

## ***Interactive comment on “NO<sub>2</sub> observations over the western Pacific and Indian Ocean by MAX-DOAS on *Kaiyo*, a Japanese research vessel” by H. Takashima et al.***

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We would like to thank the reviewer for providing constructive comments on the manuscript. A detailed description of our revisions is provided below.

General:

\*Results are obtained by making use of a MAX-DOAS type of instrument. Please discuss in more detail the reason to use for this study a MAX-DOAS instrument, whereas other methods are available. For example, the in-situ method used in reference 1 (see below) appears to have a much lower (better) detection limit. Reference 1 reports, al-  
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though for a different geographical region, NO<sub>2</sub> volume mixing ratios of approximately 10 pptv for long periods in a row (see e.g. Fig. 8 in this paper). These values are almost a factor of 10 lower than the mode of the PDF reported in this work.

>We agree with the reviewer that there are better instruments for NO<sub>2</sub> measurement. Our primary aim is to establish an automated measurement system (without any calibration system) for use on a vessel by employing a low-power/low-cost spectrometer with an active-type gimbal. As described in the text (p. 6071, lines 122–123), in this study we focus only on NO<sub>2</sub> because it is somewhat easier to retrieve than other components, although MAX-DOAS has the potential to perform simultaneous measurements on several components. After evaluating NO<sub>2</sub> retrieved by this system on a vessel, we develop a retrieval method for other components. Another point is that MAX-DOAS is useful for validating chemical transport models, because it can provide a mean/typical value for a layer (e.g., the mean value for the 0–1 km layer) and it is easy to compare the content with the model results, whereas in situ direct measurement can only obtain data at a point and it may be difficult to compare such measurement data with model results.

\*It is mentioned that the detection limit of the method used in this study is approximately 0.1 ppbv (p.6078, l.13-15). This would imply that the MAX-DOAS method cannot be used to determine a reliable PDF of the NO<sub>2</sub> volume mixing ratio below this value. From this perspective, Fig. 11 may cause some confusion, since it suggests that values between 0.00 and 0.05 ppbv are very unlikely to be found and that the PDF has a mode around 0.1 ppbv (p. 6078, l.21). This value for the mode seems to quite high when compared to the values reported in reference 1 (despite the fact that a different region is studied). The mode around 0.1 ppbv may therefore not reflect the mode of the real PDF over the remote ocean, but rather be due to the relatively high detection limit of the measurement technique. Perhaps Fig. 11 could be modified, such that a single bin is used below 0.1 (or 0.2) ppbv.

>Figure 8 shows good agreement (no systematic differences) between the two analy-

ses (460–490 nm and 338–370 nm). At low NO<sub>2</sub> concentrations, both analyses show a maxima/mode around ~0.1 ppbv, with variability of ~0.05–0.1 ppbv. This blurry structure in the case of low concentrations over the remote ocean is also clear in Fig. 11. Consequently, we may say that the background level over the remote ocean for the 0–1 km layer is ~0.1±0.1 ppbv. The variability of ~0.05–0.1 ppbv may correspond to the random error of NO<sub>2</sub> concentration over the remote ocean, although the calculated value in Table 2 is much smaller than this (i.e., smaller than ~0.05–0.1 ppbv). Because our original description of “detection limit” may have led to misunderstanding, we revised the relevant text.

\*Compared to in-situ observations, a major strength of MAX-DOAS is the ability to measure tropospheric columns. In relation to transport of NO<sub>2</sub> (p.6071, l.3), this quantity is just as relevant as the volume mixing ratio. Please provide a figure with tropospheric NO<sub>2</sub> columns (preferably similar to Fig. 11 and for the same subset of all observations), or otherwise mention why it is decided not to include this MAX-DOAS product despite its relevance in this context.

>We agree with the reviewer. We added the tropospheric column values in Table 2.

\*A negative correlation is reported between NO<sub>2</sub> (425-450 nm) and H<sub>2</sub>O (p.6076, l.9-10). This finding is used to support the claim that H<sub>2</sub>O has a negative impact on the DOAS fitting in this particular fitting window. Alternatively, one could hypothesize that the negative correlation is due to a chemical reaction (or a mechanism of reactions) involving both NO<sub>2</sub> and H<sub>2</sub>O. Please provide correlations between H<sub>2</sub>O from this fitting window (425-450) and NO<sub>2</sub> from the two other fitting windows (338-370nm and 460-490nm) to exclude this alternative hypothesis, or otherwise please comment on this point.

>The problem is that the NO<sub>2</sub> DSCD value for 425–450 nm was commonly negative and lower at lower elevation angles (Fig. 7), whereas it was positive for 460–490 nm (figure not shown). The negative values at lower ELs are unlikely to occur in the real

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atmosphere. The correlation between NO<sub>2</sub> for 460–490 nm (or NO<sub>2</sub> 338–370 nm) and H<sub>2</sub>O was positive (figure not shown), as expected from the fact of a longer light path at lower EL. Therefore, the negative correlation in Fig. 7 is not due to a chemical reaction. Note that good agreements were observed between H<sub>2</sub>O for 425–450 nm and H<sub>2</sub>O for 460–490 nm, and between NO<sub>2</sub> for 460–490 nm and NO<sub>2</sub> for 338–370 nm (Fig. 8). Note also that similarly negative and low DSCD values were found for the 425–490 nm fitting window (see the reply to the relevant comment by referee #2). These findings are explained in the revised manuscript.

Specific comments:

\*p.6074, l.22-25 (Here, DSCD ... collision complex.): Please rephrase.

> Change made as suggested.

\*p.6076, l.4: Please replace 'cause' by 'case'.

> Change made as suggested.

\*p.6077, l.8-10: This sentence is a bit confusing, especially the part: 'the temperature dependence of trace gas in tropospheric'. Please rephrase.

> Change made as suggested.

\*Fig. 9: The dashed line is not mentioned in the caption or in the text. Please explain it. Is it a linear fit? Which fitting method is used? It does not seem to describe well the higher values.

> We added the relevant information in the caption to Fig. 9.

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