Author's Response to Referee #3

In this response, the referee comments (in black) are listed together with our replies (in blue) and the changes to the original manuscript (in red).

This manuscript does an overall good job of describing an instrument designed to measure NOx, NOy, and particulate nitrate by thermal dissociation – CRDS. The most useful aspect of the work is the demonstration of problems with the use of activated carbon denuders for removing gas-phase NOy compounds. This will be of great use to many other researchers who use these types of denuders and activated carbon in general!
I recommend it be published after addressing the minor comments below.
We thank the referee for the positive review of our paper and the helpful comments which we address in this response.

The detection limits are listed in the abstract (98 ppt for NOx with 1 min averaging) but strangely are not described elsewhere in the manuscript. Is this for a signal-to-noise ratio of 2? 3? How the LOD is defined and these numbers are determined should be in the main text somewhere. Given how sensitive CRDS can be to NO2, I am surprised that the LODs are as high as they are – I would have expected that with a minute of averaging the LOD would be quite a bit lower. Is this a result of the large correction (116 ppt) that must be made to account for the difference in Rayleigh scattering when sampling humid ambient air vs. dry zero air? In addition to that correction that must be made to account for the differences in humidity between sampling and zero measurements, doesn't the change in humidity also change the reflectivity of the mirrors (due to the change in the index of refraction of air), and thus the ring-down times?
We added a paragraph in Sect. 2.2 about the performance of the instrument and a critical comparison with other instruments (including LODs) in Sect. 3.4. The LOD we listed previously was from the AQABA campaign, where the ship's motions caused significant fluctuations in the ring-down times. We now list the performance obtainable on a stationary platform.
For NO_2, the performance of the instrument was first described by Thieser et al. (2016), who reports a measurement uncertainty of 6 % + (20 pptv*RH/100) which is dominated by uncertainty in the effective cross section of NO_2 and the wavelength stability of the laser diode. The NO_x detection limit of 40 pptv (2σ, 1 minute average) for the present instrument (laboratory conditions) was derived from an Allan variance analysis and is worse than that reported by Thieser et al. (2016) (6 pptv at 40 s) due to degradation of the mirror reflectivity. Corrections applied to take into account humidity and pressure changes are discussed in Sect. 2.1. The total uncertainty in NO_y will depend on the uncertainty in the conversion to NO_x of both gaseous and particulate nitrate and thus depends on the individual components of NO_y in the air sampled. For purely gaseous NO_y, the major problem is likely to be related to loss of sticky molecules at the inlet and we choose to quote a "worst case" uncertainty of 15%.

We have amended the LOD we quote to that obtained on a stationary platform (the one mentioned in the last version was derived from the AQABA dataset obtained on a ship):

In this context we note that the deployment on a ship resulted in a degradation in performance (LOD was ≈ 100 pptv) owing to the ship's motions, especially in heavy seas, which resulted in drifts in the instrument zero.

The zero air used for zeroes is "CAP 180, Fuhr GmbH"- please clarify what this means– is it compressed zero air from a cylinder, or is it from a zero air generator? Rather than deal with the effects of ambient sampling vs. dry zeroes, why not use humidity-matched air? (e.g., ambient air that has been scrubbed of NO2 via purafil or a catalyst?)
The zero-air generator has been described in more detail. Generally, the humidity correction is ≤ 100 pptv and has small associated uncertainties. Essentially we are scrubbing ambient air that has passed through a compressor.
k_0 is typically determined every five minutes (for one minute) by overflowing the inlets with zero air from a commercial zero air generator (CAP 180, Fuhr GmbH) attached to a source of compressed ambient air.

pg 6, last line - define BET pg 13, "However, when the main dilution flow was humidified significant," This sentence appears to missing a word. Or perhaps the last word should actually be "significantly".
Both corrected.
[…] we calculate a BET (Brunauer-Emmett-Teller (Brunauer et al., 1938)) surface area […]

[…] humidified ~~significant~~ […]

References

Brunauer, S., Emmett, P. H., and Teller, E.: Adsorption of gases in multimolecular layers, J. Amer. Chem. Soc., 60, 309-319, 1938.