

We like to thank the three reviewers for their helpful comments. We are grateful for the opportunity to improve our manuscript. Below we describe point-by-point the changes we have made based on these comments. The reviewers' comments are in black, our responses are in red, and the changes to the manuscript in blue. We think that we have addressed all comments by the reviewers and are confident that the revised manuscript can be accepted for publication in ACP.

Reply to reviewer 1

1. The particle measurement method is not described. Although the nucleation events and particle concentrations as well as the correlation with IO columns are mentioned in the abstract, which assigns some importance to the particle measurements, only very little information is given in the text of the article, in three lines within the site description paragraph. The method should be properly explained in an individual paragraph in section 2 (and possibly additional paragraphs also in section 3 and/or 4 on data analysis and discussion of the results).

The declared correlation between particle concentration and IO mixing ratios is an important result of the manuscript. This result is emphasized in abstract and conclusions and should therefore be supported by a clear figure. When comparing Fig.2 with Figs. 5 and 6, this correlation can be estimated, the data set is thus present and it should be possible to demonstrate the mentioned correlation directly. An additional, purposeful plot would be helpful, at least showing particle concentration and IO mixing ratios together.

We included an additional section (Section 2.3) on particle measurements:

Particle measurements were carried out by a nano scanning mobility particle sizer (nano-SMPS), covering the size range from 3 nm to 20 nm, and a standard SMPS, covering the 10 nm to 100 nm range. Both, the nano-SMPS and SMPS were standard Thermo Systems Inc. (TSI) systems [Wang and Flagan, 1990], with the nano-SMPS using the TSI 3025a condensation particle counter (CPC) as a detector and the SMPS using a TSI 3010 CPC as a detector. The instruments were located about 150 m away from the low tide region and sampling was conducted through a 3 m long, stainless steel inlet tube. Size resolved concentrations were corrected for diffusional losses, which were calculated on the basis of tube diameter and residence time in the sample tube using the equations given by Seinfeld and Pandis [2006].

Figure 2 was replaced by a figure, where both, particle concentration and IO column densities of the long light path are shown. Additionally we added a reference to Huang et al. (2010) where a plot of IO column densities vs. particle concentration is already published. We now write on p. 21380, l. 24 ff.

An example for the correlation between enhanced IO column densities measured along the long light path and particle formation is illustrated in Figure 2. The particle burst (upper panel) at noon coincides with elevated IO column densities (lower panel). A plot with total particle concentration vs. IO column densities can be found in Huang et al. (2010).

2. Section 2.1 and 2.2: Please give some information on the integration times for both techniques. In the LP-DOAS case, please state the time difference between the alternating paths.

In the revised version we give more information on the integration time for both techniques. In Section 2.1 we included the following text:

A full LP-DOAS measurement sequence took about 12 minutes for good visibility conditions. A measurement sequence started with a lamp reference spectrum with 15 scans, followed by the measurement of the atmospheric spectrum with maximum 30 scans within 30 seconds and a second lamp reference spectrum of 15 scans. Afterwards background spectra were taken with a fixed integration time of 10 seconds. In the process of the analysis, the two lamp reference spectra were added to one shortcut spectrum for each atmospheric spectrum, in order to provide the ideal sensitivity. Since the detection limit of the longer light path was expected to be lower for homogeneous distributed trace gases, we performed measurements on the short light path during every fourth measurement cycle.

And in Section 2.2 we changed the text to:

Spectra were taken under 2°, 4°, 6°, 10°, 20°, and 90° elevation angle for a fixed integration time of 300 seconds for each angle. Thus a full sequence took 30 minutes.

3. In section 4.1 the polynomial order and an “offset” within the MAX-DOAS retrieval are discussed as influencing the retrieval of OIO and I₂. The meaning of the “offset” in the retrieval is not explained or mentioned. Please add some information in section 3.1 on this (stray light?).

We specify the term offset and now state in Section 3.2 of the revised manuscript:

To detect the absorbers of interest, a 90° reference spectrum of the same sequence, a Ring spectrum, a polynomial to account for Rayleigh and Aerosol scattering, an intensity offset (polynomial of degree 2) to account for possible instrument stray light, and cross sections of all other absorbers were fitted to the atmospheric spectrum.

4. Section 4.1 discusses the increase of IO dSCDs for decreasing viewing angle and a steep vertical gradient of IO is identified in the interpretation of this finding. In this argumentation, the authors should be more specific and quantitative. Generally, a decrease of dSCD with increasing elevation angle is also expected for a constant profile (box profile) up to a certain altitude. Here, the analysis and the finding of a “strong vertical gradient” (p. 21379, 1.5) depends on the strength of the increase, so that some quantitative information becomes necessary. How strong does the IO dSCD increase for increasing viewing angle? E.g. also: which increase in IO dSCD would still agree with a box profile of the IO mixing ratio? I believe that it is difficult to retrieve specific profile information of IO from these measurements, but even more the authors should be somewhat more precise and cautious than in their interpretation. The statement in p.21379, 1.12 is unfortunately not true, otherwise the authors could quantify the vertical gradient. I am convinced, that the vertical gradient of IO mixing ratios influences their measurements and may lead to the observed decrease in dSCDs for increasing elevation angle - however, the vertical gradient is not measured directly, which is unfortunately quite a difficult task. Maybe the sentence can be reformulated.

We agree that we cannot directly prove a strong vertical gradient, since we are not able to distinguish between the differences between the different elevation angles due to a vertical gradient of the trace gas or a longer light path through the atmosphere. However, we now

include a plot (Figure 10) of the IO/O₄ ratio. Since the O₄ profile is essentially invariable (except for small changes due to changing air density) and changes in the dSCDs of O₄ are thus caused by changes of the light path due to scattering, one can use the O₄ dSCDs as an approximation for the length of the light path. Figure 10 shows that the ratio for 2° elevation angle is significantly higher, indicating a strong vertical gradient of IO. We changed Section 4.1 to read (page 21379, line 5 ff.):

...the dSCDs decrease rapidly for higher elevation angles. The decrease of dSCDs for higher elevation angles is generally expected for tropospheric absorbers, because the light path through the trace gas layer is longer for lower elevation angles. However, the separation of the different elevation angles can also be caused by a vertical gradient of the trace gas itself. Since the source of IO is located on the ground, one would expect a vertical gradient of the IO concentration. To distinguish between the two effects radiative transport modelling would be necessary. The quite heterogeneous distribution of IO and rather high noise would make the radiative transfer modelling and its interpretation a quite difficult task that is beyond the scope of this work. However, we used an approximation via O₄ to obtain information about the vertical distribution of IO: The profile of O₄ is essentially invariable (except for small changes due to changing air density) and therefore changes in the dSCDs of O₄ are caused by changes of the light path due to scattering in the atmosphere. For a constant profile of IO in the lowest few hundred meters, one would expect the IO/O₄ ratio to be rather constant for different elevation angles, whereas for a vertical gradient with higher IO concentrations close to the ground, the ratio should be higher for lower elevation angles. Figure 10 shows the IO/O₄ ratio for the five days of measurement. For 2° elevation angles, the ratio is significantly higher, indicating a strong vertical gradient

5. p. 21381, l.13-15: What is the difference between the two fibres used? In writing “well mixed” - are the authors referring to polarization issues? Do the fibres have different lengths? Why does this cause a negative bias for the IO results? Please just give some more information on this.

Spectroscopy requires homogeneous illumination of the grating of the spectrometer used. This is achieved by using a quartz glass fibre which ‘mixes’ the receiving light (image of the single retro reflectors) to obtain a homogeneous illumination at the spectrometer (so-called ‘mode mixing’). Different fibres (different doping materials, diameters and processing) can exhibit different mixing properties. The mixing can be improved by exerting mechanical stress on the fibres (in a unit called ‘mode mixer’), but is often limited depending on the fibre characteristics. A good mixing of the light is especially important for very short light paths, since then the image of the retro reflectors is sharper, or if the applied light source features strong spectral structures in the respective wavelength range (as it is the case for IO). If the mixing is not perfect (which is the typical case) small spectral structures may arise, which can correlate/ anti-correlate with absorption structures. This can cause bias concentrations (positive or negative). We included some more information on this and on p. 21381, l.15ff. now write:

Spectroscopy requires a homogeneous illumination of the spectrometer grating. To obtain this, quartz glass fibres are used in order to ‘mix’ the light and thus obtain a homogeneous illumination of the spectrometer. Different fibres can exhibit different mixing properties. The mixing can be improved by exerting mechanical stress on the fibres applying a mode mixer (Stutz and Platt, 1997), but mixing is often limited depending on the fibre characteristics. A good mixing of the light is especially important for very short light paths, as then the image of

the retro reflectors is sharper, or if the applied light source features strong spectral structures in the respective wavelength range (which is the case in the evaluation wavelength range of IO). If the mixing is not perfect, small spectral structures may arise, which can correlate/ anti-correlate with absorption structures. This can cause a bias in the observed concentrations and therefore the column densities observed after 31 August, 14:00 might be underestimated.

6. The structuring of section 4 is not ideal, as 4.2 contains the results from the LPDOAS and additionally also the comparison with the MAX-DOAS and the interpretation following from this (starting p.21381, l.17). I suggest an additional paragraph 4.3 for the comparison and interpretation.

We separated Section 4.2 by adding a further paragraph 4.3 for the discussion and comparison of the results. Section 4.3 starts on page 21380, line 5 with the discussion of the LP-DOAS data followed by the comparison with the MAX-DOAS results. We also included some more information on the IO results and now write on p. 21380, 14 ff:

IO was detected on both light paths on each day. The average detection limit was $2.2 \cdot 10^{13}$ molec/cm² or 2.2 ppt on the long light path and $3.6 \cdot 10^{13}$ molec/cm² or 14 ppt on the short light path. The maximum observed column densities were $8.0 \cdot 10^{13}$ molec/cm² and $7.4 \cdot 10^{13}$ molec/cm², respectively.

7. Section 4.2 (maybe becoming 4.3?) might need some revision. Little information is given and one part of this section is by content repeated twice: parts (p. 21381, l.22-29) and (p.21382, l.10-15) contain very similar information and nearly the same wording.

We moved the content of (p.21382, lines10-15) to (p. 21381, lines 22-29). On p.21382, l. 10ff. we now write:

Figure 9 shows the correlation between the 2° MAX-DOAS dSCDs and the LP-DOAS column density along the long light path. The data show strong scatter, but although the intertidal area in front of the instrument is crossed just once, the MAX-DOAS shows higher column densities most of the time. This occurs mainly during low tide (see Fig. 8). There are probably two source regions, which explain the higher MAX-DOAS signal: First, the MAX-DOAS instrument probably also probes the intertidal area on the other side of Finish Island (Figure 3 shows that in the deeper water behind Finish Island *Laminaria Hyperborea* is located, which is a very strong emitter of iodine precursors (Ball, 2009)), and second, it could be possible, that light reflected from the surface and therefore passing a layer with very high IO concentrations, causes the higher signal.

In the comparison between LP-DOAS and MAX-DOAS (starting p. 21381, l.17) it needs to be made clear right at the beginning, that agreement between the column densities is not expected, as the light paths between the two methods are considerably different. The statement on p. 21381, l.19-21 is not entirely correct, as far as I understand. The MAX-DOAS dSCDs are not generally higher than the LP-DOAS column values. This statement is mainly true for the MAX-DOAS 2_ direction. However, the 4_ elevation values are rather similar or lower than the LP-DOAS results. In this part, some more detailed and careful comparison of the measurement results will help.

We changed p. 21381, l 17ff. to read:

Figure 8 shows a comparison of the 2° and 4° MAX-DOAS IO dSCDs (blue stars and black circles, respectively) with the column densities of the LP-DOAS instrument (long light path: black crosses, short light path: pink triangles). An absolute agreement between the active and the passive data is not expected, since the light paths are significantly different. However, the data correlate well and it can be seen that the 2° MAX-DOAS dSCDs are largely higher than the LP-DOAS column densities, while the 4° results are of about the same magnitude or lower than the LP-DOAS results. A striking feature is that the 2° MAX-DOAS results are often a factor of 2 higher than the 4° results. This is not only because of a longer light path, but also due to a vertical gradient of IO (see Figure 10).

In the discussion of higher IO columns in the MAX-DOAS results, the statement that “light reflecting from the surface and passing very high concentrations, causes the higher signal” might need more reflection, ideally some quantitative information. I agree that this light path contributes to the measured signal. However, is a major contribution to the signal expected? Radiative transfer calculations should be able to estimate the contribution from this pathway. Can the authors provide block air mass factors for their measurements?

Generally the albedo of water can be estimated at 4-6%, which is not high, but depending on the concentration above the seaweed, might have an impact. We could provide a box air mass factor for the measurements. However, we think that this would be misleading: a box air mass factor could not account for the heterogeneity of IO, which is shown by the LP-DOAS data. Generally, radiative transfer modelling is hard to implement, as the measurements are somewhat under-determined. To obtain sufficient information about the 3-dimensional distribution of IO that would be needed to perform useful radiative transfer modelling the dataset of one instrument is not sufficient. However, we included some information about the albedo and now write on p.21381, l.22ff:

First, the MAX-DOAS instrument probably also probes the intertidal area on the other side of Finish Island (Figure 3 shows that in the deeper water behind Finish Island Laminaria Hyperborea is located, which is a very strong emitter of iodine precursors (Ball, 2009)), and second, it is possible, that light reflected from the surface and therefore passing a layer with very high IO concentrations, causes the higher signal. While water only has an albedo below 10% very high concentrations of IO close to the surface could have a significant impact on the results. Additionally light not reflected by the surface, but scattered by, e.g. aerosols close to the ground contributed to the measured IO signal.

8. In the Conclusions, the sequence of argumentation seems somewhat confused (starting p. 21382, l. 22). Shouldn't the line of logic should run in the following way? IO column results from the LP-DOAS long light path agree well with those from the short distance. Hence, one understands that the IO distribution is inhomogeneous along the long light path, meaning that a confinement of IO to the intertidal area can be inferred. In addition one can suspect, that on the short light path, the IO also might be inhomogeneously distributed (which cannot be seen directly from the measurements, but might well be the case). If the IO is not homogeneously distributed over the intertidal area, then the previously found model results on IO concentrations and nucleation events agree with the findings in the present study.

We restructured the conclusions according to the reviewer's suggestions and now write:

IO could clearly be detected, with the observation angle dependence of the observed dSCDs indicating a vertical gradient in the IO concentration. Comparing the IO column densities of two different LP-DOAS light paths, where one light path was just crossing intertidal area, suggests that IO is almost exclusively located in this area. Assuming that its distribution in the intertidal area is also inhomogenous, we feel that we can confirm the so-called "hot-spot-theory"...

B. Additional Comments

- p. 21373, l.25: As not all RHS are listed in Peters et al (2005), maybe rewrite "... observations of RHS can be found in Peters et al. (2005)." to read "... observations of important RHS (IO, OIO, I2 and BrO) can be found in Peters et al. (2005)."

We changed the text as suggested.

- p. 21373, l.26: It is not clear that the expression "All above mentioned measurements..." actually only refers to the few lines above and not to the first half of this page 21373, where studies are cited which actually use other than LP-DOAS techniques. Please be more specific here, saying (e.g.) that the measurements which were conducted at Mace Head were using the LP-DOAS method.

We agree and reformulated the sentence on p.21373, l. 26ff. We now write:

Most RHS measurements were made using active LP-DOAS.

- p. 21375, l.3: Here, "f" is used for the f-number (f/) and below (l.21) for the focal length. Please write in l.3 "(f/6.9,...)" or "(f-number = 6.9,...)" and in addition give the focal length (f = 500 mm, I guess?) of the Action 500pro.

We changed the text as suggested.

- p. 21376, l.8: As Fig.1 does not show the seaweed density, the reference to Fig.1 should be relocated to earlier in the sentence, e.g., to behind "Mweenish Island".

We reformulate the sentence and now write:

The MRI is located in front of Mweenish Island (see Fig.1), an island with a very high seaweed density.

- pp. 21377-21378: In the description of the electronic-vibrational absorption bands of IO, OIO and I2, for IO and I2 the term electronic transition (with spectroscopic notations given) is used, while for OIO it says vibrational bands (without giving the spectroscopic notation). This is correct, but misleading. As the absorption bands are in any of the three cases electronic transitions into different vibrational levels, please use similar descriptions for each, e.g. write "vibrational bands" on p. 21377, l. 17 and p. 21378, l.1 for IO and I2, respectively, as for OIO.

We changed the text as suggested.

- p. 21380, l.12-13: Do you mean "with only minor differences" or "although also some differences are found"?

We reformulated the sentence and now write:

A striking feature is the observation that the column densities on the long light path are about the same as on the short although also some differences are found.

- p. 21380, l.29: The sentence “Until 2 Sep, the column densities are about the same.” would be better understandable if one would add “as for the long light path”.

We changed the sentence and now write:

Until 2 September , the column densities are about the same as for the long light path.

- Fig.1: Please give a legend to the colours of the figure, probably the green colour is the intertidal area, but definite assignment is always helpful. (If possible a higher quality figure would be desirable.)

Unfortunately the figure is not available in higher quality. However, we now state that the green area is the intertidal area around Mweenish Island.

- Fig.2, figure caption: The name/abbreviation “Nano-SMPS” is not explained.

We introduce the term nano-SMPS in the new Section 2.3, where we explain the particle measurements.

- Fig.2: The axis label is missing on the x-axis (Julian Day) as well as the label on the colour bar (particle concentration cm^{-3}). The information is given in the caption but should always be placed on the figure also.

We included the missing labels.

C. References:

p. 21372, l.26: I think in this place, reference to the article from Barrie et al. 1988 would be appropriate. (Reference: Barrie, L. A., Bottenheim, J. W., Schnell, R. C., Crutzen, P. J., and Rasmussen, R. A.: Ozone destruction and photochemical reactions at polar sunrise in the lower Arctic atmosphere, Nature, 334, 138–141, 1988.)

p. 21373, l.4: Please give references for the statement that at mid latitudes and Antarctica IO plays a role in the ozone destruction process, e.g. Read et al. 2008, or others. (Read, K. A., Mahajan, A. S., Carpenter, L. J., Evans, M. J., Faria, B. V. E., Heard, D. E., Hopkins, J. R., Lee, J. D., Moller, S. J., Lewis, A. C., Mendes, L., McQuaid, J. B., Oetjen, H., Saiz-Lopez, A., Pilling, M. J., and Plane, J. M. C.: Extensive halogenmediated ozone destruction over the tropical atlantic ocean, Nature, 453, 1232–1235, 2008.)

Both references were included

p. 21373, 1.19-20: Please give a reference to this statement.

We included the publication of McFiggans et al. 2004.

Technical Corrections

p. 21375, 1.24: “stabilized” should read “stabilize”

p. 21377, 1.17: “band” should read “bands”. For better reading maybe include the word “situated” or similar at the end of the sentence.

p.21378, 1.8: “ring” should read “Ring”

p.21380, 1.26: either “concentrations ... are” or “concentration ... is”

p. 21382, 1.3: either “from intertidal areas” or “from the intertidal area”

We included all corrections.

Reply to reviewer 2

1. The greater IO slant column densities in the 2 degree MAX-DOAS viewing angle compared to higher angles certainly suggests a strong ground-based source for this relatively short-lived species. This is discussed well qualitatively, but it seems to me (I’m not a MAX-DOAS expert) that there is a lot of quantitative data contained within the slant column differences which has not been exploited. For example, what is required to retrieve vertical profiles of IO? Can this be done within the scope of this paper? It would be very interesting.

We agree, that a vertical profile would be of great interest. However, as we already discussed in the reply to reviewer 1, we do not have enough measurements to retrieve a vertical profile of IO. Even if one would determine the light path the light traveled before it reaches the detector of the MAX-DOAS instrument (by using radiative transfer models), we know from the LP-DOAS measurements that there is a strong horizontal heterogeneity. To account for this heterogeneity, more than one MAX-DOAS instrument would be needed, because with just one instrument the system is under-determined. We choose not to give an air mass factor, since this would be misleading: The heterogenous spatial distribution of IO has to be taken into account when deriving IO concentrations, which, unfortunately with our data set is not possible. However, we included the IO/ O₄ ratio (Figure 10), which can be used as an approximation (see answer to comment 4 of Reviewer 1)

2. It is a shame that I₂ and OIO could not be measured above the present instruments' detection limits. OIO is notoriously difficult to detect, and although I₂ has a helpfully structured absorption spectrum it too has often not been observed by previous investigators even in the presence of strong IO signals. Thus the absence of I₂ and OIO data is not a substantial flaw in this work. However discussion on this topic would be improved considerably by including a summary of the I₂ and OIO concentrations observed by previous studies (with references), and thus whether the authors might expect to have seen I₂ and OIO above the detection limits of their various instruments. [It is not acceptable simply to refer readers to Peters et al (2005) in line 25 p21373 and expect them to form their own conclusions]. For example, the I₂ detection limit quoted for the shorter LP-DOAS path (282 pptv; top of page 21380) is rather larger than any ambient I₂ mixing ratio detected to my knowledge.

We agree that the manuscript can be improved by including a summary of the so far reported measurements of I₂ and OIO and a detailed discussion of our OIO/I₂ measurements. We now write on p21379, l.22ff:

OIO measurements are reported from various sites: At Mace Head up to 6.7 ppt were reported (Hebestreit, 2001; Saiz-Lopez and Plane, 2004), Allan et al. (2001) report up to 3.0 ppt from Cape Grim, Tasmania, Stutz et al. (2007) saw up to 30 ppt in the Gulf of Maine and Mahajan et al. (2009) saw a maximum of 8.7 ppt in Roscoff, France. For our measurements the OIO mixing ratios are below the detection limit of our instrument when measuring along the long light path. Therefore our results are not in contradiction to earlier studies. However, since Mweenish Bay is an area of high seaweed density, one could speculate that the OIO mixing ratios at Mweenish Bay could be higher than those reported from, e.g. Mace Head.

We also included a discussion about I₂ measurements and now write on p. 21380, 14ff:

I₂ has so far been observed at Mace Head (Saiz-Lopez and Plane, 2004, Bitter et al., 2005, Peters et al., 2005, Huang et al., 2009), at Roscoff, France (Mahajan et al., 2009) and at Mweenish Bay (Saiz-Lopez et al., 2006) where maximum mixing ratios of 140.7 ppt, 52.3 ppt and 193.3 ppt are reported, respectively. Of special interest for our measurements are the results of Huang et al. (2009), who measured up to 193.3 ppt of I₂ at the MRI when also LP-DOAS measurements were performed. However, Huang et al. used an in-situ technique and were measuring very close to the ground in the intertidal area. Although our detection limit on the long light path is considerably lower than the reported concentrations, 50 ppt along the

entire light path and at about 5 meters height would be required So the fact, that we were not able to detect I₂, is probably an indication that it is not homogenously distributed.

3. I agree with Referee #1 that much more could and should have been included about the particle observations and their relationship to the measured IO time series. After all, this is a major reason for doing this work! It is too important to limit to just one sentence in the site description when introducing figure 2 and one further sentence in section 4.2 discussing the LP-DOAS measurements

We already gave an answer to this remark in our reply to question 1 of Reviewer 1.

4. The paragraph “The most likely source of reactive iodine:::source of RHS” needs references (line 16 onwards on p21373).

We included a reference to the publication of McFiggans et al. 2004.

5. Line 26 p21373: “All of the above mentioned measurements were carried out using active LP-DOAS”. Not true. Bale et al used resonance fluorescence of iodine atoms and Saiz-Lopez et al includes measurements of I₂ by denuder tubes and broadband cavity ringdown spectroscopy.

We apologize for our mistake and changed the sentence to:

Most RHS measurements were made using active LP-DOAS.

6. Line 25 p21374: It is not necessary to reference the Merten (2008) PhD thesis if the pertinent information can be found in the Merten et al 2009 publication. Line 1 p21375: are the prisms 63 mm dia each, or the retro-reflectors? I agree with Ref #1’s comment about f-numbers. Please also include the focal length of the spectrometer, typical spectral resolution and typical integration times for each observation (these could equally go into section 3).

Since Merten et al. is not published yet, we also refer to the thesis. For f-numbers and integration times see additional comments and question number 2 of reviewer 1. The prisms are 63 mm diameter each. We specify this by writing on p.21375, l. 1:

(63 mm diameter each prism)

7. Line 15 p 21376: Only limited justification is given for choosing to concentrate on the core five-day period. What happened outside these days? – any useful data here too? Spectral fitting for IO (second paragraph in section 3.1): there is also an absorption band of H₂O around 445nm. Was this included in the fit?

We only had five days of measurements where we were able to detect IO on both light paths. So for the remaining days, no information regarding the spatial distribution of IO could be derived. We now write on p.21376, l. 15 ff.:

We concentrate on a core period of five days of measurements between 30 August and 4 September, since only during those five days we were able to detect IO on both LP-DOAS light paths.

We actually did not account for the H₂O band and therefore are thankful for the comment. We did a re-analysis of the data, including H₂O and O₄ in the fit. In the revised manuscript only data are shown, where H₂O and O₄ were accounted for in the analysis. Table 2 was updated.

8. Line 5 et seq p 21380: I agree that the observation of similar IO column densities in the two LP-DOAS paths suggests a common localised source. Yet for the lowest tide on the 30th Aug, systematic differences were observed and attributed to another seaweed bed becoming uncovered at the far end of the longer DOAS path (top of p 21381). Is it possible to use these data and the seaweed habitat map (Fig 3) to deduce approximate source strengths (e.g. per unit length) for the two emitting areas? After all, they comprise different seaweed species which might be expected to emit differently.

We agree that source strengths for different seaweed species would be desirable. However, the seaweed habitat map in Figure 3 is more of qualitative than of quantitative nature. To actually derive emission rates, one would need detailed information about the seaweed density along the light path. While in front of the MRI the seaweed was pretty much homogeneously distributed, the seaweed in front of Finish Island was more dispersed. We therefore do not have enough information to derive source strengths. Moreover the seaweed usually does not grow as separately as Figure 3 suggests, which would make an analysis of source strengths even more delicate.

9. Line 17 et seq 21380: comparisons with simultaneous IO measurements at Mace Head, whilst interesting background information, cannot be considered quantitative because IO is too short-lived to be transported between these measurement sites.

We did not intend to quantitatively compare the two measurement sites. However, we agree, that the sentence is misleading and now write:

This is in good agreement with modelling studies of Burkholder et al. (2004), who state that 50 to 100 ppt of IO are needed to explain nucleation events with particle concentration of 10⁶/cm³ similar to those that were observed each of the 5 days (T. Neary, University of Galway, personal communication, 2007).

Reply to reviewer 3

1. For instance, an example of a spectroscopic fit for the MAX-data is not provided, nor is there a discussion of visibility with the help of O₄ slant columns. Did the authors check the influence of the water Ring on the retrievals, since reflection from the surface is considered as one explanation for the large IO columns? Do the authors expect to observe OIO and I₂, both highly photolabile species, with the MAX-DOAS technique?

We are thankful for this comment. In the revised manuscript, we include a fit of the MAX-DOAS data (Figure 5) and the IO/ O₄ ratio (Figure 10). Since the albedo for water is only ~4%-6% the water ring effect is negligible and would already be covered by the normal (air) ring spectrum, as the water ring does not differ much from it. Thus all ring effects are already corrected with the ring spectrum. Additionally the water ring effect would also arise during days with low concentration what could not be seen in the data.

OIO was observed during the day at Mace Head (Peters et al., 2005) and at Appledore Island (Stutz et al., 2007). I₂ was detected during the day at Mace Head (Saiz-Lopez and Plane, 2004) and Roscoff, France (Mahajan et al., 2009). See also answer to comment 2 in the reply to Reviewer 2.

2. LP-DOAS fit: What do the authors gain from fitting the 4th line just after the gap in the wavelength window? This line is the most poorly fitted one. In the region of this fourth line, water and O₄ have strong absorption features as well. Why are those spectra not included in the fit? What about glyoxal? I'm puzzled by the drift in base line of the LPdata, which seems to be about 10 ppt over 5 days.

In the revised manuscript we included H₂O and O₄ in the fit (see also answer to comment 7 in the reply to Reviewer 2). We did a run with glyoxal for LP-DOAS and MAX-DOAS, but could not detect it. The detection limits are $4.0 \cdot 10^{15}$ molec/cm² (corresponding to 0.4 ppb) and $7.6 \cdot 10^{15}$ moelc/cm², respectively. We included the 4th line, because we figured out that this could improve the negative shift we see. The drift of the baseline is likely due to a degradation of the fibre (see answer to comment 5 of Reviewer 1).

What do the authors mean when stating that the light is not as well mixed with the new fibre (p. 21381, l. 14)? Since the individual spectra are normalized by lamp spectra, shouldn't that remove instrumental features such as drifts over time?

See answer to question 5 of Reviewer 1.

3. Why is the detection limit estimated from the statistical errors and not from the residual of the fit (p. 21377, l.14)?

The statistical error is calculated from the residuum of the fit using the approach described by Stutz and Platt, (1996). Thus the direct estimation from the residual and from the statistical

error leads to same detection limit. But as the detection limit is defined as twice the measurement error (in this case the observed statistical error) we chose this direct method. For details see Stutz and Platt (1996).

4. The polynomial in the DOAS fit does not only account for broad band structures due to scattering, but also due to broad band absorption and broad band instrumental features (p. 21377, l. 12).

That is correct and we include the following sentence in the revised manuscript (p.21377, l.12):

In addition a polynomial was included to account for broad band absorption structures, broad band structures due to scattering in the atmosphere and broad band instrument features.

5. The references are not up to date, e.g. Dixneuf et al. (2009), Mahajan et al. (2009), Read et al. (2008), Schönhardt et al. (2008), Rhamble special issue in ACPD as well now. It is not appropriate to cite a paper from 2005 (Peters et. al) for an overview of measurements made hitherto considering the progress over the last few years in our knowledge of iodine chemistry.

In our revised manuscript we include the above mentioned publications, which were not already included in the manuscript.

6. References that need to be added: p. 21373, l. 18: should reference one of Carpenter's papers

We included a reference to Carpenter et al. (2003).

p. 21373, l. 23: should reference Saiz-Lopez and Plane (2004), who made the first reported atmospheric observations of I₂ (at Mace Head!).

We are aware of that and therefore cite the paper in the previous line.

p. 21374, l. 5. Saiz- Lopez et al. (2006a) also deduced this from modelling the OIO/IO ratio and comparing with DOAS observations at Mace Head.

That is correct. However, we do not think that a change to the manuscript is necessary.

p. 21380, l 10: cite Mahajan (2009) as well as Peters

We included a reference to Mahajan et al. (2009).

p. 21380, l 20: Saiz-Lopez et al. (2006a) deduced these levels over the intertidal zone at Mace Head.

That is correct. However, we do not think that a change to the manuscript is necessary.

p. 21382, l. 13: Laminaria Hyperborea [...] which is a very strong emitter of iodine precursors

We included a reference to Ball et al. (2009).

p. 21382, l. 16: need to state here that this is in contrast to Saiz-Lopez and Plane (2004), who reported OIO at night and I₂ at low tides during both day and night.

Since they reported this from Mace Head this is not in contradiction to our statement.

p.21382, l. 25: add Saiz-Lopez et al., 2006a

We include a reference to Saiz-Lopez et al., 2006.

7. The authors cite a number of Heidelberg Master and PhD theses whereas many of these are irrelevant for this study, e.g. Stein; Martin.

We included the respective theses, because we think they provide relevant information directly related to the presented observations and therefore prefer to keep them.

8. Figures 1 and 7 need more explanations. Is the green shaded area in fig 1 the area that is covered with seaweed or is that the area that falls dry during lowest tide?

The caption of Figure 1 was updated. The Y-axis in Figure 7 shows which areas are exposed for which water level.

9. Figure 5 needs O₄ SCs to further investigate diurnal variations.

We included the IO/ O₄ ratio (Figure 10). Table 2 was updated for the O₄ analysis.

10. Minor typographical corrections: p. 21373, l. 24: hitherto

We changed the text as recommended.

p. 21374, l. 8: just crossed the intertidal

We changed the text as recommended.

p. 21376, l. 9: been the object of

We changed the text as recommended.