions were assumed to be the only error sources of ozone modeling. For each error scenario, cross-variable adjustment of the NOx emissions through assimilating the artificial O_3 observations with the EnKF was conducted. Figures 4(a-c) show the O_3 concentrations and NOx emissions before and after DA, with their ensemble samples before DA at 12:00 August 12, 2008.

Figure 4a presents the results under the first scenario with 10% underestimation of NOx emissions (S1). The analyzed O_3 concentration and NOx emission after DA were close to their true state, suggesting an improvement of the NOx emission estimation from the cross-variable assimilation. Figure 4b shows the results under the second scenario with 30% underestimation of NOx emissions (S2). The DA inefficiently reduced the error in NOx emission, since large errors (about 20%) still

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persisted in the optimized NOx emission. Ensemble samples of O_3 concentrations shown in Fig.4b were obtained from the ensemble runs of the box model that were derived from the ensemble samples of NOx emissions (also shown in Fig.4b). Obviously, the ensemble forecasts of O_3 concentrations presented high nonlinear responses to the perturbations of NOx emissions. This suggests that the EnKF with Monte Carlo simulations can properly predict the nonlinear evolutions of error statistics of the O_3 modeling. At the analysis step, the ensemble samples of O_3 concentrations and NOx emissions were integrated into the EnKF to calculate the background error covariance in Eq. (5). The linearized relationship between the O_3 concentrations and the NOx emissions is presented in Fig. 4b. Noticeable discrepancies appear between the nonlinear relationship denoted by the ensemble samples and the linearized relationship at the analysis step. This significantly weakens the performance of the EnKF in the cross-variable adjustment.

In the third scenario (S3) with NOx emissions underestimated by 50%, enhanced deterioration of the NOx emission estimations was observed (Fig. 4c). The DA closely adjusted the simulated O_3 concentration to the true state, but induced additional bias to previously underestimated NOx emission. Such negative DA impact on NOx emission estimation was similar to the phenomenon observed on the daytime NO_2 forecast over some urban stations in the RDA experiment. From the results in Fig. 4(a-c), the most plausible cause of the negative DA impact on NOx emission estimation is the linearizing analysis of the EnKF in dealing with the cross-variable (O_3 to NOx emission) DA problem of a highly nonlinearly chemical system. With large bias in the a priori estimation of NOx emissions, the cross-variable assimilation may induce enhancement of the bias in NOx emissions. The results of the three IDA experiments (i.e., positive DA impact under the first and second scenarios and negative impact under the third scenario) confirm the mixed effects of the cross-variable assimilations observed in the RDA experiments, and suggest a strong link between the mixed effects and the linearization process at the analysis step of the EnKF over strongly nonlinear chemical system.

In order to consider error scenarios with overestimations of NOx emission, four idealized DA experiments in which NOx emission was assumed to being overestimated by 10%, 30%, 50% and 100% respectively were performed. The results are shown in Fig. 5(a-d). In the first three experiments with 10%, 30% and 50% overestimations of the a priori NOx emission, the DA worked well and significantly reduced the biases of the emission. In the fourth experiment with the largest bias in the a priori emission estimation, the DA enhanced the bias of the emission estimation in daytime. These

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mixed DA effects under different biases of the a priori emission estimation are similar to those observed in previous idealized experiments conducted with underestimate scenarios. Both underestimate and overestimate scenarios clearly confirm the mixed effects of the DA.

Note that above IDA experiments do not consider the complex model errors (e.g., errors in boundary layer or transport modeling). In the real case, model errors exist, and the DA scheme needs to properly quantify model uncertainties and deal with the nonlinearity between assimilated observations and adjusted variables simultaneously. Model errors may affect the results of the real DA. Thus, in order to investigate the DA performance of adjusting NOx emissions under the presence of biases on other factors, we assumed that the NO_2 photolysis rate was overestimated by 20% in the idealized box modeling, since the errors of the NO_2 photolysis rates were found to be the top five uncertainty sources of ozone modeling over Beijing and surrounding areas during the Beijing Olympic Games (Tang et al., 2010a).

Firstly, we were blind to the bias of the simulated NO_2 photolysis rate, so that no perturbation was operated on it in the DA experiment. The NOx emission was adjusted in the same way as the above-idealized experiments. Fig. 6a displays the results of the DA experiment under the error scenario of 30% overestimation in the a priori NOx emission. The DA corrected the NOx emission, but led to an underestimation of the emission. This over-correction of NOx emission by the DA could be associated with the bias in simulated NO_2 photolysis rate. Therefore, in the second experiment (Fig. 6b), we considered the uncertainty of the simulated NO_2 photolysis rate and perturbed the NO_2 photolysis rate in the DA. The error scenario was the same as in the first experiment. Under that condition, the DA performed better than that of the first experiment, without over-correction of NOx emission. The results of above experiments suggest that considering the model errors is crucial for the assimilation performance; otherwise the DA leads to over-correction to the state variable. In order to deal with this issue, simulated NO_2 photolysis rates and vertical diffusion coefficients (considered as the key uncertainty sources of the O_3 modeling) were perturbed to account their uncertainties into the real DA experiment. The third DA experiment is quite similar to the second one, but we increased the bias of the a priori NOx emission to 100% overestimation. The results are shown in Fig. 6c. Under large bias in the a priori NOx emission, the DA deteriorated NOx emission estimation. In short, in sight of considering the influence of the model errors, the limitations of the DA method in dealing with the large bias of a highly nonlinear system are still persistent.

To investigate the DA impacts on the NOx emissions in night and morning, variations of O_3 concentrations and NOx emissions before and after DA and their ensemble samples before DA at 8:00 August 13, 2008 (morning time) are shown in Figs. 7(a-c). Similar trends (not shown here) were obtained for other night and morning times. In Figs. 7(a-c), different level errors (10%, 30% and 50% underestimations) in NOx emissions were significantly reduced through the cross-variable assimilation with the EnKF. The ensemble forecasts of morning O_3 concentrations show near-linear responses to the uncertainties (or perturbations) of NOx emissions; the linearization of the EnKF at the analysis step worked properly to correct the biases in NOx emissions. The positive DA impacts on the NOx emission estimation in IDA experiments in night and morning were consistent with the improvement of the NO_2 forecasts after data assimilation in RDA experiment. In comparison with the mixed effects of the DA in daytime, the positive DA impacts in night and morning in both RDA and IDA experiments indicate that the assimilation of O_3 observations with the EnKF might be useful in optimizing NOx emissions and NO_2 forecasts in night and morning. Furthermore, the ensemble forecasts of O_3 concentrations show strong nonlinear responses to the perturbations of NOx emissions during daytime in Figs. 4(a-c) but present near-linear responses in night and morning in Figs. 7(a-c). This suggests the variability of nonlinearity of the chemical system leads to different DA impacts during different periods of the day.

4. Conclusion and discussion

The impacts of cross-variable adjustment of NOx emissions on NO_2 forecasts were investigated through assimilating O_3 observations with a variant of the EnKF (proposed by Houtekamer and Mitchell, 2001) over Beijing and surrounding areas during the 2008 Beijing Olympic Games. Both real DA experiments with a 3-dimensional chemical transport model and ideal DA experiments with a simplified box chemical model were performed.

The results of the data assimilation experiments revealed mixed effects of the cross-variable assimilation with the EnKF. The DA worked properly in improving the NO_2 forecasts and optimizing the NOx emissions in night and morning when the uncertainties of O_3 concentrations were almost linearized to those of NOx emissions. During daytime, the data assimilation resulted in positive DA impacts on NO_2 forecasts over some urban sites, negative over other urban sites and weak impacts over suburban sites. Through idealized DA experiments, the mixed effects were found to be strongly

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associated with the difficulty in dealing with highly nonlinear DA problem especially under large model biases. The results highlighted critical limitation of the EnKF for the chemical DA despite its strong performance for improving ozone forecasts (e.g., Tang et al., 2011).

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The results suggest that bias correction is crucial for the application of the EnKF in highly nonlinear chemical DA problem. Alternatively, avoiding the cross-variable DA between two strong-nonlinearly related variables such as NOx emissions and O_3 is also a possible way to overcome this issue. For example, assimilating NO_2 observations directly to optimize NOx emissions might produce better result than assimilating O_3 observations to improve the NO_2 forecasts and NOx emission estimations. Nevertheless, strong nonlinearity issue remains a critical challenge in the chemical DA. In sum, DA approaches that enable dealing with high nonlinearity in both model evolution and analysis step are needed. Particle filters as nonlinear filter method (e.g., Moral et al., 1996; van Leeuwen, 2009; 2010) might have potential in this field if its limitation for high dimensional system application (Stordal et al., 2011) can be overcome.

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Acknowledgements

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Figures

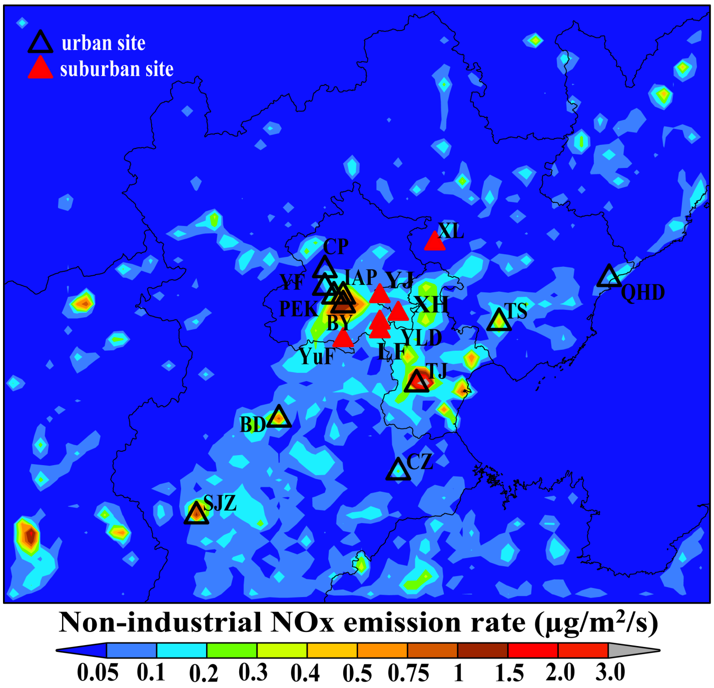


Figure 1 Distribution of the observation stations and non-industrial NOx emission rates in the third model domain (9km resolution) that covers Beijing and its surrounding areas. The non-industrial NOx emission rates (μg/m^2/s) are divided into different bins (<0.05; 0.01-0.1; 0.1-0.2; 0.2-0.3; 0.3-0.4; 0.4-0.5; 0.5-0.75; 0.75-1.0; 1.0-1.5; 1.5-2.0; 2.0-3.0) and represented by different shaded colors. The urban areas with high non-industrial NOx emission rates are marked by the brown and red colors, and the suburban or rural areas with low non-industrial NOx emission rates are marked by the green or blue colors. The 11 urban sites are denoted by the black triangles, and the 6 suburban stations are represented by the red triangles. The abbreviations of the station names are displayed close to the marks.

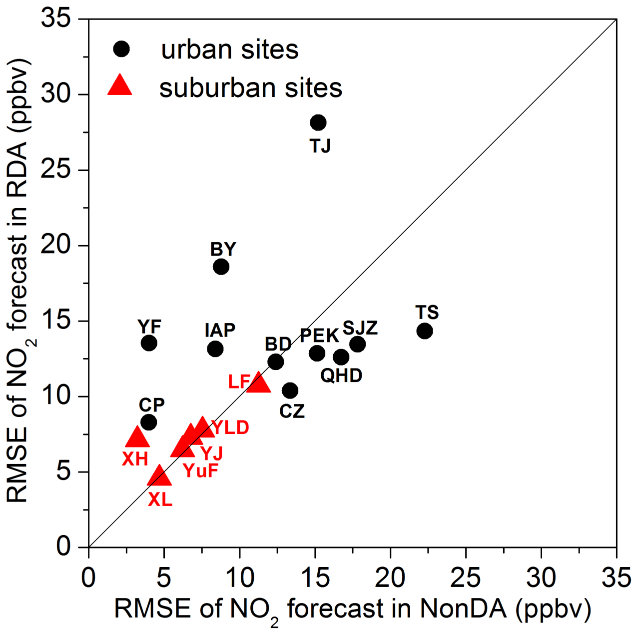
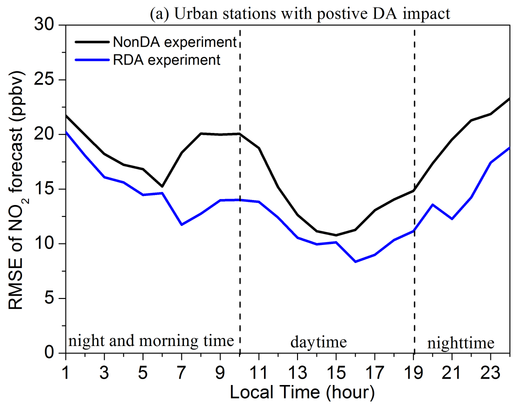
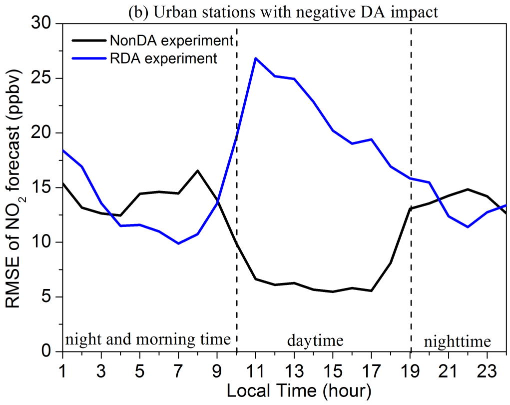


Figure 2 Comparison of the root mean square errors (RMSEs) (ppbv) of 1h NO_2 forecasts at the 17 stations of Beijing and its surrounding areas during the period of 00:00 LT 9 August to 00:00 LT 23 August in 2008 in the real data assimilation (RDA) experiments and those in the reference (NonDA) experiment with a free run of the model. The comparisons at urban sites are denoted by the dots and those over suburban stations are represented by the triangles. The abbreviations of the station names are displayed close to the marks. The number of the valid observations used for the calculation is 336 at QHD, SJZ, TS, IAP, LF, YF and XH, and the numbers are 292, 226, 326, 317, 326, 320, 333, 321, 311, 323 at BD, PEK, BY, CZ, CP, TJ, XL, YJ, YLD and YuF respectively.





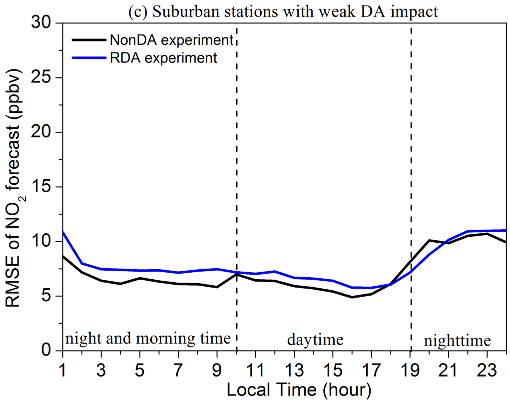
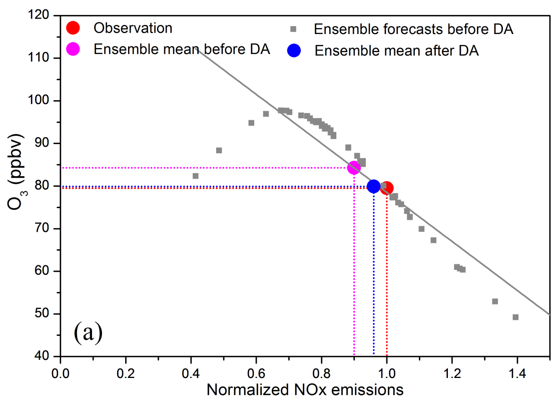


Figure 3 Daily variation of the 1h NO_2 forecast RMSEs (ppbv) in the real data assimilation (RDA) experiments (blue line) and the reference (NonDA) experiment with a free run of the model (black line) over: (a) urban stations (CZ, PEK, QHD, SJZ, and TS) with positive DA impacts; (b) urban sites (BY, CP, IAP, TJ and YF) with negative DA impacts; (c) suburban stations (LF, XH, YLD, YJ and YuF) with weak DA impacts.



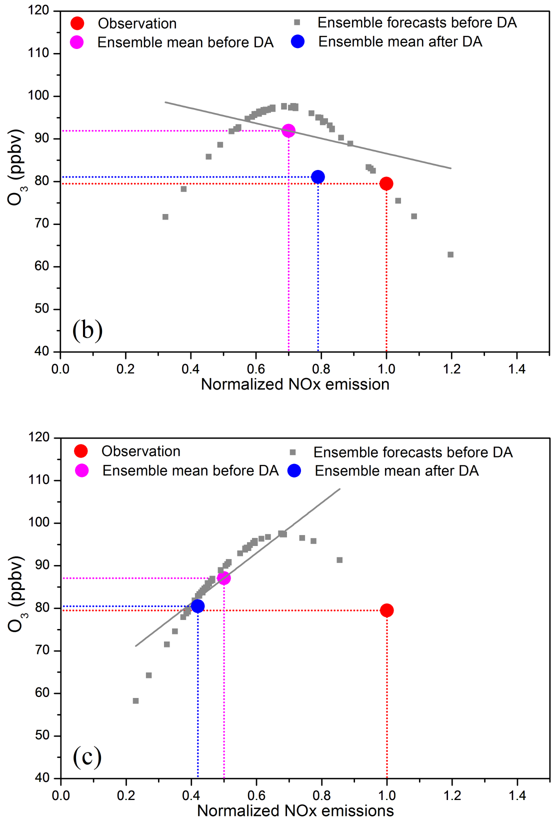


Figure 4 (a-c) O_3 concentrations (ppbv) and NOx emissions (no unit, normalized by the true NOx emission) before and after data assimilation (DA) and their ensemble samples before DA at 12:00 LT on August 12, 2008 in the three ideal ozone data assimilation experiments with the prior NOx emissions underestimated by 10% (a), 30% (b) and 50% (c) respectively. The grey squares denote the ensemble forecast O_3 concentrations corresponding to the perturbations of the NOx emissions (ensemble forecasts before DA), and the magenta dot represents the result of the ensemble mean of the grey squares (ensemble mean before DA). The gray line represents a linear relationship calculated from the ensemble samples of O_3 concentrations and NOx emissions. The red dot represents the true state of NOx emission and the observed O_3 concentration. The analyzed O_3 concentration and NOx emission are denoted by the blue dot.

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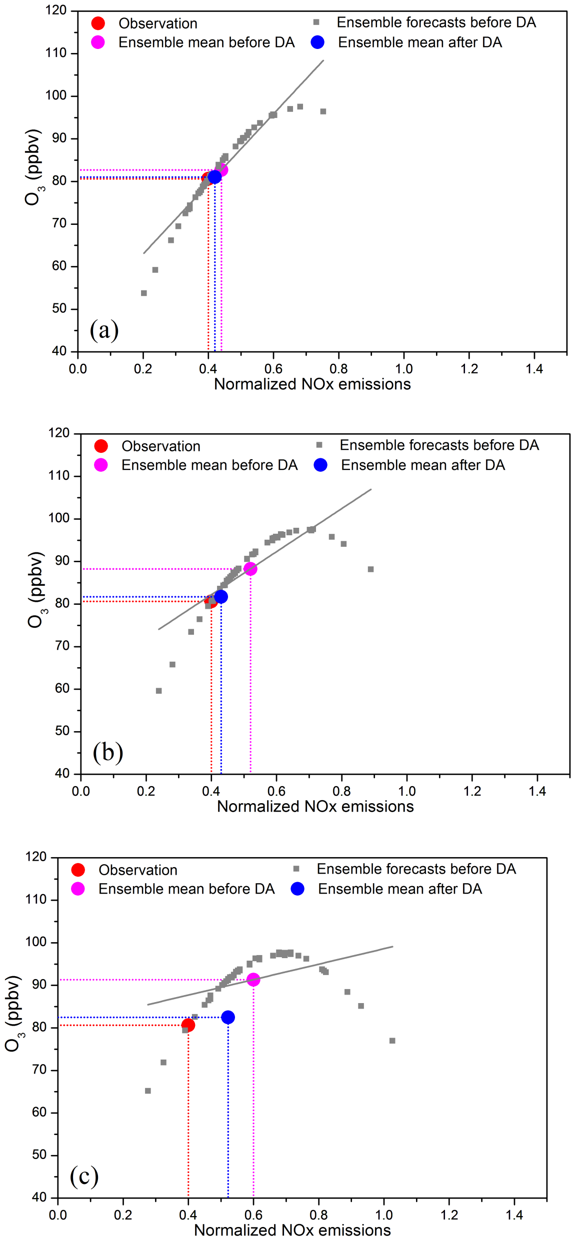

Observation
Ensemble forecasts before DA
Ensemble mean before DA
Ensemble mean after DA
O_3 (ppbv)
Normalized NOx emissions
(a)
(b)
(c)

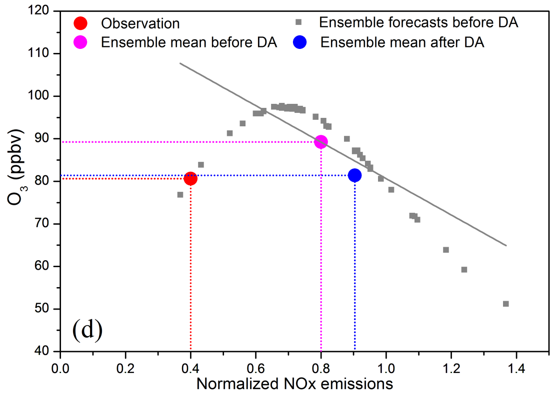
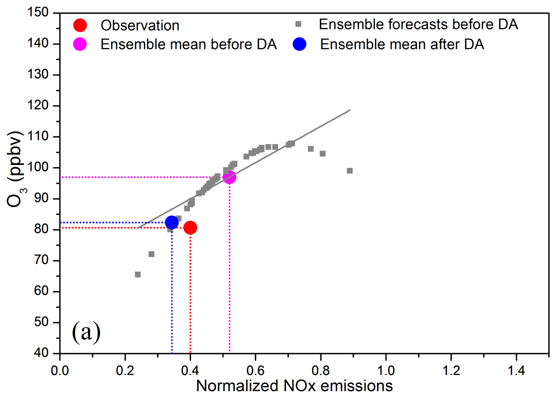


Figure 5 (a-d) O_3 concentrations (ppbv) and NOx emissions (no unit, normalized by the true NOx emission) before and after data assimilation (DA) and their ensemble samples before DA at 12:00 LT on August 12, 2008 in the four idealized DA experiments. (a) DA experiment with 10% overestimation in the a NOx emission estimation; (b) DA experiment with 30% overestimation in the a priori NOx emission estimation; (c) DA experiment with 50% overestimation in the a priori NOx emission; (d) DA experiment with 100% overestimation in the a priori NOx emission. The magenta dot, the gray squares, the gray line, the red dot and the blue dot represent the same as in Fig. 4.



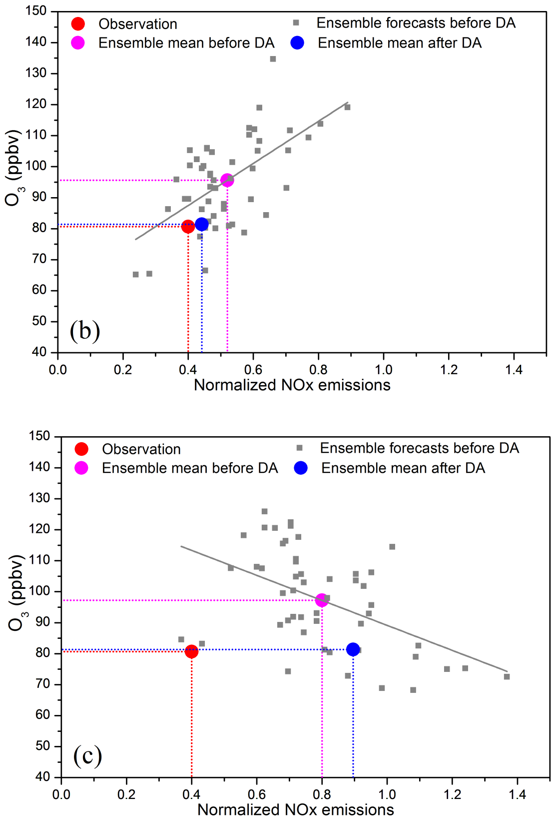


Figure 6 (a-c) O_3 concentrations (ppbv) and NOx emissions (no unit, normalized by the true NOx emission) before and after data assimilation (DA) and their ensemble samples before DA at 12:00 LT on August 12, 2008 in the three ideal DA experiments. The NO_2 photolysis rate is assumed to being overestimated by 20%. (a) The prior NOx emission is overestimated by 30% and adjusted by the DA, but the uncertainty of the NO_2 photolysis rate is missed (without perturbations on the NO_2 photolysis rate) in the DA. (b) The same as the DA experiment in (a), but the uncertainty of the NO_2 photolysis rate is taken into account through perturbing it. (c) The same as the DA experiment in (b), but the bias in the prior NOx emission is increased to 100%. The magenta dot, the gray squares, the gray line, the red dot and the blue dot represent the same as in Fig. 4.

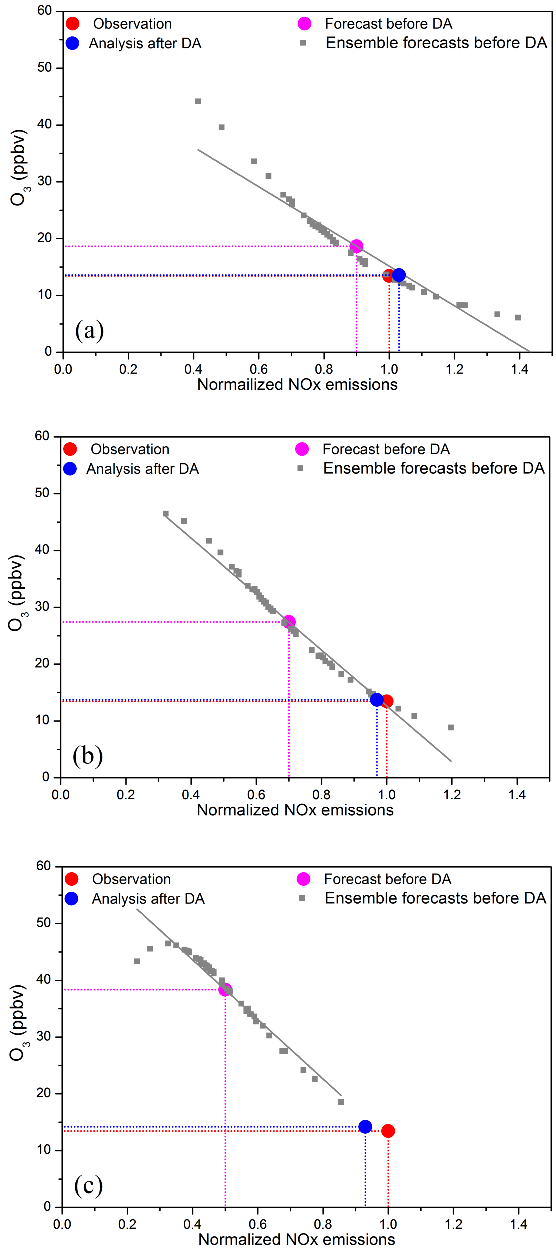


Figure 7 (a-c) O_3 concentrations (ppbv) and NOx emissions (no unit, normalized by the true NOx emission) before and after data assimilation (DA) and their ensemble samples before DA 08:00 LT on August 12, 2008 in the three ideal ozone data assimilation experiments with the prior NOx emissions underestimated by 10% (a), 30% (b) and 50% (c) respectively. The magenta dot, the gray squares, the gray line, the red dot and the blue dot represent the same information as Figs. 4.