Response to Referee 1:

Can the authors rule out any measurement uncertainty or bias that may explain the discrepancies? It is noted that no discussion is provided in this paper on the uncertainty or quantitative accuracy of the measurement data.

Readers are directed to the QC-TILDAS characterization paper (Ellis et al., 2010) for detailed information on the instrument figures of merit. Uncertainty in our measurements may arise from uncertainties in the calibration source (NH_3 permeation tube), stability of the permeation oven, the measurement of the temperature and pressure in the optical cell and the spectroscopic data given in the HITRAN database. Of all these, the biggest source of uncertainty should be the output of the permeation tube source, which can be determined offline using an ion chromatograph to better than 10%. The instrument has been compared to 10 other measurements techniques (von Bobrutzki et al., 2010) and does not exhibit any discernible bias. The biggest discrepancy between the model and measurements is in the gas fraction distribution, and a systematic measurement bias would not resolve this disagreement.

Data is averaged to 5 minutes to produce a clearer picture of trends in the time series in Figure 1. When averaging to 5 minutes our detection limit is 42 ppt with a precision of 14 ppt. We have included these numbers in the revised manuscript.

Is it possible that there is a significant component of ammonium associated with the coarse particle mode, which could influence observed gas phase NH3 levels and hence the measured gas fraction.

While we do not have measurements of ammonium content for particle sizes larger than $PM_{2.5}$, we do not anticipate a significant amount of ammonium in coarse mode aerosol. Size-resolved particle composition measurements in Southern Ontario described in VandenBoer et al., (2010) and Zhang et al., (2008) show negligible contributions of ammonium in particles larger than 2.5 microns. Additionally, AURAMS predicts 3 orders of magnitude less NH_4^+ in the coarse mode than in $PM_{2.5}$.

Could it not be argued that based on Fig 9, which compares the ammonia gas fraction versus sulfate, that the measurements and model discrepancy is systemic and that the authors should not just focus on the discrepancies at either sulfate concentration extreme (which is the basis for the bi-directional flux from/to plants as a function of gas ammonia concentrations). One could argue that the model overall does a poor job and thus is likely lacking in many respects. It is noted that even when a bi-directional flux is included (though it may be a crude representation), the improvements were marginal.

Figure 9 shows that the model underestimates gas fraction at high sufate, and overestimates it at low sulfate, so there is not a systematic discrepancy, but rather one that appears to depend on the chemical composition of the atmosphere. Thus while there could be several reasons for the discrepancy between measurements and model (emissions too low, deposition too high, incorrect representation of gas-particle partitioning, advection), we believe that the full implementation of a bi-directional flux parameterization will be most successful at reducing the discrepancy. Our uncoupled representation of bi-directional flux was successful at significantly reducing the very low gas fraction population in the model distribution but further improvement may need a more sophisticated treatment.

As a side question, why do plants attempt to keep gas phase ammonium in a certain concentration range? Could the proposed missing source realistically provide the amount of missing ammonia needed to bring models in agreement with observation during high sulfate conditions?

The physiological control of gas phase ammonia exchange between plants and the atmosphere is based on Henry's Law and dissociation equilibrium between the aqueous phase ammonia and ammonium in the soil water or plant apoplast or cuticles. As long as a sufficient pool of ammonium exists at the surface, this flux can be maintained. Detailed information is given in the references offered in the manuscript.

Pg, 21901 line 10, give the calculated cut size of the inertial separator used to remove particle interferences. The NH3 inlet configuration is also not clear; does the complete inlet only consist of a 10cm quartz tube, or are there additional sections. How high was the inlet located above ground level and how was it located relative to activities around the site?

Added the theoretical cut size of particles larger than 300 nm.

The whole inlet is made of quartz, 10 cm in length, with three 2.5 cm long protrusions on one side for attachment of zero, cal and sample lines. A 3 m PFA sample line connected the quartz inlet and the instrument optical cell. For more information on inlet configuration please see Ellis et al., (2010).

Added, "During BAQS-Met, the inlet was mounted on top of the measurement trailer, at a height of 2.5 m above the ground, and connected to the QC-TILDAS via a 3 m long, 3/8 inch PFA sample tube."

Pg 21902, clarify the sentence: ½" OD Teflon-coated aluminum tube breaching a 4" diameter PVC pipe. Also, I assume this was the same inlet used for particles. Include an estimate of particle sampling looses (or lack of them). Are there issues sampling particles through a teflon-coated tube due to electrostatic effects. No information is provided on the AIM accuracy (eg, was it compared to filters etc), measurement LODs or uncertainties.

The Teflon-coated aluminum tube is distributed by University Research Glassware and is designed for the sampling of fine particulate with minimal losses. We have added: "Based on comparisons with the AMS instrument at our site, it does not appear that there were losses of particles due to electrostatic effects in the inlet." Detection limits are provided in the manuscript. We have provided more quantitative information on accuracy: "An off-line calibration of the AIM-IC was performed in-field by directly injecting multiple ion standards into each IC, providing accuracy of better than 15 %."

Pg 21904, line 9, clarify what is meant by exact overlap, what time scales were associated with the miss-match? Line 26, give distance to road.

When spikes of ammonia occurred (mainly at night) spikes in other species did not occur at the same time, but either before or after separated by several minutes.

Added distance to road, approximately 40 m.

Pg 21905, lines 4 and 5, but was there a direct correlation between the field work and measured NH3, this is implied but not explicitly stated. Line 24, missing "be" (to be expected?).

Unfortunately the field work was not logged and we cannot discern whether this activity had any effect on the ammonia concentrations. We simply offer this as a possible contribution to variability in local ammonia emissions.

Added "be".

Pg 21908, regarding the discussion of variability between 10:00 to 15:00, numbers are needed to justify the statement that partitioning contributed to NH3 decrease during this time (line 15). In Fig 5a, it appears that there is a 10% decrease in NHx, is this significant relative to the decrease in NH3?

Added "During the hours of 10:00 – 15:00, ammonia decreased by 30% while only a 10% decrease was observed in NH_x"

Pg 21910 line 18-20, what about changes in BL height and variability in the vertical concentration profile of NHx as a process that influences NHx concentrations in each model grid cell.

To clarify, we are reporting the model results in the lowest grid cell which extends from the surface to 6.89 m. The depth of the mixed layer may affect the predicted NHx levels if the PBL height predicted by the weather forecast model is too high, resulting in more significant vertical diffusion from the lowest grid cell, making the concentrations too low. However, by the same argument, all of the other pollutants should be too low in concentration as well (e.g. NO_x, CO should be low in the model output). This was not observed, and in fact the model was biased high for NO_x and CO (Makar et al., 2010). So the model and observations for other species do not support the idea that NHx underestimates are due only to PBL height overestimates - the other species are not being affected the same way.

Pg 21911 and 21912 regarding the discussion of bi-directional flux of NH3 involving ambient NH3 concentrations and plant apoplastic fluid. The argument is that plants attempt to keep some form of equilibrium between apoplastic fluid and ambient NH3 levels and that this explains the discrepancy between the model and observations.
Is it reasonable to assume that the plants can adjust that quickly and with sufficient emissions to account for the large differences observed?

As discussed in these references (Farquhar et al., 1980; Sutton et al., 1993; Sutton et al., 1995; Asman et al., 1998; Nemitz et al., 2001) given in the manuscript, the equilibrium is established based on the Henry's Law constant and acid dissociation and is thus a temperature dependent function of the ratio of NH_4^+/H^+ in solution. The timescale on which this equilibrium can be achieved should depend on the timescales of vertical mixing in the atmosphere, which in the lowest grid of the model should be on the order of minutes.

In the Conclusions definitive statements are made but were not completely proven in the body of the paper. Eg, Pg 21913 line 5, I don't believe that spikes at night were proven to be from cars, I thought it was only speculated. Other statements should be considered and qualified if need be.

We have restated our conclusions to take into account the comments of both reviewers.

Response to Referee 2:

Morning peak in ammonia

The authors rule out that desorption of ammonia from plant surfaces is responsible for the rise in NH3 in the morning. They argue that air temperature rises at a later time in the day, which would take time to increase surface temperatures. However, in fact it is the rise in the surface temperature (by inception of solar radiation) that drives the increase in temperature in the morning. Thus leaf surface layers can evaporate as soon as they are illuminated by the sun, while the air temperature increases more slowly. Desorption of NH3 has been observed following high humidity periods, even if no visible dew formation took place. Microscopic leaf water layers which form at RH well below 100% can store significant amounts of NH3 (e.g. Flechard et al., 1999; in your reference list). Thus I do not think it can be ruled out that desorption of NH3 from surfaces into a boundary layer that is still shallow contributes to the rise of NH3 in the morning, at least initially.

The reviewer raises a valuable point and we have altered the manuscript accordingly: “It is possible that the temperature of the soil increases slightly earlier than the temperature of the atmosphere at 2 m (the height of our RH/T probe), but we have no measurements of the soil temperature. While the rate of increase of NH_3 in the morning was not substantially higher following nights with formation of dew, Flechard et al., (1999) states that significant amounts of ammonia can be stored in leaf water layers formed at relative humidities below 100%. Without measurements of surface wetness and surface temperature, we cannot conclusively confirm or rule out volatilization from the surface.”

Role of nitrate partitioning.

One principal concern about the manuscript is that ignores the interaction of NH3 with HNO3 to form NH4NO3, although nitrate should be available from the AIM-IC. Nitric acid would be helpful for the interpretation, but I understand that the AIM-IC inlet was not suitable to measure this compound. Because the NH3-HNO3-NH4NO3 equilibrium is highly dynamic it is likely that HNO3 has a strong influence on NH3 concentrations in the gas phase, especially since the measurements indicate the presence of significant excess free NH3 during the entire period. This would imply that the aerosol was probably neutralised during the period. However, the current paper does not provide any information on

(a) whether the aerosol was neutralised or acidic

Figure 4 has been updated to include the mole equivalents of NH_3, NH_4^+, SO_4^{2-} and NO_3^- and shows that the aerosol was neutralized except for some afternoons with very high sulfate loadings. The model predicted significant amounts of acidic aerosol, throughout the study, which often led to predictions of zero ammonia.

and (b) what the importance was of ammonium nitrate compared with the ammonium sulphates. A plot comparing modelled and measured concentrations of NH3, NH4+, NO3- and SO4= would be helpful to form a fuller picture of the interactions. A comparison of the acidity of the measured and modelled aerosol would equally be very helpful. I suspect that the offline run of ISORROPIA predicts an acidity that is much closer to the measurements than predicted by AURAMS?

The updated Figure 4 also demonstrates that nitrate was an insignificant constituent of the aerosol. This is consistent with the analysis of Markovic et al. (2010) who used AMS data from the same site and saw that nitrate was only important in the early morning hours. AURAMS predictions of nitrate also peaked in the morning and were of similar magnitude to the observations. Markovic et al. (2010) also includes model-measurement comparisons and offline ISORROPIA runs which show aerosol acidity much closer to measurements. In the mid-afternoon when we saw the largest discrepancies between the model and measurements the formation of ammonium nitrate was not important.

For a neutralised aerosol, NH3 uptake onto the aerosol should not be governed by the aerosol sulphate loading, but by the in-situ production of sulphuric acid (are SO2 data available to estimate H2SO4 concentrations?) and by HNO3 concentrations. Under these conditions, large SO4= concentrations per se do not provide a large condensation sink for NH3. The NH3 needs to be neutralised somehow. Thus, the decrease of NH3 during the day could be associated with an increase in HNO3, which ties up more NH3 into the aerosol phase. This process appears to be a more likely explanation for the diurnal cycles observed than the neutralisation of advected SO4= aerosol.

The data shows that the formation of ammonium nitrate is not favoured in the afternoon and while we do not have measurements of HNO_3 it is unlikely that it is acting as a condensation sink of ammonia. As discussed above, in some instances of high sulfate mass loadings the aerosol was acidic and net uptake of ammonia would be expected. On days with neutralized aerosol, as the reviewer points out, the net uptake rate of ammonia is governed by the production rate of H_2SO_4. We estimated this using observed midday SO_2 of 2 ppb, assuming 5×10^6 OH, and obtained a production rate of 0.04 ppb h^{-1}. Since two equivalents of ammonia are required to neutralize, this works out to 0.08 ppb h^{-1} or 0.056 μg m^{-3} h^{-1} loss of NH_3. This calculation is a lower limit as it does not account for heterogeneous production of H_2SO_4. The calculated production rate is significant compared to the rate of surface exchange (0.3 μg m^{-3} h^{-1}) calculated by the model for afternoon conditions. This implies that even in cases with neutral aerosol, a strong condensation sink in the atmosphere still exists and if this term was included in the model it would have an impact on the surface fluxes. This explanation has been added to the manuscript.

The current Fig. 9 is only partially informative because the entity shows in the xaxis (SO4=) is effectively part of the denominator of the entity shown on the y-axis, making it circular. Nevertheless, if the NH3 concentration is governed by the NH3-HNO3-NH4NO3 equilibrium, the NH3 concentration may show a dependence on the SO4=/NO3- ratio, because NH3 vapour pressures above mixed NO3-/SO4= aerosols are reduced compared with pure NO3- aerosol. This would also be consistent with Fig. 9. It may be interesting to use the offline version of ISORROPIA to predict the HNO3 concentration based on the measured values and compare this with HNO3 diurnal cycles in the literature.

We plotted the NH_3 concentration (and gas fraction) versus the SO_4^{2-}/NO_3^- ratio as the reviewer suggested but the graphs did not provide any additional information. As in our discussion above, the observed ammonia concentrations were not governed by the NH_3-HNO_3-NH_4NO_3 equilibrium in the afternoon. Because there is such little nitrate in the aerosol, observations do not provide a useful constraint to predict HNO_3 concentrations from ISORROPIA.

If the modelled aerosol is often acid, whereas the measured aerosol was not (as Fig. 8 suggests), this would probably suggest that NH3 emissions upwind of the measurement site are underestimated or (less likely) that the time-scale for neutralisation (NH3 uptake onto the aerosol) is overestimated. This underestimation in NH3 emissions may reflect missing plant emissions (from plant canopy compensation points), but it could equally reflect underestimated agricultural emissions.

The reviewer is correct that if the emissions in upwind adjacent grid squares were higher, the predicted aerosol would not be as acidic. However, scaling up agricultural emissions would result in uniformly higher NH_x, which would reduce the number of zero gas fraction points but not the high gas fraction population. Therefore this explanation does not appear to resolve the model-measurement discrepancy.

Similarly, it would be interesting to investigate under what conditions, the model predict higher gas fractions than the measurements. These could be conditions, under which the NH4+ is dominated by NH4NO3 and the model does not predict sufficient NH3 and HNO3 for NH4NO3 production. This could indicate that HNO3 formation from NOx is underestimated during these conditions.

Figure 9 shows that the high gas fractions occur when the SO_4^{2-} is low. There is no dependence of the gas fraction on nitrate.

The analysis of the different operators affecting the NH3 concentration at the site is interesting, but since the model does not predict the acidity of the aerosol correctly, I don't believe the results bear any relationship to reality. The model should be adjusted to get the results correct first, before this analysis is undertaken.

The mass tracking operators were investigated in an attempt to discern the extent to which processes in the model govern ammonia concentrations at our site. We did not intend to use the model to explain real world behaviour because, as the reviewer points out, the model results to do match observations. We have clarified this in the manuscript.

Compensation point modelling.

The temperature dependent compensation point, based on a constant value of gamma, is clearly the more correct approach to modelling bi-directional transport. If this does not solve the model/measurement discrepancy it is not the (only) missing process.

Foliar compensation points will always act to raise day-time concentrations, because during night temperatures are lower and stomata are closed. It is unclear from the description, what parameterisations were used for simulating atmospheric (Ra, Rb), stomatal and cuticular exchange resistances.

With our temperature dependent compensation point approach, we also used ISORROPIA to iteratively partition the NH_3 in the same manner as with the constant value compensation point. We did not use a resistance based parameterization in our approach. This has been clarified in the text. While a foliar compensation point may also be relevant, the reviewer suggests it would raise daytime concentrations whereas our simple model approach is already biased high during the day.

Changes that would increase night-time concentrations include use of lower boundary layer height and larger attribution of the NH3 emissions to the night in the diurnal emissions cycle.

Reducing the boundary layer height or changing the diurnal cycle of NH_3 emissions are ways of changing the ammonia concentrations in the model without invoking the bi-directional flux. However these methods would not improve the discrepancy in the gas fraction distribution. If bi-directional flux is indeed occurring then a change in boundary layer height will have no impact assuming that concentrations quickly converge to the compensation point.

In general, the single-point offline approach taken by the authors assumes that concentrations are in full equilibrium with each other (as predicted by ISORROPIA) and in equilibrium with the surface (as predicted by the bi-directional exchange model). In reality, both processes interact with each other as pointed out earlier in the manuscript with reference to the work of Wolff et al. (2010).

This is what we were referring to in the statement "Thus atmospheric gas phase ammonia is simultaneously participating in two equilibria, one with atmospheric particulate ammonium, and one with a pool of ammonium at the surface. If the condensation sink increases (e.g. by an increase in aerosol sulfate), the atmosphere is depleted of ammonia, potentially lowering the ambient mixing ratios below the compensation point, and triggering emissions from the plants and soil."

Abstract. The authors should add a sentence on the conclusions drawn from the implementation of a bi-directional NH3 flux parameterisation into the model.

Added "A simple representation of an offline bi-directional flux parameterization using ISORROPIA was successful at reducing the population of zero gas fraction points, but not higher gas fraction points."

P21897, l. 10ff: Some valuable plant communities only survive because of N limitation, because at higher N inputs they would be out-competed by nitrophilic plant communities. Thus, the statement that NH3 is beneficial to N limited ecosystems needs to be qualified.

Changed statement to "Deposition of NH_3 and NH_4^+ to the Earth's surface can fertilize nitrogen-limited ecosystems, and have detrimental effects such as eutrophication, soil acidification, and biodiversity loss in sensitive ecosystems"

P21898, l. 18ff: Wolff et al. (2010) were not the first to study the interactions between surface/atmosphere exchange and gas-particle partitioning. More pivotal papers include Brost et al. (1988), Kramm & Dlugi (1994), Harrison et al. (1989).

Included suggested references.

P21898, l.26 ff. It may be worth including references to some other simultaneous NH3/NH4+ measurements (e.g. Nemitz et al., 2004; Trebs et al., 2004; Schaap et al., 2010)

Included suggested references.

P21901, l.4. Has the effect of the heated inlet on NH3/NH4+ phase partitioning been characterised? The heating could volatilise some NH4NO, which is obviously important in the context of the present analysis.

The heated inlet has not been tested for volatilization but theoretical calculations indicate that the temperature is not high enough and the residence time through the inlet is too short (due to a high flow of 9 L min^{-1}) to volatilize NH_4NO_3 before the virtual impactor.

P12901, l. 15. The manuscript describes the fast response of the QC-TILDAS in some detail, but only 5 minute values are used for the analysis. Thus detection limit & precision for 5 minutes are more relevant here than for 1 s.

Data is averaged to 5 minutes to produce a clearer picture of trends in the time series in Figure 1. When averaging to 5 minutes our detection limit is 42 ppt with a precision of 14 ppt. We have included these numbers in the revised manuscript.

Section 2. Please provide information on length of sampling inlets for QC-TILDAS and AIM-IC as well as inlet height.

The QC-TILDAS and AIM-IC inlets were co-located at a height of 3 m above the ground. We have added: "During BAQS-Met, the inlet was mounted on top of the measurement trailer, at a height of 3 m above the ground, and connected to the QC-TILDAS via a 2.5 m long, 3/8 inch PFA sample tube." Information on the QC-TILDAS inlet was further clarified "Following the campaign, it was determined that the sampling inlet (1/2 inch OD Teflon-coated aluminum tube (URG, North Carolina) breaching a 4 inch diameter PVC pipe) was not appropriate for the quantitative sampling of gases, especially soluble ones such as HNO_3 and NH_3, so only the $PM_{2.5}$ measurements are used in this analysis. Based on comparisons with the AMS instrument at our site, it does not appear that there were losses of particles due to electrostatic effects in the inlet."

P21904, l. 9ff. Good correlation between NH3 and other compounds does not necessarily prove common sources, but could just reflect a common control through boundary-layer dynamics.

We are referring to correlation between species on very fast time scales (seconds to minutes) which is faster than the change in boundary layer height.

Figure 2. The freely available OpenAir software provides a nice means to display exactly these kinds of relationships (www.openair-project.org).

We believe the relationships are conveyed well by the graphs in their current format.

P21905, l.17ff. The statement that deposition rates to water should be lower than to vegetation is counterintuitive because deposition rates of NH3 to vegetation are often dominated by deposition to microscopic water layers on plant surfaces and increase as these water layers get thicker at high relative humidity. However, vertical mixing and therefore transport is less efficient above smooth surfaces such as water bodies.

Clarified statement to "On the other hand, the deposition velocity of surface reactive gases such as ammonia is lower over water than over land due to less vigorous vertical mixing over water, possibly extending the lifetime of ammonia against deposition"

P21906, l. 10. Do the authors have any information on soil wetness? Very wet soil conditions suppress NH3 evaporation because the liquid pools can store a lot of NH4+, reducing the likelihood for NH3 volatilisation. Only in otherwise very dry conditions would I expect precipitation to enhance denitrification to an extent that it overrules this effect. You could also refer to Nemitz et al. (2000) and McCalley and Sparks (2008), both of whom demonstrated NH3 increases with humidity/precipitation, from leaf litter and desert soil, respectively. However, have the authors investigated alternative explanations for the increase in NH3 after rainfall, such as changes in air mass associated with frontal activity?

Unfortunately we do not have measurements of soil wetness. From a regional analysis which identified changes in air mass at the site, there were no air mass changes during those times. Of the 4 instances of NH_3 increase after rainfall, the local wind direction changed on 2 occasions, but this was characterized by very low wind speeds, indicating stagnant air and not a major frontal shift. We have included this information in the revised manuscript.

P21906, l. 1ff. Is the spike removal really justifiable? The aerosol measurement may also have included spikes, but these could not be resolved. Similarly, the model provides a grid-cell average, which also includes areas near sources.

The spatial allocation for emissions in the model is not appropriate to resolve emissions from individual vehicles, so the removal of spikes is required in this case. It is true that the hourly measurement from the AIM-IC may have included spikes in aerosol ammonium, but it is not clear whether the short duration spikes would be in equilibrium with NH_3 and thus have an impact on the ammonia concentrations. Most of our analysis is based on diurnal cycles and there is no statistical difference between diurnal plots of ammonia with spikes and without spikes. Also the spikes occurred mostly at night, while our analysis is concentrated on daytime concentrations.

P21908, l. 9f. I do not agree with the statement that if total NHx is considered, the effect of phase partitioning is removed. Due to the difference in the deposition rate of NH3 and NH4+, the phase partitioning dictates the atmospheric lifetime and thus concentration of total NHx.

We agree with the reviewer and have clarified statement to: “At equilibrium, a decrease in NH_3 would result in an increase in NH_4^+, and a plot of NH_x would be constant assuming no additional sources or sinks. Thus, the effect of gas-particle partitioning is removed in plots of NH_x, and the diurnal variability is thus only controlled by transport and vertical exchange.”

Fig. 5. It would be good to see the results of the ISORROPIA offline run also.

Figure 5 has been updated to include the ISORROPIA run.

Fig. 6. It is unclear to me whether the operator allocation is for the local grid cell only or for a larger area within the model.

The allocation is for the local grid cell over Harrow at 2.5 km resolution. This has been added to the manuscript.

Technical Corrections

Numerous spaces are missing throughout the manuscript as already noted by the editorial team.

Numerous occurrences of ‘. . .’ before citations, which do not appear to make sense.

Both errors were introduced in the processing of the manuscript and will be corrected in the final draft.

Introduce a space between values and their units throughout (e.g. “3 L/min” rather than “3L/min”)

Done.

P21901, l.25, change ‘outfitted’ to ‘fitted’?

Done.

P21904, l. 28, ‘stated’ instead of ‘states’?

Done.

P21905, l. 24. ‘. . . expected to be the highest . . .’

Done.

P21911, l. 23. ‘dependant’ should be ‘dependent’.

Done.

Caption Fig. 8. Suggestion for alternative wording: “While the measured frequency distribution of the gas fraction is clustered in the middle of the histogram, the model is largely bi-modal, often attaining extreme values.”

Changed wording as suggested.

References

Ellis, R. A., Murphy, J. G., Pattey, E., van Haarlem, R., O'Brien, J. M., and Herndon, S. C.: Characterizing a Quantum Cascade Tunable Infrared Laser Differential Absorption Spectrometer (QC-TILDAS) for measurements of atmospheric ammonia, Atmos. Meas. Tech., 3, 397-406, doi:10.5194/amt-3-397-2010, 2010.

Makar, P. A., Zhang, J., Gong, W., Stroud, C., Sills, D., Hayden, K. L., Brook, J., Levy, I., Mihele, C., Moran, M. D., Tarasick, D. W., and He, H.: Mass tracking for chemical analysis: the causes of ozone formation in southern Ontario during BAQS-Met 2007, Atmos. Chem. Phys. Discuss., 10, 14241-14312, doi:10.5194/acpd-10-14241-2010, 2010.

Markovic, M. Z., Hayden, K. L., Murphy, J. G., Makar, P. A., Ellis, R. A., Chang, R. Y.-W., Slowik, J. G., Mihele, C., and Brook, J.: The effect of meteorological and chemical factors on the agreement between observations and predictions of fine aerosol composition in Southwestern Ontario during BAQS-Met, Atmos. Chem. Phys. Discuss., 10, 24781-24820, doi:10.5194/acpd-10-24781-2010, 2010.

VandenBoer, T. C., Petroff, A., Markovic, M. Z., and Murphy, J. G.: Size distribution of alkyl amines in continental particulate matter and their online detection in the gas and particle phase, Atmos. Chem. Phys. Discuss., 10, 27435-27477, doi:10.5194/acpd-10-27435-2010, 2010.

von Bobrutzki, K., Braban, C. F., Famulari, D., Jones, S. K., Blackall, T., Smith, T. E. L., Blom, M., Coe, H., Gallagher, M., Ghalaieny, M., McGillen, M. R., Percival, C. J., Whitehead, J. D., Ellis, R., Murphy, J., Mohacsi, A., Pogany, A., Junninen, H., Rantanen, S., Sutton, M. A., and Nemitz, E.: Field inter-comparison of eleven atmospheric ammonia measurement techniques, Atmos. Meas. Tech., 3, 91-112, doi:10.5194/amt-3-91-2010, 2010.

Zhang, L., Vet, R., Wiebe, A., Mihele, C., Sukloff, B., Chan, E., Moran, M. D., and Iqbal, S.: Characterization of the size-segregated water-soluble inorganic ions at eight Canadian rural sites, Atmos. Chem. Phys., 8, 7133-7151, doi:10.5194/acp-8-7133-2008, 2008

Updated Figures

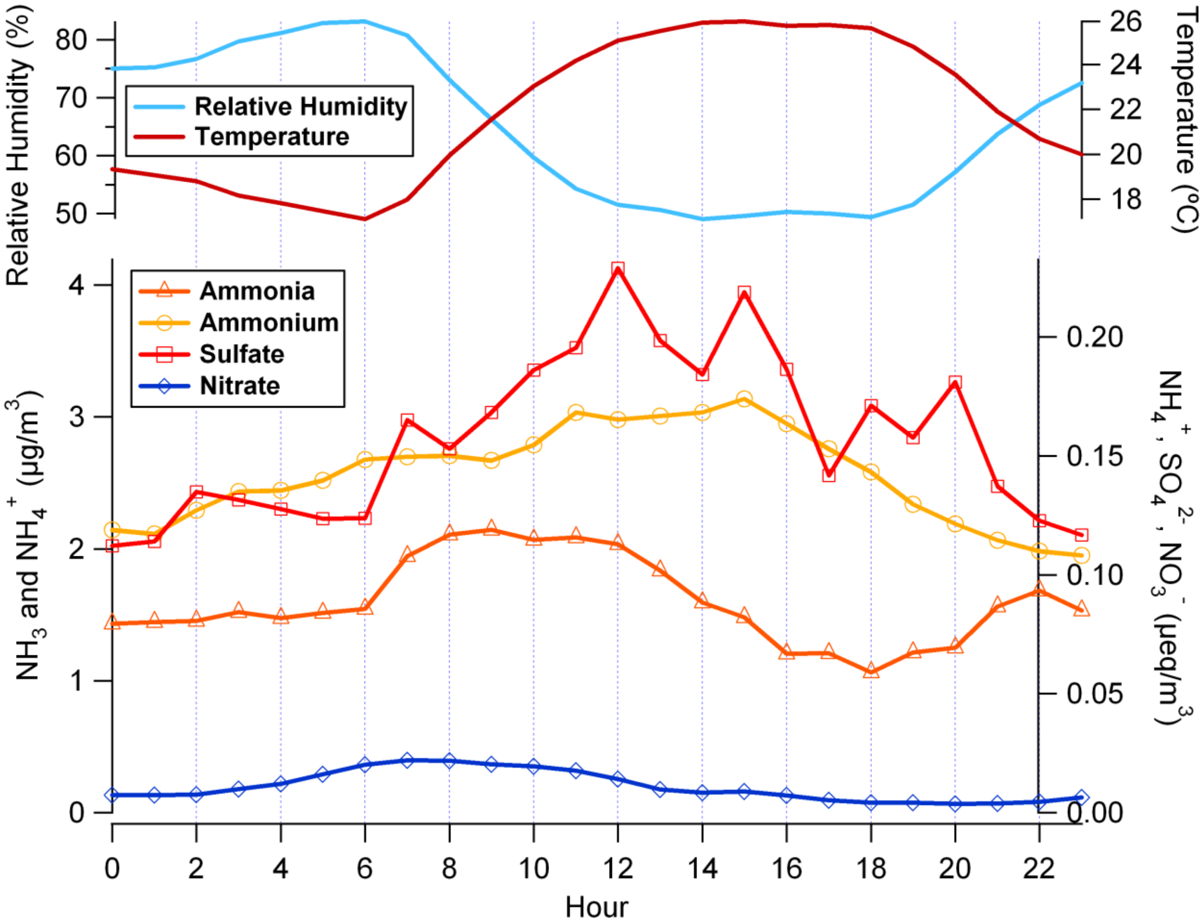

Figure 4: Average diurnal cycles of relative humidity and temperature with ammonia and ammonium concentrations. Ammonia concentrations display a rise in the early morning hours, followed by decrease in the afternoon. Mole equivalents of ammonium, sulfate and nitrate are plotted on the right axis and show the relative importance of ammonium sulfate vs ammonium nitrate.

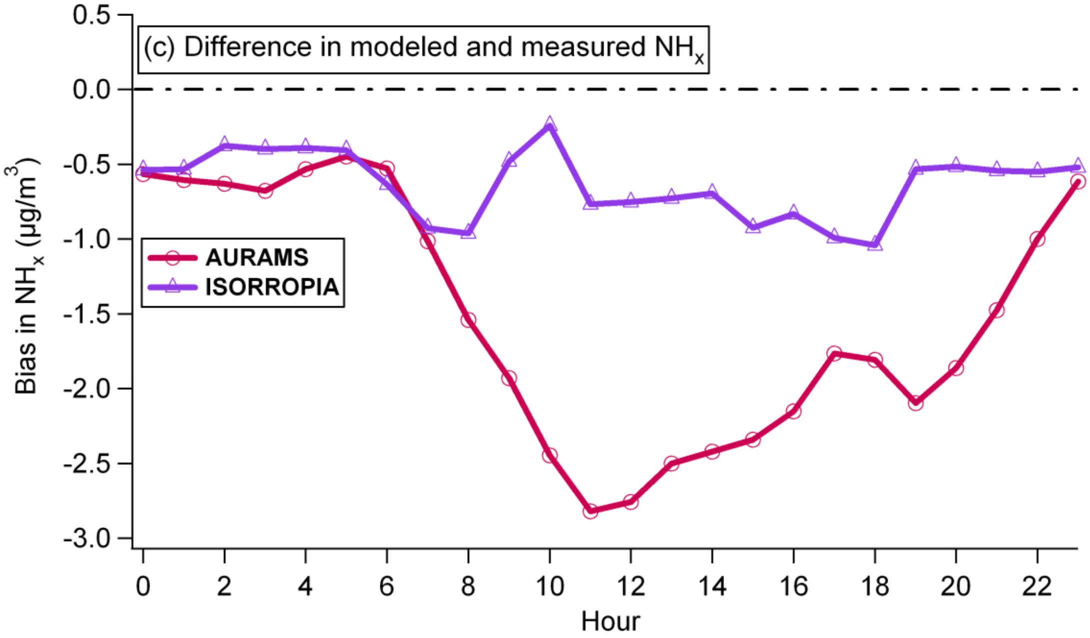

Figure 5(c): The difference between the modeled and measured NH$_x$ is show in (c) and indicates the AURAMS model is biased low in NH$_x$, especially in the afternoon. Repartitioning the observations at equilibrium using ISORROPIA removes the large afternoon bias, but is still overall biased low.