

We thank the two reviewers for their constructive and helpful comments. In the following we address the comments and suggestions by the reviewer's point-by-point.

Reviewer 1

- The nomenclature of the BIPM for the expression of uncertainty should be followed.

-> Authors: We use systematic and random effects in the miniDOAS measurements following the BIPM guidelines. We characterise the uncertainty of systematic and random effects and avoid the expressions precision and accuracy. The overall uncertainty will be derived from a combination of the two. The uncertainty of systematic effects includes uncertainty in differential absorption cross-sections (σ_i) derived from calibration. It also includes uncertain estimation of concentration offsets ($c_{ref,i}$). Random uncertainty is quantified by the concentration standard error (se_i) statistically derived from the multiple linear fit. The se_{NH3} depends linearly on the concentration (see figure 6). The lower boundary is given by the uncertainty of the intercept of the linear regression of se_{NH3} versus concentration. The random uncertainty estimate is supported by the scatter between the two miniDOAS instruments during the parallel measurements at the BTEP campaign.

- Page 1 Line 24: The instrument precision is given in "%". It should be indicated, however, that this is relative to the determined concentrations.

-> Authors: We agree and accordingly change the text

Page 1 Line 24: The statement "Accuracy is larger than precision" is trivial and should be deleted.

-> authors: We take out this sentence; at a first glance it seems trivial, but it is possible that the precision can be lower than the accuracy-. This could e.g. be the case for low light levels when the signal to noise ratio in the measured spectrums is getting very small.

Page 1 Line 24-26: It would be preferable for the reader to have a "combined uncertainty" including the different systematic and random sources of error, to estimate the potential of the presented technique. The most important error sources might be given in addition.

-> Authors: agreed, see comment above

Page 1 Line 26: The acronym "I0" is defined as the light intensity emitted by the light source (page 3 Line 23), which is common definition (i.e. Beer Lambert law). Here the term "reference spectrum" is used for I0, please correct. In addition the term I0 is used throughout the manuscript for I with "baseline concentrations", this leads to severe misunderstanding and should be corrected – it might be best to introduce an additional acronym.

-> Authors: we agree and catch up with the reviewer, introducing I_{ref} as our 'reference spectrum'. I0 was exchanged by I_{ref} throughout the manuscript (but not in Eq.1, since this is the Lambert-Beer Law). In order to improve consistency and readability, we present the

most important evaluation steps in form of Equations and refine our acronyms and definitions as stated in the following table.

Old naming	New naming	Meaning	Remarks
I_0		initial intensity of the light beam emitted by the light source	we introduce a new acronym (see row 3) for the "reference spectrum" used for our DOAS evaluation, so that I_0 can remain exclusively for the presentation of the Lambert-Beer law
	I_{dark}	dark spectrum, recorded with blocked spectrometer inlet	same dimensions as I_{ref} (see next row); represents the spectrometer background signal at given temperature and CCD integration time
I_0	I_{ref}	spectrum derived from measurements with the instrument, used as reference spectrum; I_{ref} is corrected for I_{dark}	in case of the used DOAS: a vector with the length of 274, covering the wavelength window from 203.7 to 227.8 nm with 274×58 CCD pixels
I		measurement spectrum: spectrometer reading as a result the beam passing through a layer of thickness L_{path} ; I is corrected for I_{dark}	same dimensions as I_{ref}
I_{div}		$I_{\text{div}} = I / I_{\text{ref}}$	
σ_i		absorption cross-section of an absorbing gas i	
	{...}	brackets used to indicate high-pass filtering according to our presented procedure	
local regression baseline filter / rfbaseline	REBS	REBS = "robust extraction of baseline signal"	this is the expression used in the original

			publication by Ruckstuhl et al.
	<...>	brackets used to indicate low-pass filtering according to the presented procedure	low-pass filter meaning loess(RBES(...))
$I_{div} = I_{div} / \langle I_{div} \rangle_{lowp}$	$I_{div} / \langle I_{div} \rangle$	we omit I_{div} , but term this intermediate step $I_{div} / \langle I_{div} \rangle$ directly	we will present these steps in the form of equations instead of mentioning it in the text
$I_{div} = I_{div} / \langle I_{div} \rangle_{lowp}$	$\{I_{div}\} = I_{div} / \langle I_{div} \rangle / \langle I_{div} / \langle I_{div} \rangle \rangle$	we also omit I_{div} and use $\{I_{div}\}$ to indicate high-pass filtering of I_{div} according to our procedure	
differential absorption cross-sections	Θ_i	obtained from known amount of absorbing gas: $\Theta_i = \ln(\{I_{div}\}) / c_{cal,i}$, where $c_{cal,i}$ is known from calibration by the amount of calibration gas i over the cuvette path ($c_{cal} = C_{cuvette} * \text{length}_{cuvette}$)	
differential optical density	$D = \ln(\{I_{div}\})$		we switch to the term "optical depth" instead of "optical density" here, i.e. differential optical depth = $D = \ln(\{I_{div}\})$
C_{0i}	$C_{ref,i}$	mean concentration of absorbing gas on L_{path} during the definition period of I_{ref}	
C_i		mean concentration of absorbing gas on L_{path} as determined during the period of measurement of I	
concentration standard error of gas i	se_i	standard error of the estimates of c_i as derived from the multiple linear regression	R functions: lm (package: stats), arima (package: stats)

Page 1 Line 27: The term “minimum accuracy” might lead to misinterpretation, please replace.

-> Authors: see comments above.

Page 1 Line 27: The term “the limit of detection against I_0 ” is not correct and might not be used, please rephrase.

-> Authors: changed into “the limit of detection against c_{ref} ”

Page 2 Line 32: The manuscript switches between “ $\mu\text{g m}^{-3}$ ” and “ppb”, please unify here and elsewhere in the text.

-> Authors: we are using “ $\mu\text{g m}^{-3}$ ” throughout the manuscript

Page 2 Line 49: The term “on its breadboard” might be deleted.

-> Authors: done

Page 2 Line 60: “. . . DOAS data evaluation procedure”?

-> Authors: As suggested we changed to “. . . DOAS data evaluation procedure”

Page 2 Line 63: What the meaning of “finite” in this respect?

-> Authors: We meant constrained emission areas, such as small fields or experimental plots as opposed to the micrometeorological idealised infinite source/sink area. For clarification, we delete the word “finite”.

Page 4 Line 103: In the experiment W3 the authors claim they used three miniDOAS downwind, here it is stated that two instruments were used, what is correct? Is the numbering of the instruments (S2 and S5) relevant or just for internal use, otherwise it is confusing the readers only, why not replacing it by N1, N2, . . . for the new instruments and O1 for the old one.

-> Authors: We characterise the newest Swiss miniDOAS model by presenting results obtained with two identical instruments (N1/N2), but we have to refer to the older Swiss model (O1) in experiment W2, where it measured the inflow concentration in addition to N1/N2. We rephrase:

“In this article, we generally present measurements with two instruments (called “N1” and “N2”). Only one experiment (W2, Sect. 2.3) includes additional measurements with an older miniDOAS version (“O1”) ...”

Page 4 Line 123: Please rephrase the term “The purpose is the elimination of P . . .” to something like “This is accomplished by high-pass filtering of I_{diff} . . .”

-> Authors: done, we will use the suggested phrase

Page 4 Line 125: Cross-sections were fitted to the measurement spectra I assume?

-> Authors: Since we now introduced I_{ref} , we describe $I_{div} = I / I_{ref}$. Above, we define the term “differential optical depth” $= D = \ln\{I_{div}\}$, now used consistently in the manuscript. For the

concentration calculation, "differential absorption cross-sections" (Θ_i) are fitted to the differential optical depth. We re-write the section in order to clarify these circumstances.

Page 4 Line 126: For which parameters was the gas cell controlled? Please specify.

-> Authors: this now reads: "... were derived by calibration with a gas cell containing known amounts and can be compared to literature values. For NH_3 we used a concentration of $163000 \mu\text{g m}^{-3}$ in a cuvette with a length of 0.075m. The ambient pressure was 963 mbar and the temperature 298°K"

Page 5 Line 130: The phrase "across that range" is unclear and might be corrected.

-> Authors: We delete "across that range"

Page 5 Line 138: The term "alpha sampler" might not be familiar to every reader, please change to "alpha passive ammonia samplers" or similar. You might state here that you used active and passive sampling devices for inter-comparison.

-> Authors: agreed, we changed the passive sampler statement as suggested. Since this passage is about our approach to determine c_{ref} (done with the alpha passive samplers), we mention the active devices elsewhere.

Page 6 Line 175ff: The abbreviations for the field experiments are seem arbitrary. It might be better to number the experiments consecutively.

-> Authors: yes, we clarified this.

Page 6 Line 188ff: Could you please give more details on the impinger measurements, e.g. flow rates, acid etc. or at least cite corresponding literature?

-> Authors: The citation of Häni et al. (2016) had accidentally been moved to p.12 l. 359. Häni et al. (2016) provide details about the used devices. We re-located the citation to Sect. 2.3.1 and added that we used the devices similar to Häni et al.

Page 7 Line 202: The sentence "and in parallel alpha passive samplers" is incomplete – no verb.

-> Authors: added another "run"

Page 7 Line 215: The term "IHF" should be defined when used first, but is defined later.

-> Authors: We changed that into the right order

Page 7 Line 218ff: Which experiments are described here? I assume W3. . .

-> Authors: Yes, we added the ID.

Page 8 Line 218ff: As details on the NH_3 emissions are available and given in the text this would be the ideal experiment to compare to NH_3 emission estimates determined by the DOAS technique – Why the comparison is limited to concentrations and not emissions?

-> Authors: The goal in the current manuscript was to show an example of a vertical concentration differences (caused by emissions) and showing simultaneous measurements of the volcano plume rising the SO_2 concentrations during the experiments. This offered a good opportunity to demonstrate the instrument's capacity for simultaneous NH_3 and SO_2 measurements (the latter could be compared to monitoring stations) and allowed for an additional interference check. We will, present this experiment in more detail in a forthcoming publication.

Page 8 Line 254: The term “slightly” is qualitative, what does this mean in numbers?

-> Authors: In an ideal case differential absorption cross-sections should be independent of the absorption strength (i.e. number of molecules on the path) they have been derived from. But in practice they can depend on the used high-pass filter due to filter performance. Figure 3 shows two types of high-pass filters for NH_3 absorption: a) the moving average-type filter as used by Volten et al. (2012) and b) the robust extraction of baseline signal (RBES) for 7, 70 and $700 \mu\text{g m}^{-3}$. The deviations are non-linear and will depend on the moving average filter's parameters (such as the width of the running window). They are in the order of up to 3% over the analysed range.

Page 10 Line 290-292: Spectral interferences are given in “%”, which is arbitrary, it would be better to give it in “ $\mu\text{g m}^{-3}$ / $\mu\text{g m}^{-3}$ ” or similar.

-> Authors: The unit will be changed to “ $\mu\text{g m}^{-3}$ / $\mu\text{g m}^{-3}$ ”.

Page 10 Line 302: The term “the scatter between both instruments” should be rephrased.

-> Authors: We changed the text to «and a scatterplot of the two concentrations»

Page 10 Line 307: It's hard to understand how the standard errors was estimated; I assume it's based on the AMRA or OLS fit; but should be mentioned here also.

-> Authors: Standard errors were determined from the OLS and ARMA fit, respectively. We will explain this also in section 2.2

Page 10 Line 312: It is not a “concentration difference between S2 and S5” but a “difference in concentration determined with S2 and S5”.

-> Authors: We changed the text as suggested

Page 11 Line 324: On line 317 it was stated that the precision is 0.8 to 1.4 %, here a precision of 1.4 % is given, which value is correct?

-> Authors: The two instruments showed different se_{NH_3} . For N2 it was evaluated to 0.8%, whereas for N1 it was 1.4% of the concentration.

Page 11 Line 324: The term “the LOD $\mu\text{g m}^{-3}$ ” seems awkward.

-> Authors: Overall, the NH_3 limit of detection against c_{ref} , approximated as three times the median standard error, is $0.2 \mu\text{g m}^{-3}$. Unfortunately higher integration times e.g. up to 30 minutes did not reduce this value.

Page 11 Line 329: What is an “alpha sampler batch” please clarify?

-> Authors: The ALPHA sampler “batches” are passive samplers (Tang et al. 2001) deployed in triplicates. Once the filter paper within each sampler has been analysed for ammonium, and the laboratory blank concentrations have been subtracted, the coefficient of variation between the NH₄ concentrations gives their precision. The CV values are 6.9%, 2.6% and 3.9% for the A, B & C ALPHA sets along the miniDOAS path. This gives an average of 4.5% which is a very good level of precision.

Page 11 Line 339: The sentence “Since the signal-to-noise ratio with the baseline and I₀ is inevitably larger than with I and I₀, ...” is hard to follow, please rephrase.

-> Authors: Instead of using I_{ref} for the calculation of I_{div} , one could also use another baseline of I to substitute I_{ref} in the calculation of I_{div} . This baseline was derived with the low-pass filter (loess(RBES(...))), i.e. the alternative version of $I_{div} = I / \langle I \rangle$. $\langle I \rangle$ is smooth, hence this alternative I_{div} has a smaller signal to noise ratio than $I_{div} = I / I_{ref}$. We clarify this sentence.

Page 11 Line 341: The term “the absorption present in I₀” is confusing as I₀ is the intensity of light emitted by the light source so without absorption.

-> Authors: The changes introduced in the table above clarify this point.

Page 12 Line 362: “... which allows an independent concentration determination”?

-> Authors: changed to: which allows an independent comparison of the concentrations

Page 13 Line 414: The correct term is “Swiss national air pollution monitoring network”.

-> Authors: corrected

Page 14 Line 423: The term “NH₃-SO₂-NO cross-interference” is not correct as no NO variation was observed and only NH₃ and SO₂ varied.

-> Authors: we agree and changed to “NH₃-SO₂ cross-interference”.

Page 14 Line 443: The authors mention the possibility to use a trace dilution technique with SO₂ or NO dosing. What is the LOD and the toxicity (e.g. MAK) for this components? I assume this approach might be too dangerous, please give a statement.

-> Authors: The LOD for NO and SO₂ is ca. 4 times higher as for NH₃ (on a molecular density base). Using an artificial source in a similar way as in the Witzwil experiment, the concentration of NO and SO₂ need to be increased from 5% to about 20% to get similar precision as for NH₃ for an identical setup. NO concentration easily are affected by traffic emissions. The handling of mixing ratios of 20% NO and SO₂ would also need safety requirements and cannot be done e.g. in closed rooms.

Reviewer 2:

It seems that reviewer 2 refers to an older manuscript version that we submitted at the beginning, so that line numbers indicated in his review deviate from the corresponding line

numbers in the discussion paper. We tried to find the given statements from the manuscript version that we think reviewer 2 refers to and hope to have addressed the right points. When we refer to page or line numbers in our replies, we refer to the manuscript as published in AMTD.

- Section 2.4.: The order of the field experiments in Table 1 and in the text is different, and different types of information are given for each experiment. The authors should give a name for each experiment, or use the abbreviations (W2, W3, R1, R3, HAFL) also in the text, to prevent confusion.

-> Authors: We will re-name the experiments and use the abbreviations consistently.

- Table 1: More than one mini DOAS instruments were used in most experiments, but the number of instruments is not given in Table 1. There are also discrepancies between path lengths given in Table 1 and in the text in Section 2.4 (e.g. 72 m and 76 m for the experiment W3).

-> Authors: we will specify the exact path lengths used and heights for the three DOAS systems going from low middle to high (Path lengths: 72.6m, 73.6m, 76.4m Heights: 0.49m, 1.25m, 3.01m)

- Figure 2b: Please give the concentration of SO₂ and NO as well.

-> Authors: SO₂ = 76.3 mg/m³, NO = 594.0 mg/m³; we put that into the fig. annotation; also changed in the text (p.3, l.83).

- Figures 2, 3 and 4: Can you evaluate, whether the differential absorption cross sections above 0 are within the expected uncertainties, or do they show a significant systematic bias of the instrument?

-> Authors: Differential optical depth values > 0 occur due to the behaviour of the high-pass filter and are not avoided - even with our advanced procedure. Differential absorption cross-sections from calibration are fitted to the differential optical depth from ambient measurements. Both curves are created using the same method so that no bias is introduced. We show in fig. 3 that this behaviour is consistent across a large concentration range. It does not represent a systematic bias of the instrument but is a typical feature of the evaluation of the concentration from a measured spectrum I. We add a clarifying statement in Sect. 2.2 and introduce the expression 'differential optical depth' for the measurement curves.

- Figure 4: Could you give differential absorption cross sections instead of differential optical density, to make the figure directly comparable to Figures 2 and 3?

-> Authors: no, since we would need knowledge of the true concentration a-priori for that. However, fig. 4 (vertical middle panel) displays measurement differential optical density (black) and fitted differential absorption cross-section from calibration. This already could be the required comparison as it demonstrates how both curves correspond to each other. This point should become clearer by the introduction of 'differential optical density'.

- Page 11 lines 4-5: How do you determine the errors from the 1 minute spectra? For which instruments do these errors apply? Are the errors for both instruments comparable?

-> Authors: These are derived the individual standard errors from the multiple linear fit, as indicated on p.10, l. 1-2. Fig. 6 shows the standard errors from both instruments versus NH_3 concentration. Slopes are 0.8 and 1.4 %.

- Page 11 line 19: The intercept is indeed rather small, however, “practically zero” would mean that zero is within the 95 % confidence interval limits. This is not the case in the presented example.

-> Authors: True; the 95% confidence interval of the intercept is -0.011 to -0.003 ug/m^3 , which is very close to zero. We change 'practically' to 'close to'.

- Page 12 lines 1-3: Is it possible to record the zero spectrum at a remote place, and use it afterwards in experiments carried out at other sites? Or does re-location or transport of the instrument affect the zero spectrum? How often do you recommend checking the zero spectrum?

-> Authors: It could be used in other experiments, because broadband deviations caused by various changes would be filtered out. However, it is our experience that the measurement is more precise when the zero spectrum resembles conditions close to those during the measurements. Different reflectors can show different broadband characteristics and consequently, we recommend that spectra should always be recorded with identical optical components. We also recommend to use a similar path length for both, the reference spectrum and for a specific measurement series in order to achieve lowest measurement uncertainties. The quantification of c_{ref} is a challenge and the practical solution for this depends on experimental requirements for precision and accuracy. For best results we recommend using a local 'zero' spectrum for each experiment and determine c_{ref} with a reference measurement (e.g. passive samplers) of known accuracy. With long-term measurements it seems desirable to collect experience with the applied system by repeating checks periodically, e.g. every 2 months, and then adjust the frequency according to the results of these checks.

- Section 3.3.1.: In my opinion, comparison of the DOAS instruments to an impinger system gives information about the instrument performance. I would recommend moving this part to Section 3.2.

-> Authors: agreed

- Figure 7: What does NH_3 on the y axis mean? Is it ammonia concentration or ammonia gradient? If it is concentration, with which instrument did you measure it? If it is gradient, the unit should be $\mu\text{g/m}^3\text{m}$, i.e. concentration difference divided by the distance of the instruments.

-> Authors: it's the concentration difference which was indicated in the original figures. We didn't discover during submission that special characters were not rendered in the final pdf. They will be there in the final manuscript. We replace the term concentration gradient with concentration differences to avoid any misunderstandings.

- Figure 9: How do you explain the negative NO concentrations? Is it a cross-sensitivity to SO_2 ?

-> Authors: Negative concentration can occur here, since we didn't correct for c_{ref} as indicated in the figure annotation. However, the change in NO with NH_3 from the gas-release could be interpreted as interference. This is discussed on p.14. l.425-429.

- Please make sure that you use uniform units for concentration throughout the paper. In the current version ppb, ppm, $\mu g/m^3$ and $\mu mol/m^3$ are alternately used, making it difficult for the reader to compare the different values.

-> Authors: partly done (fig. 3 and according text passage: ppb exchanged with ug/m^3). In fig. 9 we compare concentration time courses of the three gas species and also discuss their cross-interference behaviour from that. The better comparability between the gas species was the reason to switch to $\mu mol/m^3$ here. In fig. S4, standard error ranges from the three gases are contrasted in $\mu mol/m^3$ for the same reason. In the text, where we address the performance of the instrument for the gases individually, we stick to $\mu g/m^3$.

Technical comments:

-> Authors: We followed the reviewer's recommendations for technical issues as suggested and are not specifying these corrections further.