Responses to comments by Anonymous Referee #1

1. General comments, on quantitative discussion: In this paper, we presented descriptions and performance assessments of systems for the measurements of HONO concentration and HONO flux, and provide detailed descriptions/discussion of the measurement results, including a complete time series over the measurement intensive and a case study. As pointed out by the referee, we did not provide much quantitative calculations leading to a conclusion on surface nitric acid photolysis as a HONO source, which had been done and published elsewhere along with canopy surface nitric acid/nitrate data (Zhou et al., 2011). We will modify the discussion in the manuscript, and add appropriate citations to avoid the impression that we derive that conclusion based on the time-series or case study alone.

2. Page 7277, line 6, HONO removal by the Na_2CO_3-coated denuder: We have conducted tests in the field and in the laboratory: (1) we have compared the blank signal for reagent solutions (i.e., the system running with the coil sampler passed) with the "denuder-zero air" signal, and found that they were mostly identical, except during some high-NO_x episodes in the day, during which the denuder-zero air signal was slightly higher by a few pptv. (2) We have compared the signals of "zero-HONO" air generated by one denuder and by two denuders connected in series, as suggested by the referee; we did not observe discernible difference between the two. These results confirm that HONO is quantitatively removed by the Na_2CO_3-coated denuder. In addition, the denuder manufacturer reports that the collection efficiency is 99% at a flow rate of 10 L min^{-1} (http://www.urgcorp.com/library/datasheets/). Small percentages of PAN and NO_2 in ambient air may be removed by the Na_2CO_3-coated denuder. However, the effect on HONO measurement should be negligible, since the interference from these species on HONO sampling is not significant even at elevated levels (i.e., similar to the urban concentrations) (Huang et al., 2002).

3. Page 7280, line 11, the location of meteorological measurements: Temperature, relative humidity, and dew point were acquired through the website (http://www.wunderground.com) for Pellston Airport, about 5.5 km from the PROPHET site. The information will be added to the manuscript as suggested.

4. Page 7284 line 17, canopy/soil pH and HONO emission: The surface acidity (or pH) of canopy (and soil) plays an important role in HONO exchange between the surface (and soil) and the atmosphere. However, it is difficult to quantitatively assess canopy surface pH or acidity, and we are not aware of any direct surface pH measurement in literature. We used the precipitation pH distribution and patterns to compare the pH difference on surface at the sites in Michigan and in California, since they have been well established and have been recorded by the National Atmospheric Deposition Program (NADP) (http://nadp.sws.uiuc.edu). While the precipitation pH does not reflect the actual canopy acidity, it provides useful contract between the two regions: Low industrial SO_2 and high agricultural NH_3 emissions in California result in relatively low aerosol acidity and thus relatively neutral pH on canopy surface as well as in precipitation (at or slightly above pH 5.5 controlled by CO_2 buffer system). On the other hand, high industrial SO_2 emission in the Midwest results in excess acidity in aerosols (mainly as HSO_4^-) and precipitation (pH 3-4.7), and thus acidify the canopy surface. The effect of surface is not just on HNO_2/NO_2^- equilibrium and distribution, but more importantly on the photolysis rate of surface HNO_3 (or nitrate) as a HONO source. Laboratory results have shown that photolysis rate is much slower for nitrate salt than HNO_3 on surfaces, e.g., by a factor of >10 for $NaNO_3$

compared to HNO_3 on Pyrex surface (Gao, 2011). Photolysis of deposited HNO_3 on canopy surface is probably the dominant daytime HONO source at the PROPHET site (Zhou et al., 2011), but might not be that important at the sites in California (i.e., the CalNex Bakersfield site and BEARPEX site, Ren et al., 2011) due to much slower nitrate photolysis on the surface at or near neutral pH. HONO emission from soil is also highly dependent on soil pH, as reported by Su et al. (2011). Soil pH is determined by the acidity/alkalinity of the soil matrix as well as the acidity of precipitation. We are preparing a manuscript for the same special issue of ACP based on HONO gradient measurements in 2009, and we will demonstrate that the forest soil was not a net HONO source at the PRHOPHET site. Significant revision will be made to that paragraph to include the above discussion.

5. Page 7286 lines 3 and 10, HONO source calculation: The overall HONO source of ~350 pptv hr^{-1} was estimated to balance photolysis and other reaction losses of HONO at the observed concentration of ~70 pptv. The 43% volume production contribution was not calculated from the sum of HONO production rates from gaseous and aerosol phase reactions, since we did not have all the information needed to make such calculation. Instead, we derived the value from the difference between the overall HONO source required (~350 pptv hr^{-1}) and the contribution from canopy surface (~200 pptv hr^{-1}), which was calculated based on the flux information. Detailed discussion has been given in Zhou et al. (2011).

6. Page 7287 line 1, NO_x as a HONO precursor: We did not attempt to argue for or against homogeneous reactions as a HONO source in this paragraph. We intended to explore the importance of NO_x as a HONO precursor by examining the correlations of NO_x with HONO and HONO flux. During the day, NO_x was not a significant precursor for HONO produced at the forest canopy, as suggested by the poor and negative correlation between NO_x and daytime HONO flux (r^2=0.01, slope = -1.0×10^{-10} mol m^{-2} hr^{-1} (pptv NO_x)$^{-1}$). The weak but positive correlation between NO_x and daytime HONO (r^2=0.01, slope =0.016) reflected weak but positive contribution from the volume HONO production under high-NO_x conditions during southerly flow. During the night, the correlation significantly improved with a positive slop (r^2=0.37, slope =1.1×10^{-10} mol m^{-2} hr^{-1} (pptv NO_x)$^{-1}$), suggesting that reactions of NO_x on the canopy surfaces have some but not a major contribution to the observed HONO at 32 m. The stronger and positive correlation between the nighttime concentrations of HONO and NO_x (r^2 = 0.56, slope =0.02) suggests that NO_x was the major HONO precursor when photolysis of HNO_3 on surface was absent. Removing four high-NO_x data points significantly lowers the correlation, as pointed out by the referee, suggesting that importance of NO_x as a HONO precursor increases with the NO_x concentration. The measured nighttime HONO might be produced at the local forest surfaces (indicated by a generally upward HONO flux) and within the air mass (volume production), and may be transported to the measurement site from other source locations.

7. Page 7287, line 4, Evidence for heterogeneous NO_x reactions on aerosol surface as a HONO source: Starting at line 4 on page 7287 is the following sentence: "The correlation between NO_x and the daytime HONO concentration is slightly improved but still weak (r^2 = 0.10), reflecting some degree of contribution from the volume HONO production under high-NO_x conditions during southerly flow." We did not attempt to argue for or against the heterogeneous NO_x reactions on aerosol surfaces as a HONO source. Indeed, we did not have the aerosol information to examine the heterogeneous NO_x reactions on aerosol surface as a HONO source.

8. Conclusion 4: The conclusion will be revised to address the referee's comment. Any discussion on HNO_3 photolysis on canopy surface as a daytime HONO source will be referred to our previous paper (Zhou et al., 2011) in this manuscript.

Responses to comments by Anonymous Referee #2

1. Photosensitized NO_2 reaction as a daytime HONO source: We have followed the Referee's suggestion and examined the correlation between HONO flux and $[NO_2]\times J_{NO2}$, and found it to be poor, with a r^2 of 0.01, similar that between HONO flux and $[NO_x]$. The poor correlation thus suggests that the photosensitized NO_2 reaction was not a significant daytime HONO production mechanism responsible for the measured HONO flux at this low-NO_x site. The NO_x level was significantly higher at CalNex Bakersfield site (mean noontime ~ 3 ppbv in summer 2010, Ren et al., 2011) than that at the PROPHET site (mean noontime ~ 0.6 ppbv in summer 2008), and thus NOx became a more important precursor for HONO at CalNex Bakersfield site. Another difference in the PROPHET site and CalNex Bakersfield site may be the pH of the surface: the overall lower acid precipitation in the western US including California than in the mid-western and northeast U.S., by ~2 pH units (National Atmospheric Deposition Program (NADP), http://nadp.sws.uiuc.edu). Laboratory results have shown that photolysis rate on surface is much slower for nitrate salt (e.g., $NaNO_3$) than HNO_3, by a factor of 10 (Gao, 2011). Photolysis of deposited HNO_3 on acidic canopy surface is probably the dominant daytime HONO source at the PROPHET site (Zhou et al., 2011), but might not be that important at CalNex Bakersfield site due to slower nitrate photolysis on the surface of higher pH. As a result, the measured daytime HONO flux may reflect different HONO production mechanisms, i.e., surface HNO_3 photolysis at the PROPHET site, and photosensitized NO_2 reactions at CalNex Bakersfield site.

2. Evidence for photolysis of HNO_3 deposited in the forest canopy is the major daytime source of HONO: As pointed out by the referee, that conclusion is mainly based on the analysis reported in our previous paper (Zhou et al., 2011). We will change the conclusion accordingly in our next revision.

3. Page 7275, line 5, typo: The typo will be corrected.

Responses to Comments by Anonymous Referee #3

1. Measured flux *vs* the actual surface exchange: Photolysis is the dominant removal pathway for atmospheric HONO. The HONO photolytic life time was about 10 min around noontime. We have estimated that the actual daytime HONO flux from the forest canopy may be slightly greater than that that measured at 11m above the canopy, owing to the photolytic loss of HONO during diffusive turbulent transport, by an upper limit of ~10% around noontime, assuming a J_{HONO} of ~6 hr^{-1} (~10 min lifetime) and an eddy diffusivity coefficient *Kz* of ~3.6×10^4 m^2 h^{-1} (or ~1×10^5 cm^2 s^{-1}). We did discuss this potential photolytic loss of HONO in our earlier paper (Zhou et al., 2011). We will include this discussion in the manuscript in the next revision.

2. Page 7274, lines 2-3, writing style: We have often seen passive writing style in the Abstract of a paper. We would leave the editor to decide which style is more suitable.

3. Section 2.3, dynamic dead-band and β: A dynamic threshold (deadband) $w_T = \sigma_w/2$ was used to increase the concentration difference between the up and down components and to achieve a more constant β coefficient (Bowling et al., 1998). We lose the air temperature data due to some technical problems with our software, and thus we calculated the β coefficient using the vertical wind measurement:

$$\beta = \frac{\overline{w'w'}}{\sigma_w \Delta w}$$

where w' is the fluctuating components of the instantaneous measured vertical wind velocity (w), σ_w is the standard deviation of w, and Δw the difference in the conditionally sampled w. We derived a β value of 0.442±0.016 from 3-hour vertical wind data around noontime on July 15, 2008, and used it for the HONO flux calculation. The derived value is in good agreement with the value of 0.43 calculated based on:

$$\beta = \beta_o \exp(-0.75 \frac{w_T}{\sigma_w})$$

with a β_o of 0.627 at a zero threshold and a w_T of $0.5\sigma_w$ (Businger and Oncley, 1990; Westberg et al., 2001). It is also in good agreement with the reported values of 0.471±0.007 and 0.438±0.017 at the PROPHET site (Westberg et al., 2001), and 0.410±0.018 at CalNex Bakersfield site and 0.441±0.008 at the BEARPEX site (Ren et al., 2011). The description and discussion presented here will be added to the manuscript in the next revision.

4. Figure 4, concentrations for dead-band: We did not specifically measure the HONO concentration in dead-band air. The "zero-HONO" air was fed to both updraft and downdraft samplers during a dead-band period. The ambient HONO concentration integrated over the 20-min sampling period (including all the updraft, downdraft and deadband periods) indeed fell between the updraft and downdraft concentrations.

5. Page 7285, line 14, typos: The typos will be corrected in the revision.

6. Page 7288, line 19, Do you mean that the w was below the threshold the whole period? Yes, the night was very calm, with vertical wind mostly below the initial threshold w_T^o of 0.1m s^{-1} during several 20-min periods (or >95% of the time).

Responses to comments by I. Trebs, A. Moravek and M. Sörgel

We appreciate the critical comments by Trebs et al., and we take the opportunity to reexamine our systems and the data collected more carefully and critically. The point-by-point responses below address the concerns and comments raised by the authors. We will incorporate the discussion/information below into the manuscript in our next-step revision.

1 Analyzer precision

We agree with the authors that high measurement precision of concentrations is essential for reliable REA measurements. We have modified our HONO measurement system and have significantly improved the performance of the technique, as reported in this paper. The side-by-side HONO concentrations measurements by two or three systems, made at the PROPHET site and in the laboratory, showed excellent agreement, better than 2% in the concentration range of 50 pptv - 5 ppbv. Below 50 pptv, the agreement was still excellent, with data points from the two systems falling on one another; however we were unable to claim the agreement to be within 2%, since detection limit of the method was 1 pptv (3 times the noise signal when sampling "zero-HONO air). We would like to point out that the estimated overall uncertainty of $\pm(1 + 0.05$ [HONO]) pptv is a measure for deviation of the measured value from the true value, not for measurement precision, taking into consideration such uncertainties associated with standard preparation/calibration, gas and liquid flow rate measurements/variation, collection efficiency measurements/variation. Some of these uncertainties would be largely cancelled out when the difference was taken between the updraft and downdraft concentrations for flux calculation, since both systems shared the same inlet, they were calibrated by the same standards, and the sampling flow rates were calibrated by the same flow calibrator.

A flux measurement detection limit of $\pm 8\times10^{-8}$ mol m^{-2} hr^{-1} and an overall uncertainty of $\pm (8\times10^{-8} + 0.15\ F_{HONO})$ mol m^{-2} hr^{-1} were estimated, combining the uncertainties in HONO concentration measurements, vertical wind measurement, and sampling periods and sampling volumes in up and down components. The authors are correct that at low concentrations, it is difficult to measure the flux accurately. Indeed, we found large measurement uncertainties at low flux periods, such as during the night. At the measured flux of 0.1×10^{-6} mol m^{-2} hr^{-1}, the uncertainty would be $\pm 0.095\times10^{-6}$ mol m^{-2} hr^{-1}, i.e., a relative uncertainty was 95%, not 15% as stated by the authors. During the day, the observed flux was most several times greater than the measurement uncertainty.

As pointed out by the authors, the occasionally measured downward HONO fluxes under rainy and foggy conditions may be affected by HONO being trapped on inlet walls under high humidity conditions. Potential wall effect might attenuate and smooth the HONO signals. Since the updraft and downdraft systems shared the same inlet, some of the effect might be partially canceled out when taking the concentration difference for flux calculation. Therefore, we believe that the observed negative fluxes during the rain and fog events were real, although the actual deposition rate might be somewhat underestimated.

As implied by Equation 1, the HONO flux is positively proportional to σ_w, and thus higher σ_w should lead to higher HONO flux if HONO concentration difference stayed relatively constant. The diurnal cycles of HONO flux and σ_w were similar, because they are related to a common factor, the solar radiation. The measured HONO flux reflected the HONO production rate from HNO_3 photolysis at the canopy surface, while the vertical mixing is enhanced by the surface heating of solar radiation. However, a closer examination of the measurement time series (Figure 5) reveals that higher σ_w did not always result in higher HONO flux, and the highest HONO fluxes were not associated with the highest σ_w values. The highest HONO fluxes occurred at the intermediate σ_w, values (Figure 6).

As mentioned above, we did conduct several side-by-side measurements using 2 or 3 systems to evaluate the system performance, and the obtained precision was ~2%, far smaller than the

concentration difference between the updraft and downdraft air masses during majority of the time. When ambient concentration was too low or the concentration difference was too small for our systems to detect accurately, the derived flux was usually within or close to the detection limit of $\pm 8\times10^{-8}$ mol m^{-2} hr^{-1}, and the relative uncertainty was large. We are confident that the reported HONO flux, especially during the day, was not a result of systematic difference.

2. Flow conditions in the sampling tubes

Indeed, the Reynolds number is calculated to be about 2700, and thus the flow in the inlet line was in the transitional regime. We did not monitor the temperature and pressure in the inlet line, and thus were unable to evaluate the smearing effect on fluctuating signals. However, our inlet (2.2 m) was much shorter than that (30 m) by Ammann et al. (2006), resulting in a much shorter air residence time (200 ms *vs* 4.3 s). The resulting attenuation in turbulent fluctuation was probably a fraction of the air residence time in the inlet tube, which was much shorter than the median duration time of 2-3 s for a turbulent cell (please see more discussion in point 6). Therefore, the fluctuation of signals should not be affected to any significant degree by the flow conditions in the inlet tube.

Our "zero-HONO" air was generated by pulling ambient air (taken from the same inlet) through a Na_2NO_3-coated denuder (URG-2000-30x150-3CSS). Since minimal pressure drop was over the denuder, the air pressures at the incoming ports of the 3-way valve should be mostly the same for the ambient air and the "zero-HONO" air, and thus pressure fluctuation when the valve switching should be minimal. I could see that pressure fluctuation may affect the air sampling flow if a pressurized zero air source is used.

4. Effect of inlet tubing on HONO

We did not specifically investigate artificial formation or loss of HONO on inlet tubing during this campaign. However, we have previously examined this sampling interference many times at this and other low-NO_x sites (Zhou et al., 2001, 2002; He et al., 2006). We found that the exposure of glass and Teflon inlets to sunlight led to significant artificial HONO signal. No significant interference was observed when the inlet was protected from the sunlight, as demonstrated by the excellent agreement (i.e., within analytical uncertainty of 2-3%) between the signals of two channels, one without any extra inlet and the other with a long inlet (up to 30 m). In this study, the inlet tubing was 2.2 m and the residence time was 200 ms, both relatively short; the inlet artifact on HONO measurement should be negligible. Even if the inlet artifact existed, the effect on measured HONO concentrations would likely be cancelled out by taking the concentration difference.

4 Delay time

The delay time was 206 ms for air to flow through a volume of 20.6 mL in a down facing Teflon elbow (~1.9 mL), a 220-cm 0.32-cm ID Teflon common inlet tubing (~17.7 mL), and Teflon cross fitting splitting the flow (1.0 mL). A delay time of 200 ms was set to trigger the sampling valve, without taking into consideration the time lag of ~100 ms due to data acquisition at 10 Hz. As a result, there was a time offset of about 100 ms in the actual sampling of the up or down component. That means that our systems could not catch the high frequency signals, and might cause underestimations in the concentration difference and thus the calculated flux. However, the error should be relatively minor, considering the median duration of 2-3 s for updraft and

downdraft air parcels (see more discussion below in **6 Additional comments**). We will add more discussion in the manuscript in the next revision.

5 Determination of β-factor

As discussed above in point 3 under **Responses to Comments by Anonymous Referee #3**, we lose the air temperature data due to some technical problems with our software, and thus we calculated the β coefficient using the vertical wind measurement. We will add more details to the manuscript in our next revision.

6 Additional comments

Page 7277, line 7, HONO may evaporate from aerosol nitrite: A "zero-HONO" air was generated by pulling ambient air through a Na_2CO_3-coated denuder to remove gas-phase HONO, and the signal was used as a baseline to correct for the potential interference in the air, including aerosol nitrite and the HONO released from aerosol nitrite. We did reagent blank check from time to time with coil sampler bypassed (i.e., not contact with air), and found it to be identical or only slightly lower (a few pptv) than the "zero-HONO" air signal, suggesting that interference from atmospheric species, including aerosol nitrite, is insignificant.

Page 7278, lines 14-16, flux calculation: In every 30-min cycle (i.e., 10-min "zero-HONO" air and 20-min measurement), one flux data point was calculated from the 20-min integrated σ_w, C_{up} and C_{down}. Details were described in Page 7282, line 19-27, and were illustrated in Figure 4. The 20-min σ_w was calculated from 10-Hz vertical wind data; and the C_{up} and C_{down} were calculated by integrating the respective 1-min HONO data over the sampling period. In the very calm period, when |w| was smaller than the initial w_T^o of 0.1m s^{-1} for >95% of the sampling time and "zero-HONO" air was sampled for >19 min in the 20-min measurement period, the flux data point was not valid and not included in the reported flux result.

Page 7282, line 7, coordinate rotation method: Double rotation method was used for coordinate transformation.

Page 7282, line 14, duration of eddy parcels: Following the authors' suggestion, we have examined the vertical wind data collected over the 24-hr period on July 25 (with a typical calm night and a strong vertical-mixing day). The range of duration was from 0.1 - 27 s, with the median between 2 to 3 seconds for both updraft and downdraft. The duration shorter than 0.5 s accounted for only 17% of the sampling time for the updraft downdraft components, respectively; the duration longer than 1 s accounted for about 70%. The error of ~100 ms in lag time would cause only minor uncertainty in the flux measurement.

Page 7282, line 20, HONO sampling: The sampling units were not temperature-controlled. It has been demonstrate that quantitative collection (>97%) was achieved at an air sampling flow rate of ≤ 4 L min^{-1} and a scrubbing solution flow rate of ≥0.24 mL min^{-1} at room temperature of 22 ^oC (Zhou et al., 1999; Huang et al., 2002). In this study, an air sampling flow rate of 2.0 L min^{-1} and a scrubbing solution flow rate of 0.35 mL min^{-1} were used; the collection efficiency was expected to be improved and quantitative. At this high collection efficiency, the error in collection efficiency introduced by temperature fluctuation should be negligible.

3-way valves: All-PTFE 3-way miniature solenoid valves (series 1, Parker) were used in this study. The wetted material was inert PTFE and should have minimal effect on HONO

concentrations during the sampling. We will redraw the Figures 1 and 2 in the revision as suggested to make them easier to understand.

Sampling cycle: As pointed out by the authors, the 20-min sampling cycle might not be able to catch the contribution of very low frequency drafts. However, we have not seen a duration of either updraft or downdraft longer than 1 min.

Figure 4: Figure 4 shows only 2 hr of data at various processing stages to illustrate what the raw data look like and how they are processed to derive the final concentration and flux values. During the 2 hours in the early morning before sunrise on July 25, 2008, relative stable σ_w and concentration difference were maintained, resulting in relatively small variation in HONO flux.

Result interpretation: we will modify and add more discussion on HONO sources, as suggested. Soil may be a potentially important HONO source as reported by Su et al. (2011). However, our gradient measurement results in 2009 summer showed lower daytime HONO concentrations near the forest ground than above the canopy, suggesting that soil was not a significant HONO source for the overlying atmosphere at the PROPHET site. A manuscript is being prepared for ACP in the same special issue.

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