

## ***Interactive comment on “Replacing the AMOR by the miniDOAS in the ammonia monitoring network in the Netherlands” by A. J. C. Stijn Berkhout et al.***

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(comment from referee)

The manuscript by Berkhout et al. titled “Replacing the AMOR by the miniDOAS in the ammonia monitoring network in the Netherlands” describes a network of the LPDOAS instruments for measuring ammonia installed as a part of the Ditch National Air Quality Monitoring Network. The DOAS instruments were replacing an outdated AMOR instruments, which quality ammonia in-situ using wet chemistry. The manuscript also analyzes a dataset collected during a 16-months-long intercomparison measurements between DOAS and AMOR instruments at six sites throughout the country. Overall, I was pleased to learn about the Dutch air authorities using optical remote sensing

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(ORS) technology for air monitoring. ORS methods, including DOAS, offer a number of advantages over “traditional” measurement techniques, however, they have been largely underutilized by governmental agencies. This manuscript provides an example of how DOAS method can be successfully used for contentious, long-term, near real-time measurements by a regulatory agency; and hence, this article is interesting for publication. I also envision that this paper will be of interest for scientists who rely on the Dutch ammonia network data for their research. However, in my opinion, this paper has a number of deficiencies that have to be addressed, before paper can be published. Sections below provide a list of specific comments for authors to address. Additionally, I believe that this manuscript can greatly benefit from English language editing. I encourage the authors to take advantage of the English editing services offered by AMT, or have the manuscript edited by a native English speaker.

(author’s response)

AMT offers English language copy-editing for final revised papers. We will gladly make use of this service.

(comment from referee)

Finally, I think that paper can be significantly strengthened by expanding the analysis of the collected data, data interpretation, and discussion of the results.

(author’s response)

The aim of this paper is to present a comparison between the two instruments, in order to assess any differences in performance and to understand a discontinuity in the ammonia dataset at the changeover point, if one should arise. Further analysis is certainly interesting but is outside the scope of this paper. Please note that the entire dataset is published, as a supplement to this paper, so anyone interested in doing a further analysis can do so.

(comment from referee)

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Major comments Section 2.3.2 – Add a figure with the example of the DOAS spectral fit. List all parameters included in spectral evaluation.

(author's response)

We add a Figure with all the steps in the spectral fit. This Figure is included at the end of this author's response.

(comment from referee)

One of the major strengths of the DOAS technique is that calibration is not necessary and concentrations can be determined from first principles; however, authors significantly diminished such advantage by devising a cumbersome method for measuring a reference spectra.

(author's response)

We do this by solving problems that may not be present in all DOAS applications, but they are in ours:

- By using a reference spectrum measured on the instrument itself, any pixel-to-pixel variability in sensitivity is automatically taken into account. This is a source of noise if it is ignored.
- Over the spectral range we use for our analysis, the light intensity of the observed spectrum increases by a factor of 20. Dividing by a lamp reference spectrum greatly reduces the dynamic range, this makes the approximation of the differential initial intensity  $I_0(\lambda)$  more robust.
- We do agree that the calibration procedure can be improved. That is why we envisage the construction of a laboratory facility with zero concentration over the full path length of the instrument (page 17, lines 1-2).

(comment from referee)

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Authors should explain why they chose to use reference calibration rather than spectral fitting using literature absorption cross-section convoluted with the instrument function.

(author's response)

Using literature absorption spectra is only one way to construct reference spectra, the method we use is equally well accepted (CEN, 2013). All authors of the literature we cite on page 7, lines 1-4, use the same method as we do. We agree that we do not list the factors that were decisive for us to select this method. We add two sentences to clarify this (page 7, line 6):

(author's change in manuscript)

We selected this method because the spectral modelling method offers many opportunities for errors to enter the calculations, if any of the required parameters are not precisely known. The gas cell method eliminates all these potential errors. In addition, using the gas cell method, the calibration becomes traceable to a standard with a known accuracy. This is an advantage when an instrument is to be used in a monitoring network that operates under a quality management system, e.g. ISO/IEC 17025. For the miniDOAS, we use this method as well, for the same reasons.

(comment from referee)

Authors should provide a more detailed explanation for the reference DOAS instrument, as well as highlight how it differs from the miniDOAS's used in the network.

(author's response)

A more detailed description of this instrument is given in Volten et al. (2012), we refer to this in the text. We add the differences with the miniDOASes to the text (page 7, line 28):

(author's change in manuscript)

Its differences from the miniDOAS systems are the following:

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- The RIVM DOAS has a better spectral resolution than the miniDOAS (0.0306 nm vs. 0.067 nm).
- The RIVM DOAS uses a cooled CCD detector, the CCD of the miniDOAS is not cooled.
- The wavelength calibration of the RIVM DOAS is continuously monitored by measuring the emission line of a zinc lamp. If needed, the grating of the spectrograph is adjusted. The miniDOAS has no such option.
- The RIVM DOAS reports values at 5 minute intervals, the miniDOAS at 1 minute intervals.

(comment from referee)

I am also surprised that authors did not encounter issues with using an uncooled CCD array. On page 3 authors briefly mention that use of an uncooled array leads to certain sacrifice of the instrument's performance (page 3, lines 11-12), but later on the same page (page 3, line 28) authors state that choice of the CCD instrument performance.

(author's response)

This sentence seems to be missing a part. We guess that you find our statement that the performance is not unduly affected a bit brief, we agree with that. We added a short remark (page 3, line 28):

(author's change in manuscript)

The use of an uncooled CCD increases the noise in the spectra, but we find that this does not affect the retrieval of the concentrations unduly.

(comment from referee)

It is clear that 1 minute data reporting for AMOR instruments is meaningless, I therefore recommend to remove 1min comparison between the DOAS and AMOR datasets as

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it bears no statistical significance. Instead, it should be highlighted that transition to the network of DOAS instruments will result in a dataset with much higher temporal resolution.

(author's response)

We show the 1 minute comparison to substantiate our claim that it is meaningless, we are therefore reluctant to remove it. The higher temporal resolution of the miniDOAS dataset is already highlighted in the Conclusions section, please see page 16, lines 14 to 19.

(comment from referee)

Authors should expand the description and discussion of data intercomparison with passive samplers. Table 5 should be replaced with a table of monthly averaged ammonia concentrations measured by passive samplers. Detection limits for passive samplers shall be stated. Analysis/intercomparison of passive samplers and monthly averaged DOAS and AMOR ammonia concentrations also should be presented.

(author's response)

All details of the passive samplers are given in the paper by Lolkema et al. (2015), to which we refer. We added a sentence to the paper to make this more clear (page 10, line 28):

(author's change in manuscript)

The passive samplers were operated as they are in the Measuring Ammonia in Nature network. All information on these samplers – handling, detection limits, calibration et cetera – can be found in Lolkema et al. (2015).

(author's response)

The passive sampler measurements presented here are meant only to investigate a possible difference in concentrations at various heights. An intercomparison with the

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passive samplers described in this section is not possible, because these samplers are calibrated against the monitoring network measurements (see (Lolkema et al., 2015)). A full intercomparison between AMOR, miniDOAS and another set of passive samplers is obviously possible but is outside the scope of this paper.

(comment from referee)

Authors should provide R2 values for correlation plots in Figures 9 and 12.

(author's response)

We added R2 to the figure captions of Fig. 9 (page 28, line 6, hourly concentrations at Vredepeel, R2=0.70), Fig. 10 (page 29, line 2, hourly concentrations at De Zilk, R2=0.76) and Fig. 12 (page 30, line 2, daily concentrations at Vredepeel, R2=0.81).

(comment from referee)

I imagine that such a long intercomparison of co-located AMOR and DOAS instruments was partially conducted in order to aid future interpretation of long-term ammonia concentration trends. For this purpose, it would be also interesting to see a more in-depth analysis of the collected data. For example – the analysis of seasonal and geographical trends and/or differences in AMOR and DOAS datasets. Figure 13 shows monthly averages obtained by both instruments at all stations throughout the country. It is obvious that datasets for some stations agree better than for others. I found it disappointing that analysis was only limited to a correlation plot.

(author's response)

The aim of this paper is indeed to join the datasets of AMOR and miniDOAS, so that concentration trend analyses spanning the changeover from AMOR to miniDOAS are possible. In the future, the miniDOAS dataset will be highly valuable to do analyses per season or per station, on a higher temporal resolution than is possible with the AMOR data. The interpretation of the current dataset may change if more data becomes available and if we gain more insight in the performance of the instrument over time. For

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now, a subdivision of the dataset seems not feasible, especially not the intercomparison between AMOR and miniDOAS data; per station, there are only 12 to 16 monthly data points available.

(comment from referee)

Authors should add a description of the expected maintenance schedule and expected lifetime for DOAS instruments and their major components for the DOAS instruments.

(author's response)

Thank you for this suggestion. We add a short section to the manuscript (page 5, line 12):

(author's change in manuscript)

### 2.3.2 Maintenance schedule and instrument lifetime

Based on the experience gained during this intercomparison, we now adhere to the following maintenance schedule:

- The xenon lamp is exchanged once a year. Because replacing the lamp makes measuring new reference spectra necessary (see Sect. 2.3.4), such a replacement takes place by exchanging a system with a life-expired lamp with a system with a new lamp installed and newly measured reference spectra present. This minimises the instrument downtime.
- About half a year after a system is installed at a station, it receives a service visit. The system is cleaned and the alignment of the optics is checked.
- The quartz window in the station housing is cleaned every four weeks.
- The parabolic sender mirror is replaced after 5 years of continuous use.

We estimate the lifetimes of the other optical components – the receiver mirror, the folding mirror and the interference filter – to be between 5 and 10 years. The electronic

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parts – the alignment correction motors, the spectrograph and the instrument computer – also have lifetimes between 5 and 10 years. The remaining parts – the breadboard and the mirror mounts – have lifetimes of more than 25 years.

(comment from referee)

Minor comments Replace “Uptime” with “Data capture rate”

(author’s response)

We propose to ask the English language editor for advise on this change.

(comment from referee)

Replace “life-expired” with outdated in reference to old instrumentation; and with “burned-out” in reference to burned-out LP-DOAS lamp

(author’s response)

“Outdated” expresses the opinion that the instrument is no longer state-of-the-art, or even that it is old-fashioned and no longer suitable for use. We intended to denote that the instruments reached the end of their economical lifespan, with repairs becoming more frequent and more costly. We propose to ask the English language editor for advise on this change.

“burned-out”: we replace the lamps preferably before they burn out, so this is probably not a good change to make.

(comment from referee)

Page 2, line 16 (and through the rest of the document) - abbreviations shall be presented in parenthesis when first used in the text, and not another way around. E.g. – Differential Optical Absorption Spectroscopy (DOAS).

(author’s response)

We changed all instances, on page 2, line 16; page 3, line 11; page 4, line 7 and page

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5, line 9.

(comment from referee)

Page 4 – combine sections 2.2.1 through 2.2.4 into single section 2.2.

(author’s response)

Thank you for this suggestion, we merged these sections.

(comment from referee)

Page 6, Equation 6 - should be  $2l$  in denominator.

(author’s response)

In this equation, as in equations 1 to 5,  $l$  is the full optical length, i.e. twice the distance between the instrument and the retroreflector. We made this more explicit in the definition, on page 5 line 20:

(author’s change in manuscript)

...  $l$  is the length of the full optical path. In a monostatic system like ours,  $l$  is twice the distance between the instrument and the retroreflector.

(comment from referee)

Page 10, line 28 – replace “overview of the situation” with “measurements setup”.

(author’s response)

We made this change.

(comment from referee)

Pages 13-15 – combine section 4.1 and 4.2, and 4.4. Pages 14-15 – eliminate subsection in 4.3 and make a more fluid narrative (e.g. section 4.3.1 can be reduced to a single sentence or illuminated completely as it was already discussed in section 3.2).

(author's response)

Thank you for the suggestion. We made both these changes. The revised Section 4 is included as a supplement to this comment.

(comment from referee)

Please provide explanation for describing the DOAS instruments deployed in the network as miniDOAS. What is the main feature that differentiates the miniDOAS from a "traditional" LP-DOAS instrument?

(author's response)

That is indeed not clear from this paper. We added a short explanation on page 5, line 11:

(author's change in manuscript)

It was developed from a much larger system, also described in Volten et al. (2012). By using smaller and less expensive parts, the physical dimensions, the power consumption and the price tag of the miniDOAS were much smaller than the original system, hence the name. See Sect. 2.3.4 and Volten et al. (2012) for an overview of the differences between the systems.

(comment from referee)

Table 1 can be augmented to include physical dimensions of the different instruments.

(author's response)

With the exception of the miniDOAS, no dimensions were published for any of the instruments listed in Table 1. This makes adding the dimensions to the Table not feasible. We agree that information on the size of the instrument can be useful, we therefore added a photograph of the instrument (Fig. 3, referred to on page 5, line 11), with the physical dimensions in the Figure caption. This Figure is included at the end

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of this author's response.

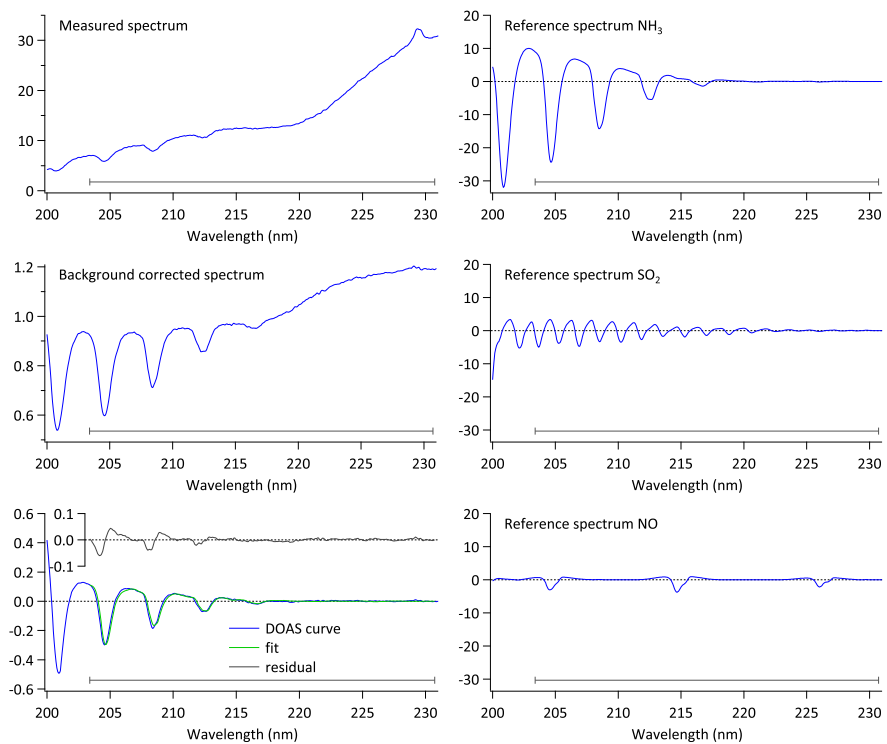
Please also note the supplement to this comment:

<https://www.atmos-meas-tech-discuss.net/amt-2016-348/amt-2016-348-AC2-supplement.pdf>

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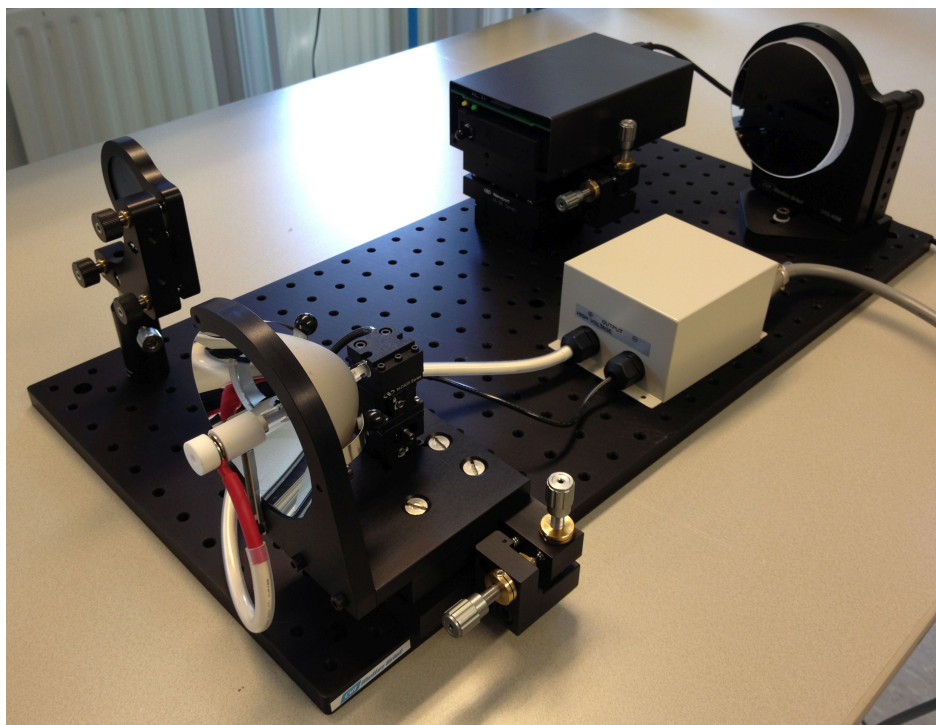
Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2016-348, 2017.

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**Fig. 1.** (see the Supplement to this comment for the full figure caption).

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**Fig. 2.** Photo of the miniDOAS. The instrument is 380 mm wide, 600 mm long and 180 mm high. Not shown: the lamp power supply (282 × 144 × 90 mm) and the instrument computer.

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