Response to Anonymous Referee #3

Thank you very much for your time and constructive comments. Here are our responses to your comments.

1. Concerns regarding estimation of effective particle surface area (A_{es})

Estimation of available surface area (A_{es}, in this manuscript) for heterogeneous reactions is an ongoing challenge in the field. However, I have two broad issues related to the method (described in Section 2.4) used to estimate A_{es}.

The first is related to the use of the filter geometric surface area to approximate A_{es} at high particle loadings (P5721L10–13). From what I understand, the assumption here is that, in the "plateau regime", where Lf no longer changes with particle mass, the affective surface area can be approximated by the geometric surface area. However, in the Bedjanian et al. (ACP 2013) paper that's cited, it is explicitly acknowledged that the geometric surface area is a lower limit for the available surface area, and that uptake coefficients calculated using this value are thus upper limits. I don't think that it's reasonable to simply assume that, in cases where $Lf > Lf_c$, $A_{gs} = A_{es}$.

The second issue concerns differences between the experiments reported here and those reported in the Bedjanian paper (and in other papers looking at gas-dust interactions using flow tube techniques, Knudsen cell, etc.), and the appropriateness of applying the conceptual framework from these latter papers in this study.

In these other types of experiments, a linear increase in γ implies that the addition of more substrate results in a proportional increase in surface area available for uptake (whether the "entire" surface area is available for uptake is another issue entirely: see the Interactive Comment from Anonymous Referee #2 for the Bedjanian *et al.* paper), and a levelling off of γ at high sample masses implies that the gas-phase species can't access all of the sample over the timescale of the uptake experiment (i.e. that uptake is limited by diffusion into the lower layers of the "thick" sample).

In the present work, however, PAA and H_2O_2 are passing directly through the filters and thus theoretically should be passing through, or "accessing", all of the sample, even if it is "thick". So, the plateau regime in these experiments means something else entirely: to me, it implies an issue with particle agglomeration, which is becoming more prominent as more $PM_{2.5}$ is collected on the filter, i.e. that PAA/H_2O_2 aren't accessing the “interior” of the agglomerations. In my mind, this agglomeration should also be an issue, albeit a lesser one, at lower particle masses.

In short, I think that the “low-mass regime = linear”, “high-mass regime = plateau” is too simplistic a conceptual framework to employ here. Indeed, the linear fit to the left-most data points in Figure 2 is questionable at best.

On a more specific note, it isn’t clear to me how Equations 10 and 11 (P6722L56) were derived, since Lf wasn’t measured for ADS and ATD as a function of particles mass (from what I understand; only two masses were used). This requires explanation, especially if these A_{es} values were used to calculate for these samples.

In summary, I think that the paper would be greatly strengthened by a reevaluation of the approach used to estimate A_{es}. It might be useful to provide estimates of higher and lower bounds for this quantity, and then to calculate the range of associated γ values. In the end, all γ values should be accompanies by a description of the surface area used in their calculation.

A: As you point out, how to accurately estimate the effective surface area (A_{es}) for heterogeneous reactions is a challenge. Brunauer-Emmett-Teller (BET) surface area (A_{BET}) and geometric surface area (A_{gs}) are usually considered to be the upper limit and the lower limit for A_{es} respectively. The uptake coefficient (γ) estimated by the A_{gs} is several orders of magnitude higher than that by A_{BET} (Shen et al., 2013). In the field observations in Beijing, the lifetime of PAA is about 4.1−5.8 h in summer (Zhang et al., 2010; Liang et al., 2013). Taking OH radical reaction, photolysis and deposition into account, the lifetime of PAA is 44.2 h on haze days and 17.6 h on non-haze days, which is much longer than the observed results. Therefore, the heterogeneous reaction is potentially important to the removal of PAA. If we use the total surface area of $PM_{2.5}$ particles by size distribution (A_{ps}) to calculate γ, the corresponding average value of γ_{PAA} is $(1.10\pm0.56)\times10^{-5}$, and the estimated lifetime of PAA is 28.3 h on haze days and 16.6 h on non-haze days. Apparently, this estimated lifetime is 3−7 times of the observations. When using the method in this study to estimate A_{es}, the corresponding average value of γ_{PAA} is $(2.70\pm0.59)\times10^{-4}$, and the estimated lifetime of PAA is 3.0 h on haze days and 7.1 h on non-haze days, which fits well with the observed results. Therefore, although the estimated A_{es} in this study is a lower limit, it is consistent with the actual situation.

Besides, due to the loss of surface area during stacking, A_{ps} are not appropriate to estimate A_{es}. Assume there is no any other process (e.g. agglomeration and deliquesce) occurring during stacking, then we can use a simple cubic packing model to estimate the loss during stacking (Keyser et al., 1993):

$$A'_{ps} = \rho_b S_g (h_e + h_i) \tag{1}$$

$$S_g = \frac{6}{\rho_t d} \tag{2}$$

$$\rho_b = \frac{\pi}{6} \rho_t \tag{3}$$

$$h_e = \frac{d}{2} \tag{4}$$

$$h_i = d(N_L - \frac{1}{2}) \tag{5}$$

$$\mathrm{L} = \frac{A_{ps} - A'_{ps} A_{gs}}{A_{ps}} \times 100\% \tag{6}$$

where A'_{ps} is the total surface area of stacked $PM_{2.5}$ particles per unit geometric area, $cm^2\ cm^{-2}$; ρ_b is the bulk density, $g\ cm^{-3}$; S_g is the specific surface area, $cm^2\ g^{-1}$; h_e is the external thickness of $PM_{2.5}$ particles, cm; h_i is the internal thickness of $PM_{2.5}$ particles, cm; ρ_t is the true density of $PM_{2.5}$ particles, 1.42 $g\ cm^{-3}$ on haze days and 1.96 $g\ cm^{-3}$ on non-haze days; N_L is the number of $PM_{2.5}$ particle layers; L is the loss percentage.

The calculated L is about 22%. Besides, $PM_{2.5}$ particles in Beijing summer contains large portion of soluble inorganic components which are hygroscopic. (see Table 5 in the revised manuscript) shows the components of $PM_{2.5}$ particles in our study. It turns out that the concentration of SO_4^{2-}, NO_3^- and NH_4^+ ions take up 66.6% on haze days and 48.1% on non-haze days. The deliquescence relative humidity (DRH) is suggested to be 79% for $(NH_4)_2SO_4$, 39% for NH_4HSO_4 and 62% for NH_4NO_3 at 298K (Cziczo et al., 1997; Lightstone et al., 2000), and the DRH of $PM_{2.5}$ is even lower than that of the individual salt particles (Seinfeld and Pandis, 2006). Li and Shao (2009) used the transmission electron microscopy (TEM) observation to ascertain that even the most hydrophobic particles (e.g., mineral dust, some organics, fly ash, soot) can be coagulated or coated with hygroscopic components (e.g., nitrates, sulfates) on haze days.

Based on the relative humidity (RH) data during our sampling period, the average

RH during daytime is (63±9)%, ranging from 51% to 91%; and the average RH during nighttime is (83±8)%, ranging from 72% to 94% (website: http://classic.wunderground.com/). Under the condition of such high RH, $PM_{2.5}$ particles might agglomerate severely on the filter, and the gas is difficult to go through the internal layers. In summary, we suggest using the estimation method in this study to calculate the value of A_{es}. Although this A_{es} is probably close to the lower limit, it can fit well with the observation results. The quantification of A_{es} still has large uncertainty. How to estimate the precise value of A_{es} is still a challenge. More works are still needed in future researches.

Although the experiment method in this study is different from that used by Bedjanian et al., a mass-dependent uptake of peroxides was evidently observed. Specifically, the uptake of peroxides increases linearly with mass in low mass regime and is independent of the mass in high mass region.

The agglomeration of particles on the filter has a strong impact on A_{es} of particles for the uptake of peroxides. In the low-mass regime, agglomeration of particles is not important, and the uptake is linearly correlated with the particle mass. As the particle mass keep growing, the agglomeration of particles becomes gradually important, and the increase of A_{es} with the particle mass is less effective than that in the low-mass regime. When the particle mass reaches a critical mass, the particles agglomerate severely with each other, and the A_{es} of particles does not increase apparently with the particle mass but rather approach a plateau. Therefore, a conceptual framework for the estimation of A_{es} similar to the study by Bedjanian et al. can be applied to the present study.

We have measured the Lf of ADS and ATD as a function of mass, i.e., 0.18, 0.37, 0.81, 1.05, 1.16, 1.63, 1.86 and 2.46 mg for ADS and 0.27, 0.48, 0.83, 1.07, 1.36, 1.58, 1.76, 2.02, 2.57 and 3.00 mg for ATD. We have added these into the revised manuscript.

As per your suggestion, we have used A_{ps} of $PM_{2.5}$ particles to and A_{BET} of ADS and ATD particles to estimate the lower limit of γ on the corresponding particles. The results are listed in Table 3 (in the revised manuscript).

Scientific/technical comments and questions

1. P5716L23, were these aqueous solutions?

A: Yes, they are aqueous solutions. We have changed “solution” to “aqueous solution”.

2. P5717L10, how were the concentrations of PAA/H_2O_2 determined? Reference should be made here to Section 2.3.

A: H_3PO_4 solution (5×10^{-3} M) was used to scrub gaseous peroxides in a glass scrubbing coil. The collection efficiency was 85% for PAA and 100% for H_2O_2 (Hua et al., 2008; Liang et al., 2013). Then the peroxide-containing scrubbing solution was rapidly sent to an online high-performance liquid chromatography (HPLC) instrument. The method was described in detailed in Sect. 2.3.

3. P5718L6, how were the ADS particles collected?

A: ADS particles were collected at PKU campus in April 17, 2006 after a strong dust storm. The ADS particles deposited on a glass plate and then were collected and kept in a glass bottle. We have added this information into the revised manuscript.

4. P5718L8, a brief description of the resuspension apparatus (rotating brush generator?) should be provided.

A: We have added the description of resuspension apparatus in the revised manuscript as “The resuspension apparatus consists of three parts, i.e., glass inlet, stainless filter holder and vacuum pump. First, we put a known amount of mineral dust particles into the glass inlet and then turned off the inlet. Secondly, we turned on the vacuum pump and a negative pressure was then formed in this resuspension system. Finally, we turned on the inlet, and the particles were resuspended with the help of airflow and collected onto the filter”.

5. P5718L25, some information regarding the timescale/resolution of an uptake measurement should be provided here. If the RH was “continuously” increasing, then presumably the uptake values shown in Figure 3 were obtained over a range of RH? Or, was the RH stepped to the values shown in Figure 3, and then held at those values for long enough to achieve (and measure) a steady-state peroxide concentration (and thus steady-state uptake coefficient)? How often were measurements of peroxide concentrations made?

A: The uptake experiment at a certain RH took 2 h for PAA and 1 h for H_2O_2; including the time for the balance of peroxide on blank filter and particles-loaded filter. The balance concentrations of PAA/H_2O_2 have been detected at least for three times. The RH was then changed to a different value with no any treatment for the filter samples. The resolution of PAA and H_2O_2 concentration measurements corresponds to the HPLC retention time of PAA and H_2O_2, which is 8.9 min and 4.0 min, respectively, and the concentrations of PAA and H_2O_2 are measured every 10 min and 5 min, respectively. We have added a detailed description into the revised manuscript.

6. P5722L18-20, if the uptake coefficient is scaled to A_{es}, why would there be dependence on $PM_{2.5}$ mass?

A: We didn't state the uptake coefficients are dependent on $PM_{2.5}$ mass. What we said is that although the $PM_{2.5}$ mass varied significantly on haze days and non-haze days, there was no obvious difference between the uptake coefficients.

7. P5722L21, I would suggest plotting this empirical fit in Figure 4 rather than just connecting the mean uptake values (and would suggest the same for Figure 5).

A: Yes. We have revised it and combined the previous two figures into one (see Fig. 4 in the revised manuscript).

8. P5723L13-16, the El Zein et al. (JPCA) paper cited here reported an inverse dependence of the initial γ value, rather than the steady-state value, on relative humidity. In addition, the steady-state uptake coefficients reported in this study were calculated using BET surface areas. So I'm not sure how appropriate this comparison is from both a qualitative/quantitative perspective.

On this note, I would suggest including a summary table showing how the uptake coefficients/RH dependencies obtained in this study fit into those obtained previously.

A: We have deleted the comparison here. We have added a summary table showing the value of γ_{H2O2} and its RH dependence on mineral dust in literature data. Table 4 (in the revised manuscript) shows the summary of γ_{H2O2} and its RH dependence on mineral dust in literature data. Apart from γ_{H2O2} on authentic Gobi dust, authentic Saharan dust and aged particles, all γ_{H2O2} values show a negative RH dependence.

9. P5723L17, to me, it looks as though the samples obtained on haze days were much more variable than those obtained on non-haze days.

A: Yes, the $R_{\gamma PAA}$ values are more variable on haze days than those obtained on non-haze days. However, by taking an average, $R_{\gamma PAA}$ has no obvious difference at different times and under different weather conditions. We have changed as

"Although the $R_{\gamma PAA}$ values are more variable on haze days than those obtained on non-haze days, the average value of $R_{\gamma PAA}$ show no obvious difference at different times and under different weather conditions".

10. P5723L17-22, were uptake measurements of both PAA and H_2O_2 made on the same samples? Is it possible that previous exposure of samples to PAA could have influenced H_2O_2 uptake? If not, which samples were used for H_2O_2 uptake measurements? Or, were PAA/H_2O_2 uptake measurements made simultaneously? This needs to be clarified in the text.

A: Yes, the uptake coefficients of PAA and H_2O_2 were measured on the same $PM_{2.5}$ samples. We measured the uptake coefficients of PAA first, and then those of H_2O_2. We have compared the measured uptake coefficient of H_2O_2 on two $PM_{2.5}$ samples: one had been used to measure the uptake coefficient of PAA and the other had not been used for any measurements. The results showed that the relative error between γ_{H2O2} on these two samples was 0.97−7.37% at different RH (3−90%). Therefore, there is no obvious difference between the uptake coefficients of H_2O_2 on used and unused $PM_{2.5}$ samples. We have added a detailed description of this into the revised manuscript.

11. P5724L4-6, I don't see how the larger value of γ for H_2O_2 at lower RH implies a physical process.

A: Thank you for pointing out this. In this study, we suggest that chemical process dominate the uptake of peroxide rather than physical process and $R_{y,PAA}$ is larger than $R_{y,H2O2}$. However, γ_{H2O2} at low RH is larger than γ_{PAA}, we once considered this might suggest the importance of physical process in low humidity. Now, we find this statement is not appropriate and we have deleted it in the revised manuscript.

12. P5724L9-10, what do the error bars in Figure 3 represent, exactly? If there are only two data points (ascending and descending RH values), I think that the spread in data would be better represented using different symbols for the ascending/descending γ values.

A: These error bars represent relative standard deviation of γ_{PAA} values on $PM_{2.5}$ with the ascending and descending RH. In the revised manuscript, we have used different symbols for γ values obtained with ascending and descending RHs (see Fig. 3 in the revised manuscript).

13. P5725L1, again, what do these errors represent? In addition, these values appear to be for ADS_l. The PAA uptake coefficient for ADS_h at 3% RH is higher than that for ADS_l, while the opposite is true for ATD (i.e. the uptake coefficient for ATD_l is higher than that for ATD_h). If these uptake coefficients are scales to A_{es} (or some proxy for it), then why are the values for high/low loadings so different?

A: These errors represent standard deviation of γ on mineral dust of the ascending and descending RH. The difference of γ between ADS_l, ADS_h and ATD_l, ATD_h are mainly caused by two reasons: the uncertainty of the A_{es} estimation and the experimental error. We have calculated the uptake coefficients of peroxides on ADS and ATD by the BET area to represent its lower limit in the revised manuscript (see Table 3).

14. P5725L18, "The value of γ_{H2O2} on ADS changed from $(1.25 \pm 0.5) \times 10^{-4}$ at 3% RH to $(4.54 \pm 0.12) \times 10^{-4}$ at 90% RH". I don't see this trace on Fig.7 at all! In addition, the presentation of values for both "high" and "low" dust loadings is confusing in the absence of explanation of differences/similarities between results obtained at the surface of these samples.

A: This value is the average of γ_{H2O2} values at low and high mass loading. We have changed it to "By taking the average of γ values at low and high mass loading, γ_{H2O2} on ADS increases from $(1.10 \pm 0.31) \times 10^{-4}$ at 3% RH to $(2.44 \pm 0.69) \times 10^{-4}$ at 90% RH". Although the values of γ_{H2O2} at low and high mass loading are not identical, all γ_{H2O2} values on ADS show a positive correlation with RH, and all γ_{H2O2} values on ATD show a negative correlation with RH. We have added this information and an explanation of the differences/similarities of γ values into the revised paragraph.

15. P5727L7, does this value represent a bulk pH? It may be possible that some individual particles are basic, and that measured uptake to $PM_{2.5}$ is an average value that includes contributions from both unreactive and reactive particles types.

A: Yes, this value is a bulk pH. We have measured compositions of $PM_{2.5}$ (see Table 5 in the revised manuscript). The concentrations of acidic ions such as SO_4^{2-} and NO_3^- make up 60.9% of $PM_{2.5}$ on haze days, and 41.3% on non-haze days. Even though there are some basic components, we believe they are already neutralized or acidified. However, in any cases, the measured uptake to $PM_{2.5}$, as you pointed out, is an average value that includes contributions from both unreactive and reactive particles types.

16. P5728L1-2, is there a reference for this?

A: Thank you for pointing out this. We have added a reference to the revised manuscript.

17. P5728L14, the Rubasinghege et al. paper looks at the acid-induced dissolution of α-FeOOH in solutions (PH 2) of HNO_3, HCl, and H_2SO_4, and investigates the effect of the anion on dissolution. Would these effects still be valid at the pH values in the aerosol samples used in these experiments?

A: Rubasinghege et al. (2010) have pointed out that in the presence of light, acids with pH=2 promote iron dissolution. While in the dark, the total iron dissolution is related to the strength and coordination mode of anions. Because sulfate and nitrate are both polyatomic oxyanions, they have bidentate and bridging coordination mode, and both of these modes can enhance iron dissolution. In our study, we detected high concentration of SO_4^{2-} and NO_3^- in $PM_{2.5}$ and ADS particles, and our study was carried out in the absence of light. Therefore, we think the presence of SO_4^{2-} and NO_3^- in $PM_{2.5}$ and ADS particles can potentially enhance iron dissolution.

We have changed the sentence “anions can enhance the dissolution of Fe cation” to “anions can potentially enhance the dissolution of Fe cation”.

18. P5729L8-10, I think that this statement is too definitive, and should be qualified somewhat, since no specific direct evidence for each of these pathways exists.

A: We have added “probably” into this statement to make it less definitive.

19. P5731L16-20, I am not sure that I understand the purpose of this passage. Presumably these larger particles are of crustal origin and contribute to PAA uptake?

A: The estimation of mean diameter of $PM_{2.5}$ is based on the literature. This literature only measured the size distribution of PM_1. So, this passage aimed to evaluating the contribution of PM_1-$PM_{2.5}$ to the total mean diameter of $PM_{2.5}$.

Minor typos/usage errors

1. P5714L6, “the aerosols” should perhaps read “aerosol-phase processes”?

Also, it’s not entirely clear to me from this sentence what the field observations suggest: that PAA is important? That the PAA budget is potentially influenced by aerosol processes? I’m assuming you mean the latter, but the sentence should be revised to make this clearer.

A: We have changed “the aerosols” to “aerosol-phase processes”. The field studies have suggested that PAA is important organic peroxide with typical concentration of several tens to hundreds of pptv, and its budget in the atmosphere is potentially influenced by aerosol processes. We have rewritten the sentence to make it clearer.

2. P5714L7, “onto the ambient” should read “onto ambient”

A: We have revised it.

3. P5714L9, “at the uptake coefficient” should read “with an uptake coefficient”

A: We have revised it.

4. P5714L14, I would specify that mineral dust is one of the main components of $PM_{2.5}$ “in this region”

A: We have changed the previous sentence to “Considering that mineral dust is one of the main components of $PM_{2.5}$ in Beijing”.

5. P5715L10-15, to my understanding, the Mao et al. and Liang et al. articles cited here focus primarily on the influence of HO_2 uptake (and, specifically, on HO_2 uptake mechanism that don’t produce H_2O_2). I think that inclusion of these papers-and, by extension, the transition metal-catalyzed HO_2 uptake story- here is somewhat of a distraction.

A: We have deleted the Mao et al. and Liang et al. articles here.

6. P5715L15-20, with reference to the previous comment, what do these studies show regarding the relative contribution of direct H_2O_2 uptake to H_2O_2 concentrations (i.e. vs. HO_2-mediated H_2O_2 loss)? I think that it would make sense here to more clearly describe the conclusions of the studies that have “ascertained the importance” of this process.

A: Thank you for your constructive suggestion. We have revised it as “a series of laboratory studies have addressed the importance of the heterogeneous reaction of H_2O_2 on model or authentic mineral dust particles (Pradhan et al., 2010a; Pradhan et al., 2010b; Wang et al., 2011; Zhao et al., 2011a; Zhao et al., 2011b; Romanias et al., 2012; Zhou et al., 2012; Romanias et al., 2013; Zhao et al., 2013; El Zein et al., 2014). For example, Pradhan et al. (2010a) have indicated that the heterogeneous reaction of H_2O_2 on dust aerosols could compete with its photolysis and significantly affect HO_x radical budget. Romanias et al. (2012, 2013) have confirmed that the heterogeneous reaction of H_2O_2 on mineral dust had an important effect on the fate of HO_x radicals. El Zein et al. (2014) also suggested that the lifetime of H_2O_2 removed by heterogeneous reaction was comparable with its photolysis on severe dust storm period. Our recent study has indicated that H_2O_2 could enhance the uptake of oxygenated volatile organic compounds (OVOCs) onto the surface of mineral dust particles (Zhao et al., 2014)".

7. P5715L16, "on the model" should read "on model"

A: We have revised it.

8. P5715L28, this is too broad: which temperate areas?

A: We have changed "temperate area" to "Mazhuang, a rural site in Shandong Province, China".

9. P5716L1, "that the heterogeneous" should read "that heterogeneous", i.e. "the" is not necessary.

A: We have deleted "the".

10. P5716L3-4, this sentence is awkwardly phrased, and should perhaps be revised to more clearly indicate that PAA is used as representative organic peroxide.

A: We have rewritten this sentence as "Therefore, we undertake PAA as representative organic peroxide to investigate its kinetics and discuss its mechanisms of the heterogeneous reactions on ambient $PM_{2.5}$ as well as mineral dust particles over a wide range of relative humidities (3–90%)".

11. P5716L9, should read "kinetics of H_2O_2 uptake on $PM_{2.5}$"

A: We have revised it.

12. P5716L10, "to ensure the constant" should read "to ensure a constant"

A: We have revised it.

13. P5717L15, "particle loaded" should be read "particle-loaded"-there are also a number of other instances in the manuscript where hyphens are missing from compound adjectives

A: We have revised them.

14. P5717L17, "PKU located" should read "PKU is located"

A: We have revised it.

15. P5718L9, “sampled in” should read “sampled on”

A: We have revised it.

16. P5718L14, “gaseous peroxide” should read “gaseous peroxides”

A: We have revised it.

17. P5718L19-22, I think it would be clearer if this were places after equation 4, since, from what I understand, $[C]_{in}$ in this equation was approximated by $[C]_{out}$, blank.

A: We have moved this sentence to the place after equation 4.

18. P5720L7, “at the flow rate” should read “at a flow rate”

A: We have revised it.

19. P5720L14, “challenge for the γ determination” should be read “challenge for the determination of γ”

A: We have revised it.

20. P5721L22, should this be 4.89×10^{-1}?

A: Yes, we have corrected it.

21. P5722L15-17, this sentence is unclear and should be rephrased.

A: We have rephrased this sentence to make it clearer.

22. P5723L14, “on the ATD” should read “on ATD”.

A: We have deleted “the”.

23. P5726L5, “to the physical processes” should read “to physical processes”.

A: We have deleted “the”.

24. P5728L12, “for the aqueous reaction” should read “for aqueous reactions”.

A: We have deleted “the”.

25. P5728L15, “by the Fe catalysis” should read “by Fe catalysis”.

A: We have deleted “the”.

26. P5728L16, the Mishra reference is missing``but I don’t necessarily think that its

conclusions are valid for application here, since PAA/H_2O_2 are weak acids.

A: Thank you for your constructive suggestion. PAA and H_2O_2 are weak acids, so their dissociation is quite limited. We have deleted this citation.

27. P5730L5, “The field observations” should read “Field observations”.

A: We have revised it.

28. P5731L15, what value was used for the mean diameter of the total particles?

A: $\overline{Dp}$ is 114.6 nm for haze days $PM_{2.5}$ particles and 62.4 nm for non-haze $PM_{2.5}$ particles. We have added it into the revised manuscript.

29. P5732L16, “the urban area” should read “urban areas”

A: We have revised it.

30. P5732L21, “from the urban area” should read “from urban areas”; “affecting the oxidants” should read “affecting oxidant”.

A: We have revised them.

31. Figure 5, the y-axis should be expanded (i.e. it should start at zero).

A: We have revised it.

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