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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Observations of iodine monoxide (IO) columns from satellite" *by* A. Schönhardt et al.

A. Schönhardt et al.

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(Referring to the Interactive Comment of Anonymous Referee ♯1 from 1 November 2007)

The Interactive Comment of Anonymous Referee ♯1 is devided into 5 separate Comments, which will be addressed in the following in unchanged order.

Comment 1

The authors raise a rather interesting point when pointing out that cloud retrievals over ice and snow are currently not available (p. 12966, line 15-17). Cloud screening

is therefore a major issue when retrieving trace gases in the polar troposphere. Furthermore, when this works highlights that IO is hardly detectable in areas other than the surroundings of Antarctica. Therefore, the paper would definitely benefit from a more thorough description of how clouds are dealt with and the potential impact upon the retrieved IO columns.

The influence of clouds on the retrieval of iodine monoxide (IO) columns from satellite is a very important point. Clouds can interfere with tropospheric measurements from space considerably. Whenever possible, cloud selection criteria are usually applied in order to sort out cloud contaminated measurements from data sets and retain only (nearly) cloud free scenes.

In the present study, the focus is set on retrieval of IO above the Antarctic region, where the underlying area is mostly ice covered. Cloud screening for ice covered regions is still a very difficult task, and not easily available. Therefore, all data that satisfy a fit quality criterion are currently used for the maps showing IO slant columns, but no cloud screening is applied.

Nevertheless, it is important to estimate the potential impact of clouds on the retrieval process and the resulting trace gas column amounts. In general, there are two aspects that play a role and have to be considered. One aspect is the change in radiative transfer due to the clouds and its influence on the retrieved trace gas amount. The second aspect are potential retrieval errors that might be introduced due to altered conditions.

- The type of influence caused by a change in radiative transfer, is determined by the location of the trace gas with respect to the cloud. In general, there are three different cases. The trace gas can be situated either mainly above, mainly below

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or within the cloud.
If the IO is located above the cloud, the impact of the clouds depends on the relative surface spectral reflectance of the cloud and the underlying ground. In the case of an ice covered surface below, the surface spectral reflectance and with that the sensitivity of the measurement is comparable. No significant impact arises in this case.
For trace gases located below a cloud cover, sensitivity for detection of the species is lost. The light captured by the satellite will mainly originate from above the cloud and will not have passed through the layers close to the ground. In effect the trace gas amount will be underestimated.
The situation is more complex if the IO is situated partly within a cloud layer. For thick clouds, sensitivity is lost just as in the case before, but for thin clouds, due to multiple scattering the effective path length in the cloud and with this the sensitivity for the detection of the respective trace gas will be enhanced.
As most studies suggest that IO is mainly located close to the ground, it is probable that in most cases, the IO will be below the cloud, and some underestimation might occur.

- In addition, the retrieval has to be safe from potential retrieval errors. For ice free regions, tests with cloud screening have been preformed. They show, that no systematic retrieval error is introduced when fitting spectra that have been measured over clouds. This is a very important test, in the case that cloud screening cannot be applied in the region of interest.
Recently, a method has been developed at our institute using the PMD (Polarisation Measurement Devices) channels from SCIAMACHY, classifying clouds and surface types and enabling the identification and separation of clouds and ice covered surfaces. This dataset is yet unpublished, but a paper describing the algorithm and data will be submitted for publication in the near future. The cloud and surface type of the pixel is flagged, e.g., as desert, ice, cloud, ice cloud

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or water. Comparisons of the pixels marked as cloudy show good agreement with results of other cloud products and also the surface type characterisations of, e.g., desert and ice show promising results. From a study using this data product, the following further conclusions can be drawn. Clouds are a regular phenomenon over the Antarctic, but there still is a significant number of cloud-free scenes. The regions of enhanced IO values do not correlate with cloudy scenes, neither for ice free regions on the Earth as mentioned above nor for the Antarctic ice covered areas. Eliminating cloudy pixels over ice and snow using these preliminary cloud information does not have a large effect on the IO maps. The scatter in the data is slightly enhanced as a result of the reduced number of data points, but neither the absolute amount, nor the spatial pattern of IO is changed systematically.

In conclusion, the use of cloudy pixels in the data set does not lead to significantly larger retrieval errors, but may lead to a small underestimation of the IO slant columns. In response to the reviewers' comments we have added a short discussion of the effect of clouds and our recent findings to the manuscript.

Comment 2

The authors suggest that transport, and subsequent recycle, could account for the high IO levels observed in the interior of the Antarctic continent. However, a close examination of panel b) in Fig. 7 shows that the IO columns retrieved as far as $\geq$ *85 degrees south are as large (e.g.* $\approx 1 \times 10^{13}$ *molec cm*$^{-2}$*) as those over the Weddell Sea. This paper argues that the maximum columns are found over the Weddell Sea areas and also speculates that the sources must be from oceanic and/or sea-ice covered areas. Then, how can such a short-lived species be transported thousands of miles into the interior of Antarctica and still be in comparable concentrations to*

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those measured over the sources areas?. What is the sensitivity or reliability of the IO retrievals at large solar zenith angles?. Could cloud contamination or other factors in the retrieval method influence this surprising finding?. The authors should elaborate more on these points.

We agree with the comment, that the high amounts of IO over the Antarctic continent are surprising. There is no evident explanation for this finding up to now. Sources of IO on the Antarctic continent are not known, but neither can they be ruled out at this stage.

In the case that transport of iodine components is one reason for enhanced IO values on the continent, this does not necessarily have to be transport of the species IO itself only.

On a daily basis, the scatter and variability in the IO data is fairly large. This makes the tracing of a single transport event difficult. But it is important to note, that transport events for BrO, which also exhibits a short lifetime, are observed to extend extraordinarily far inland. On single days, enhanced values of BrO are detected in a large plume several hundreds of kilometres into the continent. This is shown nicely on a daily map of BrO on "http://www.iup.physik.uni-bremen.de/doas/scia_data_browser.htm?gas=bro:year=2007:month=10:day=10:view=sh" for example for the recent days Nov 6th, 2007, and Oct 10th, 2007, as well as for monthly averages such as for example in December 2005 or 2006. In general, also short lived molecules can be transported over far distances if efficient recycling mechanisms exist.

The following hypothesis may further explain the observed occurrence of IO over inland ice. Organic I is produced, e.g., by the maritime biosphere in the ocean, below and in the ice. Indeed, the exact processes for this still have to be proven. When openings in the ice occur, organic iodine components can be released, and eventually IO forms.

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At enhanced concentrations, IO produces higher oxides of I, which preferentially being hygroscopic attach to aerosols or themselves are aerosol condensation nuclei. The iodine in the aerosol or later cloud phase is then transported and deposited at the surface. In these regions snow photochemistry ($HNO_3 + h\nu \rightarrow OH + NO_2$) results in local high IO amounts via oxidation of HIO_3 by OH ($OH + HIO_3 \rightarrow H_2O + IO_3$, $IO_3 \rightarrow IO + O_2$). This hypothesis implies, that IO is recycled and reformed from the aerosol phase also at further distance from the sea ice. This way, the iodine is easily transported onto the Antarctic continent. These considerations cannot serve as a proof of this process, but give a possible explanation. The suggested process and the corresponding chemistry are subject of further research.

Saiz-Lopez et al. [2007b] have also taken recycling of iodine from higher oxides and through sea-salt aerosol into account in their chemical transport model. They demonstrate that these processes are required to explain observations of IO at, e.g., Halley Station [Saiz-Lopez et al. 2007a], Antarctica, at 12 km from the coast and the observed well-mixed amounts in the boundary layer, whereas without additional recycling processes the effective lifetime of IO would be too short to explain current measurements.

In the revised version, we have included a more detailed explanation of how IO may be transported towards the continent, following the above considerations. This comprises the discussion of transport of iodine species other than IO, especially the transport of iodine containing aerosols and subsequent recycle and snow photochemistry. We have included the remark, that direct transport events for the short-lived BrO reaching far into the Antarctic continent are indeed observed.

It is true that the SZA influences the quality of the DOAS fitting procedure. With larger SZA, usually the signal-to-noise ratio drops and the quality reduces. Therefore, our IO retrieval is restricted to SZA $\leq 84^\circ$ in general. It is important to note, that no sudden quality loss is encountered above a certain SZA value within this range. The retrieval

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residual shows the expected usual behaviour of rising residual with higher SZA, but no systematic influence on the retrieved IO amount above some certain value. If the SZA had such a negative influence on our retrieval leading to artefacts in the resulting IO amounts as considered possible by the referee, then this would necessarily have to be the case for the Northern and Southern Hemisphere in the same way. As this is not the case, and no sudden quality loss is encountered in our results, we conclude, that retieval issues related to the SZA do not lead to inaccurate results.

The impact of clouds is certainly an important issue for satellite retrievals, as discussed above. For our analysis, clouds do not lead to high IO values in general. So the presence of clouds does not lead to the enhanced IO values above Antarctica. For other regions, where cloud screening is possible more easily, comparisons with mainly cloud free results show, that the pattern of IO is not altered systematically when cloudy scenes are removed or included in the analysis.

Comment 3

The comparison of the IO columns with the Chlorophyll a measurements for October 2005 is interesting but hardly provides any information regarding the sources of iodine due to the limitation to make measurements over ice covered areas. This paper deals mostly with IO retrievals from space. Therefore I find the sections regarding atmospheric significance and sources too brief and somehow out of context. The atmospheric significance of iodine has already been dealt with in much more detail in different papers in the literature and hence this paper does not provide new insights. The section about the sources of iodine is a touch speculative and not well supported by modelling, laboratory or field work. For keeping the focus of the paper on the IO retrievals, as the title states, I would recommend the authors to shorten the mentioned sections.

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The Chlorophyll-a map from the MODIS instrument shown in the section on the interpretation of the IO satellite results hardly provides useful information in ice covered regions. This problem was addressed in the text. The MODIS map has data gaps in areas that would be particularly interesting for the discussion of IO sources.

As a consequence, the MODIS map has been discarded in the revised version. The discussion related to organic precursors has not been erased completely, as we consider it generally important and relevant. Nevertheless, the respective sections have been shortened as requested by the reviewer.

The sections regarding the atmospheric significance of IO have been revised and shortened.

Comment 4

Throughout the paper there are several references to comparisons of the IO columns presented here with ground-based observations made by Saiz-Lopez et al., 2007a. After a second read of the paper, the reference to this comparison becomes a little repetitive and it is difficult for the reader to visualize how good the comparison is. It would have been much easier for the reader to have a figure of the IO columns correlated with the mentioned ground-based data set included in this paper.

In our manuscript, we compare a time series of retrieved satellite IO amounts around Halley Station, Antarctica, with ground-based measurements published by [Saiz-Lopez et al. 2007a]. Following the suggestions of both, the editor and referee ♯1, this ground-

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based data from the CHABLIS campaign has been shown as an additional figure in the revised version of the manuscript for better comparison. The exact figure as printed in the cited paper has been displayed.

As the referencing to the comparison of the satellite results with the CHABLIS data from the publication by [Saiz-Lopez et al. 2007a] is slightly repetitive, the respective sections have been revised and some repetition has been removed.

Minor point

The IO columns are compared with those retrieved by Saiz-Lopez et al., 2007c however I do not find this reference in the introduction; if other measurements have been reported they should be included in the Introductory Section where the authors mention about previous IO observations.

The citation of [Saiz-Lopez et al. 2007c] was not included in the Introductory Section, but this has been done in the revised version of the manuscript. At the same point in the Introduction, we have taken the liberty of citing our previous work of [Schönhardt et al. 2007] instead of at a later position in the text.

References

[Saiz-Lopez et al.(2007a)] Saiz-Lopez, A., Mahajan, A. S., Salmon, R. A., Bauguitte, S. J.-B., Jones, A. E., Roscoe, H. K., and Plane, J. M. C.: Boundary layer halogens in coastal Antarctica, Science, 317, 348, doi:10.1126/science.1141408, 2007a.

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