Interactive comment on "Monitoring shipping emissions in the German Bight using MAX-DOAS measurements" by André Seyler et al.

Anonymous Referee #1

Received and published: 23 February 2017

General comments

The manuscript entitled "Monitoring shipping emissions in the German Bight using MAX-DOAS measurements" presents remote sensing observations of NO2 and SO2 along a main shipping route towards the harbour of Hamburg. Ship emissions significantly contribute to the air pollution in these areas, and a monitoring of the air quality, in particular with respect to the impacts of the recent regulations of the fuel sulphur content, is of high scientific and political relevance. Therefore the topic of the manuscript is well suited for publication in ACP.

The paper is well structured and the scientific approach is clearly described. It provides a comprehensive introduction into the subject of ship emissions. The data is interpreted regarding the contribution of land- and seaborne emissions in a systematic way and the impact of the reduction of fuel sulphur content on atmospheric SO2 levels is discussed on the basis of statistical analyses. However, there are several aspects regarding the interpretation of the data which need to be revised. In particular, my impression is that the impact of horizontal inhomogeneities on the measurements and the fact that the remote sensing measurements average over a certain altitude range need to be considered more carefully. It appears to me that the latter is the main reason for the discrepancy between MAX-DOAS and in situ, which can and should be corrected for by accounting for the different vertical distribution of O4 and the target gases and thus different AMFs, as done during previous studies (e.g., Sinreich et al., AMT, 2013).

First, we would like to thank Anonymous Referee #1 for his/her helpful comments. Below, we reply point-by-point to the specific comments . As far as possible, we have considered the suggestions in the revised manuscript.

Specific comments

L56: Do you refer to fuel consumption and emission per vessel or in total (the latter would be obvious given the large increase in the number of ships).

We (and the cited studies) are referring to total fuel consumption and emissions. Following your suggestion, we included the word "total" to make this clearer:

"At the same time, *total* fuel consumption and emissions increased as well (Corbett and Koehler, 2003; Eyring et al., 2005a,b; Eyring et al., 2010b)."

Section 1.2:

Maybe the discussion of halogen chemistry should be removed since it is not of relevance for the present study. To my knowledge, the role of halogen radicals in polluted air is not well understood, and it is unclear whether the NO + XO reaction is of importance. In clean air, the conversion of XO to X proceeds either via self-reaction or reaction with HO2. In polluted air, reaction with NOx is likely to lead to a removal of halogen radicals by formation of halogen nitrates.

We removed the corresponding paragraph.

Section 3.1:

It could be mentioned that Equation (2) follows from (1) if the temperature and pressure dependence of the absorption cross section can be neglected.

Thank you very much for your thorough proof-reading, we had forgotten to include this important information. The corresponding paragraph now reads:

"Neglecting the temperature and pressure dependence of the absorption cross sections, polynomial and differential cross sections are fitted to the measured optical thickness ln (I/I0) in the linearized so-called DOAS equation:"

Section 3.2:
Please provide technical information on the fibre bundle (number of fibres, diameter, arrangement at both sides).
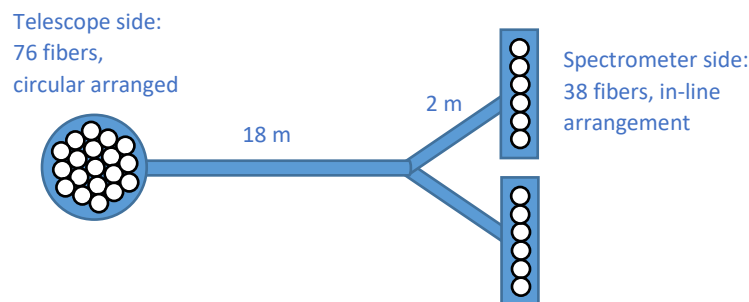
Type: Y-shaped quartz fiber bundle
Number of fibers: 2 x 38 = 76 single fibers
Diameter: 150µm each,
Length: 20m
Arrangement sketch:

Telescope side:
76 fibers,
circular arranged

2 m

Spectrometer side:
38 fibers, in-line
arrangement

18 m

We added this information to the text of the manuscript.

What are the wavelength ranges of both spectrographs?

We added the following sentence:

"The UV instrument covers the wavelength range 304.6-371.7 nm, the visible spectrometer covers 398.8-536.7 nm."

I am confused about the definition of the elevation angle. Usually, it should be the centre of the field of view, but here the definition is unusual and rather unspecific, being something like the lower edge of the field of view, which yields an offset to the common definition of 0.6_. Please specify the elevation angle as the centre of the field of view throughout the paper.

We changed the elevation angles to the actual values (center of field-of-view) throughout the paper.

Section 3.3:
It is important to include a discussion of the fit errors, detection limits and RMS residual for the target gases.

We added a paragraph on this in Section 3.3:

"Under good conditions, the typical fit RMS is around 1×10^4 for NO_2 in the visible, 2×10^4 for NO_2 in the UV and 5×10^4 for SO_2. By assuming that an optical density of twice the RMS can be detected (Peters, 2013), it is possible to estimate the detection limit of our instrument regarding the different trace gases. The differential absorption cross section of NO2 is in the order of 1×10^{-19} cm^2/molec, for SO_2 in the order of 2×10^{-19} cm^2/molec. Combining this yields a NO_2 detection limit of around 1×10^{15} molec/cm^2 corresponding to 0.05 pbb in the visible and 2×10^{15} molec/cm^2 corresponding to 0.1 pbb in the UV. The SO_2 detection limit is around 2.5×10^{16} molec/cm^2 corresponding to 0.2 pbb.

The typical absolute fit errors are 2-3 * 10^14 molec/cm² for NO_2 in the visible, 5-6 * 10^14 molec/cm² for NO_2 in the UV and 2 * 10^15 molec/cm² for SO_2, which is a factor of 5 to 10 smaller than the detection limit."

L345ff: Apart from the ozone absorption, a limitation for a retrieval window at shorter wavelengths is the lower light intensity.

This sentence was changed to the following:

"This results from the need to find a compromise between *the low light intensity caused by* the strong ozone absorption around 300nm on the one hand and the rapid decrease of the differential absorption of SO_2 at higher wavelengths on the other hand, limiting the choice of the fitting window."

Section 3.4:
The definition of the volume mixing ratio and its calculation from number concentration is well known and there is no need to discuss this here.

We removed the unnecessary explanation of volume mixing ratios and have completely rewritten this section focussing on the O_4 scaling approach and its limitations.

The uncertainties of the O4 scaling approach need to be discussed. For example, O4 usually has a profile shape very different from NO2 and SO2, which violates the basic assumption that the O4 dSCD is a good proxy for the light path through the NO2 and SO2 layers. Other studies use correction factors from radiative transfer calculations to account for this (Sinreich et al., AMT, 2013). Furthermore, the resulting near-surface volume mixing ratios will not be representative for the amount of trace gases directly at the surface, but for some kind of average over a certain height range in the boundary layer. There is also "light dilution", i.e. light scattered into the line of sight between the instrument and the trace gas plume (see e.g. Kern et al., Bulletin of Volcanology, 2010), which further reduces the measured SCDs . My impression is that these are the main reasons for the discrepancies between in situ and MAX-DOAS, and not horizontal inhomogeneities as speculated later in the paper by the authors (these would cancel out when averaging the data). The discussion of the data needs to be revised accordingly in order to account for the influence of these aspects.

We have completely rewritten this section discussing in detail the limitations of the O_4 scaling approach according to Sinreich et al. (2013)[1] and Wang et al. (2014)[2] as well as explaining the reasons why the suggested correction factors have not been applied to the data in this study: The height of the NO_2 and SO_2 layer is unknown and no additional measurements of the layer height exist. Furthermore, a comparison to our in-situ measurements indicates that the layer height and therefore the correction factors vary from day to day as well as over the course of individual days. In addition to that, an extensive RTM study like it was performed by Sinreich et al. (2013) and Wang et al. (2014) was out of the scope of this publication and the comparision to our in-situ instrument not the main point of the paper.

Equation 5: It should be mentioned that nO4 is the O4 concentration at the surface.

We added this information.

The remarks regarding the elevation angle from Section 3.2 are repeated at the end of this section. See my comments above. A deviation of 0.5_ in elevation angle is certainly not negligible at very low elevation angles.

[1] Sinreich, R., Merten, A., Molina, L., & Volkamer, R. (2013). Parameterizing radiative transfer to convert MAX-DOAS dSCDs into near-surface box-averaged mixing ratios. *Atmospheric Measurement Techniques*, *6*(6), 1521–1532. https://doi.org/10.5194/amt-6-1521-2013

[2] Wang, Y., Li, A., Xie, P. H., Wagner, T., Chen, H., Liu, W. Q., & Liu, J. G. (2014). A rapid method to derive horizontal distributions of trace gases and aerosols near the surface using multi-axis differential optical absorption spectroscopy. *Atmospheric Measurement Techniques*, *7*(6), 1663–1680. https://doi.org/10.5194/amt-7-1663-2014

We changed the elevation angles to the actual values (center of field-of-view) throughout the paper and removed the remarks. We also added a discussion on the limitations of the O4-scaling due to non-consideration of correction factors (see also above).

L474: It is not obvious to me why a thicker trace gas layer should lead to a reduction of the ratio between dSCDs near the horizon and in zenith. Wouldn't horizontal inhomogeneities, with more NO2 over the shipping lane than over the instrument, be a much more likely explanation for these findings?

This Paragraph is not about differences between SCDs near the horizon and in zenith sky direction, but about systematically higher NO_2 and SO_2 DSCDs in 0.5° Elevation compared to the 2.5° Elevation for wind from the shipping lane, indicating a low pollution layer over the shipping lane. For southerly winds, on average the NO_2 and SO_2 DSCDs in 0.5° and 2.5° Elevation are nearly equal. We have rephrased the paragraph to make it clearer:

"The highest NO_2 and SO_2 DSCD in the lowest elevation angle (0.5°, blue bars) in relation to DSCDs in higher elevations are measured especially for wind from all northern directions, in a sector ranging from WSW to ESE. These directions coincide with the course of the main shipping lane, which comes from the WSW direction (the English Channel, the Netherlands, East Frisian Islands), passes the island in the north and runs close to the city of Cuxhaven (ESE direction) into the river Elbe. This indicates that these enhanced columns in the 0.5° elevation angle is pollution emitted from ships in a surface-near layer.

For southerly wind directions, where no larger shipping lane is in the direct surrounding and land-based pollution sources dominate, the average DSCDs in 0.5° and 2.5° elevation are nearly the same for both NO_2 and SO_2."

Section 4.2:
The title of this section is too long and complicated. I suggest to replace it by something like "Volume mixing ratios of NO2 and SO2"

Done.

Section 4.5:
As already stated above, the fact that MAX-DOAS averages over a large horizontal distance should cancel out on temporal average when comparing to in situ measurements. Instead, a more probable explanation for the systematically lower mixing ratios is the fact that MAX-DOAS averages over a certain altitude range and that the differences in O4 and target gas profile shapes has not been considered. Light dilution will also play a certain role. The argument that MAX-DOAS yields lower values when the plume is orthogonal to the viewing direction does not seem convincing to me, because in this case the polluted air is also not transported towards the in situ instrument, which means that the in situ instrument might even miss particular plumes which are detected by MAX-DOAS.

This section has been completely rewritten discussing again the systematic deviations produced by the non-consideration of correction factors for the different profile shapes. Also a remark about light dilution as an uncertainty source reducing the actual measured SCDs was incorporated.

Section 4.6:
L655ff: NO2 concentrations at a particular location strongly depend on local sources, such as traffic, industry, domestic heating, etc., as well as on the distance to these sources and on the rate of vertical mixing. Therefore, the fact that amount of NO2 in background air observed in the Arctic is similar to the present study might be mere coincidence.

We have removed this paragraph.

Section 4.7:
L667: Detection limits are mentioned here for the first time. They should instead be discussed in Section 3.3.

We added a paragraph about detection limits in Section 3.3.

"By assuming that an optical density of twice the RMS can be detected (Peters et al., 2013), it is possible to estimate the detection limit of our instrument regarding the different trace gases. The differential absorption cross section of NO_2 is in the order of 1×10^{-19} cm^2/molec, for SO_2 in the order of 2×10^{-19} cm^2/molec. Combining this yields a NO_2 detection limit of around 1×10^{15} $molec/cm^2$ corresponding to 0.05 pbb in the visible and 2×10^{15} $molec/cm^2$ corresponding to 0.1 pbb in the UV. The SO_2 detection limit is around 2.5×10^{16} $molec/cm^2$ corresponding to 0.2 pbb."

Section 4.8:
An attempt is made here to separate shipping emissions from other sources by classifying the data according to the wind direction. The limitations of this approach need to be discussed more carefully. While I agree that northerly winds are little affected by background pollution, I strongly doubt that shipping emissions do not influence the measurements significantly when the wind is coming from the south. Data is filtered for light paths longer than 5 km, which means that for most observations the light path crosses the main shipping lane and probes air polluted by ship traffic. You reach this conclusion yourself in Section 4.1 (L438ff) in the context of the discussion of Figure 6, which shows that peaks from ship emissions clearly occur when air polluted by landbased sources is present. Thus, air masses classified as "Land" are likely to be partly affected by ship emissions.

You are right, air masses classified as "land" usually contain small fractions of shipping emissions as well. So the land source is slightly overestimated in these cases. How prominent ship emission peaks are in our measurements and how strong the contribution is compared to land based emissions depends on the wind direction. The wind sector classification is sketched in the following figure:

Blue sector: wind from open North Sea, shipping is the only pollution source

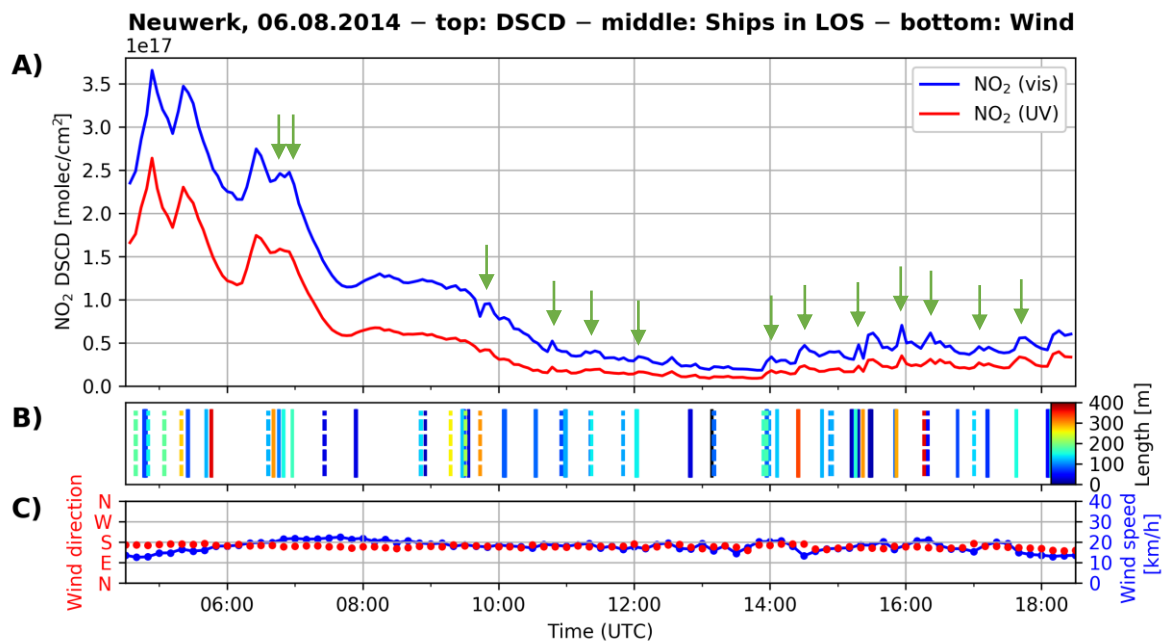Green sector: mainly land-based air pollution (traffic, industry, …)

Yellow sector: air mass contains shipping emissions as well as land-based air pollution (mixed origin)

On 23 July 2014, the day in Figure 6 which you are referring to, in the morning the wind was coming from NE-ENE, and later turned towards NNE. Those wind directions are not included in the sector we chose for pollution coming from land. Wind from those directions clearly can contain large fractions of shipping emissions as well as pollution from land and are therefore classified as directions with "mixed" pollution origin in the study.
When the wind is coming from southerly directions, ship emission peaks are much less prominent in our measured time series of NO_2 or SO_2. A day which illustrates this nicely is 17 July 2014, shown in the plot below. Until noon, wind was coming from the south and later changed to northerly directions. Measurements were done in 0° elevation towards north. In the morning, although a lot of ships are present, as can be seen from the colored bars in Panel B, ship emission peaks are very small and hardly visible. A few are still visible, marked by the green arrows. As expected, the peaks are higher in the visible than in the UV, because wind is blowing the ship plumes away from the radar tower and our instrument. In the afternoon, ship emission peaks are higher and much more prominent. The contribution of shipping emissions to the overall NO_2 measured is certainly much higher in the afternoon than in the morning.

Neuwerk, 17.07.2014 − top: DSCD − middle: Ships in LOS − bottom: Wind
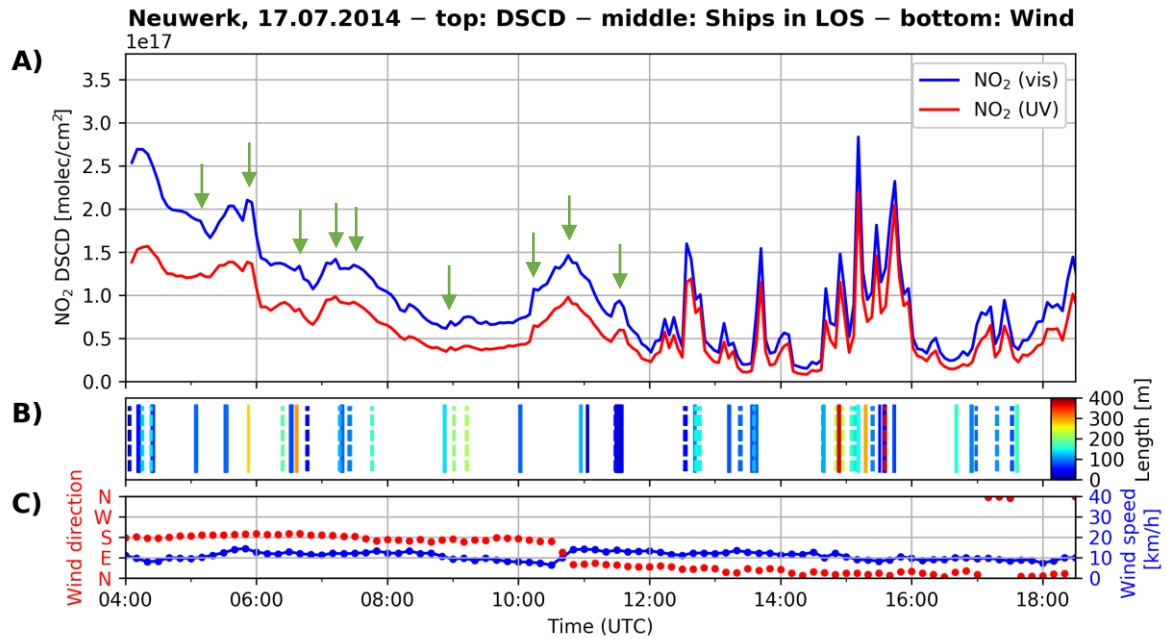
Another example is shown in the next plot for 6 August 2014.

Neuwerk, 06.08.2014 − top: DSCD − middle: Ships in LOS − bottom: Wind

To quantify the overestimation of the land source sector, the fraction of shipping emissions on the overall emissions on such a day has to be compared to the fraction of land sources. This is shown in the next plot: It is another example day with wind from southerly directions, 6 August 2014. Blue and red line show the NO$_2$ DSCD measured in the visible and UV. The cyan and magenta lines show the signal with removed ship emission peaks.

Neuwerk, 03.07.2014 − top: DSCD − middle: Ships in LOS − bottom: Wind
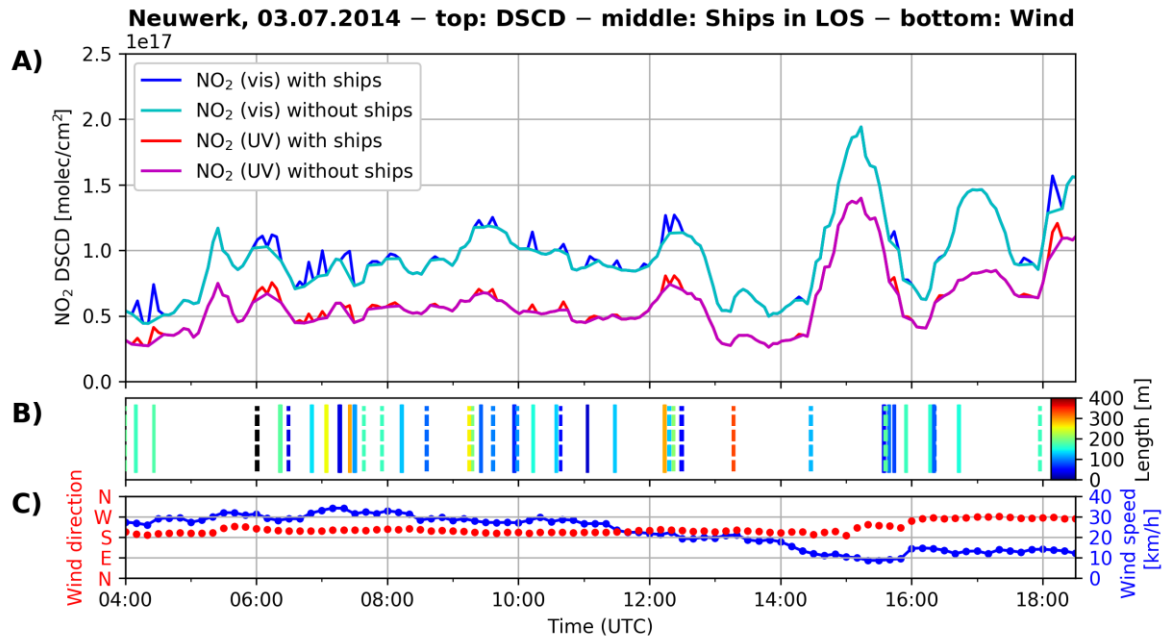
In the UV, the difference in the integral between "with ships" and "without ships" is around 1.4%, in the visible it is around 1.6%. So on this day, the NO₂ classified as "land source" is overestimated by around 1.5% due to shipping emissions which are still contained in the data set. In other measurement directions, to the NE or NW for example, this overestimation is be a bit higher, but never exceeds 3%.

So this overestimation is a small error and was therefore neglected in the study.

Section 4.9:
Given that SO2 scatters around a smooth (near-zero) background level, it is surprising to see that no negative SO2 to NO2 ratios were derived. It seems that negative values have been set to zero (Panel D of Fig. 20), which would significantly (and falsely) affect the statistics.

To address this shortcoming in our study we have completely redone the SO₂ to NO₂ ratio peak analysis. The baseline determination has improved substantially (using a second running median filter applied to the lower 50% quartile when necessary) and the positive bias in the measurements since 2015 is now gone. In addition to that, the section has been rewritten taking into account your comments. Also, the importance of NO to NO₂ titration especially for the comparison to other studies is now mentioned in this section.
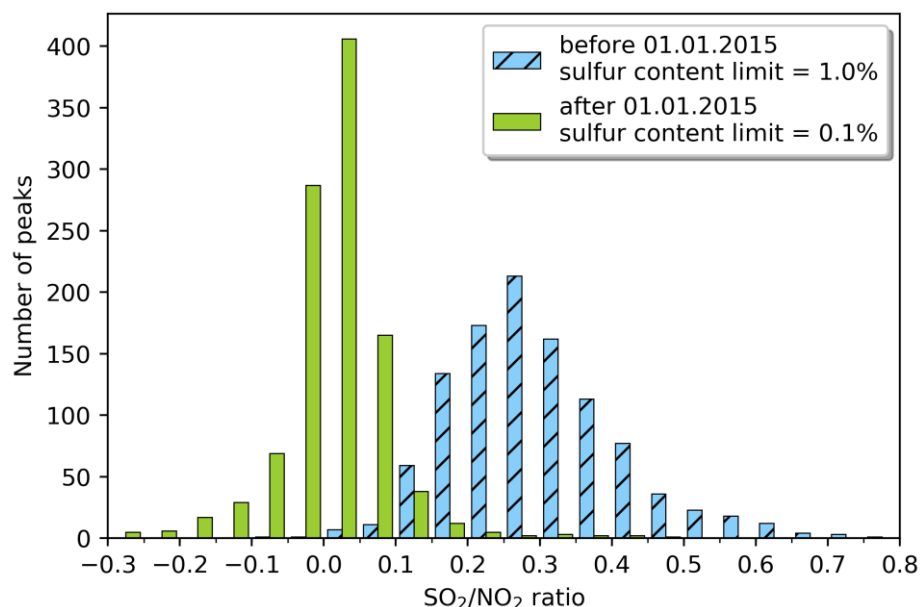
Updated plots:

For an example day (23.07.2014) before the change in regulations:
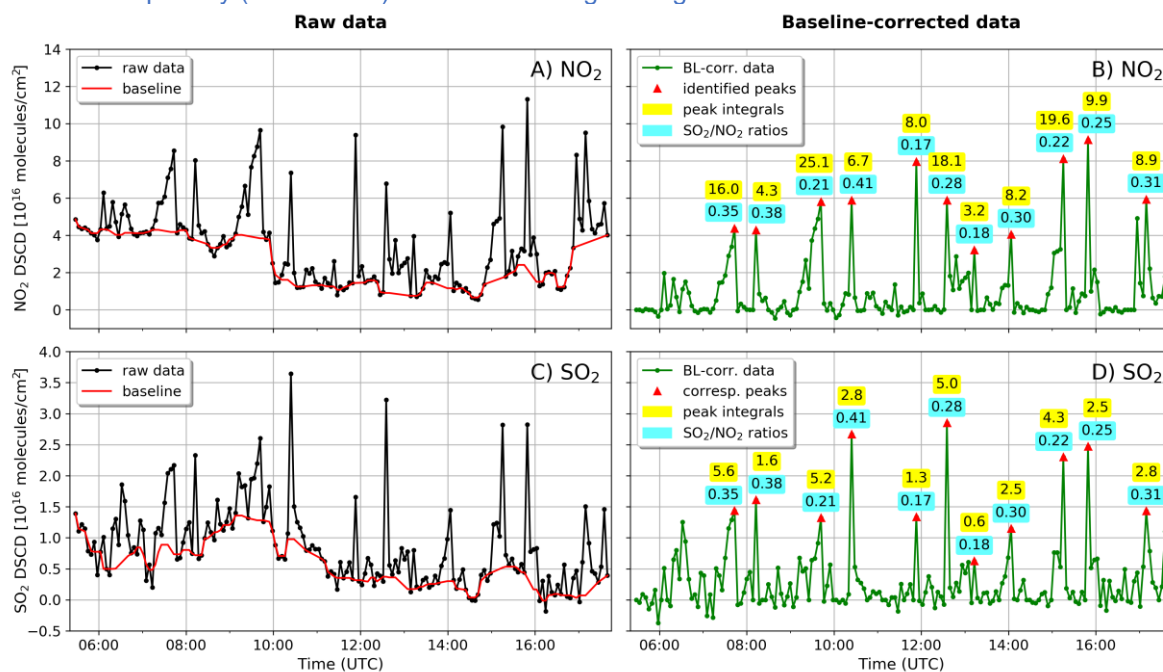
For an example day (03.07.2015) after the change in regulations:
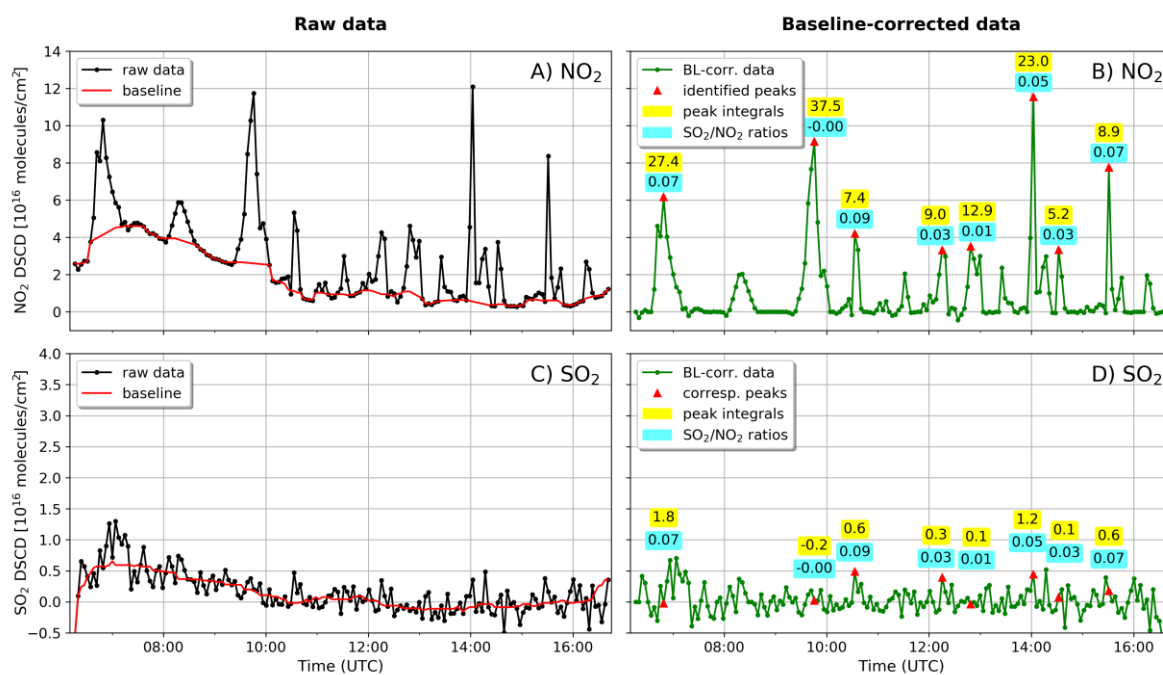
And the updated histogram:

Technical corrections
Equations: Please use single characters for variables (e.g., "S" instead of "SCD", "R" instead of "RESIDUAL", "H" instead of "MLH"). There is a difference between an abbreviation (e.g., "SCD" for slant column density) and the according mathematical symbol (e.g., "S").

This is a matter of personal taste. In the DOAS community, using variable names like "SCD" or "AMF" is quite common. Checking the most recent final revised papers in ACP and AMT containing "DOAS" in the title we found 20 papers using multi-letter variable names like "SCD", "AMF", "AOD", 1 paper using single letter variable names "S" and "M" etc. (and 8 papers without any such equations).

L11: Provide a number for the distance between measurement site and shipping lane

We added the distance:

"The island is located in the German Bight, close to the main shipping lane *(in a distance of 6-7 km)* into the river Elbe towards the harbor of Hamburg."

L13: The fact that the site is close to the shipping lane is repeated. Delete "which is a few kilometres from the shipping lane"

Done.

L17: "retrieved from NO2 retrievals" -> "determined from NO2 retrievals"

Done.

L54: ". . . from around 31 000 . . .. over 52 000 . . . to 89 000 . . ."

Done.

L68: ". . . molecular nitrogen (N2) and oxygen (O2) . . ."

Done.

L81: comma after "radicals"

This paragraph was removed.

L81: ". . . hydroperoxyl (HO2) or organic peroxy radicals (RO2) or halogen oxides (XO, were X = Cl, Br or I)

This paragraph was removed.

L84: "X atoms" -> "halogen atoms"

This paragraph was removed.

L85: "reacts" -> "react"; "reaction" -> "reaction rate"

This paragraph was removed.

L86: "Owing to the lack of photolysis, NO reacts rapidly . . .. during the night"

Done.

L87: "In addition, the nitrate radical (NO3) is formed. . ."

Done.

L110: Comma after "regions"

Done.

L112: "ecosystem" -> "ecosystems"

Done.

L121: Put "3" in "m3" into superscript

Done.

L162: Comma after "emissions"

Done.

L173: Comma after "example"

Done.

L206: Incomplete sentence. Replace, e.g., by ". . . first the measurement site is described, followed by a presentation of the wind statistics and data availability."

Done.

L215: ". . . were taken on Neuwerk, a small island in the North Sea with the size of . . ."

Done.

L221: "island of Neuwerk" or simply "Neuwerk" (here and anywhere else). Delete "where our measurement site is located" (repetition)

Done.

L225: Do you refer to a specific document from the "Statistische Ämter" or can you

provide an url to the data?

We are referring to a specific document/URL from the "Statistische Ämter…" from the year 2015. The URL to the document can be found in the bibliography under "Statistische Ämter… (2015)". The correct way how to cite this information is specified on the web page from the Statistische Ämter.

L224: Is this height above sea level?

No, this is height above ground. But the difference to height above sea level, as you can see from the photo, might be 1-2 meters and therefore negligible.

L248: "site for the measurements" -> "site"

Done.

L285: To "inject" light into the fibre sounds strange since this term suggests that the light is somehow transported actively. Replace by something like "focused on the entrance of the optical fiber"

Done.

L286: "opening angle" -> "field of view"

Done.

L307: Define what "SCD1" and "SCD2" refer to. Replace by variable names consisting of single letters.

We rephrased the sentence to make this more clear.

Regarding the single letter variable names: This is a matter of personal taste. In the DOAS community, using variable names like "SCD" or "AMF" is quite common. Checking the most recent final revised papers in ACP and AMT containing "DOAS" in the title we found 20 papers using multi-letter variable names like "SCD", "AMF", "AOD", 1 paper using single letter variable names "S" and "M" etc. (and 8 papers without any equations).

L316: It should be mentioned that a spatially limited plume directly over the instrument leads to an underestimation of the retrieved dSCDs.

Done.

Table 1: Only list the polynomial degree, not the number of coefficients.

I listed both since the definiton of the polynomial degree can be ambiguos, according to whether you count the lowest order linear term as index 0 or 1.

L399: "filtered" -> "discarded"

Done.

L439: Delete "the pollutant"

Done.

L441: ". . . difference between NO2 in the UV (red curve) and in the visible (blue curve). . ." (the discussion is about NO2 and not about the colors of the curves)

Done.

L442: "more intense" -> "stronger"

Done.

L445: "By comparing SO2 (black curve) with NO2 (red and blue curves), it can be seen. . ."

Done.

L447: Delete "A more dirty"

Done.

L454: "points in time" -> "times"

Done.
L497: "The difference between UV and visible peak values depends. . ."

Done.

L498: "A short distance of the plume to the instrument and its complete coverage by the shorter UV path leads to higher values in the UV. . ."

Done.

The title of section 4.3 does not make sense. It implies that the approach is statistically evaluated. Instead, the data is statistically evaluated. Replace with something like "Statistical evaluation of the NO2 and SO2 data"

This section is not about NO_2 and SO_2 but about NO_2 in UV and visible for DSCDs and VMRs. We changed the title to "Statistical evaluation of UV and visible NO_2 data"

L507: ". . .all single pairs of simultaneous measurements" -> "all single pairs of DSCD measurements. . ."

Done.

L508: "the left subplot in the upper row" -> "Panel A"

Done.

L509: "both measurements" -> "NO2 and SO2 DSCDs"

This section is not about NO_2 and SO_2 but about NO_2 in UV and visible. We changed „both measurements" to "NO_2 DSCDs in UV and visible".

L513: "The right subplot in the upper row" -> "Panel B"

Done.

L518: "the left subplot in the bottom row" -> "Panel C"

Done.

L523: "(right plot)" -> "(Panel D of Fig. 9)"

Done.

L533: "applied on mountains" -> "applied to mountain-based measurements"

Done.

L535: Delete "However"

Changed "However" to "In contrast to our site".

L537: "This should lead" -> "This leads" (the enhancement in path length in a cleaner and less dense atmosphere is obvious)

Done.

L541: "various" -> "detailed" or "comprehensive"

Done.

L543: Delete "emitting"

Done.

Figure 10: Mark the three panels as "A", "B" and "C" (from top to bottom)

Done.

L544: "Measurements from Wednesday, 9 July 2014 are shown in Figure 10. Panel A shows. . ."

Done.

L545: "The middle one" -> "Panel B"

Done.

L549: "The lower sub-plot" -> "Panel C"

Done.

L563: "The differences of both measurement techniques need to be considered for such a comparison:"

Done.

L565: "at one point" -> "at a single location"

Done.

L568: Insert "the" before "line-of-sight"

Done.

L569: delete "line-of-sight" (it is already mentioned at the beginning of the sentence)

Done.

L579: Delete "From the Figure, it can be easily identified that"

Done.

L581: Delete "nicely"

Done.

L586: Delete "It is also clearly visible, that"

Done.

L591: "it's" -> "its"

Done.

L592: Delete the first sentence of this paragraph

Done.

L594: "the upper subplots" -> "Panel A"; Add ", respectively" to the end of the sentence.

Done.

L606: "makes no sense" -> "is of little use"

Done.

L603: "the lower subplot" -> "Panel B"

Done.

L613: Delete "As can be seen in the figures"; delete "usually"

Done. Done.

L614: What do you mean with "progression of both curves"?

We mean "curve shape" or "course of the curves". We changed the formulation to the latter.

L623: Insert comma after "combustion"

Done.

L643: "The mean NO2 volume mixing ratios for each weekday shown in Fig. 16 illustrate the influence of land-based road traffic."

Done.

L647: "There is only little weekly cycle for air masses coming from the open North Sea. Measurements . . ."

Done.

L665: "single day measurements" -> "Single day of measurements"

Done.

L704: Delete comma after "This implies"

Done.

L738: "like expected" -> "as expected"

Done.

L743: "It can be seen that this increase for the land source sector is only a relative increase by comparing. . ."

Done.

L765: "roll" -> "role"

Done.

L767: "A monitoring of emissions from single ships requires the analysis of individual plume peaks in the NO2 and SO2 data sets."

Done.

L780: I am not familiar with the term "emission factor". Do you mean "emission rate"?

Both terms mean more or less the same, in the sense of an emission intensity. However, in the community of shipping emission measurements, the term "emission factor" is more commonly used.

780: delete "both"

Done.

L796: "one can get rid of the background pollution" -> "the background pollution can be removed"

Done.

L801: "To achieve a better signal-to-noise ratio, the integrals . . .. in the last step"

Done.

L803: "one" -> "an"

Done.

L804: "In both the NO2 and SO2 signal" -> "Both the NO2 and SO2 signal show"

Done.

L805: delete "are visible"; delete "measured"; "The shape of the peaks is also often quite similar" -> "Most of the peaks are of similar shape"

Done. Done. Done.

L807: "The SO2 to NO2 ratio can vary strongly for different ships. For example, the plume of the ship passing the line of sight around 12:00 UTC has a high NO2 content, but is low in SO2, whereas the opposite is true for the ship passing at 12:30 UTC, indicating that the second ship. . ."

Done.

L811: Delete "In contrast to this,"

Done.

L813: "High NO2 peaks also occur on this day. However,. . ."

Done.

L818: "From this plot one can also see that" -> "As can be seen from this plot, "

Done.

L819: "overestimate" -> "overestimates"

Done.

L826: "retrieved" -> "analyzed"

Done.

L836: Insert comma before "indicating"

Done.

L839: "and for 2015 and 2016, one gets a mean value of . . . " -> ", and a mean value of . . . for 2015 and 2016"

Done.

L842: "leading to overestimation" -> "leads to an overestimation"

Done.

L850: "from" -> "by"

Done.

L851: "SO2 and NO2 emission ratios can also be derived from. . ."

Done.

L858: "the dependency of SO2 to NO2 ratio to fuels sulfur content"

Done.

L863: "Island Neuwerk" -> "Island of Neuwerk"

Done.

L865: "into" - > "and"

Done.

L871: Delete "also"

Done.

L882: "NO2" -> "daily averaged NO2"

This sentence is not about daily averages but about the weekly cycle (averages according to weekday) and diurnal cycle (averaged values according to the hour of the day) of NO_2.

L908: Insert "can" after "ratios"

Done.

Thanks again for your thorough proof-reading. This helped us a lot.