

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

### **Anonymous Referee #1**

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### **General comments**

The manuscript entitled “Monitoring shipping emissions in the German Bight using MAX-DOAS measurements” presents remote sensing observations of NO<sub>2</sub> and SO<sub>2</sub> 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

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impact of the reduction of fuel sulphur content on atmospheric SO<sub>2</sub> 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 O<sub>4</sub> and the target gases and thus different AMFs, as done during previous studies (e.g., Sinreich et al., AMT, 2013).

### 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).

#### 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 HO<sub>2</sub>. In polluted air, reaction with NO<sub>x</sub> is likely to lead to a removal of halogen radicals by formation of halogen nitrates.

#### 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.

#### Section 3.2:

Please provide technical information on the fibre bundle (number of fibres, diameter, arrangement at both sides).

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What are the wavelength ranges of both spectrographs?

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  $0.6^\circ$ . Please specify the elevation angle as the centre of the 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.

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

### 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.

The uncertainties of the O<sub>4</sub> scaling approach need to be discussed. For example, O<sub>4</sub> usually has a profile shape very different from NO<sub>2</sub> and SO<sub>2</sub>, which violates the basic assumption that the O<sub>4</sub> dSCD is a good proxy for the light path through the NO<sub>2</sub> and SO<sub>2</sub> 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

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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.

Equation 5: It should be mentioned that  $n_{O_4}$  is the  $O_4$  concentration at the surface.

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  $0.5^\circ$  in elevation angle is certainly not negligible at very low elevation angles.

#### Section 4.1:

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  $NO_2$  over the shipping lane than over the instrument, be a much more likely explanation for these findings?

#### 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  $NO_2$  and  $SO_2$ "

#### 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  $O_4$  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.

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#### Section 4.6:

L655ff: NO<sub>2</sub> 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 NO<sub>2</sub> in background air observed in the Arctic is similar to the present study might be mere coincidence.

#### Section 4.7:

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

#### 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 land-based sources is present. Thus, air masses classified as “Land” are likely to be partly affected by ship emissions.

#### Section 4.9:

Given that SO<sub>2</sub> scatters around a smooth (near-zero) background level, it is surprising to see that no negative SO<sub>2</sub> to NO<sub>2</sub> 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.

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## 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*”).

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

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

L17: “retrieved from NO<sub>2</sub> retrievals” -> “determined from NO<sub>2</sub> retrievals”

L54: “. . . from around 31 000 . . . over 52 000 . . . to 89 000 . . .”

L68: “. . . molecular nitrogen (N<sub>2</sub>) and oxygen (O<sub>2</sub>) . . .”

L81: comma after “radicals”

L81: “. . . hydroperoxyl (HO<sub>2</sub>) or organic peroxy radicals (RO<sub>2</sub>) or halogen oxides (XO, were X = Cl, Br or I)

L84: “X atoms” -> “halogen atoms”

L85: “reacts” -> “react”; “reaction” -> “reaction rate”

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

L87: “In addition, the nitrate radical (NO<sub>3</sub>) is formed. . .”

L110: Comma after “regions”

L112: “ecosystem” -> “ecosystems”

L121: Put “3” in “m<sup>3</sup>” into superscript

L162: Comma after “emissions”

L173: Comma after “example”

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

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

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

L225: Do you refer to a specific document from the “Statistische Ämter” or can you provide an url to the data?

L224: Is this height above sea level?

L248: “site for the measurements” -> “site”

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”

L286: “opening angle” -> “field of view”

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

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

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

L399: “filtered” -> “discarded”

L439: Delete “the pollutant”

L441: “. . . difference between NO<sub>2</sub> in the UV (red curve) and in the visible (blue curve). . .” (the discussion is about NO<sub>2</sub> and not about the colors of the curves)

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L442: “more intense” -> “stronger”

L445: “By comparing SO<sub>2</sub> (black curve) with NO<sub>2</sub> (red and blue curves), it can be seen. . .”

L447: Delete “A more dirty”

L454: “points in time” -> “times”

L497: “The difference between UV and visible peak values depends. . .”

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. . .”

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 NO<sub>2</sub> and SO<sub>2</sub> data”

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

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

L509: “both measurements” -> “NO<sub>2</sub> and SO<sub>2</sub> DSCDs”

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

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

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

L533: “applied on mountains” -> “applied to mountain-based measurements”

L535: Delete “However”

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

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L541: “various” -> “detailed” or “comprehensive”

L543: Delete “emitting”

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

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

L545: “The middle one” -> “Panel B”

L549: “The lower sub-plot” -> “Panel C”

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

L565: “at one point” -> “at a single location”

L568: Insert “the” before “line-of-sight”

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

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

L581: Delete “nicely”

L586: Delete “It is also clearly visible, that”

L591: “it’s” -> “its”

L592: Delete the first sentence of this paragraph

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

L606: “makes no sense” -> “is of little use”

L603: “the lower subplot” -> “Panel B”

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

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

L623: Insert comma after “combustion”

L643: “The mean NO<sub>2</sub> volume mixing ratios for each weekday shown in Fig. 16 illustrate the influence of land-based road traffic.”

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

L665: “single day measurements” -> “Single day of measurements”

L704: Delete comma after “This implies”

L738: “like expected” -> “as expected”

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

L765: “roll” -> “role”

L767: “A monitoring of emissions from single ships requires the analysis of individual plume peaks in the NO<sub>2</sub> and SO<sub>2</sub> data sets.”

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

L780: delete “both”

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

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

L803: “one” -> “an”

L804: “In both the NO<sub>2</sub> and SO<sub>2</sub> signal” -> “Both the NO<sub>2</sub> and SO<sub>2</sub> signal show”

L805: delete “are visible”; delete “measured”; “The shape of the peaks is also often

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quite similar” -> “Most of the peaks are of similar shape”

L807: “The SO<sub>2</sub> to NO<sub>2</sub> 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 NO<sub>2</sub> content, but is low in SO<sub>2</sub>, whereas the opposite is true for the ship passing at 12:30 UTC, indicating that the second ship. . .”

L811: Delete “In contrast to this,”

L813: “High NO<sub>2</sub> peaks also occur on this day. However, . . .”

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

L819: “overestimate” -> “overestimates”

L826: “retrieved” -> “analyzed”

L836: Insert comma before “indicating”

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

L842: “leading to overestimation” -> “leads to an overestimation”

L850: “from” -> “by”

L851: “SO<sub>2</sub> and NO<sub>2</sub> emission ratios can also be derived from. . .”

L858: “the dependency of SO<sub>2</sub> to NO<sub>2</sub> ratio to fuels sulfur content”

L863: “Island Neuwerk” -> “Island of Neuwerk”

L865: “into” -> “and”

L871: Delete “also”

L882: “NO<sub>2</sub>” -> “daily averaged NO<sub>2</sub>”

L908: Insert “can” after “ratios”

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Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-1153, 2017.

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