Editor’s comments

Dear Wang and co-authors,

Your revised manuscript has now been reviewed by two referees. Both referees still have some major concerns on the revised manuscript, particularly regarding observational evidence for the dynamical feedback between aerosol and mixing height.

Both referees still have major concerns about the main conclusion of your paper that you show observational evidence for a dynamical control of aerosol on mixing height. The observations clearly show evidence that aerosol concentration and mixing height are related. The evidence that this is controlled through a dynamical feedback of aerosol on mixing height, rather than purely the control of mixing height on aerosol concentration, is weaker.

Before the manuscript can be accepted for publication we would need to see a convincing argument for this dynamical feedback based on your observational analysis. Please include this additional analysis in your revised manuscript, in addition to any comments in the response to review.

I look forward to receiving your revised manuscript.

Regards,

Dominick Spracklen

Dear Dominick,

We thank you very much for the patient and encouragement on this resubmission.

In this revised version, we did major revisions about the manuscript. Firstly, modeling results of surface relative humidity, temperature and mixing layer height from WRF model were added. The model results clearly indicated a vital role of aerosol direct radiative forcing in development of mixing layer height (Line 188-Line 201).Then, we added more discussions on aerosol-boundary layer feedback mechanism carefully before we ascribe our measured phenomenon as aerosol-boundary layer feedback (Line 236-Line 246). Finally, we explained more about our fitting results about PM2.5-MLH relationship (Line 216-222). Please see details for more information.

On the basis of our observed results, we hope the revised manuscript has addressed your and reviewers' concerns. Meanwhile, we are happy to revise the manuscript further, if you have more comments and suggestions.

Best wishes,

Yonghong Wang on behalf of all co-authors

A point to point response to the reviewers' comments

We thank the two reviewers for their comments, and we do think their comments and suggestions improved our manuscript a lot. Here are the points to points responses (in blue colored), accordingly, we also revised manuscript.

Report#1

The revised manuscript has addressed many of my minor concerns, but my principal criticism has not been addressed adequately yet. The manuscript provides valuable observational evidence for an inverse relationship between aerosol and mixing height, but this is not sufficient to demonstrate the presence of a feedback without further supporting evidence.

In their response, the authors reassert the existence of the dynamical feedback. The theoretical basis for this is sound, but the potential strength of this study is in providing observational evidence for it. The authors are too quick to demonstrate that the observations are consistent with the theory, and neglect the much more valuable goal of testing the theory based on the observations. This is evident in their response to my request for justification for fitting an exponential curve rather than a reciprocal. The authors present an interesting analysis in the response to reviewers, but have not provided a justification or made any changes to the manuscript. What relationship fits the observations best, and what implications does this have for the theory about a feedback?

Response: We thank you for the very important comments. Aerosol-boundary interaction and related feedback mechanisms have been subjected intensive studies due to the vital role in air pollution. Most of these studies were conducted by model simulation combined with some measurements data. In our study, we presented vertical measurements of aerosol concentration combined with mixing layer height with three years of measurement. The two measurements were connected by turbulent kinetic energy (TKE) obtained from 140 m observation plate to demonstrate aerosol-boundary layer feedback. In our opinion, the novelty of the work is the unique measurements with such a long date sets. Also, to the best of our knowledge, this is the first time we show a negative correlation about mixing layer height and TKE observed above surface, (as shown in Figure 5), which is a key process in aerosol -boundary layer feedback loop. We were able to quantify the feedback by compare increased $PM_{2.5}$ with the increased amounts of NOx. According to your comments, we did major revision on the manuscript: first, we added WRF model results during the intensive haze periods as we shown in Figure 1 to show the importance of aerosol in developing of mixing layer height. Basically, we conducted control experiment and test experiment by considering or without considering aerosol direct radiative forcing in the model. The results clearly showed that the consideration of aerosol direct radiative forcing into the model lead to decreased surface air temperature, increased relative humidity and suppressed development of mixing layer height in urban Beijing (Line 188- Line202). The second major revision was that we discussed aerosol-boundary layer feedback mechanism carefully before we ascribe observed phenomenon as aerosol-

boundary layer feedback (Line 236- line246).

Finally, we got an exponential curve rather than a reciprocal in our Figure 4 was that the reciprocal fitting overestimated the $PM_{2.5}$ concentration when the mixing layer height was very low compared to the exponential fitting function (Figure. 4), which also indicated that a much higher $PM_{2.5}$ concentration was needed in order to obtain a very low mixing layer height without the positive feedback. We revised this part and added more statements. (Line 216-Line 222)

The conclusions of the paper remain weak, as noted in my original review, but have not been altered in the revised version.

Response: we added more statements in our conclusion about the feedback mechanism as the reviewer commented. (Line 270-Line 277).

As I commented in my original review, the final sentence of the abstract needs revision: most good air quality models have included this feedback for many years (albeit without strong observational support, which this study could provide) so it is too late to "suggest that the feedback mechanism should be considered". The study just reconfirms that it should be considered. The changes made to the final paragraph of the introduction (l.80-85) have improved it and now more accurately summarise the findings of the study, but the abstract does not reflect this yet.

Response: We thank you for the comment, we revised the statement in our abstract. (Line 45,46)

L.183: As noted in my point above, the fitting of an exponential to Figure 3 still isn't explained. The new text at L.195 is not adequate to explain it. No reasoning or support is provided for ascribing the observed behaviour to a positive feedback (L.200-201).

Response: Thanks for the comment. We added more explanations about this part. (Line216-Line 222)

The English language is generally reasonable but needs further polishing before the paper is suitable for publication.

Response: We revised the manuscript carefully in this version, we hope it can meet the scope.

For an English Language journal it would be appropriate to replace the "D" and "G" in Figure S1 with "L" and "H" (the figure should be in English, not Chinese).

Response: Thanks for the reminder, we revised these symbols in the revised version.

A number of the references remain incomplete, e.g., missing journal for Wang et al. 2018, missing details/date for Petaja et al. and Ding et al., and formatting errors for several other references.

Response: Thanks for the corrections and we revised them in the current version.

The English in the title has been corrected, but in practice the title does not accurately reflect the topic or findings of the paper.

Response: We revised the title as ‘Rapid formation of intense haze episodes via aerosol-boundary layer feedback in Beijing’, which we hope it suitable for the paper.

Report#2

The main conclusion of this study is the two-way feedback between aerosol pollution and mixing layer height. However, neither the reviewer #1 nor I believed that the authors had provided extensive evidence to support this feedback mechanism in their previous manuscript. Unfortunately, the authors did not add any new data or appropriate analysis to validate their conclusion in the revised manuscript. Therefore, I cannot recommend the publication of this manuscript in its current version in ACP.

Response: Thank you for your comments, which motivated us to revise the manuscript considerable.

In this revised version, we added modeled results of surface relative humidity, temperature and mixing layer height from WRF model. The model results clearly indicated the vital role of aerosol direct radiative forcing in development of mixing layer height (Line 188-Line 201). Secondly, we added more discussions carefully on aerosol-boundary layer feedback mechanism before we ascribe our measured phenomenon as aerosol-boundary layer feedback (Line 236-Line 246). Thirdly, we explained more about our fitting results about PM2.5-MLH relationship (Line 216-222).

Finally but the most importantly, to the best of our knowledge, this is the first time that three years of the vertical air pollutants measurements were presented, combined with mixing layer height information and turbulent kinetic energy results, we do think our results provided here benefit current information of boundary layer- aerosol feedbacks in highly polluted urban cities.