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.

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 NO_2 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 NO_2 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 NO_2 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 NO_2 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/NO_y chemical aging clock, this analysis would also strengthen their ability to identify the potential contributions of direct HONO BB emissions in this dataset.

Figures with Induced Dependence

Of significant concern are the plots made for the second reviewer of HONO/NO_2/surface area vs $K^+/PM_{2.5}$. These plots demonstrate surface area vs $K^+/PM_{2.5}$ are likely dependent on each other, yet the authors present two new figures with the exact same issue in the manuscript: i) surface area/$PM_{2.5}$ versus $PM_{2.5}$, and ii) HONO/NO_2/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.

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. R^2, 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.

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.

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.

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.

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

Lines 9-11: NO_2 is also directly emitted from BB processes. This is not a sound conclusion to be making. Reconsider the wording of this sentence.

Line 27: O_3 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.

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

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

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.

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.

Line 81: Correct this to 'Field'.

Line 95: chromatography → chromatograph

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.

Line 111: Was the NO_y molybdenum converter used in any way to correct for the interference given in the previous sentence? Why mention the interference of NO_y in the NO_2 measurement and then not be clear about how you did or did not account for that possibility?

Line 124: A reference needs to be provided here.

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

Line 139: measurement → measured

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

Line 145: depression → deposition

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.

Line 166: Delete 'slightly'. This is an unnecessary modifier.

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.

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.

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.

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.

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.

Line 187: Delete 'significantly'

Line 188: Delete 'in turn' and 'a series of'

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

Line 194: of → for

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’.

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.

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.

Line 213: Are these ‘precursor concentrations’ HONO precursor concentrations? The authors only consider NO_2, so this contradicts the rest of the statement.

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.

Lines 240 – 243: This figure is internally dependent between the axes. Particle surface area versus $PM_{2.5}$ mass loading is the appropriate plot to discuss and the scales should be linear, not logarithmic.

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.

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

Line 264: testified → tested

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 $HONO/NO_2$ versus particle surface area. The current justifications for the surface area filter should be rethought and rewritten.

Line 301: poorly → no

Line 301: Give the value of the correlation.

Line 309: deployed → used

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?

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

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 $NO_2 \rightarrow HONO$ on the ground if the plume was in contact with the surface. Can the nitrate to NO_y 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?

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 NO_2 conversion efficiency) readily apparent.

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.

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

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.

Figure 2b: Change the scale on the $HONO/NO_2$ and get rid of the units since the quantity is unitless.

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.

Figure 4: Given the capacity of the authors to create this figure, Figure 7, and the availability of the nitrate and NO_y 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.

Figure 5: Why is no correlation data given? There appears to be a reasonable positive correlation here. What is the R^2 value of an error-weighted linear regression?

Figure 6: The authors want to keep this figure as displayed to show a correlation between HONO and NO_2. To me, this is very well established in the literature and the novel component of this work would be showing that HONO/NO_2 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/NO_2 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.

Figure 7: This needs to be completely reconsidered. The conversion of NO_2 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.

Figure 8a: This is just 1/$PM_{2.5}$ versus $PM_{2.5}$ with the noise from particle surface area overlaid on top. Make this into particle surface area versus $PM_{2.5}$ and keep the axes linear instead of log-log.

Figure 9: Fix the conflicting orders of magnitude on the vertical axis and relabel the horizontal axis to 'K^+/$PM_{2.5}$'. This looks like a reasonably good positive correlation to me. What are the statistics?

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 NO_2 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.

Figure 11c: What if you put the nitrate to NO_y 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.

References

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