

Interactive comment on "Long-term Time-series of Arctic Tropospheric BrO derived from UV-VIS Satellite Remote Sensing and its Relation to First Year Sea Ice" *by* Ilias Bougoudis et al.

Anonymous Referee #1

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This study presents a 22-year time series of Arctic tropospheric BrO from different satellite observations and discusses possible long-term changes, in particular in relation to changes in the distribution of first year sea ice. This is a pressing topic and the study is an important, highly relevant and timely contribution. As such I recommend publication in Atmos. Chem. Phys. after consideration of the general and specific comments. The manuscript is largely well written, but can be improved in parts for clarity of presentation. I have made specific comments and in addition would like to ask the senior coauthors to help improving the presentation where necessary.

General comments

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A central hypothesis of this study is that the transition from multi-year to first year sea ice in the Arctic ocean affects the distribution, number and intensity of bromine explosion events and thus Arctic tropospheric BrO. The potential mechanism(s) behind should be made a bit clearer in the Introduction and the discussion of results should be made under these assumptions. To be more specific: Multi-year ice that has survived at least one melt cycle has less bulk salinity compared to first year sea ice. The increase in first-year sea ice could potentially favour bromine explosion events by providing more sea salt bromine. To test this hypothesis, relating first year sea ice area with (excess) tropospheric BrO, as done in Fig. 11 is very useful and the implications of this relation should be stated accordingly. The correlation between first year sea ice area and BrO seems to be significant (by looking at Fig.11, I couldn't find a statistical test for significance) although of course not perfect. How much this relation explains (in terms of variance, trend, shift in geographic distribution) should be stated clearly, together with its limitations. Phrases like "linked in a complex way" is hide more than they reveal.

There may be other factors how sea ice age impact on tropospheric BrO. E.g. through a reduction in sea ice thickness. So while for the point above the distinction between first year and multi-year ice is most important (and the distinction between say 5 or 6 year old ice is not so important), for other factors the actual age may be critical. Please be specific in the discussion and in the presentation. Example 1: the colour scale in Fig. 9 makes it difficult to clearly identify if the majority of the ice in recent years is first year ice or 2 year old ice. Example 2: the age trend in Fig. 13: Even a relatively small trend may have resulted in a shift from multi-year to first year ice over the years. Try to be specific in the discussion of the implications.

Section 3.1 contains a general description of the DOAS method used to retrieve the BrO SCDs. I am not sure if a general description of the DOAS method is needed here, but in its current form there are too many mistakes and omissions to make it useful (specific points below). Please carefully check.

All these points as well as the specific points below should be easily improved so that I consider the changes needed as mostly minor in terms of time needed but nevertheless important to improve the manuscript.

Specific comments

P1,L11: First sentence of abstract is a bit disconnected here. Would be better in the Introduction. More generally I feel that at many places (in the abstract and elsewhere in the manuscript) "Arctic Amplification" could better be replaced by "Arctic warming" because what matters in this context is the warming, not so much the amplification (although this may be seen as pettifoggery).

P1, L15: "e.g. Hg": any other metals?

P1, L15: "22 year": suggestion: name range of years already here

P1,L19/20: "magnitude of BrO...of about 1.5%/year": what exactly increases with 1.5%/year? Tropospheric columns? Please be specific.

P1, L25: It is true that the understanding of Arctic Amplification is inadequate, but a few citations on Arctic Amplification may be useful. E.g. Pithan and Mauritsen, Nature Geosc., 2014.

P1, L24/25: "loss of sea ice" and "reduction of ice extent" are of course not independent. Maybe say loss of ice resulting in reduction of ice extent, thickness and reduced fraction of multi-year ice?

P2, L10: "over 30 years ago"

P2, L12: introduce "O3" when first used as "ozone (O3)"

P2, L13: I think this point should be made a bit clearer: O3 and OH are decreasing, but bromine radicals instead could act as oxidising agents. Are there references how the oxidising capacity overall changes?

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P2, L21: wicket sentence

P3, R4,R8: should that be "->" instead of "="?

P3, L19: Closing bracket missing.

P3, L19: This idea needs a bit more explanation: Transport of BrO plumes over large distance by deposition and reactivation due to release from snow pack and blowing snow?

P3, L24: "Polar Regions" -> "polar regions" (at many places in the manuscript)

P3, L24: "hostile" for what or whom? In spite of difficulties numerous studies have performed in-situ measurements or ground-based DOAS measurements in the Arctic. Satellite measurements are not "unique".

P3, L33: It is good to provide context by citing previous studies, but better cite what has been learned rather only what has been done.

P4, L2: "them" = "BrO explosion events" ?

P4, L10: at some stage you should mention that first year sea ice is more saline than multi-year ice

P4, L26: remove word "results"

P5, L9: This statement is too general. There were other satellite instruments before GOME, depending on what you mean by "many" and "key trace gases"

P5, L16: what is the difference between "near IR" and "short wave IR"?

P6, L5: throughput mentioned twice

P6, L9: This sentence seems odd. "Long changing polymers" -> "long chained polymers"?

P7, Eq.1: In eq.(1) the concentration of the gas j is missing.

P7, L14: really ABSORPTION cross sections or SCATTERING cross sections?

P7, L31: is a "four degree" polynomial a polynomial of fourth-order, i.e. with five degrees of freedom? Please be specific and consistent to avoid confusion.

P8, Eq.5: There is something wrong with eq. (5). The sum should be under the root. And not on the LHS.

P8, L16: Latitudes and longitudes need to be specified more consistently. E.g. "- $180^{\circ}E$ " should be " $180^{\circ}W$ " and "- $50.0^{\circ}S$ " should be " $50^{\circ}S$ ". If you use east and west longitudes please avoid " $235^{\circ}E$ to $270^{\circ}E$ " and use instead " $90^{\circ}W$ to $125^{\circ}W$ ".

P10, L1: Please mention this drift already in the instrument overview in Section 2.

P11, L2: This first sentence does not make much sense and is redundant.

P13, L13: what is the meaning of the word "high" here?

P13, L15: you can remove "due to Arctic Amplification" here. See also my general comment on Arctic Amplification.

P14, L10: There are also real stratospheric BrO trends due to changes in anthropogenic emissions!

P15, L1: I don't think you need to explain that temperatures in summer are higher because of increased solar insolation (it does not matter in this context), but you could specify in which month the maximum temperature and in which month the sea ice minimum are reached.

P15, L7: Looking at the spatial distribution may tell if there are (few and/or small) areas of bromine explosion in September or only a gradual increase in the background. Would it make sense to include somewhere also maps with BrO in autumn?

P22, L4: "in the early years, most of the BrO is found in the region of the Barents and Kara Sea": I am confused. Richter et al. (1998) show largest BrO enhancements in

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March 1997 over the Canadian Archipelago, Hudson Bay and north-west of Greenland.

P24, Fig11: I don't fully understand what are the points shown in the right hand side panels? Related to this: what is the correlation coefficients for the data shown in the time series on the left hand side? The same as given in the right hand side plots? Did you check if the correlations are significant? (By eye, the correlation between Arctic MAM mean BrO and 1st year ice extent in Fig. 11a seems significant.)

P24, L11: the phrase "fresh" ice is misleading, as first year ice has a higher salinity, i.e. is not "fresh"

P24, L15: What is the correlation coefficient? (See comment above.) A rigorous statistical analysis is more useful than the "anecdotal evidence" given in the following sentences.

P25, L4-7: I find these sentences confusing. My impression from Fig. 11a is that there is a positive correlation between first year sea ice and BrO, although this is clearly not the only factor. But then you say this agrees with Choi et al. who found even a negative correlation? I would say this is in contrast to Choi et al. and part of this difference may be attributed to a possible degradation of the OMI instrument?

P25, L22: Please specify the value of the autocorrelation used. What exactly is the meaning of the "period M"? Generally I am not convinced that the trend model with harmonics is an ideal choice as you don't have data during winter. For calculating trends in individual months as in Table 4 the harmonics are not needed at all.

P25, Table 4: It is a bit unusual to have the units in a column, instead of in the header.

P28, L10: First paragraph of Summary is redundant, largely repeats introduction

P29, L7: did you show that the correlation in Fig. 11a is not significant?

P29, L18: This statement on temperatures in 2016 is too vague. You could mention here that you have not considered changes in temperature and this may be another

factor affecting tropospheric BrO to be investigated in future studies.

P30, Author contributions: Sounds a bit strange that not all authors are named for their contributions here (not even for "providing insight and knowledge").

P31: Barrie and Platt listed twice

P32: Claas listed twice.

P32, L23: something is missing here

P33: Fickert listed twice

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