General Comments

The authors have made a modest effort to address the concerns raised in previous comments. They have suitably addressed concerns regarding the suitability of the MARGA HONO measurement from a quality assurance perspective and conservatively corrected their dataset for further analysis. They have also simplified their data analysis to exclude confounding factors introduced by photolytic loss of HONO by reducing their analysis to nocturnal observations. However, major issues remain to be addressed concerning the presence of directly emitted HONO and competitive heterogeneous formation on the ground surface in this observational dataset and a more general lack of specificity when discussing their results.

Response: First of all, we would like thank the referee for the constructive and detailed comments, which did help us to improve the manuscript a lot. In the revised manuscript, we estimated the contribution of direct emission from BB to the observed HONO concentration, and discussed the possible role of ground surface. And we also revised the manuscript according to the specific comments. The modifications in the revised manuscript were highlighted in blue color.

Major Comments

Exclusion of Direct HONO Emissions and Ground Surface HONO Production in Observations

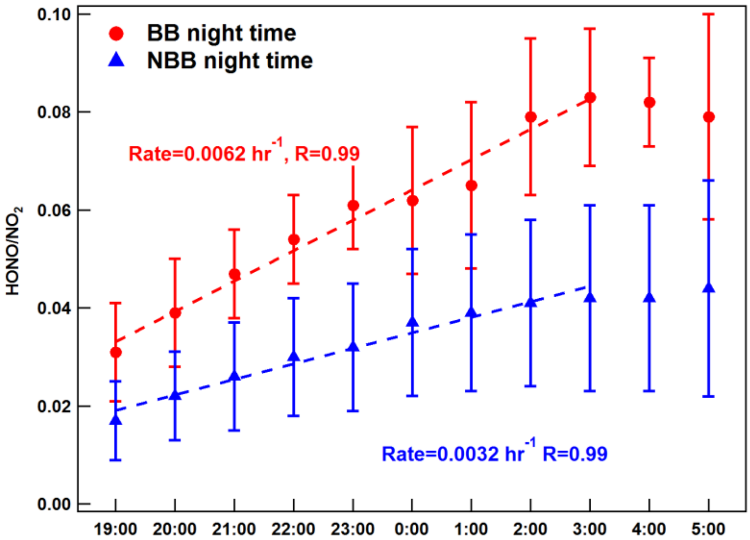
This topic of the paper has not been sufficiently addressed by the authors' responses. In many cases they have misleadingly directed substantial comments to a single response wherein they estimate the nocturnal lifetime of HONO that has significant weaknesses. The chemical lifetime calculations presented are the right direction for discerning the relative role of aerosol HONO production versus other production and loss processes. Unfortunately, the lifetime limiting nature ascribed to HONO loss on aerosol surfaces is inconsistent with the literature and physical properties of this weak acid. The authors use an uptake coefficient of 10-3 for HONO onto aerosols despite wide reports in the modern literature that this is an unlikely process on secondary inorganic or organic aerosols[*Kleffmann et al.*, 1998; *Stemmler et al.*, 2006]. The work of Wong et al.[*Wong et al.*, 2011] - cited by the authors - used a tuneable aerosol uptake to achieve agreement between their model and observations. Uptake to aerosol at such a magnitude was not directly observed, but based on HONO uptake to wet surfaces, not aerosols, nor aerosol proxies[*Wong et al.*, 2011]. The value of 10-3 is also well above those reported for HONO uptake to soil[*Donaldson et al.*, 2014] and ground[*VandenBoer et al.*, 2013] surfaces, which have greater surface areas, higher pH values and water content[*Su et al.*, 2011], and potentially large reservoirs of reactive components in comparison to biomass burning aerosols. Reasonable values for this process could range from 10-5 to 10-4 if one assumes BB aerosols are similar to soil surfaces. Given that soils are much different in composition, the uptake and loss of HONO onto BB aerosols seem more likely to be well below 10-5 from the best available investigations in the literature. Furthermore, conversion of NO2 on a variety of aerosol surfaces suggests that HONO is not retained on these substrates, but rapidly partitions to the gas phase[*Bröske et al.*, 2003; *Finlayson-Pitts*, 2009; *Finlayson-Pitts et al.*, 2003; *Kleffmann et al.*, 1998]. Given the much lower, or potentially not existent, uptake of HONO to aerosols would result in a longer HONO lifetime after direct BB emissions at night compared to the value the authors calculate. Thus, the 1.5 hour lifetime of HONO is underestimated by at least a factor of 2. This means that the dominant HONO loss process at night by their calculations is instead dry deposition.

Dry deposition loss of HONO to the ground surface is a nocturnal loss process that would limit direct emissions from influencing their measurements, but the lifetime is about 3.5 hours by the presented estimate and assumes the plume is always in contact with the surface. Regardless, this lifetime is certainly within the transport distance of the fire plumes presented in this work, with detectable amounts of HONO easily transported from up to 8 hours upwind, which is nearly the entire duration of the night. More likely, given the convective processes of a fire, the plume is aloft for part of the transit and HONO is not lost by dry deposition at all, giving it an even longer lifetime of up to 55 hours from the OH loss estimate presented. One concern that is in particular need of addressing is that NO2 would be converted to HONO on these same ground surfaces during transport when in contact with the surface on the way to their observation site. This is a HONO production term that is easily obtained from the literature via a dry deposition term and conversion efficiency and should be included in the calculation of production terms for these observations. Given that the nocturnal HONO lifetime is at least 3.5 hours, the authors certainly need to consider NO2 conversion on the ground in their analysis by considering the relative contributions of the ground surface production of HONO or provide a clear acknowledgement that they are unable to exclude this source in their analysis and that the conversion of NO2 to HONO on BB aerosols may be one of several processes contributing to their observations.

Finally, the potential utility of CO data and exploration of HONO/CO in comparison to the literature values for direct biomass burning (BB) emissions have not been attempted this analysis and would be strong data to include in the arguments for or against directly emitted HONO observed at the SORPES site. Also, the use of K+ as a biomass burning tracer is neither stated as an accepted tracer, nor appropriately referenced in the manuscript. Demonstrating that K+ correlates well with CO or another BB-specific tracer would suffice in the usage of K+ to track BB plumes and would support additional analyses presented using the K+ selection criterion for BB events. When used in conjunction with the nitrate/NOy chemical aging clock, this analysis would also strengthen their ability to identify the potential contributions of direct HONO BB emissions in this dataset.

Response: We really thank the reviewer for this specific comment, which could be separated into three issues. The first one is the life time calculation of HONO during the night time. The second one is that if the K^+ is a suitable tracer of BB. The third one is the role of ground surface to the observed HONO concentrations during BB periods.

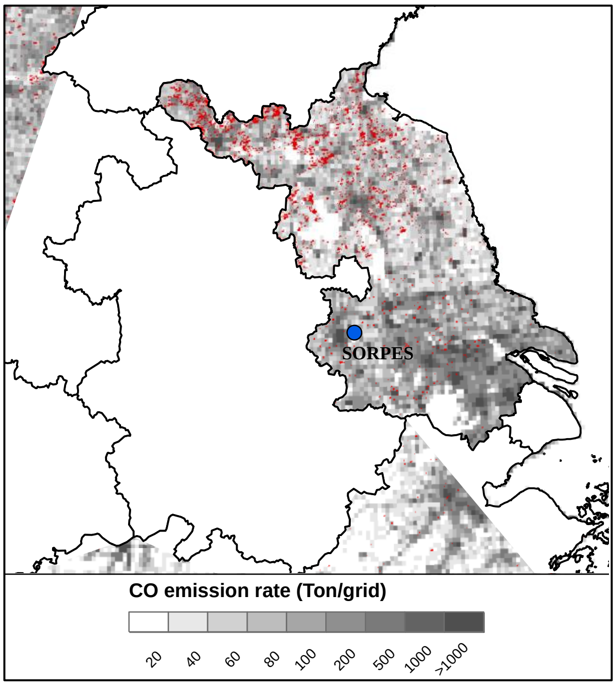
First of all, we do not think the heterogonous production of HONO on BB aerosols is the only source to the observed HONO concentration during BB periods, but probably is the major contributor to the enhancement of HONO concentrations during BB period. In the revised manuscript, we estimated the contribution of BB emission to the observed HONO using the method of K^+ tracer. We also added a new figure (as follows) to describe the difference of the HONO-to-NO_2 rates during nighttime, which can further demonstrate the higher potentials of BB aerosols to convert NO_2 to HONO. We rewrote this part in the revised manuscript.



HONO/NO_2 ratios during nighttime of both BB and Non-BB samples. Error bars are the standard deviations.

For the first question of HONO life time calculation, we agree with the reviewer's comment that we made an un-reasonable estimation in last manuscript. We re-calculated the HONO life time, and changed the description in the manuscript. Given that only the nighttime dataset was used in this work, the boundary layer during the selected periods should be low and stable, and the air masses inside the boundary layer and in the free troposphere were difficult to be exchanged. That means the BB plumes we observed during the nighttime always transported in the boundary layer, and probably contact to the ground surface. Therefore, we took the deposition of HONO on ground into consideration in the calculation of HONO life time. In such case the calculated nighttime HONO lifetime is 3.3 hours.

For the referee's suggestion that to calculate the accurate transport time of BB plumes from source regions to SORPES station, it is really a difficult job due to 1) the transport of air plume is actually an issue of gas diffusion, which is hard to define the beginning time and ending time; 2) the exact source region (fire point on the map) is hard to identified. Some episodes maybe influenced by several source regions on the transport pathway. But we believe it need quite a long time before the BB plumes transporting to SORPES station as 1) there was few fire points which were very close to the station; 2) the air plumes of several episodes, such as 9-11 June and 12-13 June, have been demonstrated transporting several days before arriving the station (Fig. 9d and 9e in Ding et al., 2013). Therefore, in the revised manuscript, we assumed the mean transport time were about 3.3 hours (the lifetime of HONO during nighttime) by BB can arrive SORPES station, which actually overestimated the contribution of BB for most episodes).



Fire points and CO emission map of YRD in June of 2012.

For the second question that if potassium ion (K^+) is a suitable tracer for biomass burning episodes. We agree that BB can emit some amount of CO. And CO can be a BB tracer in the region with few other CO sources. However, SORPES station is located in YRD, which is one of the best developed and most polluted regions. There are many CO sources other than BB, such as the power plant, industry and traffic (as showed in the figure above). These sources can contribute a lot for the CO loading even during the BB season. Instead, K^+ is well recognized and widely used BB tracer in aerosol phase (ANDREAE, 1983; Ma et al., 2003; Reid et al., 2005). And moreover, there are no other significant sources of K^+ around this region. Therefore, we believe that K^+ is a more suitable tracer for this work. In the revised manuscript, we added a new section and several references to address this point.

For the 3rd issue that if the ground surface plays any role in the observed HONO concentration during BB periods. Actually, we cannot totally exclude the influence of the ground surface to the HONO concentration. As it is difficult to ensure the exact transport pathway of different plumes and the land use/ land cover information, we cannot make an accurate calculation of ground surface to HONO concentration (both the emission and heterogeneous reaction on the ground surface). But we try to calculate the "footprint" of the plumes during their 8 hour's (during which the emitted HONO can be total consumed) transport before arriving SORPES station. And the results showed the possibility of air masses contacting to the ground surface of BB plumes was even 10 percent higher than Non-BB plumes, indicating that the ground surface did not play a key role in the observed enhancement of HONO concentrations during BB periods. In the revised manuscript, we state that we cannot totally get rid of the influence of ground surface, but the results tend to support the heterogeneous reaction of NO_2 on the surface of BB aerosols were the major contributors to the observed increase of HONO concentration during BB periods.

Reference

ANDREAE, M. O.: Soot Carbon and Excess Fine Potassium: Long-Range Transport of Combustion-Derived Aerosols, Science, 220, 1148-1151, 10.1126/science.220.4602.1148, 1983.

Ma, Y., Weber, R. J., Lee, Y. N., Orsini, D. A., Maxwell-Meier, K., Thornton, D. C., Bandy, A. R., Clarke, A. D., Blake, D. R., Sachse, G. W., Fuelberg, H. E., Kiley, C. M., Woo, J. H., Streets, D. G., and Carmichael, G. R.: Characteristics and influence of biosmoke on the fine-particle ionic composition measured in Asian outflow during the Transport and Chemical Evolution Over the

Pacific (TRACE-P) experiment, Journal of Geophysical Research: Atmospheres, 108, 8816, 10.1029/2002JD003128, 2003.

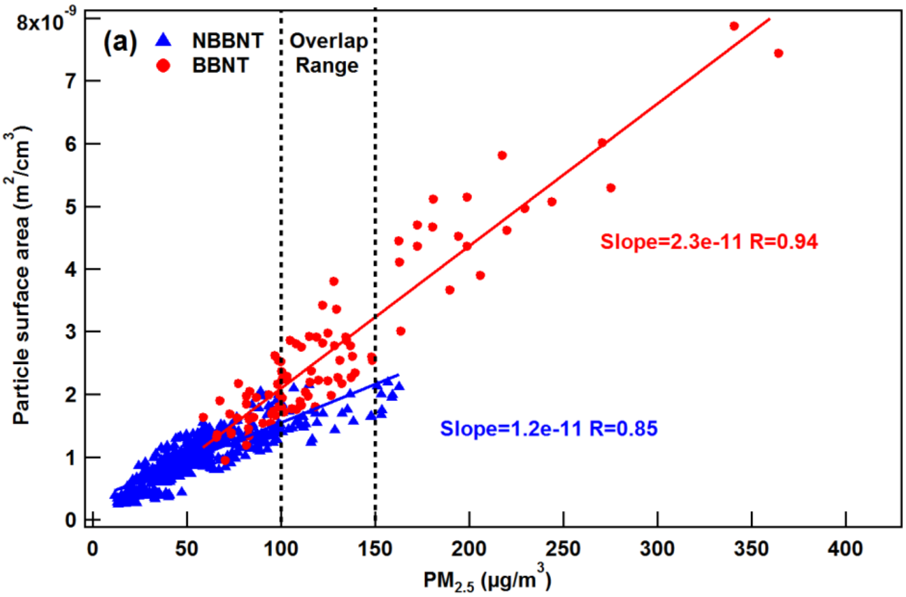
Reid, J. S., Koppmann, R., Eck, T. F., and Eleuterio, D. P.: A review of biomass burning emissions part II: intensive physical properties of biomass burning particles, Atmos. Chem. Phys., 5, 799-825, 10.5194/acp-5-799-2005, 2005.

Ding, A. J., Fu, C. B., Yang, X. Q., Sun, J. N., Zheng, L. F., Xie, Y. N., Herrmann, E., Nie, W., Petäjä, T., Kerminen, V. M., and Kulmala, M.: Ozone and fine particle in the western Yangtze River Delta: an overview of 1 yr data at the SORPES station, Atmos. Chem. Phys., 13, 5813-5830, 10.5194/acp-13-5813-2013, 2013.

Figures with Induced Dependence

Of significant concern are the plots made for the second reviewer of HONO/NO2/surface area vs K+/PM2.5. These plots demonstrate surface area vs K+/PM2.5 are likely dependent on each other, yet the authors present two new figures with the exact same issue in the manuscript: i) surface area/PM2.5 versus PM2.5, and ii) HONO/NO2/surface area versus surface area. The presentation and interpretation of the data in these figures needs to be drastically reconsidered. In each case, the data of interest have been transformed as a scalar multiple on a log-log plot of 1/x vs x, which will yield a negative correlation even when spurious data is used to scale the relationship.

Response: we agree with referee's comment. In the revised manuscript, we replaced Fig 8a to the plot of particle surface area versus $PM_{2.5}$ mass loading (as follows), which can also support the conclusion that BB aerosols have a higher specific aerosol area.



We removed the Fig. 10a, and rewrote this part in the revised manuscript. What we want to show is the difference of the NO_2 conversion efficiency (which can be represented by HONO/NO_2/surface area) between on BB aerosols and on Non-BB aerosols. However, in the ambient air, the HONO/NO_2 ratio is balanced to both ground surface and aerosol surface. So the comparison should be aided for the ratios of (HONO/NO_2) / (ground surface + aerosol surface) of the balanced samples to see the differences between BB and non-BB aerosols. In this case, if we assume the same area of related ground surface for both BB and non-BB plumes, the ratios of (HONO/NO_2) / (ground surface + aerosol surface) can only be compared when the aerosol surface areas of BB and non-BB aerosols are the same. This is why we chose the overlap part of the surface area to make the comparison.

As the areas of ground surface are unknown, and assumed their values are at the same level for BB and non-BB plumes, the differences of (HONO/NO_2) / (ground surface + aerosol surface) can be represented by the ration of (HONO/NO_2) / aerosol surface. Therefore, we compared the values of (HONO/NO_2) / aerosol surface to instead the NO_2 conversion efficiency.

Use of Correlative Statements

Throughout the manuscript the authors rely on correlative analyses to interpret their data, but they consistently do not report their statistical approach (e.g. linear least squares fit, orthogonal least distance fit, etc.) or the resulting statistics of that analysis (e.g. R2, slope, etc.) that are required to make statements about 'significance'. In particular, the use of orthogonal least distance or a weighted linear regression that accounts for the error in both datasets under comparison must be used for the relationships to be properly assessed.

Response: We agree with the referee's comments, and added all the needed statistical results in the revised manuscript, including the R, slope and the t-test results for the comparison. We also removed the word "significant" or "significance", if there is no statistical result.

Specific and Technical Comments

General Lack of Specificity

The authors rely on qualitative descriptions of their data throughout the manuscript when the quantitative values would greatly improve the impact of this work. Many of these are noted in detail below, but all identifiable locations for this improvement should be attempted.

Response: We really thank the reviewer for his/her patience to give so many specific and helpful comments. We revised the manuscript accordingly, and try to present the quantitative values at all identifiable locations.

English Language

The authors should have the revised manuscript thoroughly reviewed for typos and grammar by a native English speaker with particular attention paid to the use of the words 'the' and 'significantly'. The latter requires the use of a statistical test and many other wording choices can better describe the data presented.

Response: We thank the referee again for kindly and helpful comments, which do help us to largely improve the manuscript. We revised the manuscript thoroughly for the English language issues by an English native speaker.

Lines 5-6: An example of where the authors can be far more specific. Inclusion of the range, max, and min in addition to the mean is more informative.

Response: We have added the range of ambient HONO concentration.

Lines 6-8: Provide quantities and statistical results here that demonstrate the significance.

Response: Agree, we have added the statistical results in both the abstract and section 3.1 in the revised manuscript.

Lines 9-11: NO2 is also directly emitted from BB processes. This is not a sound conclusion to be making. Reconsider the wording of this sentence.

Response: Agree, we have changed the description of sentence and added the estimated result of HONO contribution from BB emissions. For detailed information, please refer to the response of the major comments.

Line 27: O3 is given before defined as ozone (line 29). Define with the written word at first use, as has been done with the other compounds above.

Response: Agree and thanks for pointing out these errors. We have double checked them in the revised manuscript.

Line 32: Su et al.[*Su et al.*, 2008] simulated soil pore water release of HONO, not the ground surface. Correct this statement.

Response: Agree and have corrected.

Line 43: There is an 'ö' in Sorgel. Update any misspelled author names throughout the referencing.

Response: Agree and thanks for pointing out these errors. We have double checked them in the revised manuscript.

Lines 49-50: The debate is showing that there is not a single process that dominates.

This sentence should be changed to reflect the mechanistic variety by which HONO can be made heterogeneously in the atmosphere.

Response: Agree, and have changed the description in the revised manuscript.

Line 58: The authors state that there are 'other sinks'. What are the other sinks? The must be important or the dilution argument could also be applied to aerosols. I suspect that the only sink of concern in a BB plume is photolysis, with deposition more important when plumes reach the surface.

Response: Agree and have changed "other sinks" to "photolysis".

Line 81: Correct this to 'Field'.

Response: Have corrected in the revised manuscript.

Line 95: chromatography chromatograph

Response: Have corrected in the revised manuscript.

Line 96: An example of where the exact number is more informative that 'more than 1500 samples'. Simply provide the exact number of samples collected for the dataset that was analyzed.

Response: Agree and have provided the exact number (1608) in the revised manuscript.

Line 111: Was the NOy molybdenum converter used in any way to correct for the interference given in the previous sentence? Why mention the interference of NOy in the NO2 measurement and then not be clear about how you did or did not account for that possibility?

Response: Molybdenum converters (MC) are used in both NO_2 and NO_y analyzers. The difference is the location where the molybdenum converter is installed. For the NO_2 analyzer, the MC is placed inside the analyzer (at the end of the sampling tube). For NO_y analyzer, the MC is placed outside the analyzer (at the beginning of the sampling tube).

According to previous studies, the interference of NO_2 measurement induced by the molybdenum is obvious at daytime with strong photochemical smog but is minor and can be ignored at nighttime. We added this statement in the revised manuscript.

Line 124: A reference needs to be provided here.

Response: Agree and have added.

Line 126: Delete ‘aroused by this interference’. It is unnecessary and confusing.

Response: Agree and have deleted the words in the revised manuscript.

Line 139: measurement   measured

Response: Agree and have changed.

Line 140: Delete ‘mainly’, the descriptor ‘tend’ has already been used. Also, change ‘deployed’ to ‘used’

Response: Agree and have changed.

Line 145: depression   deposition

Response: Agree and have changed.

Lines 145 – 160: The entirety of section 2.4 needs to be carefully rethought, the assumptions clearly reasoned in the text, and the calculated lifetimes given in addition to the equations so the dominant loss pathways can be easily discerned.

Response: Agree and have modified this part and changed the uptake coefficient of HONO on aerosol to 10^{-5}.

Line 166: Delete ‘slightly’. This is an unnecessary modifier.

Response: Agree and have deleted.

Line 171: What are the authors trying to say with the term ‘amplitude’ here? This seems like they are trying to describe the average difference between the diurnally averaged minimum and maximum HONO observations. What exactly is the point of interest in these amplitude values? Maybe a comparison to reports in the literature could help build some better context here.

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

Line 174 and 175: Change ‘exceeding’ to ‘up to’ and give the exact number of the maximum observation. This provides better factual boundaries for the context of the discussion and is another example of where qualitative language detracts from the quality of this work.

Response: Agree and have changed.

Line 176: Delete ‘interestingly’. It is up to the reader to determine what is and is not interesting based on the facts presented. Also delete ‘also significantly’ unless you are going to support this senctence with data from a statistical test that was performed.

Response: Agree and have changed the description in the revised manuscript. The statistical results have also been presented in the following text.

Line 179: A preface to K+ as a biomass burning tracer needs to be put in here. The authors jump right into using it to define BB versus nonBB events, but that fact, along with appropriate referencing, has not been provided anywhere at this point. This description and referencing were previously requested in earlier revisions.

Response: We have added a new section 2.5 and several references (refer to the response of the first major comment) to descript why we choose K^+ as a biomass burning tracer in this work.

Line 184: Delete ‘significantly’ and give the exact factor that each term was higher during BB versus nonBB periods instead of the more vague ‘about a factor of 2’. If a statistical test for significance is performed, provide the appropriate metrics and ‘significantly’ can be retained.

Response: Agree and have presented the statistical results in the revised manuscript.

Line 187: Delete ‘significantly’

Response: Agree and have deleted.

Line 188: Delete ‘in turn’ and ‘a series of’

Response: Agree and have changed.

Lines 190 and 191: Delete ‘on average’ inside the brackets and add ‘average’ before ‘observed’ on Line 190

Response: Agree and have changed.

Line 194: of   for

Response: Agree and have changed.

Lines 199-202: As stated above, this calculation requires some additional work and the conclusions drawn from them need to be rethought for the discussion. Also, HONO is not ‘highly reactive’, but it is ‘photolabile’.

Response: Agree and have re-calculated the HONO lifetime in the revised manuscript. We have also changed the description of this sentence.

Line 204: Can a more explicit range other than ‘several’ be given here? Is it 4-10 hours? The data generated for the HYSPLIT figure should allow the authors to have a definite range and transport time window for nocturnal biomass burning plumes to their site and that should be explicitly provided as it will allow them to contrast their lifetime calculations for HONO losses against arrival at the site. This is another way that better limits could be placed on how much HONO is arriving at the SORPES site as direct emissions versus secondarily produced HONO on BB aerosols and the ground.

Response: Please refer to the response of the main comment 2.

Lines 206-207: This is speculative and is not sufficiently justified if the individual plume travel times are not known. Once recalculated, it is likely that direct HONO emissions are important to consider for this dataset and CO observations will help to justify this.

Response: We have calculated the contribution of HONO from BB emission to the observed HONO concentrations using the method of K^+ trace by taking account the HONO lifetime and plume transport time.

Line 213: Are these 'precursor concentrations' HONO precursor concentrations? The authors only consider NO2, so this contradicts the rest of the statement.

Response: Agree and have changed the statement in the revised manuscript.

Lines 222 – 232: One lifetime against loss is only a reduction in the initial HONO concentration by a factor of 2.7. After recalculating the nocturnal loss processes, these retroplumes and subsequent discussion should be revisited as I expect a more reasonable lifetime will be on the order of 8 hours once plume time spent in contact with the surface is considered as a weighting factor in HONO dry deposition and the HONO loss to aerosols is recalculated with uptake coefficients which are more representative of organic and/or secondary inorganic aerosols.

Response: Agree. We have re-calculated the HONO lifetime in the revised manuscript. And re-draw the Fig. 4 to show the 8-hr retroplume.

Lines 240 – 243: This figure is internally dependent between the axes. Particle surface area versus PM2.5 mass loading is the appropriate plot to discuss and the scales should be linear, not logarithmic.

Response: Agree and have replaced this figure and the descriptions in the revised manuscript.

Lines 250 – 253: This also tells you that the particle number is much higher. This particular point is touched on at lines 276-277 and should be first presented here.

Response: Agree and have added this information in the revised manuscript.

Lines 256-257: The correlation statistics must be given here to demonstrate the robustness of the relationship. This is currently too qualitative.

Response: Agree and have given the statistical results in new Fig.9 in the revised manuscript.

Line 264: testified   tested

Response: Agree and have re-written this part in the revised manuscript.

Lines 267-268: The plot, again, is inherently going to have a negative dependence and needs to have that removed and reassessed from the plot of HONO/NO2 versus particle surface area. The current justifications for the surface area filter should be rethought and rewritten.

Response: Agree and have removed this figure, and changed the statements in the revised manuscript.

Line 301: poorly   no

Response: Agree and have changed.

Line 301: Give the value of the correlation.

Response: Agree and have given the information in the revised manuscript.

Line 309: deployed   used

Response: Agree and have changed.

Lines 310-313: Some of these points were similar, but many were higher. When did these points arrive at the site? Was it a continuous series of points or were they arriving at different times throughout the event? This plot is clearly showing greater plume age for this event over the rest of the dataset than many other correlative plots presented in this paper. Why have the authors not discussed this? If these populations were truly statistically similar, then what test was done, what was compared, and what were the statistical results?

Response: Here the photochemical age of is represented by the ratio of nitrate to NO_y (the slop showed in Fig. 11b), but not the nitrate concentrations. In Fig. 11b and 11c, what we want to show is whether the data points of both BB and mix plumes were in the same regime (if most data points lie around the same regression line), and whether the ratios of nitrate to NO_y were similar or not.

And the results clearly showed that the nitrate to NO_y ratios were similar between BB and mix plumes (the ratios in BB plumes were even slightly higher than those in mix plumes), suggesting the photochemical age of the plume in June 10th was similar to other BB plumes.

In the revised manuscript, however, we have removed this part and Fig. 11. Actually, the nitrate to NO_y ratio is proper parameter to estimate the photochemical age for the general air plumes. But we cannot ensure if it can be used for BB plumes. In this part, what we want to discuss was whether the photochemical age for June 10 case longer than other BB plumes? Was it the major contributor to the enhanced HONO concentration during the case?

In the revised manuscript, we presented the changes of $HONO/NO_2$ ratios during the nighttime for both BB and non-BB samples (Fig. 6 in the revised manuscript). The result showed that it only needs 8 hours to get balanced between HONO and NO_2, and reach a steady state for both BB and non-BB plumes. The balanced $HONO/NO_2$ ratios were 0.083 ± 0.014, which were still much lower than those in June 10 case (0.17 ± 0.046), suggesting some other factors other than the plume age enhanced the HONO concentrations during 10 June.

We have added these statements in the revised manuscript.

Line 314: Get rid of the ‘beginning stage’. It adds nothing to the discussion.

Response: Agree and have changed the description in the revised manuscript.

Lines 321-323: The longer photochemical age supports this conclusion. Why was this not discussed? The longer photochemical age also means there was more time for NO2 HONO on the ground if the plume was in contact with the surface. Can the nitrate to NOy plot be used to approximate the plume age? If yes, why is this not presented, nor the relevant literature cited? The authors need to choose a defendable method to describe the plume age if they are insistent that all the HONO is coming from secondary processes. The comparison criteria here aren't clearly stated. Is the figure comparing a similar period of data collected as is presented for the June 10 case? Or is this comparing all the other BB data and potentially biasing the analysis?

Response: We have removed this part and Fig. 11 in the revised manuscript. Actually, the nitrate to NO_y ratio is proper parameter to estimate the photochemical age for the general air plumes. But we cannot ensure if it can be used for BB plumes. In this part, what we want to discuss was whether the photochemical age for June 10 case longer than other BB plumes? Was it the major contributor to the enhanced HONO concentration during the case?

In the revised manuscript, we presented the changes of $HONO/NO_2$ ratios during the nighttime for both BB and non-BB samples (Fig. 6 in the revised manuscript). The result showed that it only needs 8 hours to get balanced between HONO and NO_2, and reach a steady state for both BB and non-BB plumes. The balanced $HONO/NO_2$ ratios were 0.083 ± 0.014, which were still much lower than those in June 10 case (0.17 ± 0.046), suggesting some other factors other than the plume age enhanced the HONO concentrations during 10 June.

We have added these statements in the revised manuscript.

Lines 324-326: As presented in Section 2.4, heterogeneous loss processes are surface area dependent, not dependent on mass loading. The authors need to be clear that they used mass loading here as a proxy for surface area. Figure 13c presents the best case for this, but also doesn't exclude the ground surface. Have the authors tried to derive a relationship between surface area and mass loading for their BB plume aerosol populations that they could apply to the transformation of data in this figure? It would make the property being tested (chemical nature of NO2 conversion efficiency) readily apparent.

Response: We agree that the heterogeneous loss processes are surface area dependent. Here the ratios of $HONO/NO_2$ to $PM_{2.5}$ was used to represent the potential of aerosols to convert NO_2 to HONO, which combined parameter of both NO_2 to HONO conversion efficiency ($HONO/NO_2$/surface area) and aerosol specific surface area (surface area/mass concentration). Therefore, the mass loading we chose here was not used as a proxy for the surface area. In the manuscript, we referred the information in the end of former paragraph (the 2rd paragraph of Section 3.3), before the discussion in Lines 324-326.

In the revised manuscript, we presented the relationship of particle surface area in new Fig. 8a (showed as follows). The results show that data points of BB and non-BB samples did not fit the same regression line. If we use this relationship to estimate the particle surface area of the aerosols in the June 10 case, one assumption that the particle specific surface area of aerosols in June 10 case are the same as that of other BB aerosols. However, as we referred in the manuscript, June 10 case was a mixed episode of both BB and FF plumes. The role of this kind of mixture in the particle specific area is not clear. But the formation of secondary coating on the BB aerosol would probably change the particle size distribution and morphology, and in turn influence the particle specific area. Therefore, we did not estimate the aerosol surface area by the particle mass loading.

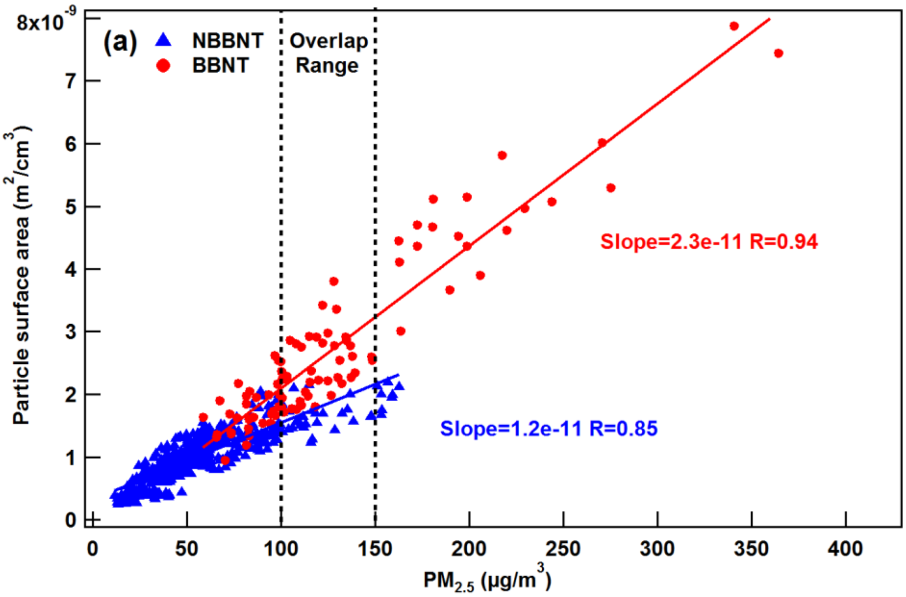


Fig. 8a Scatter plot between the particle surface area and $PM_{2.5}$ for nighttime samples during BB and Non-BB periods.

Lines 352-354: This could just as easily be due to an increase in available surface water[*Stutz et al.*, 2004]. Based on known chemical mechanisms[*Finlayson-Pitts et al.*, 2003], that is more likely than any proof presented here for SOA formation by mixing the aerosol populations.

Response: We agree with the referee's viewpoint. The related description was provided in the next paragraph. We changed some description in that paragraph in the revised manuscript.

Line 365: The conclusions need a full update based on the revisions to the paper. They have not been updated after the first revision.

Response: we have changed the conclusions part accordingly, and changed title of this part from "conclusions" to "conclusions and implications". And we also revised the abstract part.

Figure 1: The authors were requested by both reviewers to shade or somehow denote the periods they classified as biomass burning events. The authors do not want to add shaded regions, but some notation is critical to communicating the frequency of BB

versus nonBB events intercepted at this site.

Response: Agree, and we shaded the period when the BB was frequently occurred (28 May to 13 June) in the new Fig. 1 in the revised manuscript.

Figure 2b: Change the scale on the HONO/NO2 and get rid of the units since the quantity is unitless.

Response: Agree and have changed in the revised manuscript, and thank the referee for the reminder on the error of the unit.

Figure 3: Each of these plots can be tested statistically for discussion of elevated conditions in the BB plume versus when there was no BB plume detected. As requested previously, the number of data points in each panel need to be provided. Also, move 'between biomass burning period and non-biomass burning period' to follow after 'Comparisons' to improve clarity of this caption.

Response: Agree. We have added the p value of t-test result to the revised manuscript for all these comparison, and provided the number of the data points.

Figure 4: Given the capacity of the authors to create this figure, Figure 7, and the availability of the nitrate and NOy data, plume travel times to the site should be possible to calculate. At the least the range of travel times should be possible to estimate, if not specifically for each plume intercepted.

Response: Please refer to the response to the major comment 1.

It is actually a difficult job due to 1) the transport of air plume is actually an issue of gas diffusion, which is hard to define the beginning time and ending time; 2) the exact source region (fire point on the map) is hard to identified. Some episodes maybe influenced by several source regions on the transport pathway.

In the revised manuscript, we estimated the contribution of BB emission to the observed HONO concentration during BB periods using the method of K^+ tracer, and assumed the averaged transport time was 4 hours, which actually overestimated the contribution of BB for most episodes.

Figure 5: Why is no correlation data given? There appears to be a reasonable positive correlation here. What is the R2 value of an error-weighted linear regression?

Response: We have added the correlation efficiency of the least square liner regression method in the revised manuscript

Figure 6: The authors want to keep this figure as displayed to show a correlation between HONO and NO2. To me, this is very well established in the literature and the novel component of this work would be showing that HONO/NO2 is enhanced with increasing surface area, which is completely consistent with the main hypothesis of this work. The authors should strongly reconsider changing this plot to HONO/NO2 versus particle surface area and retaining the BB and nonBB point differentiation. This could also support their hypothesis of different chemical mechanisms affecting the conversion efficiency if the BB points represent a population statistically distinct from that of nonBB aerosols.

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

Figure 7: This needs to be completely reconsidered. The conversion of NO2 to HONO occurs regardless of the HONO lifetime, so the actual plume source and transport time is essential to determining how much HONO can be made in a plume interacting with the ground surface. Again, since the lifetime is e-folding, only a factor of 2.7 of the initial HONO is lost over the period of a lifetime. About five lifetimes would have to have passed for the complete loss of the initial emission of HONO. Given the more reasonable, but probably still too short, lifetime of 3.5 hours, at least 8-12 hours of transport over the surface are necessary to approximate the ground surface production.

Response: Agree. Please refer to the response of the former comments.

Figure 8a: This is just 1/PM2.5 versus PM2.5 with the noise from particle surface area overlaid on top. Make this into particle surface area versus PM2.5 and keep the axes linear instead of log-log.

Response: We have changed this figure to the suggested one, which is particle surface area versus $PM_{2.5}$.

Figure 9: Fix the conflicting orders of magnitude on the vertical axis and relabel the horizontal axis to 'K+/PM2.5'. This looks like a reasonably good positive correlation to me. What are the statistics?

Response: Agree and have changed in the revised manuscript.

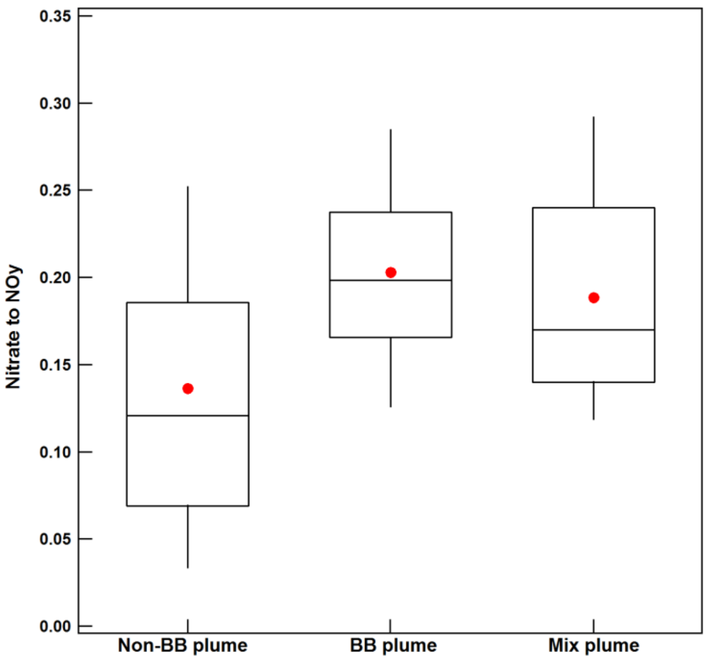
Figure 10a: Same problem as Figure 8a. Fix it. The authors are clearly trying to demonstrate that there is more than just the surface area that is important in converting NO2 to HONO, which is fantastic, but the approach to depicting this a simpler figure. The log-log scales draw attention to small differences in otherwise not very different data, which makes the comparison misleading.

Response: Agree. We have removed this figure and changed the description in the revised manuscript. Please refer to the response of the major comment 2.

Figure 11c: What if you put the nitrate to NOy for nonBB periods on this figure? Are those data any different from these? Again, the number of data points in each box-and-whisker plot need to be provided.

Response: Please refer to the former comments. We have removed the whole Fig. 11 in the revised manuscript.

The results of nitrate to NO_y for non-BB, BB and mix periods are showed as follows.



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